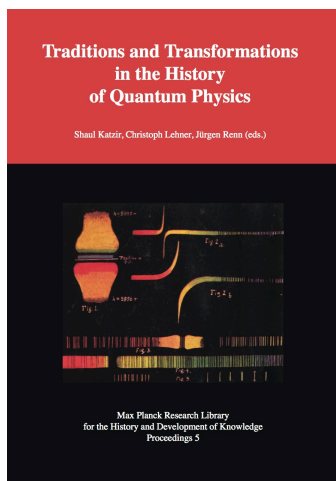


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Dieter Fick and Horst Kant:

The Concepts of Light Atoms and Light Molecules and Their Final Interpretation



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Chapter 4

The Concepts of Light Atoms and Light Molecules and Their Final Interpretation

Dieter Fick and Horst Kant

In 1900, Max Planck (1858–1947) obtained *his* famous radiation formula, see eq. (4.1) (Planck 1913, §156, eq. 275), for the energy per volume and frequency interval emitted by a black body by rather “obscure means” (Darrigol 2009):

$$u_\nu(T) = \frac{8\pi\nu^2}{c^3} \frac{h\nu}{e^{h\nu/kT} - 1}. \quad (4.1)$$

This fitted perfectly with the data over the whole frequency-temperature range investigated at that time (Warburg 1913; Rubens 1913). The way Planck found this formula, partly by “ingenious mathematical manipulations” (Cassidy 2005), how he was driven to the assumption and, later on, to the acceptance of the discrete energy quanta of the oscillators within his model black-body radiator has been related so many times that we may disregard it here.¹ We would like to comment here that the derivation of the two factors in eq. (4.1) show quite different problems. The first factor is connected to the dynamics of the oscillators within a black-body radiator; the second derives from the combinatorial assumptions of how energy elements are distributed over resonators. Its various forms and the related controversies were discussed extensively in publications by Olivier Darrigol (1988; 1991). Satyendranath Bose (1894–1974) was the first to put both factors on equal footing (Bose 1924).

In what follows, we concentrate exclusively on the interpretation, *not the derivation*, of the second factor in eq. (4.1) in terms of light particle concepts. Albert Einstein (1879–1955) opened discussion on the particle nature of black-body radiation as early as 1905 (Einstein 1905). In discussing the entropy of a black body as function of volume, he showed that black-body radiation behaves in the Wien limit like a diluted gas consisting of light quanta. The energy of light appeared in some kind of “granular structure” (Darrigol 1988, 20). Here, we will mainly follow the historical development of the light atom and light molecule concepts, giving some emphasis to the two main actors, Mieczysław

¹For a comprehensive review, see (Kuhn 1987).

Wolfke (1883–1947) and Walther Bothe (1891–1957), as well as to their relationships with Planck, Einstein, Max von Laue (1879–1960) and Louis de Broglie (1892–1987). The early part of this story, up to the beginning of the 1920s, has already been dealt with in part by Silvio Bergia and Darrigol (Bergia, Ferrario, and Monzini 1985; Darrigol 1988; 1991).

4.1 First Corpuscular Concepts of Light

Einstein always denied interpreting light in general as being composed of independent quanta. In a letter to Hendrik A. Lorentz (1853–1928) dated 23 May 1909, Einstein writes:

[...] I am not at all of the opinion that light has to be thought of as being composed of mutually independent quanta localized in relatively small spaces. To be sure, that would be the most convenient way to explain the Wien end of the radiation formula. But the splitting of light rays on the surface of refracting media already makes this approach absolutely inadmissible. A light ray splits, but a light quantum cannot split without a change in frequency. (Klein, Kox, and Schulmann 1993, 193; for German original see: Klein, Kox, and Schulmann 1993, 123)²

In his 1916 and 1917 papers, “Zur Quantentheorie der Strahlung,” (Einstein 1916; 1917)³ Einstein showed that, analogous to particles, each light quantum in a radiation bundle carries a momentum of $h\nu/c$. However, before Einstein, a number of researchers had already understood these as real atoms. An early summary of these activities can be found in Harry Bateman’s (1882–1946) 1923 publication (Bateman 1923).

On 27 September 1910 Abram F. Ioffe (1880–1960) presented a talk, “Zur Theorie der Strahlungserscheinungen” (Ioffe 1911), at a meeting of the Physical Division of the Russian Physico-Chemical Society, the content of which he had already discussed a few weeks earlier with Planck.⁴

The headline of the second part of this publication (Ioffe 1911) “Atomistische Struktur der Strahlung” and the headline of §2 “Strahlungsquanten” both

²Einstein always maintained this point of view, as seen in a letter sent to Wolfke in 1946, see sec. (4.4).

³Since the text in both publications is identical, we will refer to the more easily accessible 1917 publication only.

⁴“I [Ioffe] tried to build [at that time] a theory of radiation energy analogous to the kinetic energy of gases” (Ioffe 1983, 63). Ioffe further aimed to discuss his “at that time heretical ideas” with Planck and to this end visited him at his resort at Lake Chiemsee in Upper Bavaria at the end of August 1910. Ioffe’s former teacher, Wilhelm Conrad Röntgen (1845–1923), arranged the meeting. Ioffe also notes:

point toward an interpretation of light in terms of an atomistic concept. Indeed, §2 in part two starts with the sentence:

Since the appearance of Einstein's article a series of facts have been discovered and discussed, which find their *simplest* explanation in an atomistic concept of radiation, or at least of its emission.⁵ (Ioffe 1911, 546–547)

A list of seven points supporting this claim followed.

Around the same time, Johannes Stark (1874–1957) tried to gain experimental insight into the description of X-rays as light quanta, or alternatively as ether waves (Stark 1910).⁶ Theoretically, he discussed the momentum conservation in electron collisions with matter in great detail.⁷ Experimentally, he analyzed the forward-backward asymmetry of X-rays emitted in electron collisions on a thin, low Z anode (charcoal). In formulating the conditions for momentum conservation under the assumption that the X-rays are light quanta, he explicitly used a vector of length $h\nu/c$ for the momentum of the emitted X-rays and predicted a pronounced forward-backward asymmetry of their momentum (energy) and intensity distributions. In contrast, if X-rays were ether-waves, he found that electrodynamics demanded an isotropic distribution (Abraham 1905). The very demanding experiment revealed pronounced forward-backward asymmetries in X-ray intensities and energies, clearly favoring the light-quantum hypothesis.⁸

In 1913, Wolfke, at the time a *Privatdozent* (private lecturer) in Zurich, was probably the first to introduce the item “light atom” (*Lichtatom*) as a center with energy ϵ , referring to Stark and to Einstein's 1905 article (Stark 1910; Einstein

[...] He [Planck] deemed my article interesting. However, he urgently insisted that I abandon the use of photons since they are inconsistent with Maxwell's electromagnetic theory of light. (Ioffe 1983, 63)

In his reminiscences, Ioffe used the term “photon,” even though it was introduced in 1926 by Lewis (G. N. Lewis 1926). Nevertheless, it was published somewhat later in *Annalen der Physik* of which Planck was an editor. During his visit, Planck had assured Ioffe:

[...] that he will not oppose the publication of the manuscript. However, he does not intend to ruin with his own hands the principles of Maxwell's construct. For him personally, the manuscript is certainly lamentable [...]. (Ioffe 1985, 425)

⁵“Seit dem Erscheinen des Aufsatzes von A. Einstein ist eine Reihe von Tatsachen entdeckt und diskutiert worden, die ihre *einfachste* Erklärung in der atomistischen Auffassung der Strahlung, oder wenigstens ihrer Emission, finden.” Unless otherwise indicated all English translations are by the authors.

⁶These are now called electromagnetic waves.

⁷At that time the German word for momentum was *Bewegungsgröße*.

⁸Even though in 1910 there was growing evidence that “X-rays and light are manifestations of the same phenomena” only a few, such as Stark, believed this was so (Wheaton 1983, 169).

1905; M. Wolfke 1913a). Light energy U was thought to be localized in a large but finite number N of these centers:

$$U = N\epsilon. \quad (4.2)$$

Wolfke chose the term “light atom” to indicate that they cannot come into existence or decay by themselves, for example, N stays constant when reflected off a moving, perfect mirror. He then used this hypothesis to derive the dispersion relation for light atoms with the following arguments: according to classical electrodynamics, the ratios of beam energies U_{in} and U_{ref} and of the frequencies ν_{in} and ν_{ref} for an incoming and reflected beam of light respectively, are identical, see (Abraham 1905, §40; Planck 1913, §77, eqs. 86/87). Thus

$$\frac{U_{in}}{U_{ref}} = \frac{\nu_{in}}{\nu_{ref}} = \frac{N\epsilon_{in}}{N\epsilon_{ref}} = \frac{\epsilon_{in}}{\epsilon_{ref}} \quad (4.3)$$

follows, since the number of light atoms N in a beam ought not to change while the beam is reflected from a perfect mirror. Therefore,

$$\frac{\epsilon_{in}}{\nu_{in}} = \frac{\epsilon_{ref}}{\nu_{ref}} = \text{const.} \quad (4.4)$$

The light atoms thus follow the dispersion relation

$$\frac{\epsilon}{\nu} = \alpha, \quad (4.5)$$

α being a universal constant.⁹ Wolfke commented on this result as follows: “The equation forms the main equation of the light atom”¹⁰ (M. Wolfke 1913a, 1125).

Using this atomistic picture of light, Wolfke interpreted radiation pressure as being caused by the hits of light atoms on a mirror (M. Wolfke 1913b). In doing so, he allocated a mass to a light atom of energy $\epsilon = h\nu$ via the relativistic relation $m = \epsilon/c^2$ for the first time. Before arriving at the heart of the problem, he related the number of light atoms per unit time and volume to the power (energy per time) of the light beam. Then he used a formula found in Planck’s book on heat radiation (Planck 1913, §60, 58, uppermost formula). It connected the radiation pressure \mathfrak{F} , with the number of light atoms hitting the mirror per unit area, with their allotted mass m and with the scattering angle Θ . This formula was obtained by Planck in a quantum picture of light assuming correctly $p = mc$ for the momentum of a light

⁹Because of a mistake in the derivation of Planck’s formula, Wolfke realized only in a subsequent publication (M. Wolfke 1913b) that α is identical to Planck’s constant.

¹⁰“Die Gleichung bildet die Hauptgleichung des Lichtatoms.”

quantum.¹¹ Wolfke finally eliminated the mass through the correct relativistic relation $m = \epsilon/c^2 = hv/c^2$ and arrived thus within a quantum picture of light at the classical relation $\mathfrak{F} = (2 \cos \Theta/c)J$ between radiation pressure \mathfrak{F} and light power J .

At this point, we pause to present Wolfke's involvement up until 1920 in the "light atom" concept in further detail, in particular his controversy with Yuriy Aleksandrovich Krutkov (1890–1952), Paul Ehrenfest's (1886–1930) pupil in St. Petersburg and at that time his visitor in Leiden (Frenkel 1971). This involvement did not lead to any considerable insight and was discussed in detail by Darrigol (1991, 254–255), as well as by Luis Navarro and Enric Perez (2004, 130–132).

First, we briefly report on Wolfke's background.¹² Wolfke was a native Pole. Under Otto R. Lummer (1860–1925), he received a doctorate in Breslau in 1910 with a dissertation on optics (M. Wolfke 1911). At the beginning of 1913, Wolfke moved to the Eidgenössische Technische Hochschule (ETH)¹³ in Zurich. He must have soon met Einstein, since Einstein served as referee in Wolfke's *Habilitationskommission* (habilitation committee) (K. Wolfke 1980). On the basis of a publication that had already appeared in 1912 in *Annalen der Physik* (M. Wolfke 1912), as well as of the positive appraisal of his personality and abilities by Einstein and others,¹⁴ he was promoted on 8 May 1913 to *Privatdozent* at the ETH.

According to a personal report of his son Karol Wolfke, Einstein often visited the Wolfkes and "played violin with father's piano accompaniment" (Sredniawa 2006, 261). It is thus rather probable that Wolfke discussed the light quantum problems with Einstein. And indeed Wolfke notes in one of his publications, at the time of his debate with Krutkov, that Einstein brought certain facts to his attention (M. Wolfke 1914b). This hypothesis is corroborated by several remarks by Darrigol, indicating that Einstein supported Wolfke in his struggle with Krutkov (Darrigol 1991, 254–259).

¹¹This remark is important: Planck assumed, within the same paragraph, that the energy of a light quantum is related to a mass through the Newton relation $\epsilon = \frac{1}{2}mc^2$, and not through the relativistically correct relation $\epsilon = mc^2$. Therefore, Planck obtained twice the classical value for the radiation pressure as Maxwell did for the first time. (For a detailed derivation of the classical radiation pressure, see (Planck 1913, §58, eq. 64)).

¹²For more details, as well as information on Wolfke's relationship with Einstein over the years, see (Kiejna 2002).

¹³Swiss Federal Institute of Technology, Zurich.

¹⁴Letter [from an unknown author, handwritten] of Section VIII from 26 May 1913 addressed to the Chair of the Swiss School Council in Zurich regarding the request for the habilitation of Wolfke: "Professors Einstein and Weiss both agree in their favorable appraisal of the submitted scientific publication and of the professional qualifications and the character of the applicant, and accept the habilitation [...]." "Die Herrn Proff. Einstein und Weiss sprachen sich übereinstimmend günstig über die eingereichten wissenschaftlichen Arbeiten, die Vorbildung und die Persönlichkeit des Gesuchstellers aus, und begrüßten die Habilitation [...].", III-71.21, 23, APAN.



Figure 4.1: Mieczysław Wolfke. Courtesy of the Archives of the Polish Academy of Sciences (APAN).

In the fall of 1922, Wolfke became a professor of physics at the Technical University in Warsaw. There, he worked experimentally on a variety of problems concerning optics, high voltages, properties of liquid helium, and obviously also on what he called “light molecules.” From a report in *Nature* on the Fifth Congress of Polish Physicists in Poznań (Anonymous 1930, 660), we learn that Wolfke presided over the congress, and we read further that “special interest was aroused by papers on association of light quanta by Wolfke.”

4.2 Light Molecules, Static Concepts

4.2.1 Wolfke 1921

At the beginning of 1914, Wolfke published a brief third note, entitled “Zur Quantentheorie” (M. Wolfke 1914a), within a series of papers released in *Verhandlungen der DPG*. Here, he speculated qualitatively on how the transition from the classical Rayleigh-Jeans limit (large radiation density) to the Wien limit (small radiation density) could proceed. A few years later while still in Zurich, he resumed this topic.¹⁵ In a publication entitled “Einsteins Lichtquanten und die räumliche Struktur der Strahlung”, he intended to demonstrate that “[...] black-body radiation [...] consists of thermodynamically-independent parts [...]”¹⁶ (M. Wolfke 1921, 378) with energy density contents of $u_{\nu,s}$, $s = 1, 2, 3, \dots$.¹⁷

His starting point was the then well-known identity for the energy density per unit frequency interval in Planck’s formula, see eq. (4.1)

$$u_{\nu}(T) = u_{\nu} = \frac{8\pi\nu^2}{c^3} \frac{h\nu}{e^{h\nu/kT} - 1} = \sum_{s=1}^{\infty} u_{\nu,s} \quad (4.6)$$

with

$$u_{\nu,s} = \frac{8\pi\nu^2}{c^3} h\nu e^{-sh\nu/kT}, \quad (4.7)$$

the $u_{\nu,s}$ following Wien’s radiation law.^{18,19}

Using this expansion and generalizing Einstein’s method from the 1905 paper (Einstein 1905) to the full radiation spectrum, Wolfke showed that the sum of the partial entropy densities $s_{\nu,s}$ calculated for the partial energy densities $u_{\nu,s}$ adds up to the well-known expression for the entropy density of black-body ra-

¹⁵We can only guess why Wolfke took so long to publish on the light quantum problem. Einstein had already left Zurich in 1913, and during World War I, Wolfke and his family (wife and two children) faced severe economic problems with no regular income (K. Wolfke 1980).

¹⁶“[...] die schwarze Strahlung aus [...] voneinander thermodynamisch unabhängigen Teilstrahlungen besteht [...]”

¹⁷Different from the notations of the publications to be discussed, we additionally label the expansion coefficients and their associated quantities with the frequency ν to which they refer. This is necessary for Appendix 4.4, in which processes that change the frequency of light molecules, such as the Compton effect, are treated.

¹⁸Jun Ishiwara (1881–1947) discussed this expansion as early as 1912 (Ishiwara 1912).

¹⁹To follow the mathematical manipulations throughout this manuscript more easily, the following identities are useful: $\sum_{k=0}^{\infty} z^k = 1/(1-z)$; $\sum_{k=1}^{\infty} k \cdot z^k = z/(1-z)^2$; $\sum_{k=1}^{\infty} k^2 \cdot z^k = z \frac{d}{dz} (\sum_{k=1}^{\infty} k \cdot z^k) = z(1+z)/(1-z)^3$.

diation $\sum_{s=1}^{\infty} \varepsilon_{v,s} = \mathfrak{E}_v$. Thus he interpreted the partial radiation densities $u_{v,s}$ as thermodynamically independent of each other.

For a black body of volume V_0 , $u_{v,s}V_0 = \varepsilon_{v,s}$ may denote the energy per frequency interval of the s th partial radiation. Following Einstein's methodological tools, Wolfke obtained this radiation energy for the probability $W_{v,s}$ within a subvolume, V

$$W_{v,s} = \left(\frac{V}{V_0} \right)^{(\varepsilon_{v,s}/shv)}. \quad (4.8)$$

He therefore interpreted this relation such that the s th partial wave consists of $\varepsilon_{v,s}/shv$ spatially independent radiation quanta shv . He named these objects light molecules and finished his considerations with the remark:

[...] that black-body radiation, as seen from the point of view of Einstein's light-quantum hypothesis, consists of spatially independent light molecules $hv, 2hv, 3hv$ [...].²⁰ (M. Wolfke 1921, 378)

Finally, Wolfke analyzed the number density $n_{v,s}$ of light molecules per unit frequency interval:²¹

$$n_{v,s} = \frac{u_{v,s}}{shv} = \frac{8\pi v^2}{c^3} \frac{1}{s} e^{-shv/kT}, \quad (4.9)$$

yielding for the ratio of successive number densities:²²

$$\frac{n_{v,s+1}}{n_{v,s}} = \frac{s}{s+1} e^{-hv/kT}. \quad (4.10)$$

For large values of hv/kT (Wien limit), the ratio $n_{v,2}/n_{v,1}$ becomes very small in comparison to 1. Thus the radiation field consists of independent light quanta (light atoms) only, as stated by Einstein in his 1905 paper (Einstein 1905). Ap-

²⁰“[...] daß die Hohlraumstrahlung, vom Standpunkt der Einsteinschen Lichtquantenhypothese aus betrachtet, aus voneinander räumlich unabhängigen Lichtmolekülen $hv, 2hv, 3hv$, [...] zusammengesetzt ist.”

²¹Following Wolfke, for this presentation we rewrote all quantities that depend on volume and frequency such that their value per volume, v , and frequency interval, dv , is always quoted. Thus, $n_{v,s}$ is really a number density per frequency interval, dv , and not, as Wolfke mistakenly called it, the numbers of the various light molecules. In all the other publications discussed here, the n are always connected to the number of light molecules, but refer differently to the volume and the frequency interval. For example, in Bothe's early manuscript (Bothe 1923), the n denote the number of light molecules per frequency interval, even though this is not evident at first glance. In his last publication, Bothe defined n as a dimensionless number (Bothe 1927b), as did de Broglie in his brief publication (de Broglie 1922b) and Darrigol in his review article (Darrigol 1991).

²²In his paper, Wolfke discusses the inverse ratio.

proaching the classical Rayleigh-Jeans limit with decreasing values of $h\nu/kT$, the ratios $n_{\nu,2}/n_{\nu,1}$ increase. Wolfke concluded:

We see from this that with growing radiation density there occurs an association of light quanta into increasingly complex molecules, until finally—for very high radiation densities satisfying the Rayleigh-Jeans formula—the quanta agglomerate to form a continuous distribution in space. On the other hand, with decreasing radiation density the radiation continuum dissociates into simpler and simpler light molecules, until it ultimately dissolves into discrete light atoms!²³ (M. Wolfke 1921, 378)

4.2.2 Louis de Broglie 1922

About one year later, without citing Wolfke's publication, de Broglie published two short notes dealing with the particle properties of light and with Einstein's expression for the energy fluctuations, the variance $\overline{\Delta_\nu^2}$ of black-body radiation (de Broglie 1922a; 1922b). He found on purely formal grounds that the expansion of Planck's formula into $h\nu \cdot e^{-sh\nu/kT}$, $s = 1, 2, 3, \dots$ ²⁴ does not contradict Einstein's fluctuation formula (Einstein 1909):

$$\overline{\Delta_\nu^2} \cdot V d\nu = (h\nu) \cdot E_\nu + \frac{E_\nu^2}{Z_\nu}, \quad (4.11)$$

with

$$E_\nu = u_\nu \cdot V d\nu, \quad (4.12)$$

the time average of the energy at frequency ν in a volume V . According to Peter Debye (1884–1966):

$$Z_\nu = \frac{8\pi\nu^2}{c^3} V d\nu \quad (4.13)$$

denotes the number of elementary states²⁵ within a frequency interval, $d\nu$, for a black body of volume V (Debye 1910).

²³“Wir sehen daraus, wie mit zunehmender Strahlungsdichte eine Assoziation von Lichtquanten zu immer komplizierteren Lichtmolekülen stattfindet, bis schließlich die Quanten bei sehr großen Strahlungsdichten, im Gültigkeitsbereich des Jeans-Rayleighschen Strahlungsgesetzes, sich zu einem Kontinuum zusammenballen! Umgekehrt, mit abnehmender Strahlungsdichte dissoziiert das Strahlungskontinuum in immer einfachere Lichtmoleküle, bis es sich schließlich in diskrete Lichtatome auflöst!”

²⁴Equations resembling Wolfke's eqs. (4.6), (4.7).

²⁵De Broglie did not use the quantity Z_ν explicitly.

This is important historically because the two parts in Einstein's fluctuation equation at that time were often literally connected to the *particle* ($h\nu$) $\cdot E_\nu$ and *wave* aspect E_ν^2/Z_ν of light. In his results, de Broglie intuitively saw the physical picture of "coherent photons" (mono, duo, triple correlated photons) in the expansion of Planck's formula, see eqs. (4.6), (4.7).²⁶ Therefore, he was convinced that "if the theory of light-quanta ever succeeds in interpreting interference, it will require such agglomeration of quanta" (Darrigol 1991, 260).

4.3 Dynamical Treatments of Light Multiples

4.3.1 Bothe 1923

Citing Wolfke's 1921 publication only in passing, Bothe,²⁷ Planck's former student, submitted a manuscript entitled "Die räumliche Energieverteilung der Hohlraumstrahlung" to *Zeitschrift für Physik* in 1923 (Bothe 1923).²⁸ At the time, Bothe worked at the Physikalisch Technische Reichsanstalt (PTR) in Berlin-Charlottenburg. Aside from his experimental work (Fick and Kant 2009), from mid-1923 to the end of 1926, he published a few theoretical papers, all of which dealt with the "light quantum problem." In the acknowledgments and footnotes, more often than not, he thanked von Laue, who was a full professor at the Berlin University, a member of the Königlich Preußische Akademie der Wissenschaften (Prussian Academy of Sciences) and deputy director of the Kaiser Wilhelm Institut für Physik during that time (Hoffmann 2010, and references therein). In addition, Einstein was very influential. Einstein was also a member of the Prussian Academy. Beginning in 1914, he lived and worked in Berlin, and in 1916, he became a member of the *Kuratorium* (board of trustees) of the PTR. Since the light quantum problem was a primary concern of Bothe's, he certainly kept in touch with Einstein.²⁹ In his Nobel lecture, Bothe recalls this period: "Dur-

²⁶A detailed discussion of de Broglie's concerns can be found in (Bergia, Ferrario, and Monzini 1985).

²⁷Details on Bothe's vita up to the 1920s and his experimental achievements in the study of the wave-particle duality of light during the 1920s can be found in a recent article entitled "Walther Bothe's Contributions to the Understanding of the Wave-Particle Duality of Light" (Fick and Kant 2009).

²⁸"The Spatial Energy Distribution of Black-Body Radiation." The Archive of the Max Planck Society in Berlin (AMPG) retains, at the back of another manuscript, a handwritten manuscript (Bothe manuscript, III/6/104,1, AMPG) which coincides in large part with the printed version of Bothe's publication in the *Zeitschrift für Physik*. The differences in the printed version are marginal, as far as the content is concerned. On a few additional pages (microfilm pages 1211, 1213, 1224), Bothe also discusses the question of whether the structure of radiation emitted by a black body changes under processes that do not change the temperature of the radiation (reflection, refraction, absorption, etc.). He concluded that at his time such questions could not be answered experimentally.

²⁹See also the end of this section.

ing this time I had the singular good fortune of being able to discuss the [wave particle] problem constantly with Einstein” (Bothe 1964, 274).

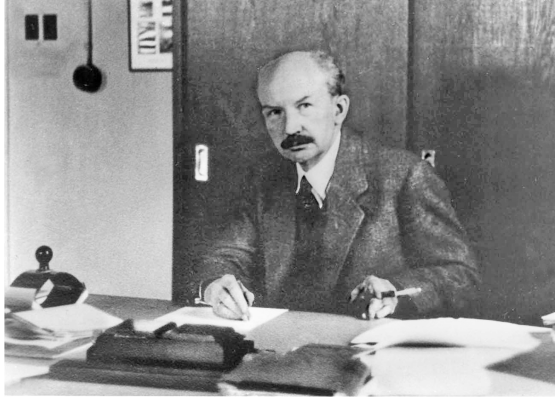


Figure 4.2: Walther Bothe. Courtesy of the Archives of the Max Planck Society, Berlin.

Beginning with Einstein’s formulation of emission and absorption processes (Einstein 1917), in his paper, Bothe studied how a two-level object (for example, an atom) achieves thermal equilibrium in a black-body radiation field. He characterized the atom by states 1 and 2 with energies ϵ_1 and ϵ_2 ($\epsilon_2 > \epsilon_1$) and occupation numbers N_1 and N_2 . According to Einstein, the atoms can undergo two types of emission processes by emitting a light quantum of energy $h\nu = \epsilon_2 - \epsilon_1$: a spontaneous one proportional to aN_2 and an induced one proportional to bN_2 . The absorption of a light quantum with energy $h\nu$, inducing transitions from state 1 to state 2 is then proportional to bN_1 .³⁰ The constants a and b are characteristic for the transitions involved.

To introduce the concept of a “quantum multiple” for a fixed frequency ν , Bothe reminds the reader that according to Einstein (1917), induced emission and absorption are, spatially, perfectly correlated processes. Moreover for stimulated emission, the inducing and stimulated quanta are perfectly correlated. They possess identical directions and energies (phases, polarizations).³¹ He noticed later that:

³⁰Without a lack of generality, we choose the statistical weights of the states 1 and 2 to be the same and equal to one, since they will not appear in the final results. This is why the b -coefficients for induced transitions and for absorption of a light quantum are equal here.

³¹We will return to this point at the end of sec. (4.3.3).

Both [quanta] are coupled seemingly; only seemingly, since in truth no forces exist between both, the dissociation energy [...] is zero. If one of both quanta is absorbed, the fate of the other is not influenced at all; this is a consequence of the assumption that the probability of an induced process is simply proportional to the spatial density of the quanta. We shall therefore better speak about *quantum pairs*. If the inducing quantum already belongs to a pair, a triple will emerge, and so on.³² (Bothe 1923, 147)

It is clear that Bothe's quantum multiples have only formally common aspects to Wolfke's and de Broglie's light molecules. Whereas both interpreted them more or less as real particles, Bothe in particular had the correlation aspect in mind. In our present understanding, we would instead call them "quasiparticles."

Denoting the number density per frequency interval dv of single quanta, pairs, triples, ... s -fold multiples, of light quanta by $n_{v,1}, n_{v,2}, n_{v,3}, \dots, n_{v,s}, \dots$, Bothe first studied the thermal equilibrium conditions.³³ Within a time interval dt , single light quanta are produced with a probability of aN_2 by spontaneous emission, and with a probability of $bN_1(2n_{v,2}h\nu)$ by absorption from a quantum pair, since $2n_{v,2}h\nu$ is the fraction of the total radiation density u_v , which belongs to quantum pairs. These are the two source terms. On the other hand, single quanta disappear with a probability $bN_1(n_{v,1}h\nu)$ through an absorption process, and with a probability of $bN_2(n_{v,1}h\nu)$ through conversion into a quantum pair in a stimulated emission process. These are the two drain terms. Since in thermal equilibrium the number of single quanta should be stationary, one obtains as a condition:

$$aN_2 + bN_1(2n_{v,2}h\nu) - bN_1(n_{v,1}h\nu) - bN_2(n_{v,1}h\nu) = 0, \quad (4.14)$$

which couples singlets and doublets of quanta with the spontaneous decay (zero quantum).

³²“Beide [Quanten] sind scheinbar gekoppelt; nur scheinbar deshalb, weil in Wahrheit keine Kräfte zwischen ihnen wirken, die Dissoziationsarbeit [...] ist Null. Wird etwa eins der beiden Quanten absorbiert, so wird das Schicksal des anderen hierdurch in keiner Weise beeinflusst; dies ist eine Konsequenz der Annahme, dass die Wahrscheinlichkeit eines Einstrahlungsprozesses einfach proportional der mittleren räumlichen Dichte der Quanten ist. Wir werden deshalb besser von Quantenpaaren sprechen. Gehört das auslösende Quant selbst schon einem Paar an, so entsteht ein Tripel, usf.”

³³In the manuscript, Bothe denotes by $n_{v,1}, n_{v,2}, n_{v,3}, \dots, n_{v,s}$ the number of light quanta $h\nu$ which form singlets, pairs, triplets, ... respectively. This definition differs from the one used by Wolfke (1921), and also from the definition used later by Bothe himself (1924). In what follows, we use Wolfke's definition (1921) of the $n_{v,s}$. It relates to the definition in the work discussed here through the relation $(n_{v,s})_{\text{Bothe}} = (s \cdot n_{v,s})_{\text{Wolfke}}$. Thus, here, the fraction of the total radiation density $u_{v,s}$, which belongs to s -fold quantum multiples, is $u_{v,s} = sn_{v,s}h\nu$.

The next higher-order rate equation, coupling singlets, doublets, and triplets with each other

$$bN_2(n_{v,1}hv) + bN_1(3n_{v,3}hv) - bN_1(2n_{v,2}hv) - bN_2(2n_{v,2}hv) = 0, \quad (4.15)$$

consists again of two source and two drain terms. The previous source term converting a doublet into a singlet now becomes a drain term for doublets, and the previous drain term for singlets now becomes a source term for doublets. This is the general structure of all higher-order rate equations. Two of the contributions always change sign in the next order equation.

Therefore, summing up these equations to the order of s , most of the terms cancel each other out. One obtains a rate equation, which couples an $(s + 1)$ - and an s -fold quantum state with a single-quantum state and the term for the spontaneous decay:

$$aN_2 + bN_1(s + 1)n_{v,s+1}hv - bN_2sn_{v,s}hv - bN_1n_{v,1}hv = 0. \quad (4.16)$$

Only the spontaneous decay process, aN_2 , cannot depend on the radiation densities for the various multiple quantum states. Since the total number of light quanta $\sum_{s=1}^{\infty} sn_{v,s}$ has to be finite, the number densities $n_{v,s}$ have to approach zero with increasing s , yielding finally:

$$-bN_1n_{v,1}hv + aN_2 = 0. \quad (4.17)$$

According to Boltzmann's law, the occupation numbers N_i ($i = 1, 2$) of the atomic states can be expressed as function of temperature³⁴

$$N_i = e^{-\epsilon_i/kT}. \quad (4.18)$$

Following Einstein (1917), the ratio a/b is then given by

$$a/b = 8\pi\nu^2/c^3. \quad (4.19)$$

Bringing everything together,³⁵ one obtains for the s th multiple:

$$n_{v,s} = \frac{8\pi\nu^2}{c^3} \frac{1}{s} e^{-sh\nu/kT} \quad s = 1, 2, 3, \dots \quad (4.20)$$

³⁴As mentioned in footnote 30, we chose without lack of generality the statistical weights of states 1 and 2 to be the same and equal to one, since they anyway will not appear in the final results.

³⁵With eq. (4.19), from eq. (4.17), one obtains $n_{v,1}$. Inserting it into eq. (4.14) yields $n_{v,2}$, and so on.

These number densities are formally identical to the ones obtained by Ishiwara and Wolfke through the expansion of Planck's formula³⁶ (Ishiwara 1912; M. Wolfke 1921). Therefore, summing up all the terms has to yield Planck's formula again. However, as pointed out above, Bothe's light multiples are correlated light quanta (quasiparticles), whereas Wolfke interpreted them as real objects.

Citing Lorentz (1916, 59), Bothe began the introduction to the publication with the remark that the energy variation (variance) of a black body holds the key to understanding radiation theory. Therefore, unlike Wolfke (1921), it was essential for him to address this topic as well. To do so, he extended Einstein's analogy between an ideal gas and the light quantum gas consisting of independent constituents to the s -fold multiples. Consequently, he assumed that the average number of s -fold multiples $n_{\nu,s}$ fluctuates in time in a Gaussian way as $\sqrt{n_{\nu,s}}$. Thus, the mean energy fluctuation squared of an s -fold multiple (energy $sh\nu$) is $\overline{\Delta_{\nu,s}^2} = (sh\nu)^2 n_{\nu,s}$. Summing over all s and using eq. (4.19), one obtains as variance

$$\overline{\Delta_{\nu}^2} = \sum_{s=1}^{\infty} \overline{\Delta_{\nu,s}^2} = \sum_{s=1}^{\infty} (sh\nu)^2 n_s = (h\nu)^2 \sum_{s=1}^{\infty} s^2 n_{\nu,s} = \frac{8\pi h^2 \nu^4}{c^3} \frac{e^{h\nu/kT}}{(e^{h\nu/kT} - 1)^2}. \quad (4.21)$$

Finally, by replacing the temperature T with u_{ν} , the energy density per frequency interval, Planck's formula, see eq. (4.1), one indeed obtains Einstein's fluctuation formula (Einstein 1917)³⁷:

$$\overline{\Delta_{\nu}^2} = h\nu \cdot u_{\nu} + \frac{u_{\nu}^2}{Z_{\nu}}. \quad (4.22)$$

Bothe demonstrated at that time that the concept of light multiples allows a correct reproduction of Einstein's fluctuation formula, in contrast to both classical theory and to simple light-quantum concepts. Even a wave concept was unnecessary to obtain the correct radiation (Planck) and fluctuation (Einstein) formulas, as long as spontaneous emission as well as phase-correlated induced emission and absorption are properly taken into account.

Nevertheless, there was at least one point that was not understood: why are there no $s = 0$ contributions in the expansion of Planck's formula, see eq. (4.6), or in other words, why must the spontaneous decay be treated separately? This

³⁶See eq. (4.9) in this contribution.

³⁷For the definition of Z_{ν} in the equation below, see eq. (4.13).

hampered an interpretation of the light quantum multiples in physical terms. We now address this question.

4.3.2 Bothe 1925a

Bothe resumed this topic a few years later in an as-yet unpublished manuscript entitled “Zur Struktur der Strahlung.”³⁸ The manuscript is undated, but he worked on this until at least February 1925 when Planck, in a meeting of the Prussian Academy of Sciences, presented a paper “Zur Frage der Quantelung einatomiger Gase” (Planck 1925), which Bothe duly cited. Bose’s seminal paper had been published in the December 1924 issue of *Zeitschrift für Physik* (Bose 1924), just a couple of months earlier.

The main aim of the unpublished work was to find a bridge between the quantum picture and the wave picture of radiation. Bothe began the text with the following sentences:³⁹

Einstein’s derivation of the radiation formula [variance of energy in a black body] (Einstein 1917) stands out due to its outstanding clarity. If one attempts to develop it further [...], one obtains the strange picture that the light quanta within the stationary radiation field are in general not independent of each other, but rather bunched. Only by accounting for the bunching of the light quanta does one arrive at the correct expression for the energy fluctuations of the radiation field (Bothe 1923; M. Wolfke 1921). Unfortunately until now, no one has succeeded in building a bridge from this point to the classical wave theory, e. g., in specifying the number of degrees of freedom within a radiation volume, or even in interpreting the interferences.^{40,41}

³⁸Bothe (ca. 1925). *Zur Struktur der Strahlung*, unpublished manuscript, AMPG III/6/105.4.

³⁹It is surprising that Bothe cited Wolfke, even though Wolfke did not discuss the energy variation in his paper at all.

⁴⁰See footnote 38.

⁴¹“Die Einsteinsche Herleitung der Strahlungsformel (Einstein 1917) zeichnet sich bekanntlich durch außerordentliche Anschaulichkeit aus. Versucht man diese weiterzubilden, ohne ihr diesen Charakter zu nehmen, so gelangt man zwangsläufig zu der merkwürdigen Vorstellung, dass im stationären Strahlungsfelde die Lichtquanten im allgemeinen nicht unabhängig voneinander, sondern zu Aggregaten vereinigt sind. Durch Berücksichtigung dieser Assoziationen der Lichtquanten gelangt man erst zum richtigen Ausdruck für die Energieschwankungen im Strahlungsfeld (Bothe 1923; M. Wolfke 1921). Leider ist es bisher nicht gelungen, von diesem Punkte aus die Brücke zu der klassischen Wellentheorie der Strahlung zu schlagen, z.B: die Anzahl der Freiheitsgrade eines Strahlungsvolumens richtig anzugeben, oder gar die Interferenzen zu deuten.” (The bibliographic references in parentheses were footnotes in Bothe’s original manuscript).

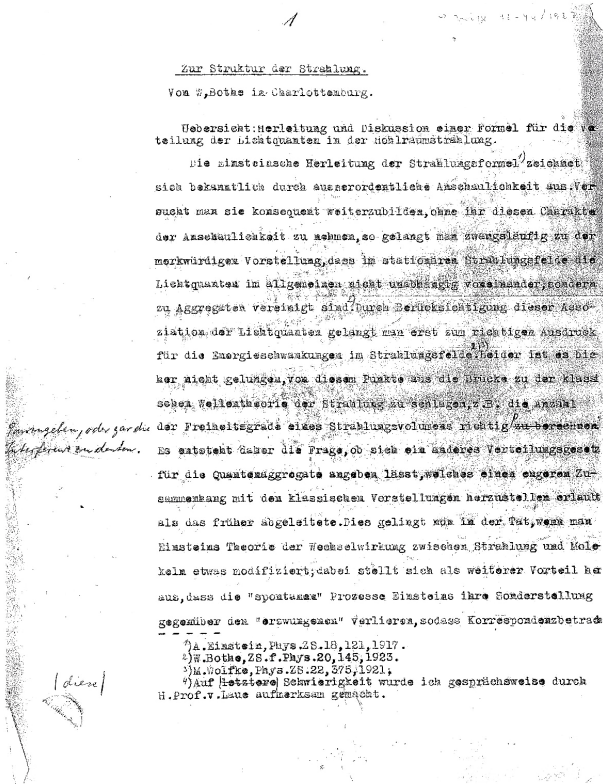


Figure 4.3: Bothe manuscript, (n.d., ca. 1925). "Zur Struktur der Strahlung." AMPG, III/6/105.4.

As we will see below, this drawback is closely connected to the missing $s = 0$ term in the expansion of Planck's formula in terms of Wienian-type s -fold light multiple energy distributions, see eqs. (4.6), (4.7), (4.20).

Bothe's main idea was to use a heuristic argument to put the spontaneous and induced emission on equal footing. With a black-body radiation field consisting of s -fold light multiples according to sec. (4.2) in mind, he started his consid-

erations again by questioning if and how thermal equilibrium is reached for his favored model system, the interaction of black-body radiation with two-level objects (atoms, molecules). He examined two processes:

1. The object is excited through interaction with an $(s + 1)$ -fold multiple from the energetically lower state with energy ϵ_1 into the energetically higher state with energy ϵ_2 , whereby the $(s + 1)$ -fold light multiple of frequency $\nu = (\epsilon_2 - \epsilon_1)/h$ is transformed into an s -fold light multiple of the same frequency ν . Using additional Boltzmann statistics for the population of the states 1 and 2, see eq. (4.18), the incidence of such a process is, as before, proportional to $(s + 1)n_{\nu,s+1}e^{-\epsilon_1/kT}$, since $(s + 1)n_{\nu,s+1}$ is the fraction of the total radiation density u_ν , which belongs to $(s + 1)$ -fold multiples.⁴²
2. The reverse process, the de-excitation of an excited molecule through interaction with an s -fold light multiple, transforms it into an $(s + 1)$ -fold one. To avoid a special role of the spontaneous transitions with respect to the induced ones, see section (4.3.1), Bothe now assumed on heuristic grounds that these transitions are proportional to $(s + 1) \cdot n_{\nu,s}$ instead of $s \cdot n_{\nu,s}$, surmising correctly, as early as 1925, a small part of modern field theory.⁴³ The incidence of these processes is then proportional to $(s + 1)n_{\nu,s}e^{-\epsilon_2/kT}$. Bothe himself commented on his ansatz very cautiously:

The addend $+1$ in the bracket replaces to some extent the spontaneous transitions, since it allows now also for emission processes which start from a zero quantum multiplet.^{44,45}

One benefit of this is that zero-fold multiples are formally allowed and would today be termed a “physical vacuum.” Thanks to Paul Dirac’s seminal work (Dirac 1927, 251/261, eq. 10), the factor $(s + 1)$ in front of terms describing the creation of light quanta (and of Bosons in general) is absolutely necessary to guarantee a symmetric wave function.⁴⁶

To achieve thermal equilibrium, both rates have to be equal, yielding

$$n_{\nu,s+1} = n_{\nu,s}e^{-h\nu/kT}, \quad (4.23)$$

⁴²Since the number density of the s -fold light multiples differs in definition from the ones of the previous section, we denote them now with a gothic n instead of a latin n .

⁴³For an early interpretation of Bothe’s choice, see Jordan’s (1928, in the very last paragraph of section I) discussion.

⁴⁴Bothe’s unpublished manuscript p. 3 (see footnote 38).

⁴⁵“Der Summand $+1$ in der Klammer ersetzt gewissermaßen die spontanen Übergänge, da er z.B. auch nullquantigen Aggregaten die Auslösung von Emissionsprozessen ermöglicht.”

⁴⁶This fact was, for example, discussed highly pedagogically in volume III of the *Feynman Lectures on Physics* (Feynman, Leighton, and Sands 1965, Chap. 4.4, Emission and absorption of photons).

for all $s \geq 0$, from which

$$n_{\nu,s} = n_{\nu,0} e^{-sh\nu/kT} \quad (4.24)$$

immediately follows. Before we report on the consequences of Bothe's ansatz, we anticipate a small detail of Bothe's 1927 publication (Bothe 1927b) discussed at the end of section (4.3.3). This detail concerns the ratio δ between the total rate of induced and spontaneous transitions in a black body. Drawing from Bothe's less precise interpretation of the two parts in the factor $(s + 1)$ (see quotation above), he obtained

$$\delta = \frac{\sum_{s=0}^{\infty} s \cdot n_{\nu,s}}{\sum_{s=0}^{\infty} 1 \cdot n_{\nu,s}} = \frac{1}{e^{h\nu/kT} - 1} \quad (4.25)$$

by means of eq. (4.24) for this ratio.

More recently, δ is called "degeneracy parameter."⁴⁷ It approaches "0" (no induced transitions) for the extreme Wien limit ($h\nu/kT \rightarrow \infty$) and "1" for the extreme Raleigh-Jeans limit ($h\nu/kT \rightarrow 0$).

In contrast to Wolfke's (1921), de Broglie's (1922b) and his own previous considerations (Bothe 1923), Bothe used his heuristic trick and found a possibility to also formally treat ($s = 0$)-multiples. Furthermore, using the last eq. (4.25), he gained an advanced interpretation of the total number density n_{ν} of light multiples, per frequency interval $d\nu$, including the zero-quantum ones. To reach this point, he identified $n_{\nu} d\nu V$, the total number of light multiples per frequency interval $d\nu$ and volume V with the number of degrees of freedom of a black body Z_{ν} , see eq. (4.13):⁴⁸

$$\sum_{s=0}^{\infty} n_{\nu,s} = \frac{Z_{\nu}}{V d\nu} = \frac{8\pi\nu^2}{c^3} = \underbrace{\sum_{s=0}^{\infty} n_{\nu,0} e^{-sh\nu/kT}}_{= n_{\nu}} \quad (4.26)$$

Performing the sum within the underlined part of the equation yields:

$$n_{\nu,0} = \frac{8\pi\nu^2}{c^3} \frac{1}{\sum_{s=0}^{\infty} e^{-sh\nu/kT}} = \frac{8\pi\nu^2}{c^3} (1 - e^{-h\nu/kT}), \quad (4.27)$$

⁴⁷One year later, Reinhold Fürth (1893–1979) discussed the same issue in other contexts (Fürth 1928a; 1928b).

⁴⁸We discuss the physical implications of this assumption in the following section, after obtaining formal results (4.3.3).

and thus

$$n_{\nu,s} = \frac{8\pi\nu^2}{c^3} (1 - e^{-h\nu/kT}) e^{-sh\nu/kT}, \quad s = 0, 1, 2, \dots \quad (4.28)$$

This equation can be rewritten as

$$n_{\nu,s} = \frac{8\pi\nu^2}{c^3} \cdot \frac{e^{-sh\nu/kT}}{\sum_{s=0}^{\infty} e^{-sh\nu/kT}}, \quad (4.29)$$

a required expression if the s -fold light multiples in a black body are distributed thermodynamically according to Boltzmann's law.⁴⁹ Thus the n_s can now be interpreted thermodynamically as occupation number densities for the s -fold multiples in a black body. Bothe was probably aware of this since he commented on his result as follows:

This is the new distribution law of the quantum multiples. From it one can easily derive the total radiation energy u_ν at frequency ν [...], that is Planck's formula.^{50,51}

$$u_\nu(T) = \sum_{s=0}^{\infty} sh\nu \cdot n_{\nu,s} = \frac{8\pi\nu^2}{c^3} \frac{h\nu}{e^{h\nu/kT} - 1}. \quad (4.30)$$

After having obtained this result, Bothe reminds the reader that, following Debye (1910), the number of degrees of freedom of a black-body radiator at frequency ν is nothing but the number of eigenmodes of the black body at that frequency ν . Each of these eigenmodes can be replaced by a Planck oscillator of frequency ν , and the number of eigenmodes with energy $sh\nu$ can be identified according to the quantum rules with $n_{\nu,s}$. Indeed, eq. (4.28) is identical to the statistical distribution law of the energies of Z_ν Planck oscillators. He ends with the remark:

After this, we can interpret each quantum multiple in black-body radiation as the energy of an eigenmode of the black body.^{52,53}

⁴⁹Einstein used this kind of reasoning for the first time in his 1911 talk at the Solvay Conference (Einstein 1913).

⁵⁰Bothe's unpublished manuscript p. 3 (see footnote 38).

⁵¹"Dies ist das neue Verteilungsgesetz der Quantenaggregate. Aus ihm ergibt sich leicht die gesamte Strahlungsenergie E_ν von der Frequenz ν [...] d.i. die Plancksche Formel."

⁵²See footnote 38.

⁵³"Wir können hiernach also jedes Quantenaggregat in der Hohlraumstrahlung deuten als die Energie einer Eigenschwingung des Hohlraums." Sentence underlined on p. 10 in Bothe's manuscript.

In 1912, Ishiwara already had a similar interpretation in mind (Ishiwara 1912), starting from the power expansion of Planck's formula, see eq. (4.6), and relying on Debye's (1910) interpretation of phase space quantization. However, he did not get very far, since at that time in the expansion of Planck's formula ($s = 0$)-terms were only previously discussed by Einstein at the 1911 Solvay Conference (Einstein 1913).⁵⁴

Bothe finally returns to the question of whether the correct expression of the energy fluctuations in a black body follows from the distribution law in eq. (4.28). He did not doubt this since the distribution law in eq. (4.28) correctly yields Planck's formula. Nevertheless, he demonstrates this explicitly in a few lines.⁵⁵

4.3.3 Bothe 1927

One can only speculate why Bothe did not publish the manuscript at the beginning of 1925. One reason was certainly his workload at the PTR. Hans Geiger (1882–1945), the director of the laboratory for radioactivity, had left the PTR around that time, and Bothe succeeded him. In Germany, the PTR was responsible for all official calibrations of radioactive samples.

The appearance of the Bose statistics and its implications might also have caused a delay in the publication of the manuscript. Moreover, apart from his daily duties, between 1926 and 1927, Bothe performed two crucial coincidence experiments which dealt with resonance fluorescence of X-rays (Bothe 1926) and later with Compton scattering in an X-ray interference field (Bothe 1927a). Both experiments were highly complex and very time consuming.⁵⁶ It might therefore be that Bothe simply did not have enough time to finish the manuscript.

Using Compton scattering, Bothe found in the latter experiment that light quanta from an interference field still carry a momentum of $h\nu/c$. Like Einstein,⁵⁷ he considered his result to be extremely important, as we know from the handwritten notes for a talk he presented at the end of October 1926.⁵⁸ At that time, he discussed standing waves of an interference field, which definitely represents a classical situation with a vanishing energy and momentum transport in

⁵⁴See also footnote 49.

⁵⁵Quite recently one of the authors (D. Fick) presented an analysis of Bothe's 1925 heuristic assumption (Fick in print), on which sections 4.3.1 and 4.3.2 are based.

⁵⁶For details, see the extended report by Fick and Kant, "Walther Bothe's Contributions to the Understanding of the Wave-Particle Duality of Light" (Fick and Kant 2009).

⁵⁷A brief report on a talk by Einstein presented at Berlin University (renamed Humboldt University in 1948) entitled "Theoretisches und Experimentelles zur Frage der Entstehung des Lichtes" (Anonymous 1927) explicitly mentioned this experiment. However, there were also critical questions concerning its relevance (Kirchner 1930, 467–468).

⁵⁸Bothe (ca. 1926). *Lecture Notes*, unpublished, AMPG III/6/105.3.

total. Light quanta in an interference field, as well as in a directed (needle) beam, nevertheless possess a momentum of $h\nu/c$.

Gathering the main ideas of his unpublished manuscript, Bothe reacted to this situation with a short publication, again reinterpreting the concept of light multiples (Bothe 1927b). Since a monochromatic unidirectional needle beam is a non-physical object, he used von Laue's (1914) light bundles of a finite cross section and of a very small frequency and opening angle interval as "elementary cells," in which the light quanta are embedded. He considered these bundles, together with the embedded quanta, as independent entities of the radiation statistics. Each bundle could contain an arbitrary amount of light quanta (including zero), whereby the number of elementary bundles per volume V and frequency range $d\nu$ was identical to the number of degrees of freedom $Z_\nu = (8\pi\nu^2/c^3)V \cdot d\nu$, see eq. (4.26). In this way, Bothe followed the new Bose statistics, which led to a sort of coupling between the light quanta, as had already been noted by a few others (Einstein 1924; Ehrenfest 1925; Landé 1925). With this interpretation in mind, Bothe demonstrated that not only the number of light quanta for a black body in thermal equilibrium is stationary but so is their grouping.

As frequently emphasized, Bothe considered the understanding of Einstein's fluctuation formula, see eq. (4.22), to be extremely important. He therefore concluded this publication with an alternative derivation which used the dual concept of light directly. Since its formal procedure is only of minor interest here, we outline the main idea and present the final result. Following Lorentz (1916), Bothe began with the remark that in an elementary bundle, the (classical) wave energy ϵ fluctuates around its average $\epsilon_0 = (u_\nu V \cdot d\nu)/Z_\nu$ with a variance (root mean square fluctuation) δ_{wave} equal to the average wave energy itself.⁵⁹

$$\overline{\delta_{wave}^2} = \epsilon_0^2. \quad (4.31)$$

In classical physics, this would be the total fluctuation of the averaged energy. However, if one assumes that the wave energy is distributed discontinuously over "light particles" (quanta), the number of light particles i itself fluctuates around its average value $i_0 = \epsilon/h\nu$ with a variance of i_0 if a Poisson distribution is assumed. The variance of the energy due to the fluctuation of the number of light particles is therefore $i_0(h\nu)^2 = \epsilon h\nu$. Furthermore, averaging over the energy ϵ itself, one obtains for the variance (root mean square fluctuation) of this contribution:

$$\overline{\delta_{particles}^2} = \epsilon_0 h\nu. \quad (4.32)$$

⁵⁹See also (Jordan 1929), in particular, the discussion of eqs. 9 and 10.

The tricky aspect of the problem now is that both variances are coupled, since the fluctuation of the number of particles depends on the fluctuation of the (wave) energy. Nevertheless, in an appendix Bothe proved that these two variances simply add to each other:

$$\overline{\delta^2} = \overline{\delta_{particles}^2} + \overline{\delta_{wave}^2} = \epsilon_0 h\nu + \epsilon_0^2. \quad (4.33)$$

Multiplying the variance $\overline{\delta^2}$ with the number of independent elementary bundles per volume and frequency interval $(Z_\nu)/(V \cdot d\nu)$, one obtains the known result, see eq. (4.22), which we repeat here for easy comparison:

$$\overline{\Delta_\nu^2} = h\nu \cdot u_\nu + \frac{u_\nu^2}{Z_\nu}. \quad (4.34)$$

As Bothe pointed out, this equation followed from a systematic treatment of the dualistic nature of light and does not follow from a plain addition of the fluctuations of the separate results that treat light first as a wave and then as a light particle. As we now know, Poissonian distributed particle numbers, for example light quanta in a black body, always end up in energy fluctuations “expressible as the sum of contributions from the fluctuations of classical particles and the contributions of classical wave fields” (Mandel and Wolf 1965, 271, eq. 6.19 and its discussion; Born 1949, 79–82).⁶⁰

With this manuscript, Bothe completed his research on the nature of light quanta for the rest of his scientific career. He never returned to the concept of light multiples. Nevertheless, one aspect of Bothe’s manuscript stood the test of time and is still cited today, his discussion of the degeneracy parameter δ , see eq. (4.25). Due to its definition, it may also be considered the average number of light quanta that are in the same quantum state. There is a third interpretation of δ as a ratio between the multi-light quantum states ($s \geq 2$) and the one-light quantum states ($s = 1$) of thermal radiation

$$\delta = \frac{\sum_{s=2}^{\infty} s \cdot n_{\nu,s}}{n_{\nu,1}} = \frac{1}{e^{h\nu/kT} - 1}. \quad (4.35)$$

Multi-light quantum states ($s \geq 2$) are generated by various sequences of induced transitions. According to Einstein (1917), light quanta from induced transitions are perfectly correlated in direction, energy, phase and polarization. Due to the

⁶⁰To make this clear, we deliberately denoted the two contributions to eq. (4.33) with “wave” and “particle” and not as Bothe did with “wave” and “quantum.” See also “Reconstruction of and commentary on Jordan’s derivation of Einstein’s fluctuation formula” in (Duncan and Janssen 2008, sec. 3).

latter interpretation of δ , it is therefore comprehensible that correlation functions in space are proportional to the degeneracy parameter δ .⁶¹

In their seminal experiments during 1957–1958, Robert Hanbury Brown and Richard Twiss were the first to provide experimental evidence of space correlations between the outputs of two photoelectric detectors illuminated by partially-correlated light waves of a mercury lamp in a Michelson configuration. In such experiments, the magnitude of the correlation coefficient is proportional to δ .⁶²

We end this section with the remark that in 1958 Hanbury Brown and Twiss determined the correlation of light of the star Sirius A, a black body of about 10,000K surface temperature for the first time (Mandel and Wolf 1995, Chap. 9.10 and references therein). The observation of correlations is an experimental proof that the concept of light molecules à la Ishiwara (1912), Wolfke and the early Bothe, see secs. (4.2.1) and (4.3.1), is useless. It has long been known that the expansion of Planck's formula into a sum of the Wienian terms $u_{\nu,s} = (8\pi\nu^2/c^3)h\nu e^{-sh\nu/kT}$, $s = 1, 2, 3, \dots$ (eqs. (4.6), (4.7)) can be interpreted in terms of thermodynamically-independent objects (light molecules) with energy $sh\nu$, since their partial entropies ε_i add up to the total entropy \mathfrak{E} of a black body (sec. (4.2.1)). If this holds, then correlations of light from such an object should not exist; the light should be completely incoherent and thus unable to produce any interference effects at all (Laue 1906).^{63,64}

⁶¹For an extended introduction, see the lucid description of the physics involved in sec. 4.2 of (Mandel and Wolf 1965).

⁶²References to these experiments and as well its semiclassical as its field theoretical treatment can be found in (Mandel and Wolf 1995, sec. 9.9 and 14.6.1).

⁶³Because of their brevity and beauty, we repeat here von Laue's arguments, which refer partly to Planck: "The entropy \mathfrak{E} of a system is [...] connected with its probability W through the equation $\mathfrak{E} = k \cdot \log W$. For two subsystems 1 and 2, which add up to the total one, the partial entropies are accordingly: $\mathfrak{E}_1 = k \cdot \log W_1$ and $\mathfrak{E}_2 = k \cdot \log W_2$. Out of these three equations, the addition theorem for entropies $\mathfrak{E} = \mathfrak{E}_1 + \mathfrak{E}_2$ ensues if and only if $W = W_1 \cdot W_2$. A complete independence of each of the two systems would be a necessary and sufficient condition. [...] coherent light bundles are, however, not independent of each other at all. Therefore the addition theorem is not valid" (Laue 1906, 374, fn. 4). "(Die Entropie \mathfrak{E} eines Systems ist [...] mit seiner Wahrscheinlichkeit W durch die Gleichung $\mathfrak{E} = k \cdot \log W$ verknüpft. Für zwei Teilsysteme, aus denen das ganze bestehen soll, gilt entsprechend $\mathfrak{E}_1 = k \cdot \log W_1$ und $\mathfrak{E}_2 = k \cdot \log W_2$. Aus diesen drei Gleichungen folgt das Additionstheorem dann und nur dann, wenn $W = W_1 \cdot W_2$ ist; notwendige und hinreichende Bedingung dafür ist, dass die beiden Teilsysteme voneinander vollkommen unabhängig sind, [...]. Kohärente Strahlenbündel sind aber nicht voneinander unabhängig, also gilt für sie das Additionstheorem nicht.)"

⁶⁴Further work on this topic was published at around this time by von Laue (1907a; 1907b) and Ehrenfest (1925, 364, fn. 1). See also sec. 4.7 "Entropy of an Optical Field" in (Mandel and Wolf 1965) for further historical information.

4.4 Obituary

In 1930, Indian theorists R. J. Majumdar and D. S. Kothari published a thorough derivation of the ratio of Einstein's A and B coefficients, following the ideas of Bose's brief first 1924 manuscript (Bose 1924). In passing, they also mention Bothe's considerations to obtain this ratio through the requirement of achieving thermal equilibrium in a black body (Bothe 1923). Here, we will not deal with their work, but instead with their remark that "Bothe created the fairly new concept of light molecules and arrived thus at Planck's formula" (Majumdar and Kothari 1930).⁶⁵ It was probably common knowledge, at least in India, that the Indian theorist Kulesh Ch. Kar also shared the same opinion (Kar 1927). Wolfke, now a professor at the Technical University in Warsaw, reacted angrily in a short note (M. Wolfke 1930) listing all the authors who had used the concept of "light atoms" or "light molecules": Ioffe (1911), Wolfke (1921), de Broglie (1922b), Bateman (1923) and Bothe (1923).

Around this time, Wolfke still maintained his relations with Einstein. The Einstein Archive Online lists a total of nine letters exchanged between Wolfke and Einstein from 1925 to 1931. Wolfke's letter dated 13 December 1925 refers to a recent meeting with Einstein in Berlin.⁶⁶ Some of the other letters deal with Wolfke's ideas to experimentally find a signal pointing to the existence of (static) light molecules. Moreover, at Wolfke's request, Einstein presented an investigation entitled "Über die statistischen Eigenschaften der Strahlung" (Einstein 1930) at a meeting of the Prussian Academy of Sciences in which Einstein arrived at a fluctuation formula with a factor two in front of the "wave part", see eq. (4.22). The corresponding manuscript never appeared.

In two letters dated 27 and 29 December 1930,⁶⁷ Wolfke tried to corroborate Einstein's result with his own calculations. In his second letter, he was almost headed in the right direction. He started with Bose's expression for the occupation probability of a phase space cell with s light quanta (Bose 1924) and identified this probability with the number density n_s of s -fold light multiples with energy $sh\nu$.⁶⁸ By this means, he arrived at Bothe's expression for n_s , see eq. (4.28), however, with one essential difference. According to his interpretation of the s -fold multiples in terms of static light molecules, he assumed that s runs only from

⁶⁵"Bothe schuf das einigermaßen neue Konzept der Lichtmoleküle und gelangte so zur Planck'schen Formel."

⁶⁶Wolfke to Einstein, 13 December 1925, 23 507, Einstein Archive Online.

⁶⁷Wolfke to Einstein, 27 and 29 December 1930, 23 517 and 23 519, Einstein Archive Online.

⁶⁸Since this approach is similar to Bothe's unpublished manuscript, see footnote 38 in sec. (4.3.2), we denote the number densities again using fractured letters.

1 to ∞ , instead of from 0 to ∞ as Bothe interpreted. Calculating the total energy density by

$$u_\nu(T) = \sum_{s=1}^{\infty} sh\nu \cdot n_{\nu,s} \quad (4.36)$$

and its averaged energy fluctuation density squared $\overline{\Delta_\nu^2}$ through (Bothe 1923, eq. 1)

$$\overline{\Delta_\nu^2} = \frac{1}{V} kT^2 \left(\frac{du_\nu}{dT} \right) \quad (4.37)$$

he obtained the correct result for the total energy density, but a factor of two in front of the wave part of the averaged energy fluctuation density squared, see eq. (4.22), as had Einstein. One can straightforwardly identify this odd result with the missing $s = 0$ term and thus with the interpretation of the n_s . Wolfke drew the proper conclusion “[...] that on the basis of the Bose – Einstein statistics it is not permissible to interpret the radiation field as consisting of *multiple quanta*.”⁶⁹

Einstein probably made the same mistake, since we know from a brief remark at the bottom of Wolfke’s 29 December 1930 letter that he calculated the energy fluctuations assuming a mixture of locally independent multiple quanta.⁷⁰ After returning to Berlin, Einstein responded in detail in a letter dated 10 April 1931⁷¹ by saying “At first, I was very impressed by your argument. But then I found a problem.”⁷² Einstein then showed explicitly that the concept of static light molecules violates Boltzmann’s law.⁷³ No reply from Wolfke is known.

The story of “light molecules” related here ends finally in 1946. Wolfke survived the German occupation of Poland in Warsaw. Here he taught, partly at a lower level, at the Polytechnic University where he was a professor. After the war, he was eager to resume his contacts with the West, in particular to Sweden, Switzerland and, if possible, to the United States. From Stockholm, he sent a manuscript entitled “Über Multiphotone”⁷⁴ to Einstein in Princeton asking him for critical remarks. Wolfke, still drawing from his 1921 publication on black-body radiation (M. Wolfke 1921), showed that the number of phase cells that

⁶⁹ “[...], dass es auf Grund der Bose-Einsteinschen Statistik nicht zulässig ist das Strahlungsfeld, als aus “Mehrfachquanten” bestehend, zu interpretieren” (see footnote 67).

⁷⁰ Wolfke to Einstein, 29 December 1930, 23 521, Einstein Archive Online.

⁷¹ Einstein to Wolfke, 10 April 1931, 23 522, Einstein Archive Online.

⁷² “Ihr Argument hat zu erst großen Eindruck auf mich gemacht. Aber ich finde eine Schwierigkeit.”

⁷³ This letter prompted us to explicitly show that Bothe’s interpretation of the $n_{\nu,s}$ as quantum multiples, including the $s = 0$ ones, is in accordance with Boltzmann statistics.

⁷⁴ “About Multiphotons”, APAN: III–71, 23, 5–12. According to the then general use of the word “photon,” rather than “light quantum,” he used the phrase “multiphoton” rather than “light molecule” (M. Wolfke 1946a).

contain s quanta of energy $h\nu$ equals the number density of multiphotons n_s with energy $sh\nu$, see eq. (4.9) according to Bose-Einstein statistics.⁷⁵ Subsequently, he discussed potentially feasible experiments using the light emitted by black bodies of extremely high temperatures, such as fixed stars, to enhance the ratio of $s = 2$ to $s = 1$ multiphotons, see eq. (4.10).

4 1

Stockholm. Sweden.
Odengatan 46.
Larssons-Pensionat.

Stockholm, d. 10. VII. 1946.

Herrn Prof. Dr. A. EINSTEIN,
Princeton N.Y. U.S.A.

Sehr geehrter Herr Professor!

Bezugnehmend auf unsere Korrespondenz im Jahre 1930 über die "Lichtmoleküle", erlaube ich mir Ihnen die beiliegende Abhandlung zur gefälligen Einsichtsnahme zu übersenden. Ich wäre glücklich, wenn Sie dazu Zeit und Lust finden würden mir Ihre Meinung mitteilen zu wollen.

Ich bin in Stockholm auf Durchreise, bleibe aber hier noch einige Wochen.

Mit vorzüglicher Hochachtung

Wolke

Figure 4.4: Letter from Wolfke to Einstein dated 10 July 1946, APAN, III-71/IV. 23,5.

⁷⁵Wolfke neglected—for whatever reason—Bothe's (1927b) dynamical treatment and thus the fact that Bothe obtained the same result almost two decades earlier.

In September 1946, Wolfke presented his ideas at a meeting of the Schweizerische Naturforschende Gesellschaft in Zurich.⁷⁶ Previously, on 18 January, de Broglie⁷⁷ had already presented a French version of the manuscript⁷⁸ at the meeting of the Académie des Sciences, Paris, which was published in *Comptes Rendus* (M. Wolfke 1946c).

Einstein answered Wolfke's letter one week later on 17 July 1946.⁷⁹ In this correspondence, Einstein argued against the existence of "light molecules" in discussing the passage of light molecules through a semi-reflecting plate. In doing so, he elaborated at length his argument from his 1909 letter to Lorentz⁸⁰ that "a light ray splits, but a light quantum cannot split without a change in frequency." Obviously Einstein had forgotten his much more convincing argument from his 1931 letter to Wolfke that the existence of static light molecules is not in accordance with Boltzmann's law.

Wolfke replied to Einstein's letter on 17 August 1946 without really responding to Einstein's critical remark. Wolfke passed away in Zurich on 4 May 1947 after suffering a heart attack. After Bothe's withdrawal in 1927, this incident finally brought to an end the discussion on the concept of "light molecules" as a tool for dealing with the correlations in a light quantum gas.

Appendix

Frequency Continuum and Light Molecules

Wolfgang Pauli 1923

In 1923, Wolfgang Pauli (1900–1958) tried to understand whether, and if so how, free electrons with a Maxwellian velocity distribution can achieve thermal equilibrium in a black-body radiation field (Pauli 1923). He assumed that the (relativistically-treated) Compton effect is the mechanism which drives the system into equilibrium, since it allows for a change of the energy and momentum of both light quantum and electron. Of course, the relativistically-formulated conservation laws of energy and momentum had to be fulfilled as well. He identified statistical equilibrium through the condition that each elementary process occurs

⁷⁶ APAN: Wolfke manuscript, 1941, III–71, 20, 10–11. In a report to his authorities in Warsaw, Wolfke writes: "On 8. IX. [1946] I lectured at a meeting of the Schweizerische Naturforschende Gesellschaft in Zurich 'On multiple quanta in Planck's radiation,'" an abbreviated version of which was published in 1946 (M. Wolfke 1946b).

⁷⁷ Wolfke probably had personal relations with de Broglie, who visited Wolfke's institute in Warsaw in 1933 (K. Wolfke 1980).

⁷⁸ APAN: III–71, 20, 10–11.

⁷⁹ Einstein to Wolfke, 17 July 1946, APAN, III–71, 24, 7.

⁸⁰ See the beginning of sec. (4.1).

as often as the time-reversed process. To substantiate this requirement, Pauli devoted quite some effort to the formal description of what a time-reversed process means relativistically.

Apart from such dynamical variables as the momenta of electron and photon, and apart from the solid angle, the rates depend then on a “rate function” (*Häufigkeitsfunktion*) F which ought to depend explicitly on u_ν , the spectral density of an arbitrary radiation field at frequency ν with which the electrons are interacting. At first Pauli tested the plausible ansatz

$$F = Au_\nu, \quad (4.38)$$

whereby the function A ought to depend on the kinematical variables of electron and the light quanta, such as the momenta and energies, but be independent of temperature T . Pauli found that this ansatz leads unambiguously to Wien’s radiation law.

Guided by a qualitative analysis of this apparently limited ansatz, he analyzed the consequences of the, at first glance, rather strange ansatz

$$F = Au_\nu + Bu_\nu u_{\nu'}, \quad (4.39)$$

in which $u_{\nu'}$ denotes the radiation density at the frequency of the Compton-scattered light quantum. Pauli himself commented on this as follows: “At first glance this postulation seems odd, since an interaction of radiation bundles is assumed here, possibly with widely different frequencies”⁸¹ (Pauli 1923, 284). This ansatz causes the Compton scattering process $\nu \rightarrow \nu'$ to occur more frequently if the frequencies ν as well as ν' are present in the radiation field. And indeed, this leads to Planck’s radiation law for a black body.

Einstein and Ehrenfest reacted immediately in the next issue of *Zeitschrift für Physik* (Einstein and Ehrenfest 1923). Obviously, they had had access to Pauli’s manuscript in advance and had submitted their manuscript prior to the appearance of Pauli’s. In subsequent steps, Einstein and Ehrenfest employed a transparent formalism which allowed equilibrium conditions to be described for absorption and emission of radiation, even when the matter part has continuous energy spectra, as in Compton scattering, for example, or for moving atoms or molecules. By defining what “time reversal” in the statistical average might mean, Einstein and Ehrenfest arrived at transition probabilities that were nonlinear in the energy density of the radiation field, in general, up to any order.⁸²

⁸¹“Beim ersten Augenblick könnte diese Forderung befremdend erscheinen, weil hier eine Wechselwirkung von Strahlenbündeln mit unter Umständen weit verschiedenen Frequenzen angenommen wird.”

⁸²For further more recent discussions, see (H. R. Lewis 1973; Lanyi 2003).

Bothe 1924

Bothe also reacted to Pauli's investigations with a formulation of the problem in terms of quantum multiples. He aimed at a proof in which the "B-processes" in Pauli's sense, see eq. (4.39), are formally dispensable (Bothe 1924). We will not enter the formal details of his arguments since they were based on the light multiple concept without the $s = 0$ terms, which as we saw, is problematic, see secs. (4.2) and (4.3.1).

Bothe's final argument was based on two recollections:

1. Pauli (1923) showed that with the A-term alone, see eq. (4.38), the thermalization of a light-quantum gas in a black body through Compton scattering on a thermalized electron gas (Boltzmann distribution) will unambiguously lead to a Wienian energy distribution.
2. Referring to Wolfke (1921), see sec. (4.2.1), de Broglie (1922a), see sec. (4.2.2), and to his own work (Bothe 1923), see sec. (4.3.1), he reminded the reader that Planck's formula may be written as an infinite sum of Wienian terms, each describing an s -fold light multiple:

$$u_{\nu,s} = \frac{8\pi\nu^2}{c^3} h\nu e^{-sh\nu/kT}, \quad s = 1, 2, 3, \dots \quad (4.40)$$

Thus it will suffice to deal with Compton scattering of s -fold quantum multiples as a whole, taking into account only A-processes, see eq. (4.38). Formally, such a process might be written as $sh\nu + e \rightarrow sh\nu' + e'$. The number of s -fold multiples will be conserved and only their frequency will vary. One just has to formally replace in Pauli's calculations (Pauli 1923, 281–282) h with sh , which will result with Wienian terms in $u_{\nu',s}$, the sum of which will yield Planck's formula again and thus thermalized black-body radiation.

Bothe then added a more formal and detailed support of this brief argument. He finished the paper with the remark that in Pauli's derivation, as well as in his own, the number of light quanta is conserved. Moreover, the achievement of thermal equilibrium is independent of the prefactor $8\pi\nu^2/c^3$ in Planck's law.⁸³

Bothe 1925b

In his unpublished manuscript,⁸⁴ Bothe set about remedying this problem. He used the heuristic trick described in sec. (4.3.2). Still using the concept of s -fold

⁸³We take this opportunity to point to Pauli's article in *Handbuch der Physik*, where he discussed the heat balance between atoms and radiation in much more detail in sec. I.2. "Wärmegleichgewicht zwischen Atomen und Strahlung" (Pauli 1926, 9–22).

⁸⁴See footnote 38.

light multiples, he again assumed that the rate of processes in which an s -fold light multiple loses a light quantum is proportional to its energy density $s n_{v,s} h\nu$, but proportional to $(s + 1) n_{v,s} h\nu$ for those processes in which it gains one.⁸⁵

Apart from this assumption, he followed Pauli's strategy (Pauli 1923) and discussed the following two elementary steps in the interaction of light quanta with Maxwellian-energy-distributed electrons: in an interaction of an electron (momentum \mathfrak{G}) with an r -fold light multiple $r h\nu$ of frequency ν the latter loses one light quantum with momentum \mathfrak{F} which is added with momentum \mathfrak{F}_1 to a s -fold light multiple $s h\nu_1$ of frequency ν_1 . The electron scatters under an angle Θ_0 and thus takes care of the momentum and energy conservation ($\mathfrak{G} \rightarrow \mathfrak{G}_1$).⁸⁶ Applying once more the heuristic trick described in sec. (4.3.2), Bothe assumed that the rate of the process is proportional to the energy density $r n_{v,r} h\nu$ of the multiples which lose a light quantum, but proportional to $(s + 1) n_{v_1,s} h\nu_1$ for those which gain one. Denoting for a Maxwellian velocity distribution at temperature T the number of electrons per unit volume within the momentum interval $d\mathfrak{G}$ by $N d\mathfrak{G}$, the rate of these processes will be

$$H = B \cdot r n_{v,r} h\nu \cdot (s + 1) n_{v_1,s} h\nu_1 \cdot d\mathfrak{F} \cdot N d\mathfrak{G} \cdot d\Theta_0. \quad (4.41)$$

The strength B depends on all the relevant variables necessary to describe the process, except for the temperature T . The rate for the time reversed process can be parameterized accordingly

$$H_1 = B_1 (s + 1) n_{v_1,s+1} h\nu_1 \cdot r n_{v,r-1} h\nu \cdot d\mathfrak{F}_1 \cdot N_1 d\mathfrak{G}_1 \cdot d\Theta_0. \quad (4.42)$$

For the argument to be made, it is important only that B and B_1 do not depend on temperature T . To achieve thermal equilibrium it is sufficient that

$$H = H_1. \quad (4.43)$$

Pauli (1923, 281, eq. 25) deduced, from momentum and energy conservation for the electron-light quantum scattering, that the ratio N/N_1 obeys the relation

$$\frac{N}{N_1} = \frac{e^{h\nu/kT}}{e^{h\nu_1/kT}}. \quad (4.44)$$

⁸⁵As before, $s n_{v,s} h\nu$ denotes the fraction of the total radiation density that belongs to s -fold light multiples, see sec. (4.3.2).

⁸⁶All quantities are defined within the normal coordinate system of the individual process.

Now using the distribution for the number of multiples, see eq. (4.28)

$$n_{\nu,s} = \frac{8\pi\nu^2}{c^3} (1 - e^{-h\nu/kT}) e^{-sh\nu/kT}, \quad s = 0, 1, 2, \dots, \quad (4.45)$$

which we have already seen is in accordance with Boltzmann's law, one finds rather easily that the temperature disappears under the condition $H = H_1$, see eq. (4.43). That means thermal equilibrium will be reached independently of the very form of the functions B and B_1 .

We conclude this section of the appendix with an interpretation of the result in terms of light bundles of finite cross section and of very small frequency and opening angle interval as "elementary cells," in which the light quanta are embedded, see sec. (4.3.3). The process denoted by H , see eq. (4.41), describes a scattering process in which, out of an elementary bundle with r light quanta of frequency ν , one quantum is scattered into another bundle (called bundle 1) already hosting s light quanta with frequency ν_1 . The electron takes care of the energy and momentum conservation. H_1 describes the time-reversed process. It is now important that also ($r = 0$)-terms contribute to H , which means that in the bundles of H_1 , light quanta may appear spontaneously. For example, in the time-reversed process denoted by H_1 , a light quantum disappears completely for $r = 1$ and is created spontaneously for $s = 0$.

Abbreviations and Archives

APAN	Archive of the Polish Academy of Sciences, Warsaw
AMPG	Archive of the Max Planck Society, Berlin
Einstein Archive Online	Albert Einstein Archives and the David and Fela Shapell Digitization Project at the Jewish National & University Library, the Hebrew University of Jerusalem, http://www.alberteinstein.info/database/

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