The Theory of Photon Packets and the Lennuier Effect.

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(ricevuto il 6 Dicembre 1959)

Summary. - The quantum electromagnetic field of radiation is studied for the case of a spectral distribution of energy and momentum. Packets of photons are introduced and classified. Some of their coherence properties are discussed by associating equivalent classical fields with each packet. The emission and dispersion of such packets are studied with special emphasis on the fluorescence resonance of frequencies near the atomic frequency. A scattered packet is shown to contain frequencies not present in the incident radiation, as found experimentally by R. LENNUIER, and the apparent non-conservation of energy is seen to be a consequence of the Heisenberg uncertainty principle.

1. – Introduction.

Since the very beginning of wave mechanics, quantum phenomena concerning electromagnetic waves have been widely studied. However, no complete formulation of the problem has been given to cover the situation when the wave contains a range of wave vectors k, except for some special problems such as the natural line breadth. The existence of experimental data, such as those obtained by R. LENNUIER (1) on resonance fluorescence, shows the need for such a formalism to describe processes involving non-monochromatic radiation.

We first introduce a formalism for photon packets in Section 2. These involve linear combination of many-photon functions, but since in physical

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(1) R. LENNUIER: *Ann. Phys.*, **2**, 233 (Paris, 1947).

processes the number of photons is conserved or changes by a definite number (one or two) we do not mix states with different numbers of photons. Some of the properties of the photon packets can be discussed by introducing the associated electromagnetic fields, as was first done by BECK [2] in a different context. This is necessary in order to define the momentum distribution of an arbitrary photon packet. In the limit of large numbers of photons, the associated fields also allow us to give a meaning to the classical concept of a coherent radiation, and to extend this concept to packets with a small number of photons.

The formalism is first applied to photon emission by an atom in Section **3**, and it is shown explicitly for the first time that a system containing a large number of similar radiating atoms gives a beam of coherent radiation as is very well known experimentally in classical optics. The second application (Section **4**) is to dispersion phenomena in which a packet changes its original distribution. This is applied in Section **5** to the dispersion of a one-photon packet by a bound electron (resonance fluorescence). LENNUIER observed that the scattered resonance fluorescence radiation contains frequencies not present in the incident radiation. This phenomenon is discussed in detail and the apparent non-conservation of energy is seen to be a consequence of the Heisenberg uncertainty principle.

2. – Photon packets.

When the radiation field is quantized and the photons are introduced, it is necessary to have a system of generalized functions to describe the field. We shall be interested in states of the field

$$\Gamma_k^n|0\rangle\,, \tag{1}$$

$$\Gamma_k^n = (\text{const.})\exp[-in\omega_k t](a_k^+)^n\,,$$

where a_k^+ is the creation operator of the photon k, k a multiple index that defines the wave vector and polarization; $|0\rangle$ is the state vector of the vacuum; the constant is chosen in order to normalize (1) to unity.

The state representing an arbitrary one-photon packet is therefore written

$$\Gamma^1|0\rangle = \sum_k \lambda_k \Gamma_k^1|0\rangle\,, \tag{2}$$

(2) G. BECK: *Nuovo Cimento*, **1**, 70 (1950).

where the λ_k's satisfy the normalization relation

$$\sum_k |\lambda_k|^2 = 1 . \tag{3}$$

In future we shall drop $|0\rangle$ and use Γ to represent states of the system. Γ^1 is not an eigenstate of the energy and k vector, but has a momentum and polarization distribution

$$f(k) = |\lambda_k|^2 .$$

An n-photon packet is similarly defined by

$$\Gamma^n = \sum_{[n_i]} L_n(n_1, n_2, n_3, ...)\Gamma_{k_1}^{n_1}\Gamma_{k_2}^{n_2}\Gamma_{k_3}^{n_3} ... , \tag{4}$$

where

$$L_n(n_1, n_2, n_3, ...) = 0$$

if

$$\sum_i n_i \neq n$$

and

$$\sum_{[n_i]} |L_n(n_i)|^2 = 1 . \tag{5}$$

We shall restrict the name photon packet to states such as (4) which contain a definite number n of photons, *i.e.* are eigenstates of the photon number operator $\sum_k a_k^+ a_k$.

As mentioned in the introduction, it is useful to associate a classical field with a photon packet. We first define the spectral distribution operator

$$D = \sum_{kk'} a_k^+ a_{k'} \exp\left[- i(\boldsymbol{k} - \boldsymbol{k}')\boldsymbol{r}\right] . \tag{6}$$

This is, for a photon packet, the diagonal part of the square of the vector potential operator, except that we omit for convenience a numerical coefficient $2\pi c^2\hbar(\omega_k\omega_{k'})^{-\frac{1}{2}}$ and the null field divergence. Similarly D is related to the energy density and the action density of the system except for some coefficients. The Fourier components of this operator, namely $D(k, k') = a_k^+ a_{k'}$, have for a photon packet Γ^n the expectation value

$$\begin{cases} \langle D(k, k')\rangle_n = \sum_{[n_i]} n_k |L_n(n_i)|^2 & k = k', \\ \qquad\qquad\quad = \sum_{[n_i]} \left[(n_k + 1)n_{k'}\right]^{\frac{1}{2}} L_n^*(n_i) L\ (n_i + \delta_{ki} - \delta_{k'i}) & k \neq k'. \end{cases} \tag{7}$$

Now a general electromagnetic field can be expressed in terms of the vector potential

$$(8)\qquad \boldsymbol{A}_c = \sum_k \boldsymbol{e}_k \sqrt{4\pi c^2}\left[q_k \exp\left[ik_\mu x_\mu\right] + q_k^* \exp\left[-ik_\mu x_\mu\right]\right] \qquad \mu = x, y, z, ict.$$

where $\boldsymbol{e}_k$ and q_k are respectively the polarization versor and the amplitude of the k Fourier component. It is convenient to define the classical quantity

$$(9)\qquad \boldsymbol{D}_c = 2/\hbar \sum_{k,k'} \sqrt{\omega_k \omega_{k'}}\, q_k^* q_{k'} \exp\left[i(k'_\mu - k_\mu)x_\mu\right],$$

which is essentially the intensity of the field, and is the classical dynamical variable corresponding to the operator $\boldsymbol{D}$ (6). Its Fourier coefficients are similarly

$$(10)\qquad \boldsymbol{D}_c(k, k') = 2/\hbar \sqrt{\omega_k \omega_{k'}}\, q_k^* q_{k'},$$

Thus the classical field which we shall associate to each photon packet is defined by the condition

$$(11)\qquad \langle \boldsymbol{D}(k, k')\rangle_n = \boldsymbol{D}_c(k, k').$$

Although (11) always defines $\boldsymbol{D}_c$ the set of equations (10) do not necessarily have a solution for the q's and thus it is not always possible to invert (9) and (8) to derive the corresponding $\boldsymbol{A}_c$. Although $\boldsymbol{A}_c$ is completely arbitrary, $\boldsymbol{D}_c$ has to have the form of the square of an arbitrary vector and is not itself arbitrary. It is of course necessary to work with the intensities of the field $\boldsymbol{D}$ and $\boldsymbol{D}_c$ rather than with the corresponding $\boldsymbol{A}$ and $\boldsymbol{A}_c$ because $\langle \boldsymbol{A}\rangle$ is always zero. Physically (11) always defines the spectral distribution of the energy, but this distribution does not always correspond to a single field $\boldsymbol{A}_c$ of well determined intensity and phase. In fact, if a quantum field admits its classical associate *i.e.* if $\boldsymbol{D}_c$ corresponds to a single $\boldsymbol{A}_c$, it means that the quantum packet corresponds to a coherent field in which the Fourier components are well defined in magnitude and relative phase. Inspection shows that packets satisfying this condition are:

a) Packets containing only one state k but any number of photons;

b) One-photon packets;

c) Packets of n photons having the same distribution as a one-photon packet.

The last is the most general case and includes the previous two as particular cases. Two packets are said to have equivalent distribution when the expectation values of the D operator are proportional, the factor of proportionality being the ratio of the total number of photons. If the one-photon packet is expressed by (2), it can easily be shown that a packet of n photons has an equivalent distribution if and only if

$$L_n(n_i) = \frac{n!}{n_1!\, n_2!\, n_3! \dots n_n! \dots}\, \lambda_1^{n_1} \lambda_2^{n_2} \lambda_3^{n_3} \dots \lambda_n^{n_n} \dots \tag{12}$$

Only these packets correspond to one coherent radiation field.

For a more general packet, it may be possible to associate M classical fields in such a way that the expectation value of D is equal to the sum of the M classical D_c magnitudes. Consider for example the packet

$$\Gamma^2 = \sum_{kk'} \lambda_k \mu_{k'} \Gamma^1_k \Gamma^1_{k'}$$

such that $\lambda_k \neq 0$ only if $\mu_k = 0$ and vice versa. This two-photon packet is the superposition of the two one-photon packets defined by λ_k and μ_k with non-overlapping spectral distribution. With it we can associate two classical fields, *i.e.* those belonging to the two one-photon packets, but on going through the analysis it is found that the relative phase of these two fields is completely arbitrary. If the two packets are not orthogonal $(\sum_k \lambda_k^* \mu_k \neq 0)$, then even the two fields are to some extent arbitrary. However, as will appear later, the physically important fact is the minimum number of separate fields required.

3. – Photon emission.

In this section we shall consider the emission of a photon by an electron making a transition between two bound states of energy E_1 and E_2, and apply the general theory of damping phenomena formulated by HEITLER [3].

In the case of spontaneous emission, the general theory gives immediately the formation of the photon packet

$$\Gamma^1 = \sum_k \alpha_k \Gamma^1_k , \tag{13}$$

where

$$\alpha_k = \frac{A H'_{k,0}}{\hbar(\omega_k - \omega_0 + i\gamma/2)} ,$$

[3] W. HEITLER: *Quantum Theory of Radiation*, 3rd edition (Oxford, 1954), p. 163, 181, 196.

$H_{k,0}$ is the matrix element of the interaction Hamiltonian between the electron and the electromagnetic field, taken between the state k, consisting of the electron in the ground state and one photon k and the state 0, consisting of the electron in the excited state and no photon; ω_0 is the frequency defined by $\hbar\omega_0 = E_1 - E_2$ and γ is the natural line breadth; A is some normalization factor.

A more interesting case is the emission by an atom in the presence of the one-photon packet (3) with distribution λ_k, *i.e.* stimulated emission. The result in this case is a packet of two photons with expansion coefficients

$$(14)\qquad \begin{cases} L_2(n_k = 1,\ n_{k'} = 1) = A(\lambda_k \alpha_{k'} + \lambda_{k'} \alpha_k) \\[2mm] L_2(n_k = 2) = A\sqrt{2}\,\lambda_k \alpha_k\,, \end{cases}$$

where α_k is again given by (13) and A is the normalization facor

$$(15)\qquad A^2 = [1 + |\beta|^2]^{-1}$$

and

$$(16)\qquad \beta = \sum_k \alpha_k^* \lambda_k$$

is the scalar product of the two one-photon packets α_k and λ_k. This two-photon packet has, in general, two associated fields that can be taken to be the fields associated with the one-photon packets (*).

$$(17)\qquad \begin{cases} \nu_{1k} = \left[\dfrac{1 - |\beta|^2}{1 + |\beta|^2}\right]^{\frac{1}{2}} \alpha_k \exp[i\varphi_1]\,, \\[4mm] \nu_{2k} = \left[\dfrac{1}{1 + |\beta|^2}\right]^{\frac{1}{2}} [\beta\alpha_k + \lambda_k] \exp[i\varphi_2]\,. \end{cases}$$

If $|\beta| \neq 1$ then (17) shows that the associated fields contain two arbitrary phases φ_1 and φ_2. Physically, with arbitrary incident radiation, the emitted packet is partly phase incoherent with the incident one. If $|\beta| = 1$, (17) reduces to one classical field, *i.e.* the emitted radiation coherent with incident. The condition $|\beta| = 1$ implies that the incident photon is equal to the photon emitted in vacuum (**).

(*) As mentioned before, the association is not unique, and in this case, for example, an exchange of α_k and λ_k in (17) yields another possible association.

(**) $|\beta| = 1$ implies $\lambda_k = \alpha_k \exp[i\varphi]$. The arbitrary constant phase difference has no physical significance and is completely undetermined since the phase is the canonical conjugate variable of the number of photons which is diagonal in our case.

Similarly, if the incident packet is a coherent n-photon packet with spectral distribution equal to the photon (13) emitted in the vacuum, it is found that the emission of an additional photon is stimulated. This photon is added coherently to the wave. Instead of the factor $\sqrt{2}$ in (14), a factor $\sqrt{(n_k+1)}$ comes in, corresponding to the usual enhancement factor for stimulated emission. A packet of the form (12) is built up, so that the emitted photon is added coherently to the wave. Thus the emission from an assembly of many radiating identical atoms results in a final coherent n-photons packet with one coherent associated classical field, as is well known experimentally from classical interference experiments.

4. – Dispersion of photon packets.

In this section we shall study phenomena in which packets change their structure, *i.e.* the scattering of photons by electrons. As regards scattering by free electrons (Compton effect), the calculations are quite straightforward with the following results. With any type of incident packet, it is verified that the scattered packet contains only wave vectors consistent with the conservation of energy and momentum. The main result of interest is that the scattered photon is incoherent with the unscattered part of the incident packet, even when the incident packet is coherent.

As regards scattering by a bound electron, consider an incident one-photon packet with spectral distribution λ_k containing frequencies in the region of the excitation frequency ω_0 to an excited state of the electron. The outgoing packet contains some unscattered incident radiation plus a dispersed component. As is shown in the Appendix, the dispersed part has spectral distribution

$$\mu_k = \frac{AH_{k,0}}{\omega_k - \omega_0 + i\gamma_0/2} \cdot \sum_{k'} \frac{\lambda_{k'} H_{0,k'}}{\omega_k - \omega_{k'} + i\gamma_{k'}/2}\,, \tag{18}$$

where

$$\gamma_{k'} = \frac{2i}{\hbar^2} \frac{|H_{k',0}|^2}{\omega_k - \omega_0 + i\gamma_0/2}\,, \tag{19}$$

$H_{k,0}$ is the matrix element for the emission of a photon k from the atom when it goes from the excited state to the ground state; γ_0 is the natural line width of the excited state.

Eq. (18) has been evaluated for several different initial distributions and the results are summarized as follows:

i) For an incident monochromatic packet of frequency ω near ω_0, the outgoing packet is practically monochromatic with line width γ_k ($\ll \gamma_0$) defined by (19).

ii) If the ingoing packet is a «white» beam, *i.e.* it has a uniform distribution over a very wide range containing ω_0, the dispersed photon has the same distribution as the natural line emitted by the atom in the excited state, *i.e.* a line with natural width γ_0 centred about ω_0.

iii) If the original packet has the same distribution as a packet emitted in vacuum by the atom in the state concerned, *i.e.* a line with maximum at ω_0 and width γ_0, then the dispersed radiation has a stronger maximum at ω_0 and is somewhat narrower than the incoming one. The intensity distribution is in fact proportional to the square of the spectral distribution of the incident radiation.

iv) If the excitation packet has a maximum at ω near ω_0 and a natural width γ, the fluorescent packet shows two maxima, the first near ω_0, the second near ω. The distribution is in fact just proportional to the distribution of case ii) above multiplied by the intensity distribution of the incident packet, as one would expect.

Cases i) and ii) are the well-known cases of excitation by a very sharp line and excitation by an uniform distribution respectively, discussed by HEITLER. However the present formalism allows one to make explicit calculations for the first time for cases iii) and iv).

5. – The Lennuier effect.

Our most important application of formula (18) is the account for the effects observed by LENNUIER. In his experiments, mercury $\lambda = 2537$ Å radiation was obtained from a hot source containing several isotopes, so that the line was very broad with a width of about $4.7 \cdot 10^{-2}$ Å. The central part of the band containing the fundamental frequency was then eliminated by means of a suitable filter. This beam, with the characteristic frequency missing, was used to excite another sample of mercury, and the fluorescence radiation was analysed after eliminating the coherent radiation due to normal dispersion. The outgoing beam showed three maxima instead of two, the third being at the fundamental atomic frequency. The appearance in the final state of frequencies not found in the original radiation and the apparent non-conservation of energy were at first sight very puzzling. However, we shall show that the effect can be deduced directly with the use of eq. (18).

The exact shape of the incoming packet is in general very complicated. However, the most important fact of the appearance of the third maximum can be explained with a simple approximation to the real distribution. We have chosen $|\lambda_k|$ to have a distribution consisting of two triangles with maxima at $\omega_0 \pm 2\gamma_0$, each one of total width $2\gamma_0$ as shown in Fig. 1. The total width

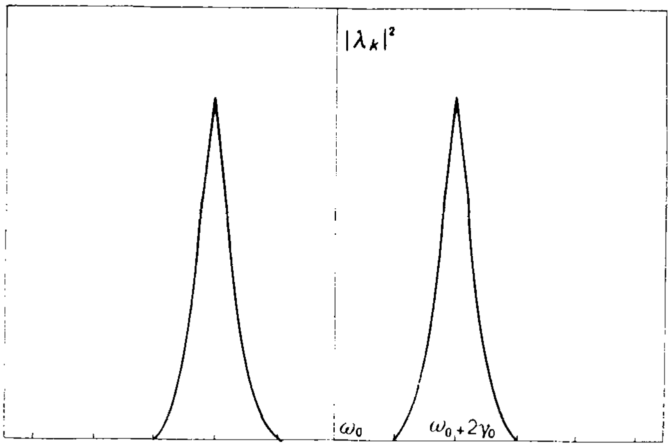

Fig. 1. – Approximate intensity distribution of the incident radiation in Lennuier's experiment.

of the spectrum, $6\gamma_0$, represents the width of the original broad line, and the missing central frequencies in a range $2\gamma_0$ about ω_0 are those eliminated by the filter. We assume a constant phase in each of the two regions of the spectrum, but a phase difference of φ between the two parts. When this distribution is substituted into (18), the value of the summation depends sensitively on the phase difference φ between the two parts. The resultant spectra for the two extreme cases $\varphi = 0$ and π are plotted in Figs. 2 and 3 and it is seen that while the first spectrum resembles the incident radiation, the second one shows a very high peak at ω_0. In the actual Lennuier experiment, the two zones of the initial spectrum probably came from the emission by different isotopes. The phase difference between them is therefore quite arbitrary and the observed spectrum is the intensity distribution averaged over all φ. The resultant spectrum is shown in Fig. 4 and displays the three maxima of approximately the same height as observed by LENNUIER.

The appearance of the third maximum can be explained simply by the Heisenberg uncertainty principle. When the atom is excited by a monochromatic wave that is spread through all space, the time at which the excitation takes place is completely unknown. Consequently the energy can be exactly determined and the outgoing packet is therefore practically mono-

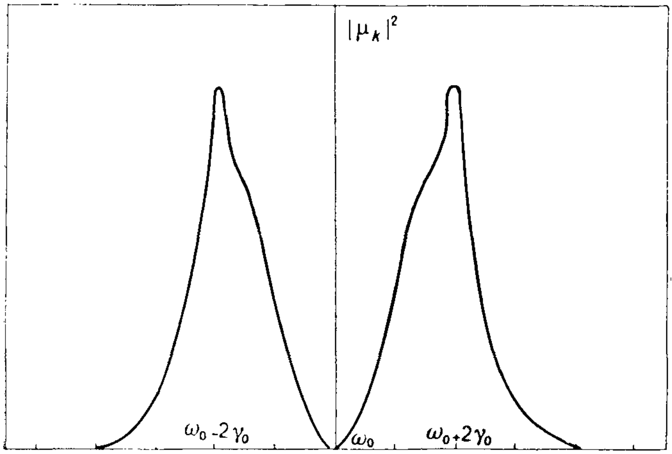

Fig. 2. – Fluorescent packet for $\varphi = 0$.

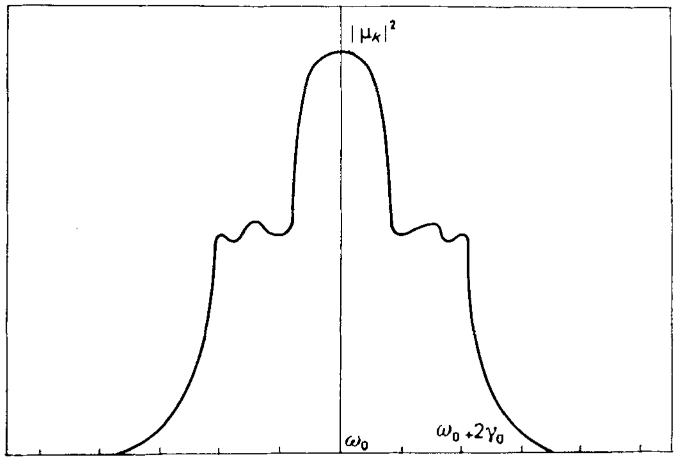

Fig. 3. – Fluorescent packet for $\varphi = \pi$.

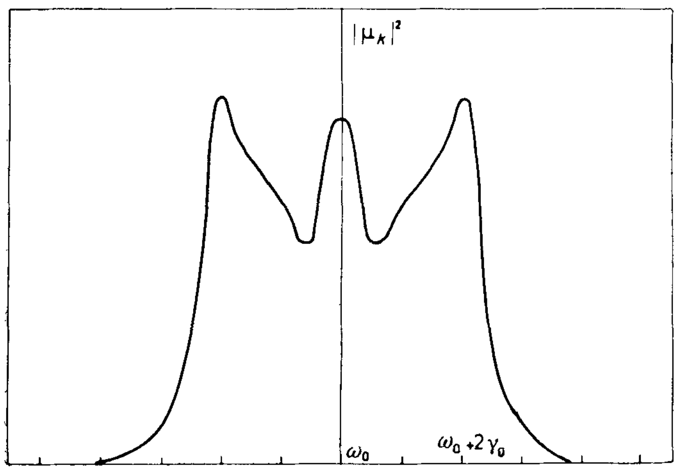

Fig. 4. – The final observed intensity when averaged over all possible phase differences.

chromatic. When the excitation is produced by a uniform distribution, the incoming packet is well localized in space, and the time of the excitation of the atom can be determined with high certainty. As the mean life of the excited state is of the order $\hbar\gamma_0^{-1}$ and is the only time uncertainty in the process, the energy uncertainty in the fluorescent packet is of the order γ_0. The spectral distribution is that of the characteristic line. The other cases of Section 4 and the Lennuier experiment are intermediate examples. The atomic process reduces the time uncertainty and the fluorescent spectrum is then broadened. The frequency ω_0, favoured by resonance phenomena, appears then in the outgoing packet.

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The author is deeply indebted to Dr. J. A. BALSEIRO for suggesting this investigation and for his constant advice and illuminating discussions. He wishes also to acknowledge some useful discussions with Dr. V. HEINE and Dr. J. WESTERKAMP and the help of Dr. M. BALANZAT in the solution of some mathematical difficulties.

APPENDIX

To study the fluorescent dispersion of a one photon packet we consider the solution of the time-dependent Schrödinger equation with the initial conditions of Table I.

TABLE I. – *States, energies and initial conditions for fluorescence resonance.*

State			Initial condition $b_n(0)$	Energy E_n
Label	Atom	Photon		
ki	Ground state	k	λ_k	$\hbar\omega_k$
0	Excited state	none	0	$\hbar\omega_0$
kf	Ground state	k	0	$\hbar\omega_k$

The states ki and kf are the same states of the system, but for the sake of clarity we add i or f according to whether the state is appearing as a component of the initial or final packet. The Schrödinger equation becomes

$$i\hbar\,\dot{b}_n(t) = \sum_m H_{n,m} b_m(t) \exp\left[i(E_n - E_m)t/\hbar\right] + i\hbar\, b_n(0)\,\delta(t)\,. \tag{A.1}$$

In terms of the Fourier transform

$$b_n(t) = -\frac{1}{2\pi i}\int_{-\infty}^{\infty} \mathrm{d}E\, G_n(E)\exp[i(E_n - E)t/\hbar], \tag{A.2}$$

the equations becomes

$$(E - E_k)G_{ki}(E) = H_{ki,0}\,G_0(E) + \lambda_k\,, \tag{A.3}$$

$$(E - E_0)G_0\,(E) = \sum_k H_{0,ki}G_{ki}(E) + \sum_k H_{0,kf}G_{kf}(E)\,, \tag{A.4}$$

$$(E - E_k)G_{kf}(E) = H_{k',0}G_0(E)\,. \tag{A.5}$$

We now put

$$G_0 = \sum_k U_{0,ki}G_{ki}\zeta(E - E_0)\,, \tag{A.6}$$

$$G_{k'f} = \sum_k U_{k'f,ki}\,G_{ki}\,\zeta(E - E_{k'})\,, \tag{A.7}$$

where

$$\zeta(x) = \frac{\mathscr{P}}{x} - i\pi\,\delta(x)\,.$$

Substituting (A.6) and (A.7) into (A.4) and (A.5), and using

$$x\zeta(x) = 1$$

we obtain

$$U_{k'f,ki} = \frac{H_{k'f,0}H_{0,ki}}{E - E_0 + i\hbar\gamma_0/2}\,, \tag{A.8}$$

$$U_{0,ki} = \frac{(E - E_0)H_{0,ki}}{E - E_0 + i\hbar\gamma_0/2}\,, \tag{A.9}$$

where

$$\gamma_0 = 2i/\hbar \sum_k |H_{0,kf}|^2\zeta(E - E_k) \tag{A.10}$$

is the natural line breadth of the excited atomic state. Substituting (A.9) and (A.6) into (A.4) we obtain

$$G_{ki} = \frac{\lambda_k}{E - E_0 + i\hbar\gamma_k/2}\,, \tag{A.11}$$

where

$$\gamma_k = 2i/\hbar\,\frac{|H_{ki,0}|^2}{E - E_0 + i\hbar\gamma_0/2}\,, \tag{A.12}$$

and where we have neglected terms of the order $e^2/\hbar c$. We are interested in the values of $b_{kf}(t=\infty)$ which are proportional to the coefficients in the final packet. From (A.2), (A.7), (A.8) and (A.11), letting $t \to \infty$, and using

$$\lim_{t\to\infty} \zeta(x) \exp[-ixt] = -2\pi i\, \delta(x)\,,$$

the final result (18) is obtained.

RIASSUNTO (*)

Si studia il campo elettromagnetico quantistico della radiazione per il caso della distribuzione spettrale dell'energia e dell'impulso. Si introducono e si classificano i pacchetti di fotoni. Si discutono alcune delle loro proprietà di coerenza associando ad ogni pacchetto un campo fisico equivalente. Si studia l'emissione e la dispersione di tali pacchetti sottolineando la risonanza di fluorescenza a frequenze prossime a quella atomica. Si mostra che un pacchetto che ha subito lo scattering contiene frequenze che non sono presenti nella radiazione incidente, come è stato trovato sperimentalmente da R. LENNUIER, e si vede che l'apparente non-conservazione dell'energia è una conseguenza del principio di indeterminazione di Heisenberg.

(*) *Traduzione a cura della Redazione.*