On the verge of *Umdeutung* in Minnesota: Van Vleck and the correspondence principle ⋆

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Abstract

In October 1924, *The Physical Review*, a relatively minor journal at the time, published a remarkable two-part paper by John H. Van Vleck, working in virtual isolation at the University of Minnesota. Van Vleck used Bohr's correspondence principle and Einstein's quantum theory of radiation to find quantum formulae for the emission, absorption, and dispersion of radiation. The paper is similar but in many ways superior to the well-known paper by Kramers and Heisenberg published the following year that is widely credited to have led directly to Heisenberg's *Umdeutung* paper. As such, it clearly shows how strongly the discovery of matrix mechanics depended on earlier work on the application of the correspondence principle to the interaction of matter and radiation.

Key words: Dispersion theory, John H. Van Vleck, Correspondence Principle, Bohr-Kramers-Slater (BKS) theory, Virtual oscillators, Canonical perturbation theory, Matrix mechanics

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1 Americans and quantum theory in the early 1920s

"[A]lthough we did not start the orgy of quantum mechanics, our young theorists joined it promptly" (Van Vleck, 1964, 24).[1] This is how John Has-

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⋆ This paper was written as part of a joint project in the history of quantum theory of the *Max Planck Institut für Wissenschaftsgeschichte* and the *Fritz-Haber-Institut* in Berlin.

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[1] Quoted and discussed in (Coben, 1971, 456)

brouck Van Vleck (1899–1980), our main protagonist,[2] described the American participation in the quantum revolution of the mid-1920s for an audience in Cleveland in 1963. Van Vleck spoke as the first recipient of an award named for America's first Nobel prize winner in physics, Albert A. Michelson (1825–1931). Van Vleck was fond of the "orgy"-metaphor, which he had picked up from the German-American physicist Ralph Kronig (1904–1995). In his Michelson address he mentioned how he had used it off-handedly a few years earlier during a press conference at Harvard on Russian contributions to science, only to find himself quoted in a newspaper as saying that there had been a "Russian orgy in theoretical physics" (ibid.). He was selling himself and his countrymen short, however, by characterizing the American contribution to the quantum revolution as simply a matter of joining an orgy started by the Europeans and in full swing by the time the Americans arrived on the scene.

Eight years later, Van Vleck, in fact, took exception to what sounds like a similar characterization given by another leading American physicist of his generation, Isidor I. Rabi (1898–1988). Van Vleck (1971, 7) quoted a comment that Rabi made in a TV documentary about Enrico Fermi (1901–1954):

> We had produced a large number of people who had been brought up to a certain level, then needed some help, some leadership to get over the hump. Once they were over the hump they were tremendous. People of my generation brought them over the hump, largely from attitudes, tastes, and developments which we had learned in Europe.

As Thomas S. Kuhn and others have emphasized, Rabi's point was that American physicists returning from Europe rather than European émigrés were mainly responsible for the coming of age of American physics.[3] This issue has been hotly debated in the history of physics literature.[4] Our study of early American contributions to quantum theory supports the observation by

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[2] For an (intellectual) biography of Van Vleck, known to his colleagues simply as 'Van', see the superb dissertation of Fred Fellows (1985) and a biographical memoir by Phil Anderson (1987), one of Van Vleck's many graduate students. Anderson shared the 1977 Nobel prize with Van Vleck and Sir Nevill Mott "for their fundamental theoretical investigations of the electronic structure of magnetic and disordered systems." Van Vleck won the prize for work begun in the early 1930s that earned him the title of "father of modern magnetism."

[3] See p. 20 of the transcript of the last of five sessions of Kuhn's interview with George E. Uhlenbeck (1900–1988) for the *Archive for the History of Quantum Physics* (AHQP) on December 9, 1963.

[4] For a concise summary and detailed references to the older literature, see (Moyer, 1985, 171–173). Whereas our focus will be on American contributions to atomic physics, Assmus (1992, 1999) has argued that American theoretical physics came of age in molecular physics (cf. note 28 below).

Sam Schweber (1986, 58) that in the 1930s theoretical physics was "already a thriving enterprise in the United States. The refugee scientists resonated with and reinforced American strength and methods: they did not create them."

Commenting on Rabi's remark, Van Vleck (1971, 6) reiterated the point of his Michelson address that "quantum mechanics was a basically European discovery,"[5] but, he added, "there has been too much of an impression that American physicists, even in the application of quantum mechanics, were effective only because they had the aid of European physicists, either by going to Europe, or because of their migration to America." Van Vleck, who was proud to be a tenth-generation American,[6] received his entire education in the United States. He hardly had any contact with European physicists before 1925, although he did meet a few on a trip to Europe with his parents in the summer of 1923. In Copenhagen, he called on Niels Bohr (1885–1962), who suggested that he get in touch with Hendrik A. (Hans) Kramers (1894–1952), Bohr's right-hand man and one of the central characters in our story.[7] Kramers was not in Denmark at the time but in his native Holland. Decades later, when he received the Lorentz medal from the Royal Dutch Academy of Sciences, Van Vleck recalled how he had searched for Kramers all over the Netherlands. After he had finally tracked him down—it can no longer be established whether this was in Bergen aan Zee or in Schoorl—the two men went for a long walk in the dunes along the North-Sea coast: "This was the beginning of a friendship that lasted until his passing in 1952" (Van Vleck, 1974, 9). Unfortunately, Van Vleck does not tell us what he and Kramers talked about.

1.1 Education

Van Vleck learned the old quantum theory of Bohr and Arnold Sommerfeld (1868–1951) at Harvard as one of the first students to take the new course on quantum theory offered by Edwin C. Kemble (1889–1984), the first American physicist to have written a predominantly theoretical quantum dissertation. Kemble's course roughly followed (Sommerfeld, 1919), the bible of the old quantum theory. Van Vleck supplemented his studies by reading (Bohr, 1918) and (Kramers, 1919) (Fellows, 1985, 17). Van Vleck was part of a remarkable cohort of young American quantum theorists, which included Gregory Breit

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[5] In 1928, he had characterized it as "the result of the reaction of mind on mind among European talent in theoretical physics" (Van Vleck, 1928, 467).

[6] He could trace his ancestry back to the fifteenth century, to a certain Johan van Vleeck of Maastricht. One of the latter's descendants, Tielman van Vleeck (or von Fleck), left Bremen for New Amsterdam in 1658 (Fellows, 1985, 5–6)

[7] See p. 14 of the transcript of the first of two sessions of Kuhn's AHQP interview with Van Vleck in October 1963.

(1899–1981),[8] John C. Slater (1900–1976),[9] Harold C. Urey (1893–1981), and David M. Dennison (1900–1976).[10] Just as Van Vleck was the first to write a purely theoretical dissertation at Harvard in 1922, Dennison was the first to do so at the University of Michigan in 1924.[11]

The older generation—men such as Michelson and Robert A. Millikan (1868–1953)—recognized that the United States badly needed to catch up with Europe in quantum physics. The Americans were already doing first-rate experimental work. One need only think of Millikan (1916) verifying the formula for the photo-electric effect (Stuewer, forthcoming) or of Arthur H. Compton

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[8] Breit was born in Russia and came to the United States in 1915. "John Wheeler relates a story told to him by Lubov [Gregory's sister] that she and Gregory were vacationing on the sea when the call to leave Russia came, and they 'came as they were.' For Gregory this meant dressed in a sailor suit with short pants; he was still wearing it when he enrolled in Johns Hopkins (at age sixteen!). Wheeler attributes some of Gregory's subsequent reticence to the ragging he took at the hand of his classmates for his dress" (Hull, 1998, 29–30). In the run-up to the Manhattan Project, Breit served as "Coordinator of Rapid Rupture," but resigned in May 1942 over security concerns bordering on paranoia (Goodchild, 1980, 48). He was replaced by J. Robert Oppenheimer (1904–1967). True to form, Breit declined to be interviewed for the AHQP (see the correspondence between Breit and Kuhn included in the folder on Breit in the AHQP). In a memorandum dated April 8, 1964 (included in the same folder), Kuhn describes how they met for lunch, but did not get beyond "casual reminiscences." Kuhn ends on a positively irritated note: "we broke off amicably but with zero achievement to report for the project."

[9] On Slater, see, e.g., (Schweber, 1990).

[10] Reminiscences about the early days of quantum physics in the United States can be found in (Van Vleck, 1964, 1971) and in (Slater, 1968, 1973, 1975). It is also an important topic of conversation in Kuhn's AHQP interviews with Van Vleck, Slater, Dennison, and Kemble. These interviews need to be handled with care. In the case of Slater and Van Vleck, one can say, roughly speaking, that the former had a tendency to exaggerate the importance of American contributions, especially his own, while the latter tended to downplay their importance. The interviews with Slater and Van Vleck and part of the interview with Kemble all took place in October 1963. Van Vleck was present at the first session of the interview with Slater and at the second session of the interview with Kemble. It was only natural for Van Vleck to get involved in Kuhn's project. As a young physicist right after World War II, Kuhn had worked with Van Vleck (Anderson, 1987, 518), a collaboration that resulted in a joint paper (Kuhn and Van Vleck, 1950).

[11] See p. 10 of the transcript of the first of three sessions of Kuhn's AHQP interview with Dennison in January 1964. Dennison could take advantage of the presence of Oskar Klein (1894–1977), who was a visiting faculty member in the physics department in Michigan from 1923 to 1925 (Sopka, 1988, 321). This is where Klein came up with what is now known as the Klein-Gordon equation; it is also where he made his contribution to what is now known as the Kaluza-Klein theory (see p. 13 of the transcript of the interview with Uhlenbeck cited in note 3).

(1892–1962) (Compton, 1923) producing strong evidence for the underlying hypothesis of light quanta (Stuewer, 1975). Theory, however, was seriously lagging behind. In 1963, Van Vleck's teacher, Ted Kemble, recalled:

> [T]he only theoretical physicists in the country at that time were really men on whom the load of teaching all the mathematical physics courses lay, and they all spent their time teaching. It wasn't, as I remember, a constructive occupation. [12]

When quantum theory arrived on the scene, some experimentalists actually tried their hands at teaching it themselves (Coben, 1971, 444). In this climate, young American physicists with a knack for theory became a hot commodity. They received fellowships to learn the theory at the feet of the masters in Europe and offers of faculty positions straight out of graduate school with provisions protecting them from the drudgery of undergraduate teaching so that they could concentrate on advanced graduate courses and research. [13]

1.2 Postdocs and faculty positions

The careers of the young theorists listed above amply illustrate the new opportunities in the mid-1920s. Slater went to Europe on a Sheldon fellowship from Harvard and spent the first half of 1924 with Bohr and Kramers in Copenhagen. During this period, Urey and Frank C. Hoyt (1898–1977) were in Copenhagen as well, Urey on a small fellowship from the American-Scandinavian Foundation, Hoyt on a more generous *National Research Council* (NRC) fellowship paid for by the Rockefeller foundation. [14] Among the visitors the Americans got to meet in Bohr's institute were Werner Heisenberg (1901–1976) and Wolfgang Pauli (1900–1958). [15] Hoyt, a promising student

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[12] P. 4 of the transcript of the second session of the AHQP interview with Kemble on October 1, 1963. See also p. 10 of the transcript of the first session on May 11, 1962. Kemble allowed one exception, Arthur Gordon Webster (1863–1923), but Webster, Kemble said, "just couldn't keep up with what was going on when the quantum theory began. I always understood that the reason he killed himself was simply because he discovered that suddenly physics had gone off in a new direction and he was unable to follow, and he couldn't bear to take a seat in the back and be silent" (p. 12 of the transcript of the first session).

[13] For further discussion of quantum physics in America before the mid-30s, see (Coben, 1971), (Seidel, 1978), (Kevles, 1978, 168–169), (Weart, 1979), (Schweber, 1986), (Holton, 1988), and, especially, (Sopka, 1988).

[14] See (Robertson, 1979, 157), (Sopka, 1988, 71, 97), and Slater to Van Vleck, July 27, 1924 (AHQP)

[15] In the letter to Van Vleck cited in the preceding note, Slater painted an unflattering portrait of his illustrious contemporaries: "Heisenberg is a very nice red haired unassuming young chap, talks a little English, and everybody likes him. Pauli is

who never reached the level of distinction of the cohort immediately following him,[16] was in Copenhagen for almost two years, from October 1922 to September 1924, Urey for less than one, from September 1923 to June 1924, and Slater only for a few months, from December 1923 to April 1924. In October 1924, Dennison arrived in Copenhagen, on an *International Education Board* (IEB) fellowship, another fellowship paid for by the Rockefeller Foundation.[17] The state of quantum theory in America was already beginning to change at that point. Like Hoyt, Dennison was awarded a NRC fellowship, but was told that he could only spend the money at an American institution.[18]

Van Vleck and Slater, who both started graduate school at Harvard in 1920 (Van Vleck in February, Slater in September) and lived in the same dormitory,[19] at one point discussed going to Copenhagen together upon completion of their Ph.D. degrees in 1923. In the end, Van Vleck decided to accept the offer of an assistant professorship at the University of Minnesota, where he started in the fall of 1923. When the university hired Van Vleck it also hired Breit so that its new recruits would not feel isolated.[20] Breit and Van Vleck replaced W. F. G. Swann (1884-1962) who had left for Chicago, taking his promising graduate student Ernest O. Lawrence (1901–1958) with him. As Van Vleck (1971, 6) notes wryly: “A common unwitting remark of the lady next to me at a dinner party was “Wasn’t it too bad Minnesota lost Swann—it took two men to replace him!”” Just as Minnesota hired both Breit and

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as different as he could be, a big fat Jew, with a very good opinion of himself and a great liking to hear himself talk. Still, he is a good natured and accomodating person, and well liked.”

[16] He wrote several papers on applications of Bohr’s correspondence principle (Hoyt, 1923, 1924, 1925a,b). The first two are cited in (Van Vleck, 1924b, 334) and all but the second are cited in (Van Vleck, 1926a, 124, 146). (Ladenburg and Reiche, 1924, 672) cites the second paper, referring to the author as “W. C. Hoyt.” Hoyt also translated Bohr’s Nobel lecture into English (Bohr, 1923a). Hoyt ended up making a career in weapons research rather than in academic physics. After the war, he worked at Argonne National Laboratory, Los Alamos, and Lockheed. John L. Heilbron interviewed Hoyt for the AHQP but he did not remember much of the early days of quantum theory.

[17] Bohr arranged for one of these fellowships to pay for Heisenberg’s visit to Copenhagen in the fall of 1924 (Cassidy, 1991, 180, 183). See also the acknowledgment in (Heisenberg, 1925b, 860).

[18] See p. 12 of the transcript of the first of three sessions of the interview with Dennison cited in note 11. In 1923, the NRC had likewise rejected the proposal of Robert S. Mulliken (1896–1987) to go work with Ernest Rutherford (1871–1937) in Cambridge. Mulliken became a NRC research fellow at Harvard instead (Assmus, 1992, 23).

[19] See Van Vleck, *1920–1930. The first ten years of John Slater’s scientific career.* Unpublished manuscript, American Institute of Physics (AIP), p. 2.

[20] See p. 14 of the transcript cited in note 7.

Van Vleck in 1923, the University of Michigan hired not one but two students of Paul Ehrenfest (1880–1933) in 1927, Uhlenbeck and Samuel A. Goudsmit (1902–1978) (Coben, 1971, 460).[21] In addition Michigan hired Dennison, its own alumnus, upon his return from Copenhagen. Ann Arbor thus became an important center for quantum theory, especially in molecular physics (Assmus, 1992, 4, 26, 30)

While Uhlenbeck and Goudsmit essentially remained in Ann Arbor for the rest of their careers, neither Breit nor Van Vleck stayed long in Minneapolis. Breit left for the Carnegie Institution of Washington after only one year, Van Vleck for the University of Wisconsin, his *alma mater*, after five. Van Vleck agonized over the decision to leave Minnesota, where he had been promoted to associate professor in June 1926 and, only a year later, to full professor (Fellows, 1985, Ch. VII). Moreover, his wife, Abigail Pearson (1900–1989), had strong ties to Minneapolis.[22]

To replace Van Vleck, Minnesota made the irresistible offer of a full professorship to the young Edward U. Condon (1902–1974). Minnesota had offered Condon an assistant professorship the year before. At that point, Condon had received six such offers and had decided on Princeton (Condon, 1973, 321). His laconic response to this embarrassment of riches: "The market conditions for young theoretical physicists continues [sic] to surprise me" (Coben, 1971, 463). Before his first Minnesota winter as a full professor, Condon already regretted leaving New Jersey. He returned to Princeton the following year. Condon, Rabi, and Oppenheimer[23] were the leaders of the cohort of American quantum theorists graduating right after the quantum revolution of 1925. The cohort most relevant to our story graduated right before that watershed.

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[21] See also (Sopka, 1988, 149) and the AHQP interview with Dennison mentioned in note 11. The recruiting was done by Walter F. Colby (1880–1970) and Harrison M. Randall (1870–1969).

[22] This is how Anderson (1987, 525) characterized the couple: "They were inseparable, and her wit was an excellent foil to his, slightly more personal and acerbic, occasionally expressing the impatience with people that Van never permitted himself." Abigail made a generous donation to the University of Minnesota to support the Abigail and John van Vleck Lecture Series. Phil Anderson gave the inaugural lecture in 1983 and the series has brought several Nobel prize winners to Minneapolis since. The main auditorium in the building currently housing the University of Minnesota physics department is also named after the couple.

[23] Oppenheimer enrolled as an undergraduate at Harvard in 1922, two years after Slater and Van Vleck started graduate school there.

It was during Van Vleck's tenure in Minnesota that his senior colleague John T. Tate (1889–1950) took over as editor-in-chief of *The Physical Review* (Sopka, 1988, 142–145, 203, note 11). Van Vleck (1971, 7–8) stressed the importance of this event, describing it as "another revolution" in the "middle of the quantum revolution." Van Vleck was very appreciative of Tate's role: "He published my papers very promptly, and also often let me see manuscripts of submitted papers, usually to referee." Thanks in no small measure to Van Vleck and other young whippersnappers in quantum theory, Tate turned what had been a lack-luster publication into the prestigious journal it still is today.[24] Van Vleck (1964, 22, 24) recalled the transformation:

> *The Physical Review* was only so-so, especially in theory, and in 1922 I was greatly pleased that my doctor's thesis [Van Vleck, 1922] was accepted for publication by the *Philosophical Magazine* in England . . . By 1930 or so, the relative standings of *The Physical Review* and *Philosophical Magazine* were interchanged . . . Prompt publication, beginning in 1929, of "Letters to the Editor" in *The Physical Review* . . . obviated the necessity of sending notes to *Nature*, a practice previously followed by our more eager colleagues [see, e.g., (Breit, 1924b), (Slater, 1924, 1925c)].

Van Vleck's impression is corroborated by two foreign-born theorists who made their careers in the United States, Rabi and Uhlenbeck (Coben, 1971, 456). Rabi was born in Galicia[25] but moved to New York City as an infant. Rabi liked to tell the story that, when he returned to Europe to study quantum theory in Germany in 1927, he discovered that *The Physical Review* "was so lowly regarded that the University of Göttingen waited until the end of the year and ordered all twelve monthly issues at once to save postage" (ibid.). On other occasions, Rabi told this story about Hamburg University (Rigden, 1987, 4). He told Jeremy Bernstein (2004, 28) that "in Hamburg so little was thought of the journal . . . that the librarian uncrated the issues only once a year." In a talk about Condon, Rabi (1975, 7) elaborated on the mediocrity of *The Physical Review*:

> it was not a very exciting journal even though I published my dissertation in it. And we felt this very keenly. Here was the United States, a vast and rich country but on a rather less than modest level in its contribution to physics, at least per capita. And we resolved that we would change the situation.

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[24] Tate edited the journal from 1926 to 1950. It is largely in recognition of this achievement that the current Minnesota physics building is named after Tate.

[25] Which makes him an *Ostjude* and not an "Ausjude" (whatever *that* may be) as Bird and Sherwin (2005, 76) claim in a bizarre passage in their otherwise excellent Oppenheimer biography.

> And I think we did. By 1937 the *Physical Review* was a leading journal in the world.

Uhlenbeck remembered how as a student in Leyden he viewed *The Physical Review* as "one of the funny journals just like the Japanese."[26] His initial reaction to the job offer from Michigan suggests that, at least at the time, his disdain for American physics journals extended to the country as a whole: "If it had been Egypt or somewhere like that, I would have gone right away, or China, or even India, I always wanted to go to exotic places [Uhlenbeck was born in Batavia in the Dutch East Indies, now Jakarta, Indonesia]; but America seemed terribly dull and uninteresting"(Coben, 1971, 460).[27]

1.4 Van Vleck, the correspondence principle, and matrix mechanics

Given the disadvantage they started out with, American theorists in the early 1920s would have done well had they just absorbed the work of their European counterparts and transmitted it to the next generation of American students. They did considerably better than that. Even before the breakthrough of Heisenberg (1925c) they started making important contributions themselves.[28] Slater was one of the architects of the ill-fated but highly influ-

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[26] See p. 20 of the transcript cited in note 3.

[27] In the AHQP interview with Uhlenbeck (see note 3), one finds no such disparaging comments about America. In fact, Uhlenbeck talks about how he had reluctantly agreed to return to the Netherlands in 1935 to replace Kramers, who had left Utrecht for Leyden to become Ehrenfest's successor after the latter's suicide (see p. 9 of the transcript cited in note 3). Uhlenbeck went back to Ann Arbor in 1939.

[28] According to Alexi Assmus (1992, 8), "[a]tomic physics was shark invested waters and was to be avoided; U.S. physicists would flourish and mature in the calmer and safer tidepools of molecular physics" (see also Assmus, 1999, 187). She sees the early contributions of Slater and Van Vleck to atomic physics, which will be the focus of our study, as exceptions to this rule: "Van Vleck and Slater viewed themselves as the younger generation, as central figures in the "coming of age" of U.S. physics. They had been given the knowledge that Kemble and his generation could provide and felt themselves capable of pushing into areas where the physics community in the United States had not dared to venture. Still, after experiences had muted their youthful exuberance, they turned to the by-then traditional problems of American quantum physics problems that addressed the building up of matter rather than its deconstruction" (Assmus, 1992, 22). We hope to show that American work in atomic physics was significantly more important—if not in quantity, then at least in quality—than these remarks suggest. At the same time, Assmus is probably right that the Americans contributed more to molecular than to atomic physics. This would fit with the thesis of Schweber (1990, 398–406) that "Americans contributed most significantly to the development of quantum mechanics in quantum chemistry."

ential Bohr-Kramers-Slater (BKS) theory (Bohr, Kramers, and Slater, 1924) (see sec. 2.2). Less well-known is the contribution of Van Vleck. In 1924 he published a remarkable two-part article in *The Physical Review* on the emission, absorption, and dispersion of radiation (Van Vleck, 1924b,c), in which he gave quantum analogues of classical relations guided by Bohr's correspondence principle (Bohr, 1918) and the quantum theory of radiation of Albert Einstein (1879–1955) (Einstein, 1916a,b, 1917).[29] At the suggestion of Pascual Jordan (1902–1980), Bartel Leendert van der Waerden (1903–1996) included the quantum part of the paper (Van Vleck, 1924b) in his well-known anthology on the development of matrix mechanics (Van der Waerden, 1968, preface).[30] Van Vleck told Kuhn in his AHQP interview that Born and Jordan, Born's assistant at the time, were studying this paper of his when the paper by Heisenberg (1925c) introducing matrix mechanics arrived in Göttingen.[31] This is something of an embellishment. (Born and Jordan, 1925a), largely inspired by (Van Vleck, 1924b,c),[32] was actually submitted to *Zeitschrift für Physik* on June 11, 1925, a few weeks before Heisenberg's breakthrough (Cassidy, 1991, 198).

Incidentally, Van Vleck (1971, 7) pointed to these pre-1925 contributions by

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[29] Originally, Van Vleck's paper was to have three parts. Van Vleck did not finish the third part at the time. As he explained in a letter to Max Born (1882–1970) on November 30, 1924 (AHQP): "Part III which is not yet ready relates to classical black body radiation rather than quantum theory." It was only toward the end of his life that he returned to the masterpiece of his youth. Three years before he died he published a paper, co-authored with D. L. Huber, that can be seen as a substitute for part III. As the authors explain: "Part III was to be concerned with the equilibrium between absorption and emission under the Rayleigh-Jeans law. It was never written up for publication [. . .] The idea occurred to him [i.e., Van Vleck] to use the 50th anniversary of Parts I and II as the date for publishing a paper which would start with Part III and might even bear its title. Although he did not succeed in meeting the deadline, it still provided a partial motivation for collaborating on the present article" (Van Vleck and Huber, 1977, 939).

[30] See also (Sopka, 1988, 110–111).

[31] See p. 24 of the transcript cited in note 7.

[32] See Jordan to van der Waerden, December 1, 1961 (quoted in Van der Waerden, 1968, 17), and (Jordan, 1973). We quote from the latter: "Van Vleck gave a derivation of Einstein's laws of the relation between the probabilities of spontaneous emission and positive and negative absorption. This result of Einstein's had been looked upon for a long time in a sceptical manner by Niels Bohr; now it was highly interesting to see, just how from Bohr's preferred way of thinking, a derivation of Einstein's law could be given. Born and I performed a simplified mathematical derivation of the results of Van Vleck. Our article on this topic [Born and Jordan 1925a] *did not contain anything new apart from our simpler form of the calculation*, but by studying this topic we both came to a more intimate understanding of Bohr's leading ideas" (Jordan, 1973, 294, our emphasis). See secs. 3.2, 3.3 and 4.1 below for discussion of Van Vleck's correspondence principles for emission and absorption.

Slater and himself to demonstrate the inaccuracy of Rabi's characterization of American work in quantum theory quoted earlier. Even at the time, Van Vleck had felt that the Europeans had not given the work of the Americans its due. He complained about this in a letter to Born of October 19, 1925, a draft of which can be found in the AHQP. The letter gives some impression of the breadth of the early American contributions to atomic physics, not just by Van Vleck himself but also by his teacher Kemble:

> I am writing this letter regarding some of the references to my work in your articles. I fully realize that an occasional error in a reference is unavoidable, for I have made such mistakes myself. I would gladly overlook any one error, but inasmuch as there are two or three instances, it is perhaps worth while to call them to your attention. On p. 332 of your treatise on "Atommechanik" [Born, 1925], the reference to my work on the crossed-orbit model of the normal helium atom is given as [Van Vleck, 1923]. This reference is only to the abstract of some work on *excited* helium and the references to my articles on *normal helium* are [Van Vleck, 1922a] (abstract of a paper delivered before the American Physical Society in Dec. 1921), and especially [Van Vleck, 1922b], where the details of the computations are given. This incorrect reference to a paper on another subject published a year later makes it appear as though my computation was published simultaneously or later than that of Kramer[s] [(Kramers, 1923), cited in the same footnote as (Van Vleck, 1923) in (Born, 1925, 332)]. The same error is also found in your article [Born, 1924b] on perturbation theory . . . Also in your book on Atommechanik [(Born, 1925, 332), the sentence with the footnote referring to (Kramers, 1923) and (Van Vleck, 1923)] you say "das raumliche [sic] Modell ist ebenfalls von Bohr vorgeschlagen" [the spatial model has also been proposed by Bohr], without any mention of the name Kemble, who proposed the crossed-orbit model in [Kemble, 1921] before [Bohr, 1922].

Van Vleck then comes to the most egregious case, Born's failure to properly acknowledge his two-part paper on the correspondence principle in (Born and Jordan, 1925a). Especially in view of Jordan's later remarks on the importance of Van Vleck's work for their own, which we quoted above (see note 32), the authors were very stingy in giving Van Vleck credit. Of course, Jordan probably knew about Van Vleck's complaint to Born on this score and may have wanted to make up for their lack of generosity. Van Vleck wrote:

> I was much interested in your recent article on the Quantization of Aperiodic Systems, in which you show that the method of Fourier integrals gives many results obtained by "Niessen and Van Vleck" [Born and Jordan, 1925a, 486], placing my name after Niessen's [Kare Frederick Niessen (1895–1967)], even though his paper [Niessen, 1924] did not appear until Dec. 1924 while the details of my computations were given in the Physical Review for Oct. 1924 [Van Vleck, 1924b, 1924c] and a preliminary notice published in the

> Journal of the Optical Society for July 1924 [Van Vleck, 1924a], before Niessen's article was even submitted for publication. I think you wrote me inquiring about my work shortly after the appearance of this preliminary note, and so you must be aware that it was the first to appear . . . inasmuch as Niessen's discussion is somewhat less general than my own, it seems to me that it scarcely merits being listed first (Van Vleck to Born, October 19, 1925 [AHQP]).

Writing from Cambridge, Massachusetts, where he was visiting MIT, Born apologized to Van Vleck on November 25, 1925 (AHQP).[33] Born had indeed written to Van Vleck concerning (Van Vleck, 1924a), albeit a little later than the latter remembered. On October 24, 1924. Born had written:

> While we already came close to one another in the calculation of the Helium atom, I see from your paper "A Correspondence Principle for Absorption" [Van Vleck, 1924a] that we now approach each other very closely with our trains of thought . . . I am sending you my paper "On Quantum Mechanics" [Born, 1924], which pursues a goal similar to yours.

This goes to show—Rabi's anecdotal evidence to the contrary notwithstanding—that at least some European physicists did keep up with theoretical work published in American journals, the *Journal of the Optical Society of America* in this case, even if they were not particularly generous acknowledging the importance of such work in print.

Van Vleck was justifiably proud of his two-part paper of 1924 on the correspondence principle. As in the case of the better-known paper by Kramers and Heisenberg (1925), which was completed a few months later, covers much the same terrain and exploits similar methods, one is struck in hindsight by how close Van Vleck came to anticipating matrix mechanics.[34] In his 1963 interview for the AHQP, Kuhn reminded Van Vleck of a remark two years earlier to the effect that if he had been "a little more perceptive" he "might have taken off from that paper to do what Heisenberg did." "That's true," Van Vleck conceded, but added with characteristic modesty: "Perhaps I should

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[33] Born had been less generous in the case of a similar complaint from America a few years earlier (see note 57 below). When Van Vleck found out that Born was in the US, he tried to arrange for Born to visit Minneapolis, but by the time the administration of the University of Minnesota got its act together, Born could no longer fit a trip to Minnesota in his schedule (see Van Vleck to Born, January 8, 1926 [draft] and Born to Van Vleck, January 13, 1926 [AHQP]).

[34] Max Dresden (1987, 275), one of Kramers' student and his biographer, calls (Kramers and Heisenberg, 1925) "the direct, immediate, and exclusive precursor to the Heisenberg paper on matrix mechanics." Martin J. Klein (1970, 31) is more restrained but does say that "this work was the immediate predecessor of Heisenberg's new quantum mechanics."

say *considerably* more perceptive."[35] In the biographical information he supplied for the AHQP, Van Vleck noted: "In the two or three years after my doctorate . . . my most significant paper was one on the correspondence principle for absorption . . . It was somewhat related to considerations based on the correspondence principle that led Heisenberg to the discovery of quantum mechanics, but I did not have sufficient insight for this." This modest assessment is reflected in the discussion of the relation between Van Vleck's work on the correspondence principle and Heisenberg's matrix mechanics by Van Vleck's biographer, Fred Fellows (1985, 74–81). Phil Anderson (1987, 506) is less reserved: "This paper comes tantalizingly close to the kind of considerations that led to Heisenberg's matrix mechanics."

Van Vleck did not pursue his own research any further in 1924 and instead spent months writing—and, as he jokingly put it, being a "galley slave" (Fellows, 1985, 100) of—a *Bulletin* for the NRC on the old quantum theory (Van Vleck, 1926a).[36] With his masterful survey he would surely have rendered a great service to the American physics community had it not been for the quantum revolution of 1925–1926. Like Pauli's *Handbuch* article (Pauli, 1926), the *Bulletin* was, as Van Vleck (1971, 6) himself put it, "in a sense . . . obsolete by the time it was off the press."[37] One might well wonder what would have happened, had Van Vleck continued to ponder the interaction between radiation and matter and the correspondence principle instead of fulfilling his duties as a newly minted member of the American physics community.

It is fair to say that Van Vleck—like Kramers, who added Heisenberg's name to (Kramers and Heisenberg, 1925) mainly as a courtesy (Dresden, 1987, 273–

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[35] See p. 24 of the transcript cited in note 7. Kuhn's recollection is that Van Vleck's earlier remark was made during a meeting in Philadelphia, February 17, 1961, to plan for the AHQP (Kuhn *et al.*, 1967, p. vii–viii).

[36] In the clause left out of the quotation in note 29 above, Van Vleck and Huber (1977, 939) explain that the former never got around to writing the last part of his trilogy, "because in 1925 the author was busy writing his book [Van Vleck, 1926a] and of course the advent of quantum mechanics presented innumerable research problems more timely than a purely classical investigation." As Clayton Gearhart has pointed out (private communication), Van Vleck also continued to worry about the problem of the specific heat of molecular hydrogen.

[37] For the reception of Van Vleck's *Bulletin*, see (Fellows, 1985, 88-89). Van Vleck's *Bulletin* and Pauli's *Handbuch* article were not the only treatises on the old quantum theory that were out of date before the ink was dry. Books on atomic mechanics by Born (1925) and Birtwistle (1926) suffered the same fate. George Birtwistle (1877–1929) was a Cambridge theorist who never made good on his promise as senior wrangler in the tripos, the famous Cambridge exam, in 1899. Two years after his book on the old quantum theory, he published a book on the new quantum theory (Birtwistle, 1928).

274)[38] —came as close to matrix mechanics as, say, H. A. Lorentz (1853–1928) and Henri Poincaré (1854–1912) came to special relativity. The comparison seems particularly apt, as the breakthroughs of both Einstein and Heisenberg consisted, to a large extent, in reinterpreting elements already present in the work of their predecessors and discarding some of the scaffolding in that work. Einstein reinterpreted Lorentz invariance and discarded the ether (Janssen, 2002). In the case of (Heisenberg, 1925c), the element of reinterpretation—*Umdeutung* in the original German—is emphasized in the title of the paper. Heisenberg's starting point was to reinterpret the frequency and amplitude of radiation accompanying transitions between stationary states in formulae for emission, absorption, and dispersion and to discard the electron orbits representing those states.[39] Heisenberg (1971, 63) himself famously compared his first paper on matrix mechanics to Einstein's first paper on special relativity. He argued that what his work had in common with Einstein's was its insistence on allowing only observable quantities into physical theory. As already indicated above, the analogy between the breakthroughs of Einstein and Heisenberg is considerably richer than that.

In the collective memory of the physics community, major discoveries understandably tend to get linked to singular events even though they are almost invariably stretched over time. The "discovery" of the electron by J. J. Thomson (1856–1940) in 1897 or the "discovery" of the quantum of action by Max Planck (1858–1947) in 1900 are well-known examples of this phenomenon. Special relativity is another good example of a "discovery" that came to be associated with a single flash of insight, Einstein's recognition of the relativity of simultaneity, and a single emblematic text, "On the electrodynamics of moving bodies" (Einstein, 1905). Much the same can be said about Heisenberg's famous trip to Helgoland in June 1925 to seek relief from his seasonal allergies and the *Umdeutung* paper resulting from his epiphany on this barren island. The way in which such stories become part of physics lore can be seen as a manifestation of what Robert K. Merton (1968) has dubbed the "Matthew effect," the disproportional accrual of credit to individuals perceived (retroactively in many cases) as leaders in the field.[40] We do, of course, recognize

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[38] See (Mehra and Rechenberg, 1982–2001, Vol. 2, 178–179) for Heisenberg's side of the story. These volumes by Mehra and Rechenberg bring together a wealth of information and we shall frequently refer to them. However, they need to be used with caution (Heilbron, 1985).

[39] For a penetrating and exceptionally lucid analysis of Heisenberg's *Umdeutung* paper, see (Landsman, 2005).

[40] The effect is named for the following passage from the Gospel According to St. Matthew: "For unto everyone that hath shall be given, and he shall have in abundance: but from him that hath not shall be taken away even that which he hath." Whereas Einstein was completely unknown in 1905, Heisenberg was already seen as a leader in the field by 1925. As Uhlenbeck put it: "Everything which Heisenberg did had to be taken seriously, because Heisenberg, Pauli, and, of course, Bohr were

the singular importance of the contributions of Einstein to special relativity and of Heisenberg to matrix mechanics. But there is no need to exaggerate the extent of their achievements. They may have been the first to enter the promised land, to use another admittedly strained biblical metaphor, but they would never have laid eyes on it without some Moses-figure(s) leading the way. It is in this sense that we feel that Van Vleck, in the passages quoted in the beginning of this section, was selling himself short.

In his biography of Kramers, Dresden (1987) makes a convincing case that Kramers deserves more credit for matrix mechanics than he received: "Kramers certainly hoped and probably expected to be the single author of the Kramers-Heisenberg paper. It is probably futile to speculate how the credit for the discovery of matrix mechanics would have been distributed in that case. There would be an indispensable preliminary paper by Kramers alone, followed by a seminal paper by Heisenberg; this might well have altered the balance of recognition" (Dresden, 1987, 252). Citing this passage, Dirk ter Haar (1998, 23), like Dresden one of Kramers' students, raises the question whether Kramers would have shared Heisenberg's 1932 Nobel prize in that case. In a curmudgeonly review of Dresden's book, however, Nico van Kampen, another one of Kramers' students, takes issue with the pattern of "near misses" that Dresden (1987, 446–461) sees in Kramers' career, the discovery of matrix mechanics being one of them (Dresden, 1987, 285–288). Van Kampen (1988) asks: "Is it necessary to explain that, once you have, with a lot of sweat and tears, constructed a dispersion formula on the basis of the correspondence principle, it is not possible to forget that background and that it takes a fresh mind to take the next step?" Similar claims can be made and similar questions can be raised in the case of Van Vleck, even though his work, unlike that of Kramers, did not directly influence Heisenberg (though it did, as we have seen, influence Born and Jordan).

Van Vleck's contribution has receded even further into the background in the history of quantum mechanics than Kramers'. Although the first (quantum) part is included in (Van der Waerden, 1968), (Van Vleck, 1924b,c) is not discussed in any of the currently standard secondary sources on quantum dispersion theory and matrix mechanics, such as (Jammer, 1966), (Dresden, 1987), or (Darrigol, 1992). Nor is it mentioned in Vol. 2 of (Mehra and Rechenberg, 1982–2001) on the discovery of matrix mechanics, although it is discussed briefly in Vol. 1 (pp. 646–647) on the old quantum theory.[41] That he worked in faraway Minnesota rather than in Copenhagen or Göttingen, we surmise, is a major factor in this neglect of Van Vleck. Whatever its reasons, the neglect is regrettable. Of the various papers in the general area of dispersion theory

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the gods" (p. 14 of the transcript of the third session of the AHQP interview with Uhlenbeck on April 5, 1962 [cf. note 3]).

[41] It is also mentioned in (Van der Waerden and Rechenberg, 1985, 330–331).

published in this period, including (Born, 1924) and (Kramers and Heisenberg, 1925), (Van Vleck, 1924b,c) stands out for its exceptionally clear exposition of the mathematics and for the breadth and cogency of its physical arguments. It is therefore much easier to see in (Van Vleck, 1924b,c) than in the later better-known paper by Kramers and Heisenberg that matrix mechanics did not come as a bolt out of the blue, but was the natural outgrowth of earlier applications of the correspondence principle to the interaction of radiation and matter.

Aitchison *et al.* (2004) have recently given a detailed reconstruction of the notoriously opaque mathematics of (Heisenberg, 1925c). By way of motivating their enterprise, they quote the confession of Steven Weinberg (1992, 67) that he has "never understood Heisenberg's *motivations* for the mathematical steps in his paper" (our emphasis). Aitchison *et al.* (2004) clearly explain the mathematical steps. The motivations for these steps, however, cannot be understood, we submit, without recourse to the dispersion theory leading up to his paper. And if we want to retrace Heisenberg's steps on his sojourn to Helgoland, Van Vleck may well be our best guide.

2 Dispersion theory as the bridge between the old quantum theory and matrix mechanics

From the point of view of modern quantum mechanics, the old quantum theory of Bohr and Sommerfeld—especially in the hands of the latter and members of his Munich school—was largely an elaborate attempt at damage control. In classical physics the state of a physical system is represented by a point in the phase space spanned by a system's generalized coordinates and momenta (q^i, p_i). All its properties are represented by functions $f(q^i, p_i)$ defined on this phase space. In quantum mechanics the state of a system is represented by a ray in the Hilbert space associated with the system; its properties are represented by operators acting in this Hilbert space, i.e., by rules for *transitions* from one ray to another. In the old quantum theory, one bent over backward to retain classical phase space. Quantum conditions formulated in various ways in (Sommerfeld, 1915a), (Wilson, 1915), (Ishiwara, 1915), (Schwarzschild, 1916), and (Epstein, 1916) only restricted the allowed orbits of points in phase space. These conditions restricted the value of so-called action integrals for every degree of freedom of some multiply-periodic system to integral numbers of Planck's constant h:

$$\oint p_i dq^i = n_i h, \tag{1}$$

where the integral is extended over one period of the generalized coordinate q^i (there is no summation over i).

Imposing such quantum conditions on classical phase space would not do in the end. As the picture of the interaction of matter and radiation in the old quantum theory already suggests, more drastic steps were required. In Bohr's theory the frequency $\nu_{i\to f}$ of the radiation emitted when an electron makes the transition from an initial state i to a final state f is given by the energy difference $E_i - E_f$ between the two states divided by Planck's constant h. Except in the limiting case of high quantum numbers, this radiation frequency differs sharply from the frequencies with which the electron traverses its quantized orbits in classical phase space before and after emission. This was widely recognized as the most radical aspect of the Bohr model. Erwin Schrödinger (1887–1961), for instance, opined in 1926 that this discrepancy between radiation frequency and orbital frequency[42]

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[42] Imre Lakatos (1970, 150–151) gives a lengthy quotation from an obituary of Planck by Born (1948), in which the same point is made more forcefully. The quotation even repeats some of the language of Schrödinger's letter: "That within the atom certain quantized orbits ... should play a special role, could well be granted; somewhat less easy to accept is the further assumption that the electrons moving on these curvilinear orbits ... radiate no energy. But that the sharply defined frequency

> . . . seems to me, (and has indeed seemed to me since 1914), to be something so *monstrous*, that I should like to characterize the excitation of light in this way as really almost *inconceivable.*[43]

One area of the old quantum theory in which this "monstrous" element became glaringly and unavoidably apparent was in the treatment of optical dispersion, the differential refraction of light of different colors. It was in this area that physicists most keenly felt the tension between orbital frequencies associated with individual states (the quantized electron orbits of the Bohr-Sommerfeld model) and radiation frequencies associated with *transitions* between such states. The key point of Heisenberg's *Umdeutung* paper was to formulate a new theory not in terms of properties of individual quantum states but in terms of transitions between states *without even attempting to specify the states themselves.* What, above all, prepared the ground for this move, as we shall demonstrate in detail below, was the development of a quantum theory of dispersion by Rudolf Ladenburg (1889–1953), Fritz Reiche (1883–1969), Bohr, Kramers, Van Vleck, Slater, and others.

By comparison, many of the other preoccupations of the old quantum theory, such as a detailed understanding of spectral lines, the Zeeman and Stark effects, and the extension of the Bohr-Sommerfeld model to multi-electron atoms (in particular, Helium) mostly added to the overall confusion and did little to stimulate the shift to the new mode of thinking exemplified by the *Umdeutung* paper.[44] The same is true—as we shall see, *pace* (Stuewer, 1975)—for the broad acceptance of Einstein's 1905 light-quantum hypothesis following the discovery of the Compton effect in 1923. What *was* crucial for the development of matrix mechanics were the A and B coefficients for emission and absorption of Einstein's quantum theory of radiation, *despite* its use of light quanta. Physicists working on dispersion theory were happy to use the A and B coefficients but they were just as happy to continue thinking of light as consisting of waves rather than particles.[45]

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of an emitted light quantum should be different from the frequency of the emitting electron would be regarded by a theoretician who had grown up in the classical school as *monstrous* and *almost inconceivable*" (our emphasis). Unfortunately, the passage quoted here by Lakatos is nowhere to be found in (Born, 1948)!

[43] Schrödinger to Lorentz, June 6, 1926 (Klein, 1967, 61).

[44] For detailed analyses of some of these bewildering developments, see, e.g., (Serwer, 1977; Forman, 1968, 1970).

[45] John Hendry (1981, 197) makes the provocative claim that "since Sommerfeld was the only known convert to the light-quantum concept as a result of the Compton effect whose opinions were of any real historical importance, this places Stuewer's thesis on the importance of the effect in some doubt." It is our impression that the Compton effect *did* convince many physicists of the reality of light quanta, just as (Stuewer, 1975) says it did, but we agree with Hendry (1981, 6) that this made surprisingly little difference for the further development of quantum physics.

Dispersion can boast of a venerable history in the annals of science, which reaches back at least to Descartes' rainbow and Newton's prism. The old quantum theory was certainly not the first theory for which dispersion presented serious difficulties. Neither Newtonian particle theories of light of the 18th century nor the wave theory of the early-19th century provided convincing accounts of dispersion (Cantor, 1983). In the wave theory of Thomas Young (1773–1829) and Augustin Jean Fresnel (1788–1827), the index of refraction of transparent matter is related to the density of the luminiferous ether, the medium thought to carry light waves, inside of it. To account for dispersion, i.e., for the dependence of the refractive index on the color of the refracted light, proponents of the wave theory had to assume that the ether density in transparent matter was different for different colors! Similarly, one had to assume that matter contained different amounts of ether for the ordinary and the extraordinary ray in double refraction. The problem likewise affected the optics of moving bodies (Janssen and Stachel, 2004; Stachel, 2005). To account for the absence of any signs of motion of the earth with respect to the ether, Fresnel (1818) introduced what is known as the "drag" coefficient. He assumed that transparent matter with index of refraction n carries along the ether inside of it with a fraction $f = 1 - 1/n^2$ of its velocity with respect to the ether. Although it was widely recognized in the 19th century that the drag coefficient was needed to account for the null results of numerous ether drift experiments, many physicists expressed strong reservations about the underlying physical mechanism proposed by Fresnel, since it implied that, because of dispersion, matter had to drag along a different amount of ether for every frequency of light!

One of the great triumphs of Lorentz's elaboration of the electromagnetic theory of light in the early 1890s was that he could derive the drag coefficient without having to assume an actual ether drag (Lorentz, 1892). In Lorentz's theory, the ether is immobile, the ether density is the same everywhere, inside and outside of matter, and the index of refraction is related, not to ether density, but to the polarization of harmonically-bound charges, later to be identified with electrons, inside transparent matter. The resonance frequencies of these oscillating charges correspond to the material's experimentally-known absorption lines. Lorentz's dispersion theory was further developed by Paul Drude (1863–1906). This classical Lorentz-Drude dispersion theory was remarkably successful in accounting for the experimental data.[46] Two centuries after Newton, there finally was a reasonably satisfactory theory of dispersion. Only two decades later, however, the model of matter underlying this theory

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[46] In 1896, Lorentz was also able to account for the Zeeman effect on the basis of this same picture of the interaction of matter and radiation (Kox, 1997).

was called into question again with the rise of the old quantum theory. The electrons oscillating inside atoms in the Lorentz-Drude model were replaced by electrons orbiting the nucleus in the Rutherford-Bohr model. As we shall see, the Lorentz-Drude theory nonetheless played a key role in the development of a quantum theory of dispersion in the early 1920s.

The basic model of dispersion in the Lorentz-Drude theory is very simple.[47] Suppose an electromagnetic wave of frequency ν (we are not concerned with how and where this wave originated) strikes a charged one-dimensional simple harmonic oscillator with characteristic frequency ν_0. We focus on the case of so-called *normal dispersion*, where the frequency ν of the electromagnetic wave is far from the resonance frequency ν_0 of the oscillator. The case where ν is close to ν_0 is called *anomalous dispersion*. We can picture the oscillator as a point particle with mass m and charge $-e$ (where e is the *absolute value* of the electron charge) on a spring with equilibrium position $x = 0$ and spring constant k, resulting in a restoring force $F = -kx$. The characteristic angular frequency $\omega_0 = 2\pi\nu_0$ is then given by $\sqrt{k/m}$. The electric field E of the incident electromagnetic wave[48] will induce an additional component of the motion at the imposed frequency ν. This component will be superimposed on any preexisting oscillations at the characteristic frequency ν_0 of the unperturbed system. It is this additional component of the particle motion, coherent with the incident wave (i.e., oscillating with frequency ν), that is responsible for the secondary radiation that gives rise to dispersion. The time-dependence of this component is given by:

$$\Delta x_{\text{coh}}(t) = A \cos \omega t, \tag{2}$$

where $\omega = 2\pi\nu$. To determine the amplitude A, we substitute eq. (2) into the equation of motion for the system. As long as we are far from resonance,

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[47] Classical dispersion theory is covered elegantly in ch. 31 of Vol. 1 of the Feynman lectures (see also ch. 32 of Vol. 2). Feynman makes it clear that this theory remains relevant in modern physics: “we will assume that the atoms are little oscillators, that is that the electrons are fastened elastically to the atoms ... You may think that this is a funny model of an atom if you have heard about electrons whirling around in orbits. But that is just an oversimplified picture. The correct picture of an atom, which is given by the theory of wave mechanics, says that, *so far as problems involving light are concerned*, the electrons behave as though they were held by springs” (Feynman *et al.*, 1964, Vol. 1, 31-4).

[48] We need not worry about the effects of the magnetic field B. The velocity of electrons in typical atoms is of order αc, where c is the velocity of light and $\alpha \simeq \frac{1}{137}$ is the fine-structure constant. The effects due to the magnetic field are thus a factor $\frac{1}{137}$ smaller than those due to the electric field and can be ignored in all situations considered in this paper.

radiation damping can be ignored and the equation of motion is simply:[49]

$$m\ddot{x} = -m\omega_0^2 x - eE\cos\omega t, \tag{3}$$

where dots indicate time derivatives and where we have made the innocuous simplifying assumption that the electric field of the incident wave is in the x-direction. Substituting $\Delta x_{\text{coh}}(t)$ in eq. (2) for $x(t)$ in eq. (3), we find:

$$-m\omega^2 A\cos\omega t = (-m\omega_0^2 A - eE)\cos\omega t. \tag{4}$$

It follows that

$$A = \frac{eE}{m(\omega^2 - \omega_0^2)}. \tag{5}$$

The central quantity in the Lorentz-Drude dispersion theory is the dipole moment $p(t) \equiv -e\Delta x_{\text{coh}}(t)$ of the oscillator induced by the electric field of the incident electromagnetic wave. From eqs. (2) and (5) it follows that:

$$p(t) = -e\Delta x_{\text{coh}}(t) = \frac{e^2 E}{4\pi^2 m(\nu_0^2 - \nu^2)}\cos 2\pi\nu t. \tag{6}$$

For groups of f_i oscillators of characteristic frequencies ν_i per unit volume, this formula for the dipole moment naturally generalizes to the following result for the polarization (i.e., the dipole moment per unit volume):

$$P(t) = \frac{e^2 E}{4\pi^2 m}\sum_i \frac{f_i}{\nu_i^2 - \nu^2}\ \cos 2\pi\nu t. \tag{7}$$

The polarization, in turn, determines the index of refraction (see, e.g., Feynman *et al.*, 1964, Vol. 1, 31-5).[50]

An early and influential attempt to bring dispersion theory under the umbrella of the old quantum theory was made by Sommerfeld (1915b, 1917) and

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[49] In sec. 3.3, we show how to take into account the effects of radiation damping.

[50] An indication that the Lorentz-Drude picture was not quite right was that it gave the wrong value for e^2/m. For dispersion in gases the number densities f_i in eq. (7) can be related to Avogadro's number on the assumption that dispersion only involves valence electrons. The value e^2/m extracted from dispersion measurements in gases on the basis of the Lorentz-Drude formula and this plausible assumption differed by as much as 25% from other determinations of e^2/m (Sommerfeld, 1917, 498). On balance, however, the treatment of dispersion was seen as an important success for Lorentz's classical theory.

by his former student Peter Debye (1884–1966) (Debye, 1915).[51] Clinton J. Davisson (1881–1958), then working at the Carnegie Institute of Technology in Pittsburgh, also made an important contribution (Davisson, 1916).[52] The Debye-Sommerfeld theory, as it came to be known, was based on the dubious assumption that the secondary radiation coming from small perturbations of a Bohr orbit induced by incident radiation could be calculated on the basis of ordinary classical electrodynamics, even though, by the basic tenets of the Bohr model, the classical theory did *not* apply to the original unperturbed orbit. In other words, it was assumed that while the large accelerations of the electron moving on a Bohr orbit would produce no radiation whatsoever, the comparatively small accelerations involved in the slight deviations from this orbit would.[53] Otherwise, the theory stayed close to the Lorentz-Drude theory, substituting small deviations in the motion of electrons from their Bohr orbits for small deviations from the vibrations of simple harmonic oscillators at their characteristic frequencies.

Both the Swedish physicist Carl Wilhelm Oseen (1879–1944) and Bohr severely criticized the way in which Debye and Sommerfeld modeled their quantum dispersion theory on the classical theory of Lorentz and Drude. Oseen (1915, 405) wrote:

> Bohr's atom model can in no way be reconciled with the fundamental assumptions of Lorentz's electron theory. We have to make our choice between

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[51] For other historical discussions of the development of quantum dispersion theory, see, e.g., (Darrigol, 1992, 224–230), (Dresden, 1987, 146–159, 215–222), (Jammer, 1966, 165 and sec. 4.3, especially 188–195), (Mehra and Rechenberg, 1982–2001, Vol. 1, sec. VI.1; Vol. 2, sec. III.5, 170–190), and (Whittaker, 1953, Vol. 1, 401, Vol. 2, 200–206). Van Vleck (1926a, sec. 49, 156–159) briefly discusses the early attempts to formulate a quantum theory of dispersion in his review article on the old quantum theory. We focus on the theory of Debye and Sommerfeld. Van Vleck also mentions theories by Charles Galton Darwin (1887–1962), Adolf Gustav Smekal (1895–1959), and Karl F. Herzfeld (1892–1978). All three of these theories make use of light quanta. In addition, strict energy conservation is given up in the theory of Darwin (1922, 1923), while in the theories of Smekal (1923) and Herzfeld (1924) orbits other than those picked out by the Bohr-Sommerfeld condition (1) are allowed, a feature known as "diffuse quantization." For other (near) contemporary reviews of dispersion theory, see (Pauli, 1926, 86–96), (Andrade, 1927, 669–682), and (Breit, 1932). Stolzenburg (1984, 17–18) briefly discusses Bohr's critical reaction to Darwin's dispersion theory.

[52] In 1927 at Bell Labs, Davisson and his assistant Lester H. Germer (1896–1971) would do their celebrated work on electron diffraction (Davisson and Germer, 1927), another great American contribution to (experimental) quantum physics for which the authors received the 1937 Nobel prize (Kevles, 1978, 188–189).

[53] Sommerfeld (1915b, 502) realized that this assumption was problematic and tried (unconvincingly) to justify it.

> these two theories … A theory based on Bohr's hypothesis as well as on Lorentz's electron theory must suffer from inconsistencies. Such a theory is Debye-Sommerfeld's theory of the dispersion of light.[54]

Bohr agreed. The central problem was that in Bohr's theory the link between radiation frequencies and orbital frequencies had been severed. As Bohr explained to Oseen in a letter of December 20, 1915, if the characteristic frequencies involved in dispersion

> … are determined by the laws for quantum emission, the dispersion cannot, whatever its explanation, be calculated from the motion of the electrons and the usual electrodynamics, which does not have the slightest connection with the frequencies considered (Bohr, 1972–1996, Vol. 2, 337).

Bohr elaborated on his criticism of the Debye-Sommerfeld theory in a lengthy paper intended for publication in *Philosophical Magazine* in 1916 but withdrawn after it was already typeset.[55] Bohr argued (we leave out the specifics of the experiments on dispersion in various gases that Bohr mentions in this passage):

> [E]xperiments … show that the dispersion … can be represented with a high degree of approximation by a simple Sellmeier formula[56] in which the characteristic frequencies coincide with the frequencies of the lines in the … spectra … [T]hese frequencies correspond with transitions between the normal states of the atom … On this view we must consequently assume that the dispersion … depends on the same mechanism as the transition between different stationary states, and that it cannot be calculated by application of ordinary electrodynamics from the configuration and motions of the electrons in these states (Bohr, 1972–1996, Vol. 2, 448–449).

In the next paragraph, Bohr added a prescient comment. Inverting the line of reasoning in the pasage above that dispersion should depend on the same mechanism as the transition between states, he suggested that the transition between states, about which the Bohr theory famously says nothing, should depend on the same mechanism as dispersion:

> [i]f the above view is correct … we must, on the other hand, assume that this mechanism [of transitions between states] shows a close analogy to an

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[54] Quoted and discussed in (Bohr, 1972–1996, Vol. 2, 337)

[55] The paper, dated January 1916 and entitled "On the application of the quantum theory to periodic systems," can be found in (Bohr, 1972–1996, Vol. 2, 433–461). For further discussion of Bohr's early views on dispersion, see (Heilbron and Kuhn, 1969, 281–283).

[56] This is a formula of the form of eq. (7) derived on the basis of an elastic-solid theory of the ether in (Sellmeier, 1872) (Jammer, 1966, 189).

> ordinary electrodynamic vibrator. This is indicated by the fact that the dispersion in many cases can be closely represented by a Sellmeier formula (ibid.).

As we shall see, in the quantum dispersion theory of the 1920s, the Lorentz-Drude oscillators were grafted onto the Bohr model. For the time being, however, it was unclear how to arrive at a satisfactory quantum theory of dispersion. The quasi-classical Debye-Sommerfeld theory led to a formula for the induced polarization of the form of eq. (7) but with resonance poles at frequencies corresponding to the Fourier decomposition of the mechanical motion of the original unperturbed Bohr orbit. As Oseen and Bohr pointed out, this was in blatant contradiction with the experimental data, which clearly indicated that the induced polarization should be a sum of resonance terms with poles at frequencies corresponding to absorption lines of the dispersive medium—in terms of the Bohr model, at frequencies corresponding to *differences* of energies of two distinct stationary states of the atom or molecule.

This criticism is repeated in more sophisticated form in a paper by Paul Sophus Epstein (1883–1966) with the subtitle "Critical comments on dispersion." This paper is the concluding installment of a trilogy on the application of classical perturbation theory to problems in the old quantum theory (Epstein, 1922a,b,c). Epstein, a Russian Jew who studied with Sommerfeld in Munich, was the first European quantum theorist to be lured to America. Millikan brought him to the California Institute of Technology in Pasadena in 1921.[57] To deal with the kind of multiply-periodic systems that represent hydrogenic atoms (i.e., atoms with only one valence electron) in the old quantum theory, Epstein customized techniques developed in celestial mechanics for computing the perturbations of the orbits of the inner planets due to the gravitational pull of the outer ones. The perihelion advance of Mercury due to such perturbations, for instance, is more than ten times the well-known 43″ per century

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[57] For further discussion of Epstein's position at Caltech, see (Seidel, 1978, 507–520). In his 1926 review article Van Vleck emphasizes the importance of the work of his colleague at Caltech and notes that it "is rather too often overlooked" (Van Vleck, 1926a, 164, note 268), to which one might add: "by European physicists." As we saw in sec. 1.4, Van Vleck felt the same way about his own contributions. Like Van Vleck, Epstein apparently complained about this lack of recognition to Born. This can be inferred from a letter from Born to Sommerfeld of January 5, 1923: "When you talk to Epstein in Pasadena and he complains about me, tell him that he should show you the very unfriendly letter he wrote to me because he felt that his right as first-born had been compromised by the paper on perturbation theory by Pauli and me [Born and Pauli, 1922, which appeared shortly after Epstein's trilogy]. Also tell him that I do not answer such letters but that I do not hold a grudge against him because of his impoliteness (to put it mildly) . . . In terms of perturbative quantization we are ahead of him anyway" (Sommerfeld, 2004, 137). We are grateful to Michael Eckert for drawing our attention to this passage.

due to the gravitational field of the sun as given by general relativity. Such calculations in classical mechanics are also the starting point of the later more successful approach to dispersion theory by Kramers and Van Vleck. Epstein clearly recognized that these calculations by themselves do not lead to a satisfactory theory of dispersion. In the introduction of his paper, Epstein (1922c, 92) explains that he discusses dispersion mainly because it nicely illustrates some of the techniques developed in the first two parts of his trilogy. He warns the reader that his approach is essentially the same as that of the Debye-Sommerfeld theory, and emphasizes that "this point of view leads to internal contradictions so strong that I consider the Debye-Davysson [sic] dispersion theory [as Epstein in Pasadena referred to it] to be untenable" (ibid.). The central problem is once again the discrepancy between radiation frequencies and orbital frequencies. As Epstein (1922c, 107–108) wrote in the conclusion of his paper:

> the positions of maximal dispersion and absorption [in the formula he derived] do not lie at the position of the emission lines of hydrogen but at the position of the mechanical frequencies of the model . . . *the conclusion seems unavoidable to us that the foundations of the Debye-Davysson* [sic] *theory are incorrect.*

Epstein (1922c, 110) recognized that a fundamentally new approach was required:

> We believe that to obtain formulae in good agreement [with the data] dispersion theory must be put on a whole new basis, in which one takes the Bohr frequency condition into account from the very beginning.

Quantum dispersion theory began to emerge from the impasse noted here by Epstein through work by Ladenburg and Reiche in Breslau,[58] which was already under way by the time Epstein wrote these lines. Ladenburg (1921)[59] was the first to use one of two key ingredients needed for a satisfactory treatment of dispersion: the emission and absorption coefficients of Einstein's 1916 quantum theory of radiation. The other critical ingredient, as we shall see below, was Bohr's correspondence principle.

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[58] Reiche came to Breslau in 1921. From 1918 to 1921 he had been at the *Kaiser Wilhelm Institut für Physikalische Chemie und Elektrochemie* in Berlin. In 1924, Ladenburg left Breslau to take up a position at the *Kaiser Wilhelm Institut.* Ladenburg emigrated to the United States in 1931, spending the rest of his career at Princeton University. Reiche only came to the United States in 1941, eventually becoming an adjunct professor at New York University (Mehra and Rechenberg, 1982–2001, Vol. 1, 633, 636). See (Bederson, 2005) for more on Reiche.

[59] Ladenburg (1921, 140, note) prominently acknowledged Reiche's help with this paper. They co-authored two follow-up papers (Ladenburg and Reiche, 1923, 1924).

As we shall show in detail in sec. 5.1, the dispersion formula that Ladenburg found for quantum systems in their normal or ground state is still valid in modern quantum mechanics, as is the more general formula for dispersion in quantum systems in arbitrary states first published by Kramers (1924a,b) in two notes in *Nature* and further clarified by Van Vleck (1924b,c) in his comprehensive study of applications of the correspondence principle to the interaction of matter and radiation. Ladenburg's formula is a special case of Kramers' formula. But not only does Ladenburg's formula lack the generality of the Kramers formula, Ladenburg's derivation of his formula also lacks the cogency of the derivations of the Kramers formula by Kramers or Van Vleck.

Ladenburg obtained his formula by equating results derived for what would seem to be two mutually exclusive models of matter, a classical and a quantum model. He calculated the rate of absorption of energy both for a collection of classical oscillators à la Lorentz and Drude, resonating at the absorption frequencies, and for a collection of atoms à la Bohr and Einstein with transitions between discrete energy levels corresponding to these same frequencies. Ladenburg set the two absorption rates equal to one another. In this way, he obtained a formula of the form of eq. (7) for the induced polarization of a group of N_r atomic systems in their ground state r with resonance poles at the right frequencies $\nu_{s\to r}$ corresponding to the allowed transitions from excited states s down to the ground state r:

$$P_r(t) = \frac{N_r c^3 E}{32\pi^4} \sum_s \frac{A_{s\to r}}{\nu_{s\to r}^2(\nu_{s\to r}^2 - \nu^2)} \cos 2\pi\nu t. \tag{8}$$

The factors $A_{s\to r}$ are Einstein's absorption coefficients for these transitions.[60] Ladenburg's formula was verified experimentally by Ladenburg and Reiche (1923).

Ladenburg's paper initially did not attract much attention. It is not mentioned in Epstein's trilogy the following year, but then Epstein was working in faraway California. More surprisingly, quantum physicists in Göttingen, Munich, and Copenhagen, it seems, also failed to take notice, even though Ladenburg was well-known to his Göttingen colleagues Born and James Franck (1882–1964). Bohr also knew Ladenburg personally: Ladenburg had attended Bohr's colloquium in Berlin in April 1920 and the two men had exchanged a few

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[60] The ratio e^2/m occurs in the expression for the absorption coefficient (as we shall show in detail in secs. 3.2 and 4.1). Ladenburg's work thus explained the anomalous values for e^2/m mentioned in note 50 above. The dependence of polarization on e^2/m is not quite as simple as that given by the Lorentz-Drude formula (7). The value of e^2/m, however, can be inferred from dispersion measurements through the values of the absorption coefficients, which in turn can be determined by measuring the lifetimes of excited states.

letters since (Bohr, 1972–1996, Vol. 4, 709–717).

Heisenberg later attributed the neglect of Ladenburg in Göttingen and Munich to the problem of connecting Ladenburg's work, which was closely tied to Einstein's radiation theory, with the dominant Bohr-Sommerfeld theory (Mehra and Rechenberg, 1982–2001, Vol. 2, 175–176). According to Heisenberg, it was only when Kramers (1924a,b) rederived Ladenburg's formula a few years later that its significance was widely appreciated. Ladenburg's own derivation had been unconvincing, at least to most physicists.[61] He offered no justification for equating the result for his classical model to that for his Einsteinian quantum model of matter. Commenting on Ladenburg's paper, van der Waerden (1968, 10) suggests that Ladenburg implicitly appealed to Bohr's correspondence principle: "By the principle of correspondence, this relation must hold in the limit for large quantum numbers. Hence, Ladenburg's guess is, that it be generally true." However, there is simply no mention of the correspondence principle anywhere in Ladenburg's paper. Given that Ladenburg only considered dispersion in atoms in their ground state and *not* in atoms in highly excited states where classical and quantum theory may be expected to merge in the sense of the correspondence principle, invoking the correspondence principle would have been unwarranted in any case. Still, if Heisenberg's later recollections are to be trusted, it might have helped the reception of Ladenburg's paper had he made some reference to the correspondence principle.

Unlike his colleagues in Göttingen and Munich, Bohr, in fact, did take notice of the paper early on. He was just slow expressing himself about it in print. As noted in (Hendry, 1981, 192), Bohr referred to (Ladenburg, 1921) in the very last sentence of "Application of the quantum theory to atomic problems in general" (Bohr, 1972–1996, Vol. 3, 397–414), a manuscript he did not date and never finished but which can reliably be dated to 1921 on the basis of several similarities to letters Bohr wrote to Darwin and Ehrenfest around the same time. In a paper submitted in November 1922, Bohr (1923b, 162) finally discussed Ladenburg's work in print. After repeating some of the observations about dispersion made in the passages of his unpublished 1916 paper quoted above, Bohr, in his usual verbose style, made some remarks that anticipate aspects of the 1924 Bohr-Kramers-Slater theory (see sec. 2.2):

> the phenomena of dispersion must thus be so conceived that the reaction of the atom on being subjected to radiation is closely connected with the

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[61] As Kuhn puts it in his interview with Slater for the AHQP (see note 10): "Of course, there was a good deal that appeared to most physicists as pretty totally *ad hoc* about the Reiche-Ladenburg work, and the whole question as to why it was the transition frequencies that occurred in the denominator rather than the orbital frequencies." Slater disagreed: "This seemed to me perfectly obvious . . . " (p. 41 of the first session of the AHQP interview with Slater).

> unknown mechanism which is answerable [the German has *verantwortlich*: responsible] for the emission of the radiation on the transition between stationary states. In order to take account of the observations, it must be assumed that this mechanism ... becomes active when the atom is illuminated in such a way that the total reaction of a number of atoms is the same as that of a number of harmonic oscillators in the classical theory,[62] the frequencies of which are equal to those of the radiation emitted by the atom in the possible processes of transition, and the relative number of which is determined by the probability of occurrence of such processes of transition under the influence of illumination. A train of thought of this kind was first followed out closely in a work by Ladenburg [1921] in which he has tried, in a very interesting and promising manner, to set up a direct connection between the quantities which are important for a quantitative description of the phenomena of dispersion according to the classical theory and the coefficients of probability appearing in the deduction of the law of temperature radiation by Einstein (Bohr, 1972–1996, Vol. 3, 496).

A letter from Bohr to Ladenburg of May 17, 1923 offers further insights into Bohr's developing views on the mechanism of radiation:

> to interpret the actual observations, it ... seems necessary to me that the quantum jumps are not the direct cause of the absorption of radiation, but that they represent an effect which accompanies the continuously dispersing (and absorbing) effect of the atom on the radiation, even though we cannot account in detail for the quantitative relation [between these two effects] with the usual concepts of physics (Bohr, 1972–1996, Vol. 5, 400).

At the beginning of this letter, Bohr mentioned the vagueness of some of his earlier pronouncements on the topic. After the passage just quoted he acknowledged "that these comments are not far behind the earlier ones in terms of vagueness. I do of course reckon with the possibility that I am on the wrong track but, if my view contains even a kernel of truth, then it lies in the nature of the matter that the demand for clarity in the current state of the theory is not easily met" (ibid.). Bohr need not have been so apologetic. His comments proved to be an inspiration to Ladenburg and Reiche. On June 14, 1923, Ladenburg wrote to Bohr:

> Over the last few months Reiche and I have often discussed [the absorption and scattering of radiation] following up on your comments in [Bohr 1923b] about reflection and dispersion phenomena and on my own considerations [Ladenburg 1921] which you were kind enough to mention there (Bohr, 1972–1996, Vol. 5, 400–401).

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[62] Note the similarity between Bohr's description here to Feynman's observation (quoted in note 47 above) that atoms behave like oscillators "so far as problems involving light are concerned."

In this same letter, Ladenburg announced his forthcoming paper with Reiche in a special issue of *Die Naturwissenschaften* to mark the tenth anniversary of Bohr's atomic theory. In the conclusion of their paper, Ladenburg and Reiche (1923, 597) wrote:[63]

> Surveying the whole area of scattering and dispersion discussed here, we have to admit that we do not know the true [*eigentlich*] mechanism through which an incident wave acts on the atoms and that we cannot describe the reaction of the atom in detail. This is no different by the way in the case of the true [*eigentlich*] quantum process, be it that an external wave ν_0 lifts electrons into higher quantum states, or be it that a wave ν_0 is sent out upon the return to lower states. We nevertheless believe on the grounds of the observed phenomena that the end result of a process in which a wave of frequency ν acts upon the atom should not be seen as fundamentally different from the effect that such a wave exerts on classical oscillators.

Ladenburg and Reiche (1923, 588, 590) introduced the term "*Ersatz*-oscillators" for such classical oscillators representing the atom as far as its interaction with radiation is concerned. They credited Bohr with the basic idea.[64] As we shall see in sec. 2.2, these *Ersatz*-oscillators became the "virtual oscillators" of the Bohr-Kramers-Slater theory.

Unlike Ladenburg in 1921, Ladenburg and Reiche prominently mentioned both Bohr's atomic theory and the correspondence principle in their 1923 paper. The authors' understanding and use of the correspondence principle, however, are still tied strongly to Einstein's quantum theory of radiation. Their "correspondence" arguments apply not to individual quantum systems, for which Bohr's correspondence principle was formulated, but to collections of such systems in thermal equilibrium with the ambient radiation. The authors also do not limit their "correspondence" arguments to the regime of high quantum numbers (Ladenburg and Reiche, 1923, especially secs. 4–5, 586–589). These problems invalidate many of the results purportedly derived from the correspondence principle in their paper. Drawing on earlier work by Planck, they derived a result for emission consistent with the correspondence principle (i.e., merging with the classical result in the limit of high quantum numbers), but their attempts to derive similar results for absorption and dispersion were unconvincing. In fact, it may very well be that these dubious attempts were the direct inspiration for Van Vleck to formulate correspondence principles for emission, absorption, and dispersion (see sec. 3.3 for further discussion).

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[63] Quoted and discussed in (Hendry, 1981, 192).

[64] See also (Ladenburg and Reiche, 1924, 672). Van Vleck (1926a, 159, note 260) reports that Lorentz made a similar suggestion at the third Solvay congress in 1921 (Verschaffelt *et al.*, 1923, 24), but does not mention Ladenburg and Reiche in this context, attributing the idea to (Slater, 1924) instead. (Herzfeld, 1924, 350) mentions *Ersatz*-oscillators and does cite (Ladenburg and Reiche, 1923).

Given Bohr's strong interest in dispersion theory, it is not surprising that it was his first lieutenant Kramers who took the next big step after Ladenburg and Reiche.[65] Ladenburg's formula, as we mentioned above, only holds for systems in the ground state. The correspondence principle only applies to highly excited states with large quantum numbers. Kramers (1924a,b)[66] and Van Vleck (1924b,c) showed that the correspondence principle requires a formula with *two* terms,

$$P_r(t) = \frac{N_r c^3 E}{32\pi^4}\left(\sum_{s>r}\frac{A_{s\to r}}{\nu^2_{s\to r}(\nu^2_{s\to r}-\nu^2)} - \sum_{t<r}\frac{A_{r\to t}}{\nu^2_{r\to t}(\nu^2_{r\to t}-\nu^2)}\right)\cos 2\pi\nu t, \quad (9)$$

where r refers to a highly excited state to which the correspondence principle can be applied, and s and t to states above and below r, respectively (see secs. 3.2 and 4.2 for details). The first term is Ladenburg's formula, which represents the full answer if we assume that the result (9) can be extended to the low-quantum-number regime unaltered, and if we give r the value for the ground state, with no further states t below it. Van Vleck (1924b) clarified the physical interpretation of the second term by comparing this formula for dispersion to a similar formula he derived for absorption (see sec. 3.3).

The construction of the dispersion formula (9) by Kramers—and by Van Vleck—required, as a prelude to the application of the correspondence principle, a derivation of the classical formula for the dipole moment of an arbitrary (non-degenerate) multiply-periodic system. This is where Ladenburg and Reiche came up short. Kramers and Van Vleck, like Epstein before them, used canonical perturbation techniques from celestial mechanics to derive this formula (see secs. 3.1 and 4.2 for details). Unlike Epstein, however, they were able to make dramatic progress by fully exploiting the power of both Einstein's radiation theory and Bohr's correspondence principle. They considered transitions between neighboring states with high quantum numbers, replaced derivatives with respect to the action occurring in the classical dispersion formula by difference quotients,[67] amplitudes by Einstein coefficients $A_{i\to f}$, and orbital frequencies by transition frequencies $\nu_{i\to f}$ (see secs. 3.2 and 4.2 for details). Explicitly appealing to the kind of logic mistakenly attributed to Ladenburg by Van der Waerden, Kramers and Van Vleck took the leap of faith that their difference formula (9)[68] merging with the classical formula for

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[65] Hendry (1984, 46) goes as far as calling Kramers' theory "the Bohr-Kramers dispersion theory."

[66] In addition to the literature cited in note 51, see (Ter Haar, 1998, 23–30) for discussion of Kramers' work on dispersion theory.

[67] This replacement is known as "Born's correspondence rule." It was in fact found independently of Born both by Kramers and by Van Vleck. We shall return to this point in sec. 3.2.

[68] This formula is not uniquely determined and it took some inspired guesswork

high quantum numbers would hold all the way down to states with low quantum numbers including the ground state. It would take several years before the terms for atoms in excited states in the general dispersion formula were verified experimentally—and then only in an indirect manner—by the Indian physicist Chandrasekhara Venkata Raman (1888–1970) and by the Russians Grigoriaei Samjuilovich Landsberg (1890–1957) and Leonid Isaakovich Mandelstam (1879–1944) (Raman, 1928; Landsberg and Mandelstam, 1928).[69]

The quantum dispersion theory of Kramers and Van Vleck was a crucial step in the transition from the old quantum theory to matrix mechanics, and thereby in the transition from classical phase spaces to Hilbert spaces. As was noted by Kramers (1924b, 311) in his second note to *Nature*—written in response to a letter by Minnesota's Gregory Breit (1924b)—the dispersion formula (9)

> only contains such quantities as allow of a direct physical interpretation on the basis of the fundamental postulates of the quantum theory ... and exhibits no further reminiscence of the mathematical theory of multiple [sic] periodic systems.

This point is amplified in (Kramers and Heisenberg, 1925, 234):

> we shall obtain, quite naturally, formulae which contain only the frequencies and amplitudes *which are characteristic for the transitions*, while all those symbols which refer to the mathematical theory of periodic systems will have disappeared (our emphasis).

Orbits do not correspond to observable quantities, but transitions do, namely to the frequency $\nu_{i\to f}$ of the emitted radiation, and, through the Einstein coefficients $A_{i\to f}$, to its intensity. In the introduction of his *Umdeutung* paper, Heisenberg (1925c, 262) explained that he wanted "to establish a theoretical quantum mechanics, analogous to classical mechanics, but in which only relationships between observable quantities occur." In the next sentence he identified the Kramers dispersion theory as one of "the most important first steps toward such a quantum-theoretical mechanics" (ibid.). Rather than using classical mechanics to analyze features of electron orbits and translating the end result into quantities associated with transitions between orbits on the basis of the correspondence principle, as Kramers and Van Vleck had done, Heisenberg set out to formulate a new theory directly in terms of transitions

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guided by the correspondence principle to arrive at the one that would prove to be correct in modern quantum theory.

[69] See (Mehra and Rechenberg, 1982–2001, Vol. 2, 188). See (Singh, 2002) for the history of the work leading up to Raman's discovery of the effect named after him. Ladenburg (1926, 1213) emphasized the desirability of experimentally demonstrating the presence of the terms for atoms in excited states in the Kramers dispersion formula.

between states without bothering to find a representation for the states themselves.

With the elimination of orbits, the Bohr-Sommerfeld quantization condition (1) formulated in terms of orbits in phase space can no longer be used. Using the general recipe introduced in the *Umdeutung* paper for translating classical formulae into quantum-mechanical ones, Heisenberg (1925c, 268) converted (a derivative of) the Bohr-Sommerfeld condition into an equation that contains only amplitudes and frequencies.[70] As Heisenberg points out, this equation had been found earlier in dispersion theory. It is the so-called Thomas-Kuhn sum rule (Thomas, 1925; Kuhn, 1925). A detailed derivation of Thomas' result was given by Reiche and Thomas (1925), which is why the sum rule is also known as the Thomas-Kuhn-Reiche sum rule.[71]

Van Vleck had, in fact, been the first to find the sum rule (Sopka, 1988, 135, note 184). As he wrote in his NRC *Bulletin*:

> Eq. (62a) [a version of the sum rule] appears to have been first incidentally suggested by the writer [Van Vleck 1924c, 359–360, footnote 43] and then was later and independently much more strongly advanced by Thomas ... Kuhn ... and Reiche and Thomas (Van Vleck, 1926a, 152).

Van Vleck is referring to a footnote in the section on dispersion in the classical part of his paper. In this footnote he mentioned two objections that explain why he did not put greater emphasis on the sum rule himself. Van Vleck's idea—which he calls "tempting (but probably futile)" (Van Vleck, 1924c, 359, footnote 43)—was that the sum rule would allow him to compute the Einstein A coefficients. He was under the impression, however, that "such a method

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[70] Since Heisenberg's theory only deals with transitions between states, the absolute value of the action J is irrelevant. Only the difference in J-value between two states matters. Hence the derivative.

[71] Interestingly, Reiche and Thomas use the notion of *Umdeutung* in describing their goal in their paper which was *submitted* to the *Zeitschrift für Physik* in early August 1925 about a month before (Heisenberg, 1925c) *appeared* in the same journal: "We use ... the correspondence principle in the same way in which it was applied by Kramers in the derivation of the dispersion formula by reinterpreting (*umdeuten*) the mechanical orbital frequencies as radiation frequencies, the Fourier coefficients as the "characteristic amplitudes" that determine the quantum radiation, and, finally, in analogy to the Bohr frequency condition, differential quotients as difference quotients. In the realm of high quantum numbers the classical and quantum-theoretical representations become identical. We try to arrive at a general relation, by maintaining the reinterpretation (*Umdeutung*) of classical quantities into quantum-theoretical ones for all quantum numbers" (Reiche and Thomas, 1925, 511–512). We also note that Reiche and Thomas (1925, 513) cite (Van Vleck, 1924b, 359).

is hard to reconcile with the [experimental] work of F. C. Hoyt [1923, 1924]" on X-ray absorption and that it "would lead to transitions from positive to negative quantum numbers, which can scarcely correspond to any physical reality" (ibid.).

As Heisenberg (1925c, 269–270) shows briefly in his paper, the sum rule follows from the Kramers dispersion formula (9) if one takes the limit in which the frequency ν of the incident radiation is much greater than any of the absorption frequencies $\nu_{i \to j}$. That the quantization condition obtained by massaging the Bohr-Sommerfeld condition can be derived from the Kramers dispersion theory, widely recognized as one of the most secure parts of the old quantum theory, clearly bolstered Heisenberg's confidence in the translation procedure of his *Umdeutung* paper. So, in addition to inspiring the general approach of the *Umdeutung* paper, the Kramers dispersion theory furnished Heisenberg's new theory with its fundamental quantization condition. It was left to Born and Jordan (1925b) to replace the Thomas-Kuhn sum rule with the now standard commutation relations for position and momentum. These commutation relations can be extracted from the sum rule, as we shall show in detail in sec. 5.1. That Heisenberg stopped short of making this move, we conjecture, is because it requires a way of looking at the sum rule that runs counter to its interpretation in dispersion theory.

The positivism that gets pride of place in the introduction of Heisenberg's *Umdeutung* paper probably came from a variety sources. Pauli, Heisenberg's fellow student and frequent discussion partner (both in person and in writing), was a devoted follower of his godfather Ernst Mach (1838–1916).[72] As Pauli had written to Bohr, for instance, on December 12, 1924:

> We must not . . . put the atoms in the shackles of our prejudices (of which in my opinion the assumption of the existence of electron orbits in the sense of the ordinary kinematics is an example); on the contrary, we must adapt our concepts to experience (Bohr, 1972–1996, Vol. 5, 35–36).

We already mentioned in sec. 1.4 that Heisenberg himself later claimed that his positivist attitude came in part from his reading of Einstein's 1905 special relativity paper.[73] The most important source, however, was undoubtedly Kramers' dispersion theory, on which, after all, he and Kramers had just published a joint paper (Kramers and Heisenberg, 1925). In this context positivism was not a blanket injunction against unobservable quantities in general but had a very specific set of increasingly problematic unobservables as its target,

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[72] On Pauli's positivism, see, e.g., (Hendry, 1984, 19–23) and (Gustavson, 2004).

[73] See, e.g., (Holton, 2005, 26–31) for discussion. Cassidy (1991, 198) makes the suggestive observation that Born and Jordan (1925a, 493), in a paper completed by June 11, 1925, not only emphasized the observability principle but also appealed to Einstein's analysis of distant simultaneity in support of it.

viz. the electron orbits of the Bohr-Sommerfeld theory.

As Helge Kragh (1999, 162) notes: “there was no royal road from the observability principle to quantum mechanics.” This truism is nicely illustrated by a conversation between Einstein and Heisenberg reported years later by the latter. The following exchange supposedly took place in Berlin in the spring of 1926:

> “But you don’t seriously believe,” Einstein protested, “that none but observable magnitudes must go into a physical theory?” “Isn’t that precisely what you have done with relativity?” I asked in some surprise . . . “Possibly I did use this kind of reasoning,” Einstein admitted, “but it is nonsense all the same” (Heisenberg, 1971, 63).[74]

As Einstein complained in 1917 in a letter to his friend Michele Besso (1873–1955), referring to the excessive Machian positivism of their mutual acquaintance Friedrich Adler (1879–1960): “He is riding the Machian nag [*den Machschen Klepper*] to exhaustion.” In a follow-up letter he elaborated: “It cannot give birth to anything living, it can only stamp out harmful vermin.”[75] This is very much true in the case of matrix mechanics as well. Heisenberg’s positivist attitude, whatever its sources, would have been perfectly sterile had it not been for Kramers showing that the problematic Bohr orbits were fully dispensable in dispersion theory.

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[74] Quoted and discussed, for instance, in (MacKinnon, 1977, 185) and in (Holton, 2005, 30–31). For another version of the same anecdote, see (Heisenberg, 1983, 113–114). In the 1940s, Heisenberg (1943a,b, 1944) once again tried to force a theoretical breakthrough, this time with his S-matrix theory, by restricting himself to observable quantities, with the qualification that he had taken to heart Einstein’s lesson that, in the end, it is the theory that determines what the observables are (Heisenberg [1971, 63] has Einstein make this point a few sentences after the passage quoted above and acknowledged it as a source of inspiration for his uncertainty principle). Nearly two decades after the *Umdeutung* paper, Heisenberg (1943a, 514) wrote: “in this situation it seems useful to raise the question which concepts of the present theory can be retained in the future theory, and this question is roughly equivalent to a different question, namely which quantities of the current theory are “observable” . . . Of course, it will always only be decided by the completed theory which quantities are truly “observable”.” See (Cushing, 1990) and (Dresden, 1987, 453–458) for discussion of Heisenberg’s S-matrix program.

[75] Einstein to Besso, April 29 and May 13, 1917, respectively (Einstein, 1987–2004, Vol. 8, Docs. 331 and 339). For further discussion, see, e.g., (Holton, 1968).

2.2 Dispersion theory, virtual oscillators, and the Bohr-Kramers-Slater (BKS) theory

The *Ersatz*-oscillators of (Ladenburg and Reiche, 1923) became the "virtual oscillators" of (Bohr, Kramers, and Slater, 1924), submitted to *Philosophical Magazine* on January 21, 1924.[76] As we saw in the preceding section, the idea of virtual oscillators was essentially Bohr's, although it is often attributed to Slater, both by contemporaries (Van Vleck, 1924b, 330) and by later historians (see, e.g., Stuewer, 1975, 291, 303). Bohr was probably also responsible for the fanciful new name. Despite their close association with the BKS theory, the main use of virtual oscillators continued to be in dispersion theory. They formed the basis for the physical interpretation of the Kramers dispersion formula (9) in the year and a half before Heisenberg's *Umdeutung* paper.

In the abstract of (Van Vleck, 1924b, 330) it is noted that the Kramers dispersion formula "assumes the dispersion to be due not to the actual orbits but to Slater's 'virtual' or 'ghost' oscillators having the spectroscopic rather than orbital frequencies."[77] In the last section of this part of his paper, Van Vleck cautioned:

> The introduction of these virtual resonators is, to be sure, in some ways very artificial, but is nevertheless apparently the most satisfactory way of combining the elements of truth in both the classical and quantum theories. In particular this avoids the otherwise almost insuperable difficulty that it is the spectroscopic rather than the orbital frequencies ... which figure in dispersion (Van Vleck, 1924b, 344).

In his second *Nature* note, Kramers (1924b, 311) made a similar cautionary remark about the use of virtual oscillators:[78]

> In this connexion it may be emphasized that the notation 'virtual oscillator' used in my former letter [Kramers, 1924a] does not mean the introduction of any additional hypothetical mechanism, but is meant only as a terminology suitable to characterise certain main features of the connexion between

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[76] See (Dresden, 1987, 221) for a helpful chronology of events in 1923–1925 pertaining to BKS and dispersion theory. For discussions of the BKS theory, see, e.g., (Klein, 1970, 23–39), (Stuewer, 1975, 291–305), (Mehra and Rechenberg, 1982–2001, Vol. 1, sec. V.2), (Hendry, 1981), (Dresden, 1987, 159–215), the essay by Klaus Stolzenburg (1984) in (Bohr, 1972–1996, Vol. 5, 3–96) and the dissertation of Neil Wasserman (1981).

[77] See also (Van Vleck, 1926a, 163).

[78] In the work that led to (Kramers and Heisenberg, 1925), however, Kramers, according to Hendry (1981, 202), "ignored their virtual nature altogether and treated the oscillator model as naively as he had the orbital model."

> the description of optical phenomena and the theoretical interpretation of spectra.

Despite such disclaimers, Kramers and Van Vleck—as well as Slater, Born, Breit and others working in the general area of dispersion theory in 1924–1925—used a model of the atom in which the electron orbits of the Bohr-Sommerfeld theory were supplemented by an "orchestra of virtual oscillators"[79] with characteristic frequencies corresponding to each and every transition that an electron in a given orbit can undergo. The dual representation of the atom as simultaneously a quantum system à la Einstein and Bohr and a set of oscillators à la Lorentz and Drude, implicit in (Ladenburg, 1921) and made explicit, under Bohr's influence, in (Ladenburg and Reiche, 1923), thus became widely accepted. Thanks to virtual oscillators—to paraphrase Heisenberg's succinct statement to van der Waerden (1968, 29) in 1963—at least *something* in the atom was vibrating with the right frequency again.

What was meant by the word "virtual" in this context is not exactly clear. As the BKS authors were putting the final touches on the German version of their paper, Bohr was anxious to ensure that Pauli approved of "the words "communicate" and "virtual", for after lengthy consideration, we have agreed here on these basic pillars of the exposition."[80] In this letter, Bohr first announced that the manuscript would be submitted that same day to *Zeitschrift für Physik* and that he would enclose a copy, then added a postscript saying that there had been further delays and that he would send it later. Amused, Pauli wrote back a few days later:

> I laughed a little (you will certainly forgive me for that) about your warm recommendation of the words "communicate" and "virtual" and about your postscript that the manuscript is still not yet completed. On the basis of my knowledge of these two words (which I definitely promise you not to undermine), I have tried to guess what your paper may deal with. But I have not succeeded. In any case, it will interest me very much to read it and if I can help a little with the language, I should gladly do so.[81]

The term "virtual" also puzzled the group of physicists in Ann Arbor studying the BKS paper with Bohr's former associate Klein, who wrote to Bohr on June 30, 1924: "Colby [cf. note 21], who is also most interested in it, asked me about the meaning of the term 'virtual radiation'" (Stolzenburg, 1984, 29). Whatever its exact meaning, the designation 'virtual' does serve as a warning that these oscillators do not behave classically. They are, as Darrigol (1992, 257) aptly

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[79] The term "virtual orchestra" comes from (Landé, 1926, 456) (Jammer, 1966, 187).

[80] Bohr to Pauli, February 16, 1924 (Bohr, 1972–1996, Vol. 5, 409).

[81] Pauli to Bohr, February 21, 1924 (Bohr, 1972–1996, Vol. 5, 412). Our reading of this letter differs from that of Hendry (1981, 202), who characterized it as "mocking."

put it, "nothing but a condensed expression of their effects, which could be deduced from the correspondence principle piece by piece but could not be synthesized in any classical model."

The BKS theory grew around an idea that Slater hit upon shortly before he left for Europe in late 1923.[82] Slater suggested that the wave and particle aspects of the behavior of light might be reconciled by having an electromagnetic field guide corpuscular light quanta.[83] When Slater arrived in Copenhagen around Christmas 1923, his idea was cannibalized by Bohr and Kramers, who stripped it of all reference to light quanta. Against his better judgment—as he wrote decades later in a letter of November 4, 1964 to van der Waerden (1968, 13)—Slater went along and his idea entered the literature via the BKS paper. In a short letter that Slater sent to *Nature* a week after this joint paper had been submitted, he explained how Bohr and Kramers had convinced him of their point of view and proceeded to explain his idea couched in BKS terms:

> Any atom may, in fact, be supposed to communicate with other atoms all the time it is in a stationary state, by means of a virtual field of radiation originating from oscillators having the frequencies of possible quantum transitions and the function of which is to provide for the statistical conservation of energy and momentum by determining the probabilities for quantum transitions (Slater, 1924, 307).

The final clause about the statistical conservation of energy and momentum was foisted upon Slater by Bohr and Kramers.[84] Bohr had been contemplating such a move for several years, as can be seen, for instance, from correspondence with Ehrenfest in 1921 in connection with the third Solvay congress held that year (Klein, 1970, 19) and with Darwin in 1922 (Stolzenburg, 1984, 13–19). Slater's concept of virtual radiation emitted while an atom is in a stationary state fit nicely with a number of ideas Bohr had been mulling over for some time, especially with the tentative ideas developed in the context of dispersion theory about the mechanism of emission and absorption of radiation by atoms (see sec. 2.1).

Even though Slater had been opposed to giving up strict conservation of energy and momentum, the junior co-author of BKS ended up defending the theory with the zeal of the converted. In a lengthy paper signed December 1, 1924, and published in the April 1925 issue of *The Physical Review*, he tried to work

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[82] See Slater to his mother, November 8, 1923 (quoted in Dresden, 1987, 161); Slater to Kramers, December 8, 1923 (AHQP). For discussions of Slater's idea, see (Klein, 1970, 23), (Stuewer, 1975, 291–294), (Hendry, 1981, 213–214), (Stolzenburg, 1984, 6–11), and (Darrigol, 1992, 218–219).

[83] Slater was probably unaware that Einstein and Louis de Broglie (1892–1987) had already made similar suggestions (Hendry, 1981, 199; Darrigol, 1992, 218).

[84] See also (Bohr, Kramers, and Slater, 1924, 160).

out a "consistent detailed theory of optical phenomena" based on the BKS theory (Slater, 1925a, 395).[85] In the introduction of the paper, he presents the dilemma that led him to embrace Bohr's statistical conservation laws.[86] The problem, he argues, is that

> in the quantum theory the energy of atoms must change by jumps; and in the electromagnetic theory the energy of a radiation field must change continuously ... Two paths of escape from this difficulty have been followed with more or less success. The first is to redefine energy [i.e., to adopt Einstein's light-quantum hypothesis]; the second to discard conservation. Optical theory on [the first interpretation] would be a set of laws telling in what paths the quanta travel ... [One way to do this is] to set up a sort of ghost field, similar to the classical field, whose function was in some way to guide the quanta. For example, the quanta might travel in the direction of Poynting's vector in such a field. The author was at one time of the opinion that this method was the most hopeful one for solving the problem ... The other direction of escape from the conflict between quantum theory and wave theory has been to retain intact the quantum theory and as much of the wave theory as relates to the field, but to discard conservation of energy in the interaction between them (Slater, 1925a, 396–397).

Slater then sketches some difficulties facing this second approach, but makes it clear that this is the approach he now favors:

> An attempt was made by the writer, in a note to Nature [Slater, 1924], enlarged upon in collaboration with Bohr and Kramers [BKS], to contribute slightly to the solution of these difficulties. In the present paper, the suggestions made in those papers are developed into a more specific theory (ibid., 398).

Slater then proceeds to describe more carefully how to picture the interaction between matter and radiation in the BKS theory and makes it clear that the proposed mechanism is incompatible with strict energy conservation. According to Slater, the "one ... essentially new" suggestion of BKS (note that he

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[85] Slater presented this work at a meeting of the *American Physical Society* in Washington, D.C., in December 1924 (Slater, 1925b). At this same meeting, Van Vleck (1925) talked about (Van Vleck, 1924b,c) and Breit (1925) talked about (Breit, 1924a). The AHQP contains some correspondence between Slater and Van Vleck regarding this meeting and regarding (Slater, 1925a) (Slater to Van Vleck, December 8, 1924; Van Vleck to Slater, December 15, 1924). Slater sent a copy of his paper to Bohr in December 1924 and defended his elaboration of the BKS theory in a letter to Bohr of January 6, 1925 (Bohr, 1972–1996, Vol. 5, 65–66).

[86] See also the brief discussion of the BKS theory in (Van Vleck, 1926a, 285–286), the *Bulletin* for the NRC, written after the decisive experimental refutation of the theory.

does not claim credit for the concept of virtual oscillators) was:

> that the wavelets sent out by an atom in connection with a given transition were sent out, not as a consequence of the occurrence of the transition, but as a consequence of the existence of the atom in the stationary state from which it could make that transition.[87] On this assumption, the stationary state is the time during which the atom is radiating or absorbing; the transition from one state to another is not accompanied by radiation, but so far as the field is concerned, merely marks the end of the radiation or absorption characteristic of one state, and the beginning of that characteristic of another. The radiation emitted or absorbed during the stationary state is further not merely of the particular frequency connected with the transition which the atom is going to make; it includes all the frequencies connected with all the transitions which the atom could make ... Although the atom is radiating or absorbing during the stationary states, its own energy does not vary, but changes only discontinuously at transitions ... It is quite obvious that the mechanism becomes possible only by discarding conservation (ibid., 397–398).

On the next page, Slater inserts a disclaimer similar to the ones by Van Vleck and Kramers quoted above:

> It must be admitted that a theory of the kind suggested has unattractive features; there is an apparent duplication between the atoms on the one hand, and the mechanism of oscillators producing the field on the other. But this duplication seems to be indicated by the experimental facts, and it is difficult at the present stage to see how it is to be avoided (ibid., 399).

At least for the time being, Slater had turned his back on Einstein's light quanta and was completely sold on the highly problematic BKS theory.[88]

BKS has often been portrayed as Bohr's last stand against light quanta after the Compton effect had finally convinced most other physicists that they were unavoidable (Klein, 1970, 3). Upon his return from a trip to Canada and the USA in late 1923, Bohr wrote ruefully to his old Manchester mentor Rutherford:

> In Chicago ... I met Michelson who I believe found in me a more conservative scientist than he had expected, at any rate decidedly more so than the

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[87] Note the similarity with the comments of Bohr to Ladenburg quoted in the preceding section: "the quantum jumps are not the direct cause of the absorption of radiation, but ... represent an effect which accompanies the continuously disperging (and absorbing) effect of the atom on the radiation" (Bohr, 1972–1996, Vol. 5, 400).

[88] For detailed criticism of the physics of the BKS theory, see (Dresden, 1987, 178–185).

> younger school of American physicists who, as the Comptons [Arthur H. and Karl T. (1887–1954)], notwithstanding their great achievements, take views simply horrifying for a man who spend [sic] his life in studying the most refined interference phenomena and for whom the wave theory is a creed (Bohr, 1972–1996, Vol. 5, 6).

In the BKS theory the Compton effect is explained without light quanta. The frequency shift between incoming and scattered X-rays is attributed to a Doppler shift in the X-ray wave fronts instead. Compton (1923), it seems, considered this possibility, but did not pursue it because, as he showed in the paper in which he introduced the effect now named after him, the recoil velocity needed to get the Doppler shift to come out right is different from the recoil velocity needed to ensure the conservation of energy and momentum in the process. Compton naturally assumed that these two velocities had to be the same since in both cases it is the electron that is doing the recoiling. The BKS theory, however, has room for two different recoil velocities, one for the electron, one for the orchestra of virtual oscillators associated with it.[89] The Compton effect can be interpreted as a Doppler shift if the appropriate recoil velocity is assigned to the virtual oscillators. Energy and momentum are conserved, at least statistically, averaged over many electrons involved in Compton scattering, if the appropriate recoil velocity is assigned to the electrons themselves. Bohr and his co-authors wasted few words on the justification of this startling maneuver:

> That in this case the virtual oscillator moves with a velocity different from that of the illuminated electrons themselves is certainly a feature strikingly unfamiliar to the classical conceptions. In view of the fundamental departures from the classical space-time description, involved in the very idea of virtual oscillators, it seems at the present state of science hardly justifiable to reject a formal interpretation as that under consideration as inadequate (Bohr, Kramers, and Slater, 1924, 173).

This is almost as bad as pieces of glass dragging along different amounts of ether for different colors of light in early-19th-century ether theory!

The BKS theory was decisively refuted in experiments by Walther Bothe (1891–1957) and Hans Geiger (1882–1945) in Berlin and by Compton and Alfred Walter Simon in Chicago. These experiments showed that energy-momentum is strictly conserved in Compton scattering (i.e., event by event) and not just statistically (Stuewer, 1975, 299–302; Stolzenburg, 1984, 75–80).

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[89] What Compton (1923, 487) actually said in his paper is very suggestive of this option: “It is clear . . . that so far as the effect on the wave-length is concerned, we may replace the recoiling electron by a scattering electron” with an “effective velocity” different from that of the recoiling electron (quoted and discussed in Stuewer, 1975, 230).

The experiments were begun shortly after the BKS paper was published (see Bothe and Geiger, 1924), but the final verdict did not come in until the following year. Bothe and Geiger (1925a,b) published their results in April 1925. The paper by Compton and Simon (1925) is signed June 23, 1925, and appeared in September 1925.[90] On April 17, 1925, Geiger sent Bohr a letter forewarning him of the results of his experiments with Bothe. When Geiger's letter arrived in Copenhagen four days later, Bohr was in the process of writing to Ralph H. Fowler (1889–1944). In a postscript to his letter, Bohr wrote: "there is nothing else to do than to give our revolutionary efforts as honourable a funeral as possible" (Stuewer, 1975, 301).

When Slater found out about the experimental refutation of the BKS theory, he dashed off another letter to *Nature* (dated July 25, 1925), announcing that he had changed his mind once more: "The simplest solution to the radiation problem then seems to be to return to the view of a virtual field to guide corpuscular quanta" (Slater, 1925c). Kramers and Bohr concurred: "we think that Slater's original hypothesis contains a good deal of truth."[91] Slater thus reverted to the position that, as he reminds the reader, he had been talked out of by Bohr and Kramers. He also noted that Swann had argued for this view during the December 1924 meeting of the *American Association for the Advancement of Science*, unaware that he, Slater, had been thinking along the same lines.[92] The following year, Bohr mentioned in passing in a letter to Slater that he had "a bad conscience in persuading you to our view." Slater told him not to worry about it.[93]

The way in which the BKS paper had come to be written, however, had left Slater with a bitter taste in his mouth (Schweber, 1990, 350–356). His disenchantment with Copenhagen shines through in the letter to Van Vleck, written on his way back to the United States, from which we quoted earlier (see note 15). Off the coast of Nantucket, a few hours before his ship—The Cunard R.M.S. "Lancastria"—docked in New York, he wrote:

> Don't remember just how much I told you about my stay in Copenhagen.

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[90] Stuewer (1975, 301) draws attention to a footnote in this paper that makes it clear that the experiment had been discussed even before Slater's arrival in Copenhagen: "The possibility of such a test was suggested by W. F. G. Swann in conversation with Bohr and one of us [Compton] in November 1923" (Compton and Simon, 1925, 290, note 6). Swann, the reader may recall, had just started in Chicago that fall, leaving the vacancy in Minnesota that was filled by Breit and Van Vleck (see sec. 1.2).

[91] Kramers to Urey, July 16, 1925, quoted by Stolzenburg (1984, 86).

[92] Cf. (Swann, 1925). See (Stuewer, 1975, 321–322) for discussion of Swann's proposal.

[93] Bohr to Slater, January 28, 1926; Slater to Bohr, May 27, 1926 (Bohr, 1972–1996, Vol. 5, 68–69).

> The paper with Bohr and Kramers was got out of the way the first six weeks or so—written entirely by Bohr and Kramers. That was very nearly the only paper that came from the institute at all the time I was there; there seemed to be very little doing. Bohr does very little and is chronically overworked by it . . . Bohr had to go on several vacations in the spring, and came back worse from each one.[94]

The second sentence of this passage is quoted by Dresden (1987, 165) in the course of his detailed discussion of Slater's reaction to his experiences in Copenhagen. In his AHQP interview, Slater was particularly down on Bohr and his institute. When he found out afterwards that Copenhagen would be one of the depositories for the AHQP materials, he wrote to Kuhn urging him to keep the interview out of the copy to be sent to Denmark.[95]

Initially, Slater was angry with both Bohr and Kramers, but his attitude toward the latter later softened (Dresden, 1987, 168–171). This was probably under the influence of his wife, fellow-physicist Rose Mooney (1902–1981) (Dresden, 1987, 527–528). Before Ms. Mooney became Mrs. Slater in 1948, she had been close to Kramers, whom she had met at a summer school in Michigan in 1938. The two of them almost certainly had an affair. Kramers was profoundly unhappy in his marriage to Anna 'Storm' Petersen, a Danish singer he had met in artistic circles in Copenhagen and married in 1920 after she got pregnant.[96] In one of the most memorable passages of his book, Dresden (1987, 289–295; see also 170–171) reveals that Kramers had told Storm many years after the fact that he himself had on at least one occasion been railroaded by Bohr. Kramers apparently thought of the Compton effect around 1920, well before Compton and Debye did. Bohr, however, detested the notion of light quanta so much that he worked on Kramers until he recanted. According to what Storm told Dresden, Kramers had to be hospitalized after one of these sessions with Bohr! Bohr's victory was complete. Even more strongly than Slater in the case of BKS a few years later, Kramers joined Bohr's crusade

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[94] Slater to Van Vleck, July 27, 1924 (AHQP). On the very same day, Slater wrote to Bohr, thanking him for his "great kindness and attention to me while I was in Copenhagen. Even if we did have some disagreements, I felt very well repaid for my time there, and I look back to it very pleasantly" (Bohr, 1972–1996, Vol. 5, 494). This sounds disingenuous in view of his comments to Van Vleck, but Slater had also been very positive about Bohr writing to his teacher Bridgman on February 1, 1924 (Schweber, 1990, 354).

[95] Slater to Kuhn, November 22, 1963, included in the file on Slater in the AHQP.

[96] Kramers was on the rebound at the time from the on-again-off-again relationship with his Dutch girlfriend, Waldi van Eck. Dresden's description of Kramers' relationship with van Eck (not to be confused with Van Vleck) conjures up the image of a virtual oscillator: "no commitments were made, no decisions were taken, the relationship was never defined, it was certainly never consummated, nor ever terminated" (Dresden, 1987, 525).

against light quanta with "all the passion of a repentant convert" (Dresden, 1987, 171). Slater may well have found out about this episode from his wife, Kramers' former mistress. Whether or not he did, in his autobiography, as Dresden (1987, 528) points out, Slater (1975, 233) refers to his BKS co-author as "my old friend Kramers."

Virtual oscillators survived the demise of the BKS theory and happily lived on in the dispersion theory from which they originated. As Van Vleck emphasizes in his NRC *Bulletin*, written after the Bothe-Geiger and Compton-Simon experiments, the rejection of the BKS theory and the acceptance of the light-quantum hypothesis

> does not mean that Slater's concept of virtual oscillators is not a useful one. We may assume that the fields which guide the light-quants come from a hypothetical set of oscillators rather than from the actual electron orbits of the conventional electrodynamics.[97] In this way the appearance of the spectroscopic rather than the orbital frequency in dispersion can be explained, and the essential features of the virtual oscillator theory of dispersion . . . can still be retained. There is an exact conservation of energy between the atoms and the actual corpuscular light-quants, but only a statistical conservation of energy between the atoms and the hypothetical virtual fields (Van Vleck, 1926a, 286–287).

Since virtual oscillators made their official debut in the context of BKS, the Kramers dispersion theory has been portrayed, in most older and even in some more recent historical literature, as an application of the BKS theory. Max Jammer (1966, 184), for instance, writes that the BKS theory "was the point of departure of Kramers's detailed theory of dispersion." And Mara Beller (1999, 23) still characterized (Kramers and Heisenberg, 1925) as a paper that "spelled out, in a rigorous mathematical way, the ideas only roughly outlined in the presentation of Bohr, Kramers, and Slater." Beller should have known better given that Dresden (1987, 144–146, 220–221) had already set the record straight. Darrigol (1992, 225) duly emphasizes that the Kramers dispersion theory was developed before and independently of the BKS theory. Even before Dresden, Hendry (1981) had already made it clear that BKS got its virtual oscillators from dispersion theory—the *Ersatz*-oscillators of (Ladenburg and Reiche, 1923)—and not the other way around.

To a large extent, Kramers and Slater have themselves to blame for the widespread impression that dispersion theory and virtual oscillators were bound up with BKS. As we saw above, (Slater, 1925a) is presented explicitly as an elaboration of the BKS theory. Kramers also presented his dispersion theory as an application of the BKS theory, both in his *Nature* notes (Kramers,

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[97] At this point, the following footnote is appended: "This viewpoint has been advocated by Slater during the printing of the present Bulletin. See [Slater, 1925a]."

1924a,b) and in (Kramers and Heisenberg, 1925).[98] In the abstract of their paper, Kramers and Heisenberg (1925, 223) announce that

> The arguments are based throughout on the interpretation of the connection of the wave radiation of the atom with the stationary states advocated in a recent paper by Bohr, Kramers and Slater, and the conclusions, should they be confirmed, would form an interesting support for this interpretation.[99]

In contrast, Van Vleck (1924b,c) used virtual oscillators without buying into the rest of the BKS theory.

The case of a closely related paper by Max Born (1924) is more ambiguous. At the beginning of the paper, Born writes:

> Recently . . . considerable progress has been made by Bohr, Kramers and Slater on just this matter of the connection between radiation and atomic structure . . . How fruitful these ideas are, is also shown by Kramers' success in setting up a dispersion formula . . . In this situation, one might consider whether it would not be possible to extend Kramers' ideas, which he applied so successfully to the interaction between radiation field and radiating electron, to the case of the interaction between several electrons of an atom . . . The present paper is an attempt to carry out this idea (Born, 1924, 181–182).

A footnote appended to this passage reads: "By a happy coincidence I was able to discuss the contents of this paper with Mr. Niels Bohr, which contributed greatly to a clarification of the concepts." Bohr had visited Born and Heisenberg in Göttingen in early June 1924 (Cassidy, 1991, 177–179). Heisenberg had already told Born all about the BKS theory and Born had expressed his enthusiasm for the theory in a letter to Bohr of April 16, 1924.[100] Bohr's visit must have further solidified this enthusiasm. A week later, however, Einstein passed through Göttingen and trashed the BKS theory.[101] In a letter to Born and his wife Hedi (1892–1972) of April 29, 1924, Einstein had already delivered his oft-quoted put-down that, should BKS turn out to be correct, he "would rather have been a shoemaker or even an employee in a gambling casino than a physicist" (Klein, 1970, 32). As a result of Einstein's onslaught, Born hedged

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[98] In addition, Kramers defended the BKS theory is several unpublished manuscripts written in 1924–1925, which have been preserved (Stolzenburg, 1984, 41).

[99] We use the translation of Stolzenburg (1984, 87) at this point, which is more accurate than the standard translation in (Van der Waerden, 1968, 223).

[100] See (Bohr, 1972–1996, Vol. 5, 299), discussed in (Mehra and Rechenberg, 1982–2001, Vol. 2, 143).

[101] See Heisenberg to Pauli, June 8, 1924 (Pauli, 1979, Doc. 62). For discussions of Einstein's objections to BKS, see (Klein, 1970, 32–35), (Wasserman, 1981, 255–263), and (Stolzenburg, 1984, 24–28, 31–34).

his bets and did not throw in his fate with the more controversial aspects of the BKS theory (see Mehra and Rechenberg, 1982–2001, Vol. 2, 144; Cassidy, 1991, 179). At the beginning of sec. 3 of his paper, he writes:

> it will be profitable to make use of the intuitive ideas, introduced by Bohr, Kramers and Slater ... but our line of reasoning will be independent of the critically important and still disputed conceptual framework of that theory, such as the statistical interpretation of energy and momentum transfer (Born, 1924, 189).[102]

Born, however, continued to be a true believer in BKS and took the collapse of the theory harder than Bohr himself. On April 24, 1925, he wrote to Bohr:

> Today Franck showed me your letter [of April 21, 1925, the day that Bohr had received word from Geiger about the results of the Bothe-Geiger experiment] ... which interested me exceedingly and indeed almost shocked me, because in it you abandon the radiation theory that obeyed no conservation laws (Bohr, 1972–1996, Vol. 5, 84).

In contrast to Born, Pauli, in a letter to Kramers of July 27, 1925,[103] called the demise of BKS "a magnificent stroke of luck" (*ein ungeheures Glück*). As is stressed by Darrigol (1992, 244), Pauli clearly recognized that dispersion theory remained unaffected. In the same letter, he alerted Kramers to a footnote in (Pauli, 1925, 5), which emphasizes

> that the formulas of [Kramers and Heisenberg, 1925] used here are independent of the special theoretical interpretation concerning the detailed description of the radiation phenomena in the quantum theory taken as a basis by them [i.e., the BKS theory], since these formulas only apply to averages over a large number of elementary phenomena.

Pauli wanted to distance himself from the notion put forward by Kramers and Heisenberg (1925) in the abstract of their paper (the passage quoted above) that evidence for their dispersion theory would also count as evidence for the BKS theory. He sneered:

> if I had not added the footnote in question, it would also have been true that the conclusions of *my* paper, if they should be confirmed, 'would form an interesting support for this interpretation' [this is a direct quote from

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[102] This illustrates the importance of what Beller (1999) has called the "dialogical approach" to the history of quantum mechanics (an approach adopted *avant la lettre* by Hendry [1984]): to resolve the tension between the two quoted passages in Born's paper, it is important to be attuned to the voices of both Bohr and Einstein in his text.

[103] (Pauli, 1979, 232–234) or (Bohr, 1972–1996, Vol. 5, 87).

> the abstract of Kramers and Heisenberg, 1925]. This impression I had, of course, to counteract! (Pauli to Kramers, July 27, 1925).

This letter was written after Heisenberg's *Umdeutung* paper, which was much more to Pauli's liking. In cruel Pauli fashion, he proceeded to berate Kramers for pushing the BKS theory:

> I now feel less lonely than about half a year ago when (spiritually and spatially) I found myself rather alone between the Scylla of the number-mystical Munich school and the Charybdis of the reactionary Copenhagen Putsch, propagandized by you to fanatical excesses! Now I only hope that you will no longer delay the process of recovery of Copenhagen physics which, as a result of Bohr's strong sense of reality, cannot fail to take place . . . Finally, kind regards to your wife, to whom I let the following be said: every time you—either out loud or to yourself—throw down all the curses of the Deuteronomy upon those who much prefer freedom of thought over any kind of "true faith", she shall sing merry tunes of spring, beauty and love to you! (ibid.).

Pauli apparently did not know at the time about the earlier clash between Bohr and Kramers over light quanta. And when Bohr commiserated with him a few years later about Kramers' lingering bitterness over this episode, Pauli appears to have had little sympathy for Kramers (Dresden, 1987, 294).[104] Despite his venom, however, Pauli's animus against Kramers, it seems, did not run all that deep. A few months after the scathing letter from which we quoted above, he told another correspondent (Stolzenburg, 1984, 91)

> [m]any greetings also to Kramers, whom I am very fond of after all, especially when I think of his beautiful dispersion formula (Pauli to Kronig, October 9, 1925).

There would have been far less confusion about the relation between dispersion theory and BKS, if Kramers had published his dispersion theory *before* the BKS paper, especially if he had done so availing himself of the Ladenburg-Reiche *Ersatz*-oscillators. As it happened, (Kramers, 1924a) was only submitted to *Nature* on March 25, 1924, nearly two months *after* the BKS paper. This note moreover contains only the briefest of hints as to how Kramers found his dispersion formula. It was not until July 22, 1924, that Kramers submitted a second note to *Nature* with at least an outline of the derivation of the formula. And he did not get around to publishing the full derivation until his paper

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[104] Pauli later told his colleague Res Jost (1918–1990) at the ETH in Zurich that he had consoled Bohr by arguing that discovering the Compton effect was hardly an impressive feat since both Compton and Debye had done so independently of one another.

with Heisenberg in early 1925.[105] The basic results, however, were in place by the time Slater arrived in Copenhagen in December 1923. Slater's letter to Van Vleck, from which we quoted above, continues:

> Kramers hasn't got much done, either. You perhaps noticed his letter to Nature on dispersion [Kramers, 1924a]; the formulas & that he had before I came, although he didn't see the exact application; and except for that he hasn't done anything, so far as I know. They seem to have too much administrative work to do. Even at that, I don't see what they do all the time. Bohr hasn't been teaching at all, Kramers has been giving one or two courses (Slater to Van Vleck, July 27, 1924 [AHQP]).

Van Vleck had to deal with the effects of Kramers' procrastination later in 1924. He finished his two-part paper on the application of the correspondence principle to the interaction of matter and radiation in June 1924. He had only seen Kramers' first note to *Nature* at that point. It was not clear on the basis of this short report exactly what Kramers had and had not yet done. Van Vleck thought he could extend some of Kramers' results. As he explained to Kramers in a letter of September 22, 1924 (AHQP):

> [my] article was ready to send to the printer about the time we received the copy of Nature containing your dispersion formula. In your note [Kramers, 1924a] I did not understand you to state how generally you had verified the asymptotic connection with the classical dispersion from the actual orbit, and it immediately occurred to me that this question could easily be investigated by the perturbation theory method I had previously developed in connection with what I call the "correspondence principle for absorption". I therefore inserted two sections (# 6 and # 15 . . .) showing that your formula merged into the classical one [in the limit of high quantum numbers]. Inasmuch as the classical dispersion formula had apparently not been developed for the general non-degenerate multiply periodic orbit, and as you did not give this in your note to Nature, I conjectured that you had verified the asymptotic connection only in special cases, such as a linear oscillator, so that my computations on dispersion would not be a duplication of what you had done. However, while visiting at Cambridge, Mass. last week I learned from Dr. Slater that your calculation of the asymptotic connection was almost identical with my own in scope and generality. I have therefore altered the proof of my Physical Review article to include a note [Van Vleck 1924b, 345] stating that you have also established the correspondence theorem in the general case.

The independent derivation of Kramers' results by Van Vleck, who certainly

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[105] This paper was completed over the Christmas break of 1924, received by *Zeitschrift für Physik* on January 5, 1925, and published two months later (Mehra and Rechenberg, 1982–2001, Vol. 2, 181).

was not under the spell of the BKS theory, underscores that dispersion theory and virtual oscillators were independent of the BKS theory.

As long as we think of the Kramers dispersion theory as part and parcel of BKS, we see matrix mechanics as replacing a decisively refuted theory. Once we recognize that the Kramers dispersion theory was developed before and independently of BKS, we see matrix mechanics growing naturally out of an eminently successful earlier theory. The BKS theory and its refutation by the Bothe-Geiger and Compton-Simon experiments are then seen as a sideshow distracting from the main plot line, the transition from dispersion theory to matrix mechanics. A corollary to this last observation is that the acceptance of the light-quantum hypothesis was largely irrelevant to the development of matrix mechanics. Compton scattering provided convincing evidence for the light-quantum hypothesis and against the BKS theory, but it had no bearing on dispersion theory. This is not to deny that the light-quantum hypothesis indirectly played a role in dispersion theory: the work of Ladenburg, Kramers, and Van Vleck crucially depends on Einstein's A and B coefficients, the derivation of which of course did involve light quanta.

After having spent most of this section trying to disentangle dispersion theory from BKS, we want to draw attention to one problem, common to these two theories, that may have played a role in the transition to matrix mechanics. This is the troublesome feature of the BKS explanation of the Compton effect that the electron and the virtual oscillators associated with it recoil with different velocities. This problem carries over to the dispersion theory based on the dual representation of atoms in terms of classical orbits and virtual oscillators, as is acknowledged, if only in passing, by Kramers and Heisenberg (1925, 229): "We shall not discuss in any detail the curious fact that the centre of these spherical waves moves relative to the excited atom." This sharpened the problem of the Bohr-Sommerfeld orbits in the theory. Not only were they responsible for the discrepancy between orbital frequencies and radiation frequencies, they also make it harder to picture an atom in space and time. After all, the system of electron orbits does not even move in concert with its orchestra of virtual oscillators.

Edward MacKinnon (1977, 1982) has argued forcefully that the resulting problem of combining these different pictures of the atom into one coherent picture forced Heisenberg to make a choice between them (see also Beller 1999, 23). Since the orchestra of virtual oscillators carries all the physical information about the system while the electron orbits are completely unobservable, the choice is obvious. MacKinnon (1977, 138) has gone as far as describing Heisenberg's *Umdeutung* paper as proposing a theory of virtual oscillators. Heisenberg, MacKinnon (1977, 155–156) suggests, merely suppressed all talk of virtual oscillators as a response to Pauli's objections—despite his earlier assurances to Bohr on this score—to the "virtualization" of physics. Einstein may have had a hand in this as well. Both Heisenberg and Pauli got an earful from Einstein on the BKS theory, Heisenberg, as we saw above, in Göttingen in June 1924, Pauli during the annual meeting of the *Gesellschaft Deutscher Naturforscher und Ärzte* in Innsbruck in September 1924.[106] As MacKinnon points out, Heisenberg (1925a) nonetheless made extensive use of virtual oscillators in a paper on the polarization of fluorescence radiation published a few months before the *Umdeutung* paper.

MacKinnon's reconstruction would also explain Heisenberg's later strikingly positive assessment of BKS. When Heisenberg first read the BKS paper, he was not impressed: "Bohr's paper on radiation is certainly very interesting; but I do not really see any fundamental progress."[107] He subsequently warmed to the theory, writing to Copenhagen on April 6, 1924 that he hoped Bohr had meanwhile convinced Pauli.[108] To Sommerfeld he wrote on November 18, 1924: "Maybe Bohr's radiation theory is a most felicitous [*sehr glücklicke*] description of this dualism [i.e., the wave-particle duality of radiation] after all" (Sommerfeld, 2004, 174, quoted in Wasserman, 1981, 251). Five years later, Heisenberg was praising BKS effusively:

> This investigation represented the real high point in the crisis of quantum theory, and, although it could not overcome the difficulties, it contributed, more than any other work of that time, to the clarification of the situation in quantum theory (Heisenberg, 1929, 492; translated and quoted in Stuewer, 1975, 291).

And thirty years later, Heisenberg (1955, 12) remembered the BKS theory as "the first serious attempt to resolve the paradoxes of radiation into rational physics" (quoted in Klein, 1970, 37). The profuse praise of 1929 and 1955 becomes understandable, if we assume that what Heisenberg was praising in retrospect was not the BKS theory as a whole but mainly the notion of virtual oscillators, which he had put to good use in (Heisenberg, 1925a), in (Kramers and Heisenberg, 1925), and, if MacKinnon is right, in his *Umdeutung* paper as well.

What speaks in MacKinnon's favor is that Heisenberg saw a draft of his 1977 paper and apparently approved of it (cf. MacKinnon 1977, 149, note 29). We have to keep in mind, however, that we are dealing here with recollections about fifty years after the fact. Then again, it is hardly far-fetched to talk

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[106] See Pauli to Bohr, October 2, 1924 (Pauli, 1979, Doc. 66), quoted and discussed in (Wasserman, 1981, 260–263).

[107] Heisenberg to Pauli, March 4, 1924 (Pauli, 1979, Doc. 57); quoted by Dresden (1987, 202) and Wasserman (1981, 250).

[108] See (Bohr, 1972–1996, Vol. 5, 354–355), cited by David Cassidy (1991, 176) to support his claim that "by the end of his March 1924 visit to Copenhagen, Werner was a convert."

about matrix mechanics in terms of virtual oscillators. In fact, Landé (1926, 456) introduced the phrase "virtual orchestras" to describe not BKS but matrix mechanics![109] The imagery, if not exactly the language, of an "orchestra of virtual oscillators" was also used in early popular expositions of matrix mechanics. In a popular book of the 1930s that went through many editions and was endorsed by Max Planck in a short preface, Ernst Zimmer wrote:[110]

> The state of an atom should no longer be described by the unobservable position and momentum of its electrons, but by the measurable frequencies and intensities of its spectral lines . . . Regardless of the nature of the real musicians who play the optical music of the atoms for us, Heisenberg imagines assistant musicians [*Hilfsmusiker*]: every one plays just one note at a certain volume. Every one of these musicians is represented by a mathematical expression, q_{mn}, which contains the volume and the frequency of the spectral line as in expressions in acoustics familiar to physicists. These assistant musicians are lined up in an orchestra [*Kapelle*] according to the initial and final states n and m of the transition under consideration. The mathematician calls such an arrangement a "matrix" (Zimmer, 1934, 161–162).

Zimmer's *Kapelle der Hilfsmusiker* was clearly inspired by Landé's *Ersatzorchester der virtuellen Oszillatoren.* Virtual oscillators thus not only survived the demise of the BKS theory but also the transition to matrix mechanics. In fact, as we shall see in sec. 5.1, the features captured by the notion of virtual oscillators can still readily be identified in the formalism of modern quantum mechanics.

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[109] Landé had worked with Heisenberg in 1924 (Cassidy, 1991, 177), resulting in a joint paper (Landé and Heisenberg, 1924).

[110] We are grateful to Jürgen Ehlers for drawing our attention to Zimmer's book.

3 Van Vleck and the application of the correspondence principle to the interaction of matter and radiation

In the two-part study that forms the focal point of our paper, Van Vleck (1924b,c) explored in a systematic and physically cogent fashion the implications of the correspondence principle for several aspects of the interaction of matter and radiation. The paper is signed June 19, 1924 and appeared in the October 1924 issue of *The Physical Review.* In this paper, Van Vleck gives a detailed derivation of the correspondence principle for absorption, which he had introduced in a short note in the *Journal of the Optical Society in America*, signed April 7, 1924 (Van Vleck, 1924a). In addition, he thoroughly examined the issues involved in connecting Einstein's A and B coefficients to features of classical electron orbits. Finally, as we mentioned in sec. 2.2, he showed that, in the limit of high quantum numbers, Kramers' quantum formula for polarization merges with the classical formula for polarization in arbitrary non-degenerate multiply-periodic systems.

In part I of his study, reproduced in (Van der Waerden, 1968), Van Vleck (1924b) discussed the transition from quantum-theoretical expressions for emission, absorption, and dispersion to corresponding classical expressions that one expects to hold in the limit of high quantum numbers. It is only in part II, not included in (Van der Waerden, 1968), that Van Vleck (1924c) derives the classical expressions for absorption and dispersion of radiation by a general non-degenerate multiply-periodic system, using standard methods of canonical perturbation theory in action-angle variables. Van Vleck could assume his audience to be thoroughly familiar with these techniques. This is no longer true today. For the sake of clarity of exposition, we therefore invert the order of Van Vleck's own presentation.

In sec. 3.1, we present the basic elements of the canonical formalism in action-angle variables and use it to rederive the classical formula (6) for the dipole moment of a charged one-dimensional simple harmonic oscillator. Though much more complicated than the derivation given in sec. 2.1, this new derivation has two distinct advantages. First, it suggests a way of translating the classical formula into a quantum formula with the help of Bohr's correspondence principle and Einstein's A and B coefficients. Secondly, both the derivation of the classical formula and its translation into a quantum formula can easily be generalized to arbitrary non-degenerate multiply-periodic systems.

In sec. 3.2, we translate the classical formula for the dipole moment of a simple harmonic oscillator into a quantum formula. In sec. 3.3, we similarly convert classical formulae for emission and absorption by a simple harmonic oscillator to the corresponding quantum formulae, Both the mathematical manipulations and the physical interpretation are particularly transparent in the case of a simple harmonic oscillator, and Van Vleck himself frequently used this example for illustrative purposes. The generalization of the various results to arbitrary non-degenerate multiply-periodic systems, which is a primary focus of Van Vleck's paper, will be deferred to sec. 4.

3.1 Deriving the classical formula for the dipole moment of a simple harmonic oscillator using canonical perturbation theory

In this subsection we rederive formula (6) for the dipole moment of a charged one-dimensional simple harmonic oscillator, using canonical perturbation theory in action-angle variables. Like Kramers, Van Vleck was a master of these techniques in classical mechanics. As Van Vleck recalled fifty years after the fact:

> In 1924 I was an assistant professor at the University of Minnesota. On an American trip, Ehrenfest gave a lecture there . . . [He] said he would like to hear a colloquium by a member of the staff. I was selected to give a talk on my "Correspondence Principle for Absorption" . . . I remember Ehrenfest being surprised at my being so young a man. The lengthy formulas for perturbed orbits in my publication on the three-body of the helium atom [Van Vleck, 1922b] had given him the image of a venerable astronomer making calculations in celestial mechanics (Van Vleck, 1974, 9).[111]

We begin by reviewing some of the mathematical tools we need.[112] Consider a classical Hamiltonian system with phase space coordinates (q^i, p_i), $i = (1, 2, \ldots N)$ and Hamiltonian $H(q^i, p_i)$, which does not explicitly depend on time. Hamilton's equations are

$$\dot{q}^i = \frac{\partial H}{\partial p_i}, \quad \dot{p}_i = -\frac{\partial H}{\partial q^i}. \tag{10}$$

Consider a contact transformation $(q^i, p_i) \to (q'^i, p'_i)$ preserving the form of Hamilton's equations, in the sense that there exists a new Hamiltonian H' such that

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[111] Van Vleck failed to conform to Ehrenfest's image of a young physicist in another respect. In an interview in 1973, "Van Vleck recalled, "I shocked Ehrenfest . . . when I told him I liked popular music." Ehrenfest, he said, "thought that was completely irreconcilable with my having written any respectable papers."" (Fellows, 1985, 54)

[112] This material is covered in standard graduate textbooks on classical mechanics, such as (Goldstein, 1980), heavily influenced by (Born, 1925) (Goldstein, 1980, 429, 493, 540). We recommend (Matzner and Shepley, 1991).

$$\dot{q}'^i = \frac{\partial H'}{\partial p'_i}, \ \ \dot{p}'_i = -\frac{\partial H'}{\partial q'^i}. \tag{11}$$

Since Hamilton's equations (10) and (11) must hold simultaneously, the variational principles

$$\delta \int\limits_{t_1}^{t_2} \left(\sum_i p_i \dot{q}^i - H(q^i, p_i) \right) dt = 0, \ \ \delta \int\limits_{t_1}^{t_2} \left(\sum_i p'_i \dot{q}'^i - H'(p'_i, q'^i) \right) dt = 0 \tag{12}$$

for arbitrary times t_1 and t_2 must also hold simultaneously. This implies that the difference between the two integrands in eq. (12) must be a total time derivative

$$\left(\sum_i p_i \dot{q}^i - H(q^i, p_i) - \sum_i p'_i \dot{q}'^i + H'(p'_i, q'^i) \right) dt = dF, \tag{13}$$

which will not contribute to the variation of the action. The apparent dependence of F on the $4N + 1$ variables $(q^i, p_i, q'^i, p'_i, t)$ can be reduced to $2N + 1$ variables via the equations for the contact transformation $(q^i, p_i) \rightarrow (q'^i, p'_i)$. If we choose to write F as a function of the initial and final coordinates, $F = F(q^i, q'^i, t)$, then the partial derivatives of F can be read off directly from eq. (13):

$$\frac{\partial F}{\partial t} = H' - H, \quad \frac{\partial F}{\partial q^i} = p_i, \quad \frac{\partial F}{\partial q'^i} = -p'_i. \tag{14}$$

By solving (at least in principle!) the second of these three equations for q'^i as a function of (q^i, p_i), and then substituting the result in the third to obtain p'_i, we see that the function F encodes the full information of the transformation $(q^i, p_i) \rightarrow (q'^i, p'_i)$. This function is called the *generating function* of the transformation. Given F the form of the new Hamiltonian H' can be obtained (again, in principle!) from the first of eqs. (14).

A special case of great interest occurs when the generating function F can be chosen so that the resulting Hamiltonian is independent of the new coordinates q'^i (which are then called *ignorable*). Hamilton's equations then immediately imply that the associated momenta p'_i are time-independent, and that the new coordinates q'_i are linear in time. In this circumstance the new momenta are usually called *action variables*—the notation J_i is conventional for these—while the new coordinates are dubbed *angle variables*, with the conventional notation w^i.

To illustrate the above with a concrete example, which we shall be using throughout this section, consider a one-dimensional simple harmonic oscillator with Hamiltonian:[113]

$$H = \frac{p^2}{2m} + \frac{1}{2}m\omega_0^2 q^2. \tag{15}$$

Consider the transformation induced by

$$F = \frac{1}{2}m\omega_0 q^2 \cot q'. \tag{16}$$

This function does not explicitly depend on time, so $H' = H$ (see eq. (14)). Eq. (14) also tells us that

$$p = \frac{\partial F}{\partial q} = m\omega_0 q \cot q', \quad p' = -\frac{\partial F}{\partial q'} = \frac{1}{2}m\omega_0 q^2 \csc^2 q'. \tag{17}$$

From the latter equation it follows that $q^2 = (2p'/m\omega_0)\sin^2 q'$ or that

$$q = \sqrt{\frac{2p'}{m\omega_0}}\sin q'. \tag{18}$$

Inserting this expression for q into the expression for p, we find

$$p = \sqrt{2m\omega_0 p'}\cos q'. \tag{19}$$

Substituting eqs. (18)–(19) for q and p into eq. (15) we find

$$H = \omega_0 p'. \tag{20}$$

Since $H' = H$, this means that the new coordinate variable q' is *ignorable*, as desired. Hamilton's equations for (q', p') are:

$$\dot{q}' = \frac{\partial H}{\partial p'} = \omega_0, \quad \dot{p}' = -\frac{\partial H'}{\partial q'} = 0, \tag{21}$$

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[113] A short digression on the (almost inevitable) notational confusions lurking in this subject is in order. We shall continue to use the conventional notation ω to denote angular frequencies, with the ordinary frequency (reciprocal period) denoted by the Greek letter ν. Unfortunately, Van Vleck uses ω to denote ordinary frequency! Moreover, there is the embarrassing similarity of the angle variables w^i to the frequencies ω_i. Also, there is the need to distinguish between the frequencies of the isolated mechanical system ($\omega_0 = 2\pi\nu_0$ for the simple harmonic oscillator) and the frequency of an applied electromagnetic wave, which we shall denote as $\omega = 2\pi\nu$ throughout.

from which it follows that $q' = \omega_0 t + \epsilon$ and that $p' = H/\omega_0$ is time-independent.

Instead of the canonically conjugate variables (p', q') it is customary to employ rescaled action/angle variables

$$J \equiv 2\pi p', \quad w \equiv \frac{1}{2\pi} q', \tag{22}$$

which reduce to $J = H/\nu_0$ and $w = \nu_0 t + \epsilon$ (appropriately redefining the arbitrary phase ϵ) for our one-dimensional oscillator.

The connection to the terminology *action variable* is easily seen in this example. In this simple case, the action is defined as the area enclosed by a single orbit of the periodic system in the two-dimensional phase space spanned by the coordinates (p, q):

$$J = \oint p dq \tag{23}$$

(cf. eq. (1)). Inserting eqs. (18) and (19) into the integrand, we find

$$\oint \left(\sqrt{2m\omega_0 p'} \cos q' \right) d \left(\sqrt{\frac{2p'}{m\omega_0}} \sin q' \right) = \int_0^{2\pi} 2p' \cos^2 q' dq' = 2\pi p', \tag{24}$$

which is just the expression for J in eq. (22).

The result (18) represents, of course, the solution of the equation of motion of the oscillator

$$q(t) = D \cos 2\pi\nu_0 t = D \cos 2\pi w, \tag{25}$$

where we have chosen the phase shift ϵ to start the oscillator at maximum displacement at $t = 0$, and where the amplitude is a function of the action variable

$$D = \sqrt{\frac{J}{m\pi\omega_0}}. \tag{26}$$

We now turn to our basic model for dispersion, i.e., a charged one-dimensional simple harmonic oscillator subjected to the periodically varying electric field of an electromagnetic wave. In sec. 2.1, we used elementary techniques of classical mechanics to analyze this system (see eqs. (2)–(6)). Although such methods are physically transparent, they depend on an explicit treatment of the equations of motion of a specific and completely specified Hamiltonian.

The same results can be obtained by the methods of canonical perturbation theory, where general formulas can be obtained for the perturbation in the coordinate(s) of the system *completely independently of the specific nature of the dynamics.* As Van Vleck (1924c, 350) put it:

> If we were to study the perturbations in the motion produced by the incident wave purely with the aid of [Newton's second law] it would be impossible to make further progress without specializing the form of the potential function [such as, e.g., $\frac{1}{2}m\omega_0^2q^2$ in eq. (15)] ... However, it is quite a different story when we seek to compute the perturbations ... in the "angle variables" w_1, w_2, w_3 and their conjugate momenta J_1, J_2, J_3 ... In fact by using them rather than x, y, z, which is the essential feature of the present calculation, the periodic properties of the system come to light even without knowing the form of [the potential].

Using canonical perturbation theory in action-angle variables, we rederive eq. (6) of sec. 2.1 for the polarization of a one-dimensional charged simple harmonic oscillator. In sec. 4, we turn to the general case of an arbitrary non-degenerate multiply-periodic system.

The Hamiltonian is now the sum of the Hamiltonian H_0 given by eq. (15) and a perturbative term H_{int} describing the interaction between the harmonic oscillator and the electromagnetic wave:[114]

$$H = H_0 + H_{\text{int}} = \frac{p^2}{2m} + \frac{1}{2}m\omega_0^2x^2 + eEx\cos\omega t. \tag{27}$$

The subscript '0' in ν_0 or ω_0 refers to the characteristic frequency of the unperturbed oscillator. Without subscript ν and ω refer to the frequency of the external electric field.

Absent a perturbing field ($E = 0, H = H_0$), we can write $x(t)$ in terms of the action-angle variables J and $w = \nu_0 t$:

$$x(t) = \sum_{\tau=\pm1} A_\tau(J)e^{2\pi i\tau w}, \tag{28}$$

where A_τ has to satisfy the conjugacy relation $A_\tau = A^*_{-\tau}$ to ensure that $x(t)$ in eq. (28) is real ($x(t) = x^*(t)$). Note that we have changed notation somewhat

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[114] As before, we assume that the electric field is in the direction of motion of the oscillator (cf. eq. (3)). It follows from eq. (27) that the force $F = -\partial V/\partial x$ of the electric field on the charge is $-eE\cos\omega t$, in accordance with eq. (3) (recall that we use e to denote the absolute value of the electron charge).

compared to eq. (25). We returned to Cartesian coordinate notation (x instead of q), and the amplitude has been redefined:[115]

$$D = 2|A_\tau|. \tag{29}$$

The action-angle variables $J = H_0/\nu_0$ and $w = \nu_0 t$ satisfy Hamilton's equations (cf. eqs. (21)–(22)):

$$0 = -\dot{J} = \frac{\partial H_0}{\partial w}, \qquad \frac{\partial H_0}{\partial J} = \dot{w} = \nu_0. \tag{30}$$

It is a special feature of the simple harmonic oscillator that the frequency ν_0 is independent of the amplitude of motion (and thereby of the action). The generating function for the contact transformation from (x, p) to (w, J) is time-independent (cf. eq. (16)), so eq. (14) implies that the old and new Hamiltonians coincide in value (i.e., one simply reexpresses the original Hamiltonian in the new variables). Even with the perturbation turned on *we shall continue to use the same contact transformation*, computing the perturbations $(\Delta w, \Delta J)$ induced by the applied field in the action-angle variables (w, J) as an expansion in E. These are *not* action-angle variables for the full Hamiltonian $H_0 + H_{\text{int}}$, only for the unperturbed Hamiltonian H_0 (cf. Van Vleck 1926a, 200–201).

Eventually, we are interested in the displacement Δx in the particle coordinate (to first order in E) induced by the applied field. To first order, Δx is given by

$$\Delta x = \frac{\partial x}{\partial J}\Delta J + \frac{\partial x}{\partial w}\Delta w. \tag{31}$$

Using eq. (28) to evaluate $\partial x/\partial J$ and $\partial x/\partial w$, we can rewrite this as:

$$\Delta x = \sum_\tau \left(\frac{\partial A_\tau}{\partial J}\Delta J + 2\pi i \tau A_\tau \Delta w \right) e^{2\pi i \tau w}. \tag{32}$$

Assuming the external field to be switched on at time zero, the first-order shifts Δw and ΔJ are given by:

$$\Delta J = \int_0^t \Delta \dot{J} dt, \quad \Delta w = \int_0^t \Delta \dot{w} dt. \tag{33}$$

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[115] Inserting $A_\tau = |A_\tau| e^{i\varphi}$ into eq. (28), we find $x(t) = (|A_\tau| + |A_{-\tau}|)\cos(2\pi w + \varphi)$. Since $A_\tau = A^*_{-\tau}$, $|A_\tau|^2 = A_\tau A^*_\tau$ is equal to $|A_{-\tau}|^2 = A_{-\tau}A^*_{-\tau}$. The phase factor φ is immaterial.

where the integrands $\Delta\dot{J}$ and $\Delta\dot{w}$ are determined by Hamilton's equations.

The perturbation in eq. (27) will induce a time-dependence in the action variable, as Hamilton's equation for the action variable in the presence of the perturbing field now reads

$$\dot{J} = -\frac{\partial H_0}{\partial w} - eE\frac{\partial x}{\partial w}\cos 2\pi\nu t = -eE\frac{\partial x}{\partial w}\cos 2\pi\nu t. \tag{34}$$

Note that we still have $\partial H_0/\partial w = 0$, so $\Delta\dot{J} = \dot{J}$. At this point it is convenient to go over to complex exponentials and replace $\cos 2\pi\nu t$ by $\frac{1}{2}(e^{2\pi i\nu t} + e^{-2\pi i\nu t})$. Inserting eq. (28) into eq. (34), we find

$$\Delta\dot{J} = -\pi ieE\sum_{\tau=\pm1}\tau A_\tau\left(e^{2\pi i(\tau w+\nu t)} + e^{2\pi i(\tau w-\nu t)}\right). \tag{35}$$

To obtain the polarization, which is a linear effect in the applied field E, we only need ΔJ and Δw to first order in E. This means that the angle variables w in the exponents in eq. (35) can be taken to zeroth order, i.e., $w = \nu_0 t$. Integrating $\Delta\dot{J}$ we find:

$$\Delta J = \int_0^t \Delta\dot{J}dt = \frac{eE}{2}\sum_{\tau=\pm1}\tau A_\tau\left\{\frac{1-e^{2\pi i(\tau\nu_0 t+\nu t)}}{\tau\nu_0+\nu} + \frac{1-e^{2\pi i(\tau\nu_0 t-\nu t)}}{\tau\nu_0-\nu}\right\}. \tag{36}$$

Next, we need to compute the first order shift Δw in the angle variable w. Hamilton's equation for the angle variable w in the presence of the perturbation is:[116]

$$\begin{aligned}\dot{w} &= \frac{\partial H_0}{\partial J} + eE\frac{\partial x}{\partial J}\cos 2\pi\nu t\\ &= \nu_0 + \frac{eE}{2}\sum_{\tau=\pm1}\frac{\partial A_\tau}{\partial J}\left(e^{2\pi i(\tau w+\nu t)} + e^{2\pi i(\tau w-\nu t)}\right).\end{aligned} \tag{37}$$

Once again, w may be replaced by $\nu_0 t$ in the exponentials in eq. (37). Integrating the second term in eq. (37), which gives the shift $\Delta\dot{w}$ due to H_{int}, we find:

$$\Delta w = \int_0^t \Delta\dot{w}dt = \frac{ieE}{4\pi}\sum_{\tau=\pm1}\frac{\partial A_\tau}{\partial J}\left\{\frac{1-e^{2\pi i(\tau\nu_0 t+\nu t)}}{\tau\nu_0+\nu} + \frac{1-e^{2\pi i(\tau\nu_0 t-\nu t)}}{\tau\nu_0-\nu}\right\}. \tag{38}$$

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[116] It is a special feature of the simple harmonic oscillator that the characteristic frequency ν_0 is independent of the amplitude and thus of the action variable J (see eq. (26)). In general, ν_0 will be a function of J. The first term on the right-hand side of eq. (37) would then become $\partial H_0/\partial J = \nu_0(J) = \nu_0 + (\partial\nu_0/\partial J)\Delta J$.

Substituting expressions (36) and (38) for ΔJ and Δw into eq. (32), we find

$$\Delta x = \frac{eE}{2} \sum_{\tau'=\pm1} \sum_{\tau=\pm1} \left\{ \frac{\partial A_{\tau'}}{\partial J} \tau A_\tau - \tau' A_{\tau'} \frac{\partial A_\tau}{\partial J} \right\} \frac{1 - e^{2\pi i(\tau\nu_0 t - \nu t)}}{\tau\nu_0 - \nu} e^{2\pi i \tau' \nu_0 t} \qquad (39)$$
$$+ \ (\nu \to -\nu),$$

where "$(\nu \to -\nu)$" here and below is shorthand for: "the same term with ν replaced by $-\nu$ everywhere." The coherent contribution to the polarization comes from the terms in eq. (39) with the same time-dependence as the applied field, i.e., from terms in which the time-dependence is given by the factor $e^{\pm 2\pi i\nu t}$. In the terminology of Van Vleck (1924c, 361): "the part of the displacement which is resonant to the impressed wave." These are the terms in which the summation indices, which in the case of the simple harmonic oscillator only take on the values ±1, have opposite values, i.e., $\tau = -\tau'$. The contribution of such terms to the first-order displacement is

$$\begin{aligned} \Delta x_{\text{coh}} &= \frac{eE}{2} \sum_{\tau=\pm1} \left\{ \left(\frac{\partial A_{-\tau}}{\partial J} \tau A_\tau + \tau A_{-\tau} \frac{\partial A_\tau}{\partial J} \right) \frac{-e^{-2\pi i\nu t}}{\tau\nu_0 - \nu} + (\nu \to -\nu) \right\} \\ &= \frac{eE}{2} \sum_{\tau=\pm1} \tau \frac{\partial |A_\tau|^2}{\partial J} \left\{ \frac{e^{-2\pi i\nu t}}{\nu - \tau\nu_0} - \frac{e^{2\pi i\nu t}}{\nu + \tau\nu_0} \right\}. \qquad (40) \end{aligned}$$

The imaginary part of this expression is a sum over the product of odd and even functions of the index τ,

$$-\frac{eE}{2} \sum_{\tau=\pm1} \tau \frac{\partial |A_\tau|^2}{\partial J} \left(\frac{1}{\nu - \tau\nu_0} + \frac{1}{\nu + \tau\nu_0} \right) \sin 2\pi\nu t, \qquad (41)$$

and therefore vanishes, leaving only the real part:

$$\begin{aligned} \Delta x_{\text{coh}} &= \frac{eE}{2} \sum_\tau \tau \frac{\partial |A_\tau|^2}{\partial J} \left(\frac{1}{\nu - \tau\nu_0} - \frac{1}{\nu + \tau\nu_0} \right) \cos 2\pi\nu t \\ &= \frac{eE}{2} \sum_\tau \tau \frac{\partial |A_\tau|^2}{\partial J} \left(\frac{2\tau\nu_0}{\nu^2 - \tau^2\nu_0^2} \right) \cos 2\pi\nu t. \qquad (42) \end{aligned}$$

Since $|A_\tau|^2 = |A_{-\tau}|^2$ (see note 115) and since τ only takes on the values ±1 in the case of the simple harmonic oscillator, $\tau^2 = 1$ and the two terms in the summation over τ are identical. Although in this special case the derivative with respect to J only acts on $|A_\tau|^2$, we are free to include the expression $2\nu_0/(\nu^2 - \nu_0^2)$ within the scope of the derivative (recall that ν_0 does not depend on J in this case). Eq. (42) then becomes

$$\Delta x_{\text{coh}} = 2eE\frac{\partial}{\partial J}\left(\frac{\nu_0}{\nu^2-\nu_0^2}|A_\tau|^2\right)\cos 2\pi\nu t. \tag{43}$$

The resulting expression for the dipole moment, $p(t) = -e\Delta x_{\text{coh}}$, of a one-dimensional charged simple harmonic oscillatoris a special case of the expressions for the dipole moment of a general non-degenerate multiply-periodic system with the same charge given by Kramers and Van Vleck. Kramers (1924b, 310, eq. 2*) denotes this quantity by P and gives the following formula:

$$P = \frac{E}{2}\sum\frac{\partial}{\partial I}\left(\frac{C^2\omega}{\omega^2-\nu^2}\right)\cos 2\pi\nu t. \tag{44}$$

In the special case of a one-dimensional charged simple harmonic oscillator, ω, I, and C correspond to ν_0, J, and $2|A_\tau|$ in our notation, respectively. There appears to be a factor e^2 missing in Kramers' formula. We shall derive the corresponding formula (41) in (Van Vleck, 1924c, 361) in sec. 4.2.

Eq. (43) is equivalent to eq. (6), the result of our much simpler derivation in sec. 2.1. Recalling that (cf. eqs. (25)–(26), eqs. (28)–(29) and note 115)

$$x(t) = 2|A_\tau|\cos 2\pi\nu_0 t = \sqrt{\frac{J}{2\pi^2 m\nu_0}}\cos 2\pi\nu_0 t, \tag{45}$$

we have $|A_\tau|^2 = J/(8\pi^2 m\nu_0)$, and eq. (43) reduces to

$$\Delta x_{\text{coh}} = \frac{eE\cos 2\pi\nu t}{4\pi^2 m(\nu^2-\nu_0^2)}. \tag{46}$$

The dipole moment is thus given by:

$$p(t) = -e\Delta x_{\text{coh}} = \frac{e^2E}{4\pi^2 m(\nu_0^2-\nu^2)}\cos 2\pi\nu t, \tag{47}$$

in agreement with eq. (6).

The preceding discussion employs a version of canonical perturbation theory in which a single set of action-angle variables, chosen for the unperturbed Hamiltonian, is used throughout the calculation, even after the time-dependent perturbation is switched on. Accordingly, the new action variables are no longer constant, and the new angle variables are no longer linear in time. The same classical polarization result is derived in a somewhat different manner by Born

(1924) and by Kramers and Heisenberg (1925). Born performs a contact transformation in which the generating function F (cf. eq. (13)) is chosen as a function of the old coordinates and the new momenta $F(q^i, p'_i)$, which is then evaluated systematically order by order in the perturbation to maintain the constancy of the new action variables. In (Kramers and Heisenberg, 1925) the same procedure is followed, but as only the first order result is needed, it suffices to use the infinitesimal form of the contact transformation.[117]

3.2 Converting the classical formula for dispersion to a quantum formula in the special case of a simple harmonic oscillator

Using Bohr's correspondence principle as our guide, we now 'translate' the classical formula (43) for displacement (and thence for polarization) into a quantum formula. Two main ingredients go into this particular application of the correspondence principle: (1) a rule—commonly attributed to Born (1924)[118] but found and applied earlier by Kramers and Van Vleck (see below)—for replacing derivatives with respect to the action variables in classical formulae by difference quotients involving neighboring quantum states; (2) the A and B coefficients of Einstein's quantum theory of radiation. In general, the 'translation' of a classical formula into a quantum formula involves a third step. The orbital frequencies need to replaced by transition frequencies. The case of a simple harmonic oscillator has the special features that the only relevant transitions are between adjacent states and that the transition frequency $\nu_{i\to f}$ coincides with the mechanical frequency ν_0. Another special feature is that the correspondence between quantum and classical results for large quantum numbers continues to hold all the way down to the lowest quantum numbers, due to the extremely simple form of the energy spectrum, with uniformly spaced levels.

Using the rule for replacing derivatives by difference quotients, the quantum formula for polarization is obtained from (43) by the formal correspondence replacement

$$\left.\frac{\partial F}{\partial J}\right|_{J=rh} \to \frac{1}{h}(F(r+1) - F(r)), \tag{48}$$

where $F(r)$ refers to any dynamical quantity associated with the quantum state specified by the integer quantum number r. In the correspondence limit

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[117] For a discussion of infinitesimal canonical transformations, see Ch. 11 of (Matzner and Shepley, 1991).

[118] See, e.g., (Jammer, 1966, 193), and, following Jammer, (Aitchison *et al.*, 2004, 1372), or (Cassidy, 1991, 178, 186, 188).

where r gets very large, the difference between the values rh and $(r+1)h$ for the action variable J become so small that the difference quotient to the right of the arrow in eq. (48) becomes equal the derivative on the left. With this prescription, the classical formula eq. (43) turns into a quantum expression for the coherent part of the displacement of the particle in quantum state r:

$$\Delta x^r_{\text{coh}} = \frac{2eE}{h}\left(\frac{\nu_0|A^{r+1}|^2}{\nu^2-\nu_0^2} - \frac{\nu_0|A^r|^2}{\nu^2-\nu_0^2}\right)\cos 2\pi\nu t. \tag{49}$$

The amplitudes A^r correspond to the A_τ (with $\tau = \pm 1$) in eq. (43), and are related to the amplitudes D_r in eq. (26) for an oscillator in state r by $D_r = 2|A^r|$ (see eq. (45)). As we saw in sec. 2.1, Ladenburg (1921) showed how these amplitudes can be connected to the Einstein A coefficients for spontaneous emission (not to be confused with the amplitudes A^r).

At this point we briefly review Einstein's quantum theory of radiation (Einstein, 1916a,b, 1917), using the notation of (Van Vleck, 1924b). Imagine an ensemble of atoms—or indeed, any conceivable quantized mechanical system, such as one-dimensional quantized oscillators—in interaction and statistical equilibrium with an ambient electromagnetic field of spectral density $\rho(\nu)$. If we label the stationary states of the atoms by indices $r, s, \ldots$, the number of atoms in state r (of energy E_r) by N_r, and recall the Bohr frequency condition $\nu_{rs} = (E_r - E_s)/h$, Einstein's analysis gives an average rate of energy emission of light of frequency ν_{rs} for an atom in state r as

$$\frac{dE_{r\to s}}{dt} = h\nu_{rs}\left(A_{r\to s} + B_{r\to s}\rho(\nu_{rs})\right), \tag{50}$$

and the rate of energy absorption of light of frequency ν_{rs} by an atom in state s as

$$\frac{dE_{s\to r}}{dt} = h\nu_{rs}B_{s\to r}\rho(\nu_{rs}). \tag{51}$$

Einstein's analysis of the requirements for thermodynamic equilibrium and comparison with Planck's law of black-body radiation then yields the critical relations

$$B_{r\to s} = B_{s\to r} = \frac{c^3}{8\pi h\nu_{rs}^3}A_{r\to s}. \tag{52}$$

For a charged simple harmonic oscillator, the only allowed transitions amount to changes in the action by one unit of Planck's constant h, so there is only a single Einstein coefficient for spontaneous emission from the state $r+1$, namely $A_{r+1\to r}$. The correspondence principle dictates that we associate the rate of spontaneous energy emission for *high* quantum numbers,

$$\frac{dE_{r+1\to r}}{dt} = h\nu_0 A_{r+1\to r} \tag{53}$$

(cf. eq. (50), in the absence of external radiation) with the classical result for the power emitted by an accelerated (in this case, oscillating) charge, given by the Larmor formula (Jackson, 1975; Feynman *et al.*, 1964, Vol. 1, Ch. 32):

$$P = \frac{2}{3}\frac{e^2}{c^3}\dot{v}^2. \tag{54}$$

For an oscillator in state r, with $x(t) = D_r \cos\omega_0 t$, this becomes, for the instantaneous power emission P_r in state r

$$P_r = \frac{2}{3}\frac{e^2}{c^3}\omega_0^4 D_r^2 \cos^2\omega_0 t, \tag{55}$$

the time average of which, $\frac{1}{3}(e^2/c^3)\omega_0^4 D_r^2$, then gives the desired connection between the amplitudes $D_r = 2|A^r|$ appearing in eq. (49) and the Einstein coefficient $A_{r+1\to r}$:

$$\begin{aligned} h\nu_0 A_{r+1\to r} &= \frac{4}{3}\frac{e^2}{c^3}\omega_0^4 |A^{r+1}|^2 \\ |A^{r+1}|^2 &= \frac{3hc^3}{64\pi^4 e^2 \nu_0^3} A_{r+1\to r}. \end{aligned} \tag{56}$$

Van Vleck (1924b, 333) refers to this connection as the "correspondence principle for emission." Multiplying the displacement Δx in eq. (49) by the charge $-e$ to obtain the dipole moment per oscillator, and by $n_{\rm osc}$, the number density of oscillators, we obtain the following result for the polarization induced by the electric field E:

$$P_r = 3\frac{n_{\rm osc} c^3}{32\pi^4} E \left(\frac{A_{r+1\to r}}{\nu_0^2(\nu_0^2 - \nu^2)} - \frac{A_{r\to r-1}}{\nu_0^2(\nu_0^2 - \nu^2)} \right) \cos 2\pi\nu t. \tag{57}$$

Of course, for the special case of the ground state of the oscillator, $r = 0$, the second term in eq. (57) cannot be present. Ladenburg's quantum formula for dispersion accordingly only had the equivalent of the first term in eq. (57) (Ladenburg, 1921). The full equation corresponds to eq. (5) in (Kramers, 1924a), and to eq. (17) in (Van Vleck, 1924b), except for a factor of 3, as we have not assumed random orientation of the oscillators (Van Vleck, 1924b, footnote 25).

One may easily guess that the corresponding formula for a more general, multiply-periodic system will take the form of (Van Vleck, 1924b, eq (17)), in analogy to (57) (cf. eqs. (6)–(7)):

$$P_r = 3\frac{n_{\rm osc}c^3}{32\pi^4}E\left(\sum_s \frac{A_{s\to r}}{\nu_{sr}^2(\nu_{sr}^2-\nu^2)} - \sum_t \frac{A_{r\to t}}{\nu_{rt}^2(\nu_{rt}^2-\nu^2)}\right)\cos 2\pi\nu t, \tag{58}$$

where the sum over s (resp. t) corresponds to states higher (resp. lower) than the state r, and where ν_{ij} is Van Vleck's notation for the transition frequency $\nu_{i\to j}$. In the correspondence limit where r is very large and neither s nor t differ much from r, the transition frequencies ν_{sr} and ν_{rt} become equal to the orbital frequencies in the orbits characterized by the values rh, sh, and th for the action variable J. For the harmonic oscillator, the sums in eq. (58) degenerate to a single term each (with $s = r+1$, $t = r-1$), and the transition frequencies ν_{sr}, ν_{rt} are all equal to the mechanical frequency ν_0. In sec. 4.2 we shall explain Van Vleck's derivation of eq. (58) in detail.

From the point of view of classical dispersion theory, the terms in the second sum in eq. (58) for polarization, corresponding to transitions to lower states, have no direct physical interpretation. They appear to correspond to oscillators of negative mass (Kramers 1924a, 676; 1924b, 311)! In the early spring of 1924 Van Vleck had already derived an expression for absorption with a structure similar to that of eq. (58) for polarization (see sec. 3.3). In the case of absorption, the terms with transitions to lower states are readily recognized as corresponding to "negative absorption," i.e., to the process of stimulated emission introduced by Einstein.

As we indicated above, there is some disagreement in the historical literature as to who was or who were responsible for the key move in the construction of the quantum dispersion formula on the basis of the correspondence principle, viz. the replacement (48) of derivatives with respect to the action variable by difference quotients. Jammer (1966, 193) and Mehra and Rechenberg (1982–2001, Vol. 2, 173) suggest that Kramers got the idea from Born via Heisenberg. The rule can be found in (Born, 1924), published well before Kramers first used it in print in (Kramers and Heisenberg, 1925). Dresden (1987, 222) convincingly argues, however, that Kramers and Born found this rule independently of one another, with Kramers finding it well before Born. Recall that Slater wrote to Van Vleck in July 1924 that Kramers already had the derivation of his dispersion formula in December 1923 (see sec. 2.2). It turns out that Van Vleck discovered the replacement (48) of derivatives by difference quotients for himself as well. Since Van Vleck (1924a) announced the correspondence principle for absorption, which he could not have derived without this rule, in a paper submitted in April 1924, whereas (Born, 1924) was not received by *Zeitschrift für Physik* until June 1924, Van Vleck clearly could not have taken the rule from Born's paper. Writing to Born later in 1924, Van Vleck sounds slightly annoyed at Born's insinuation that he, Van Vleck, somehow did not realize that one needs to replace derivatives by difference quotients to get from classical to quantum-theoretical expressions. In a letter of October 24, 1924, from which we already quoted in sec. 1.4, Born had written:

> I am sending you my paper On Quantum Mechanics [Born 1924], which pursues a goal similar to yours. While you limit yourself to the correspondence with high quantum numbers, I conversely aim for rigorous laws for arbitrary quantum numbers.

To which Van Vleck replied:

> Thank you for your letter and reprint relating to "Quantum Dynamics, etc" . . . I have read with great interest your important, comprehensive article. There is, as you say, considerable similarity in the subject matter in your article and mine, especially as regards to dispersion[119] . . . As noted in your letter you mention more explicitly than do I the fact that formulas of the quantum theory result from those of the classical theory by replacing a derivative by a difference quotient. I have stressed the asymptotic connection of the two theories but I think it is clear in the content of my article that in the problems considered the classical and quantum formulas are connected as are derivatives and difference quotients (Van Vleck to Born, November 30, 1924 [AHQP]).

That Kramers, Van Vleck, and Born independently of one another hit upon the same idea, underscores that the rule (48) for replacing derivatives by difference quotients is so natural that it readily comes to mind when one is trying to construct quantum-theoretical expressions out of classical ones on the basis of the correspondence principle.

3.3 Emission and absorption classically and quantum-theoretically in the special case of a simple harmonic oscillator

Before we present Van Vleck's "correspondence principle for absorption" (for the special case of a simple harmonic oscillator), we gather some useful results from the classical theory of a charged oscillator (of natural frequency ν_0) coupled to a Maxwellian electromagnetic field. Such an oscillator (i) emits electromagnetic radiation of frequency ν_0 in the absence of an external field,

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[119] Van Vleck seems to be talking here about (Van Vleck, 1924b,c), whereas Born was talking about (Van Vleck, 1924a). Born asked Van Vleck to send him "an offprint of your extensive calculations." Van Vleck obliged: 'As you requested, I am sending you under separate cover a reprint of Parts I and II of my computations," presumably (Van Vleck, 1924b,c).

(ii) absorbs energy from an applied electromagnetic field of frequency ν, and (iii) undergoes a net displacement coherent with an applied electromagnetic field (or "polarization", analyzed above).

The Larmor formula (54) gives the power loss due to radiation by our charged harmonic oscillator. The energy loss of the oscillating system can be ascribed to a radiative reaction force given by

$$F_{\mathrm{rad}} = \frac{2e^2}{3c^3}\ddot{v} \equiv m\tau\ddot{v}, \tag{59}$$

where we shall assume that the characteristic time τ is very short in comparison to the mechanical period: $\omega_0\tau << 1$, so that radiation damping is very slow on the time scale of the mechanical oscillations of the system. The equation of motion of the oscillator (in the absence of external applied forces) now becomes

$$\dot{v} - \tau\ddot{v} + \omega_0^2 x = 0. \tag{60}$$

Now, to a good approximation, the coordinates and velocities of this system are still behaving as harmonic oscillations of frequency ω_0 so we may assume $\ddot{v} \simeq -\omega_0^2 v$ in (60) and obtain

$$\ddot{x} + \tau\omega_0^2\dot{x} + \omega_0^2 x = 0. \tag{61}$$

Inserting the *Ansatz* $x(t) = De^{-\alpha t}$ into equation (61), we find:

$$(\alpha^2 - \tau\omega_0^2\alpha + \omega_0^2)De^{-\alpha t} = 0. \tag{62}$$

Neglecting a term with $\tau^2\omega_0^4$ (recall that $\omega_0\tau << 1$, so that $\tau^2\omega_0^4 << \omega_0^2$),[120] we can rewrite the expression in parentheses as:

$$(\alpha - \frac{1}{2}\tau\omega_0^2 + i\omega_0)(\alpha - \frac{1}{2}\tau\omega_0^2 - i\omega_0). \tag{63}$$

It follows that:

$$\alpha \simeq \frac{1}{2}\tau\omega_0^2 \pm i\omega_0 \equiv \Gamma/2 \pm i\omega_0. \tag{64}$$

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[120] Such terms are treated incorrectly in any event by the approximation leading to eq. (61).

Thus, we have a solution of the form

$$x(t) = De^{-\Gamma t/2} \cos \omega_0 t, \tag{65}$$

from which the average rate of oscillator energy loss from the Larmor formula (54) at small times (i.e., when damping due to the $e^{-\Gamma t/2}$ factor can be ignored) is easily seen to be

$$-\frac{dE_{\text{osc}}}{dt} = \frac{e^2}{3c^3} D^2 \omega_0^4 = \frac{16\pi^4 e^2}{3c^3} D^2 \nu_0^4 \tag{66}$$

(where we used that $\dot{v} \simeq \omega_0^2 D$). The constant $\Gamma = \tau \omega_0^2$ is called the radiative decay constant. We emphasize again that the preceding discussion presupposes the narrow resonance limit, $\Gamma << \omega_0$. In terms of Γ, the basic equation of motion (61) can be written as

$$\ddot{x} + \Gamma \dot{x} + \omega_0^2 x = 0. \tag{67}$$

Now suppose that our charged oscillator is immersed in an ambient electromagnetic field, characterized by a spectral function (energy density per unit spectral interval) $\rho(\nu)$. As we are dealing with one-dimensional oscillators we shall simplify the discussion by assuming that only the x-component of the electric field is relevant as all the oscillators are so aligned. Then (using overbars to denote time averages) the average value of the electromagnetic energy density is (in Gaussian units) $(1/4\pi)\bar{\mathbf{E}}^2 = (3/4\pi)\bar{E_x}^2 = \rho(\nu)\Delta\nu$ in the frequency interval $(\nu, \nu + \Delta\nu)$. If $E_x = E \cos 2\pi\nu t$ we have $\bar{E_x}^2 = E^2/2$ so finally we have

$$E^2 = \frac{8\pi}{3} \rho(\nu) \Delta\nu. \tag{68}$$

The equation of motion (67) must be modified to include the coupling to the external field (switching back temporarily to angular frequencies, $\omega = 2\pi\nu$, and using complex notation to encode amplitude and phase information):

$$\ddot{x} + \Gamma \dot{x} + \omega_0^2 x = \frac{eE}{m} e^{i\omega t} \equiv F_{\text{app}}/m, \tag{69}$$

and the average rate of energy absorption of the oscillator from the ambient field is simply the time average $< F_{\text{app}} \dot{x} >$. This linear second order equation is solved by a sum of transients (i.e. solutions of the homogeneous equation: see eq. (67))

$$x_{\text{tr}}(t) = De^{-\Gamma t/2} \cos \omega_0 t, \tag{70}$$

plus the following particular solution coherent with the applied perturbation

$$x_{\text{coh}}(t) = \text{Re}\ \frac{eE}{m}\frac{e^{i\omega t}}{\omega_0^2 - \omega^2 + i\Gamma\omega}, \tag{71}$$

so that the desired time average $< F_{\text{app}}\dot{x} >=< F_{\text{app}}(\dot{x}_{\text{tr}} + \dot{x}_{\text{coh}}) >$ giving the energy absorption rate becomes

$$< F_{\text{app}}\dot{x} >=< eE\cos\omega t\ \frac{eE}{m}\ \text{Re}\left(\frac{i\omega e^{i\omega t}}{\omega_0^2 - \omega^2 + i\Gamma\omega}\right) > . \tag{72}$$

Note that the transient part of the particle coordinate $x_{\text{tr}}(t)$ is *not* coherent with the applied field (we assume $\omega \neq \omega_0$), and therefore does not contribute to the time average of the energy absorption. This explains why the amplitude D of the oscillations is absent from the final result, which will instead depend only on the specific energy density of the ambient field. In other words, even though the charged particle may be executing very large amplitude oscillations $x_{\text{tr}}(t)$, the only part of the full coordinate $x(t)$ responsible for a nonvanishing average absorption is the part of the displacement $x_{\text{coh}}(t)$ induced by the applied field, which is proportional to E and does not involve the amplitude D. As we shall see below, the corresponding feature of the quantum calculation in the correspondence limit led Van Vleck to the very important realization that the net energy absorption involves a *difference* in the amount of absorption and stimulated emission as described in Einstein's quantum theory of radiation.

Only the cosine part of the complex exponential in eq. (72) will contribute to the time average. Using $< \cos^2\omega t >= 1/2$ and eq. (68), we find

$$\begin{aligned} < F_{\text{app}}\dot{x} >&= \frac{e^2E^2\Gamma}{2m}\frac{\omega^2}{(\omega_0^2 - \omega^2)^2 + \Gamma^2\omega^2} \\ &= \frac{4\pi e^2}{3m}\rho(\frac{\omega}{2\pi})\Gamma\frac{\omega^2}{(\omega_0^2 - \omega^2)^2 + \Gamma^2\omega^2}\frac{1}{2\pi}\Delta\omega \end{aligned} \tag{73}$$

for the energy absorption rate due to the ambient field in the frequency interval $(\nu, \nu + \Delta\nu) = (\omega, \omega + \Delta\omega)$. Since eq. (73) contains the electric field E squared, it is apparent that the generalization of this linear simple harmonic oscillator result to an arbitrary multiply-periodic system will require a second-order canonical perturbation theory calculation, which will necessarily be more involved than the corresponding classical polarization calculation, which only involves the electric field to the first order. In the case of interest, where $\Gamma << \omega_0$, the line resonance shape in eq. (73) is highly peaked around the resonance frequency ω_0, so we may use the distributional limit

$$\frac{\epsilon}{x^2 + \epsilon^2} \to \pi\delta(x), \quad \epsilon \to 0 \tag{74}$$

with $x = \omega^2 - \omega_0^2$ and $\epsilon = \Gamma\omega$ to execute the integration over ω in eq. (73) and compute the total absorption rate:

$$\begin{aligned} < F_{\text{app}}\dot{x} > &\approx \frac{2e^2}{3m}\int \rho(\frac{\omega}{2\pi})\Gamma\frac{\pi}{\Gamma\omega}\omega^2\delta(\omega^2 - \omega_0^2)d\omega \\ &= \frac{\pi e^2}{3m}\rho(\nu_0). \end{aligned} \tag{75}$$

This classical result is found in (Planck, 1921) (Van Vleck, 1924b, 339, note 12))[121] and gives the rate at which a classical charged oscillator gains energy when immersed in an ambient classical electromagnetic field.

In eq. (56) we found the connection in the limit of high quantum numbers between the Einstein A coefficients and the amplitudes $D_r = 2|A^r|$ of the mechanical motion in the emitting state r:

$$A_{r\to s} \simeq \frac{16\pi^4 e^2}{3hc^3}D_r^2\nu_{rs}^3. \tag{76}$$

From the Einstein relation (52) this implies a corresponding result for the B-coefficients:

$$B_{r\to s} = B_{s\to r} = \frac{2\pi^3 e^2}{3h^2}D_r^2. \tag{77}$$

In the r-th quantized state of the oscillator, we have $J = rh$ so from eq. (26) the corresponding amplitude D_r^{qu} of the *quantized* motion becomes

$$D_r^{\text{qu}} = \sqrt{\frac{rh}{2\pi^2 m\nu_0}}, \tag{78}$$

and the quantum result for the A coefficients in the present case of a linear simple harmonic oscillator becomes

$$A_{r\to r-1} = \frac{8\pi^2 e^2\nu_0^2 r}{3mc^3}, \tag{79}$$

while the quantum result for the B coeffficients takes the form

$$B_{r\to r-1} = B_{r-1\to r} = \frac{\pi e^2 r}{3hm\nu_0}. \tag{80}$$

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[121] Van Vleck probably got the references to (Planck, 1921) from (Ladenburg and Reiche, 1923). Both (Van Vleck, 1924b, 339, note 12; 340, note 14) and (Ladenburg and Reiche, 1923, 588, note 19; 591, note 30) cite "equations (260) and (159)" and "section 158" in (Planck, 1921).

The Einstein analysis of A and B coefficients makes it clear that at the quantum level we must consider what Van Vleck (1924b, 340) calls the "differential absorption rate": the rate of energy absorption of the oscillator in state r going to state $r+1$ via (51) *minus* the stimulated emission induced by the ambient field and causing the transition r to $r-1$ (the B term in (50)). From eq. (80) we therefore have for the differential absorption rate of an oscillator in state r

$$\begin{aligned}\frac{dE_{\text{net}}}{dt} &= h\nu_0(B_{r\to r+1} - B_{r\to r-1})\rho(\nu_0)\\ &= h\nu_0(B_{r+1\to r} - B_{r\to r-1})\rho(\nu_0)\\ &= h\nu_0(r+1-r)\frac{\pi e^2}{3hm\nu_0}\rho(\nu_0)\\ &= \frac{\pi e^2}{3m}\rho(\nu_0), \qquad (81)\end{aligned}$$

which is precisely the classical result (75). Note that the dependence on the quantum state r (or classically, the amplitude of the motion D_r) has cancelled in the differential absorption rate, corresponding to the lack of coherence discussed previously between the transient and impressed motion.

Van Vleck derived this result in sec. 4 of his paper. He concluded:

> We thus see that in the limiting case of large quantum numbers, where [eq. (80)] is valid, the classical value [in eq. (75)] for the rate of absorption of energy is nothing but the differential rate of absorption in the quantum theory. This connection of the classical and quantum differential absorption we shall term the correspondence principle of absorption (Van Vleck, 1924b, 340).[122]

In sec. 5, he generalized the result to arbitrary non-degenerate multiply-periodic systems.

Van Vleck's correspondence principle for 'differential absorption' (i.e., the excess of absorption over stimulated emission) also clarifies the correspondence principle for dispersion. As Kramers (1924a,b) emphasized, the negative terms in the dispersion formula were difficult to account for on the basis of purely classical concepts—they somehow corresponded to a *negative* value for e^2/m for those virtual oscillators corresponding to transitions from the initial atomic level to lower energy levels. Similar negative contributions in the case of absorption are physically much more transparent: transitions to higher levels

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[122] Van Vleck points out that this "is a purely mathematical consequence of the correspondence principle for emission, which was used in deriving [eq. (80)]" (ibid.). A few pages later, Van Vleck (1924b, 343) notes that he could also have done the reverse, deriving the correspondence principle for emission from that for absorption.

result in a positive absorption of energy from the ambient electromagnetic field, whereas transitions to lower levels result in energy being returned to the field. The latter process was therefore known as "negative absorption" at the time, a term used by both Kramers (1924a, 676) and Van Vleck (1924b, 338). Noticing the greater physical transparency of his correspondence-principle results for absorption, and under the impression that Kramers' correspondence-principle arguments for the dispersion formula rested only on a treatment of harmonic oscillators, Van Vleck added sections on dispersion to his paper. Sec. 6, "The General Correspondence Principle Basis for Kramers Dispersion Formula," was added to the first quantum-theoretical part of the paper; sec. 15, "Computation of Polarization," to the classical part (see the letter from Van Vleck to Kramers of September 1924, quoted in sec. 2.2). Van Vleck was thus the first to publish a fully explicit derivation of the correspondence limit for polarization in the context of a general multiply-periodic system.

When Kuhn in his AHQP interview with Van Vleck brought up the paper on the correspondence principle for absorption, Van Vleck said: "I think that was one of my better papers." "How did you get into that?," Kuhn wanted to know. Van Vleck told him:

> Through a misunderstanding of something Gregory Breit [Van Vleck's colleague in Minnesota at the time] told me. He said that the net absorption was the difference between the fluctuations up and the fluctuations down, referred to some paper of—I think it was (Kretschmann)—but that was an entirely different thing. It was concerned with the fact that under certain phase relations the light did work on the atom and under certain phase relations the atom did work on the light. It was dealing essentially with statistical fluctuations. I misunderstood his remark and proceeded to try and get the differential effect between the absorption up from a given stationary state and a[b]sorption going down.[123]

The paper Breit was referring to is presumably (Kretschmann, 1921). In this paper, Erich Kretschmann (1887–1973), a student of Planck better known for his work in general relativity (Kretschmann, 1917), gave a purely classical discussion of the emission and absorption of radiation. What Van Vleck says here about this paper fits with its contents.

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[123] P. 22 of the transcript of the AHQP interview with Van Vleck on October 2, 1963. Van Vleck told this story in somewhat greater detail to Katherine Sopka (1988, 135, note 184, which, however, makes no mention of Kretschmann). He also explained to her why he acknowledged Breit in (Van Vleck, 1924a, 28) but not in (Van Vleck, 1924b,c): "As he [Van Vleck] remembers it, he wanted to thank Breit in the latter, but Breit objected on the ground that the phase fluctuations he had in mind were quite different from the difference effect employed by Van Vleck and so, overmodestly, felt no acknowledgment was in order."

Van Vleck's comments, however, are also very reminiscent of the following passage in (Ladenburg and Reiche, 1923):

> . . . according to Einstein's assumptions the effect of external radiation on a quantum atom corresponds to the effect a classical oscillator experiences from an incident wave. When the frequency of such a wave does not differ much or not at all from the characteristic frequency of the oscillator, the reaction of the oscillator consists in an increase or a decrease of its energy, depending on the difference in phase between the external wave and the motion of the oscillator. In analogy to this, Einstein assumes that the atom in state i has a probability characterized by the factor b_{ik} to make a transition to a higher state k under absorption of the energy $h\nu$ of the incident wave ("positive irradiation") and that the atom in state k has another probability (b_{ki}) to return to the state i under the influence of an external wave ("negative irradiation") (Ladenburg and Reiche, 1923, 586)

As we mentioned in sec. 2.1, Ladenburg and Reiche appealed to the correspondence principle to justify their quantum formulae for emission, absorption, and dispersion. Except in the case of emission, however, their arguments were fallacious. We conjecture that this is what inspired Van Vleck to use his expertise in techniques from celestial mechanics—the kind of expertise Ladenburg and Reiche clearly lacked—to derive expressions for emission, absorption, and dispersion merging with classical results in the sense of the correspondence principle. Van Vleck (1924b, 339, note 13; 344, note 21) cited Ladenburg and Reiche but gave no indication that their paper was the inspiration for his own. It is not implausible, however, that Van Vleck simply preferred to pass over their badly flawed calculations in silence rather than touting his own clearly superior results. As we mentioned in sec. 2.1, one of the problems with the "correspondence" arguments of Ladenburg and Reiche is that, following (Planck, 1921) and in the spirit of the derivation of the A and B coefficients in (Einstein, 1917), they focus on collections of atoms in thermal equilibrium rather than on individual atoms. What is suggestive of a possible influence of (Ladenburg and Reiche, 1923) on (Van Vleck, 1924b,c) is that the exact same passages of (Planck, 1921) are cited in both papers (see note 121 above) and that Van Vleck (1924b, 340, note 14) explicitly comments on the issue of many atoms in thermal equilibrium versus single atoms, noting that in Planck's discussion "no explicit mention is made of the asymptotic connection of the classical absorption and the differential absorption for a single orbit (where thermodynamic equilibrium need not be assumed) which is the primary concern of the present paper." The topic of a third installment that Van Vleck originally planned to add to his two-part paper also becomes understandable in light of our conjecture about the connection between (Van Vleck, 1924b,c) and (Ladenburg and Reiche, 1923). As Van Vleck explained in 1977 (see note 29 above): "Part III was to be concerned with the equilibrium between absorption and emission under the Rayleigh-Jeans law." If Ladenburg and Reiche did indeed stimulate Van Vleck's work, however, it is odd that he does not seem to have recognized that the virtual oscillators of BKS, which Van Vleck consistently attributed to Slater, are essentially just the *Ersatz*-oscillators of (Ladenburg and Reiche, 1923). Van Vleck, however, was hardly alone in associating virtual oscillators with BKS. We thus conclude that it is plausible that Van Vleck was inspired by (Ladenburg and Reiche, 1923) to formulate correspondence principles for emission, absorption, and dispersion. For one thing, this would explain why Van Vleck, who had not worked on radiation theory before, turned his attention to the interaction between matter and radiation.

4 Generalization to arbitrary non-degenerate multiply-periodic systems

4.1 The correspondence principle for absorption

The primary result of (Van Vleck, 1924b,c) was an extension of eq. (81) to an arbitrary non-degenerate multiply-periodic system of a single particle in three dimensions, and the demonstration that the quantum-differential absorption coincides with this more general result in the correspondence limit. Before giving Van Vleck's result we recall some basic features of multiply-periodic systems, which we shall in any event need in the next subsection when a completely explicit derivation (following, with minor notational changes, the one laid out by Van Vleck) of the corresponding formula for polarization will be provided.

The transition from one-dimensional periodic (and harmonic) systems such as the linear simple harmonic oscillator to three-dimensional multiply-periodic ones is fairly straightforward. Apart from the obvious need to introduce vector quantities, there are only two significant additional features. First, there is the appearance of multiple overtones in the general multiply-periodic expansion (so that the multiplicity variables in the analogue of eq. (28) take arbitrary positive and negative integral values, not just ±1). Second, the mechanical frequencies ν_1, ν_2, ν_3 (with $\nu_i = \partial H_0/\partial J_i$) of the separated coordinates are now in general functions of the amplitude of the classical path, which is to say, of the action variables J_i (with $i = 1, 2, 3$). We assume as before that the imposed electric field is in the X-direction so the x-coordinate of our electron is the relevant one for computing the induced coherent polarization, and in analogy to eq. (28) we now have

$$x(t) = \sum_{\vec{\tau}} A_{\vec{\tau}} e^{2\pi i \vec{\tau}\cdot\vec{w}}, \tag{82}$$

where in the absence of the external field the angle variables $\vec{w} = (w_1, w_2, w_3) = (\nu_1, \nu_2, \nu_3)t \equiv \vec{\nu}t$ and $\vec{\tau} = (\tau_1, \tau_2, \tau_3)$ with τ_i taking on all (positive and negative) integer values. It will be useful to write eq. (82) in an alternative purely real form, as a cosine expansion:

$$x(t) = \sum_{\vec{\tau},\vec{\tau}\cdot\vec{\nu}>0} X_{\vec{\tau}} \cos(2\pi\vec{\tau}\cdot\vec{\nu}t). \tag{83}$$

The complex amplitudes $A_{\vec{\tau}}$ satisfy the conjugacy condition $A_{\vec{\tau}} = A^*_{-\vec{\tau}}$ to ensure that $x(t)$ is real and we have the relation $X^2_{\vec{\tau}} = 4A_{\vec{\tau}}A_{-\vec{\tau}}$.[124]

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[124] Cf. eqs. (28)–(29) and note 115 in sec. 3.1.

As before (cf. eq. (27)), the full Hamiltonian has the form

$$H = H_0 + eEx(t)\cos 2\pi\nu t. \tag{84}$$

The subscripted mechanical frequencies ν_i with $i = 1, 2, 3$ (comprising the vector $\vec{\nu}$) must be distinguished from the single frequency ν (unsubscripted) corresponding to the applied field.

With these notations, Van Vleck's result for the absorption rate becomes (Van Vleck, 1924b, 342, eq. (16)):

$$\frac{dE_{\text{net}}}{dt} = \frac{2}{3}\pi^3 e^2 \left[\rho(\vec{\tau}\cdot\vec{\nu})\tau_k \frac{\partial G_\tau}{\partial J_k} + \rho'(\vec{\tau}\cdot\vec{\nu})G_\tau \tau_k \frac{\partial}{\partial J_k}(\vec{\tau}\cdot\vec{\nu})\right]. \tag{85}$$

where $\rho' \equiv \partial\rho/\partial\nu$ and where summation over $k = (1, 2, 3)$ is implied and where $G_\tau \equiv \vec{\tau}\cdot\vec{\nu}D^2_{\vec{\tau}}$ with $D^2_{\vec{\tau}} \equiv X^2_{\vec{\tau}} + Y^2_{\vec{\tau}} + Z^2_{\vec{\tau}}$. In the special case of the harmonic oscillator, the term with ρ', the derivative of the spectral function, vanishes as there is only a single mechanical frequency $\nu = \nu_0$, which is independent of the action variable J. In the first term, we get simply

$$\frac{dE_{\text{net}}}{dt} = \frac{2}{3}\pi^3 e^2 \rho(\nu_0)\frac{\partial}{\partial J}(\nu_0 D^2). \tag{86}$$

Using eq. (26), $D = \sqrt{J/m\pi\omega_0}$, for the amplitude, we recover the previous result, eq. (81).

Eq. (85) is the product of a highly nontrivial application of canonical perturbation techniques, where quantities of *second* order in the applied field need to be properly evaluated (cf. discussion following eq. (73) above). The polarization calculation presented in full in the next section only involves canonical perturbation theory to first order. For the absorption calculation, the variation in the action variables ΔJ_k in particular is needed to second order, and the integration of the result obtained for a monochromatic incident field needed to pass to the case of continuous radiation specified by an arbitrary spectral function $\rho(\nu)$ requires considerable care.

Slater also tried his hand at this calculation, as can be inferred from a letter from Kramers to Van Vleck of November 11, 1924. This is the reply to Van Vleck's letter of September 22, in which Van Vleck explained that he had added a discussion of dispersion to (Van Vleck, 1924b,c) unaware that Kramers had already obtained those same results but had not yet published them. This he only heard later from Slater (see sec. 2.2). Kramers wrote:

> Your note on absorption made me much pleasure and I think it very just of Providence that you got it published before hearing of our work. Slater had,

> on my request, made the same calculation, and he stated that the classical mean-absorption formula gave the right result in the limit of high quantum numbers. I did, however, not see his formula, and am not quite sure that he had not forgotten the term with $\partial\rho/\partial\nu$, without which the thing is not complete of course.

Van Vleck clearly remembered this point almost forty years later. Talking to Kuhn about his 1924 absorption papers, he mentioned: "I got the term in partial rho with respect to nu. I'm very proud of the fact that I picked that one up ... Slater, at Kramers' suggestion I guess, made a completely parallel calculation in Copenhagen which he never published." [125]

4.2 The correspondence principle for polarization

In this section we retrace the derivation given by Van Vleck (1924c) of the classical polarization formula for a general non-degenerate multiply-periodic system (with a single electron) in three dimensions. We remind the reader that this result is by no means new to Van Vleck, nor, for that matter, to Born or Kramers, who also produced derivations of the same result at around this time, using slightly different versions of canonical perturbation theory (cf. our comments at the end of sec. 3.1). The formula obtained is basically identical to a formula originally derived in celestial mechanics to compute the perturbation in the orbits of the inner planets due to the outer ones. As we saw in sec. 2.1, Epstein had been the first to use the relevant techniques from celestial mechanics in the context of the old quantum theory. As Van Vleck reminded Slater in a letter of December 15, 1924 (AHQP): "The classical formula analysis to the Kramer[s] formula appears to be first ca[lc]ulated by Epstein [1922c]."

The derivation is basically a straightforward generalization of the derivation of sec. 2.1 for the special case of a charged simple harmonic oscillator in an electromagnetic field (see eqs. (31)–(43)). The first-order perturbation in the coordinate $x(t)$ (the direction of the electric field in the incident electromagnetic wave) corresponding to the shifts $(\Delta J_l, \Delta w_l)$ in the action-angle variables is given by the three-dimensional version of eq. (31):

$$\Delta x = \frac{\partial x}{\partial J_l}\Delta J_l + \frac{\partial x}{\partial w_l}\Delta w_l. \tag{87}$$

As in sec. 2.1, we imagine that the external field is switched on at time zero, so that the shifts $(\Delta J_l,\ \Delta w_l)$ are the integrals of their time derivatives from 0

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[125] P. 22 of the transcript of the AHQP interview with Van Vleck on October 2, 1963.

to t. In analogy with eq. (36) and using eq. (82) for $x(t)$, we can immediately write down the equation for ΔJ_l to first order in E:

$$\Delta J_l = \int_0^t \dot{J}_l dt = \frac{eE}{2} \sum_{\vec{\tau}} \tau_l A_{\vec{\tau}} \left\{ \frac{1 - e^{2\pi i(\vec{\tau}\cdot\vec{\nu}+\nu)t}}{\vec{\tau}\cdot\vec{\nu}+\nu} + (\nu \to -\nu) \right\}. \tag{88}$$

All the terms inside the summation can be taken to zeroth order in the applied field. The computation of the first-order shifts Δw_l is a little more involved as new terms, not present in the harmonic-oscillator case, enter (cf. note 116). The Hamilton equation for $\dot{w}_l$ for the full Hamiltonian eq. (84) is (cf. eq. (37)):

$$\dot{w}_l = \nu_l + \frac{eE}{2} \sum_{\vec{\tau}} \frac{\partial A_{\vec{\tau}}}{\partial J_l} \left\{ e^{2\pi i(\vec{\tau}\cdot\vec{\nu}+\nu)t} + (\nu \to -\nu) \right\}. \tag{89}$$

Both terms in eq. (89) contribute to the first-order deviation $\Delta\dot{w}_l$ from the value of ν_l for the unperturbed system. Since ν_l depends on J_k, there will be a term $(\partial\nu_l/\partial J_k)\Delta J_k$ (cf. note 116). The second term is just the generalization of the corresponding term in eq. (37). Hence, we get:

$$\Delta\dot{w}_l = \frac{\partial \nu_l}{\partial J_k}\Delta J_k + \frac{eE}{2} \sum_{\vec{\tau}} \frac{\partial A_{\vec{\tau}}}{\partial J_l} \left\{ e^{2\pi i(\vec{\tau}\cdot\vec{\nu}+\nu)t} + (\nu \to -\nu) \right\}. \tag{90}$$

Upon substitution of eq. (88) for ΔJ_k this turns into

$$\begin{aligned}\Delta\dot{w}_l = {} & \frac{eE}{2} \sum_{\vec{\tau}} \left\{ \frac{\partial A_{\vec{\tau}}}{\partial J_l} e^{2\pi i(\vec{\tau}\cdot\vec{\nu}+\nu)t} + \tau_k \frac{\partial \nu_l}{\partial J_k} A_{\vec{\tau}} \frac{1 - e^{2\pi i(\vec{\tau}\cdot\vec{\nu}+\nu)t}}{\vec{\tau}\cdot\vec{\nu}+\nu} \right\} \\ & + \ (\nu \to -\nu).\end{aligned} \tag{91}$$

Integrating eq. (91), we find

$$\begin{aligned}\Delta w_l = {} & \frac{eE}{4\pi} \sum_{\vec{\tau}} \left\{ i \frac{\partial A_{\vec{\tau}}}{\partial J_l} \frac{1 - e^{2\pi i(\vec{\tau}\cdot\vec{\nu}+\nu)t}}{\vec{\tau}\cdot\vec{\nu}+\nu} \right. \\ & \left. + \tau_k \frac{\partial \nu_l}{\partial J_k} A_{\vec{\tau}} \frac{2\pi(\vec{\tau}\cdot\vec{\nu}+\nu)t - i(1 - e^{2\pi i(\vec{\tau}\cdot\vec{\nu}+\nu)t})}{(\vec{\tau}\cdot\vec{\nu}+\nu)^2} \right\} + (\nu \to -\nu).\end{aligned} \tag{92}$$

Inserting eq. (82) into eq. (87), we arrive at

$$\Delta x(t) = \sum_{\vec{\tau}'} \left(\frac{\partial A_{\vec{\tau}'}}{\partial J_l} \Delta J_l + 2\pi i A_{\vec{\tau}'} \tau'_l \Delta w_l \right) e^{2\pi i \vec{\tau}'\cdot\vec{\nu}t}. \tag{93}$$

Inserting eqs. (88) and (92) for ΔJ_l and Δw_l, respectively, into this expression, we obtain

$$\begin{aligned}\Delta x(t) = \frac{eE}{2}\sum_{\vec{\tau},\vec{\tau}'}\Bigg\{&\tau_l\frac{\partial A_{\vec{\tau}'}}{\partial J_l}A_{\vec{\tau}}\frac{1-e^{2\pi i(\vec{\tau}\cdot\vec{\nu}+\nu)t}}{\vec{\tau}\cdot\vec{\nu}+\nu} - \tau_l'\frac{\partial A_{\vec{\tau}}}{\partial J_l}A_{\vec{\tau}'}\frac{1-e^{2\pi i(\vec{\tau}\cdot\vec{\nu}+\nu)t}}{\vec{\tau}\cdot\vec{\nu}+\nu} \\ &+A_{\vec{\tau}}A_{\vec{\tau}'}\tau_k\frac{\partial \nu_l}{\partial J_k}\tau_l'\frac{2\pi i(\vec{\tau}\cdot\vec{\nu}+\nu)t+1-e^{2\pi i(\vec{\tau}\cdot\vec{\nu}+\nu)t}}{(\vec{\tau}\cdot\vec{\nu}+\nu)^2} \\ &+(\nu\rightarrow-\nu)\Bigg\}e^{2\pi i\vec{\tau}'\cdot\vec{\nu}t}. \qquad (94)\end{aligned}$$

As in sec. 2.1, we are only interested in the coherent contribution to the polarization, so we drop all terms in eq. (94) whose time dependence is not precisely $e^{\pm 2\pi i\nu t}$ and find, writing for convenience $\tau_k(\partial/\partial J_k) \equiv \vec{\tau}\cdot\vec{\nabla}_J$,

$$\begin{aligned}\Delta x_{\text{coh}} = \frac{eE}{2}\sum_{\vec{\tau}}\Bigg\{&-\vec{\tau}\cdot\vec{\nabla}_J(A_{\vec{\tau}}A_{-\vec{\tau}})\frac{e^{2\pi i\nu t}}{\vec{\tau}\cdot\vec{\nu}+\nu} \qquad (95) \\ &+\ A_{\vec{\tau}}A_{-\vec{\tau}}\vec{\tau}\cdot\vec{\nabla}_J(\vec{\tau}\cdot\vec{\nu})\frac{e^{2\pi i\nu t}}{(\vec{\tau}\cdot\vec{\nu}+\nu)^2}\Bigg\}\ +\ (\nu\rightarrow-\nu).\end{aligned}$$

Note that the coherent contribution derives from terms in which $\vec{\tau}' = -\vec{\tau}$, as otherwise the uncancelled overtones from the mechanical system would shift the spectral line (as in Raman scattering). Essentially the only additional physics of (Kramers and Heisenberg, 1925) in comparison to (Van Vleck, 1924b,c) is a detailed examination of such terms, predicted earlier by Smekal (1923). The terms in eq. (95) involving $\sin 2\pi\nu t$ vanish, as can be seen with the help of the identities

$$\begin{aligned}\sum_{\vec{\tau}}\tau_j\left(\frac{1}{\vec{\tau}\cdot\vec{\nu}+\nu}-\frac{1}{\vec{\tau}\cdot\vec{\nu}-\nu}\right)\cdot(\text{even function of }\vec{\tau}) &= 0 \\ \sum_{\vec{\tau}}\tau_j\tau_k\left(\frac{1}{(\vec{\tau}\cdot\vec{\nu}+\nu)^2}-\frac{1}{(\vec{\tau}\cdot\vec{\nu}-\nu)^2}\right)\cdot(\text{even function of }\vec{\tau}) &= 0.\end{aligned}$$

Thus eq. (95) simplifies to

$$\begin{aligned}\Delta x_{\text{coh}} &= -\frac{eE}{2}\cos 2\pi\nu t\sum_{\vec{\tau}}\left\{\vec{\tau}\cdot\vec{\nabla}_J(\frac{A_{\vec{\tau}}A_{-\vec{\tau}}}{\vec{\tau}\cdot\vec{\nu}+\nu})+(\nu\rightarrow-\nu)\right\} \\ &= -eE\cos 2\pi\nu t\sum_{\vec{\tau}}\vec{\tau}\cdot\vec{\nabla}_J\left(\frac{\vec{\tau}\cdot\vec{\nu}A_{\vec{\tau}}A_{-\vec{\tau}}}{(\vec{\tau}\cdot\vec{\nu})^2-\nu^2}\right). \qquad (96)\end{aligned}$$

With the replacement $X_{\vec{\tau}}^2 = 4A_{\vec{\tau}}A_{-\vec{\tau}}$, we may go over to the cosine form of the expansion in eq. (96) (cf. eqs. (82)–(83)), summing over only positive values of $\vec{\tau}\cdot\vec{\nu}$ (with a factor of 2):

$$\Delta x_{\mathrm{coh}} = -\frac{eE}{2}\cos 2\pi\nu t \sum_{\vec{\tau},\vec{\tau}\cdot\vec{\nu}>0} \vec{\tau}\cdot\vec{\nabla}_J \left(\frac{\vec{\tau}\cdot\vec{\nu}X_{\vec{\tau}}^2}{(\vec{\tau}\cdot\vec{\nu})^2-\nu^2}\right). \tag{97}$$

This is the generalization of eq. (43) for the harmonic oscillator.

Finally, we obtain the polarization by multiplying the displacement by N_r, the number of electrons per unit volume (the subscript r refers to the fact that we shall shortly consider only electrons in a particular quantum state r), and by $-e$ for the electron charge

$$P = N_r\frac{e^2}{2}E\cos 2\pi\nu t \sum_{\vec{\tau}\cdot\vec{\nu}>0} \vec{\tau}\cdot\vec{\nabla}_J \left(\frac{\vec{\tau}\cdot\vec{\nu}X_{\vec{\tau}}^2}{(\vec{\tau}\cdot\vec{\nu})^2-\nu^2}\right) \tag{98}$$

which is eq. (41) in(Van Vleck, 1924c; in Van Vleck's notation, $\vec{\tau}\cdot\vec{\nu}$ is written ω_τ) and equivalent to eq. (2*) in (Kramers, 1924b).

The equivalence of eq. (98) to the Kramers dispersion formula (58) in the correspondence limit is sketched in (Kramers, 1924b) and fully explained in sec. 6 of (Van Vleck, 1924b). Here we follow the latter. So we begin with eq. (58) for the polarization of a quantized system in state r, without the factor of 3 corresponding to the assumption that all oscillators be aligned with the applied field (rather than randomly in 3 dimensional space), and writing N_r instead of n_{osc}:

$$P_r = \frac{N_r c^3}{32\pi^4}E\cos 2\pi\nu t\left(\sum_s \frac{A_{s\to r}}{\nu_{sr}^2(\nu_{sr}^2-\nu^2)} - \sum_t \frac{A_{r\to t}}{\nu_{rt}^2(\nu_{rt}^2-\nu^2)}\right). \tag{99}$$

The sums over s (resp. t) refer to states higher (resp. lower) in energy than the fixed state r under consideration. In the correspondence limit, we take the state r to correspond to very high quantum numbers (n_1, n_2, n_3). The states s, t are associated to the central state r in symmetrical pairs:

$$\begin{aligned} &s \to (n_1+\tau_1, n_2+\tau_2, n_3+\tau_3),\\ &r \to (n_1, n_2, n_3),\\ &t \to (n_1-\tau_1, n_2-\tau_2, n_3-\tau_3), \end{aligned} \tag{100}$$

with $\vec{\tau}\cdot\vec{\nu} > 0$ so that the states s (resp. t) do indeed correspond to higher (resp. lower) energy states. Furthermore, we assume that $\vec{\tau}\cdot\vec{\nu} << \vec{n}\cdot\vec{\nu}$ so that the transitions $s \to r \to t$ correspond to very slight changes in the classical orbitals (and differences approximate well to derivatives). The Bohr-Sommerfeld quantization condition (1) associates action values $J_i = n_i h$ with a given quantized state, so the formal correspondence principle becomes (cf. eq. (48) in sec. 3.2):

$$\delta_{\vec{\tau}} F(\vec{n}) \equiv F(\vec{n}) - F(\vec{n} - \vec{\tau}) \to h\vec{\tau} \cdot \vec{\nabla}_J F. \tag{101}$$

In this notation, formula (99) the polarization can be written as

$$P_r = \frac{N_r c^3}{32\pi^4} E \cos 2\pi\nu t \sum_{\vec{\tau}} \delta_{\vec{\tau}} \left(\frac{A_{s\to r}}{\nu_{sr}^2(\nu_{sr}^2 - \nu^2)} \right), \tag{102}$$

with $A_{s\to r}$ given by Van Vleck's "correspondence principle for emission" (see eq. (76) and eq. (56))

$$A_{s\to r} = \frac{16\pi^4 e^2}{3hc^3} D_s^2 \nu_{sr}^3, \tag{103}$$

where $D_s^2 = (X_{\vec{\tau}}^{(s)})^2 + (Y_{\vec{\tau}}^{(s)})^2 + (Z_{\vec{\tau}}^{(s)})^2$ is the full vector amplitude squared for the Fourier component of the classical path responsible for the transition $\vec{n} + \vec{\tau} \to \vec{n}$. Substituting eqs. (101) and (103) into eq. (102) and replacing the difference frequency ν_{sr} by its classical counterpart $\vec{\tau} \cdot \vec{\nu}$, we obtain, :

$$\begin{aligned} P_r &= N_r E \cos 2\pi\nu t \frac{c^3}{32\pi^4} \frac{16\pi^4 e^2}{3hc^3} h \sum_{\vec{\tau}\cdot\vec{\nu}>0} \vec{\tau} \cdot \vec{\nabla}_J \left(\frac{\vec{\tau}\cdot\vec{\nu} D_s^2}{(\vec{\tau}\cdot\vec{\nu})^2 - \nu^2} \right) \\ &= N_r \frac{e^2}{2} E \cos 2\pi\nu t \sum_{\vec{\tau}\cdot\vec{\nu}>0} \vec{\tau} \cdot \vec{\nabla}_J \left(\frac{\vec{\tau}\cdot\vec{\nu} \frac{1}{3} D_s^2}{(\vec{\tau}\cdot\vec{\nu})^2 - \nu^2} \right). \end{aligned} \tag{104}$$

With the replacement $\frac{1}{3}D_s^2 \to X_{\vec{\tau}}^2$ appropriate for randomly oriented atoms, eq. (104) becomes identical to the classical formula (98). This shows that the Kramers dispersion formula (99) does indeed merge with the classical result in the limit of high quantum numbers, as Van Vleck set out to demonstrate.

5 Derivation of the formulae for dispersion, emission, and absorption in modern quantum mechanics

Describing the impact of the new quantum mechanics on dispersion theory, Van Vleck wrote in 1928:

> Dispersion was particularly bothersome in the old quantum theory, which could never explain why the resonance frequencies in dispersion were experimentally the spectroscopic frequencies given by the Bohr frequency condition rather than the altogether different frequencies of motion in orbits constituting the stationary states [cf. our discussion in sec. 2.1]. The new mechanics, however, yields the Kramers dispersion formula, previously derived semi-empirically from the correspondence principle . . . As the result of the masterful treatment by Dirac [1927], a mechanism has at last to a certain extent been found for the previously so mysterious quantum jumps between stationary states . . . Dirac's work brings out nicely the parallelism between matter and radiation, and their corpuscular and wave aspects, which are complementary rather than contradictory (Van Vleck, 1928, 494–495).

In this paper, we will not follow the historical developments post-*Umdeutung*. Instead we present in this section our own versions of the derivations of the quantum formulae for dispersion (sec. 5.1), (spontaneous) emission (sec. 5.2), and absorption (sec. 5.3) in modern quantum theory. Seeing how modern quantum mechanics sanctions the formulae found by Kramers, Van Vleck and others in the old quantum theory on the basis of Einstein's quantum theory of radiation and Bohr's correspondence principle will illuminate various aspects of the relation between the old and the new theory.

First, we show how the orchestra of virtual oscillators of pre-*Umdeutung* dispersion theory survives in the guise of a sum over matrix elements of the position operator. Second, we show how the diagonal matrix elements of the fundamental commutation relation for position and momentum, $[X, P] = i\hbar$, are given by the high-frequency limit of the Kramers dispersion formula, a formula known as the Thomas-Kuhn(-Reiche) sum rule (Thomas, 1925; Kuhn, 1925; Reiche and Thomas, 1925). This formula replaces the Bohr-Sommerfeld condition as the fundamental quantization condition in the *Umdeutung* paper. Heisenberg obtained the sum rule by applying the procedure introduced in the *Umdeutung* paper for translating classical quantities into quantum-theoretical ones to (a derivative of) the Bohr-Sommerfeld quantization condition. He then showed that the sum rule also obtains by comparing the high-frequency limit of the Kramers dispersion formula with the polarization of a charged harmonic oscillator in the limit where $\nu >> \nu_0$ (see our eq. (47)). In hindsight, we can see clearly in the *Umdeutung* paper how close Heisenberg came to recognizing the presence of the commutation relation between position and momentum in the sum rule serving as his quantization condition. What probably prevented him from seeing this was an inconvenient division of the sum over quantum states in the sum rule, which, though very natural from the point of dispersion theory, obscured its connection to a commutator.

It will also become clear in the course of our modern derivation that the Kramers dispersion formula is an even more general result in modern quantum mechanics than it was in the old quantum theory. In the old quantum theory, it held for any non-degenerate multiply-periodic system with an unperturbed Hamiltonian such that the unperturbed motion can be solved in action-angle variables. In modern quantum mechanics, the results holds for any system with a Hermitian Hamilton operator such that the unperturbed part has a spectrum that is at least partially discrete. This helps to explain why the Kramers dispersion formula carries over completely intact from the old quantum theory to modern quantum mechanics.

5.1 Dispersion

In this subsection, we derive the Kramers dispersion formula in time-dependent perturbation theory. We then examine the high-frequency limit of this formula and discuss the role it played in (Heisenberg, 1925c) paper as the fundamental quantization condition replacing the Bohr-Sommerfeld condition.

We consider a quantized charged system (valence electron) with states labeled by discrete indices $r, s, t, ...$, and with the Hamilton operator

$$H = H_0 + V(t) = H_0 + eEx\cos\omega t. \tag{105}$$

We want to calculate the first-order perturbation (in the electric field E) in the expectation value of the electron position in a particular state $|r, t>$. It is convenient to work in the interaction picture.[126] The state $|r, t>_{\text{int}}$ in the interaction picture is related to the state $|r, t>$ in the Schrödinger picture via:

$$|r, t>_{\text{int}} \equiv e^{iH_0 t/\hbar}|r, t> . \tag{106}$$

An operator $O_{\text{int}}(t)$ in the interaction picture is related to the corresponding

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[126] The special role of H_0 in the time dependence of states and operators in the interaction picture is analogous to the choice of action-angle variables for the free rather than the full Hamiltonian in the version of canonical perturbation theory used by Van Vleck. This is what lies behind the close similarities between the calculations in this section and those in secs. 3.1 and 4.2.

operator O in the Schrödinger picture via

$$O_{\rm int}(t) \equiv e^{iH_0t/\hbar} O e^{-iH_0t/\hbar}. \tag{107}$$

It follows that expectation values are the same in the two pictures:

$$_{\rm int}\!< r,t|O_{\rm int}(t)|r,t>_{\rm int} = < r,t|O|r,t> . \tag{108}$$

The evolution of the states in the interaction picture is given by:

$$\begin{aligned}\frac{\partial}{\partial t}|r,t>_{\rm int} &= \frac{i}{\hbar} e^{iH_0t/\hbar} H_0|r,t> + e^{iH_0t/\hbar}\frac{\partial}{\partial t}|r,t> \\ &= \frac{i}{\hbar} e^{iH_0t/\hbar}\left(H_0 - H\right)|r,t>,\end{aligned} \tag{109}$$

where in the last step, we used the Schrödinger equation

$$\frac{\partial}{\partial t}|r,t> = -\frac{iH}{\hbar}|r,t> . \tag{110}$$

Since $H_0 - H = -V(t)$ (see eq. (105)), we can write eq. (109) as:

$$\begin{aligned}\frac{\partial}{\partial t}|r,t>_{\rm int} &= -\frac{i}{\hbar} e^{iH_0t/\hbar} V(t) e^{-iH_0t/\hbar}|r,t>_{\rm int} \\ &= -\frac{i}{\hbar} V_{\rm int}(t)|r,t>_{\rm int},\end{aligned} \tag{111}$$

where we used eqs. (106)–(107). To first order in $V_{\rm int}(t)$ (i.e., to first order in the field E), the solution of (111) is

$$\begin{aligned}|r,t>_{\rm int} &= |r,0>_{\rm int} - \frac{i}{\hbar}\int_0^t d\tau V_{\rm int}(\tau)|r,0>_{\rm int} \\ &= |r,0>_{\rm int} - \frac{ieE}{\hbar}\int_0^t d\tau x_{\rm int}(\tau)\cos\omega\tau|r,0>_{\rm int} .\end{aligned} \tag{112}$$

At $t = 0$ the states (and operators) in the interaction picture coincide with those in the Schrödinger picture. From now on we thus simply write $|r>$ for $|r,0>_{\rm int}$. The dual ('bra') of the vector ('ket') in eq. (112) is:

$$_{\rm int}\!< r,t| = < r| + \frac{ieE}{\hbar}\int_0^t d\tau\cos\omega\tau < r|x_{\rm int}(\tau). \tag{113}$$

To find the dipole moment $P_r(t)$ of the system in state r to first order in E, we calculate the first-order contribution to the expectation value of the displacement $<\Delta x>_r$ in the state r induced by the field E:

$$<\Delta x>_r \equiv {}_{\rm int}\!< r,t|x_{\rm int}(t)|r,t>_{\rm int} - < r|x_{\rm int}(t)|r> . \tag{114}$$

Inserting eqs. (112)–(113) into this expression, we find:

$$<\Delta x>_r = \frac{ieE}{\hbar}\int\limits_0^t d\tau < r|\left\{x_{\rm int}(\tau)x_{\rm int}(t) - x_{\rm int}(t)x_{\rm int}(\tau)\right\}|r> \cos\omega\tau . \tag{115}$$

Writing $\cos\omega\tau = \frac{1}{2}(e^{i\omega\tau} + e^{-i\omega\tau})$, and inserting a complete set of eigenstates of the unperturbed Hamiltonian H_0 $(1 = \sum_s |s><s|)$ between the two coordinate operators, we obtain

$$\begin{aligned}
<\Delta x>_r = {} & \frac{ieE}{2\hbar}\sum_s \int\limits_0^t d\tau \Big(< r|e^{iH_0\tau/\hbar} x e^{-iH_0\tau/\hbar}|s>< s|e^{iH_0t/\hbar} x e^{-iH_0t/\hbar}|r> \\
& - < r|e^{iH_0t/\hbar} x e^{-iH_0t/\hbar}|s>< s|e^{iH_0\tau/\hbar} x e^{-iH_0\tau/\hbar}|r> \Big) e^{i\omega\tau} \\
& + \quad (\omega \to -\omega) \\
= {} & \frac{ieE}{2\hbar}\sum_s \int\limits_0^t d\tau \Big(e^{i(E_r - E_s + \hbar\omega)\tau/\hbar} e^{i(E_s - E_r)t/\hbar} \\
& - e^{i(E_r - E_s)t/\hbar} e^{i(E_s - E_r + \hbar\omega)\tau/\hbar} \Big) < r|x|s>< s|x|r> \qquad (116) \\
& + \quad (\omega \to -\omega).
\end{aligned}$$

We introduce the notation $X_{rs} \equiv < r|x|s>$ for the matrix elements of the coordinate operator. Note that these matrix elements in eq. (116) are accompanied by time-development phases $e^{i(E_r - E_s)t/\hbar}$ of *purely harmonic form*: they are the precise correlates in modern quantum mechanics of the *Ersatz*-oscillators of Ladenburg and Reiche (1923) or, equivalently, the virtual oscillators of BKS, as was clearly realized, for instance, by Landé (1926).

Performing the time integral in eq. (116), we find

$$\begin{aligned}
<\Delta x>_r = {} & \frac{eE}{2}\sum_s \Bigg[\frac{e^{i(E_r - E_s + \hbar\omega)t/\hbar} - 1}{E_r - E_s + \hbar\omega} e^{i(E_s - E_r)t/\hbar} \\
& - \frac{e^{i(E_s - E_r + \hbar\omega)t/\hbar} - 1}{E_s - E_r + \hbar\omega} e^{i(E_r - E_s)t/\hbar} \Bigg] X_{rs}X_{sr} \qquad (117) \\
& + \quad (\omega \to -\omega).
\end{aligned}$$

(cf. eqs. (36) and (38) in sec. 3.1 and eqs. (88) and (92) in sec. 4.2). The coherent terms in $< \Delta x >_r$, i.e. the terms with a time-dependence $e^{\pm i\omega t}$ (cf. eq. (40) in sec. 3.1 and eq. (95) in sec. 4.2), are:

$$< \Delta x_{\rm coh} >_r = \frac{eE}{2} \sum_s X_{rs} X_{sr} e^{i\omega t} \left[\frac{1}{E_r - E_s + \hbar\omega} - \frac{1}{E_s - E_r + \hbar\omega} \right] \tag{118}$$
$$+ \quad (\omega \to -\omega).$$

Using the Bohr frequency condition $\hbar\omega_{rs} = E_r - E_s$, we can write the expression in square brackets in eq. (118) as:

$$\frac{1}{\hbar\omega_{rs} + \hbar\omega} - \frac{1}{\hbar\omega_{sr} + \hbar\omega} = \frac{2\omega_{rs}}{\hbar(\omega_{rs}^2 - \omega^2)}. \tag{119}$$

Inserting this result into eq. (118) and noting that the terms proportional to $\sin \omega t$ vanish, we find the following result for the dipole moment of the system in state r (cf. eq. (6) or eq. (47))

$$P_r(t) = -e < \Delta x_{\rm coh} >_r = \frac{2e^2 E}{\hbar} \sum_s \frac{\omega_{sr} X_{rs} X_{sr}}{\omega_{sr}^2 - \omega^2} \cos \omega t. \tag{120}$$

The sum over s can naturally be separated into states s of higher energy than r, with $\omega_{sr} > 0$, and states t of lower energy, with $\omega_{rt} > 0$ ($\omega_{rt} = 0$ for $r = t$):

$$P_r = \frac{2e^2 E}{\hbar} \left(\sum_s \frac{\omega_{sr} X_{sr} X_{rs}}{\omega_{sr}^2 - \omega^2} - \sum_t \frac{\omega_{rt} X_{rt} X_{tr}}{\omega_{rt}^2 - \omega^2} \right) \cos \omega t. \tag{121}$$

If we recall the correspondence principle for emission (76), and identify D_s^2 with $3(X_\tau^s)^2 = 12 A_\tau A_{-\tau}$ and the Fourier coefficients $A_\tau \to X_{sr}$, $A_{-\tau} \to X_{rs}$ we get

$$A_{s\to r} = \frac{64\pi^4 e^2}{hc^3} \nu_{sr}^3 X_{sr} X_{rs}, \tag{122}$$

whence we recover the original form (58) of the dispersion formula

$$P_r = \frac{c^3}{32\pi^4} E \cos \omega t \left(\sum_s \frac{A_{s\to r}}{\nu_{sr}^2(\nu_{sr}^2 - \nu^2)} - \sum_t \frac{A_{r\to t}}{\nu_{rt}^2(\nu_{rt}^2 - \nu^2)} \right). \tag{123}$$

Of course, the above identification of classical Fourier components with matrix elements of the position operator is at the core of Heisenberg's 1925 breakthrough.

Returning for a moment to eq. (121), we see that in the Thomson limit where the frequency of incident radiation far exceeds the difference frequencies ω_{rs} for the electron states $r, s,$[127] the polarization P_r becomes asymptotically

$$P_r \simeq -\frac{2e^2E}{\hbar\omega^2}(\sum_s \omega_{sr}X_{sr}X_{rs} - \sum_t \omega_{rt}X_{rt}X_{tr})\cos\omega t. \tag{124}$$

The preceding equation is in content identical with the next to last (unnumbered) equation in sec. 2 of (Heisenberg, 1925c), where the Kramers dispersion theory is explicitly invoked. For large frequencies, we expect the polarization to approach our previously derived result (see eq. (6) or eq. (47)) for the polarization of a charged harmonic oscillator in the limit where $\nu >> \nu_0$:[128]

$$P_r = -\frac{e^2E}{m\omega^2}\cos\omega t, \tag{125}$$

Comparing eq. (124) with eq. (125) we find eq. (16) in (Heisenberg, 1925c):

$$h = 4\pi m(\sum_s \omega_{sr}X_{sr}X_{rs} - \sum_t \omega_{rt}X_{rt}X_{tr}). \tag{126}$$

This result is first obtained by Heisenberg from the Bohr-Sommerfeld quantization condition by applying the quantum-theoretical transcription procedure, which was introduced in sec. 1 of the *Umdeutung* paper and had been inspired by dispersion theory. It replaces the Bohr-Sommerfeld condition as the fundamental quantization constraint in Heisenberg's new theory. That the same result can be obtained directly from the high-frequency limit of the Kramers dispersion formula is clearly regarded by Heisenberg as strong evidence for the validity of his transcription procedure. Using eq. (126), together with the formal transcription of the classical equation of motion, $\ddot{x} + f(x) = 0$ (eq. (11) of the *Umdeutung* paper), Heisenberg asserts the possibility of "a complete determination not only of frequencies and energy values, but also of quantum-theoretical transition probabilities" (Heisenberg, 1925c, 268). As Heisenberg points out, eq. (126) is completely equivalent to the sum rules for oscillator strengths derived by Thomas (1925) and Kuhn (1925).[129]

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[127] Or, alternatively, when the incident photon energy far exceeds the energy needed to ionize the electron, so that the latter can be regarded as essentially a free, unbound particle

[128] This result is obtained in (Kuhn, 1925) by equating the energy scattered by an electron in the Thomson limit to the radiation emitted by an oscillating dipole according to the Larmor formula.

[129] Heisenberg's logic is slightly different from ours. Instead of pointing out that the high-frequency limit (124) of the Kramers dispersion formula and the well-established classical result (125) imply Heisenberg's quantization condition (126),

The realization that eq. (126) is equivalent to (the diagonal matrix elements of) the fundamental commutator relation $[P, X] = \hbar/i$ of modern quantum theory came shortly after this, in the work of Born and Jordan (1925b). The recognition of eq. (126) as a commutator is mathematically obscured by the separation of the sum into states higher (s) and lower (t) than the given state r—a separation which is very natural given the history of the Kramers dispersion formula. If Heisenberg had applied his own transcription rules for associating classical variables with quantum two-index quantities to the momentum $P \equiv m\dot{X}$ in the unnumbered equation immediately following (13) in the *Umdeutung* paper (Heisenberg, 1925c, 267), he would have found (using modern matrix notation)[130]

$$P_{rs} = im\omega_{rs}X_{rs}. \tag{127}$$

Rewriting eq. (126) as a *single* sum over all states s, but splitting the sum into two equal pieces via the identity $2\omega_{sr} = \omega_{sr} - \omega_{rs}$, we find

$$\begin{aligned} h &= 4\pi m \sum_s \omega_{sr} X_{rs} X_{sr} \\ &= 2\pi m \sum_s (X_{rs}\omega_{sr}X_{sr} - \omega_{rs}X_{rs}X_{sr}) \\ &= -2\pi i \sum_s (X_{rs}P_{sr} - P_{rs}X_{sr}), \end{aligned} \tag{128}$$

where in the last step we used eq. (127). In modern notation, this last expression is immediately recognized as the diagonal matrix element of the fundamental commutator $[X, P] = i\hbar$:

$$i\frac{h}{2\pi} = < r|XP - PX|r >$$

Heisenberg (1925c, 269–270) points out that eqs. (126) and (124) imply eq. (125). This is only a cosmetic difference. The point of the exercise is still to show that the new quantization condition, found through *Umdeutung* of the derivative of the Bohr-Sommerfeld condition, follows from well-established results in Kramers' dispersion theory and classical electrodynamics. We are nonetheless grateful to Christoph Lehner for alerting us to this point.

[130] Following Heisenberg's procedure in the *Umdeutung* paper for translating classical equations into quantum-mechanical ones, we would translate his classical equation for momentum, $m\dot{x} = m\sum_\alpha a_\alpha(n) i\alpha\omega_n e^{i\alpha\omega_n t}$, into the following quantum-mechanical equation: $P(n, n+\alpha) = ima(n, n+\alpha)\omega(n, n+\alpha)$. In modern notation, this becomes: $P_{rs} = imX_{rs}\omega_{rs}$ (no summation). That Heisenberg did not write down this equation is probably because he was thinking in terms of the Lagrange formalism (with q-s and $\dot{q}$-s) and not in terms of the Hamilton formalism (with q-s and p-s).

$$=\sum_s (< r|X|s >< s|P|r > - < r|P|s >< s|P|r >). \tag{129}$$

Although Heisenberg recognized the significance of the noncommutativity of quantum-theoretic quantities in his formalism (see the last three paragraphs of section 1), the simplicity of $x(t)p(t) - p(t)x(t)$ implied by his fundamental quantization relation (126) eluded him, probably because the origin of that relation in dispersion theory prevented him from rewriting the summations the way we did in eq. (128).

5.2 Spontaneous emission

To begin with, we note that we are dealing throughout with the dipole approximation, which is implicit in the 1924 work, corresponding to the regime where the wavelength of light is much larger than atomic dimensions (or equivalently, where photon momentum is much smaller than electron momentum). Once again, note that the notation of (Van Vleck, 1924b, eq. (1)),

$$\begin{aligned} x &= \sum_{\tau_1\tau_2\tau_3} X(\tau_1,\tau_2,\tau_3)\cos\{2\pi(\tau_1\omega_1+\tau_2\omega_2+\tau_3\omega_3)t+\ldots\} \\ &= \sum \left\{ \frac{1}{2}X(\tau_1,\tau_2,\tau_3)e^{+2\pi i(\tau_1\omega_1+\tau_2\omega_2+\tau_3\omega_3)t+\ldots} \right. \\ &\qquad\qquad \left. +\frac{1}{2}X(\tau_1,\tau_2,\tau_3)e^{-2\pi i(\tau_1\omega_1+\tau_2\omega_2+\tau_3\omega_3)t+\ldots} \right\}, \end{aligned} \tag{130}$$

implies that van Vleck's $D^2 = X^2 + Y^2 + Z^2$ (Van Vleck, 1924b, line following eq. (8)) corresponds to four times the square of the matrix element of the quantum position operator appearing in the dipole transition formulas of modern quantum mechanics. For the latter we shall follow the treatment of (Baym, 1969, Ch. 13).

In the dipole approximation, the spontaneously emitted power per unit solid angle is given by (Baym, 1969, 282, eq. 13–100), for emitted light of polarization vector $\vec{\lambda}$, in a transition from state r to state s:

$$\begin{aligned} \frac{dP}{d\Omega} &= \frac{\omega^4 e^2}{2\pi c^3} < r|\vec{\lambda}\cdot\vec{x}|s >< s|\vec{\lambda}\cdot\vec{x}|r > \\ &= \sum_{i,j=1}^{3} \frac{\omega^4 e^2}{2\pi c^3}\lambda_i\lambda_j < r|x_i|s >< s|x_j|r > . \end{aligned} \tag{131}$$

Here (unlike Baym) we take real polarization vectors $\vec{\lambda}$ (plane polarized) rather than complex (circularly polarized) ones as our basis. We want the total spontaneously emitted power in any event, summed over the two possible polarizations for any momentum vector $\vec{k}$ of the emitted photon (so the basis of photon states is irrelevant). This requires the polarization sum

$$\sum_{\lambda=1}^{2} \lambda_i \lambda_j = \delta_{ij} - \hat{k}_i \hat{k}_j, \quad (i, j = 1, 2, 3), \tag{132}$$

which follows from the fact that the two polarization vectors are any pair of orthogonal unit vectors perpendicular to the unit vector $\hat{k}$ along the photon direction. Finally, we want the total power emitted in any direction, so the polarization sum (132) must be integrated over all solid angles:

$$\int d\Omega_{\hat{k}} (\delta_{ij} - \hat{k}_i \hat{k}_j) = 4\pi (\frac{2}{3} \delta_{ij}). \tag{133}$$

The Einstein coefficient $A_{r \to s}$ in (Van Vleck, 1924b, eqs. (5) and (9)) refers to a rate of photon emission (not energy emission) so we must divide eq. (131) by $\hbar\omega$. Putting together the above results (and switching to $\nu = \omega/2\pi$), we find:

$$A_{r \to s} = \frac{1}{\hbar\omega} \int d\Omega_{\hat{k}} \frac{dP}{d\Omega_{\hat{k}}} = \frac{\omega^4 e^2}{2\pi\hbar\omega c^3} \frac{8\pi}{3} \sum_i < r|x_i|s >< s|x_i|r > . \tag{134}$$

Using the notation $X_{rs} \equiv < r|x|s >$, etc. for the matrix elements of position introduced above we can rewrite this as:

$$A_{r \to s} = \frac{\omega^4 e^2}{2\pi\hbar\omega c^3} \frac{8\pi}{3} \left(|X_{rs}|^2 + |Y_{rs}|^2 + |Z_{rs}|^2 \right). \tag{135}$$

Replacing the matrix elements X_{rs}, Y_{rs}, and Z_{rs} by the amplitude D_r in the correspondence limit as indicated in the preceding section (cf. the remarks preceding eq. (122)) and substituting $\omega = 2\pi\nu$, we arrive at:

$$A_{r \to s} = \frac{16\pi^4 e^2 \nu^3}{3hc^3} D_r^2. \tag{136}$$

D_r^2 is the amplitude defined by (Van Vleck, 1924b) immediately following eq. (8), to be replaced by $D_r(\tau_1, \tau_2, \tau_3)^2$ in eq. (9), with which eq. (136) is seen to be identical.

5.3 Absorption

The Einstein formula for absorption (Van Vleck, 1924b, eq. (6)), when combined with the stimulated emission ("negative absorption") term to yield (ibid., eq. 15)), leads directly to the correspondence limit result (ibid., eq. (16)). Here, we check the identity of eq. (15) in (Van Vleck, 1924b) (more precisely, the unnumbered equation immediately following this one) with the modern absorption calculation given in (Baym, 1969). For the rate of absorption of light leading to a transition from state s to (higher) state r, (Baym, 1969, eq. 13–40) reads (in dipole approximation, $\vec{j}_{\vec{k}} \to \vec{p}/m$):

$$\Gamma^{\rm abs}_{s\to r} = \frac{2\pi e^2}{\hbar^2 c^2}\frac{\omega^2}{(2\pi c)^3}\int d\Omega_{\hat{k}} \sum_{\lambda} < s|\vec{\lambda}\cdot\frac{\vec{p}}{m}|r >< r|\vec{\lambda}\cdot\frac{\vec{p}}{m}|s > |A_{\vec{k}\vec{\lambda}}|^2. \quad (137)$$

As usual, in dipole approximation we can use (Baym, 1969, eq. 13–98) to replace matrix elements of the momentum operator with those of the coordinate operator (using the equations of motion). For Hamiltonians of the form $H = (\vec{p}^2/2m) + V(\vec{x})$,

$$[H, x_j] = \frac{1}{2m}[p_i p_i, x_j] = \frac{1}{m}p_i[p_i, x_j] = \frac{p_i}{m}\frac{\hbar}{i}\delta_{ij} = \frac{\hbar}{i}\frac{p_j}{m}, \quad (138)$$

whence

$$\begin{aligned} < r|\frac{\vec{p}}{m}|s > &= \frac{i}{\hbar} < r|[H,\vec{x}]|s > \\ &= \frac{i}{\hbar}(E_r - E_s) < r|\vec{x}|s > \\ &= i\omega < r|\vec{x}|s >, \end{aligned} \quad (139)$$

where $\hbar\omega = E_r - E_s$. Once again, in eq. (139), we see the "monstrous" difference frequencies characteristic of quantum theory, which wreaked havoc on classical interpretations of radiation phenomena, making their appearance in the modern formalism. Accordingly, eq. (137) becomes

$$\Gamma^{\rm abs}_{s\to r} = \frac{2\pi e^2}{\hbar^2 c^2}\frac{\omega^4}{(2\pi c)^3}\int d\Omega_{\hat{k}} \sum_{\lambda} < s|\lambda_i x_i|r >< r|\lambda_j x_j|s > |A_{\vec{k}\vec{\lambda}}|^2. \quad (140)$$

Now we are going to assume that the ambient light is unpolarized and isotropic so that the squared amplitude $|A_{\vec{k}\vec{\lambda}}|^2$ is in fact independent of $\lambda, \hat{k}$, and the only angular dependence comes in via the polarization vectors. The angle average of the polarization sum in eq. (140) can then be performed as in eq. (133) to yield

$$\Gamma^{\rm abs}_{s\to r} = \frac{4\pi e^2}{3\hbar^2 c^2}\frac{\omega^4}{(2\pi c)^3} < s|x_i|r >< r|x_i|s > \int d\Omega_{\hat{k}}|A_{\vec{k}\vec{\lambda}}|^2. \quad (141)$$

Next, we need to establish the relation between the squared mode amplitudes $|A_{\vec{k}\vec{\lambda}}|^2$ and the specific energy density function $\rho(\nu)$ defined as the energy per unit volume per unit frequency interval. The mode amplitudes $A_{\vec{k}\vec{\lambda}}$ correspond to discrete modes for electromagnetic radiation in a box of volume V, with each mode contributing energy density

$$\frac{1}{V}|A_{\vec{k}\vec{\lambda}}|^2\frac{\omega}{2\pi c^2} \tag{142}$$

(Baym, 1969, eq. 13–14). As the box volume goes to infinity we have the usual correspondence

$$\frac{1}{V}\sum_k \to \int \frac{k^2 dk d\Omega_{\hat{k}}}{(2\pi)^3}, \tag{143}$$

so that the total energy density *between frequency ν and frequency $\nu + \Delta\nu$* is

$$\begin{aligned}\rho(\nu)\Delta\nu &= \frac{1}{V}\sum_{2\pi\nu<kc<2\pi(\nu+\Delta\nu)} 2|A_{\vec{k}\vec{\lambda}}|^2\frac{\omega^2}{2\pi c^2}\\ &\to \frac{1}{(2\pi)^3}\int d\Omega_{\hat{k}} \int_{2\pi\nu/c}^{2\pi(\nu+\Delta\nu)/c} dk\;\; k^2\frac{\omega^2}{2\pi c^2}2|A_{\vec{k}\vec{\lambda}}|^2. \end{aligned}\tag{144}$$

Note that although we continue to write the mode amplitudes $A_{\vec{k}\vec{\lambda}}$ as depending on polarization and momentum vector of the photon, we are really assuming that there is no dependence on the polarization or photon *direction*. Hence the factor of 2, with no remaining sum over λ. Eq. (144) gives

$$\rho(\nu)\Delta\nu = \frac{1}{(2\pi)^3}\frac{2\pi}{c}k^2\frac{\omega^2}{2\pi c^2}2\int d\Omega_{\hat{k}}|A_{\vec{k}\vec{\lambda}}|^2\Delta\nu, \tag{145}$$

or, equivalently

$$\int d\Omega_{\hat{k}}|A_{\vec{k}\vec{\lambda}}|^2 = \frac{4\pi^3c^5}{\omega^4}\rho(\nu). \tag{146}$$

Inserting eq. (146) into eq. (141) and multiplying by $\hbar\omega$ to get the rate of energy absorption (instead of the number rate of photon absorption) we find, using the usual association of squares of matrix elements of the position operator to the classical orbit amplitude $\frac{1}{4}D_r^2$,

$$\hbar\omega\Gamma^{\rm abs}_{s\to r} = \frac{4\pi e^2\omega}{3\hbar c^2}\frac{\omega^4}{(2\pi c)^3}\frac{4\pi^3c^5}{\omega^4}\rho(\nu)\frac{1}{4}D_r^2$$

$$= \frac{2\pi^3 e^2}{3h} \nu \rho(\nu) D_r^2, \tag{147}$$

which coincides with the first term in van Vleck's equation (Van Vleck, 1924b, following eq. (15)) for the part of the total absorption rate due to upward transitions. Of course, the second (negative absorption, or stimulated emission) term is of exactly the same form (with a minus sign) due to the symmetry of the Einstein B coefficients.

6 Conclusion

Our study of Van Vleck's two-part paper on the application of the correspondence principle to the interaction of matter and radiation (Van Vleck, 1924b,c) has led us to consider three clusters of questions. First, there are questions about the paper itself. What made Van Vleck decide to work in this area? He had not published on radiation theory before. And—as one is inevitably tempted to ask—why did Van Vleck not take the next step and arrive at something like matrix mechanics? That gets us to the second cluster of questions, about the developments in quantum theory that provide the natural context for Van Vleck's work, especially the transition of the old quantum theory of Bohr and Sommerfeld to matrix mechanics. What was important for this development and what was not? The third group of questions concerns the relative importance of American contributions to these developments. In this final section we collect the (partial) answers we have found to these biographical, conceptual, and sociological questions.

Let us first dispose of the issue of American contributions to early quantum theory. Since we focused on the work of only two individuals, Van Vleck and Slater, we are in no position to draw strong conclusions. Still, it seems safe to say that our study supports the thesis of Sam Schweber (1986) and others that, by the early 1920s, the United States had a homegrown tradition in quantum theory, which, to be sure, was reinforced, but certainly not created by the influx of European émigrés in the 1930s. We are less sanguine about the thesis of Alexi Assmus (1992) that American theorists contributed mainly to molecular rather than to atomic physics, although she may be right that Slater and Van Vleck are just the exception to the rule (see note 28). However, we did come across several other contributions (some admittedly minor) to atomic theory by Americans (Breit, Davisson, Hoyt, Kemble) or by Europeans working in America (Epstein, Swann). And we do want to emphasize that the contributions to atomic theory by our main protagonists were absolutely first rate, even if they did not always receive the recognition they deserved from their European colleagues (see the correspondence between Born and Van Vleck cited in secs. 1.4 and 3.2). The quickly refuted but highly influential Bohr-Kramers-Slater (BKS) theory was built around Slater's idea of a virtual radiation field emitted by an atom while in a stationary state (see sec. 2.2). The derivation of a correspondence principle of absorption for a general non-degenerate multiply-periodic system, the centerpiece of (Van Vleck, 1924b,c), is a *tour de force* that may well have been the most sophisticated application of the correspondence principle in the old quantum theory. All in all, the Americans had definitely established a presence in atomic theory by the early 1920s. In the period we examined, they were certainly more prominent than the British, not to mention the French. One could argue that this casts doubt on the famous thesis of Paul Forman (1971) about the importance of Weimar culture for the breakthrough to quantum mechanics. Our study, however, does not allow any firm conclusions on this score. Those skeptical of the Forman thesis will emphasize parallels between developments in Europe and America, such as those between the work of Kramers and Van Vleck in dispersion theory documented in our study. Those enamored of the Forman thesis, however, can counter that, despite such parallels, it was in Europe, and not in America, that ultimately the decisive steps were taken.

This brings us to the question of why Van Vleck stopped short of these decisive steps. Before we offer our best guess as to why Van Vleck did not do what he did not do, we want to say a few words about why he did what he did. His papers on the correspondence principle for absorption (Van Vleck, 1924a,b,c) constitute his first foray into quantum radiation theory. His earlier publications had dealt with such topics as the extension of Bohr's model of the atom to Helium and the specific heat of molecular hydrogen. The formulation of a correspondence principle for absorption, Van Vleck told Kuhn in his interview for the AHQP in 1963, had been triggered by a comment of his Minnesota colleague Breit (see also Van Vleck, 1924a, 28). Breit's remark, we conjectured (at the end of sec. 3.3), may have directed Van Vleck to the work of Ladenburg and Reiche (1923), who proposed quantum formulae for emission, absorption, and dispersion, invoking but not always correctly implementing the correspondence principle. Van Vleck used the correspondence principle to construct his own quantum formulae for these three phenomena and used his considerable expertise in classical mechanics to show that they merged with the classical formulae in the limit of high quantum numbers. In this way, Van Vleck independently confirmed many of the results in dispersion theory found by Kramers (1924a,b) and further elaborated in (Kramers and Heisenberg, 1925). (Van Vleck, 1924b) certainly contains all the results of (Kramers and Heisenberg, 1925) that are important for Heisenberg's *Umdeutung* paper.

So why did Van Vleck not take the next step? The trivial explanation is that he was too busy working on his *Bulletin* for the *National Research Council* on the old quantum theory (Van Vleck, 1926a) to pursue his own research. But even if he had not been burdened by this *Bulletin*, we seriously doubt that Van Vleck would have done what Heisenberg did—as he himself acknowledged both in a biographical statement prepared for the AHQP and in his interview for the project (see sec. 1.4). Van Vleck, it seems, was too wedded to the orbits of the Bohr-Sommerfeld theory to completely discard them, a prerequisite for Heisenberg's *Umdeutung*. This is clear at several points in (Van Vleck, 1924b). At the end of sec. 1, for instance, we find a formula expressing the Einstein coefficient $A_{r \to s}$ as an average over the frequencies of orbits, not allowed by the Bohr-Sommerfeld quantization condition, between the initial state r and the final state s. Sec. 2 of the paper is devoted to "a correspondence principle for *orbital distortions*" (Van Vleck, 1924b, 334, our emphasis). On the issue of how seriously one should take the orbits of the Bohr-Sommerfeld theory, Van Vleck might have benefited from direct contact with the Europeans. He had the distinct disadvantage of *reading* Sommerfeld instead of *talking* to Bohr and his circle.[131] Bohr and Pauli certainly prepared Heisenberg for the step of leaving orbits behind.

The emphasis on observable quantities in the *Umdeutung* paper, however, struck a chord with Van Vleck, who had been primed for such a positivist turn by his Harvard teacher Bridgman.[132] Explaining the new quantum mechanics in *Chemical Reviews* in 1928,[133] he wrote:

> Heisenberg's epoch-making development of the matrix theory was spurred by Born's repeated emphasis to his colleagues at Göttingen that the reason the old quantum theory was then (1925) failing was that we were all too anxious to use the same concepts of space and time within the atom as in ordinary measurable large-scale events. . . . the concepts of distance and time have a meaning only when we tell how they can be measured. This is very nicely emphasized in Bridgman's recent book, "The Logic of Modern Physics." [Bridgman, 1927] . . . one cannot use a meter stick to measure the diameter of an atom, or an alarm clock to record when an electron is at the perihelion of its orbit. Consequently we must not be surprised if within the atom the correlation of space and time is something which cannot be visualized, and that models cannot be constructed with the same kind of mechanics as Henry Ford uses in designing an automobile. . . . The goal of theoretical physics and chemistry must ever be to explain observable rather than unobservable phenomena . . . What the physicist observes about an atom is primarily its radiations . . . We may say that we have a sound atomic theory when we have a set of a small number of mathematical postulates from which these observed things can be calculated correctly, even though it forces us to discard the usual space-time models (Van Vleck, 1928, 468).

Van Vleck was thus ready enough to give up orbits once Heisenberg (and Born) had shown the way. He failed to take this step on his own.

Studying Van Vleck's paper has made us appreciate various aspects of the

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[131] According to Alexi Assmus (1992, 8, 15), Americans had a tendency to follow Sommerfeld rather than Bohr anyway.

[132] At a ceremony honoring Bridgman's 1946 Nobel prize, Slater suggested that there might even be a genetic connection between Bridgman's operationalism and Heisenberg's uncertainty principle! Schweber (1990, 391) quotes Slater as saying on this occasion: "It is very likely that this principle, so much like Bridgman's attitude, is actually derived to a very considerable extent from Bridgman's thinking."

[133] For the benefit of the chemists, Van Vleck (1928, 469) compared a matrix to a baseball schedule: "the entry in row 3 and column 2, for instance, gives information about a transition between a 3 and 2 quantum state, just as the analogous baseball entry does about the meetings between teams 3 and 2."

transition from the old quantum theory to matrix mechanics that tend to get obscured when one approaches these developments through, say, (Kramers and Heisenberg, 1925). Most importantly perhaps, following (Van Vleck, 1924b,c) rather than (Kramers and Heisenberg, 1925) or (Born, 1924, 1925), we were able to give an illuminating, transparent, and explicit version of the derivation needed to show that the crucial Kramers dispersion formula reduces to the classical formula in the limit of high quantum numbers (see secs. 3.1–3.2 for the special case of a simple harmonic oscillator, sec. 4.2 for the generalization to arbitrary non-degenerate multiply-periodic systems, and sec. 5.1 for a closely analogous derivation of the Kramers formula in modern quantum mechanics). That Van Vleck confirmed the Kramers dispersion formula without relying on the Bohr-Kramers-Slater (BKS) theory makes it particularly clear that matrix mechanics grew directly out of dispersion theory and that BKS was mainly a sideshow (see sec. 2.2). The only element of the BKS theory used by Van Vleck is the concept of virtual oscillators. We saw that this concept actually predates BKS. 'Virtual oscillators' was Bohr's new name for the *Ersatz*-oscillators introduced into dispersion theory the year before and at Bohr's suggestion by Ladenburg and Reiche (1923). In addition to popularizing the notion of virtual oscillators, BKS may have contributed to instilling skepticism about the electron orbits of the Bohr-Sommerfeld theory (MacKinnon, 1977, 1982). In that sense, it might have helped Van Vleck had he embraced BKS more wholeheartedly. Overall, however, we argued that BKS played no role in the breakthrough to matrix mechanics. The same is true for the broad acceptance of Einstein's light-quantum concept following the discovery of the Compton effect. Physicists working in dispersion theory, while accepting the Compton effect as decisive evidence for light quanta, happily continued to treat light as a wave phenomenon.

What was it about dispersion theory that made it so important for the transition from the Bohr-Sommerfeld theory to the theory of Heisenberg's *Umdeutung* paper? As we argued in sec. 2.1, the answer is that the discrepancy between orbital frequencies and radiation frequencies—one of the most radical, if not *the* most radical aspect of the Bohr model of the atom—manifested itself glaringly and unavoidably in dispersion theory. The natural approach to adapting the successful classical dispersion theory of Lorentz and Drude to Bohr's new theory inevitably led to a dispersion formula with resonance poles at the orbital frequencies (Sommerfeld, 1915b; Debye, 1915; Davisson, 1916; Epstein, 1922c), whereas experiment clearly indicated that the resonance poles should be at the radiation frequencies, associated in Bohr's theory with transitions between orbits. Employing Einstein's quantum theory of radiation and Bohr's correspondence principle (in conjunction with techniques from celestial mechanics customized to the problems at hand) and building on pioneering work by Ladenburg (1921) and Ladenburg and Reiche (1923), Kramers (1924a,b) constructed a quantum formula for dispersion with resonance poles at the transition frequencies rather than at the orbital frequencies and claimed that this formula merged with the classical formula in the limit of high quantum numbers. Van Vleck (1924b,c) was the first to publish an explicit proof that the Kramers quantum formula does indeed merge with the classical formula for dispersion in a general non-degenerate multiply-periodic system in the correspondence limit. The three key moves in translating the classical formula into a quantum-theoretical one were to (1) replace orbital frequencies by transition frequencies; (2) relate amplitudes to Einstein's A coefficients; and (3) replace derivatives with respect to the action variable by difference quotients. The first move goes back to the embryonic version of the correspondence principle in (Bohr, 1913) (Heilbron and Kuhn, 1969, 274–275). Ladenburg (1921) introduced the second move. It was made more precise by Kramers and Van Vleck (cf. Jordan's remarks quoted in note 32). Born (1924) is usually credited with the third move and the rule for replacing derivatives by difference quotients is sometimes even called "Born's correspondence rule" (Jammer, 1966, 193) or "Born's discretizing rule" (Cassidy, 1991, 181). It was found independently, however, by both Kramers and Van Vleck (see the discussion at the end of sec. 3.2).

The Kramers dispersion formula no longer contains any reference to the orbits of the Bohr-Sommerfeld theory, but only to transitions between them. This signaled to Heisenberg that orbits could be dispensed with altogether. Dispersion theory further told Heisenberg how to generate quantum formulae from classical formulae in his *Umdeutung* scheme. The procedure consisted of the same three moves listed above: one had to replace (1) classical frequencies (more specifically: the Fourier overtones of the classical mechanical motion) by quantum transition frequencies; (2) classical amplitudes associated with definite orbits by quantum transition amplitudes associated with pairs of stationary states; and (3) derivatives by difference quotients. Dispersion theory also furnished the fundamental quantization condition for Heisenberg's new theory. Heisenberg formulated this condition by applying his *Umdeutung* procedure to the Bohr-Sommerfeld quantum condition, which was no longer acceptable because of its explicit reference to orbits. That Heisenberg's new condition also emerged in the high-frequency limit of the Kramers dispersion formula (see sec. 5.1) convinced him that he had found a sensible replacement for the Bohr-Sommerfeld condition. The relevant formula had been found in quantum dispersion theory before and was known as the Thomas-Kuhn(-Reiche) sum rule (Thomas, 1925; Kuhn, 1925; Reiche and Thomas, 1925). As we saw in sec. 2.1, Van Vleck actually was the first to find this rule, even though he did not emphasize the result because he thought it was problematic. According to Roger Stuewer (private communication), Van Vleck was nonetheless very proud of this achievement and used to mention it with pride to various colleagues in his later years. The Kramers dispersion formula and its corollary, the Thomas-Kuhn sum rule, are the critical physical ingredients in the first two sections of (Heisenberg, 1925c), in which the *Umdeutung* procedure is motivated. Van Vleck was fully cognizant of these same ingredients by mid-1924.

Van Vleck can thus truly be said to have been on the verge of *Umdeutung* in Minnesota in the summer of 1924.

Acknowledgments

We are grateful to Soma Banerjee, Jeffrey Bub, Jeroen van Dongen, Michael Eckert, Jürgen Ehlers, Fred Fellows, Amy Fisher, Domenico Giulini, Lee Gohlike, Seth Hulst, David Kaiser, Mary Kenney, Klaas Landsman, Christoph Lehner, John Norton, Jürgen Renn, Serge Rudaz, Rob Rynasiewicz, Bob Seidel, Phil Stehle, Roger Stuewer, Kathreen Woyak, Carol Zinda, and especially Clayton Gearhart for comments, helpful discussion, and references. Preliminary versions of parts of this paper were presented at Seven Pines VIII (Stillwater, MN, May 5–9, 2004), the Max Planck Institute for History of Science (Berlin, July 2004), New Directions in the Foundations of Physics (College Park, MD, April 29–May 1, 2005), and at the annual meeting of the History of Science Society (Minneapolis, MN, November 3–6, 2005). The final version was presented at *hq0*, a workshop on the history of quantum physics at the Max Planck Institute for History of Science (Berlin, June 13–16, 2006). Michel Janssen gratefully acknowledges support from the Max Planck Institute for History of Science. The research of Anthony Duncan is supported in part by the NSF under grant PHY-0554660.

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