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Quantum Theory of Dispersion (Continued). Parts VI and VII

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VI. PHENOMENA NEAR RESONANCE

§1. Simplification introduced near resonance

When the frequency of a light quantum is very close to a possible absorption frequency the probabilities of absorption and of scattering become large. The formulas which we have derived so far cannot be applied because they give infinite scattering when the two frequencies coincide. The effect of interaction between matter and radiation is not small in this case so that a special consideration near resonance is necessary.

Qualitatively the effects near resonance may be described as follows: If an atom is in an excited state, it may emit a light quantum when it jumps to a lower state. The probability of finding this light quantum increases with the time. Simultaneously the probability of finding the atom in the excited state decreases. The smaller the chance of finding the atom in the excited state, the slower is the rate at which the probability of finding a light quantum increases. The larger the chance of finding a light quantum, the more likely it is that the light quantum is reabsorbed by the atom and the slower the rate of decrease of the probability of excitation. These effects are seen to cooperate in slowing down the emission.

Similarly, if a light quantum is incident on an atom, there is a chance of its being absorbed and of the atom being excited. The larger the chance that the atom is excited, the more likely it is that another light quantum (of approximately the original frequency) is emitted. This possibility of emission slows down the rate at which the excitation of the atom increases. The effect will be seen to be much the same as though the atom were a harmonic oscillator of natural frequency equal to the absorption frequency and damped by its own radiation.

We have to consider only light quanta of frequencies in the neighborhood of the absorption frequency. The probability of finding

other light quanta is relatively small. Their neglect is expected to spoil the validity of the results when the frequency of incident light differs appreciably from the absorption frequency. Under these circumstances, however, it is not necessary to take into account the damping, so that the ordinary emission, absorption and scattering formulas may be used.

The physical results may be seen by considering the probability amplitudes of states of approximately equal energy. In fact, the energy operator is constant so that the quantum mean of the energy as well as of all its powers does not change with time. If at $t=0$ the energy has a certain statistical distribution around a sharp and narrow maximum, the same distribution persists for other values of t. The interaction energy between matter and radiation is considered as small. Equal energies of the whole coupled system are, therefore, treated as approximately equal to the sums of the unperturbed energies of the coupled parts.

There is little doubt that the interaction between radiation and matter is small and that it contributes only an insignificant part to the whole energy. A closer examination of the equations of the present theory shows, however, that they give an infinite value for the interaction energy. This is a defect of the present theory. It will be discussed in VII, §6. For the present, we are satisfied with the supposition that our instinct is more correct than our equations and that the equations will give good results if we use them as though the interaction energy were small.

§2. Emission of a resonance line

We consider first the emission of light from an atom. At the time $t=0$ the atom is excited and there are no light quanta. This case has been already discussed under the heading of spontaneous emission by means of Eq. (111). This discussion applies only for times short in comparison with the mean life of an atom and long in comparison with the period of the emitted light waves. It will be remembered that these restrictions were introduced when the probability of the atom being excited was supposed to be constant. We now remove the restrictions of considering times short in comparison with the mean life. Let the excited state of the atom be n' and the normal n, their energies being $h\nu(n')$ and $h\nu(n)$, respectively, and the energy difference being written as $h(\nu(n')-\nu(n))$. The probability amplitude for the atom to be in the state n' while there are no light quanta we denote by $ce^{-2\pi i\nu(n)t}$. The probability amplitude for the atom to be in the state n, and there being one light quantum of the kind s, we denote by $c_s e^{-2\pi i\nu(n)t}$. By (110) we have to satisfy the equation

$$\begin{aligned}(d/2\pi i dt+\nu_s)c_s&=A_s c\\ (d/2\pi i dt+\nu(n'n))c&=\sum A_s{}^* c_s\end{aligned}\qquad(122)$$

where

$$A_s=\alpha^s{}_{nn'}(\nu_s V)^{-\frac{1}{2}}\cong e^{-i\mathbf{kR}}(2\pi h\nu_s V)^{-\frac{1}{2}}(\sum e_i\dot{\mathbf{r}}_i\mathbf{f}_s)_{nn'}.\qquad(122')$$

[We, thus, neglect deliberately all φ's in (110) except

$$\varphi(n';0,0,\cdots)=ce^{-2\pi i\nu(n)t}\qquad\text{and}\qquad\varphi(n;0,0,\cdots 1_s,0,0,\cdots)=c_s e^{-2\pi i\nu(n)t}$$

with $\nu_s\cong\nu(n'n)$ since these probability amplitudes refer to the only important states of approximately the initial energy.]

Approximate solutions of (122) have been found by Hoyt[29] and by Weisskopf and Wigner.[30] The method consists in supposing that to a good approximation $c=e^{-2\pi i(\nu(n'n)-i\Gamma)}$ where Γ is a real constant. This is suggested by the fact that if all c_s were zero the last equation (122) would be satisfied by $c=e^{-2\pi i\nu(n'n)t}$. The analogy to the classical oscillator leads one to expect that the absolute value of c would decrease exponentially. For this reason the factor $e^{-2\pi i\Gamma t}$ is also introduced. There are as many equations of the first type in (122) as there are kinds of light quanta. Since at the time $t=0$ every $c_s=0$, we can use this value of c_s as the initial condition in all the differential equations. Having also the right-

[29] F. Hoyt, Phys. Rev. **36**, 860 (1930).

[30] V. Weisskopf and E. Wigner, Zeits. f. Physik **63**, 54 (1930). See here discussion of emissions ending in excited states. The width of the line is shown to be the sum of the widths of the initial and final levels; V. Weisskopf, Ann. d. Physik **9**, 23 (1931).

hand side as $A_s e^{-2\pi i(\nu(n'n)-i\Gamma)t}$, the values of c_s follow at once as

$$c_s = A_s \frac{e^{-2\pi i(\nu(n'n)-i\Gamma)t} - e^{-2\pi i\nu_s t}}{\nu_s - \nu(n'n) + i\Gamma}.$$

These expressions must now be substituted into the last equation (122) because it does not follow, so far, that this equation is satisfied. On substitution we obtain

$$i\Gamma = \sum_s |A_s|^2 \frac{1 - e^{-2\pi i(\nu_s - \nu(n'n)+i\Gamma)t}}{\nu_s - \nu(n'n) + i\Gamma}. \quad (123)$$

By (109)

$$\alpha^s{}_{nn'} = (2\pi h)^{-\frac{1}{2}}(\mathbf{f}_s \sum (e_i/2)(e^{-i\mathbf{k}_s\mathbf{r}_i}\dot{\mathbf{r}}_i + \dot{\mathbf{r}}_i e^{-i\mathbf{k}_s\mathbf{r}_i}))_{nn'}$$

in the limit of a very concentrated $D^i{}_{r,r'}$. For wave-lengths long in comparison with atomic dimensions we may take

$$\alpha^s{}_{nn'} = (2\pi h)^{-\frac{1}{2}} e^{-i\mathbf{k}_s\mathbf{R}} (\mathbf{f}_s \sum_i e_i \dot{\mathbf{r}}_i)_{nn'},$$

where $\mathbf{R}$ is the vector from the origin to the center of mass of the atom. Substituting this into (122′) we have

$$|A_s|^2 = |(\mathbf{f}_s \sum e_i \dot{\mathbf{r}}_i)_{nn'}|^2/(2\pi h \nu_s V). \quad (123')$$

This number is practically independent of ν_s within the natural breadth of the spectral line because this breadth is very small in comparison with ν_s. It depends much more strongly on the polarization vector $\mathbf{f}_s$.

It is now important to realize that within a very narrow range of ν_s the polarization vector $\mathbf{f}_s$ may have all sorts of directions. This follows from the fact that there may be light quanta of the same frequency moving in arbitrary directions with arbitrary polarizations. In the summation over s we may, therefore, average first over the different directions $\mathbf{f}_s$. The resultant average is

$$\overline{|A_s|^2} = \tfrac{1}{3}\{|(\sum e_i \dot{x}_i)_{nn'}|^2 + |(\sum e_i \dot{y}_i)_{nn'}|^2 + |(\sum e_i \dot{z}_i)_{nn'}|^2\}/(2\pi h \nu_s V). \quad (123'')$$

It is seen to be practically independent of the frequency if the approximation (123′) applies. It is also practically independent of ν_s if this approximation does not apply. However, then (123″) does not hold since the retardation factors $e^{-i\mathbf{k}_s\mathbf{r}_i}$ may not be treated as constants. Taking $|A_s|^2$ outside of the summation sign the summation is replaced by integration $[\sum_s \to \int d\nu_s/\Delta\nu_s]$ where $(\Delta\nu_s)^{-1}$ is the number of possible light quanta per unit frequency range at $\nu_s[(\Delta\nu_s)^{-1} = 8\pi\nu_s^2/c^3]$. It is justifiable from the present point of view to treat ν_s^2 as a constant $= \nu^2{}_{n'n}$ and to extend the limits of integration from $-\infty$ to $+\infty$. Doing so one obtains

$$\Gamma = \pi \overline{|A_s|^2}/\Delta\nu. \quad (124)$$

This expression is independent of the time. This proves that all Eqs. (122) are satisfied, provided Γ has the value just given. For future reference we summarize the results

$$c = e^{-2\pi i(\nu(n'n)-i\Gamma)t}$$

$$c_s = A_s \frac{(e^{-2\pi i(\nu(n'n)-i\Gamma)t} - e^{-2\pi i\nu_s t})}{(\nu_s - \nu(n'n) + i\Gamma)}. \quad (125)$$

The number of light quanta present is $\sum_s |c_s|^2$. Using the expressions for c_s in (125) replacing the summation by integration as before we obtain

$$\sum_s |c_s|^2 = \frac{\pi}{\Gamma(\Delta\nu)} \overline{|A_s|^2}(1 - e^{-4\pi\Gamma t}) = 1 - e^{-4\pi\Gamma t}$$

by (124). At the same time

$$|c|^2 = e^{-4\pi\Gamma t}.$$

Thus,

$$|c|^2 + \sum_s |c_s|^2 = 1.$$

This relation follows also as a general consequence of (122) and the initial conditions. It is satisfactory to see that the approximations made have not impaired its validity.

The mean life of the atom in the excited state is $(1/4\pi\Gamma)$. At the time $t = 0$ the chance of emitting a light quantum per second is $4\pi\Gamma$. By using (123″) it is seen that $4\pi\Gamma$ is equal to the expression (112). The mean life of the atom is thus given by the reciprocal of Einstein's emission probabilities. For $t \gg (1/4\pi\Gamma)$ the number of light quanta per unit frequency range is by (125)

$$[|A_s|^2/(\Delta\nu)]/[(\nu_s - \nu(n'n))^2 + \Gamma^2].$$

The half-value breadth of the emission line is thus 2Γ or $(1/2\pi)^{\text{th}}$ part of the emission probability. This is *exactly* the *relation* which holds *for*

the *classical harmonic oscillator*. More generally, if t is not very large in comparison with the mean life, the number of light quanta per unit frequency range is given by

$$\frac{1+e^{-4\pi\Gamma t}-2e^{-2\pi\Gamma t}\cos 2\pi(\nu(n'n)-\nu_s)t}{(\nu_s-\nu(n'n))^2+\Gamma^2}\frac{|A_s|^2}{(\Delta\nu)}.$$

In particular, if $\nu(n'n)-\nu_s=0$, the numerator of this expression is $(1-e^{-2\pi\Gamma t})^2$. Thus, at the core of the line the number of light quanta per unit frequency range increases at the time $t=0$ as t^2. Throughout the whole frequency range the number of light quanta at $t=0$ increases as t the main contributions arising from regions $|\nu_s-\nu(n'n)|\gg 4\pi\Gamma$. The *energy first appears, therefore, in the wings of the line* and only later the maximum at the core is built up.[31] These relations are also exactly similar to those for a classical oscillator provided one averages over the initial phase of the oscillator.

Within the approximations of sharp resonance the relations between Einstein emission coefficient, the mean life, and the half-value breadth are independent of the assumption that the effect of retardation is negligible. The specialization to long wave-lengths made in (123′) is not essential for the validity of these relations.

§3. Absorption by a resonance line

To discuss the relations in absorption we consider one light quantum of the kind s being present at the time $t=0$. We suppose that the atom is at that time in the normal state n and that there are no other light quanta present. The frequency $\nu(s_0)$ we suppose to be in the general neighborhood of $\nu(n'n)$. Our calculation will apply if $|\nu(n'n)-\nu(s_0)|$ is of the general order of Γ (e.g., $\Gamma/100$ or 100Γ) but not in cases where it is at all comparable to $\nu(n'n)$ itself. At later times there may be a chance of the light quantum being absorbed and the atom being in the state n'. The probability amplitude for this state is c. There may also appear light quanta of other kinds $s'\neq s_0$, the atom simultaneously returning into n. The probability amplitude of these states is c_s.

We look for a solution of the type

$$c_{s_0}=e^{-2\pi i(\nu_0-i\gamma)t}, \tag{126′}$$

where ν_0 we expect to be approximately ν_{s_0} and γ we expect to be very small. Then it follows from the s_0^{th} Eq. (122) that

$$c=[(\nu(s_0)-\nu_0+i\gamma)/A_{s_0}]e^{-2\pi i(\nu_0-i\gamma)t}. \tag{126″}$$

Using this value in the equations for c_s and imposing the initial condition $c_s=0(s\neq s_0)$ we have

$$c_s=(A_s/A_{s_0})[(\nu(s_0)-\nu_0+i\gamma)/(\nu_s-\nu_0+i\gamma)][e^{-2\pi i(\nu_0-i\gamma)t}-e^{-2\pi i\nu_s t}]. \tag{126‴}$$

We substitute the expressions (126) into the last equation (122), replace the summation by integration, as before, and obtain

$$[\nu(n'n)-\nu_0-i(\Gamma-\gamma)](\nu(s_0)-\nu_0+i\gamma)=|A_{s_0}|^2e^{-2\pi i(\nu(s_0)-\nu_0+i\gamma)t}$$

having replaced the summation over *all* values of s by an integral. The solutions (126) do not satisfy the equations exactly. However, for $t\ll(2\pi)^{-1}[(\nu(s_0)-\nu_0)^2+\gamma^2]^{-\frac{1}{2}}$ they are satisfactory. We choose a γ so as to satisfy the above condition for such values of t, i.e., so as to have

$$[\nu(n'n)-\nu_0-i(\Gamma-\gamma)](\nu(s_0)-\nu_0+i\gamma)=|A_{s_0}|^2. \tag{127}$$

This gives

$$\gamma\cong|A_{s_0}|^2\Gamma/[(\nu(n'n)-\nu(s_0)^2)+\Gamma^2];$$
$$\nu_0\cong\nu(s_0)+(\gamma/\Gamma)(\nu(s_0)-\nu(n'n)). \tag{127′}$$

The maximum value of γ is

$$\frac{|A_{s_0}|^2}{\Gamma}=\frac{|A_{s_0}|^2}{\overline{|A_s|^2}}\frac{\Delta\nu}{\pi}.$$

However,

$$\Delta\nu=(\lambda/L)^2(c/8\pi L),$$

where L is the side of the fundamental cube. The time constant $(4\pi\gamma)^{-1}$ is, thus, always greater than

$$(2L/c)(L/\lambda)^2\overline{|A_s|^2}/|A_{s_0}|^2.$$

The fraction $\overline{|A_s|^2}/|A_{s_0}|^2$ is of the order of magnitude of 1. The fraction (L/c) is the time

[31] Cf. P. A. M. Dirac, *Case of Resonance*, Proc. Roy Soc. **A114**, 710 (1927).

taken by light to travel through the cube. The time during which the factor $e^{-4\pi\gamma t}$ may be replaced by 1 is seen to be exceedingly great in comparison with the time taken for light to travel through the cube.

The derivation of (127) is valid only if

$$t \ll 1/\{2\pi[(\nu(s_0)-\nu_0)^2+\gamma^2]^{\frac{1}{2}}\} \leqq [(\nu(n'n)-\nu(s_0))^2+\Gamma^2]^{\frac{1}{2}}/(2\pi|A_{s^0}|^2).$$

The minimum value of this upper limit is $\Gamma/(2\pi|A_{s_0}|^2)$, which is again of the same order of magnitude as before. There can be no physical interest in times so much larger than the time taken for light to traverse the fundamental cube because all the physical things supposed to exist in one fundamental cube are also supposed to exist in all others. Eqs. (126) together with (127) are, therefore, sufficiently good approximations.

The solutions (126) represent a condition somewhat analogous to forced oscillations of a classical harmonic oscillator under the influence of a purely sinusoidal impressed wave. The quantity c is analogous to the amplitude of classical oscillations. The scattered radiation must be thought of in this analogy to have been removed at the time $t=0$ and to build up as time goes on. It does not correspond to the case of the atom being initially in the normal state. It is clear that the latter condition can be represented by superposing (125) with a proper coefficient with the solution (126). The result is

$$c_{s_0}=e^{-2\pi i(\nu_0-i\gamma)t}-\frac{|A_{s_0}|^2}{\nu(n'n)-\nu_0-i(\Gamma-\gamma)}\frac{e^{-2\pi i(\nu(n'n)-i\Gamma)t}-e^{-2\pi i\nu(s_0)t}}{\nu(s_0)-\nu(n'n)+i\Gamma}, \tag{128'}$$

$$c=A_{s_0}\frac{e^{-2\pi i(\nu_0-i\gamma)t}-e^{-2\pi i(\nu(n'n)-i\Gamma)t}}{\nu(n'n)-\nu_0-i(\Gamma-\gamma)}, \tag{128''}$$

$$c_s=A_sA_{s_0}{}^*\left[\frac{e^{-2\pi i(\nu_0-i\gamma)t}}{[\nu(n'n)-\nu_0-i(\Gamma-\gamma)](\nu_s-\nu_0+i\gamma)}-\frac{e^{-2\pi i(\nu(n'n)-i\Gamma)t}}{[\nu(n'n)-\nu_0-i(\Gamma-\gamma)](\nu_s-\nu(n'n)+i\Gamma)}\right.$$
$$\left.+\frac{e^{-2\pi i\nu_s t}}{(\nu_s-\nu_0+i\gamma)(\nu_s-\nu(n'n)+i\Gamma)}\right] \quad (s\neq s_0). \tag{128'''}$$

Here the small number $\nu(s_0)-\nu_0+i\gamma$ has been eliminated by means of (127). The emission solutions (125) and absorption solutions (128) when superposed with proper coefficients give general solutions of (122) for arbitrary initial values of c_s, c.

The main features of the state represented by (128) may be seen from the behavior of $|c|^2$. We have

$$|c|^2=|A_{s_0}|^2\frac{e^{-4\pi\gamma t}+e^{-4\pi\Gamma t}-2e^{-2\pi(\Gamma+\gamma)t}\cos 2\pi(\nu(n'n)-\nu_0)t}{(\nu(n'n)-\nu_0)^2+(\Gamma-\gamma)^2}. \tag{129}$$

If $t\ll(2\pi\gamma)^{-1}$ we may use instead

$$|c|^2=|A_{s_0}|^2\frac{1+e^{-4\pi\Gamma t}-2e^{-2\pi\Gamma t}\cos 2\pi(\nu(n'n)-\nu(s_0))t}{(\nu(n'n)-\nu(s_0))^2+\Gamma^2}. \tag{129'}$$

This expression vanishes for $t=0$ as it should. After t exceeds $(2\pi\Gamma)^{-1}$ it attains a stationary value

$$|A_{s_0}|^2/[(\nu(n'n)-\nu(s_0))^2+\Gamma^2].$$

If we wait still longer $|c|^2$ decreases on account of the terms in $e^{-2\pi\gamma t}$, $e^{-4\pi\gamma t}$. Thus for $t\gg(2\pi\Gamma)^{-1}$, to the first power of γt

$$|c|^2\leqq\frac{|A_{s_0}|^2}{(\nu(n'n)-\nu(s_0))^2+\Gamma^2}-4\pi|A_{s_0}|^4\frac{\Gamma t}{[(\nu(n'n)-\nu(s_0))^2+\Gamma^2]^2}; \quad (t\gg(2\pi\Gamma)^{-1}, \tag{129''}$$

the approximate value of γ given by (127′) having been used. Thus, $|c|^2$ first increases to an almost stationary value following (129′). After the transient represented by the two last terms in (129′) dies down $|c|^2$ decreases slowly following (129″). According to (129′) the absorption probability at $t=0$ depends on $\nu(n'n)-\nu(s_0)$. If $\nu(n'n)=\nu(s_0)$ we have exact resonance of the incident radiation with the transition $n\rightarrow n'$. In this case the numerator of (129′) is

$$(1-e^{-2\pi\Gamma t})^2,$$

giving an initial increase proportional to t^2. If, however, (129′) is averaged over different values of the incident frequency $\nu(s_0)$ in the neighborhood of $\nu(n'n)$ we obtain

$$\overline{|c|^2}=\overline{|A_{s_0}|^2}(\pi/\Gamma)(1-e^{-4\pi\Gamma t}).$$

The initial rate of increase of $\overline{|c|^2}$ (i.e., the absorption probability) is, thus,

$$4\pi^2\overline{|A_{s_0}|^2}=4\pi\Gamma\Delta\nu.$$

The spontaneous emission probability is $4\pi\Gamma$. The ratio is in agreement with Einstein's relation between emission and absorption probabilities.

The probability amplitudes $c_s(s\neq s_0)$ determine the probability of finding a scattered light quantum. The sum of the squares of their absolute values may be determined by integration. A simple calculation shows that the summation, when extended over the scattered light quanta, gives

$$(\pi/\Delta\nu)|A_{s_0}|^2\overline{|A_s|^2}\left\{\frac{\Gamma+\gamma-\Gamma e^{-4\pi\gamma t}-re^{-4\pi\Gamma t}}{[a^2+(\Gamma+\gamma)^2]\Gamma\gamma}-4\frac{a\sin 2\pi at-(\Gamma+\gamma)\cos 2\pi at}{[a^2+(\Gamma-\gamma)^2][a^2+(\Gamma+\gamma)^2]}e^{-2\pi(\Gamma+\gamma)t}\right\},$$

where

$$a=\nu_0-\nu(n'n).$$

In taking this sum we have used expression (128‴) also for $s=s_0$. This, however, is not important because $(1/\Delta\nu)$ is a large number, so that making a mistake in counting of one light quantum too many makes little difference. The above expression is to a sufficient approximation

$$\Gamma|A_{s_0}|^2\left\{\frac{1-e^{-4\pi\gamma t}}{\Gamma[a^2+(\Gamma+\gamma)^2]}+\frac{4\pi t}{a^2+(\Gamma+\gamma)^2}-4e^{-2\pi\Gamma t}\frac{a\sin 2\pi at-\Gamma\cos 2\pi at}{[a^2+\Gamma^2]^2}\right\}.$$

For $t\gg(2\pi\Gamma)^{-1}$ the term in t predominates. In this condition the number of light quanta scattered per second is

$$4\pi\Gamma|A_{s_0}|^2/[a^2+(\Gamma+\gamma)^2],$$

which corresponds to $|A_{s_0}|^2/[a^2+(\Gamma+\gamma)^2]$ atoms emitting with the probability $4\pi\Gamma$ per second.

The expression (128‴) contains a term in $e^{-2\pi\Gamma t}$. As ν_s varies its absolute value approaches a maximum when $\nu_s=\nu(n'n)$. The sharpness of the maximum is such that the half breadth is 2Γ. This term contributes, therefore, light quanta of the emission frequency $\nu(n'n)$ with a spread of approximately Γ to either side. The term in $e^{-2\pi i(\nu_0-i\gamma)t}$ becomes large when $\nu_s=\nu_0$, i.e., when the scattered frequency is approximately equal to the incident. The half-value breadth of its absolute value is 2γ. The frequencies represented by it are, therefore, very closely grouped around the incident frequency. Finally the last term becomes large both when $\nu_s=\nu_0$ and when $\nu_s=\nu(n'n)$. It is a linear combination of two terms one of which resonates at ν_0 and another at $\nu(n'n)$. The term in $e^{-2\pi\Gamma t}$ is very similar to the *impulse oscillations* of a classical oscillator under the influence of a harmonic force applied suddenly. The vibrations may then be thought of as the result of superposing a damped oscillation with the natural frequency of the oscillator and an undamped forced vibration with the frequency of the impressed force. In the initial stages of motion the oscillator radiates waves of its own natural frequency in addition to those having the frequency of the impressed force. The distribution of energy among radiated quanta may be seen best from $|c_s|^2$ which may be put into the form

$$|c_s|^2=\frac{|A_{s_0}|^2|A_s|^2}{a^2+(\Gamma-\gamma)^2}\left|\frac{e^{2\pi i(x+i\gamma)t}-1}{x+i\gamma}-\frac{e^{2\pi i(x+a+i\Gamma)t}-1}{x+a+i\Gamma}\right|^2,$$

where $x=\nu_s-\nu(s_0)$, $a=\nu_0-\nu(n'n)$.

§4. Absorption and scattering by an atom or radiation emitted by another atom

We consider two atoms located sufficiently far from each other to make the influence of direct electrostatic and magnetic actions negligible in comparison with the effect of radiated light quanta. Initially we suppose one atom (denoted by I) to be excited to a state n_I' from which it may fall to a state n_I. The other atom (denoted by II) we suppose to be initially in its normal state n_{II} from which it may rise by absorption to an excited state n_{II}'. The frequencies which correspond to these transitions will be written as ν_I, ν_{II}, respectively. We concern ourselves with those cases in which only the transitions $n_I'\to n_I$ and $n_{II}\to n_{II}'$ are of importance, i.e., with such absorbing atoms which do not have other absorption lines from n_{II} in the neighborhood of ν_I and with emitting atoms for which ν_I is the only strong emission line from n_I'. The frequencies ν_I, ν_{II} must, therefore, be not too far apart. They may, however, be farther apart than the half-value breadth of either line.

The probability amplitudes which we must take into account are, $e^{-2\pi i(\nu(n_I)+\nu(n_{II}))t}c_s$ = probability amplitude for atoms I, II to be in normal states and for there being one light quantum of type s, $e^{-2\pi i(\nu(n_I)+\nu(n_{II}))t}c_I$ = probability amplitude for atom I to be in the excited state n_I', for atom II to be in the normal state and for there being no light quanta, $e^{-2\pi i(\nu(n_I)+\nu(n_{II}))t}c_{II}$ = probability amplitude for atom II to be in excited state n_{II}', for atom I to be in normal state and for there being no light quanta.

For any type of light quantum the two atoms have definite interaction constants A_s (see (122')). Their values for I and II, respectively, will be written A_s^I, A_s^{II}. The time dependence of the probability amplitude is now governed by the immediate extension of (122)

$$\begin{aligned}(d/2\pi i dt+\nu_s)c_s&=A_s^Ic_I+A_s^{II}c_{II},\\ (d/2\pi i dt+\nu_I)c_I&=\textstyle\sum_s(A_s^I)^*c_s,\\ (d/2\pi i dt+\nu_{II})c_{II}&=\textstyle\sum_s(A_s^{II})^*c_s.\end{aligned}\qquad(130)$$

At $t=0$, we take in accordance with the above $c_I=1$, $c_{II}=c_s=0$. Supposing for the moment that the effect of c_{II} on c_I is small, we may consider (125) as a zeroth approximation to the values of c_I and c_s.

$$\begin{aligned}c_I^{(0)}&=e^{-2\pi i(\nu_I-i\Gamma_I)t},\\ c_s^{(0)}&=A_s^I\frac{e^{-2\pi i(\nu_I-i\Gamma_I)t}-e^{-2\pi i\nu_s t}}{\nu_s-\nu_I+i\Gamma_I}.\end{aligned}\qquad(130')$$

This would be the solution of (130) if all the A_s^{II} were zero, then c_{II} would also remain zero. In the next (first) approximation we take the A_s^{II} into account. The additions to c_{II} and c_s which result, we call $c_{II}^{(1)}$, $c_s^{(1)}$. In this approximation

$$c_{II}=c_{II}^{(1)},\quad c_s=c_s^{(0)}+c_s^{(1)}.$$

The manipulation of equations (130) is dictated to us now by physical considerations. Atom II will be excited by the radiation from I represented by $c_s^{(0)}$. The probability of excitation will depend on the distance between the atoms, the spontaneous probability of absorption and also on the rate at which atom II is able to radiate its absorbed energy. For the larger this rate, the smaller we expect to be the value of c_{II}. The reaction of the radiation from II on I we expect to be small if the distance between them is large. It does not interest us at present because it may be described as the rescattering by I of the radiation scattered by II. In order to keep the calculation simple, we neglect, therefore, the changes in $c_I^{(0}$ due to $c_s^{(1)}$, thus neglecting the rescattering by I. The second equation (130) thus need not be considered any more. The defining equations for $c_{II}^{(1)}$, $c_s^{(1)}$ are, thus,

$$\begin{aligned}(d/2\pi i dt+\nu_s)c_s^{(1)}&=A_s^{II}c_{II}^{(1)},\\ (d/2\pi i dt+\nu_{II})c_{II}^{(1)}&=\textstyle\sum_s(A_s^{II})^*(c_s^{(0)}+c_s^{(1)}).\end{aligned}\qquad(130'')$$

The first of these is homogeneous in $c_{II}^{(1)}$, $c_s^{(1)}$. The second is not, containing a term of zero power in these variables, *viz.*, $\sum(A_s^{II})^*c_s^{(0)}$. The classical theory analogon of this term is the

electric intensity impressed by the light wave on the oscillator.

In the evaluation of $\sum(A_s^{II})^* c_s^{(0)}$ we encounter the combination $(A_s^{II})^* A_s^{I}$. The polarization properties of the light quanta enter only through this combination, the remaining factor of each term in the summation being a pure function of the frequency ν_s. The summation is, therefore, performed by keeping the value of ν_s within a very narrow range, averaging each term over all directions of light quanta, multiplying the result by the number of possible s per unit frequency range of ν_s and integrating.

We shall suppose for the present that the dimensions of each atom are small in comparison with the wave-length of the light. The essential features of the phenomena can be seen in this simple case. We have

$$(A_s^{II})^* A_s^{I} = e^{i\mathbf{k}_s(\mathbf{R}_{II}-\mathbf{R}_I)}(\mathbf{f}_s(et)^{II})_{n_{II}'n_{II}}(\mathbf{f}_s(et)^{I})_{n_In_I'}/(2\pi h\nu_s V)$$

where $(et)=\sum e_i t_i$.

To obtain the average of this for fixed ν_s we turn the coordinate system so as to have $\mathbf{R}_{II}-\mathbf{R}_I$ along the z axis. Denoting the polar angles of $\mathbf{k}_s$ by Θ, Φ and the polar angles of $(et)_{n_In_I'}$, $(et)_{n_{II}n_{II}'}$ by (θ_I, φ_I), $(\theta_{II}, \varphi_{II})$, respectively, we have to average

$$\tfrac{1}{2}\{\sin\theta_I \sin(\varphi_I-\Phi)\sin\theta_{II}\sin(\varphi_{II}-\Phi)+[\sin\theta_I\cos\Theta\cos(\varphi_I-\Phi)-\cos\theta_I\sin\Theta]$$
$$\times[\sin\theta_{II}\cos\Theta\cos(\varphi_{II}-\Phi)-\cos\theta_{II}\sin\Theta]\}e^{ik_sR_{II\,I}\cos\Theta}$$

over Θ and Φ. Performing the averaging and discarding terms in $R_{II\,I}^{-2}$, $R_{II\,I}^{-3}$ we obtain

$$\overline{(A_s^{II})^* A_s^{I}} = \frac{\sin(k_sR_{II\,I})}{(k_sR_{II\,I})}\frac{\sin\theta_I\sin\theta_{II}\cos(\varphi_I-\varphi_{II})}{4\pi h\nu_s V}|(et)|_{n_{II}'n_{II}}|(et)|_{n_In_I'},$$

where the matrix elements in $|\ |$ are the ratios of the vector matrices to the unit vectors (sin θ cos φ, sin θ sin φ, cos θ). It is possible to introduce the polar angles θ, φ only if the absolute values of the vector matrices have a definite meaning, i.e., only if each vector matrix may be expressed as a unit real vector times a complex number. For degenerate states this is not possible, in general. The simple modifications required by degeneracy will be discussed later. The factor $\sin\theta_I\sin\theta_{II}\cos(\varphi_I-\varphi_{II})|(et)|_{n_{II}'n_{II}}|(et)|_{n_In_I'}$ may be interpreted as the scalar product of the projections of the matrix vectors $(et)_{n_{II}'n_{II}}$, $(et)_{n_In_I'}$ on to a plane perpendicular to $\mathbf{R}_{II}-\mathbf{R}_I$. Denoting these projections by a superscript (p) we have

$$\overline{(A_s^{II})^* A_s^{I}} = (\sin k_sR_{II\,I}/k_sR_{II\,I})(et)^{II(p)}_{n_{II}'n_{II}}(et)^{I(p)}_{n_In_I'}/(4\pi h\nu_s V).$$

We substitute this value into $\sum(A_s^{II})^* c_s^{(0)}$ and treat k_s, ν_s as though they were constants as long as they do not enter in trigonometric or exponential expressions. We let

$$\tau = R_{II\,I}/c$$

and make use of the integrals

$$\int_{-\infty}^{+\infty}\frac{e^{2\pi i\nu_s a}}{\nu_s-\nu_I+i\Gamma_I}d\nu_s=0, \qquad \int_{-\infty}^{+\infty}\frac{e^{-2\pi i\nu_s a}}{\nu_s-\nu_I+i\Gamma_I}d\nu_s=-2\pi i e^{-2\pi i(\nu_I-i\Gamma_I)a}$$

($a>0$ and real).

Then

$$\int_{-\infty}^{+\infty}\sin(2\pi\nu_s\tau)\frac{e^{-2\pi i(\nu_I-i\Gamma_I)t}-e^{-2\pi i\nu_s t}}{\nu_s-\nu_I+i\Gamma_I}d\nu_s = \begin{cases}=\pi e^{-2\pi i(\nu_I-i\Gamma_I)(t-\tau)} & (t>\tau)\\ =0 & (t<\tau).\end{cases}$$

Thus $\sum_s(A_s^{II})^* c_s^{(0)}=0$ if $t<\tau$. The atom II is not affected by the disturbance from atom I before the lapse of the time $R_{II\,I}/c$. *The interaction between the atoms takes place, therefore, with the velocity of light.*

After the lapse of the time τ

$$\sum_s (A_s^{II})^* c_s^{(0)} = K e^{-2\pi i(\nu_I - i\Gamma_I)(t-\tau)},$$

where

$$K = (e\mathfrak{x})^{II(\nu)}{}_{n_{II}' n_{II}} (e\mathfrak{x})^{I(\nu)}{}_{n_I n_I'} / (R_{II\ I} h c^2). \tag{130'''}$$

With (130''') substituted into (130'') it becomes natural to look for a solution of the type

$$c_{II}^{(1)} = K' e^{-2\pi i(\nu_I - i\Gamma_I)(t-\tau)},$$

which by the first equation (130'') makes it necessary to have

$$c_s^{(1)} = K' A_s^{II} (e^{-2\pi i(\nu_I - i\Gamma_I)(t-\tau)} - e^{-2\pi i \nu_s (t-\tau)}) / (\nu_s - \nu_I + i\Gamma_I).$$

Substituting both of these into the last Eq. (130'') and replacing as usual the summation by an integration we obtain

$$K' = K / [\nu_{II} - \nu_I + i(\Gamma_I - \Gamma_{II})].$$

These solutions satisfy the correct initial conditions for $c_s^{(1)}$ but not for $c_{II}^{(1)}$. Correct initial conditions for both are satisfied by superposing the emission type of solution (125) for atom II t in (125) being replaced everywhere by $t-\tau$. Thus,

$$\begin{aligned} c_{II}^{(1)} &= K \frac{e^{-2\pi i(\nu_I - i\Gamma_I)(t-\tau)} - e^{-2\pi i(\nu_{II} - i\Gamma_{II})(t-\tau)}}{\nu_{II} - \nu_I + i(\Gamma_I - \Gamma_{II})} \\ c_s^{(1)} &= \frac{K A_s^{II}}{\nu_{II} - \nu_I + i(\Gamma_I - \Gamma_{II})} \left[\frac{e^{-2\pi i(\nu_I - i\Gamma_I)(t-\tau)} - e^{-2\pi i \nu_s (t-\tau)}}{\nu_s - \nu_I + i\Gamma_I} - \frac{e^{-2\pi i(\nu_{II} - i\Gamma_{II})(t-\tau)} - e^{-2\pi i \nu_s (t-\tau)}}{\nu_s - \nu_{II} + i\Gamma_{II}} \right]. \end{aligned} \tag{131}$$

According to (131) there is a chance of finding atom II in the excited state n_{II}' after the time τ. This chance is

$$|c_{II}^{(1)}|^2 = |K|^2 \frac{e^{-4\pi\Gamma_I(t-\tau)} + e^{-4\pi\Gamma_{II}(t-\tau)} - 2e^{-2\pi(\Gamma_I + \Gamma_{II})(t-\tau)} \cos 2\pi(\nu_I - \nu_{II})(t-\tau)}{(\nu_{II} - \nu_I)^2 + (\Gamma_{II} - \Gamma_I)^2}. \tag{131'}$$

When $t-\tau \gg (4\pi\Gamma_I)^{-1}$, $(4\pi\Gamma_{II})^{-1}$, $|c_{II}^{(1)}|^2 \to 0$. The atom (II) has then dissipated its energy by radiating it away.

Let the atom II be put now in the vicinity of a source of light containing many radiating atoms excited at random intervals. We suppose for simplicity that the atoms in the source are excited in not too great numbers so that the transient (131') has time to die down between excitations. We wish to know the chance of finding II in the excited state. Let the number of atoms excited in the source per second be N. The required probability is then

$$N \int_{t=\tau}^{\infty} |c_{II}^{(1)}|^2 dt = \frac{(\Gamma_I + \Gamma_{II}) |K|^2 N}{4\pi\Gamma_I\Gamma_{II}[(\nu_I - \nu_{II})^2 + (\Gamma_I + \Gamma_{II})^2]}. \tag{131''}$$

The maximum probability of excitation occurs when $\nu_I = \nu_{II}$. The atoms in the source are then in exact resonance with the absorbing atom II. If $\nu_I - \nu_{II}$ is varied, as is the case experimentally on account of the Doppler effect, the resonance becomes less effective. The half-value breadth is then

$$\delta\nu = 2(\Gamma_I + \Gamma_{II}) = \delta\nu_I + \delta\nu_{II}. \tag{131'''}$$

It is equal to the sum of the half-value breadths of atoms I and II. The addition of the half-value breadths may also be explained by considering the spectral distribution in the emission line which according to (125) is such that

$$|A_s^I|^2 / [(\nu_s - \nu_I)^2 + \Gamma_I^2]$$

is the chance of finding a light quantum of type s

after the emission took place. On the other hand, the effect on $|c_{II}|^2$ due to light quanta is according to (128) proportional to $[(\nu_a-\nu_{II})^2+\Gamma_{II}^2]^{-1}$. It is easily found that

$$\int_{-\infty}^{+\infty}\frac{d\nu_a}{[(\nu_a-\nu_I)^2+\Gamma_I^2][(\nu_a-\nu_{II})^2+\Gamma_{II}^2]}=\frac{\pi(\Gamma_I+\Gamma_{II})}{\Gamma_I\Gamma_{II}[(\nu_{II}-\nu_I)^2+(\Gamma_I+\Gamma_{II})^2]}$$

which agrees with (131''). The additivity of the damping constants as described by (131''), (131''') is old and well known for the case of classical oscillators.[32] It has had technical applications in radiotelegraphy.

For large t we obtain from (131)

$$|c_a^{(1)}|^2=\frac{|K|^2|A_a^{II}|^2}{[(\nu_a-\nu_I)^2+\Gamma_I^2][(\nu_a-\nu_{II})^2+\Gamma_{II}^2]},$$

which shows that the distribution of scattered energy among frequencies follows the same law in the quantum as in the classical theory. The total chance of finding a scattered quantum is

$$\sum_a|c_a^{(1)}|^2=\frac{|K|^2(\Gamma_I+\Gamma_{II})/\Gamma_I}{(\nu_{II}-\nu_I)^2+(\Gamma_I+\Gamma_{II})^2}. \quad (132)$$

This is seen to be in agreement with (131''). For if there is the number

$$N\int_{t-\tau}^{+\infty}|c_{II}^{(1)}|^2dt$$

of excited atoms and if each of them has a chance $4\pi\Gamma_{II}$ of emitting one light quantum per second the rate at which light quanta will be scattered is given by

$$4\pi\Gamma_{II}N\int_{t-\tau}^{+\infty}|c_{II}^{(1)}|^2dt$$

which by (131'') is N times $\sum|c_a^{(1)}|^2$ as given by (132), i.e., N times the number of scattered quanta due to each atom in the source. Care must be taken, however, not to conclude that the frequency of scattered quanta is the emission frequency ν_{II}.

The same result can be obtained by *analogy with classical oscillators*. Each atom is then considered as a classical oscillator with an effective value of e^2/m given by

$$(e^2/m)_{eff.}=(2/h\nu)\{|\sum(e_i\dot{x}_i)_{nn'}|^2+|\sum(e_i\dot{y}_i)_{nn'}|^2+|\sum(e_i\dot{z}_i)_{nn'}|^2\},$$

which makes the emission probability $4\pi\Gamma=(2/3c^3)(2\pi\nu)^2(e^2/m)_{eff.}$. Monochromatic radiation of frequency ν, with an electric intensity is scattered by the virtual oscillator of atom II at the rate

$$\frac{\mathcal{E}_\nu^2\Gamma_{II}}{4\pi[(\nu_{II}-\nu)^2+\Gamma_{II}^2]}(e^2/m)^{II}_{eff.}\cos^2\theta_{II},$$

where θ_{II} is the angle between the axis of the virtual oscillator and $\mathcal{E}$. On the other hand, atom I emits a total amount of energy $h\nu_I$ during a time T. This energy is divided among frequencies so that at a distance R from I the energy density between ν and $\nu+d\nu$ is

$$\frac{3h\nu_I(\Gamma_I/\pi)\sin^2\theta_I}{8\pi R^2cT[(\nu-\nu_I)^2+\Gamma_I^2]}d\nu=\frac{\mathcal{E}_\nu^2}{4\pi},$$

where θ_I is the angle between the axis of the equivalent oscillator for atom I and the line $R_{II\,I}$.

In the direction of II the radiation due to I must be considered as polarized in accordance with the usual rule of classical electrodynamics. This determines $\cos\theta_{II}$. Substituting this into the rate of scattering of energy, multiplying by T and then integrating over ν we obtain the rate of scattering energy. Dividing this by $h\nu$ we get the probability of the scattering of a light quantum. The result is the same as (132) but multiplied by $(\nu_I^2/\nu\nu_{II})$ where ν is the mean frequency of the scattered light quanta. This correction is not significant because in the derivation of (132) we have supposed that $(\nu_{II}-\nu_I)/\nu_I\ll 1$.

§4a. Interference of radiation scattered by several atoms

We can discuss scattering from several atoms by the same method. Let atom I emit as before and let atoms II_1, II_2, $\cdots$ do the scattering.

[32] V. Bjerkness, Ann. d. Physik **44**, 74 (1891); **291**, 121 (1895); M. Wien, Ann. d. Physik **25**, 625 (1908); **29**, 679 (1909).

Similarly to (130), we have a system of equations

$$(d/2\pi idt+\nu_s)c_s=A_s^{I}c_I+\sum_j A_s^{II_j}c_{II_j},$$
$$(d/2\pi idt+\nu_I)c_I=\sum_s(A_s^{I})^*c_s, \qquad (133)$$
$$(d/2\pi idt+\nu_{II_j})c_{II_j}=\sum_s(A_s^{II_j})^*c_s,$$

the meaning of the symbols being exactly similar to that in (130). We consider the solution for the condition in which the rescattering by II_2, II_3, $\cdots$ of the radiation scattered by II_1 is not important, and, similarly, we shall neglect the rescattering by I of the radiation scattered by the II_j. The zeroth approximation will again be given by (130'). It is clear that we can now solve (133) simply by superposing the solutions (131) due to the atoms II. This solution is, of course, not exact, but the error introduced is small if the scattering atoms are far apart.

The resultant $c_s^{(1)}$ is the result of summing the expression $c_s^{(1)}$ given by (131) over all the scattering atoms. For $t\gg(2\pi\Gamma_I)^{-1}$ the probability of observing a light quantum of type s is simply

$$|\sum_j(c_s^{(1)})_{II_j}|^2=\frac{1}{(\nu_s-\nu_I)^2+\Gamma_I^2}\left|\sum_j\frac{K_{II_j}A_s^{II_j}}{\nu_s-\nu_{II_j}+i\Gamma_{II_j}}\right|^2. \qquad (133')$$

The important point to note is that the position of the scattering atoms enters this formula through $A_s^{II_j}$ in exactly the way which we would expect from the virtual oscillator analogy. For each $A_s^{II_j}$ involves as a factor $e^{-i\mathbf{k}_s\mathbf{R}_{II_jI}}$.

By making Γ_I very small we approach the condition of the scattering of monochromatic radiation. We then have appreciable probabilities for light quanta only when ν_s is very close to ν_I and the solutions become equivalent to those of formulas (128).

§5. Polarization of light quanta

So far for every direction of the wave normal we had two types of light quanta, each linearly polarized, the directions of the polarizations being at right angles to each other. Otherwise there has been no restriction on the choice of the vectors $\mathbf{f}_s$. It is clear that we may carry through the theory for any choice of the polarization vectors $\mathbf{f}$, provided they are perpendicular to $\mathbf{k}$ and provided $\mathbf{f}_{\mathbf{k},1}$ is perpendicular to $\mathbf{f}_{\mathbf{k},2}$. Any choice is as legitimate as any other. However, for the discussion of some problems a special choice may be preferable. We shall make this matter clear by an example.

We consider the emission of radiation by an atom due to a transition from an excited to the normal level. The solution is given by (125). The probability amplitude for a light quantum of type s is given by c_s which according to the calculation is proportional to A_s. If A_s is zero no light quantum of type s appears. Now

$$A_s\cong e^{-i\mathbf{kR}}(2\pi h\nu_s V)^{-\frac{1}{2}}((e\dot{\mathbf{r}})\mathbf{f}_s)_{nn'}. \qquad (122')$$

Let us consider the simplest case of a linear oscillator with the axis of vibration along the x axis of coordinates so that:

$$(\sum e_i\dot{y}_i)_{nn'}=(\sum e_i\dot{z}_i)_{nn'}=0,$$

and let us look for light quanta with the propagation vector along the y axis as indicated in Fig. 1. It is to our advantage to choose the two polarization vectors $\mathbf{f}_1$, $\mathbf{f}_2$ (we omit the suffix $\mathbf{k}$ in $\mathbf{f}_{\mathbf{k},l}$ because it is irrelevant in the present discussion) along the x and z axes, respectively. The probability of finding a light quantum with

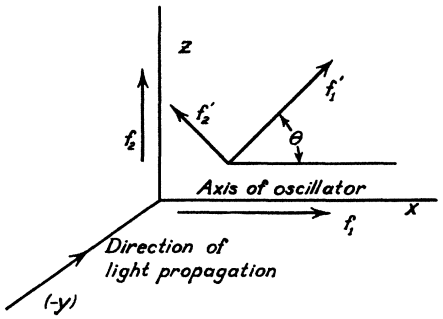

FIG. 1. Linearly polarized light quanta.

the polarization $\mathbf{f}_2$ is then zero since the corresponding A_s vanishes. The light emitted in the y direction may be said to be linearly polarized in the x direction.

We may, however, also use another set of mutually perpendicular polarization vectors $\mathbf{f}_1'$, $\mathbf{f}_2'$ and perform a calculation with them. For

each the A_s does not vanish. There is a chance of finding light quanta of polarizations $\mathbf{f}_1'$ and $\mathbf{f}_2'$. This corresponds exactly to our expectations from analogy to the classical theory. Light polarized linearly along the x axis excites vibrations in oscillators turned at an angle with respect to the x axis. In order to find out whether a light quantum of polarization $\mathbf{f}_1'$ may appear we have to test for its presence. We may do so by using an oscillator with its axis along $\mathbf{f}_1'$ and observing whether absorption will take place. The classical analogy leads us to expect that the absorption occurs for all orientations of the absorbing oscillator with the exception of $\mathbf{f}_2$. This expectation is correct, as shown by (130‴), (131). It is seen from these results that the only thing of importance in a test for the polarization of the radiation is the expression

$$((e\dot{\mathbf{t}}))^{(p)}{}_{n_{II}'n_{II}}((e\dot{\mathbf{t}}))^{(p)}{}_{n_{I}n_{I}'}.$$

If we wished to do so, we could avoid talking about the polarization of light quanta altogether, because the final experiment for their discussion eliminates the light quanta. However, there is no harm in keeping the classical terminology.

In order to complete the connection of the description by means of the $\mathbf{f}_1$, $\mathbf{f}_2$ polarizations with the $\mathbf{f}_1'$, $\mathbf{f}_2'$ representation, we must give the transformations of the radiation variables between these two systems. We must also show that these transformations lead in general to consistent results. We go back to formula (1) for the transverse part of the vector potential. For any given $\mathbf{k}$ we have two sets of radiation variables $(a_1, a_1^+)(a_2, a_2^+)$ corresponding to $\mathbf{f}_1$, $\mathbf{f}_2$. They contribute

$$(\mathbf{f}_1a_1+\mathbf{f}_2a_2)e^{-i\mathbf{kr}'}+i(\mathbf{f}_1a_1^++\mathbf{f}_2a_2^+)e^{i\mathbf{kr}'}$$

to $(V^{\frac{1}{2}}\nu/c)\mathbf{A}$. We have

$$\begin{aligned}\mathbf{f}_1'&=\mathbf{f}_1\cos\theta+\mathbf{f}_2\sin\theta,\\ \mathbf{f}_2'&=-\mathbf{f}_1\sin\theta+\mathbf{f}_2\cos\theta,\end{aligned}\tag{134}$$

so that if

$$\begin{aligned}a_1'&=a_1\cos\theta+a_2\sin\theta\\ a_2'&=-a_1\sin\theta+a_2\cos\theta\\ a_1'^+&=a_1^+\cos\theta+a_2^+\sin\theta\\ a_2'^+&=-a_1^+\sin\theta+a_2^+\cos\theta\end{aligned}\tag{135}$$

this contribution is

$$(\mathbf{f}_1'a_1'+\mathbf{f}_2'a_2')e^{-i\mathbf{kr}'}+i(\mathbf{f}_1'a_1'^++\mathbf{f}_2'a_2'^+)e^{i\mathbf{kr}'}.$$

The transformation (135) is *canonical*. It leaves $a_1^+a_1+a_2^+a_2$ invariant and, therefore, the Hamiltonian function is also invariant. The physical consequences are therefore the same whether we work with (a_1, a_2) or (a_1', a_2'), just as in Pauli's treatment of electron spin.

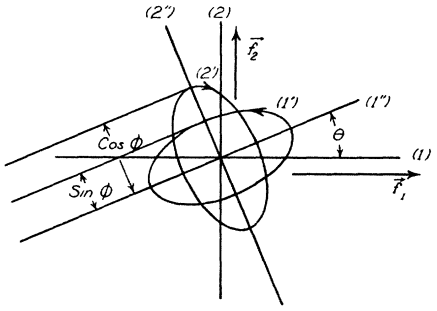

FIG. 2. Transformation to elliptical polarization.

We may similarly perform a transformation from two linear to two *elliptical polarizations*. Referring to Fig. 2 we look for two sets of variables such that

$$\begin{aligned}a_1'(\mathbf{f}_1''\cos\varphi-i\mathbf{f}_2''\sin\varphi)e^{-i\mathbf{kr}'}+ia_1'^+(\mathbf{f}_1''\cos\varphi+i\mathbf{f}_2''\sin\varphi)e^{i\mathbf{kr}'}+a_2'(\mathbf{f}_2''\cos\varphi-i\mathbf{f}_1''\sin\varphi)e^{-i\mathbf{kr}'}\\ +ia_2'^+(\mathbf{f}_2''\cos\varphi+i\mathbf{f}_1''\sin\varphi)e^{i\mathbf{kr}'}=(a_1\mathbf{f}_1+a_2\mathbf{f}_2)e^{-i\mathbf{kr}'}+i(a_1^+\mathbf{f}_1+a_2^+\mathbf{f}_2)e^{i\mathbf{kr}'}\\ =(a_1''\mathbf{f}_1''+a_2''\mathbf{f}_2'')e^{-i\mathbf{kr}'}+i(a_1''^+\mathbf{f}_1''+a_2''^+\mathbf{f}_2'')e^{i\mathbf{kr}'}.\end{aligned}$$

By (7′) the variables (a', a'^+) represent a wave in the vector potential of the form

$$\text{const. } R\{(\mathbf{f}_1''\cos\varphi-i\mathbf{f}_2''\sin\varphi)e^{2\pi i\nu t-i\mathbf{kr}'}\},$$

which is elliptically polarized. The vector potential at a fixed point describes by its end point the ellipse designated by (1′) in the direction indicated. Similarly for (2′). The major semiaxis is proportional to $\cos\varphi$, the minor to $\sin\varphi$.

The transformation formulas are:

$$a_1''=a_1'\cos\varphi-ia_2'\sin\varphi, \qquad a_1''^{+}=a_1'\cos\varphi+ia_2'^{+}\sin\varphi,$$
$$a_2''=-ia_1'\sin\varphi+a_2'\cos\varphi, \qquad a_2''^{+}=ia_1'\sin\varphi+a_2'^{+}\cos\varphi, \tag{136'}$$

$$a_1=a_1''\cos\theta-a_2''\sin\theta, \qquad a_1^{+}=a_1''^{+}\cos\theta-a_2''^{+}\sin\theta,$$
$$a_2=a_1''\sin\theta+a_2''\cos\theta, \qquad a_2^{+}=a_1''^{+}\sin\theta+a_2''^{+}\cos\theta. \tag{136''}$$

It is seen again that any succession of transformations (136) leaves invariant: the Hamiltonian, the exchange relations and $a_1^{+}a_1+a_2^{+}a_2$. We shall obtain, therefore, the same results whether we work in one set of $(a,\ a^{+})$ or in another.

In practical applications (Zeeman effect, resonance radiation) we encounter most frequently *circular polarizations*. The corresponding transformation is a special case of (136). We set $\theta=0$ and $\varphi=\pi/2$. The corresponding term in the vector potential is

$$c(2V)^{-1}\nu^{-1}\{[a_1'(\mathbf{f}_1-i\mathbf{f}_2)+a_2'(\mathbf{f}_2-i\mathbf{f}_1)]e^{-i\mathbf{kr}'}+i[a_1'^{+}(\mathbf{f}_1+i\mathbf{f}_2)+a_2'^{+}(\mathbf{f}_2+i\mathbf{f}_1)]e^{i\mathbf{kr}'}\}. \tag{137}$$

The transformation formulas are:

$$a_1=(a_1'-ia_2')/2^{\frac{1}{2}}, \qquad a_1^{+}=(a_1'^{+}+ia_2'^{+})/2^{\frac{1}{2}},$$
$$a_2=(-ia_1'+a_2')/2^{\frac{1}{2}}, \qquad a_2^{+}=(ia_1'^{+}+a_2'^{+})/2^{\frac{1}{2}}. \tag{137'}$$

We now remember that $(a)_{N,\,N-1}=(hN/(2\pi)^{\frac{1}{2}}$ and that A_s arose as the $(n\ n')$ matrix element of a $(\mathbf{fp})$. Working in the circular polarization directly we obtain then two A'^{s}: $A_{1'}$, $A_{2'}$, corresponding to the variables a_1', a_2', respectively.

Expression (137) shows that

$$A_{1'}=e^{-i\mathbf{kR}}(2\pi h\nu V)^{-\frac{1}{2}}((e\mathfrak{k})(\mathbf{f}_1-i\mathbf{f}_2)/2^{\frac{1}{2}})_{nn'},$$
$$A_{2'}=e^{-i\mathbf{kR}}(2\pi h\nu V)^{-\frac{1}{2}}((e\mathfrak{k})(\mathbf{f}_2-i\mathbf{f}_1)/2^{\frac{1}{2}})_{nn'}.$$

Thus, if $\mathbf{k}$ is along z, $\mathbf{f}_1$ along x, $\mathbf{f}_2$ along y, the emission of circularly polarized light quanta polarized in the direction of rotating x into y is determined by the matrix element of

$$((\dot{\mathbf{x}}+i\dot{\mathbf{y}})/2^{\frac{1}{2}})_{n'n}$$

(n' is the upper level) in the same way as the emission of linearly polarized quanta is determined by $(\dot{\mathbf{x}})_{n'n}$, $(\dot{\mathbf{y}})_{n'n}$, respectively.[33] Circularly polarized light quanta having the direction of rotation from y to x are similarly determined by

$$((\dot{\mathbf{y}}+i\dot{\mathbf{x}})/2^{\frac{1}{2}})_{n'n}.$$

The use of the expression (137) is most convenient for obtaining the result. It is useful, however, also to consider the matter from the point of view of the transformation (137'). We suppose the problem solved by working with the variables a_1, a_2 and their conjugates. We may then still obtain all the desired information about the variables a_1', a_2', i.e., about the probability of circularly polarized light quanta. For if we wish to know how likely it is that a circularly polarized light quantum of type 1' will be found, we simply have to find the quantum mean of $ia_1'a_1'^{+}$. By (137')

$$a_1'a_1'^{+}=\tfrac{1}{2}(a_1+ia_2)(a_1^{+}-ia_2^{+}).$$

Applying (61) and using $(a^{+})_{N,\,N+1}=-i(h/2\pi)^{\frac{1}{2}}(N$

[33] The same result can be inferred by forming a wave packet out of the wave function for the upper state and that of the lower, without making use of Dirac's theory of radiation. Let the wave packet be

$$c_{n'}u_{n'}e^{-2\pi i\nu(n')t}+c_nu_ne^{-2\pi i\nu(n)t}.$$

The variable parts of the quantum mean of x, y are then

$$2Re(c_{n'}^{*}c_nx_{n'n}e^{2\pi i\nu(n'n)t}),$$
$$2Re(c_{n'}^{*}c_ny_{n'n}e^{2\pi i\nu(n'n)t}).$$

These may be regarded as responsible for the emission of the linear light quanta polarized in the x and y directions.

If

$$y_{n'n}=\begin{cases}-ix_{n'n}\\ +ix_{n'n}\end{cases}$$

the y motion $\begin{cases}\text{lags } 90^\circ \text{ behind}\\ \text{leads } 90^\circ \text{ before}\end{cases}$ the x motion so that the resulting motion is a rotation in the sense from $\begin{cases}x \text{ to } y\\ y \text{ to } x\end{cases}$. For the y to x rotation $x_{n'n}+iy_{n'n}=0$, while for the x to y motion it is $2x_{n'n}$. Thus $x_{n'n}+iy_{n'n}$ is a measure of the purely rotational motion in the sense x to y.

$+1)^{\frac{1}{2}}(a)_{N,N-1}=(h/2\pi)^{\frac{1}{2}}N^{\frac{1}{2}}$ we have for the desired quantum mean

$$\frac{c_1^*+ic_2^*}{2^{\frac{1}{2}}}\frac{c_1-ic_2}{2^{\frac{1}{2}}}=\left|\frac{c_1-ic_2}{2^{\frac{1}{2}}}\right|^2$$

in agreement with the previous result. The probability of finding a light quantum of type (1′) thus depends on the *phase relation of the probability amplitudes* for linearly polarized light quanta. If $c_1+ic_2=0$ the *probability amplitudes* may be said to *interfere* so as *to give circular polarization.*

§6. Analogy of atoms to virtual oscillators

It has been seen above that in some special cases of emission and of scattering there is a close analogy between the behavior of atoms according to the theory of light quanta and the corresponding behavior of classical oscillators. It appears desirable to see a more general reason for this analogy than the agreement in special cases. It may, in fact, be shown that the solution of the quantum equations for a collection of atoms is connected with the solution of corresponding classical equations for oscillators in such a way that the quantum atoms may be replaced by the classical oscillators provided the frequencies dealt with are nearly in resonance with a possible emission or absorption frequency of the atoms in question. The atoms do not have to be exactly alike nor need they be far apart provided their electrostatic interaction is small. In this way it is seen that the classical dispersion theory remains valid for relatively dense matter and that the approximations of supposing the emitting and scattering atoms far apart made in VI §§4, 4a are not essential for the validity of the classical picture. If there is a strong electrostatic interaction between neighboring atoms, the analogy to the ordinary classical dispersion model is, of course, destroyed.

In the quantum treatment above it is natural to deal with the probability amplitudes c_s, $c_{\rm I}$, $c_{\rm II}$, etc. All the c's are essentially complex. In the classical theory, on the other hand, we deal with essentially real quantities, such as, the electric intensity at a point, the displacement of a classical dispersion electron from its position of equilibrium, etc. In order to obtain equivalent sets of equations in the two theories, we must introduce proper complex variables in the classical theory. We consider a number of classical oscillators. To an individual oscillator we refer by the letter j. The $j^{\rm th}$ oscillator is supposed to be located at the point R_j. Its direction of vibration is supposed to be restricted to a direction described by the unit vector $\mathbf{f}_j$; the displacement of the dispersion electron from its position of equilibrium along $\mathbf{f}_j$ is denoted by r_j. Neglecting retardation within r_j we have: [cf. I, §3 (18) (14)]

$$\begin{aligned}&[(d/2\pi i dt)+\nu_s]a_s^*=\textstyle\sum_j(2\pi)^{-1}(\nu_s V)^{-\frac{1}{2}}(\mathbf{J}_j\mathbf{f}_s)e^{-i\mathbf{k}_s\mathbf{R}_j},\\&[(d^2/dt^2)+(2\pi\nu_j)^2]r_j=2\pi(e_j/m_j)\textstyle\sum_s(\nu_s/V)^{\frac{1}{2}}(\mathbf{f}_j\mathbf{f}_s)[ia_s^*e^{i\mathbf{k}_s\mathbf{R}_j}-ia_se^{-i\mathbf{k}_s\mathbf{R}_j}].\end{aligned}\qquad\text{(C)}$$

These equations should be compared with the quantum equations for probability amplitudes:

$$\begin{aligned}[(d/2\pi i dt)+\nu_s]((h/2\pi)^{\frac{1}{2}}c_s)&=\textstyle\sum_j(2\pi)^{-1}(\nu_s V)^{-\frac{1}{2}}(\mathbf{J}^j{}_{nn'}\mathbf{f}_s)e^{-i\mathbf{k}_s\mathbf{R}_j}c_j,\\[(d/2\pi i dt)+\nu_j]c_j&=\textstyle\sum_s h^{-1}(\nu_s V)^{-\frac{1}{2}}(\mathbf{J}^j{}_{n'n}\mathbf{f}_s)e^{i\mathbf{k}_s\mathbf{R}_j}(h/2\pi)^{\frac{1}{2}}c_s,\end{aligned}\qquad\text{(Q)}$$

where $J^j{}_{nn'}$ is the matrix element of the current of the $j^{\rm th}$ atom and c_j is the probability amplitude for the $j^{\rm th}$ atom to be excited to the energy level $h\nu_j$. The frequencies ν_j are supposed to be nearly equal, and the possibility of excitation of the same atom to another energy level is neglected.

There is a similarity in the form of the first Eqs. (C) and (Q). The second Eq. (C) differs from the second Eq. (Q), inasmuch as it is of the second order. As has been just mentioned, however, it is not to be expected that the essentially real quantity r_j should be similar to the essentially complex c_j. We examine, therefore, Eqs. (C) and express them in terms of quantities having a close resemblance to the quantum quantities entering in (Q). We look for particular solutions of (C) having the nature of free vibrations. Let

$$a_s^* = \alpha_s e^{2\pi i\nu' t} + \beta_s e^{-2\pi i\nu' t}; \qquad \mathbf{J}_j = \mathbf{f}_j(A_j e^{2\pi i\nu' t} + B_j e^{-2\pi i\nu' t}),$$

$$\epsilon_j r_j = (A_j e^{2\pi i\nu' t} - B_j e^{-2\pi i\nu' t})/(2\pi i\nu').$$

Since J_j must be real

$$A_j^* = B_j.$$

Substitution into (C) gives:

$$(\nu_s + \nu')\alpha_s = \sum_j (2\pi)^{-1}(\nu_s V)^{-\frac{1}{2}}(\mathbf{f}_j\mathbf{f}_s)e^{-i\mathbf{k}_s\mathbf{R}_j}A_j; \qquad (\nu_s - \nu')\beta_s = \sum_j (2\pi)^{-1}(\nu_s V)^{-\frac{1}{2}}(\mathbf{f}_j\mathbf{f}_s)e^{-i\mathbf{k}_s\mathbf{R}_j}B_j.$$

It follows that $\alpha_s \ll \beta_s$ to the order $\Delta\nu/\nu$. Neglecting α_s in comparison with β_s Eqs. (C) give the following relations between β_s and B_j

$$\begin{aligned}(\nu_s - \nu')\beta_s &= \sum_j (2\pi)^{-1}(\nu_s V)^{-\frac{1}{2}}(\mathbf{f}_j\mathbf{f}_s)e^{-i\mathbf{k}_s\mathbf{R}_j}B_j,\\ (\nu_j - \nu')B_j &\cong \frac{\epsilon_j^2}{m_j}\frac{\nu'}{\nu_j + \nu'}\sum_s \left(\frac{\nu_s}{V}\right)^{\frac{1}{2}}(\mathbf{f}_j\mathbf{f}_s)e^{i\mathbf{k}_s\mathbf{R}_j}\beta_s.\end{aligned} \tag{C'}$$

We also look for particular solutions of (Q) in which the c's vary as $\exp(-2\pi i\nu' t)$. For such solutions

$$c_s = c_s^{(0)} \exp(-2\pi i\nu' t), \qquad c_j = c_j^{(0)} \exp(-2\pi i\nu' t)$$

where the $c^{(0)}$ are independent of the time

$$\begin{aligned}(\nu_s - \nu')(h/2\pi)^{\frac{1}{2}}c_s^{(0)} &= \sum_j (2\pi)^{-1}(\nu_s V)^{-\frac{1}{2}}(\mathbf{J}^j{}_{nn'}\mathbf{f}_s)e^{-i\mathbf{k}_s\mathbf{R}_j}c_j^{(0)},\\ (\nu_j - \nu')c_j^{(0)} &= \sum_s h^{-1}(\nu_s V)^{-\frac{1}{2}}(\mathbf{J}^j{}_{n'n}\mathbf{f}_s)e^{i\mathbf{k}_s\mathbf{R}_j}(h/2\pi)^{\frac{1}{2}}c_s^{(0)}.\end{aligned} \tag{Q'}$$

Eqs. (C') (Q') are identical in form with the exception of numerical constants which are easily adjusted and of the occurrence of $\nu'/(\nu_j + \nu')$ in the second Eq. (C'). We replace this factor by its approximate values $\frac{1}{2}$. This is permissible as long as we are interested only in a narrow range of frequencies ν' in the neighborhood of the ν_j. The general solution for c_s, c_j can be obtained by representing these quantities as sums of particular solutions of the type assumed for (Q'). The important range of values of ν' is of course in the neighborhood of the ν_j. For every ν' we let

$$\beta_s = (h/2\pi)^{\frac{1}{2}}c_s^{(0)}, \quad \mathbf{f}_j B_j = \mathbf{J}^j{}_{nn'}c_j^{(0)}.$$

The first Eq. (C') is then satisfied automatically. The second equation can also be satisfied, provided

$$\begin{aligned}B_j/c_j^{(0)} &= (\epsilon_j^2/2m_j)h\bar{\nu}_s(\mathbf{f}_j/\mathbf{J}^j{}_{n'n})\\ &= (\epsilon_j^2/2m_j)h\bar{\nu}_s(c_j^{(0)*}/B_j^*),\end{aligned}$$

where $\bar{\nu}_s$ is the average of ν_s over the range for which $c_s^{(0)}$ is appreciable. It follows that

$$|B_j|^2 = (\epsilon_j^2/2m_j)h\bar{\nu}_s|c_j^{(0)}|^2$$

and, therefore, that

$$\epsilon_j^2 \cong (2m_j/h\bar{\nu}_s)|J^j{}_{nn'}|^2.$$

To within the approximations used here we cannot distinguish between ν_j and $\bar{\nu}_s$. The result is, therefore, in agreement with the customary interpretation for the equivalent number of dispersion electrons. [Cf. Eq. 33 of preceding report.] Only in the present treatment it is more natural to think of it in terms of the effective value of the square of the charge for the equivalent virtual oscillator. We have supposed $\mathbf{f}_j$ to be real, and, therefore, we have considered only linear polarizations. The proof is easily generalized to elliptical polarizations by considerations similar to those of VI, §5.

It is, thus, seen that to within the approximations of the above derivation the atoms act as a collection of classical oscillators. The classical $\beta_s\nu'$ is connected with the energy of the radiation field in the same way as $(h/2\pi)^{\frac{1}{2}}c_s^{(0)}$ is connected with the expectation of the energy on the quantum theory. Similarly, if the energy of the equivalent classical oscillator is averaged over many periods of its own motion it becomes

$$\overline{W}_j = (2m_j/\epsilon_j^2)|B_j|^2 = h\nu|c_j^{(0)}|^2$$

again in agreement with the expectation on the quantum theory.

VII. Coherent and Incoherent Radiation; The Range of Validity of the Theory

§1. Scattering from atoms with degenerate states

Consider one scattering atom with a doubly degenerate normal state. The two states into which it splits under the action of an external field we call n and m. Let the atom be initially in the state n. After scattering, the atom may return to n, having emitted a light quantum s. It may also happen that a light quantum has been scattered but the atom is found after scattering in the state m. The probability amplitudes for the two states we write $\varphi(n, 1_s)$, $\varphi(m, 1_s)$. The two states are orthogonal, and, therefore, the total chance of a light quantum s is:

$$|\varphi(n, 1_s)|^2+|\varphi(m, 1_s)|^2.$$

Let there be two scattering atoms located sufficiently far from each other to make the rescattering by II of the radiation scattered by I negligible in comparison with the direct scattering. In the absence of II atom I, scattering by itself, would produce probability amplitudes $\varphi^{\mathrm{I}}(n_{\mathrm{I}}, 1_s)$, $\varphi^{\mathrm{I}}(m_{\mathrm{I}}, 1_s)$ for the states in which it is in the normal substates n_{I}, m_{I}, respectively, and in which there is a light quantum of type s. Similarly in the absence of I atom II, scattering by itself, would produce probability amplitudes $\varphi^{\mathrm{II}}(n_{\mathrm{II}}, 1_s)$, $\varphi^{\mathrm{II}}(m_{\mathrm{II}}, 1_s)$ for the normal substates of atom II. When both I and II are present the scattering occurs by either I or II going to an intermediate state and returning to one or the other of the normal states. The probability of both atoms changing their state on scattering is thus small and may be neglected. Supposing that the initial states of the atom are n_{I}, n_{II}, the probability amplitudes which appear after scattering are:

$$\varphi(n_{\mathrm{I}}, n_{\mathrm{II}}; 1_s)=\varphi^{\mathrm{I}}(n_{\mathrm{I}}, 1_s)+\varphi^{\mathrm{II}}(n_{\mathrm{II}}, 1_s),$$

$$\varphi(n_{\mathrm{I}}, m_{\mathrm{II}}; 1_s)=\varphi^{\mathrm{II}}(m_{\mathrm{II}}, 1_s),$$

$$\varphi(m_{\mathrm{I}}, n_{\mathrm{II}}; 1_s)=\varphi^{\mathrm{I}}(m_{\mathrm{I}}, 1_s).$$

Here $\varphi(m_{\mathrm{I}}, n_{\mathrm{II}}; 1_s)$ is the probability amplitude for atom I to be in m_{I}, for II to be in n_{II}, and for there being a light quantum s. The value of $\varphi(m_{\mathrm{I}}, m_{\mathrm{II}}; 1_s)$ is of a smaller order of magnitude as has been explained above. The total chance of finding a light quantum 1_s is, therefore,

$$|\varphi^{\mathrm{I}}(n_{\mathrm{I}}, 1_s)+\varphi^{\mathrm{II}}(n_{\mathrm{II}}, 1_s)|^2+|\varphi^{\mathrm{I}}(m_{\mathrm{I}}, 1_s)|^2+|\varphi^{\mathrm{II}}(m_{\mathrm{II}}, 1_s)|^2.$$

Similarly, for three scattering atoms the probability is

$$|\varphi^{\mathrm{I}}(n_{\mathrm{I}}, 1_s)+\varphi^{\mathrm{II}}(n_{\mathrm{II}}, 1_s)+\varphi^{\mathrm{III}}(n_{\mathrm{III}}, 1_s)|^2+|\varphi^{\mathrm{I}}(m_{\mathrm{I}}, 1_s)|^2+|\varphi^{\mathrm{II}}(m_{\mathrm{II}}, 1_s)|^2+|\varphi^{\mathrm{III}}(m_{\mathrm{II}}, 1_s)|^2.$$

The probability is seen to consist of two parts. One of these of the form (133′) is the square of a sum and implies interference of probability amplitudes. Another is a sum of independent contributions. The first part may be said to be due to coherent and the second to incoherent scattering. The scattering due to the return of atoms to the initial state is coherent. The scattering due to the transition into another atomic state is incoherent. It should be emphasized that it does not matter whether the energy of the new atomic state is different from the initial energy. It is only important that the two states be orthogonal. The fact that the transitions to n and m may give rise to different polarizations is also irrelevant, since coherent waves with mutually perpendicular polarizations may interfere when one of them is passed through a suitable optical instrument (quarter-wave plate, for example).

The above distinction between coherent and incoherent radiation is identical with the original terminology of Kramers and Heisenberg [Eqs. (48), (53)]. Before Dirac's theory of light quanta the most direct reason for supposing (48) to be the coherent part of the scattered radiation has been that it has the correct frequency. As has just been explained there exists also incoherent radiation of unchanged frequency. Thus, the summation in (48) should be thought of as including only one substate of J' if J' is degenerate.

If the degeneracy of an atom is due to the spherical symmetry of the field, it may be removed by applying a magnetic field in any direction. The atom is then space quantized. The

scattering due to the atom leaving a magnetic sublevel m of the lower state, going to a sublevel μ of the upper state and then returning to the same m, has coherence properties. This scattering contributes to the refractive index. The scattering due to a transition from μ to a different sublevel m' of the lower state is incoherent and has no effect on the refractive index. The average contribution to the refractive index per atom must be independent of the direction of the magnetic field if the field is weak [spectroscopic stability].

If the magnetic field is parallel to the electric intensity of the incident light, the absorptions $m \to \mu$ are due to the π components of the Zeeman effect and, therefore, the coherent scattering $\mu \to m$ is also due to the π components. If the magnetic field is now turned so as to be parallel to the direction of propagation of the light wave, the coherent radiation is due to σ components. For a given initial m there may be either just one coherent σ component emitted or else there may be two. In the first case the σ component must interfere with another oppositely polarized σ component due to another atom in another initial state so as to give a resultant linear polarization in the direction of the electric intensity of the incident light wave. If there are two σ components ending on the same m then it may also happen that they will combine into a linearly polarized wave for a single atom. [See example of mercury resonance radiation below.] Such a combination can happen only for the coherent part of the scattering.

If the arrangement of the scattering atoms in space is so irregular that no interference of radiations from different atoms can take place, one can still observe the scattered radiation, as is the case in experiments on resonance radiation. Again the intensity and the degree of polarization of the scattered radiation cannot depend on the direction of a very small magnetic field. This comes about through the interference of σ components due to a return of the atom to the initial magnetic sublevel m. The mathematical theory of this phenomenon is given in VII, §4 below. There are, thus, two types of possible interference between radiation in the Zeeman pattern: the type responsible for the refractive index and the whole coherently scattered radiation; and the interference which may take place in resonance radiation between two Zeeman components due to absorptions from m to μ and μ' and the return from μ and μ' to m. The first type is illustrated by Na below and the second is illustrated by Hg.

We consider the resonance radiation of mercury (neglecting the nuclear spin). The normal state has an inner quantum number $j=0$ and for the excited state $j=1$. The scattering gas is supposed to be in a magnetic field directed along the electric intensity of the linearly polarized incident beam. The magnetic quantum number for the upper state can be $m=1, 0, -1$; for the lower state $m=0$. The transitions $1 \to 0$, $-1 \to 0$ are circularly polarized (σ Zeeman components).

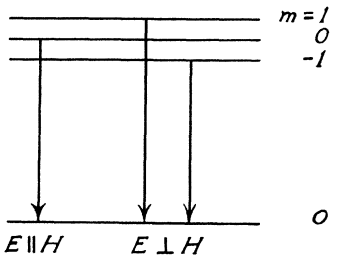

FIG. 3. Zeeman diagram for λ2537 of mercury.

The transition $0 \to 0$ is linearly polarized (π component). The scattered radiation is due to the excitation to $m=0$ and is, therefore, linearly polarized. If, on the other hand, the magnetic field is perpendicular to the incident electric intensity, the scattered radiation consists of two frequencies (the two σ components) each of which is circularly polarized. Since the atom returns in each case to the initial level, the circular components may interfere. As long, however, as the frequencies are appreciably different, the interference is ineffective even though it exists because the resultant linear vibration has a rapidly precessing axis. When the frequencies differ so little that the axis cannot precess through an appreciable angle during the time $(2\pi\Gamma)^{-1}$ the radiation is again linearly polarized this time as the result of the interference of two σ components. For small H there is thus no difference between different directions of H. This is, of course, as it should be by the

principle of spectroscopic stability.[34] We see that in this instance this principle is satisfied by the ability for interference of quanta having opposite directions of circular polarization. The experiments of Wood, Ellett and Hanle on the depolarization in weak magnetic fields may be regarded as demonstrating the interference between circularly polarized quanta of slightly different frequencies. This results in a linearly polarized quantum with a precessing polarization axis. It should be remembered, however, that from the point of view of our theory it is the probability amplitudes of light quanta having precisely the same frequency that interfere. We deal, therefore, with the overlapping of the spectral ranges of the scattered radiation due to the two σ components. The quantitative features of these phenomena will be dealt with in the section on the polarization of resonance radiation.

We consider next the scattering by sodium, and for simplicity we take into account the D_1 line only. The possible transitions and the corresponding polarization characters are shown in the Fig. 4. Initially, we may suppose half the atoms

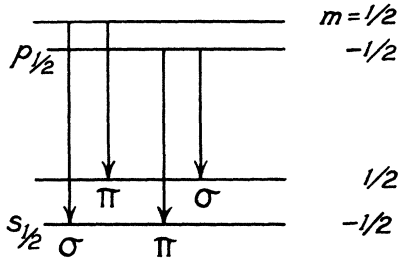

FIG. 4. Zeeman diagram of D_1 line of sodium.

to be in the s state with $m=\frac{1}{2}$ and the other half in the s state with $m=-\frac{1}{2}$. For $E||H$ we obtain coherent scattering of the π's and incoherent scattering of the σ's. For $E\perp H$ it is the σ's that are scattered coherently while the π's are scattered incoherently. If H becomes very small but $\perp E$, the polarization of the coherent part of the scattered radiation is again that of the π's, as though H were $||E$, on account of the interference between the coherent σ's.

[34] W. Heisenberg, Zeits. f. Physik 31, 617 (1925); W. Pauli, *Quantentheorie Handbuch der Physik*, Geiger-Scheel XIII, 86–108.

Only the coherent part of the scattering is of importance for the discussion of the refractive index of a gas. For any system of atoms satisfying the conservation of angular momentum we can apply a small magnetic field in the direction of the incident electric vector. We then have to consider only the π components. The plane of polarization is unchanged. There is no double refraction and no rotation of the plane of polarization. If a magnetic field of appreciable magnitude is applied in the direction of the propagation of light, the σ components are responsible for the coherent scattering and a rotation of the plane of polarization results. This is the essence of the Faraday effect of the magnetic rotation of the plane of polarization. The difference between the Faraday effect and the depolarization of resonance radiation consists, thus, in the fact that the Faraday effect is concerned only with coherent scattering while experiments on resonance radiation are affected also by the incoherent radiation.

Interference between Zeeman components as connected with the polarization of resonance radiation has been discussed by Heisenberg and Pauli[34] before the invention of quantum mechanics.

§2. Emission from atoms with degenerate states

Again, consider an atom definitely located in a certain excited state at the time $t=0$. Let the normal state be doubly degenerate as before. For

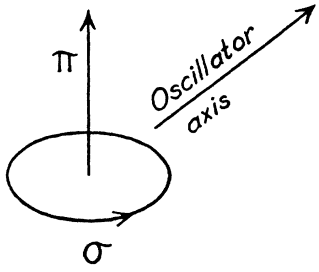

FIG. 5. Interference of linearly and circularly polarized quanta.

example, at the time $t=0$ we may have the atom in the $m=\frac{1}{2}$ magnetic sublevel of the p_1 state of sodium in the last illustration of scattering. The atom may pass into either of the two normal

levels emitting either a π or a σ component. A group of nondegenerate scattering atoms may be used to test the resultant radiation for coherence. To illustrate our point we shall use linear oscillators for the scattering atoms, the line joining the scattering atoms to the source will be perpendicular to the magnetic field, and the axis of the oscillators will be at 45° with respect to the magnetic field. Both the π and the σ components will affect the oscillators. If we were to think of the process in terms of classical electrodynamics, we should expect the amplitude of the oscillator vibrations to be a function of the phase of the π and σ vibrations. Such a phase might be difficult to ascertain, and it might be random for a collection of emitting atoms; nevertheless, it exists for any classical radiating system. The quantum theory of radiation, on the contrary, does not leave any possibility for the interference of the π and σ vibrations in their effect on the exploring oscillators. The π component may be said to be scattered coherently by the oscillators and the scattering from them will be given by (133′); similarly, for the σ components. The resultant probability of finding a scattered light quantum is the sum of the two probabilities.

Although quantum mechanics allows us to discuss the scattering from an atom in a definite state, the only meaning of the discussion is the statistical interpretation which can be made either by using a large number of identical atoms in the same state or by using the same atom and repeatedly bringing it back to the original state. The sum total of the above discussion for emission is thus that it is *impossible to prepare* an excited atomic state in such a way as to have *definite phase relationships* between the electromagnetic waves sent out when the atom jumps to two different orthogonal lower states. Formally, this is simply a restatement of the fact that energy and time are canonically conjugate variables.

We now support the above discussion by calculation. The two normal states of the *emitting atom* we call n_{I}, m_{I}, its excited state n_{I}'; the normal and the excited states of the *scattering atom* (II) n_{II}, n_{II}', respectively. We let

$$e^{2\pi i(\nu(n_{\mathrm{I}})+\nu(n_{\mathrm{II}}))t}=\theta;\quad \theta\varphi(n_{\mathrm{I}}', n_{\mathrm{II}}; 0, 0, 0, \cdots)=c_{\mathrm{I}};\quad \theta\varphi(m_{\mathrm{I}}, n_{\mathrm{II}}'; 0, 0, \cdots)=c_{\mathrm{II}m},$$

$$\theta\varphi(n_{\mathrm{I}}, n_{\mathrm{II}}'; 0, 0, \cdots)=c_{\mathrm{II}n};\quad \theta\varphi(n_{\mathrm{I}}, n_{\mathrm{II}}; 0, \cdots, 1_s, 0, \cdots)=c_{sn};\quad \theta\varphi(m_{\mathrm{I}}, n_{\mathrm{II}}; 0, \cdots, 1_s, 0, \cdots)=c_{sm}.$$

Working near resonance the equations are:

$$(d/2\pi i dt+\nu(n_{\mathrm{I}}'n_{\mathrm{I}}))c_{\mathrm{I}}=\textstyle\sum_s(A_{sm}^{\mathrm{I}*}c_{sm}+A_{sn}^{\mathrm{I}*}c_{sn}),$$

$$(d/2\pi i dt+\nu(n_{\mathrm{II}}'n_{\mathrm{II}}))c_{\mathrm{II}n}=\textstyle\sum_s A_s^{\mathrm{II}*}c_{sn},$$

$$(d/2\pi i dt+\nu(n_{\mathrm{II}}'n_{\mathrm{II}})+\nu(n_{\mathrm{I}}n_{\mathrm{I}}))c_{\mathrm{II}m}=\textstyle\sum_s A_s^{\mathrm{II}*}c_{sm},$$

$$(d/2\pi i dt+\nu_s)c_{sn}=A_{sn}^{\mathrm{I}}c_{\mathrm{I}}+A_s^{\mathrm{II}}c_{\mathrm{II}n},$$

$$(d/2\pi i dt+\nu_s+\nu(m_{\mathrm{I}}n_{\mathrm{I}}))c_{sm}=A_{sm}^{\mathrm{I}}c_{\mathrm{I}}+A_s^{\mathrm{II}}c_{\mathrm{II}m},$$

$$A_s^{\mathrm{II}}\cong e^{-i\mathbf{k}\mathbf{R}_{\mathrm{II}}}(2\pi h\nu_s V)^{-\frac{1}{2}}(\textstyle\sum_i e_i^{\mathrm{II}}\mathfrak{k}_i^{\mathrm{II}}\mathfrak{f}_s)_{n_{\mathrm{II}}n_{\mathrm{II}}'};\quad A_{sn}^{\mathrm{I}}\cong e^{-i\mathbf{k}\mathbf{R}_{\mathrm{I}}}(2\pi h\nu_s V)^{-\frac{1}{2}}(\textstyle\sum_i e_i^{\mathrm{I}}\mathfrak{k}_i^{\mathrm{I}}\mathfrak{f}_s)_{n_{\mathrm{I}}n_{\mathrm{I}}'},$$

$$A_{sm}^{\mathrm{I}}\cong e^{-i\mathbf{k}\mathbf{R}_{\mathrm{I}}}(2\pi h\nu_s V)^{-\frac{1}{2}}(\textstyle\sum_i e_i^{\mathrm{I}}\mathfrak{k}_i^{\mathrm{I}}\mathfrak{f}_s)_{m_{\mathrm{I}}n_{\mathrm{I}}'}.$$

The solution proceeds analogously to that of (130). If atom (II) were absent, we should have

$$(d/2\pi i dt+\nu(n_{\mathrm{I}}'n_{\mathrm{I}}))c_{\mathrm{I}}^{(0)}=\textstyle\sum_s(A_{sm}^{\mathrm{I}*}c_{sm}^{(0)}+A_{sn}^{\mathrm{I}*}c_{sn}^{(0)}),$$

$$(d/2\pi i dt+\nu_s)c_{sn}^{(0)}=A_{sn}^{\mathrm{I}}c_{\mathrm{I}}^{(0)},$$

$$(d/2\pi i dt+\nu_s+\nu(m_{\mathrm{I}}n_{\mathrm{I}}))c_{sm}^{(0)}=A_{sm}^{\mathrm{I}}c_{\mathrm{I}}^{(0)}.$$

A sufficiently approximate solution of these equations is

$$c_{\mathrm{I}}^{(0)}=\exp\{-2\pi i(\nu(n_{\mathrm{II}}'n_{\mathrm{I}})-i\Gamma_{\mathrm{I}})t\},$$

$$c_{sn}^{(0)}=(A_{sn}^{\mathrm{I}}/[\nu_s-\nu(n_{\mathrm{I}}'n_{\mathrm{I}})+i\Gamma_{\mathrm{I}}])[\exp\{-2\pi i(\nu(n_{\mathrm{I}}'n_{\mathrm{I}})-i\Gamma_{\mathrm{I}})t\}-\exp\{-2\pi i\nu_s t\}],$$

$$c_{sm}^{(0)}=A_{sm}^{\mathrm{I}}e^{-2\pi i\nu(n_{\mathrm{I}}n_{\mathrm{I}})t}/[\nu_s-\nu(n_{\mathrm{I}}'m_{\mathrm{I}})+i\Gamma_{\mathrm{I}}])[\exp\{-2\pi i(\nu(n_{\mathrm{I}}'m_{\mathrm{I}})-i\Gamma_{\mathrm{I}})t\}-\exp\{-2\pi i\nu_s t\}],$$

$$\Gamma_{\mathrm{I}}=(\pi|A_{sn}^{\mathrm{I}}|^2/(\Delta\nu))_{\nu=\nu(n_{\mathrm{I}}'n_{\mathrm{I}})}+(\pi|A_{sm}^{\mathrm{I}}|^2/(\Delta\nu))_{\nu=\nu(n_{\mathrm{I}}'m_{\mathrm{I}})}.$$

The damping constant for emission is seen to be the sum of the damping constants for the emission of $\nu(n_1'n_1)$, $\nu(n_1'm_1)$ independently as has been shown by Weisskopf and Wigner.[30]

Neglecting the change in c_1^0, i.e., the rescattering by (I), we have on letting

$$c_{sn}=c_{sn}^{(0)}+c_{sn}^{(1)};\qquad c_{sm}=c_{sm}^{(0)}+c_{sm}^{(1)};\qquad c_{\mathrm{II}n}=c_{\mathrm{II}n}^{(1)};\qquad c_{\mathrm{II}m}=c_{\mathrm{II}m}^{(1)}$$

the following pair of mutually independent sets of equations

$$(d/2\pi i dt+\nu_s)c_{sn}^{(1)}=A_s^{\mathrm{II}}c_{\mathrm{II}n}^{(1)},$$

$$(d/2\pi i dt+\nu(n_{\mathrm{II}}'n_{\mathrm{II}})c_{\mathrm{II}n}^{(1)}=\sum A_s^{\mathrm{II}*}(c_{sn}^{(0)}+c_{sn}^{(1)}),$$

$$(d/2\pi i dt+\nu_s)(c_{sm}^{(1)}e^{2\pi i\nu(m_1n_1)t})=A_s^{\mathrm{II}}(c_{\mathrm{II}m}^{(1)}e^{2\pi i\nu(m_1n_1)t}),$$

$$(d/2\pi i dt+\nu(n_{\mathrm{II}}'n_{\mathrm{II}}))(c_{\mathrm{II}m}^{(1)}e^{2\pi i\nu(m_1n_1)t})=\sum_s A_s^{\mathrm{II}*}(c_{sm}^{(0)}+c_{sm}^{(1)})e^{2\pi i\nu(m_1n_1)t}.$$

The solution of the first set is given by (131) directly. The same formulas apply to $c_{sm}^{(1)}e^{2\pi i\nu(m_1n_1)t}$, $c_{\mathrm{II}m}^{(1)}e^{2\pi i\nu(m_1n_1)t}$ for the second set. The probability of a light quantum of type s is given by $|c_{sn}|^2+|c_{sm}|^2$ exactly as though there two kinds of incoherent scattered radiation described, respectively, by the probability amplitudes c_{sn}, c_{sm}. We shall make use of this fact in the next section.

In the classical theory the scattering by an atom depends on the *square of the electric field* of the incident light wave. We expect that in the quantum theory it should depend on the *quantum mean* or "expectation" of this quantity. This is the case as may be seen from the following consideration.

Using expression (4) for $\mathcal{E}$ we form the component of $\mathcal{E}$ in a given direction defined by the unit vector $\mathbf{f}$

$$(\mathbf{f}\mathcal{E})=-\sum_s 2\pi i(\mathbf{f}\mathbf{f}_s)(\nu_s/V)^{\frac{1}{2}}[a_s e^{-i\mathbf{k}_s\mathbf{r}}-ia_s^{+}e^{i\mathbf{k}_s\mathbf{r}}].$$

We form the square of this quantity. In the result we interchange the position of a_s and a_s^{+} so as to have them always in the order $a_sa_s^{+}$. The resultant quantity is directly related to the probability of scattering. To evaluate it we use the values of the matrix elements.

$$\begin{cases}(a_{s'}a_{s''})_{N_{s'},\,N_{s''};\,N_{s'}-1,\,N_{s''}-1}=(h/2\pi)(N_{s'}N_{s''})^{\frac{1}{2}},\\ (a_{s'}^{+}a_{s''}^{+})_{N_{s'},\,N_{s''};\,N_{s'}+1,\,N_{s''}+1}=(h/2\pi)(N_{s'}+1)^{\frac{1}{2}}(N_{s''}+1)^{\frac{1}{2}},\\ -i(a_{s'}^{+}a_{s''})_{N_{s'},\,N_{s''};\,N_{s'}+1,\,N_{s''}-1}=(h/2\pi)(N_{s'}+1)^{\frac{1}{2}}N_{s''}^{\frac{1}{2}},\end{cases}\qquad (s'\neq s'')$$

$$(a_sa_s)_{N_s,\,N_s-2}=(h/2\pi)N_s^{\frac{1}{2}}(N_s-1)^{\frac{1}{2}};\qquad (a_s^{+}a_s^{+})_{N_s,\,N_s+2}=(h/2\pi)(N_s+1)^{\frac{1}{2}}(N_s+2)^{\frac{1}{2}},$$

$$-i(a_sa_s^{+})_{N_s,\,N_s}=(h/2\pi)N_s,\qquad -i(a_s^{+}a_s)=(h/2\pi)(N_s+1),$$

and the general formula (61) for the quantum mean. Confining ourselves to cases in which the probability amplitudes exist only for those states in which there is not more than one light quantum present we have after an easy substitution the desired quantum mean at the point $\mathbf{r}$

$$\overline{(\mathbf{f}\mathcal{E})^2}=\sum_n(4\pi h/V)\,|\,(\mathbf{f}\mathbf{f}_s)\nu_s^{\frac{1}{2}}e^{i\mathbf{k}_s\mathbf{r}}c_{ns}\,|^2,\qquad (138)$$

where the index n refers to the state of the matter.

This should be compared with the last equation (130) and the second equation (130''). The absorption by the atom (II) depends on $\sum A_s^{\mathrm{II}*}c_s$. To within the approximations made throughout the present treatment we cannot distinguish between the factors $\nu_s^{-\frac{1}{2}}$ in A_s^{II} and $\nu_s^{\frac{1}{2}}$ in (138). Otherwise these two quantities differ only by a constant factor if $\mathbf{f}$ is taken along $(\sum e_i\mathbf{r}_i)_{n_{\mathrm{II}}n_{\mathrm{II}}'}$.

Formula (138) shows again the lack of coherence of probability amplitudes referring to different states of matter. It shows also that

$$\sum_s(4\pi h\nu_s/V)^{\frac{1}{2}}e^{i\mathbf{k}_s\mathbf{r}}c_{ns}\mathbf{f}_s\qquad (138')$$

is analogous to the electric intensity of classical theory. The square of the absolute value of the vector (138') determines part of the scattering by an atom put at $\mathbf{r}$. The total intensity of scattering

is the sum of the intensities due to (138′) for different n.

(§3) Unidirectional emission of quanta

In all the preceding discussion we have supposed the nuclei of the atoms to be fixed in position. We shall now take into account the motion of the nucleus. It is well known that the emission of light leads to recoil actions on the atom. According to Einstein, whenever a light quantum is emitted the atom should suffer a change of momentum numerically equal to $h\nu/c$ and opposite to the direction of emission exactly as though the light quanta were unidirectional darts each of momentum $h\nu/c$. It is known, on the other hand, that the light emitted by an atom resembles a spherical wave in many of its properties. Thus, for example, the radiation scattered by a collection of atoms interferes as though each atom were the source of spherical wavelets. Similarly, in any discussion of interference phenomena, such as Newton's rings, it is very necessary to be able to consider the radiation emitted in different directions by the same point of the source as coherent. Experience shows that this view leads to correct results. The argument of Einstein about unidirectional emission and the classical requirements of interference are at first sight in conflict with each other.

The reconciliation of the two demands can be reached by inquiring more closely into the meaning of the conditions dealt with in both considerations. It is at once apparent that in Einstein's discussion we have to deal with conditions in which the momentum of the light quantum and the momentum of the atom are definitely known.[35] According to the principle of uncertainty this automatically excludes the knowledge of the positions of the atoms and thus makes a discussion of interference impossible. On the other hand, in discussing interference we suppose the position of the atoms to be known so that the momentum cannot be ascertained. Under these circumstances the atom may be said to emit spherical waves. We can say if we wish that the atom must be held fixed in order to emit a spherical wave.

[35] C. Eckart, Phys. Rev. **34**, 167 (1929); W. Heisenberg, *Principles of the Quantum Theory*, University of Chicago Press, 1930; G. Breit, J. Opt. Soc. Am. **14**, 324 (1927).

It is uncomfortable to have this differentiation without seeing more clearly just what determines a sufficient degree of fixedness for the atom in order that radiation emitted in different directions could be considered as coherent. The purpose of this section is to clarify this point. We shall concern ourselves only with the essential qualitative features of the problem and, therefore, shall suppose that the ratio of the nuclear to the electronic mass is large and that the wavelength of the emitted light is long in comparison with atomic dimensions. For the sake of simplicity we discuss the hydrogen atom although all the essential deductions apply to many electron atoms and molecules as well.

We denote the coordinates of the electron and nucleus by indices e and n, respectively. The relative coordinates and the coordinates of the center of mass are then given respectively by

$$\mathbf{r}=\mathbf{r}_e-\mathbf{r}_n, \quad \mathbf{R}=(m\mathbf{r}_e+M\mathbf{r}_n)/(m+M)$$

and

$$\mathbf{r}_e=\mathbf{R}+[M/(m+M)]\mathbf{r},$$

$$\mathbf{r}_n=\mathbf{R}-[m/(m+M)]\mathbf{r},$$

and where m is the mass of e and M is the mass of n.

It is well known that the Schroedinger equation is separable in the variables $\mathbf{r}$, $\mathbf{R}$. The eigenfunctions which contain $\mathbf{r}$ are exactly those for a particle of mass $\mu=mM/(m+M)$ and a fixed nucleus. The eigenfunctions containing $\mathbf{R}$ are those for a free particle of mass $M+m$. The stationary states of the whole atom, therefore, can be described completely by assigning quantum numbers to the relative coordinate states which we call n and by assigning momenta to the center of mass. The latter we denote by the letter $\mathbf{P}$ which stands collectively for the three components of momentum P_x, P_y, P_z.

Initially (at the time $t=0$) we suppose the atom to be in the excited state n'. Its momentum we shall not specify exactly. Instead we suppose that there is a probability amplitude, $C(\mathbf{P})$ which by the square of its absolute value determines the probability of there being a momentum $\mathbf{P}$. Since the momentum has a continuous range of values, $C(\mathbf{P})$ must refer to an element of volume in the momentum space (P_x, P_y, P_z). Thus, the probability of there being

a momentum in the volume element $dP_x dP_y dP_z$ shall be $|C(\mathbf{P})|^2 dP_x dP_y dP_z$. In the course of time light quanta are emitted. Correspondingly, there appear probability amplitudes $C_s(\mathbf{P}')$. These refer to states in which there is a light quantum of type s and the electronic state is n. The probability of finding such a light quantum and simultaneously of having the atom in the normal state while the momentum is in the volume element $dP_x dP_y dP_z$ shall be $|C_s(\mathbf{P})|^2 dP_x dP_y dP_z$.

In order to find the relations between the probability amplitudes we go back to the general Eq. (110). In this the state of the matter was described by the index n, which denoted collectively all the quantum numbers. In the present case we replace this index by two letters n, $\mathbf{P}$. We have, thus,

$$(2\pi h)^{\frac{1}{2}}\alpha^s_{n\mathbf{P};\,n',\,\mathbf{P}'} = (e/2)\mathbf{f}_s[(\mathbf{p}_e/m)e^{-i\mathbf{k}_s\mathbf{r}_e} - (\mathbf{p}_{nuc}/M)e^{-i\mathbf{k}_s\mathbf{r}_{nuc}}]_{n,\,\mathbf{P};\,n'\mathbf{P}'}.$$

The operators representing the velocities of the electron and the nucleus are connected by relations quite similar to those for the coordinates. For a heavy nucleus and for an atom small in comparison with the wave-length of the light we may approximate the above value of $\alpha^s_{n,\,\mathbf{P};\,n',\,\mathbf{P}'}$ by

$$(2\pi h)^{\frac{1}{2}}\alpha^s_{n,\,\mathbf{P};\,n',\,\mathbf{P}'} = (e/2m)(\mathbf{f}_s\mathbf{p})_{n,\,n'}(e^{-i\mathbf{k}_s\mathbf{R}})_{\mathbf{P};\,\mathbf{P}'}, \tag{139}$$

where $\mathbf{p}$ is the relative momentum, conjugate to the relative coordinates $\mathbf{r}$. We see that the whole matrix element splits in this case into two factors. One of these $(\mathbf{f}_s\mathbf{p})_{n,\,n'}$ is exactly as though the nucleus were fixed. The second refers to the motion of the center of mass. The eigenfunction

$$h^{-\frac{1}{2}}e^{(2\pi i/h)\mathbf{P}'\mathbf{R}}$$

represents a state of constant total momentum P'. Since

$$e^{-i\mathbf{k}_s\mathbf{R}+(2\pi i/h)\mathbf{P}'\mathbf{R}} = \int \delta\left(\mathbf{P},\,\mathbf{P}'-\frac{h\mathbf{k}_s}{2\pi}\right)e^{(2\pi i/h)\mathbf{P}\mathbf{R}}dP_x dP_y dP_z$$

we have by the general expansion theorem [cf. (70)]

$$(e^{-i\mathbf{k}_s\mathbf{R}})_{\mathbf{P},\,\mathbf{P}'} = \delta(\mathbf{P},\,\mathbf{P}'-h\mathbf{k}_s/2\pi) \tag{140}$$

and similarly

$$(e^{i\mathbf{k}_s\mathbf{R}})_{\mathbf{P},\,\mathbf{P}'} = \delta(\mathbf{P},\,\mathbf{P}'+h\mathbf{k}_s/2\pi). \tag{140'}$$

Using (139), (140), (110) and proceeding quite analogously to the treatment of emission from fixed atoms [cf. (122)] we have:

$$\left(\frac{d}{2\pi i dt}+\frac{(\mathbf{P}-\mathbf{G}_s)^2}{2Mh}+\nu_s\right)C_s(\mathbf{P}-\mathbf{G}_s) = \tilde{A}_s C(\mathbf{P}),$$
$$\left(\frac{d}{2\pi i dt}+\frac{\mathbf{P}^2}{2Mh}+\nu(n'n)\right)C(\mathbf{P}) = \textstyle\sum_s \tilde{A}_s^* C_s(\mathbf{P}-\mathbf{G}_s), \tag{141}$$

where

$$\tilde{A}_s = (2\pi h\nu_s V)^{-\frac{1}{2}}(\textstyle\sum_i e_i\mathbf{i}_i\mathbf{f}_s)_{nn'} \tag{141'}$$

and

$$\mathbf{G}_s = h\mathbf{k}_s/2\pi = (\mathbf{k}_s/k_s)(h\nu_s/c). \tag{141''}$$

The vector $\mathbf{G}_s$ is usually called the *momentum of the light quantum*. *Eqs. (141)* are seen to *connect* probability amplitudes of *states with the same total linear momentum*. Thus in the state referred to by $C(\mathbf{P})$ the linear momentum is $\mathbf{P}$, there being no light quanta; in the state referred to by $C_s(\mathbf{P}-\mathbf{G}_s)$ the light quantum has a momentum $\mathbf{G}_s$ and the matter has a momentum $\mathbf{P}-\mathbf{G}_s$, giving again $\mathbf{P}$ for the total.

Initially we have supposed all $C_s(\mathbf{P})$ to be zero. An approximate solution of (141) can be obtained for these initial conditions quite similarly to the way in which (125) was obtained from (122). Thus,

$$C(\mathbf{P}) = C^{(0)}(\mathbf{P})\exp\{-2\pi i[\nu(n'n)+(P^2/2Mh)-i\Gamma]t\}, \tag{142}$$
$$C_s(\mathbf{P}-\mathbf{G}_s) = C^{(0)}(\mathbf{P})A_s\frac{\exp\{-2\pi i[\nu(n'n)+(P^2/2Mh)-i\Gamma]t\}-\exp\{-2\pi i[\nu_s+((\mathbf{P}-\mathbf{G}_s)^2/2Mh)]t\}}{\nu_s-(\mathbf{P}\mathbf{G}_s/Mh)+(G_s^2/2Mh)-\nu(n'n)+i\Gamma},$$

where $C^{(0)}(\mathbf{P})$ is the value of $C(\mathbf{P})$ at the time $t=0$. Eqs. (142) thus describe emission in terms of the initial probability amplitudes for different momenta of the emitting atom. The damping constant Γ is a function of $\mathbf{P}$. Its dependence on $\mathbf{P}$ is slight provided M is large. In order not to complicate the discussion we shall neglect the dependence of Γ on $\mathbf{P}$.

Eq. (138) gives the effective value of the square of a component of the electric vector as observed by means of an exploring atom. In its derivation the stationary states, which have been denoted by n were supposed to be discrete. In the present problem the stationary states are continuous and are denoted by $\mathbf{P}$. The summation over n must be replaced by integration over P_x, P_y, P_z. We have

$$(\mathfrak{f}\mathfrak{E})^2=\frac{4\pi h}{V}\sum_{s',s''}(\mathfrak{f}\mathfrak{f}_{s'})(\mathfrak{f}\mathfrak{f}_{s''})(\nu(s')\nu(s''))^{\frac{1}{2}}e^{-i(\mathbf{k}_{s'}-\mathbf{k}_{s''})\mathbf{r}}\int C_{s'}^*(\mathbf{P})C_{s''}(\mathbf{P})d\Omega_{\mathbf{P}}, \tag{143}$$

where

$$d\Omega_{\mathbf{P}}=dP_x dP_y dP_z. \tag{143'}$$

The integral over $d\Omega_{\mathbf{P}}$ is seen to determine the coefficient of $e^{i(\mathbf{k}_{s''}-\mathbf{k}_{s'})\mathbf{r}}$ in the Fourier expansion of $(\mathfrak{f}\mathfrak{E})^2$. In order that $(\mathfrak{f}\mathfrak{E})^2$ be localized in space it is necessary for these integrals to have appreciable values for the probable ranges of $\mathbf{k}_{s'}$, $\mathbf{k}_{s''}$. The localization of the light intensity in space at a given time may be thought of as the result of the interference of the light quanta $\mathbf{k}_{s'}$, $\mathbf{k}_{s''}$, the effectiveness of the interference being determined by the integral over $d\Omega_{\mathbf{P}}$ in (143). The propagation of the light wave is then described as the *motion of* the *interference fringes* formed by $\mathbf{k}_{s'}$, $\mathbf{k}_{s''}$ and is brought about by the time dependence of the integrals over $d\Omega_{\mathbf{P}}$.

The same point of view can be taken also in classical electrodynamics. Here

$$(\mathfrak{f}\mathfrak{E})^2=\sum_{s',s''}(4\pi^2/V)(\nu(s')\nu(s''))^{\frac{1}{2}}(\mathfrak{f}_{s'}\mathfrak{f})(\mathfrak{f}_{s''}\mathfrak{f})[a_{s'}a_{s''}^*e^{i(\mathbf{k}_{s''}-\mathbf{k}_{s'})\mathbf{r}}+a_{s''}a_{s'}^*e^{i(\mathbf{k}_{s'}-\mathbf{k}_{s''})\mathbf{r}}$$
$$-a_{s'}a_{s''}e^{-i(\mathbf{k}_{s'}+\mathbf{k}_{s''})\mathbf{r}}-a_{s'}^*a_{s''}^*e^{i(\mathbf{k}_{s''}+\mathbf{k}_{s''})\mathbf{r}}]. \tag{143''}$$

When the motion of the emitting system has subsided the a_s depend on the time as $a_s(0)e^{2\pi i\nu_s t}$ [see (7')]. They are thus analogous to the $C_s^*(\mathbf{P})$. The first two terms in the brackets of (143'') are similar to (143). Their dependence on the time has the character

$$e^{2\pi i(\nu(s')-\nu(s''))t}.$$

The $\nu(s')$, $\nu(s'')$ cover a range of the order of magnitude of Γ. These terms in $a_{s'}a_{s''}^*$ represent, therefore, the motion of the interference fringes through space which takes place for a given point in approximately the time $(2\pi\Gamma)^{-1}$. The terms in $a_{s'}a_{s''}$, $a_{s'}^*a_{s''}^*$ depend on the time as

$$\exp[\pm 2\pi i(\nu(s')+\nu(s''))t].$$

They represent high-frequency changes in the light intensity. They have no analogon in (143). This is due to the fact that only states of approximately equal energies have been considered. The absence of high-frequency fluctuations in the light intensity is not serious since such fluctuations are usually not accessible to observation. We have:

$$\int C_{s'}^*(\mathbf{P})C_{s''}(\mathbf{P})d\Omega_{\mathbf{P}}=\bar{A}_{s'}^*A_{s''}I(s',s'')\exp[2\pi i(\nu(s')-\nu(s''))t] \tag{144}$$

$$I(s',s'')=\int C^{*0}(\mathbf{P}+\mathbf{G}_{s'})C^0(\mathbf{P}+\mathbf{G}_{s''})\left[-1+\exp\left\{-2\pi i(\nu(s')-\nu(n'n)-\frac{\mathbf{P}\mathbf{G}_{s'}}{Mh}-\frac{\mathbf{G}_{s'}^2}{2Mh}-i\Gamma)t\right\}\right]$$
$$\left[-1+\exp\left\{2\pi i\left(\nu(s'')-\nu(n'n)-\frac{\mathbf{P}\mathbf{G}_{s''}}{Mh}-\frac{\mathbf{G}_{s''}^2}{2Mh}+i\Gamma\right)t\right\}\right]\left[\nu(s')-\nu(n'n)-\frac{\mathbf{P}\mathbf{G}_{s'}}{Mh}-\frac{\mathbf{G}_{s'}^2}{2Mh}-i\Gamma\right]^{-1}$$
$$\times\left[\nu(s'')-\nu(n'n)-\frac{\mathbf{P}\mathbf{G}_{s''}}{Mh}-\frac{\mathbf{G}_{s''}^2}{2Mh}+i\Gamma\right]^{-1}d\Omega_P. \tag{144'}$$

It is useful to have an expression for $I(s', s'')$ also in terms of the wave function $\psi(\mathbf{R})$ describing the state of the center of mass of the atom at $t=0$. We have [cf. (61′)]

$$\psi(\mathbf{R})=\int h^{-\frac{1}{2}}C^0(\mathbf{P})e^{2\pi i\mathbf{PR}/h}d\Omega_P; \qquad C^0(\mathbf{P})=\int h^{-\frac{1}{2}}\psi(\mathbf{R})e^{-2\pi i\mathbf{PR}/h}d\Omega_R. \tag{145}$$

The terms in [ ] of (144′) can be expressed as a double integral of exponential functions. This substitution and the substitution of $C^*(\mathbf{P}+\mathbf{G}_{s'})$, $C(\mathbf{P}+\mathbf{G}_{s''})$ by means of (145) give $I(s', s'')$ as an integral which reduces, on integrating over $\mathbf{P}$, to

$$4\pi^2\int_0^t\int_0^t dt'dt''\int\int\psi^*(\mathbf{R}')\delta\left(\mathbf{R}'-\mathbf{R}''+\frac{\mathbf{G}_{s'}t'}{M}-\frac{\mathbf{G}_{s''}t''}{M}\right)\psi(\mathbf{R}'')\exp\left\{-2\pi i\left[\nu(s')-\nu(n'n)-\frac{\mathbf{G}_{s'}^2}{2Mh}-i\Gamma\right]t'\right.$$
$$\left.+2\pi i\left[\nu(s'')-\nu(n'n)-\frac{\mathbf{G}_{s''}^2}{2Mh}+i\Gamma\right]t''+\frac{2\pi i}{h}(\mathbf{G}_{s'}\mathbf{R}'-\mathbf{G}_{s''}\mathbf{R}'')\right\}d\Omega_{R'}d\Omega_{R''}.$$

We let

$$R=R'+\mathbf{G}_{s'}t'/M=R''+\mathbf{G}_{s''}t''/M$$

and we have

$$I(s', s'')=4\pi^2\int_0^t\int_0^t dt'dt''\int\psi^*\left(\mathbf{R}-\frac{\mathbf{G}_{s'}t'}{M}\right)\psi\left(\mathbf{R}-\frac{\mathbf{G}_{s''}t''}{M}\right)\exp\left\{\frac{2\pi i}{h}(\mathbf{G}_{s'}-\mathbf{G}_{s''})\mathbf{R}\right.$$
$$\left.-2\pi i\left[\nu(s')-\nu(n'n)-\frac{\mathbf{G}_{s'}^2}{2Mh}-i\Gamma\right]t'+2\pi i\left[\nu(s'')-\nu(n'n)-\frac{\mathbf{G}_{s''}^2}{2Mh}+i\Gamma\right]t''\right\}d\Omega_{\mathbf{R}}. \tag{146}$$

By substituting into (143) and using (144):

$$\overline{(\mathbf{f}\mathcal{E})^2}=\frac{16\pi^2h}{V}\int\left|\sum_s(\mathbf{f}\mathbf{f}_s)\nu_s^{\frac{1}{2}}\tilde{A}_s e^{i(\mathbf{k}_s\mathbf{r}-2\pi\nu_s t)}\int_0^t\psi\left(\mathbf{R}-\frac{\mathbf{G}_s\tau}{M}\right)\exp\left\{2\pi i\left(\nu_{s'}-\nu(n'n)+i\Gamma+\frac{\mathbf{G}_s^2}{2Mh}\right)\tau\right.\right.$$
$$\left.\left.-\frac{2\pi i\mathbf{G}_s\mathbf{R}}{h}\right\}d\tau\right|^2 d\Omega_{\mathbf{R}}. \tag{147}$$

If M is made very large the distance $G_s\tau/M$ can be made so small for all the important values of τ that $\psi(\mathbf{R}-G_s\tau/M)$ is practically the same as $\psi(\mathbf{R})$. For such sufficiently large values of M (147) simplifies into

$$\overline{(\mathbf{f}\mathcal{E})^2}=\frac{4\pi h}{V}\int\left|\sum_s(\mathbf{f}\mathbf{f}_s)\nu_s^{\frac{1}{2}}(A_s e^{-i\mathbf{k}_s\mathbf{R}})e^{i\mathbf{k}_s\mathbf{r}}\frac{e^{-2\pi i(\nu(n'n)-i\Gamma)t}-e^{-2\pi i\nu_s t}}{\nu_s-\nu(n'n)+i\Gamma}\right|^2|\psi(\mathbf{R})|^2d\Omega_{\mathbf{R}}. \tag{147′}$$

By (141′), (122′) $\tilde{A}_s e^{-i\mathbf{k}_s\mathbf{R}}$ is just what A_s would be for an atom with a fixed nucleus put at the point $\mathbf{R}$. Comparing now (147′) with (138) and the emission solution (125) for fixed nuclei we see that (147′) may be thought of as the result of adding the light intensities at the point r due to atoms at the points R, each intensity being given a weight $|\psi(\mathbf{R})|^2$. *In this approximation*, therefore, *the points R of the initial wave package may be considered as emitting independent and incoherent spherical waves* [cf. (130‴)]. If the wave package is made very small, the spherical waves differ very little from each other and the propagation of $\overline{(\mathbf{f}\mathcal{E})^2}$ becomes identical with that of a fixed atom.

We call the waves represented by the integrand of (147′) *spherical* because we have already seen [(130), (130‴)] that their effect on scattering

atoms is exactly as though we had a spherical wave of total energy $h\nu(n'n)$ emitted. It may also be verified directly that the summation over s leads to a wave of this character. The calculation is exactly similar to the one made for (130'''). Keeping ν_s constant and averaging over the directions of $\mathbf{f}_s$ the average value of $(\mathbf{f}\mathbf{f}_s)\tilde{A}_s e^{i\mathbf{k}_s(\mathbf{r}-\mathbf{R})}$ is found to be

$$\mathbf{f}^{(p)}(\sum e_i \mathbf{r}_i^{(p)})_{nn'}(2\pi h\nu_s V)^{-\frac{1}{2}}\frac{\sin k_s|\mathbf{r}-\mathbf{R}|}{2k_s|\mathbf{r}-\mathbf{R}|},$$

where the superscript (p) indicates that instead of the vector one takes its vector projection in a plane perpendicular to the line of $\mathbf{r}-\mathbf{R}$. Substituting and performing the integration over ν_s it is found that

$$\overline{(\mathbf{f}\mathcal{E})^2}=8\pi^2\nu^2(n'n)c^{-4}|\mathbf{f}^{(p)}(\sum e_i\mathbf{r}_i^{(p)})_{nn'}|^2\int_{ct>|\mathbf{r}-\mathbf{R}|}\left|\frac{e^{-2\pi i(\nu(n'n)-i\Gamma)(t-|\mathbf{r}-\mathbf{R}|/c)}}{|\mathbf{r}-\mathbf{R}|}\right|^2|\psi(\mathbf{R})|^2d\Omega_{\mathbf{R}},$$

the integrand being zero if $ct<|\mathbf{r}-\mathbf{R}|$ and otherwise having the value written above. Allowing $4\pi\Gamma t$ to be large, using (124), and taking the integral of $\mathcal{E}_x^2+\mathcal{E}_y^2+\mathcal{E}_z^2$ over all space we obtain

$$4\pi h\nu(n'n),$$

so that the energy of the electric field is $\frac{1}{2}h\nu(n'n)$ and the total energy is $h\nu(n'n)$.

In order to obtain the approximation (147') we had to suppose that a change in $\mathbf{R}$ of the amount $G_s\tau/M$ produced no effect on the value of $\psi(\mathbf{R})$. The expansion of $\psi(\mathbf{R})$ in terms of $C^{(0)}(\mathbf{P})$ shows that this is accomplished if the range of values of $\mathbf{P}$ for which $C^{(0)}(\mathbf{P})$ is appreciable is kept constant and if M increases indefinitely. In the limit of infinite M the atom has then the velocity $\mathbf{P}/M=0$. The limiting form (147') corresponds, therefore, to atoms of an infinite mass and at rest. The limit of infinite mass may be approached also in such a way as to represent moving atoms. This condition is obtained by keeping $C^{(0)}(\mathbf{P})$ constant for a given $\Delta\mathbf{P}$ when $P=P_0+\Delta P$. The velocity of the wave packet is then $\mathbf{P}_0/M$. This quotient is kept constant as M approaches infinity. Thus, we let

$$\psi(\mathbf{R})=e^{(2\pi i/h)\mathbf{P}_0\mathbf{R}}\chi(\mathbf{R}), \tag{148}$$

where $\chi(\mathbf{R})$ is independent of $\mathbf{P}_0$ and suffers a negligible change when $\mathbf{R}$ is changed into $R-\mathbf{G}_s\tau/M$. Substituting this $\psi(\mathbf{R})$ into (147) we have:

$$\overline{(\mathbf{f}\mathcal{E})^2}=\frac{4\pi h}{V}\int\left|\sum_s\frac{(\mathbf{f}\mathbf{f}_s)\nu_s^{\frac{1}{2}}\tilde{A}_s e^{-i\mathbf{k}_s(\mathbf{R}-\mathbf{r})}}{\nu_s-\nu(n'n)-(\mathbf{P}_0\mathbf{G}_s/(Mh))+i\Gamma}\left[e^{-2\pi i(\nu(n'n)+\mathbf{P}_0\mathbf{G}_s/Mh-i\Gamma)t}-e^{-2\pi i\nu_s t}\right]\right|^2|\psi(\mathbf{R})|^2d\Omega_{\mathbf{R}}. \tag{148'}$$

This represents the *emission from a moving atom* with neglect of the spreading of the wave packet due to different values of $\mathbf{P}/M$. If Γ is small the emitted frequencies are given by

$$\nu_s-\mathbf{P}_0\mathbf{G}_s/Mh=\nu(n'n),$$

which on using (141'') becomes

$$\nu_s=\nu(n'n)/[1-(\mathbf{P}_0/Mc)(\mathbf{k}_s/k_s)]$$

which is the Doppler effect formula for emission from a moving source. Similarly to (147'), Eq. (148') can be interpreted as the superposition of the intensities of light emitted classically by moving point sources $\mathbf{R}$ the velocity of each point being $\mathbf{P}_0/M$.

Both (147') and (148') are approximations valid only as long as $\mathbf{G}_s\tau/M$ is small in comparison with the dimensions of the wave packet $\psi(\mathbf{R})$. The important values of τ lie in the range from $\tau=0$ to values equal to several times the mean life $(4\pi\Gamma)^{-1}$. This restricting condition may be written

$$|\Delta\mathbf{R}|\gg(|\mathbf{G}_s|/M)(2\pi\Gamma)^{-1}. \tag{149}$$

It means that owing to the recoil action of the emitted light quantum the atom during its mean life should move through a distance which is small in comparison with the size of the wave packet. Supposing that the spread in the momentum $|\Delta\mathbf{P}|$ has an order of magnitude given by

$$|\Delta\mathbf{R}|\,|\Delta\mathbf{P}| \sim h/(2\pi),$$

we have

$$(|\Delta\mathbf{P}|/M)\cdot(\nu/c) \ll \Gamma. \qquad (149')$$

For ordinary wave packets the condition (149) expressed in the form (149′) means that the momentum distribution should be so sharp as to make the changes in the Doppler effect due to the spread of possible values of the momentum negligible in comparison with the line breadth. If, therefore, the line is so narrow that the Doppler effect due to $\Delta\mathbf{P}$ is readily observable the points $\mathbf{R}$ of the wave packet cease to act as sources of independent and incoherent waves.

By (146) the strength of the interference fringe between the light quanta s', s'' is determined by $I(s', s'')$. Neglecting $\mathbf{G}_s\tau/M$ in ψ of (147) is equivalent to neglecting changes in $\mathbf{P}$ in the denominators of (144′). The integration over $d\Omega_{\mathbf{P}}$ in (144′) becomes confined to the numerator. The absolute value of

$$\int C^{*0}(\mathbf{P}+\mathbf{G}_{s'})C^{0}(\mathbf{P}+\mathbf{G}_{s''})d\Omega_{\mathbf{P}} = \int \psi^{*}(\mathbf{R})e^{(2\pi i/h)(G_{s''}-G_{s'})\mathbf{R}}\psi(\mathbf{R})d\Omega_{\mathbf{R}} \qquad (150)$$

may be taken as a measure to within which the strength of the interference between s', s'' is preserved by the wave package. The most favorable condition for all frequencies is to have a very concentrated $\psi(\mathbf{R})$ and a flat $C^{0}(\mathbf{P})$. In order that interference be pronounced we, thus, want

$$|\Delta\mathbf{R}| \ll \lambda = c/\nu. \qquad (150')$$

This restriction is of an opposite kind to that of (149). If M and Γ are such that both (149) and (150′) can be satisfied the conditions for the emission from stationary atoms can be approximated. The conditions (149), (149′) are always in the way of approximating the emission from stationary atoms quite exactly. For a hydrogen atom taking $(2\pi\Gamma)^{-1}$ to be 10^{-8} sec. and $h\nu = 5$ volts the right side of (149) is of the order of 10^{-6} cm which is not very small in comparison with the wave-length.

If both (149) and (150′) are to be fulfilled then it follows *a fortiori* that

$$2\pi\Gamma \gg h\nu^{2}/Mc^{2}, \qquad (151)$$

so that there is not much meaning to the inclusion of the term

$$G_s^{2}/2Mh = h\nu_s^{2}/2Mc^{2}$$

in the exponential of (147). This term changes the frequency $\nu(n'n)$ into $\nu(n'n) - G_s^{2}/2Mh$. It represents thus the *frequency change due to* the *recoil* of a stationary atom. It is closely allied to the Compton effect. In order that this frequency change be observable it is necessary to have

$$\Gamma \ll h\nu^{2}/2Mc^{2}. \qquad (152)$$

It is, thus, impossible to observe it satisfactorily under the conditions required for approximating a spherical wave since these conditions lead to a directly opposite requirement (151). In addition we must also require

$$|\Delta\mathbf{P}| \ll h\nu/c, \qquad (152')$$

which, for wave packets satisfying $|\Delta\mathbf{R}|\,|\Delta\mathbf{P}| \sim h/(2\pi)$ leads to

$$\lambda \ll 2\pi|\Delta\mathbf{R}|. \qquad (152'')$$

The wave packet must be thus spread through a region large compared with the wave-length.

Since (152) means that the distance travelled by an atom due to recoil during the time $(1/2\Gamma)$ is large compared to λ the term $\mathbf{G}_s\tau/M$ cannot be neglected in (147) for cases where the line is sharper than the frequency shift $G_s^{2}/2Mh$. The $|\ |^{2}$ in (147) depends now not only on the value of $|\psi(\mathbf{R})|^{2}$ but on the special phase relationships between all the $\psi(\mathbf{R}-\mathbf{G}_s\tau/M)$. No single point of the wave packet can be considered now as responsible for the coefficient of $d\Omega_{\mathbf{R}}$. On the contrary all points on half a straight line $[\mathbf{R}-(\mathbf{G}_s\tau/M)(0<\tau<\infty)]$ in the initial wave packet are seen to interfere. There is no question of resemblance to spherical waves after the integration over τ has been performed. The *spherical waves* due to each $d\tau$ *interfere and give rise to unidirectional quanta* having different frequencies in different directions and described by (142) for small Γ and $|\Delta\mathbf{P}|$.

The above relations may be illustrated by means of the Gauss error curve wave packet used by Heisenberg:

$$\psi(\mathbf{R})=\pi^{-\frac{3}{4}}(\Delta X\cdot\Delta Y\cdot\Delta Z)^{-\frac{1}{2}}\exp\left[-\frac{(X-X_0)^2}{2(\Delta X)^2}-\frac{(Y-Y_0)^2}{2(\Delta Y)^2}-\frac{(Z-Z_0)^2}{2(\Delta Z)^2}+\frac{2\pi i}{h}(\mathbf{P}_0\mathbf{R})\right],$$

$$\mathbf{R}=(X,\ Y,\ Z);\quad \mathbf{R}_0=(X_0,\ Y_0,\ Z_0). \tag{153}$$

Here ΔX, ΔY, ΔZ are numbers determining the spread of the wave packet in space. $\mathbf{P}_0$ is a constant equal to the mean momentum. For the evaluation of (146) we need to perform the integration over $d\Omega_{\mathbf{R}}$. It is found that

$$\int\psi^*\left(\mathbf{R}-\frac{\mathbf{G}_{s'}t'}{M}\right)\exp\left[(2\pi i/h)(\mathbf{G}_{s'}-\mathbf{G}_{s''})\mathbf{R}\right]\psi\left(\mathbf{R}-\frac{\mathbf{G}_{s''}t''}{M}\right)d\Omega_{\mathbf{R}}$$

$$=\exp\left\{-\sum_{x,y,z}\left[\frac{(G_{xs'}t'-G_{xs''}t'')^2}{4(\Delta X)^2M^2}+\left(\frac{\pi(G_{xs'}-G_{xs''})\Delta X}{h}\right)^2\right]\right.$$

$$\left.+\frac{2\pi i}{h}\left[(\mathbf{G}_{s'}-\mathbf{G}_{s''})\left(\mathbf{R}_0+\frac{\mathbf{G}_{s'}t'+\mathbf{G}_{s''}t''}{2M}\right)+\frac{\mathbf{P}_0}{M}(\mathbf{G}_{s'}t'-\mathbf{G}_{s''}t'')\right]\right\}. \tag{153'}$$

The terms of second degree in t', t'' may be neglected in the approximation (148). Doing so and introducing $\Delta\mathbf{P}$ by

$$\Delta X\cdot\Delta P_x=h/2\pi$$

it is found that:

$$I(s',s'')=\frac{(e^{-2\pi i(x'-i\Gamma)t}-1)(e^{2\pi i(x''+i\Gamma)t}-1)}{(x'-i\Gamma)(x''+i\Gamma)}\exp\left[\frac{2\pi i}{h}(\mathbf{G}_{s'}-\mathbf{G}_{s''})\mathbf{R}_0-\sum_{x,y,z}\frac{(G_{xs'}-G_{xs''})^2}{4(\Delta P_x)^2}\right], \tag{153''}$$

where

$$x'=\nu(s')-\nu(n'n)-\frac{\mathbf{P}_0\mathbf{G}_{s'}}{Mh}+\frac{\mathbf{G}_{s'}\mathbf{G}_{s''}}{2Mh};\qquad x''=\nu(s'')-\nu(n'n)-\frac{\mathbf{P}_0\mathbf{G}_{s''}}{Mh}+\frac{\mathbf{G}_{s'}\mathbf{G}_{s''}}{2Mh}.$$

The terms $G_{s'}G_{s''}/(2Mh)$ do not have much significance since t'^2 has been dropped in (153'). The factor

$$\exp\left[-\sum_{x,y,z}\frac{(G_{xs'}-G_{xs''})^2}{4(\Delta P_x)^2}\right]=\exp\left[-\sum_{x,y,z}\left(\frac{(k_{xs'}-k_{xs''})\Delta X}{2}\right)^2\right] \tag{153'''}$$

represents the blurring effect due to the space distribution of the wave packet. By (147'), (148') it should be the same in absolute value as

$$\int\exp\left[i(\mathbf{k}_{s'}-\mathbf{k}_{s''})\mathbf{R}\right]|\psi(\mathbf{R})|^2d\Omega_{\mathbf{R}},$$

which is readily verified using (153).

§4. The polarization of resonance radiation

The polarization of resonance radiation has been treated by means of Dirac's theory of light quanta by Weisskopf.[20] The theory of the absorption and scattering by an atom from a nondegenerate state presented above in VI, §3 has been extended by him to cover the case of a degenerate normal state and a degenerate upper level. The intensity of radiation scattered due to a given frequency of incident radiation $\nu(s_0)$ is then integrated over $\nu(s_0)$. The results are applied to the calculation of the angle of maximum polarization in the special case of a nondegenerate normal state ($j=0$) and a triply degenerate upper level.

The theory is easily generalized so as to cover the possible cases of groups of hyperfine structure levels which arise in practice. The calculation can be made simpler by avoiding the integration over the incident frequencies. The resonance bulb is

supposed below to be subjected to the action of radiation from an atom having a fairly large damping constant Γ_I. We make Γ_I large in comparison with the damping constant Γ of the scattering atom and we thus approach the condition of white radiation being incident. The calculation is very similar to that of VI, §4. We refer to the sublevels of the normal state by m and to the sublevels of the upper level by μ. The scattering atom is supposed to be initially in a definite sublevel m_0. The emitting atom is taken for simplicity as having nondegenerate states. We use the following probability amplitudes: $c_{s,m}$ for light quantum s, scatterer in m, emitter in normal state; c_μ for no light quantum, scatterer in μ, emitter in normal state; $c_{I,m}$ for no light quantum, scatterer in m, emitter in excited state. The transition frequency of the emitter we call ν_I, the transition frequencies between μ and m of the scatterer $\nu_{\mu m}$; by A_s^I we mean expression (122′) for the emitter, by $A_s^{m\mu}$ the same expression for the scatterer using $n=m$, $n'=\mu$. We have to solve the equations

$$(d/2\pi i dt+\nu_s+\nu(m))c_{s,m}=A_s^I c_{I,m}+\sum_\mu A_s^{m\mu}c_\mu,$$
$$(d/2\pi i dt+\nu_I+\nu(m))c_{I,m}=\sum_s (A_s^I)^* c_{s,m},$$
$$(d/2\pi i dt+\nu(\mu))c_\mu=\sum_{s,m}(A_s^{m\mu})^* c_{s,m}.$$

If there were no scattering the solutions would be

$$c_{I,m}^{(0)}=\delta_{m,m_0}e^{-2\pi i(\nu_I+\nu_m-i\Gamma_I)t},$$
$$c_{s,m}^{(0)}=\delta_{m,m_0}A_s^I\frac{e^{-2\pi i(\nu_I+\nu_m-i\Gamma_I)t}-e^{-2\pi i(\nu_s+\nu_m)t}}{\nu_s-\nu_I+i\Gamma_I}.$$

Analogously to (130″) we have:

$$\begin{aligned}(d/2\pi i dt+\nu_s+\nu(m))c_{s,m}^{(1)}&=\sum_\mu A_s^{m\mu}c_\mu^{(1)},\\ (d/2\pi i dt+\nu(\mu))c_\mu^{(1)}&=\sum_{s,m}(A_s^{m\mu})^*(c_{sm}^{(0)}+c_{sm}^{(1)}),\end{aligned}\tag{155}$$

and similarly to (130‴)

$$\sum_s (A_s^{m\mu})^* c_{sm}^{(0)}=K_{\mu m}\delta_{mm_0}e^{-2\pi i(\nu_I+\nu(m)-i\Gamma_I)(t-\tau)},$$
$$K_{\mu m}=(e\mathfrak{x})_{\mu m}^{(p)}(e\mathfrak{x}^I)_{n_I n_I'}^{(p)}/(R_{II}hc^2);\qquad \tau=R_{II}/c.$$

We let $t'=t-\tau$ and we look for solutions of the type

$$c_\mu^{(1)}=K_\mu'\exp\{-2\pi i(\nu_I+\nu(m_0)-i\Gamma_I)t'\},$$
$$c_{sm}^{(1)}=\sum K_\mu' A_s^{m\mu}[\nu_s-\nu_I+i\Gamma_I+\nu(mm_0)]^{-1}[\exp\{-2\pi i(\nu_I+\nu(m_0)-i\Gamma_I)t'\}-\exp\{-2\pi i(\nu_s+\nu(m))t'\}].$$

Substituting these into (155) we get

$$K_\mu'(\nu_\mu-\nu_I-\nu(m_0)+i\Gamma_I)=K_{\mu m_0}+\sum_{\mu'}i\Gamma_{\mu\mu'}K_{\mu'}',\tag{156}$$

where

$$\Gamma_{\mu\mu'}=\sum_m(\pi/\Delta\nu)\overline{(A_s^{m\mu})^*A_s^{m\mu'}}\tag{156′}$$

analogously to (124). The bar indicates as before an average over the directions of s. Expression (156) is thus to within a constant factor

$$\sum_m\{(e\dot{x})^{\mu m}(e\dot{x})^{m\mu'}+(e\dot{y})^{\mu m}(e\dot{y})^{m\mu'}+(e\dot{z})^{\mu m}(e\dot{z})^{m\mu'}\}.$$

The calculation is simplified by the fact that in the important cases which may arise this sum vanishes unless μ is μ'. In fact it will be shown that

$$\Gamma_{\mu\mu'}=\Gamma\delta_{\mu\mu'},\tag{156″}$$

where Γ is independent of μ. That this is true is well known for the case of no nuclear spin and definite values of the total electronic angular momentum for the upper as well as the lower states. For it is readily shown that the matrix $\Gamma_{\mu\mu'}$ commutes with the matrices representing the three components of angular momentum for the upper state. A matrix commuting with these three matrices is known to be a constant times a unit matrix which is in agreement with (156″). If the nuclear spin **I** is coupled to the electronic angular momentum **J**, Eq. (156″) still remains correct. To see this, imagine the coupling of **I** to **J** to be first very weak. The nuclear spin orients itself in the magnetic field independently of **J**. Each sublevel μ must then be characterized by the projections m_J and m_I. In this case (156″)

is, thus, correct because the eigenfunctions are simple products of the electronic and the nuclear-spin functions. If next the nuclear spin is coupled more strongly to $\mathbf{J}$ the eigenfunctions become linear combinations of the products. The transformation from the decoupled to the coupled condition can be represented as a unitary transformation of the wave functions. There is one unitary transformation S_a for the lower state a and another one S_b for the upper one b. It is readily seen that

$$\|\Gamma_{\bar{\mu}\bar{\mu}'}\| = S_b^{-1}\|\Gamma_{\mu\mu'}\|S_b,$$

where the bar indicates a state of the coupled condition. The $\Gamma_{\mu\mu'}$ matrix transforms itself entirely as $\mathbf{J}^2$ which is a constant times a unit matrix both for the weak as well as the strong field condition. Eq. (156″) remains, therefore, correct.

We now solve (156) for K_μ' and obtain possible solutions for $c_\mu^{(1)}$, $c_{sm}^{(1)}$. These solutions are not yet quite suitable for the discussion of the problem because $c_\mu^{(1)} \neq 0$ at $t'=0$. They must be combined with emission solutions from the levels μ in such a way that $c_\mu^{(1)}$ should vanish at $t'=0$. The emission solutions are obtained similarly to VI, §2. In the derivation, use is made of the fact that $\Gamma_{\mu\mu'}$ is Γ times a unit matrix. The result of the combination with the emission solutions gives for large t

$$c_{sm}^{(1)} = \sum_\mu A_s^{m\mu} K_{\mu m_0} e^{-2\pi i(\nu_s+\nu(mi))t'}/(\nu_s-\nu_I+i\Gamma_I+\nu(mm_0))(\nu_s-\nu(\mu m)+i\Gamma). \tag{157}$$

We make Γ_I large and obtain by so doing the condition of radiation having a uniform intensity distribution in the spectrum. Then

$$|c_{sm}^{(1)}|^2 = \Gamma_I^{-2}\sum_{\mu'}{}_{,\mu} K_{\mu' m_0}{}^* K_{\mu m_0}(A_s^{m\mu'})^* A_s^{m\mu}/(\nu_s-\nu(\mu' m)-i\Gamma)(\nu_s-\nu(\mu m)+i\Gamma). \tag{157'}$$

The number of light quanta scattered into a small solid angle $\Delta\omega$ having a polarization of type s is then

$$\sum_m \frac{\Delta\omega}{8\pi\Delta\nu}\int |c_{s,m}^{(1)}|^2 d\nu_s. \tag{158}$$

This may be summed over all initial states m_0 and divided over the statistical weight g_a of the lower state a. The result is the probability of finding a light quantum of polarization s per atom of scattering substance and per atom of the source. The result is

$$I = (C/g_a)\sum\binom{m,m'}{\mu,\mu'}(e\mathbf{r}\mathbf{f})_{\mu m}(e\mathbf{r}\mathbf{f})_{m\mu'}(e\mathbf{r}\mathbf{f}_s)_{\mu' m'}(e\mathbf{r}\mathbf{f}_s)_{m'\mu}/(1-2\pi i\tau\nu(\mu',\mu)), \tag{159}$$

where $\mathbf{f}$ is a unit vector in the direction of the projection of $(e\mathbf{r}^\Gamma)_{n_I n_I'}$ on to the plane perpendicular to the line joining the emitting atom to the scatterer, i.e., $\mathbf{f}$ is a unit vector parallel to the electric intensity of the incident light wave; $\mathbf{f}_s$ is a unit vector parallel to the electric intensity which is transmitted by the nicol prism or some other analyzing apparatus; $\tau = (4\pi\Gamma)^{-1}$ and is the mean life of the atom; C is a constant involving $R_{II\,I}$, Γ_I, Γ, which is of no interest to us.

If the incident electric wave is polarized in the x direction and if the analyzing apparatus is arranged so as transmit waves polarized along a direction ξ, we may take

$$I_\xi = (C/g_a)\sum\binom{m,m'}{\mu,\mu'}(e\dot{x})_{\mu m}(e\dot{x})_{m\mu'}(e\dot{\xi})_{\mu' m'}(e\dot{\xi})_{m'\mu}/(1-2\pi i\tau\nu(\mu'\mu)). \tag{159'}$$

If all the $\nu(\mu'\mu)=0$, Eq. (159′) gives I as the sum of the diagonal elements of a product of 4 matrices. Such a sum will be denoted by Sp. The direction of the auxiliary magnetic field used as an axis for space quantization can now be changed. The matrices are then transformed canonically. As a result the Sp is left unchanged. The intensities of radiations having different directions do not depend, therefore, on the direction of the auxiliary magnetic field. Again the result is independent of whether one supposes the nuclear spin i to be coupled to the electronic angular momentum j or not. In fact, as long as there is no energy difference between the different

states μ, coupling i to j simply means working with proper linear combinations of uncoupled states. Such a linear transformation for the upper states may be represented by a unitary transformation S_b. Similarly, for the lower state there is a similar S_a. The matrix product occurring in (159′) is transformed canonically with S_b and the Sp is unchanged. We see, therefore, that if i is coupled so weakly to j that the frequency differences between different *hfs* levels in the upper level are small in comparison with the natural breadth $1/(2\pi\tau)$, the degree of polarization of the resonance radiation is the same as though there were no nuclear spin.

We consider next upper states having *hfs* separations which are large in comparison with the natural breadth. We use thus all terms in (159′) except those for which μ and μ' lie in different hyperfine structure levels. Let the *hfs* levels of b be denoted by φ and those of a by f. The matrices involving only one φ and one f are called submatrices and are written

$$x^{\varphi f}=(x^{\varphi f}_{\mu m}), \quad x^{f\varphi}=(x^{f\varphi}_{m\mu})$$

and similarly for y and z. For any direction of polarization ξ the intensity of radiation in the absence of a magnetic field is

$$I_\xi=(C/g_a)\sum_{\varphi}\sum_{ff'} Sp[(e\dot{x})^{\varphi f}(e\dot{x})^{f\varphi}(e\dot{\xi})^{\varphi f'}(e\dot{\xi})^{f'\varphi}]. \tag{159″}$$

We have the following relations[36] for any vector with components having the same commutation relations with J_x, J_y, J_z and F_x, F_y, F_z as $(e\dot{x})$, $(e\dot{y})$, $(e\dot{z})$:

$$Sp[(e\dot{x})^{\varphi f}(e\dot{x})^{f\varphi'}(e\dot{x})^{\varphi' f'}(e\dot{x})^{f'\varphi}]=Sp[(e\dot{y})^{\varphi f}(e\dot{y})^{f\varphi'}(e\dot{y})^{\varphi' f'}(e\dot{y})^{f'\varphi}]=Sp[(e\dot{z})^{\varphi f}(e\dot{z})^{f\varphi'}(e\dot{z})^{\varphi' f'}(e\dot{z})^{f'\varphi}], \tag{160}$$

$$Sp[(e\dot{x})^{\varphi f}(e\dot{x})^{f\varphi'}(e\dot{y})^{\varphi' f'}(e\dot{y})^{f'\varphi}]=Sp[(e\dot{y})^{\varphi f}(e\dot{y})^{f\varphi'}(e\dot{z})^{\varphi' f'}(e\dot{z})^{f'\varphi}]=Sp[(e\dot{z})^{\varphi f}(e\dot{z})^{f\varphi'}(e\dot{x})^{\varphi' f'}(e\dot{x})^{f'\varphi}],$$

$$Sp[(e\dot{x})^{\varphi f}(e\dot{x})^{f\varphi}(e\dot{x})^{\varphi f'}(e\dot{x})^{f'\varphi}]+2Sp[(e\dot{x})^{\varphi f}(e\dot{x})^{f\varphi}(e\dot{y})^{\varphi f'}(e\dot{y})^{f'\varphi}]=(2\varphi+1)A^{\varphi f}A^{\varphi f'}/3, \tag{160′}$$

where

$$A^{\varphi f}=\sum_{m_f}\{|(e\dot{x})^{\varphi f}_{\mu_\varphi m_f}|^2+|(e\dot{y})^{\varphi f}_{\mu_\varphi m_f}|^2+|(e\dot{z})^{\varphi f}_{\mu_\varphi m_f}|^2\}$$

is proportional to the transition probability from φ to f and is independent of μ.

The Eqs. (160) show that (159′) is in agreement with the principle of spectroscopic stability also in the case of hyperfine structure. In addition

$$\sum_f A^{\varphi f}=A, \tag{160″}$$

where A is proportional to the transition probability from any sublevel of b and is independent of φ, μ. Thus

$$\begin{aligned} I_x&=(C/g_a)\textstyle\sum_{\varphi,\,f,\,f'} Sp[(e\dot{x})^{\varphi f}(e\dot{x})^{f\varphi}(e\dot{x})^{\varphi f'}(e\dot{x})^{f'\varphi}],\\ I_y&=(C/g_a)\textstyle\sum_{\varphi,\,f,\,f'} Sp[(e\dot{x})^{\varphi f}(e\dot{x})^{f\varphi}(e\dot{y})^{\varphi f'}(e\dot{y})^{f'\varphi}], \end{aligned} \tag{161}$$

and

$$I_x+2I_y=(C/3g_a)A^2\textstyle\sum_\varphi(2\varphi+1)=Cg_bA^2/3g_a \tag{161′}$$

by (160′).

Eq. (161) shows that the total intensity of the radiation scattered in all directions does not depend on whether the nuclear spin is coupled to the electronic angular momentum or not. This is true, of course, only as long as the incident radiation forms a broad band with a uniform distribution of intensity among frequencies. Such a condition is usually designated as that of broad incident lines. It was to be expected that (161′) should hold because otherwise the absorption

[36] M. Born and P. Jordan, *Elementare Quantenmechanik*, Julius Springer, 1930, pp. 151–162. The equations used in the text are derived without difficulty by the methods of this section.

coefficient would depend on the nuclear spin. The occurrence of A^2 in (161′) is of no significance because A is involved also in C.

We now apply a magnetic field $\mathfrak{H}$ of sufficient strength to have the Zeeman splitting comparable with Γ but not large enough to make the Paschen back effect appreciable. The direction of $\mathfrak{H}$ shall be the z axis and the angle between ξ and $\mathfrak{H}$ will be called θ. It is convenient to use the standard representation of angular momentum matrices in which (ez) is diagonal and the matrix elements of $(e(\dot{x}-i\dot{y}))$ vanish except when $\mu+1=m$ while those of $(e(\dot{x}+i\dot{y}))$ vanish except when $\mu-1=m$. Substitution into (159′) shows that

$$I_\xi=\sum_\varphi\left\{(\alpha\varphi)+\beta(\varphi)\left[\frac{e^{-2i\theta}}{1-i\tau g_\varphi e\mathfrak{H}/mc}+\frac{e^{2i\theta}}{1+i\tau g_\varphi e\mathfrak{H}/mc}\right]\right\}, \tag{162}$$

where

$$\alpha(\varphi)=(C/4g_a)\sum_{ff'}\sum_{\mu\varphi}(A_\sigma)^{\varphi f}_{\mu\varphi}(A_\sigma)^{\varphi f}_{\mu\varphi}=(C/4g_a)\sum_{\mu\varphi}(\sum_f(A_\sigma)^{\varphi f}_{\mu\varphi})^2,$$

$$\beta(\varphi)=(C/4g_a)\,|\sum_f(e\dot{x})^{\varphi f}_{\mu+2,\,\mu+1}(e\dot{x})^{f\varphi}_{\mu+1,\,\mu}|^2, \tag{162′}$$

$$(A_\sigma)^{\varphi f}_\mu=2\{|(e\dot{x})^{\varphi f}_{\mu,\,\mu+1}|^2+|(e\dot{x})^{\varphi f}_{\mu,\,\mu-1}|^2\}.$$

Here $(A_\sigma)_\mu^{\varphi f}$ may be thought of as the transition probability from the magnetic sublevel μ of φ to f due to all possible σ Zeeman components. We have further

$$\begin{aligned}\alpha(\varphi)+2\beta(\varphi)&=(C/g_a)\sum_{f,\,f'}Sp((e\dot{x})^{\varphi f}(e\dot{x})^{f\varphi}(e\dot{x})^{\varphi f'}(e\dot{x})^{f'\varphi}),\\ \alpha(\varphi)-2\beta(\varphi)&=(C/g_a)\sum_{ff'}Sp((e\dot{x})^{\varphi f}(e\dot{x})^{f\varphi}(e\dot{y})^{\varphi f'}(e\dot{y})^{f'\varphi}).\end{aligned} \tag{163}$$

Adding twice the second of these to the first and using (160′) (160″)

$$3\alpha(\varphi)-2\beta(\varphi)=(C/g_a)(2\varphi+1)A^2/3 \tag{163′}$$

Eq. (163′) together with the first equation (162′) determines $\alpha(\varphi)$, $\beta(\varphi)$ in terms of the intensities of the lines of the Zeeman pattern.

By (162) the degree of polarization in the presence of a magnetic field is

$$P=\frac{I_x-I_y}{I_x+I_y}=\sum_\varphi\frac{(P_0)_\varphi}{1+(\tau g_\varphi e\mathfrak{H}/mc)^2}, \tag{164}$$

where

$$(P_0)_\varphi=2\beta(\varphi)/\sum_\varphi\alpha(\varphi). \tag{164′}$$

The calculation can be carried through either by using (162′) for both α and β, or by using the first Eq. (162′) for α and (163′) for β. The latter method is preferable because it involves only the use of transition probabilities. One can also calculate α and β using Heisenberg's rule as applied to hyperfine structure by Ellett.[37] That this is true is seen from the fact that Eqs. (163) involve only spurs; z may be, therefore, substituted for x and x for y without changing the values of the sums. Letting μ be any sublevel of φ we have from (163)

$$\begin{aligned}\alpha(\varphi)+2\beta(\varphi)&=(C/g_a)\sum_{f,\,f'}\sum_\mu|(e\dot{z})^{\varphi f}_\mu|^2|(e\dot{z})^{\varphi f'}_\mu|^2,\\ \alpha(\varphi)-2\beta(\varphi)&=(C/g_a)\sum_{f,\,f'}\sum_\mu|(e\dot{z})^{\varphi f}_\mu|^2\{|(e\dot{x})^{\varphi f'}_{\mu,\,\mu+1}|^2+|(e\dot{x})^{\varphi f'}_{\mu,\,\mu-1}|^2\}.\end{aligned} \tag{164″}$$

The interpretation of this formula is that one can think of $\alpha(\varphi)+2\beta(\varphi)$ as being due to the absorption along π components to the sublevels μ in φ followed by emissions of π components from the same μ, the absorptions being taken proportional to $|(e\dot{z})^{\varphi f}_\mu|^2$ and the emissions to $|(e\dot{z})^{\varphi f'}_\mu|^2$. The origin of $\alpha(\varphi)-2\beta(\varphi)$ is, on the other hand, first an absorption to μ with the same probability as before followed by the emission of σ components with a probability $2\{|(e\dot{x})^{\varphi f'}_{\mu,\,\mu+1}|^2+|(e\dot{x})^{\varphi f'}_{\mu,\,\mu-1}|^2\}$. The result must then be divided by 2 in order to agree with (164″). This is in agreement with the fact that σ

[37] A. Ellett, Phys. Rev. 35, 588 (1930).

components radiate only with $\frac{1}{2}$ the efficiency of π components in a direction perpendicular to the magnetic field.

Using (160), (160′), (160″) one can express α, β in terms of quantities proportional to transition probability. We let $A_\pi^{\varphi\mu}$ be a number proportional to the total transition probability from sublevel μ of φ due to π components

$$A_\pi^{\varphi\mu}=\sum_f|(e\dot{z})_\mu^{\varphi f}|^2$$

and $A_\sigma^{\varphi\mu}$ we, similarly, make proportional to the total transition probability due to σ components

$$A_\sigma^{\varphi\mu}=2\sum_f\{|(e\dot{x})_{\mu,\mu+1}^{\varphi f}|^2+|(e\dot{x})_{\mu,\mu-1}^{\varphi f}|^2\},$$

then

$$A=A_\pi^{\varphi\mu}+A_\sigma^{\varphi\mu}$$

is the total transition probability (on the same scale) and is independent of μ and φ. In terms of the σ components

$$\alpha(\varphi)=(C/4g_a)\sum_\mu(A_\sigma^{\varphi\mu})^2,\qquad \beta(\varphi)=(3C/4g_a)\sum_\mu(A_\sigma^{\varphi\mu})^2-(C/g_a)(2\varphi+1/3)A^2.$$

By using these expressions, the polarization in zero field is

$$P_0=\sum_\varphi P_0(\varphi)=3-[4A^2(2i+1)(2j_b+1)/3\sum_{\varphi,\mu_\varphi}(A_\sigma^{\varphi\mu})^2]$$

for broad incident lines. The most convenient expressions are in terms of the π components

$$\alpha(\varphi)=(C/4g_a)\{\sum_\mu(A_\pi^{\varphi\mu})^2+(2\varphi+1)A^2/3)\},\qquad 2\beta(\varphi)=(C/4g_a)\{3\sum_\mu(A_\pi^{\varphi\mu})^2-(2\varphi+1)A^2/3\},$$

$$\sum_\varphi\alpha(\varphi)=(C/4g_a)\{\sum_{\varphi,\mu}(A_\pi^{\varphi\mu})^2+A^2(2i+1)(2j_b+1)/3\}\quad(165)$$

where i is the nuclear spin and j_b is the inner quantum number of b. For broad incident lines the polarization in zero field is[38] by (165)

$$P_0=\frac{\sum_{\varphi,\mu}3(A_\pi{}^{\varphi\mu})^2-A^2(2i+1)(2j_b+1)/3}{\sum_{\varphi,\mu}(A_\pi{}^{\varphi\mu})^2+A^2(2i+1)(2j_b+1)/3}.\quad(165')$$

In order to calculate the polarization in a magnetic field, however, it is most convenient to use Eq. (165). For narrow incident lines Eq. (164″) is the simplest starting point. One must multiply then $|(ez)_\mu{}^{\varphi f}|^2$ by $I_{\varphi f}$, the intensity of incident radiation having the frequency $\nu(\varphi f)$.

The angle of maximum polarization is found from Eq. (162). It is given by

$$\tan 2\theta=\frac{\sum_\varphi P_0(\varphi)\sin 2\theta(\varphi)\cos 2\theta(\varphi)}{\sum_\varphi P_0(\varphi)\cos^2 2\theta(\varphi)},\quad(166)$$

where

$$\tan 2\theta(\varphi)=\tau g(\varphi)e\mathfrak{H}/mc.\quad(166')$$

Let now the *distance between the hfs levels of b* be *comparable with the natural breadth* of the lines. It was already seen that if the natural breadth is large Eq. (159) gives the same result as though there were no nuclear spin. For small natural breadths all terms of (159′) in which μ and μ' referred to different *hfs* levels φ and φ' were omitted because the denominators were large and Eq. (159″) followed. For intermediate cases, in the absence of an external magnetic field we have by (159′), (160′), (165)

$$I_x=\sum_{\varphi,\mu}(C/g_a)(A_\pi{}^{\varphi\mu})^2+\sum_{f,f',\varphi>\varphi'}(2C/g_a)Sp((e\dot{x})^{\varphi f}(e\dot{x})^{f\varphi'}(e\dot{x})^{\varphi' f'}(e\dot{x})^{f'\varphi})/[1+(2\pi\tau\nu_{\varphi f})^2],$$
$$I_x+2I_y=(CA^2/3g_a)\sum_\varphi(2\varphi+1)=(CA^2/3g_a)(2i+1)(2j_b+1).\quad(167)$$

The second of these equations shows that the total amount of scattered radiation does not depend on the relative values of the *hfs* separations and the natural breadth. By (160)

$$\sum_{f,f'}Sp((e\dot{x})^{\varphi f}(e\dot{x})^{f\varphi'}(e\dot{x})^{\varphi' f'}(e\dot{x})^{f'\varphi})=\sum_\mu|\sum_f(e\dot{z})_{\mu\mu}^{\varphi f}(e\dot{z})_{\mu\mu}^{f\varphi'}|^2.\quad(167')$$

[38] Pauling and Goudsmit, *Structure of Line Spectra*, p. 241, McGraw-Hill, 1930.

As an example, consider the D_2 line of Na on the supposition that the spin of the nucleus in units of $h/2\pi$ is 1. We write the electronic eigenfunctions of the $P_{\frac{3}{2}}$ state corresponding to magnetic quantum numbers $\frac{3}{2}$, $\frac{1}{2}$ $\cdots$ as $J(\frac{3}{2})$, $J(\frac{1}{2})$, $\cdots$ similarly, the nuclear spin functions as $I(1)$, $I(0)$, $I(-1)$ and the electronic eigenfunctions of $S_{\frac{1}{2}}$ as $J_{\frac{1}{2}}'$, $J_{-\frac{1}{2}}'$. The coefficients with which the products of these functions must be used in forming the proper functions for *hfs* levels with definite magnetic quantum numbers are given in Table I.

TABLE I

$\mu=5/2$	3/2		1/2				
$J(3/2)I(1)$	$J(1/2)I(1)$	$J(3/2)I(0)$	$J(-1/2)I(1)$	$J(1/2)I(0)$	$J(3/2)I(-1)$		
1	$3^{\frac{1}{2}}/5^{\frac{1}{2}}$	$2^{\frac{1}{2}}/5^{\frac{1}{2}}$	$3^{\frac{1}{2}}/10^{\frac{1}{2}}$	$6^{\frac{1}{2}}/10^{\frac{1}{2}}$	$1/10^{\frac{1}{2}}$	$\varphi=5/2$	$b=^2P_{\frac{3}{2}}$
	$2^{\frac{1}{2}}/5^{\frac{1}{2}}$	$-3^{\frac{1}{2}}/5^{\frac{1}{2}}$	$2^{\frac{1}{2}}/15^{\frac{1}{2}}$	$-1/15^{\frac{1}{2}}$	$-6^{\frac{1}{2}}/15^{\frac{1}{2}}$	$\varphi=3/2$	
			$1/6^{\frac{1}{2}}$	$-2^{\frac{1}{2}}/6^{\frac{1}{2}}$	$3^{\frac{1}{2}}/6^{\frac{1}{2}}$	$\varphi=1/2$	
$m=$	3/2		1/2				
	$J'(1/2)I(1)$		$J'(-1/2)I(1)$	$J'(1/2)I(0)$			
	1		$1/3^{\frac{1}{2}}$	$2^{\frac{1}{2}}/3^{\frac{1}{2}}$		$f=3/2$	$a=^2S_{\frac{1}{2}}$
			$-2^{\frac{1}{2}}/3^{\frac{1}{2}}$	$1/3^{\frac{1}{2}}$		$f=1/2$	

By means of Table I (transformation matrix) the values of $\sum_f$ in (167′) can be found. The calculation is simplified by the fact that the matrix element $(J(\frac{1}{2})/(e\dot{z})/J'(\frac{1}{2}))$ is equal to $(J(-\frac{1}{2})/(e\dot{z})/J'(-\frac{1}{2}))$ as follows for instance from the fact that the state $\varphi=\frac{5}{2}$, $\mu=\frac{1}{2}$ should not combine with $f=\frac{1}{2}$, $m=\frac{1}{2}$. This matrix element we write $z(\frac{1}{2}, \frac{1}{2}')$. The coefficients of $z(\frac{1}{2}, \frac{1}{2}')$ for $(e\dot{z})_{\mu\mu}{}^{\varphi f}$ are given in Tables II and III.

TABLE II.

$\mu=3/2$			
f \ φ	5/2	3/2	1/2
3/2	$(3/5)^{\frac{1}{2}}$	$(2/5)^{\frac{1}{2}}$	
1/2			

TABLE III.

$\mu=1/2$			
f \ φ	5/2	3/2	1/2
3/2	$3(1/10)^{\frac{1}{2}}$	$(2/45)^{\frac{1}{2}}$	$-1/18^{\frac{1}{2}}$
1/2	0	$-5/45^{\frac{1}{2}}$	$-2/9^{\frac{1}{2}}$

Hence the expression (167) is found to have the following values for

$(\varphi, \varphi')=$	(5/2, 3/2)	(5/2, 1/2)	(3/2, 1/2)	(5/2, 5/2)	(3/2, 3/2)	(1/2, 1/2)	
	14/25	1/10	2/5	117/50	26/25	1/2	$\times\lvert z(1/2, 1/2')\rvert^4$.

It is easily found that $\lvert z(\frac{1}{2}, \frac{1}{2}')\rvert^2=2A^2/3$. Hence by (167)

$$I_x=(C/g_a)\lvert z(\tfrac{1}{2}, \tfrac{1}{2}')\rvert^4\left\{\frac{97}{25}+\frac{28/25}{1+(2\pi\tau\nu(5/2, 3/2))^2}+\frac{1/5}{1+(2\pi\tau\nu(5/2, 1/2))^2}+\frac{4/5}{1+(2\pi\tau\nu(3/2, 1/2))^2}\right\},$$

$$I_y=(C/g_a)\lvert z(\tfrac{1}{2}, \tfrac{1}{2}')\rvert^4\left\{\frac{64}{25}-\frac{14/25}{1+(2\pi\tau\nu(5/2, 3/2))^2}-\frac{1/10}{1+(2\pi\tau\nu(5/2, 1/2))^2}-\frac{2/5}{1+(2\pi\tau\nu(3/2, 1/2))^2}\right\}.$$

If all the $\nu(\varphi, \varphi')$ are made large, one may take $I_z=97$, $I_y=64$ corresponding to $P=0.205$. If all the $\nu(\varphi, \varphi')$ are small, one, similarly, gets $P=0.60$. According to Larrick,[39] the splitting $\nu(\frac{5}{2}, \frac{1}{2})=1.44\times10^8$ sec.$^{-1}$ so that $\nu(\frac{5}{2}, \frac{3}{2})=0.90\times10^8$ sec.$^{-1}$, $\nu(\frac{3}{2}, \frac{1}{2})=0.54\times10^8$ sec.$^{-1}$ and by using $\tau=10^{-8}$ sec., $P=0.228$. According to the latest observations of Heydenburg, Larrick and Ellett,[39] the polarization for both D lines together is $16.48\pm.33$ percent which according to their calculations corresponds closely to a polarization of 20.5 percent for D_1 alone. The calculations of these authors correspond apparently to the supposition that the strengths of the incident D_1 and D_2 lines are equal. Calculating, similarly, the expected polarization for D_1+D_2 we obtain 18.5 percent which is, therefore, somewhat higher than the observed value. If, on the other hand, the incident D_2 line is taken to have its theoretical strength of twice that of D_1, we would get only 15.6 percent with the line-breadth correction and 14.0 percent without it.

Modified formula for unpolarized incident radiation. In some experiments the incident radiation is unpolarized. This practice has the advantage of giving larger intensities of scattered radiation by eliminating losses of light in the polarizing apparatus and we shall indicate briefly the necessary treatment.

The incident radiation is considered as traveling along the y axis and the directions of observation and of the magnetic field are taken to be the z axis. We consider the incident beam as a mixture of two beams polarized along OX and OZ. The intensities of the two beams are equal and phase relations between them are supposed to be absent. The intensity due to the first beam is given by (162) where α, β are obtained by means of (165). The second beam is polarized along OZ. We consider the general formula (159). We have only terms with μ and μ' lying in the same φ. For $e\mathfrak{x}$ we must now write $e\mathfrak{z}$. For this component the selection rule $\mu=m=\mu'$ applies. Thus the sum (159) involves only terms with $\mu=\mu'$ and all $\nu_{\mu'\mu}=0$ which shows that the magnetic field has no effect on this radiation. Its intensity is therefore also independent of θ. By a direct application of the formulas already given this intensity is

$$(C/2g_a)\sum_{\varphi,\ \mu}A_x^{\varphi\mu}A_z^{\varphi\mu}=\alpha(\varphi)-2\beta(\varphi)$$

as is clear also from the fact that this intensity must be equal to the intensity of the radiation scattered along z polarized along y due to radiation polarized along x. We may thus use Eqs. (162), (164), (164'), (166) provided $\alpha(\varphi)$, $\sum_\varphi\alpha(\varphi)$ are replaced by [see Eq. (165)]

$$\bar{\alpha}(\varphi)=2\alpha(\varphi)-2\beta(\varphi).$$

For zero magnetic field the polarization due to unpolarized incident radiation is found from the above to be

$$\bar{P}_0=P_0/(2-P_0)$$

where P_0 is given by (165'). By (164') $P_0(\varphi)$ contains α only in the denominator. Therefore $P_0(\varphi)$ is diminished in the same proportion as P_0. Thus the *modification* of (164), (166) for unpolarized incident radiation *consists* simply *in multiplying* the *values* given by these formulas *by* $1/(2-P_0)$ where P_0 is the polarization due to polarized incident radiation in zero magnetic field.

Modified formula for fluorescence. The formulas derived above for resonance radiation are easily extended to fluorescence. We suppose the atom to be in state a having an inner quantum number j_a, to absorb radiation taking it to state b from which it falls to various states c. The levels a, b, c are supposed to be split into hyperfine levels with fine quantum numbers f, φ, h. We are interested in the polarization of the line due to the fall from b to c. Eq. (159) applies to this case as well provided m is restricted to lie in a and m' in c. This restriction must be made for m because Γ_I of (157) is small compared with $|\nu_{ac}|$ and because we are interested only in scattered radiation near the frequency ν_{bc}. Performing calculations exactly similar to those above it is seen that (163) is changed only to the extent of replacing f by h in the last two factors in the Sp. Similarly in (164'') f' is to be replaced by h. Eqs. (165) go into

[39] L. Larrick, Phys. Rev. **40**, 1041A (1932).

$$\alpha(\varphi)=(C/4g_a)\{\sum_\mu A_\pi^{\varphi\mu}(a)A_\pi^{\varphi\mu}(c)+(2\varphi+1)A(bc)A(ba)/3\},$$
$$2\beta(\varphi)=(C/4g_a)\{3\sum_\mu A_\pi^{\varphi\mu}(a)A_\pi^{\varphi\mu}(c)-(2\varphi+1)A(bc)A(ba)/3\}$$

where $A_\pi^{\varphi\mu}(a)$, $A_\pi^{\varphi\mu}(c)$ are the transition probabilities from the magnetic sublevels μ of φ due to π components to all magnetic sublevels in a and c respectively while $A(ba)$, $A(bc)$ are similar transition probabilities due to both π and σ components from b to a and c respectively. With these changes (164′), (166) still apply. In all of the above formulas the factor $1/g_a$ is present so as to give the scattered radiation per atom of scattering substance. If one deals with a mixture of isotopes the contributions of α and β (i.e., the intensities) due to the different isotopes are proportional to their abundance and are obtained by first multiplying the expressions derived above for each isotope by its abundance and then summing the results. Calculations using this theory have been made for λ2537 of Hg by A. C. G. Mitchell to whom the writer is very grateful for the permission to reproduce the graphical comparison between theory and experiment given below.

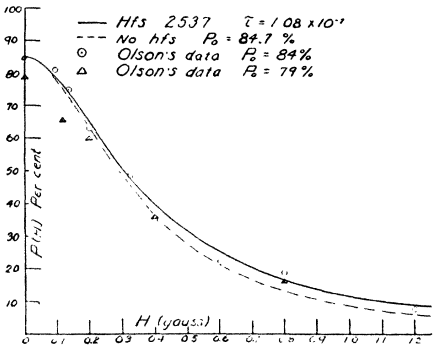

FIG. 6.

Narrow incident lines. It has been supposed above that the intensity of the incident radiation is sensibly uniform throughout the range of all possible transition frequencies between the sublevels of the levels a and b. This assumption has been introduced in passing from (157) to (157′) by making Γ_I large. The integration over ν_s involved in passing to (159) can be performed without this approximation and leads then to an expression similar to (159). The factor $1/(1-2\pi i\tau\nu_{\mu'\mu})$ is now multiplied by some constants and also by

$$i/[\nu(\mu' m_0)-\nu_I+i(\Gamma+\Gamma_I)]$$
$$+i/[\nu_I-\nu(\mu m_0)+i(\Gamma+\Gamma_I)].$$

Let Γ_I be large compared with Γ and yet small compared with the distance between two hyperfine structure components. The above factor is then appreciable only for that *hfs* component for which the absorption frequency $\nu(\mu m_0)\cong\nu_I$. The two terms combine under these conditions to give a resonance factor $1/[(\nu(\mu m_0)-\nu_I)^2+\Gamma_I^2]$ which is proportional to the intensity of the incident light per unit frequency range at the absorption frequency $\nu(\mu m_0)$. Thus for any mixture of wave trains having $\Gamma_I>\Gamma$ we can simply modify Eq. (162) by including in (163) (164″) a factor proportional to the intensity of incident radiation at the frequency $\nu_{\varphi f}$. This applies also if $\Gamma_I\sim\Gamma$ provided the incident line is broad compared to Γ. In fact in this case we must integrate over ν_I and we are again left essentially with the intensity at the frequency $\nu_{\varphi f}$. In practice incident lines are broad in comparison with the natural breadth and further refinements are not necessary.

§5. The Compton effect and Klein's method; the two-body problem

One of the most striking demonstrations of the unidirectional nature of light quanta is the Compton effect. As is well known the wave-length of x-rays scattered by free electrons initially at rest is increased on scattering. This change of wave-length is called the Compton effect. The x-ray quantum imparts to the electron a certain momentum and, therefore, also a certain amount of energy. The amount of energy available for the scattered quantum is smaller than that of the incident. The wave-length shift is derived without difficulty from the laws of conservation of energy and of momentum when applied to this collision process. As is well known, the wave-length shift is

$$\Delta\lambda=(h/mc)(1-\cos\Theta),$$

where Θ is the angle made by the direction of scattering with the direction of the incident radiation.

The theoretical justification of applying the laws of conservation of energy and momentum to this collision process is essentially the same as that used above [VII, §3] in the discussion of the unidirectional emission of light quanta. The point in the calculation at which the conservation of momentum makes its appearance is

$$(e^{-i\mathbf{k}_s\mathbf{R}})_{\mathbf{P},\,\mathbf{P}'} = \delta(\mathbf{P}, \mathbf{P}' - h\mathbf{k}_s/2\pi). \qquad (140)$$

In the discussion of the Compton effect we let $\mathbf{R} = (X, Y, Z)$ be the coordinates of the electron and $\mathbf{P} = (P_x, P_y, P_z)$ its components of momentum. $\mathbf{R}$ occurs in the interaction energy only as $e^{\mp i\mathbf{k}_s\mathbf{R}}$ the $-$ sign occurring with the radiation variable a_s and the $+$ sign with a_s^*. One has as a result a selection principle according to which there can be transitions only between states with exactly equal total momenta provided each light quantum is interpreted as having a momentum $h\nu/c$ in the direction of its propagation. [See Eq. (141).] The point at which conservation of energy is brought in is in the fact that the problem is treated by means of a Hamiltonian function. The wave-length shift is, thus, in agreement with Dirac's theory of light quanta inasmuch as it is a consequence of the laws of conservation.

The intensity of the scattered radiation can be also derived from the theory of light quanta. The formula believed in at present is that of Klein and Nishina.[40] It is derived using Dirac's relativistic equation for the electron. Previously to that Dirac[41] and Gordon derived a somewhat simpler formula by means of Schroedinger's quadratic relativistic equation without spin. The method of calculation in both papers is that of Klein[27] and is closely allied to the treatments of optical dispersion by Born, Heisenberg, Jordan and by Schroedinger presented in IV above. The relativistic electron equation of Dirac gives better results for most problems than Schroedinger's quadratic equation and the formula of Klein-Nishina is, thus, to be preferred.

The justification for using the correspondence method in the discussion of intensity relations in the Compton effect has been given by Dirac[42] and by Waller.[43] Dirac makes use of the general validity of Einstein's A/B relation [V, §6 above]. Waller performs a direct calculation using Dirac's theory of light quanta from the beginning. The calculations necessary for a presentation of the connection of the theory of light quanta with the correspondence method have been performed by us in V, §7 above. We explain only the case of the Klein-Nishina formula, since it is probably the more correct formula and since its theoretical discussion is simpler than that of Schroedinger's quadratic equation.

By using Dirac's relativistic electron equation the interaction energy between radiation and matter is

$$H' = \boldsymbol{\alpha} e\mathbf{A}, \qquad (168)$$

where the electronic charge is e, the vector formed by the three Dirac matrices α_1, α_2, α_3 is called $\boldsymbol{\alpha}$ and $\mathbf{A}$ as usual is the vector potential. In order to emphasize the connection with the nonrelativistic discussion of V above we use

$$c\boldsymbol{\alpha} = -\mathfrak{k}, \quad H' = -(e/c)\mathfrak{k}\mathbf{A}. \qquad (168')$$

The calculation for scattering is now practically the same as that of V, §7 above. Only the terms in $\beta^{\mathbf{k}',\mathbf{k}''}_{n',n''}$ are now absent. Thus, Eq. (118') applies omitting the term in β. Remembering the definition of α_{nm}^s given by (109) and making the distribution function D infinitely concentrated, we have instead of (118')

$$(\alpha^{s'})_{\text{eff.}} = \frac{e^2}{2\pi h}\left(\frac{N_{s''}}{V\nu(s'')}\right)^{\frac{1}{2}} \sum_{n'} \left\{ \frac{(\mathfrak{k}e^{-i\mathbf{k}'\mathbf{r}}\mathbf{f}')_{n_2n'}(\mathfrak{k}e^{-i\mathbf{k}''\mathbf{r}}\mathbf{f}'')^*_{n_1n'}}{\nu(n'n)_1 - \nu''} + \frac{(\mathfrak{k}e^{-i\mathbf{k}'\mathbf{r}}\mathbf{f}')_{n'n_1}(\mathfrak{k}e^{-i\mathbf{k}''\mathbf{r}}\mathbf{f}'')^*_{n'n_2}}{\nu' - \nu(n_1n')} \right\}, \qquad (169)$$

where the incident radiation is referred to by $\mathbf{f}''$, $\mathbf{k}''$, ν'', the scattered by $\mathbf{f}'$, $\mathbf{k}'$, ν', the initial state of matter by n_1, the final by n_2 and n' refers to the intermediate states. Eq. (169) gives the effective

[40] O. Klein and Y. Nishina, Zeits. f. Physik **52**, 853 (1929); Y. Nishina, Zeits. f. Physik **52**, 869 (1929).

[41] P. A. M. Dirac, Proc. Roy. Soc. **A111**, 405 (1926); W. Gordon, Zeits. f. Physik **40**, 117 (1927). For connection with correspondence principle: G. Breit, Phys. Rev. **27**, 362 (1926); Amer. Phys. Soc. Proc., December, 1925.

[42] P. A. M. Dirac, Proc. Cam. Phil. Soc. **26**, 361 (1930).

[43] I. Waller, Zeits. f. Physik **52**, 75 (1929).

value of [see Eqs. (109), (111), (114), (118)]

$$(\mathfrak{t}e^{-i\mathbf{k}'\mathbf{r}}\mathbf{f}')_{n_1n_2}^{\text{eff.}}=((2\pi h)^{\frac{1}{2}}/e)(\alpha^{s'})_{\text{eff.}}, \tag{170}$$

which should be used in emission formulas such as (111), (114) in order to obtain the probability of scattering a light quantum s'.

In order to see the relation with the classical theory, consider the equation immediately preceding Eq. (111) from which the latter was derived:

$$\left(\frac{d}{2\pi i dt}+\nu_s\right)a_{ns}e^{2\pi i\nu(n)t}=\frac{\alpha^s{}_{nn_0}}{(V\nu_s)^{\frac{1}{2}}}e^{2\pi i\nu(nn_0)t}=\frac{e}{(2\pi hV\nu_s)^{\frac{1}{2}}}(\mathfrak{t}e^{-i\mathbf{k}\mathbf{r}}\mathbf{f}_s)_{nn_0}e^{2\pi i\nu(nn_0)t} \tag{171a}$$

and compare this with Eqs. (18) of classical electrodynamics making use of (9):

$$\left(\frac{d}{2\pi i dt}+\nu_s\right)a_s^*=\frac{e}{2\pi(V\nu_s)^{\frac{1}{2}}}\int\mathbf{f}_s\mathbf{j}e^{-i\mathbf{k}\mathbf{r}}dV \qquad \text{(171b) (classical)}$$

Eqs. (171a), (171b) are very similar to each other. In (171a) the probability amplitude a_{ns} is determined by its initial value and by the matrix element standing on the right; in (171b) the radiation variable a_s^* is similarly determined by its initial value and the integral on the right. The similarity suggests that the a_{ns} are identical with the a_s^* for a proper classical radiation field. It must be remembered, however, that in (171b) $e\mathbf{j}$ is a current of classical electrodynamics and is, therefore, necessarily real, while the quantity which corresponds to it in (171a) is $u_n^*\mathfrak{t}u_{n_0}\times e^{2\pi i\nu(nn_0)t}$ and is essentially complex. We let, therefore,

$$\begin{aligned}\mathbf{j}&=\mathbf{J}e^{2\pi i\nu(nn_0)t}+\mathbf{J}^*e^{-2\pi i\nu(nn_0)t};\\ \mathbf{J}&=u_n^*\mathfrak{t}u_{n_0}e^{2\pi i\nu(nn_0)t}\end{aligned} \tag{172}$$

and we substitute this into (171b) and the corresponding equation for a_s. These equations determine a_s, a_s^* as:

$$\begin{aligned}a_s&=a_s(\mathbf{J}e^{2\pi i\nu(nn_0)t})+a_s(\mathbf{J}^*e^{-2\pi i\nu(nn_0)t}),\\ a_s^*&=a_s^*(\mathbf{J}e^{2\pi i\nu(nn_0)t})+a_s^*(\mathbf{J}^*e^{-2\pi i\nu(nn_0)t}),\end{aligned} \tag{172'}$$

where by $a_s^*(\mathbf{J}e^{i\omega t})$ is meant the result of solving (171b) with $\mathbf{J}e^{i\omega t}$ substituted for j. Thus, in (172') $a_s(\mathbf{J}e^{2\pi i\nu(nn_0)t})$, $a_s^*(\mathbf{J}e^{2\pi i\nu(nn_0)t})$ are not the complex conjugates of each other but are simply functional symbols. The left sides of the two Eqs. (172') are, nevertheless, just as usual the complex conjugates of each other because j as defined by (172) is real.

If $\nu(nn_0)<0$ and if initially $a_s(\ )$, $a_s^*(\ )$ are all zero, the only appreciable values of the functions in (172') are those of

$$a_s^*(\mathbf{J}e^{2\pi i\nu(nn_0)t}),\quad a_s(\mathbf{J}^*e^{-2\pi i\nu(nn_0)t})$$

as is seen by comparison with Eqs. (111), (114). [These two quantities are also complex conjugates.] Thus, the only part of j which is of importance in (171b) is that containing J. We let, therefore,

$$\begin{aligned}\mathbf{J}e^{2\pi i\nu(nn_0)t}&=u_n^*\mathfrak{t}u_{n_0}e^{2\pi i\nu(nn_0)t},\\ a_s^*&=(h/2\pi)^{\frac{1}{2}}a_{ns}e^{2\pi i\nu(n)t}\end{aligned} \tag{173}$$

and we see that the distribution of energy through the spectrum and among various space directions for the classical radiation caused by (172) is the same as the probable distribution in the quantum problem. For the classical energy is $2\pi\nu_s|a_s|^2$ which by (173) is the same as the expectation of the quantum energy $h\nu_s|a_{ns}|^2$.

By means of (172) we can picture the emission of radiation as due to a classical current density. What has been proved so far is only that the classical electromagnetic field emitted by j has the same energy distribution among frequencies and space directions as the expectation of the energy distribution has for the quantum problem. It has already been seen VII, §3 that in some cases the equality of a_{ns} and of a_s^* leads also to a similarity of quantum and classical theories so far as location in space is concerned. It is hardly necessary to elaborate this point. It will suffice to remember that $(\mathfrak{f}\mathfrak{E})^2$ of the classical theory is analogous to the expectation of this quantity in quantum mechanics and that the behavior of the

classical quantity is quite similar to that of its quantum analogue.

The validity of considering the radiation as due to the classical current density (172) is closely related to the *correspondence principle* discussion [see II above]. Einstein's transition probability from the state J' to the state J'' was supposed to be given by Eq. (47) in which

$$2e\tau_{J'J''} = \bar{C}_\tau(J)^{(J'J'')},$$

while the polarization was expressed as

$$\tfrac{1}{2}(C_\tau e^{2\pi i(\tau\nu)t} + C_\tau^* e^{-2\pi i(\tau\nu)t}).$$

It is clear that the current density (172) gives the same rate of radiation as (47) if the atomic dimensions are small in comparison with the wave-length. We may, thus, regard (172) as the generalization of (47) to the case of no restriction on atomic dimensions.

So far we have explained the justification of the treatment of emission by means of the classical current given by (172) in terms of the theory of light quanta. A classical current density similar to (172) may be used for the discussion of scattering. There is in fact not very much difference between spontaneous emission and scattering if one regards the incident light wave and the scattering electron as one coupled system. This point of view has been elaborated in detail in Section IV of this report. It has been shown there that it is legitimate to use the explicit time dependence of the perturbing potential and that one arrives in this manner at the same results as follow from a consideration of the coupled system. The discussion of Section IV did not include the retardation of potentials and is for this reason not altogether sufficient for our present purpose. Including the retardation effects we consider the electron under the action of an electromagnetic wave having a vector potential

$$\mathbf{A} = \mathbf{f}(be^{-i\mathbf{kr}+2\pi i\nu t} + b^* e^{i\mathbf{kr}-2\pi i\nu t}). \quad (174)$$

Let the interaction energy be given by (168), (168′). Under the action of the light wave an atomic state n_0 is changed into $\sum c_n{}^{n_0} u_n$ where

$$c_{n_0}{}^{n_0} = e^{-2\pi i\nu(n_0)t},$$

$$c_n{}^{n_0} = \frac{e}{hc}\left\{b(\dot{t}e^{-i\mathbf{kr}}\mathbf{f})_{nn_0}\frac{e^{2\pi i(\nu-\nu(n_0)t}}{\nu+\nu(nn_0)} + b^*(\dot{t}e^{i\mathbf{kr}}\mathbf{f})_{nn_0}\frac{e^{-2\pi i(\nu+\nu(n_0))t}}{\nu(nn_0)-\nu}\right\}.$$

The current density $eu_{n_1}{}^*\dot{t}u_{n_2}e^{2\pi i\nu(n_1n_2)t}$ which determines the probability of a jump from n_1 to n_2 is changed into

$$eu_{n_1}{}^*\dot{t}u_{n_2}e^{2\pi i\nu(n_1n_2)t} + \sum_{n'}\frac{e^2}{hc}\left\{\frac{(\dot{t}e^{-i\mathbf{kr}}\mathbf{f})_{n_1n'}}{\nu(n'n_1)-\nu}u_{n'}{}^*\dot{t}u_{n_2} + \frac{(\dot{t}e^{-i\mathbf{kr}}\mathbf{f})_{n'n_2}}{\nu+\nu(n'n_2)}u_{n_1}{}^*\dot{t}u_{n'}\right\}be^{2\pi i(\nu(n_1n_2)+\nu)t}$$
$$+\sum_{n'}\frac{e^2}{hc}\left\{\frac{(\dot{t}e^{i\mathbf{kr}}\mathbf{f})_{n_1n'}}{\nu+\nu(n'n_1)}u_{n'}{}^*\dot{t}u_{n_2} + \frac{(\dot{t}e^{i\mathbf{kr}}\mathbf{f})_{n'n_2}}{\nu(n'n_2)-\nu}u_{n_1}{}^*\dot{t}u_{n'}\right\}b^*e^{2\pi i(\nu(n_1n_2)-\nu)t}. \quad (175)$$

This expression will now be used in place of $e\mathbf{J}e^{2\pi i\nu(nn_0)t}$ of (172) and the effective classical current density $e\mathbf{j}$ will be given again by (172). It is maintained that this procedure is equivalent to the calculation by means of the theory of light quanta which led to (169).

In fact, we see that the part of $\mathbf{J}$ [see Eq. (175)] which contains the frequency $\nu(n_1n_2)+\nu$ is very closely related to the complex conjugate of the expression for "(α^s) eff." given by (169). [It should be remembered that in (169) the incident frequency ν was denoted by ν''.] Using (175) we compute without difficulty "(α^s) eff." by means of (170). The expression in { } involving matrix elements is obviously identical with the complex conjugate of the corresponding expression in (169). The factors by which the sum is multiplied are also readily seen to be identical, provided we let

$$b = ca_{s''}/(V\nu'')^{\frac{1}{2}}; \quad |a_{s''}|^2 = N_{s''}h/2\pi$$

in accordance with (1). The factor for $(et\dot{e}^{-i\mathbf{k}'\mathbf{r}}\mathbf{f})_{n_1n_2}{}^{\text{eff.}}$ is then by (170)

$$(e^2/(2\pi h)^{\frac{1}{2}})(N_{s''}/V\nu'')^{\frac{1}{2}} = (e^2/hc)|b|$$

in agreement with (175). We have not followed

through the analogy of the classical phases occurring in a_s, a_s^* and the quantum phases which occur in the probability amplitudes. This may also be readily done.

The use of the classical current density (172) with $\mathbf{J}$ given by (175) for the calculation of the intensity of scattered radiation is the correspondence method of Klein. In applications of the method it is not always necessary to use Eq. (175) for in some cases the perturbation of the proper functions may be computed more conveniently in other ways. The only essential point in (175) is the representation of $e\mathbf{J}$ as a sum of terms involving definite frequencies. Klein's method was formulated before the invention of the theory of light quanta. It may be looked at either as an *a priori* hypothesis or else one may prefer to justify it by means of the theory of light quanta as we have done here. The latter way has the appearance of being the better one, since it makes it possible to treat radiation and matter as a coupled dynamical system. It should be borne in mind, however, that the theory of light quanta has not gone so far much beyond the justification of the correspondence method and that it cannot claim to be logically consistent on account of the well-known divergence difficulties. Klein's formulation appears at the present time, therefore, as a particularly clear way of stating our knowledge about the probabilities of spontaneous emission and of scattering. Although it does not aspire to the same degree of finish as the theory of light quanta it is highly recommended by its simplicity and is likely to be correct quite independently of the theory of light quanta. In practical applications it has the additional advantages of avoiding too complicated calculations and of enabling one to visualize the phenomena in terms of charge and current distributions.

The result of the calculations of Klein and Nishina is that at a distance r from the scattering electron the intensity of radiation scattered in a direction making an angle Θ with the incident beam and an angle θ with the incident electric intensity is

$$I = I_0 \frac{e^4}{m^2c^4r^2} \frac{\sin^2\theta}{(1+\alpha(1-\cos\Theta))^3}\left[1+\alpha^2\frac{(1-\cos\Theta)^2}{2\sin^2\theta(1+\alpha(1-\cos\Theta))}\right]. \tag{176}$$

For unpolarized incident radiation:

$$I = I_0 \frac{e^4}{2m^2c^4r^2} \frac{1+\cos^2\Theta}{(1+\alpha(1-\cos\Theta))^3}\left[1+\alpha^2\frac{(1-\cos\Theta)^2}{(1+\cos^2\Theta)(1+\alpha(1-\cos\Theta))}\right], \tag{176'}$$

where

$$\alpha = h\nu/mc^2 = \Lambda/\lambda \tag{176''}$$

with the Compton wave-length denoted by

$$\Lambda = h/mc. \tag{176'''}$$

For small values of α, i.e., for long wave-lengths Eqs. (176), (176') are the same as follow from the theory of scattering according to classical electrodynamics as given by J. J. Thomson. The factors in front of [ ] are derivable according to Dirac and Gordon from the quadratic equation of Schroedinger. The [ ] represents the modification due to Dirac's relativistic linear equation as compared with Schroedinger's quadratic equation. Qualitatively, the important difference between the formulas of Klein and Nishina and those of Thomson is that the scattering takes place more intensely in the direction of the incident radiation. If $\Theta = 0$, the intensity of scattering is the classical intensity on any theory because for scattering in this direction the electron is at rest both before and after scattering.

Present experimental evidence speaks in favor of the validity of the Klein-Nishina formula.[44] In the experiments only light elements seem to scatter hard γ-radiation according to this formula. For heavier elements the nuclei presumably take part in the scattering[45] giving an additional effect and the photoelectric effect is also responsible for a part of the absorption.[46]

[44] G. T. Tarrant, Proc. Roy. Soc. **A128**, 345 (1930); C. Y. Chao, Proc. Nat. Acad. **16**, 431 (1930); L. Meitner and H. Hupfeld, Zeits. f. Physik **67**, 147 (1931); J. Jacobsen, Zeits. f. Physik **70**, 145 (1931); Gray, Proc. Roy. Soc. **A130**, 524 (1931).

[45] W. Heisenberg, in press.

[46] Harvey Hall and J. R. Oppenheimer, Phys. Rev. **38**, 57 (1931); F. Sauter, Ann. d. Physik **11**, 454 (1931); H. R. Hulme, Proc. Roy. Soc. **A133**, 381 (1931).

There appears to be no serious objection to the use of Dirac's linear equation in the derivation of the scattering formulas, although it must be conceded that the Klein paradoxon makes the universal validity of Dirac's equation questionable. It would appear, however, that a more serious limitation must be met with when the wave-length of the incident light is very short. In particular, one would be inclined toward special caution when the wave-length is comparable with or smaller than $a=e^2/2mc^2$ which is approximately the electronic radius demanded by the classical theory of electromagnetic mass. It is true, of course, that in the present quantum theory the electron is supposed to be a point charge and no direct meaning is assigned to the electronic radius of classical electricity and magnetism. Nevertheless, one would hardly expect quantum theory to be valid when applied to such small dimensions because the experimental facts which served as guides and tests for the formulation of the theory have to do with phenomena on a larger scale. The application of quantum theory to dimensions of the order of magnitude of a would at best appear as a speculative extrapolation of knowledge gained from larger scale phenomena. There appears to be no reason to believe in the validity of such an extrapolation and, in fact, there is strong experimental evidence against it. For it is well known that nuclear structure cannot be understood in terms of ordinary quantum mechanics. According to Heisenberg[47] an electron cannot exist in the nucleus and have anything like the experimentally observed energy. The thing in the way is the very small dimension of the nucleus. In his recent theory Heisenberg supposes, in fact, that the nucleus is made up of protons and neutrons and, thus, throws the deviations from quantum theory into the existence of Chadwick's neutron. It is certain, therefore, that quantum mechanics cannot be applied to the description of phenomena concerned with differences of properties of points separated by distances of the order of 10^{-13} cm and at first sight one would be inclined to mistrust the Klein-Nishina formula for wave-lengths of the incident radiation of this order of magnitude.

[47] W. Heisenberg, Zeits. f. Physik **77**, 1 (1932).

An important consideration of this point has been given by Bohr at the 1931 congress in Rome. According to Bohr,[48] one should expect that the Klein-Nishina formula is nevertheless correct for still shorter wave-lengths when measured in a frame for which the electron is initially at rest even though at distances of the order of 10^{-13} cm quantum theory is known to fail. Bohr's argument is that special relativity would at all events be expected to be applicable to the phenomena under consideration. By choosing a frame of reference moving with an appropriate velocity one is able to change the wave-length of the incident radiation at will on account of the Doppler effect. Simply requiring that the wave-length of the incident radiation should be larger than a certain amount therefore does not have very much meaning because by choosing a proper frame of reference one can change this wave-length at will. On the other hand, there exists a unique choice of a frame of reference in which the wave-lengths of the incident and scattered radiations are the same. This is the frame moving with such a velocity that the initial momentum of the electron is equal and opposite to the momentum of the incident light quantum. On scattering, the momenta are still equal and opposite, and on account of the conservation of energy the absolute value of either momentum is unchanged on scattering.[49] For this reference system the wave-length has a unique and simple meaning because it is the same before and after scattering while for a frame in which the electron is initially at rest the wave-length is usually much longer after scattering so that from the point of view of the scattered radiation the restriction on the wave-length would at all events be expected to be less stringent.

According to Bohr, the limitation on the shortness of wave-length should be stated for this particular reference system in which the wave-length is unchanged by scattering, and in this

[48] The writer became acquainted with these considerations through Professor Heisenberg to whom he is very much indebted also for an interesting discussion of this matter.

[49] This frame of reference has been used with great success by Pauli in a discussion of the equilibrium between black-body radiation and free electrons. W. Pauli, Zeits. f. Physik **18**, 272 (1923).

reference frame it is expected that when the wave-length becomes so small as to be comparable with $a=e^2/2mc^2$ the Klein-Nishina formula will fail.

Let the wave-length and the frequency of the incident radiation in the frame of reference at rest with respect to the initial state of the electron be λ, ν, respectively, and let the corresponding quantities in the frame of equal and opposite initial momenta be λ', ν'. The relative velocity of the two frames of reference is given by

$$v=h\nu/[mc+(h\nu/c)]$$

and

$$\nu'=\nu\left(\frac{1-v/c}{1+v/c}\right)^{\frac{1}{2}}=\frac{\nu}{(1+2h\nu/mc^2)^{\frac{1}{2}}}.$$

For high values of ν

$$\nu'=(\nu mc^2/2h)^{\frac{1}{2}};\quad \lambda'=(2\Lambda\lambda)^{\frac{1}{2}}. \tag{177}$$

The wave-length which has absolute significance for Bohr's argument is λ', and it is the geometric mean of twice the Compton wave-length and the wave-length λ. When $\lambda\ll\Lambda$, λ' is appreciably greater than λ so that λ' may be greater and λ less than a. Numerically, the difference is very large. Let us require that

$$\lambda'\geqq a$$

Then by (177)

$$\lambda\geqq\frac{a^2}{2\Lambda}=\frac{e^2}{2hc}a=\frac{a}{4\pi''137''}\leqq\frac{a}{1730}. \tag{178}$$

The Klein-Nishina formula may be, thus, expected to fail when λ has the value given by (178), i.e., when the energy of the incident light quantum referred to the electron initially at rest is

$$c/\lambda\leqq 1730\ hc/a\leqq(1730)^2mc^2\leqq 1.5\times10^{12}$$

electron-volts.

If, on the other hand, it is required that

$$\lambda=a.$$

The energy of the quantum is $hc/a\leqq 1730mc^2\leqq 8.7\times10^8$ electron-volts. Bohr's argument makes it, thus, probable that the Klein-Nishina formula is applicable for energies such as are at present considered in connection with cosmic radiation.[50] It should, of course, be regarded not as a proof of the validity of the K–N formula up to 10^{12} electron-volts but rather as a point of view. It is essential for Bohr's argument to consider the limitation as due only to the wave-length and not to be concerned with the velocity of the electron.

The interaction between two particles can also be treated according to quantum electrodynamics,[1] and it is possible[51] to explain by this means the interactions of the electron spins of two particles as well as the orbital and orbit spin interactions. It should be remembered, however, that the divergence difficulties of the theory make it impossible, according to Oppenheimer,[25] to arrive at a unique interpretation of the results. It is satisfactory to know that retardation of potentials applies to the interaction of two particles. Two limiting cases are fairly well understood. For particles moving with low velocities one may use an interaction energy

$$H'=e^2/r-(e^2/2)(\mathbf{a}^{\mathrm{I}}\mathbf{a}^{\mathrm{II}}/r+(\mathbf{a}^{\mathrm{I}}\mathbf{r})(\mathbf{a}^{\mathrm{II}}\mathbf{r})/r^3). \tag{179}$$

According to the derivation of Breit, one could only be sure of this form of the energy to the first order in e^2. According to Oppenheimer,[25] the first term e^2/r should have significance to much higher approximations. This comes about through the fact that the electrostatic field may be eliminated exactly, as has been explained in I and V above.

The second term involving Dirac's α's is of a more questionable type. There is no evidence that it is correct to higher than the first order in e^2 even though it is analogous to a similar term occurring in the classical Hamiltonian of Darwin.[52]

It is, on the other hand, possible to show a simple connection between the second term and Maxwell's equations quite independently of the details of the theory of light quanta.[53]

Thus, it follows from the equations

$$\mathcal{E}=-\partial\mathbf{A}/c\partial t+\mathcal{E}';\quad \mathcal{H}=rot\ \mathbf{A};\quad div\mathcal{E}'=4\pi\rho,$$
$$rot\ \mathcal{E}'=0$$

used in I and V above that neglecting acceler-

[50] W. Heisenberg, Ann. d. Physik **13**, 430 (1932).

[51] G. Breit, Phys. Rev. **34**, 553 (1929); see also: J. A. Gaunt, Phil. Trans. Roy. Soc. **228**, 151 (1929); Proc. Roy. Soc. **A122**, 153 (1929).

[52] C. G. Darwin, Phil. Mag. **39**, 537 (1920).

[53] G. Breit, Phys. Rev. **39**, 616 (1932).

ation and higher order effects the energy which must be added on account of the presence of transverse waves represented by the vector potential **A** is

$$\Delta E=-\frac{1}{4c^2}\int\left(\frac{\mathbf{j}_{P'}\mathbf{j}_{P''}}{r}+\frac{(\mathbf{j}_{P'}\mathbf{r})(\mathbf{j}_{P''}\mathbf{r})}{r^3}\right)dV_{P'}dV_{P''}, \tag{179'}$$

where the j's represent the electric current operators at points P', P'' and r is the distance between P' and P''. By the use of Dirac's equation, the self energy connected with ΔE may be shown to be an infinite constant in the same sense as this is the case [V, §3] for the electrostatic self energy. One feels rather certain from purely theoretical considerations, therefore, that the second term of (179) has a significance as an additive energy which must be considered in addition to the term value corresponding to the use of e^2/r alone. At the same time this term has no significance to higher orders of e^2. With this interpretation (179) is in agreement with experiment[54] leading essentially to the interaction energy set up by Heisenberg[55] by means of the spin model.

It will be noted that the use of the theory of light quanta is not essential to the understanding of (179') and that the largest claim which can be made for the theory is its consistency with experiment provided the divergence difficulties are treated in a proper way.

It followed already from the first formulation of the Heisenberg-Pauli theory of wave fields that retardation of potentials applied also to particles moving with any velocity. Thus, it was known[51] that to the first order in e^2 the interaction energy between two electrons can be represented by a formula exactly similar to the ordinary formulas using purely electrostatic interactions with the simple modification of having all exchange integrals replaced by

$$A_{st,\ ts}=\int(u_\rho^{*s}u_\rho^{t}\{1/r\}(u_\sigma^{*t}u_\sigma^{s})'-u_\rho^{*s}\alpha_{\rho\sigma}u_\sigma^{t}\{1/r\}(u_\mu^{*t}\alpha_{\mu\nu}u_\nu^{s})')dVdV', \tag{180}$$

where

$$\{1/r\}=(1/r)\cos 2\pi r/\lambda_{st};\qquad \lambda_{st}=ch/|E_s-E_t|.$$

Here s, t refer to two unperturbed electronic stationary states of energies E_s, E_t; the proper functions are u_ρ where ρ is Dirac's spin index; P, P' are two points in space; the elements of volume at P and P' are dV and dV', respectively, and, similarly, the charge and current indices with and without are taken at P' and P. By expanding the cosine in $\{1/r\}$ and using its first two terms one obtains a result in agreement with (179).

A simple derivation of (180) not involving the use of the theory of light quanta has been given by Rosenfeld.[56] The particles are described in their configuration space. Referring to one of them as I and to the other as II particle II is thought of first as subject to the field of I as though the latter were not perturbed by II. The "four potential" at $\mathfrak{r}_{II}$ due to the current and charge distribution of I is calculated using the formulas of classical electrodynamics. The unperturbed state of I is taken to be

$$\psi=\sum c_n^{I}u_n^{I},$$

where the u_n^{I} are eigenfunctions of states n of I not involving the time while each c_n^{I} involves the time in the form of $exp\ (-2\pi i\nu(n)t)$. The energy which must be added on account of the action of I and II is then found to be

$$\Delta E=e_Ie_{II}\sum_{n,\ m;\ r,\ s}c_n^{I*}c_m^{I}c_r^{II*}c_s^{II}B_{nm;\ rs}, \tag{181}$$

where

$$B_{nm;\ rs}=\int\int\{(u_n^{I*}u_m^{I})(u_r^{II*}u_s^{II})-(u_n^{I*}\alpha^{I}u_m^{I})(u_r^{II*}\alpha^{II}u_s^{II})\}e^{2\pi i\nu(mn)^{I}|\mathfrak{r}_I-\mathfrak{r}_{II}|/c}/|\mathfrak{r}_I-\mathfrak{r}_{II}|\,dV^{I}dV^{II}. \tag{181'}$$

[54] G. Breit, Phys. Rev. 36, 385 (1930).
[55] W. Heisenberg, Zeits. f. Physik 39, 499 (1926).
[56] L. Rosenfeld, Zeits. f. Physik 73, 253 (1931).

It should be noted that the above procedure implies the same time dependence of the wave functions of I as though II were not interacting with it. In this respect the theory is inaccurate. For equal particles we must deal with an antisymmetric wave function.

Let

$$\psi = 2^{-\frac{1}{2}}(\sum_n c_n^{\mathrm{I}} u_n^{\mathrm{I}} \sum_r c_r^{\mathrm{II}} u_r^{\mathrm{II}} - \sum_n c_n^{\mathrm{I}} u_n^{\mathrm{II}} \sum_r c_r^{\mathrm{II}} u_r^{\mathrm{I}}) = 2^{-\frac{1}{2}} \sum_{n,\, r} (c_n^{\mathrm{I}} c_r^{\mathrm{II}} - c_n^{\mathrm{II}} c_r^{\mathrm{I}})(u_n^{\mathrm{I}} u_r^{\mathrm{II}} - u_n^{\mathrm{II}} u_r^{\mathrm{I}}). \quad (182)$$

It is reasonable to expect that the procedure used in obtaining (181) will also apply for this antisymmetric ψ. We calculate the electromagnetic four potential due to II for the condition of the system in which I is at $\mathbf{r}_{\mathrm{I}}$. We obtain thus the addition to the energy due to the fact that I may be $\mathbf{r}_{\mathrm{I}}$, and we then average this result over $\mathbf{r}_{\mathrm{I}}$. We obtain

$$\Delta E = \tfrac{1}{2} \sum_{nmrs} c_n^{\mathrm{I}*} c_m^{\mathrm{I}} c_r^{\mathrm{II}*} c_s^{\mathrm{II}} (B_{n,\, m;\, r,\, s} + B_{r,\, s;\, n,\, m})$$

$$= \tfrac{1}{4} \sum_{nmrs} (c_n^{\mathrm{I}*} c_r^{\mathrm{II}*} - c_n^{\mathrm{II}*} c_r^{\mathrm{I}*})(c_m^{\mathrm{I}} c_s^{\mathrm{II}} - c_s^{\mathrm{I}} c_m^{\mathrm{II}})(B_{nm;\, rs} + B_{rs;\, nm}), \quad (183)$$

where the indices n, m, r, s take independently all their possible values and the symbol $B_{nm;\, rs}$ is defined by (181′) and is thus unsymmetrical with respect to an interchange of n, m, with r, s. The combinations of the B's occurring in (183) is such that n, m, may be interchanged with r, s so that (183) is symmetrical with respect to I and II. Obviously, we would have arrived at the same expression if we had taken I in the field of II. For the special case of a stationary state described by

$$(1/2^{\frac{1}{2}})(u_n^{\mathrm{I}} u_r^{\mathrm{II}} - u_n^{\mathrm{II}} u_r^{\mathrm{I}})$$

Eq. (182′) gives

$$\Delta E = B_{nn;\, rr} - \tfrac{1}{2}(B_{nr;\, rn} + B_{rn;\, nr}), \quad (183')$$

which is in agreement with (180). In presenting the above discussion of Rosenfeld we have deviated from the original by not using the method of quantized amplitudes which are known to be equivalent to the simple considerations in configuration space. Needless to say a similar treatment can be given for particles obeying Einstein-Bose statistics. The perturbation energy operator (183) is not expected to be valid except to the first order in e^2, because in the derivation the field of particle I acting on particle II was supposed to be unaffected by the interaction of the two particles.

The perturbation energies (181) and (183) may be used not only for calculations of energy changes of stationary states but also for the discussion of collisions between particles. The results are again expected to be valid only to the first order in the interaction energy and have, thus, significance for fast electrons. The method of treatment which enables one to treat the problem is due to Møller[57] who starts out in his discussion with Born's method for collision problems.

Møller's formula is essentially equivalent to (180) and is easily connected with (181). We let the proper functions u_n correspond to states of constant momentum. We may then regard $e_{\mathrm{I}} e_{\mathrm{II}} B_{nm;\, rs}$ as the matrix element of the perturbation energy in momentum space. Using the proper normalization factors h^{-3} for each particle [II, §3] and (181′) we have:

$$B_{nm;\, rs} = \int h^{-6} |\mathbf{r}_{\mathrm{I}} - \mathbf{r}_{\mathrm{II}}|^{-1} \exp\{(2\pi i/h)[(\mathbf{p}_n - \mathbf{p}_m)\mathbf{r}_{\mathrm{I}} + (\mathbf{p}_r - \mathbf{p}_s)\mathbf{r}_{\mathrm{II}}] + 2\pi i \nu(nm) |\mathbf{r}_{\mathrm{I}} - \mathbf{r}_{\mathrm{II}}|/c\}$$

$$\{(a_n^* a_m)(a_r^* a_s) - (a_n^* \boldsymbol{\alpha} a_m)(a_r^* \boldsymbol{\alpha} a_s)\} d V_{\mathrm{I}} d V_{\mathrm{II}},$$

where the a's involve Dirac's spin index so that

$$u_n = e^{2\pi i (\mathbf{p}_n \mathbf{r})/h} a_n.$$

Let

$$I = h^{-3} \int |\mathbf{r}_{\mathrm{I}} - \mathbf{r}_{\mathrm{II}}|^{-1} \exp\{(2\pi i/h)(\mathbf{p}_r - \mathbf{p}_s)\mathbf{r}_{\mathrm{II}} + 2\pi i \nu(nm) |\mathbf{r}_{\mathrm{I}} - \mathbf{r}_{\mathrm{II}}|/c\} d V_{\mathrm{II}}.$$

[57] C. Møller, Zeits. f. Physik 70, 786 (1931); H. Bethe, Zeits. f. Physik 76, 293 (1932).

Then

$$(\Delta_{\rm I}+4\pi^2\nu^2(nm)/c^2)I=-4\pi h^{-3}e^{(2\pi i/h)(\mathbf{p}_r-\mathbf{p}_s)\mathbf{r}_{\rm I}}.$$

A solution of this equation is

$$I=(h/\pi)e^{(2\pi i/h)(\mathbf{p}_r-\mathbf{p}_s)\mathbf{r}_{\rm I}}/[(\mathbf{p}_r-\mathbf{p}_s)^2-h^2\nu^2(nm)/c^2]$$

and it may be seen that this is the correct solution to use. Hence

$$B_{n,\,m;\,r,\,s}=\frac{1}{\pi h}\frac{(a_n^*a_m)(a_r^*a_s)-(a_n^*\boldsymbol{\alpha}a_m)(a_r\boldsymbol{\alpha}a_s)}{(\mathbf{p}_r-\mathbf{p}_s)^2-h^2\nu^2(nm)/c^2}\delta(\mathbf{p}_n+\mathbf{p}_r-\mathbf{p}_m-\mathbf{p}_s), \tag{184}$$

which is Møller's formula. Conservation of momentum is satisfied automatically by (184). The conservation of energy also follows in this method of treatment as a result of using a Hamiltonian. Eq. (184) is symmetrical in the two particle states because we may use not only $\mathbf{p}_r-\mathbf{p}_s=\mathbf{p}_m-\mathbf{p}_n$ but also $\nu(nm)=\nu(sr)$. A similar treatment can be given for unequal particles. Møller's result is of importance for the discussion of penetrating radiation.[58] A systematic derivation of Møller's formula from quantum-electrodynamics has recently been given by Bethe and Fermi.[59] The derivation is subject to the divergence difficulties of the theory. Disregarding these it shows that to within the first power in e^2 there is no probability of emitting light quanta.

These authors derive the interaction energy (179) from Møller's equation. This connection has been previously discussed in[51] where Eq. (180) was the starting point. Bethe and Fermi's discussion is somewhat more general not being confined to the treatment of identical particles. The essential point is again that $\cos(2\pi r/\lambda)$ when expanded according to powers of r/λ gives rise to correction terms to the electrostatic interaction. It should be remembered however that (179) applies very accurately in the region of small velocities while (180) is accurate only to the first order in e^2. It is thus impossible to really derive (179) from (180) but it is possible to demonstrate its plausibility.

Bethe and Fermi also derive both (179) and Møller's formula from the theory of light quanta. The authors obtain Møller's form by considering equations in the momentum space of the two interacting particles. The particles are supposed to have initially definite momenta and definite energies their interaction with the radiation field being neglected. This initial step presupposes that the electrostatic interaction between the particles may be considered as small for otherwise the energy and the momentum cannot be simultaneously specified. For the same reason the particles in question must be considered as free. The interaction with the radiation field is then introduced. If there were only one particle initially in an energy state n the initial state of the particle and the radiation field would be described by $\varphi(n;\ 0,\ 0,\ \cdots)$ where the zeros indicate that there are no light quanta. The interaction with the field will bring in also the states $\varphi(n';\ 0,\ \cdots 1_s,\ 0)$ in which the energy of the particle has been changed and a light quantum appeared. Such states may be said to describe to a first approximation the particle and its own electromagnetic field.

For two particles I, II the initial state is taken to be $\varphi(n_{\rm I},\ n_{\rm II};\ 0,\ 0,\ \cdots)=\varphi(n_{\rm I},\ n_{\rm II})$. The interaction with radiation brings about the states $\varphi(n_{\rm I}',\ n_{\rm II};\ 1_s)$ and $\varphi(n_{\rm I},\ n_{\rm II}';\ 1_s)$ in which the energy of one of the particles has been changed and a light quantum appeared. In this approximation we are dealing with two particles and their own fields. The interaction of the particles with each other has not yet been taken into account. It appears only in the next approximation. The state $\varphi(n_{\rm I}',\ n_{\rm II};\ 1_s)$ gives rise through the absorption of a light quantum to $\varphi(n_{\rm I}',\ n_{\rm II}',\ 0,\ 0,\ \cdots)$ and similarly the same state may arise from $\varphi(n_{\rm I},\ n_{\rm II}';\ 1_s)$. The simultaneous change of the energy of both particles without a change in the number of light quanta is thus represented in two successive steps, the first step being the emission of a quantum and a change of energy of one of the particles while in the second step the light quantum is absorbed and the

[58] W. Heisenberg, Ann. d. Physik **13**, 430 (1932).

[59] H. Bethe and E. Fermi, Zeits. f. Physik **77**, 296 (1932). See also B. Podolsky and V. A. Fock, Phys. Zeits d. Sow. **2**, 275 (1932).

second particle changes its state. On performing the calculation Bethe and Fermi make use of the fact that

$$(\mathfrak{a}_{\mathrm{I}}(\mathfrak{p}_{\mathrm{I}}'-\mathfrak{p}_{\mathrm{I}}))(\mathfrak{a}_{\mathrm{II}}(\mathfrak{p}_{\mathrm{II}}'-\mathfrak{p}_{\mathrm{II}}))-(E_{\mathrm{I}}'-E_{\mathrm{I}})^2/c^2=0$$

where $\mathfrak{p}$, E are the momentum and energy and the $'$ refers to quantities after collision. This is essentially the continuity equation for matter waves. The use of this continuity equation is common to all derivations of interaction energies from the theory of light quanta and is also involved in the derivation of (180). In order to obtain physically sensible results it has also been necessary to consider only such states n_{I}', n_{II}' for which the energy of the two particles together is equal to the sum of their energies in the states n_{I}, n_{II}. This is a weakness of the theory of light quanta and it is connected with the fact that the result is accurate only to the first power in e^2 as well as the supposition that spontaneous radiation of light quanta is not important.

It is instructive to consider the interaction energy in the six dimensional coordinate space of the two particles and to derive it from the theory of light quanta. The equations for the probability amplitudes are

$$\{(h/i)\partial/\partial t+E(n_{\mathrm{I}})+E(n_{\mathrm{II}})+\textstyle\sum_s N_s h\nu_s+e_{\mathrm{I}}e_{\mathrm{II}}/r+\sum_{n_{\mathrm{I}}',s}Ke_{\mathrm{I}}\nu_s^{-\frac{1}{2}}[(\alpha_{\mathrm{I}}^{\mathfrak{k}_s})_{n_{\mathrm{I}}n_{\mathrm{I}}'}N_s^{\frac{1}{2}}\Delta_s^-$$
$$+(\alpha_{\mathrm{I}}^{\mathfrak{k}_s})_{n_{\mathrm{I}}n_{\mathrm{I}}'}(N_s+1)^{\frac{1}{2}}\Delta_s^+]I(n_{\mathrm{I}}'/n_{\mathrm{I}})+\textstyle\sum_{n_{\mathrm{II}}',s}Ke_{\mathrm{II}}\nu_s^{-\frac{1}{2}}[(\alpha_{\mathrm{II}}^{\mathfrak{k}_s})_{n_{\mathrm{II}}n_{\mathrm{II}}'}N_s^{\frac{1}{2}}\Delta_s^-+$$
$$+(\alpha_{\mathrm{II}}^{\mathfrak{k}_s})_{n_{\mathrm{II}}n_{\mathrm{II}}'}(N_s+1)^{\frac{1}{2}}\Delta_s^+]I(n_{\mathrm{II}}'/n_{\mathrm{II}})\}\cdot\phi(n_{\mathrm{I}},n_{\mathrm{II}},N_1,\cdots,N_s,\cdots)=0$$

where

$$K=(hc^2/V)^{\frac{1}{2}},\quad (\alpha_{\mathrm{I}}^{\mathfrak{k}_s})_{n_{\mathrm{I}}n_{\mathrm{I}}'}=(\mathfrak{a}_{\mathrm{I}}\mathfrak{f}_s e^{-i\mathfrak{k}_s\mathfrak{r}_{\mathrm{I}}})_{n_{\mathrm{I}}n_{\mathrm{I}}'}$$

the operators Δ_s^-, Δ_s^+ lower and raise respectively N_s by 1 and $I(n'/n)$ converts n into n'. The approximation which involves the first power of K represents the particles with their own fields and is given by

$$\phi(n_{\mathrm{I}},n_{\mathrm{II}})=\phi^{(0)}(n_{\mathrm{I}},n_{\mathrm{II}})\exp[-2\pi i(\nu(n_{\mathrm{I}})+\nu(n_{\mathrm{II}}))t]$$

where ϕ^0 does not involve t, and by

$$\phi(m_{\mathrm{I}},n_{\mathrm{II}};1_s)=e_{\mathrm{I}}K(\alpha_{\mathrm{I}}^{\mathfrak{k}_s})_{m_{\mathrm{I}}n_{\mathrm{I}}}\phi(n_{\mathrm{I}}n_{\mathrm{II}})/h\nu_s^{\frac{1}{2}}(\nu(n_{\mathrm{I}}m_{\mathrm{I}})-\nu_s)$$

as well as similar expression for $\phi(n_{\mathrm{I}},m_{\mathrm{II}};1_s)$. When we omit a N_s we mean that $N_s=0$. The above solution represents a stationary state since the time dependence is represented by the same exponential for all ϕ's. Strictly speaking one must discuss the states before and after collision as such stationary solutions. This is however not necessary if we are interested only in such states for which $\phi(m_{\mathrm{I}},m_{\mathrm{II}})$ is $\gg\phi(m_{\mathrm{I}}',m_{\mathrm{II}}';1_s)$. The way in which the probability amplitude for any such state varies may be then taken to be the same as that for $\phi(m_{\mathrm{I}},m_{\mathrm{II}})$. From the Schroedinger equation and the first approximation we get

$$\{(h/i)\partial/\partial t+E(n_{\mathrm{I}})+E(n_{\mathrm{II}})\}\phi(n_{\mathrm{I}}',n_{\mathrm{II}}')$$
$$+e_{\mathrm{I}}e_{\mathrm{II}}\{(n_{\mathrm{I}}'n_{\mathrm{II}}'|r^{-1}|n_{\mathrm{I}}n_{\mathrm{II}})+S\}\phi(n_{\mathrm{I}}n_{\mathrm{II}})=0$$

where

$$S=\int\int u^*_{n_{\mathrm{I}}}u^*_{n_{\mathrm{II}}}Tu_{n_{\mathrm{I}}}u_{n_{\mathrm{II}}}dV_{\mathrm{I}}dV_{\mathrm{II}}$$

and

$$T=\sum_s\frac{c^2}{2\pi V}\left\{\frac{e^{i\mathfrak{k}\mathfrak{r}}}{\nu[\nu(n_{\mathrm{II}}n_{\mathrm{II}}')-\nu]}+\frac{e^{-i\mathfrak{k}\mathfrak{r}}}{\nu[\nu(n_{\mathrm{I}}n_{\mathrm{I}}')-\nu]}\right\}(\mathfrak{a}_{\mathrm{I}}\mathfrak{f}_s)(\mathfrak{a}_{\mathrm{II}}\mathfrak{f}_s).$$

Here $\mathfrak{r}=\mathfrak{r}_{\mathrm{I}}-\mathfrak{r}_{\mathrm{II}}$ and the indices s are omitted in $\mathfrak{k}$ and ν. The integration for S involves also a summation over the spin coordinates. Using the transverse polarization property of the electromagnetic waves we obtain

$$T=\textstyle\sum_{\mathfrak{k}}(c^2/\pi V)[(\mathfrak{a}_{\mathrm{I}}\mathfrak{a}_{\mathrm{II}})$$
$$-k^{-2}(\mathfrak{a}_{\mathrm{I}}\nabla_{\mathrm{I}})(\mathfrak{a}_{\mathrm{II}}\nabla_{\mathrm{II}})](\nu^2-\nu_0^2)^{-1}\cos\mathfrak{k}\mathfrak{r}$$

where we supposed conservation of energy to hold and have set $\nu_0=\nu(n_{\mathrm{II}}n_{\mathrm{II}}')=\nu(n_{\mathrm{I}}'n_{\mathrm{I}})$. The operators ∇_{I}, ∇_{II} are supposed to be applied only to the $\mathfrak{r}$ in the $\cos\mathfrak{k}\mathfrak{r}$ and not to the variables in functions u. Replacing the summation by an integration we get

$$T=-(\mathfrak{a}_{\mathrm{I}}\mathfrak{a}_{\mathrm{II}}/r)\cos k_0 r$$
$$-(\mathfrak{a}_{\mathrm{I}}\nabla_{\mathrm{I}})(\mathfrak{a}_{\mathrm{II}}\nabla_{\mathrm{II}})(1-\cos k_0 r)/k_0^2 r$$

where

$$k_0 = 2\pi\nu_0/c.$$

Substituting T into S the gradients can be transferred to the eigenfunctions by partial integration giving rise to

$$-\int (1-\cos rk_0) r^{-1} k_0^{-2} \nabla_{\rm I}(u_{\rm I}'^* \alpha_{\rm I} u_{\rm I}) \nabla_{\rm II}(u_{\rm II}'^* \alpha_{\rm II} u_{\rm II}) dV_{\rm I} dV_{\rm II}$$

summations over spin indices being indicated by ( ). By the continuity equation for matter we have

$$(E-E')(u'^* u) - (\hbar/i)\nabla(u'^* \alpha u) = 0$$

which gives

$$\nabla_{\rm I}(u_{\rm I}'^* \alpha_{\rm I} u_{\rm I}) \nabla_{\rm II}(u_{\rm II}'^* \alpha_{\rm II} u_{\rm II}) = k_0^2 (u_{\rm I}'^* u_{\rm I})(u_{\rm II}'^* u_{\rm II})$$

Hence

$$S = \int u_{\rm I}'^* u_{\rm II}'^* (1/r)\{-1+(1-\alpha_{\rm I}\alpha_{\rm II})\cos k_0 r\} u_{\rm I} u_{\rm II} dV_{\rm I} dV_{\rm II},$$

and is simply the matrix element of the quantity in $\{\ \}(1/r)$. Combining this with $1/r$ we obtain the matrix elements of the interaction energy as the matrix elements of

$$(e_{\rm I} e_{\rm II}/r)(1-\alpha_{\rm I}\alpha_{\rm II})\cos k_0 r.$$

It will be noted that the above written operator contains explicitly the unperturbed energies of the two particles [in k_0]. This is inherent to the treatment which takes into account retardation only inasmuch as the time dependence of the unperturbed wave functions of the system is known.

It will also be noted that the treatment is inapplicable to problems in which common energy differences do not exist for the two particles. Transitions for such systems involve not only the two particles but also the emission or absorption of light quanta. The part S of the interaction inergy is accurate only to the first power of e^2 while the additional term e^2/r has a much higher degree of accuracy for low velocities.

In the discussion of collisions between free particles Møller's method does not introduce transitions to states of negative energy on account of the simultaneous validity of the conservation of energy and momentum. This may be seen by considering the particles in the reference system of their center of gravity. We have then $\mathbf{p}_{\rm I} = -\mathbf{p}_{\rm II}$, $\mathbf{p}_{\rm I}' = -\mathbf{p}_{\rm II}'$ where $'$ denotes the quantity after collision. Hence

$$E_{\rm I}'^2 - E_{\rm I}^2 = c^2(p_{\rm I}'^2 - p_{\rm I}^2) = c^2(p_{\rm II}'^2 - p_{\rm II}^2) = E_{\rm II}'^2 - E_{\rm II}^2$$

and by the conservation of energy

$$E_{\rm I}' - E_{\rm I} = -(E_{\rm II}' - E_{\rm II}).$$

Hence either: (a) $E_{\rm I}' + E_{\rm I} = -E_{\rm II}' - E_{\rm II}$ which together with the conservation of energy gives $E_{\rm I} = -E_{\rm II}$, $E_{\rm I}' = -E_{\rm II}'$ or else (b) $E_{\rm I}' = E_{\rm I}$, $E_{\rm II}' = E_{\rm II}$. In the case (a) one of the particles has a positive and the other a negative energy both before and after collision. The total energy is zero. In case (b) the values of the energy do not change for either particle.

It will be seen from the above review of the work on the two electron problem that the theory of light quanta is not a very satisfactory tool for its discussion. Results can be obtained, but without additional physical considerations they are not unique. For this reason a formulation similar in point of view to that of Klein has been given by Heisenberg[60] for the treatment of the interaction of radiation with matter. In this paper Heisenberg uses quantum-electrodynamics but writes the equations in a form identical with that of classical electrodynamics. This is accomplished by the use of quantized wave amplitudes. It enables one to keep in mind the analogy to classical theory. He is able to show very elegantly the equivalence of the light quantum treatment to the visual representation of charge and current densities of Schroedinger and Klein. He also shows that radiation is travelling with the velocity of light directly from

[60] W. Heisenberg, Ann. d. Physik 9, 338 (1931); see also L. Rosenfeld, Zeits. f. Physik 71, 273 (1931).

Maxwell's equations. This method has the advantage of conciseness. Since, however, it is mathematically equivalent to the calculations which we presented above, we do not go into the details of Heisenberg's mathematical procedure. Instead we would like to emphasize Bohr's point that the applications of quantum-electrodynamics are made possible in this as well as in all other papers only by the supposition that the interaction between radiation and matter may be treated as small while according to quantum-electrodynamics it is infinite.

§6. Convergence difficulties

We have attempted above to present those parts of the theory of radiation for which the conclusions are certain and the results are likely to be physically correct. Before closing the report it is necessary to discuss briefly also the nature of the unsatisfactory features of the theory.

As is well known the theory of light quanta suffers from divergence difficulties. The infinities which arise in applications of the theory have the appearance of being of the following kinds: (a) those having to do with the zero-point energy of radiation in empty space; (b) those having to do with the finite size of the electron.

The first kind (a) is by itself not very troublesome because the zero-point energy $\sum_s h\nu_s/2$ is an additive constant in the total energy of the system. As has been explained in V above the order of factors in the Hamiltonian function can be chosen in such a way that this zero-point energy does not occur at all. Even though it may be, thus, formally removed, the peculiarities of the quantum treatment which are responsible for it are still present. We can illustrate the point by considering a single simple harmonic oscillator of frequency ν. It also has a zero-point energy and by subtracting from the Hamiltonian function $\pi\nu i(PQ-QP)$ we can remove this energy. Nevertheless, we still have a finite value of $\overline{Q^2}$ and of $\overline{P^2}$ for the normal state of the oscillator, and we see from the fact that the Schroedinger wave function is of the form $e^{-\alpha Q^2}$ that the oscillator is neither in its equilibrium position nor in a state of rest. This state of agitation for the normal modes of vibration of a crystal is, for example, demonstrated experimentally by a residual diffuseness of x-ray diffraction patterns produced by crystals even at low temperatures. We see, therefore, that, although the purely additive constant $\sum_s h\nu_s/2$ in the energy is not a real stumbling block of the theory, it is connected with a condition of the electromagnetic field in the absence of light quanta which may be described by saying that there is a state of residual agitation in the field. It is impossible to remove it just as it is impossible to find a stationary solution for the harmonic oscillator for which it can be considered as being in a state of rest. It is essentially for this reason combined with the postulated existence of an infinite number of states for any light quantum that the theory is intrinsically divergent quite apart from the question of the size of the electron. Thus, it has been shown by Rosenfeld[61] that even in the absence of matter the gravitational field due to light quanta is infinite. In this work it is supposed that the formalism of general relativity may be transferred to the discussion of the interaction of gravitational and matter waves. By taking into account terms in the first power of the gravitational constant it is found that the interaction between the gravitational and the light wave field gives no contribution to the expectation of the mutual energy and an infinite contribution to the expectation of the energy of the gravitational field. The origin of the infinity is traced to the occurrence of summations which extend over all possible frequencies and cover, therefore, also the range of infinitely high frequencies. It is, furthermore, pointed out that from the point of view of the configuration space of light quanta the divergence is attributable to the infinitesimal volume given to the light quantum. Needless to say there is a direct connection between these two aspects just as in the case of matter waves, and it is, furthermore, clear that the problem here considered is analogous to the discussion of the interaction of electrons with light quanta, with light quanta substituted for electrons and gravitational quanta for light quanta. The particular significance of this work lies in emphasizing the generality of the divergence difficulties when applied to any system of wave fields.

The divergence difficulties (b) having to do with the size of the electron have been already

[61] L. Rosenfeld, Zeits. f. Physik 69, 589 (1930).

partly discussed by us. We saw [I, §§5, 6] that classical electricity and magnetism is subject to the same defect, and [V, §3] we, further, convinced ourselves that the electrostatic part of the self energy behaves in quantum theory exactly in the same way as it does in the classical. This part of the self energy is not especially bothersome, for, as has been shown, it may be regarded as an additive constant characteristic of the distribution function $D(\mathbf{r}_i-\mathbf{r}')$. It will be recalled, however, that in the classical theory the infinite contribution to the energy of the electromagnetic field is due not to the electrostatic energy alone but also to the transverse waves; the purely electrostatic part is given by Eq. (28), while the whole energy is given by Eq. (27). Corresponding to this, there are also in the quantum theory infinite contributions due to the energy of the transverse waves. This has been shown by direct calculation by Oppenheimer[25] for stationary states and by Waller for free electrons.[62] It is especially important to note that the infinite contribution to the energy due to transverse waves cannot be considered as an additive constant. It has, in fact, been shown by Oppenheimer[25] that the difference between the self energies of two different electronic stationary states is, itself, infinite. The theory thus leads to an infinite displacement not only of spectral terms [which by itself could be harmless] but also to an infinite displacement of spectral lines and is, thus, in direct contradiction with experience. The displacements calculated by Waller[62] are, of course, of the same character. That the differences between the self energies of different spectral terms should be infinite is to be expected[25] by analogy with classical electricity and magnetism. The range of probable electronic velocities is different for different stationary states. According to the well-known expression

$$W=W_0[1+(\beta^2/3)]/[(1-\beta^2)^{\frac{1}{2}}] \qquad (27)$$

the difference $W(\beta_1)-W(\beta_2)$ becomes infinite for $W_0\to\infty$ which corresponds to a point electron.

Care should be taken, however, not to carry this analogy too far, because the particle picture is applicable only partially in quantum mechanics. As is well known, there exists a complimentary duality between waves and particles, and neither the wave nor the particle picture gives a universally appropriate description of nature.[63] This complimentarity of the wave and particle aspects of matter can be emphasized by means of the mathematical machinery of second quantization, i.e., of quantization of matter waves.[64] The point of special interest to us is that one may start either with a mathematical treatment in the configuration space of the particles or else one may start with a three-dimensional description in terms of matter waves. Dealing with the configuration space of the particles one is in a convenient position to see the analogy to the equations of motion of classical particle dynamics (Newton's laws of motion) and one sees this connection quite apart from any special form of the Hamiltonian function as has been emphasized in Part II of this report.

Connection between the particle picture of classical dynamics and the physical reality described by the equations of the quantum theory is of course given in the sense of Bohr's correspondence principle. The connection is put into the theory by arranging the quantum Hamiltonian so as to lead to the classical equations of motion for the quantum operators. Often this is done by making the quantum Hamiltonian identical with the classical one. In the mathematical formulation by means of quantized waves one starts with a Hamiltonian function analogous in form to a classical one describing the behavior of matter waves. The classical matter waves are to be thought of as smeared out in space and their interaction may be pictured as due to the mutual energy of the charge densities at different points in space. In this classical picture there is no indication of the discreteness of the number of particles, and there is also no indication of any

[62] I. Waller, Zeits. f. Physik **62**, 673 (1930).

[63] N. Bohr, Nature **121**, 580 (1928); see also W. Heisenberg, *The Physical Principles of the Quantum Theory*, University of Chicago Press, 1930.

[64] For a brief presentation of this mathematical method, see W. Heisenberg, *The Physical Principles of the Quantum Theory*, pp. 177–181, University of Chicago Press, 1930; see also: P. A. M. Dirac, Proc. Roy. Soc. **A114**, 243 (1927); P. Jordan and O. Klein, Zeits. f. Physik **45**, 751 (1927); P. Jordan, Zeits. f. Physik **45**, 766 (1927); **44**, 473 (1927); P. Jordan and E. Wigner, Zeits. f. Physik **47**, 631 (1928); V. Fock, Zeits. f. Physik **75**, 622 (1932); P. Jordan, Zeits. f. Physik **75**, 648 (1932).

particle size. It is these properties of the physical reality [the particle aspect] that are introduced by the second quantization, i.e., by the treatment of ψ^* and ψ as noncommuting quantities, as is shown by the fact that it may be proved mathematically that the quantized wave treatment is equivalent to the particle treatment by means of Schroedinger's equation in configuration space. Thus, by formulating the theory by means of quantized waves we have a convenient way of seeing the analogy to classical matter waves again in a sense *via* the correspondence principle.

Knowing the equivalence of the treatment in configuration space with that by means of quantized matter waves, we see that Bohr's principle of complimentarity finds here its proper mathematical formulation. We do not expect to have a complete description either by means of the particle or by means of the wave picture. It would also be unwarranted to expect the particle-aspect consideration made in connection with Eq. (27) to be wholly applicable. Neither should we consider the smeared-out charge distribution picture to be literally valid. According to the latter we would only expect a contribution to the magnetic energy of the order of the energy of the magnetic field due to the smeared-out current, and this energy would not be troublesome. However, the calculations of Oppenheimer, Waller and Rosenfeld show that the particle aspect plays an important part although the form of the mathematical expressions arrived at is not very reminiscent of the corresponding classical particle formulas. It, thus, comes about that the self energy due to the transverse waves is infinite, and yet it is not directly representable as Eq. (27).

It must be emphasized that the divergence difficulties are characteristic of the original Dirac theory of light quanta and that they are not due to the formalism introduced by the wave field theory of Heisenberg and Pauli. This is obvious from the fact that on eliminating the electrostatic field the wave field theory is equivalent to Dirac's theory of light quanta. It must, furthermore, be borne in mind that the use of Dirac's relativistic linear equation made in the work of Oppenheimer and Waller cannot be held responsible for the failure of the theory, because as has been shown by Rosenfeld[65] that the failure sets in even if one uses Schroedinger's non-relativistic wave equation. In this calculation Rosenfeld starts out with an initial state of the coupled system consisting of radiation and matter in an initial condition of having matter in a stationary state and no light quanta. He then calculates by the method of variation of constants the behavior of the system at later times. The matter is supposed to be a classical harmonic oscillator which is the direct opposite in its general properties to the free electron considered by Waller. He finds that with the lapse of time the expectation of the energy of the electromagnetic field becomes infinite. The infinity comes about mathematically through a summation over the possible frequencies of light quanta. He breaks off the summation at an arbitrarily chosen high frequency, and he finds that the result is a function of the vibrational quantum number for the initial state. Letting this vibrational quantum number be n and the frequency at which the sum is broken off be $\bar{\nu}$, the energy behaves as $\bar{\nu}^{n+\frac{1}{2}}$. Now a consideration of the matrix elements $\alpha_{nm}{}^s$ entering in V above shows without difficulty that breaking off the summation at $\bar{\nu}$ is nearly equivalent to dealing with an electron having a finite size described by the distribution function D and that this size is of the order of magnitude of $c/\bar{\nu}$. For different initial vibrational states, therefore, one obtains in the limit $\bar{\nu}\to 0$ infinite differences between the expectations of the energies of the radiation field.

It is instructive to consider the relation of this flux of energy into the radiation field with the principle of conservation of energy. One is, of course, sure that this principle is correct for any calculations involving quantum mechanics because of the existence of a Hamiltonian function. The total Hamiltonian may be written as

$$H = H_m + H_R + H',$$

where H_m, H_R refer, respectively, to radiation and matter alone and H' describes their interaction. For Rosenfeld's case H_m is positive definite and so under no circumstances can $\overline{H}_m = -\infty$. It has, furthermore, been seen [I, §7, V, §2] that

[65] L. Rosenfeld, Zeits. f. Physik **70**, 454 (1931).

$$H = m\dot{\mathbf{r}}^2/2 + V(\mathbf{r}) + H_R.$$

It is found that $\overline{H}_R = +\infty$. Therefore, $\overline{H} = +\infty$ because neither $\overline{V}$ nor $\overline{\dot{r}^2}$ can be $-\infty$. Therefore, initially also $\overline{H} = +\infty$. However, initially $\overline{H}_m$ was finite and $\overline{H}_R = 0$. Therefore, initially $\overline{H}' = +\infty$. Now H' itself consists of a sum of two parts one of which is linear in the vector potential $\mathbf{A}$ and the other involves $\mathbf{A}^2$. The linear part is readily seen to have an expectation value $= 0$ and, thus, the source of trouble may be said to be the term in $\mathbf{A}^2$. An easy direct calculation shows, in fact, that the expectation of $\mathbf{A}^2$ is infinite. It is curious to note that the infinity is, thus, finally traced back to the fact that the oscillators describing radiation may not be said to be in a state of absolute rest even when physically we like to deny the presence of light quanta. In this way even though we remove the infinite additive energy $\sum_s h\nu_s/2$ the origin of it—the lack of possibility of having a condition of absolute rest or a position of equilibrium—makes itself felt at another point of the theory.

Attempts at rectifying the theory by simply introducing a finite radius for the electron do not appear very rational because by so doing the relativistic invariance of the mathematical scheme is destroyed. It, furthermore, does not appear to have very much physical sense to assign to the electron any particular shape or dimensions. There is so far no evidence of a possibility of exploring such properties experimentally, and there is at present no way of explaining how such an electron would stay together. These difficulties of the classical theory remain in full force also in quantum mechanics. It is, on the other hand, likely [VII, §5] that quantum mechanics fails within the nucleus and that the representation of interactions between particles at nuclear distances is impossible by means of theories advanced so far. It, thus, appears reasonable to expect our present-day descriptions within nuclear dimensions to be fallacious, and there is from this point of view nothing very surprising in arriving at ultimate contradictions by having used calculations dealing too specifically with electrons and light quanta of zero dimensions.

An attempt at removing the difficulties by an apparently different formulation of the theory has been made recently by Dirac.[66] A more detailed investigation by Rosenfeld[67] shows, however, that the requirements laid down by Dirac are satisfied by the Heisenberg-Pauli theory of wave fields. It, thus, does not appear likely that one will arrive at a solution of the difficulties by formal methods based on present-day quantum mechanics.

On the other hand, the fact that quantum theory fails in the nucleus makes one hope that studies of nuclear physics will ultimately suggest a solution of the present difficulties of quantum-electrodynamics.

The writer is very grateful to Professors Pauli and Heisenberg for many discussions on the above subject.

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[66] P. A. M. Dirac, Proc. Roy. Soc. **A136**, 453 (1932).

[67] L. Rosenfeld, Zeits. f. Physik **76**, 729 (1932). P. A. M. Dirac, V. A. Fock and Boris Podolsky, Phys. Zeits. d. Sow. **2**, 468 (1932).