RESONANCE INTERACTION OF AN INTENSE PHOTON FLUX WITH A BOUND ELECTRON AND THEORY OF THE DELAY TIME

M. E. PEREL'MAN

Cybernetics Institute, Georgian Academy of Sciences

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Stimulated emission and elastic scattering of resonance photons in a two-level system are calculated by the methods of quantum electrodynamics and scattering theory, taking account of absorption and re-emission of the incident quanta. The rates of these reactions possess a saturation threshold with respect to the photon flux density $j_0 \sim 1/\lambda^2\tau$, where τ is the duration of an elementary scattering act. It is shown that new reaction channels may open up not as a result of increase in the energy of the particle being scattered but as a result of increase of the flux density of the incident particles. An experimental investigation of these features (and also of the upper end lower bounds, calculated for the given model, of the monochromaticity region of the stimulated radiation) could help to elucidate the question of the existence and magnitude of the delay time.

1. INTRODUCTION AND SURVEY OF THE RESULTS

IN this article a calculation is performed, using the methods of quantum electrodynamics and scattering theory, on the processes of stimulated emission and elastic scattering in a two-level system

$$n\gamma(\omega) + e_2(E_2) \to (n+1)\gamma(\omega) + e_1(E_1), \tag{1.1}$$

$$n\gamma(\omega) + e_k \to n\gamma(\omega) + e_k \quad (k = 1, 2) \tag{1.2}$$

close to resonance (e_1 and e_2 are states of the electron in the lower and upper levels of the two-level system and $|\omega - \omega_0| \lesssim \Gamma$, where $\omega_0 = E_2 - E_1$ and Γ is the half-width of the excited level), when features associated with the quantum nature of an intense electromagnetic wave are more pronounced.

Thus, the transition (1) in a flux of density j is usually regarded as a one-photon process; allowing for the photon statistics, this leads to the transition probability

$$W = A + Bj(\omega_0). \tag{1.3}$$

However, consideration of the rate of reaction (1.1)

$$dR_n = W dN_1 \ldots dN_n \tag{1.4}$$

(dN_i is the number of incident photons of the i-th species in the interval $(\omega_0, d\omega)$) leads to the necessity, close to resonance, of writing in place of (1.3) a more general "virial" expansion (Secs. 2, 3):

$$R \equiv \sum_n R_n = A + \sum B_n(\omega, \omega_0, \Gamma) j^n \tag{1.5}$$

(far from resonance $B_n \to 0$ like x^{n-1} and (1.5) goes over to (1.3)).

In (1.5) the important thing is the convergence of the series in j. The presence of a radius of convergence of (1.5) as $n \to \infty$ is interpreted in Sec. 4 by means of the concept of a delay time[1,2] in the $e\gamma$ scattering:

$$\tau \equiv \mathrm{Re}\,(d/id\omega)\ln S = {}^1/_2\Gamma[(\omega-\omega_0)^2 + \Gamma^2/4]^{-1}. \tag{1.6}$$

An examination of the resulting features could serve as the basis for an experimental investigation of the concept of a delay time[1)]. (The interactions considered, in view of the ease of obtaining intense monochromatic fluxes, are unique in the whole of elementary particle physics[2)].)

It is interesting that (1.6) elucidates the features of (1.2) even for n = 1. In fact, introducing the flux density (we are concerned with the order of magnitude)

$$j_0 = 1/\lambda^2\tau(\omega), \tag{1.7}$$

which determines (phenomenologically) the maximum flux of photons being scattered independently by one electron, we can write the reaction rate for Kramers-Heisenberg resonance scattering[5] in the form

$$R = \sigma j \approx (4\pi c^2/\omega^2)\Gamma^2 j[(\omega-\omega_0)^2 + \Gamma^2/4]^{-1} \sim \Gamma j/j_0. \tag{1.8}$$

The point $j = j_0$ is singular for R(j) and is the threshold of new processes, when the scatterings of separate photons cannot be assumed to be independent (absorption of the next photon occurs before the emission of the previous one) and it is necessary to take account of the scattering amplitude of a photon by an electron in the upper level. If the cross section for such a scattering is much smaller than (1.8), then $R(j > j_0) \approx R(j_0)$ and saturation of the transition (1.8) occurs (if this relation is not fulfilled, it will be possible to conclude that the extra re-emissions increase the resonance transition time).

Thus, a characteristic feature of processes in high-intensity beams is the presence of thresholds for new

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1)A theory of the delay time has been developed for two-particle interactions and is an alternative method of description for these. The very formulation of the problem in [1,2] and in other papers enables us neither to carry out a direct experimental check of the theory or its consequences, nor to investigate phenomena not studied by other approaches. From a general point of view, it is also important to relate the delay time to the delay region (cf. [3]).

2)We note that, in agreement with Höhler's analysis, the processes considered are similar to processes in the Lee model with a decaying V-particle: the transition (1.1) corresponds to stimulated emission of a θ-particle and the expansion (2.3) to consecutively allowing for all sectors of the model (the necessary changes in the formulas for such reactions are obvious).

processes determined not by the energy of an individual particle being scattered but by the flux density of the fixed-energy particles being scattered. Evidently, the existence of these thresholds can be established only with the help of the concept of duration of the elementary acts of interaction (their spread depends on the dispersion of τ—cf.[6]), and an experimental determination of the existence of thresholds and interaction saturation phenomena would be able to verify the correctness of the whole concept.

It seems that it should be possible to check the following conclusions: 1) the dependence, given by (4.2) and (4.7), of the monochromaticity of reaction (1.1) on j and on the duration of the stimulating pulse, and the presence of the threshold (4.3); 2) the change in the form of the refractive index close to the absorption line when $j \sim j_0$, insofar as it is determined by a reaction of the type (1.2)[7]; 3) the necessary presence in nonlinear processes with substantially multiphoton absorption (doubling of the frequency and multiphoton ionization) of a lower j-threshold, equal to $1/\lambda^2\tau_{min}$, where τ_{min} is the minimum scattering duration in the chain of absorptions which lead to ionization, multiple increase of the frequency, etc.[7] (Of course, the calculations in the article are of a model character and do not pretend to quantitative agreement with experiment—cf.[8].)

2. QUANTUM ELECTRODYNAMICS

The calculation for the processes (1.1) and (1.2) is conveniently carried out in the Furry picture, using the low Green function[5] for the bound electron:

$$G(x_1, x_2) \approx \frac{1}{2\pi i}\sum_n \psi_n(\mathbf{r}_1)\bar{\psi}(\mathbf{r}_2)\int_{-\infty}^{\infty}\frac{\exp[i\omega(t_1-t_2)]\,d\omega}{E_n+\omega-i\Gamma_n/2} \quad (2.1)$$

(the level shift is included in E_n). All the radiative corrections to the propagator line enter into (2.1), while the radiative corrections to the vertex part enter naturally into Γ_n (here we can also include all corrections for the Doppler effect etc.). Virtual photon lines which pass round two or more external vertices are realized in the form of low-frequency photons, which are not important in what follows; other possible corrections reduce to the appearance of renormalization constants in Γ_n. Thus, taking experimental values for E_n and Γ_n enables us to leave radiative corrections out of consideration throughout the following (the problem of the calculation of E_n and Γ_n as functions of j is not considered in this article).

The external electromagnetic field can be quantized in the plane-wave approximation in the usual way (V is the quantization volume):

$$A_{i,f} \to \sum_k \frac{\hat{e}_{i,f}\,a_k}{(2\omega_{i,f}V)^{1/2}}\exp[\pm i(\mathbf{k}_{i,f}\mathbf{r}-\omega_{i,f}t)]. \quad (2.2)$$

The transition amplitude for (1.1) or (1.2) can be written in the form of a cluster decomposition:

$$\langle m\gamma', e_f|S|n\gamma, e_i\rangle = 2\pi i\sum_k C_k\langle(m-n+k)\gamma', e_f|T|k\gamma, e_i\rangle\,Q_k \times\delta\left(\sum E_i-\sum E_f\right) \quad (2.3)$$

(nonphysical singularities arising because of factors of the type $\langle \mathbf{k}\gamma|\mathbf{k}\gamma\rangle$ (e.g.,[9]) can be eliminated by going over to a wave-packet or similar representation). The factor C_k corresponds to the weights of the multiple terms, and the factor Q_k corresponds to the density of states and contains the whole dependence of (2.3) on the quantization volume. We can already take into account in (2.3) (although this is usually done for the transition probabilities—cf.[1], p. 185) that in the interval $(\mathbf{k}_i, d\mathbf{k}_i)$ there are dN_i photons of the i'th species, and we can formally replace Q_k by

$$Q_k' = Q_k\prod_1^k\sqrt{\gamma dN_i} = \prod_{i=1}^k\left(\frac{dN_i}{2\omega_i V}\right)^{1/2}\prod_{f=1}^{m-n+k}\frac{1}{(2\omega_f V)^{1/2}} \to \prod_{i=1}^k\left(\frac{\rho_i(\mathbf{k})\,d\mathbf{k}_i}{2\omega_i}\right)^{1/2}\prod_{f=1}^{m-n+k}\left(\frac{d\mathbf{k}_f}{2\omega_f(2\pi)^3}\right)^{1/2}, \quad (2.4)$$

where $\rho_i(\mathbf{k})$ is the density of photons of species i; in a monochromatic directed (coherent) flux, $\rho(\mathbf{k}_i) = \rho\delta(\mathbf{k}_i - \mathbf{k}_0)$.

The degree of nonorthogonality of states with different numbers of photons is determined by means of (1.6), from the extent of overlap of the wavefunctions:

$$d \sim (\lambda/c\tau)^3 \approx (2\lambda/c\Gamma)^3[(\omega-\omega_0)^2+\Gamma^2/4]^3. \quad (2.5)$$

At resonance, $d \sim (\lambda\Gamma/c)^3 \sim 10^{-18}$, and, with allowance for (2.3), the reaction rate (1.4) for the process (1.1) acquires the form

$$dR_n = 2\pi\sum_{q=0}^n\left(\frac{j}{2\omega}\right)^q\prod_{f=1}^{q+1}\int\frac{d\mathbf{k}_f}{2\omega_f(2\pi)^3}C_q^2 \times|\langle(q+1)\gamma, e_f|T|q\gamma, e_i\rangle|^2\,\delta\left(\sum E_i-\sum E_f\right), \quad (2.6)$$

i.e., can be written in the form (1.5). (To simplify the writing, here and below we do not take into account of factors arising from averaging over the initial polarizations and summing over the final polarizations.)

We shall consider the transition (1.1) for n = 1, when one of the resonance photons incident on the excited electron directly induces its transition to the ground state. The process (1.1) is described by six diagrams, pairs of which differ by permutations of finite photon lines. The three basic diagrams are shown in Fig. 1[3].

When $|\omega-\omega_0| < \Gamma/2$ the main contribution to the transition probability is made by the resonance diagrams b; in these it is possible to neglect the A_μ^2 terms and, in the long-wavelength approximation of practical interest, replace all the radius vectors in (2.6) by the radius vector of the centre of the atom. With these assumptions, (2.6) leads to the expression

$$R_1 = \frac{64\pi}{9\omega^2}j\Gamma\frac{\Gamma}{(\omega-\omega_0)^2+\Gamma^2/4} \approx \frac{256\,\pi c^2}{9\omega_0^2}j. \quad (2.7)$$

If, however, all n photons in (1.1) interact directly,

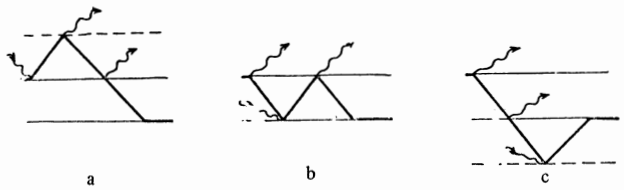

FIG. 1

[3]In all the diagrams, a continuous line is an electron line, a wavy line is a photon line, a real line represents a real electron level and a dotted line a virtual level. The chronological order of events follows the abscissa.

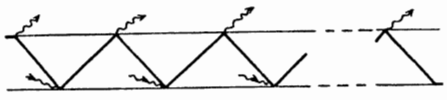

FIG. 2

i.e., by means of absorption and re-emission, with the excited electron, inducing its transition to the ground state, then the basic resonance diagram (Fig. 2) will describe the "oscillation," known in quantum mechanics[10], of an electron between levels. Allowing only for the contribution of the resonance terms ($C_n = n!\,(n+1)!/(2n+1)!$), we obtain the integral and total transition rates in a stationary flux ($n \gg 1$):

$$R_n \approx \pi\Gamma n v^n, \quad R = \sum_0^\infty R_n \approx \pi\Gamma \frac{v}{(1-v)^2} \quad \left(v = \frac{j}{j_0}\right) \tag{2.8}$$

We sum the series (2.8) for $v < 1$, i.e., for (cf. (1.7))

$$j < j_0 = \frac{\omega^2}{\pi c^2} \frac{(\omega-\omega_0)^2 + \Gamma^2/4}{\Gamma} \approx \frac{\omega_0^2 \Gamma}{4\pi c^2}. \tag{2.9}$$

Other multiphoton resonance processes can be calculated in a similar way. Thus, to generalize the expression (1.8) describing the dispersion close to resonance, we must consider (1.2). Proceeding in the same way as above, we obtain the reaction rates ($j < j_0$):

$$R_n \approx 2\Gamma n^{-1} v^n, \quad R = \sum_0^\infty R_n \approx -2\Gamma \ln(1-v). \tag{2.10}$$

Similar expressions can also be obtained for other processes, combination of frequencies, combination scattering, etc.

The reaction rate (2.8) was obtained by taking account only of resonance transitions. The presence of non-resonance transitions leads to the appearance in (2.8) of factors of the form

$$v_k \equiv \frac{j_k}{j_0} = \frac{\pi j c^2}{\omega^2} \Gamma \left[(k\omega + \omega_0)^2 + \frac{\Gamma^2}{4}\right]^{-1} \approx \frac{j}{j_0} \frac{\Gamma}{4(k+1)^2 \omega_0^4} \tag{2.11}$$

(the interference terms between a resonance term and any non-resonance term are proportional to $|\omega - \omega_0|$ and can be discarded). If q non-resonance transitions of first order (k = 1) occur between two resonance transitions, in (2.8) we must make the replacement

$$v \to v v_1^q \approx (j/j_0)^{q+1} (\Gamma^2/16\omega_0^2)^q, \tag{2.12}$$

leading to the convergence radius (2.8)

$$j < j_1(q) = j_0 (4\omega_0/\Gamma)^{2q/(q+1)}. \tag{2.13}$$

Thus, limiting the treatment to resonance terms only is equivalent to leaving in (1.5) only the terms responsible for the nearest singularity, and the justification for our approach lies in this fact.

3. DURATION OF THE INTERACTION AND UNITARITY

We shall derive the same results in another way ([1], p. 498). The duration of the transition (1.2) should depend on the parameters ω_0 and Γ of the scatterer and on the frequencies of the incident photons (we consider "elastic" interactions). If all $\omega_k = \omega_0$, the reciprocal of the duration of the resonance transitions can be expanded in a Taylor series in all the independent ω_k:

$$\tau_{Rn}^{-1}(\omega_1, \ldots, \omega_n) = \alpha + \sum [\beta_k + \gamma_k(\omega_k - \omega_0)](\omega_k - \omega_0)^2 + \cdots \tag{3.1}$$

The independence of the transitions implies that, close to resonance, when the cross terms $\sim (\omega - \omega_0)^4$ in (3.1) can be neglected, the S-matrix can be factorized and the interaction time is additive:

$$S_n = \prod_k S_{nk}'(\omega_0, \Gamma; \omega_k), \quad \tau_{Rn} = \sum_k \tau_n^{(k)}(\omega_k) = \mathrm{Re}\left\{-i \sum_1^n \frac{d}{d\omega_k} \ln S_{nk}'\right\}. \tag{3.2}$$

Integration of (3.2) taking the single-valuedness into account leads to the unitary S-matrix:

$$S_n = \exp\left\{2i \sum [\delta(\omega_k) + \nu(\omega_k)]\right\} = \prod \left(1 + \frac{i\Gamma}{\omega_0 - \omega_k - i\Gamma/2}\right) e^{2i\nu(\omega_k)}, \tag{3.3}$$

where $\delta(\omega_k) = \tan^{-1}[\Gamma/2(\omega - \omega_k)]$ is the partial phase shift.

The scattering matrix (3.3) is close to that used in Sec. 2. The extra terms in (3.3) could correspond to realizations of the transition (1.2) through chains of real processes of the type $k\gamma + e_2 \to (k+1)\gamma + e_1$, $m\gamma + e_1 \to (m-1)\gamma + e_2$ ($m + k = n$), and so on (the absence of chains of such processes in the scheme of Sec. 2 leads to an apparent violation of unitarity).

In the following, we shall need (3.3) for all $\omega_k = \omega \sim \omega_0$. In this limit, obviously, $\alpha = \Gamma/4n$, $\beta_k = \beta = (n^2\Gamma)^{-1}$, $\gamma_k = 0$ and

$$S_n = \exp(2in\delta(\omega)) = [1 + i\Gamma/(\omega - \omega_0 - i\Gamma/2)]^n, \quad \tau_{Rn} = n\tau. \tag{3.4}$$

The calculation performed is valid only for elastic processes of the type (1.2). We shall try, however, to extract the maximum information from it by generalizing the optical theorem of scattering theory ([1], Ch. 5, Sec. 1; we use the same notation) to the processes under consideration:

$$\sum_{m \neq n} P_{nm}(0) = -2\,\mathrm{Im}\,\mathcal{T}_{nn}. \tag{3.5}$$

We separate out from $\mathcal{T}_{nn}$ the factors corresponding to the quantization volumes of each of the absorbed and emitted particles. In this it is necessary to take account of the dimensionless character of the transition amplitude S_n calculated above, and therefore to write $\mathcal{T}_{nn}$ as

$$\mathcal{T}_{nn} = T_{nn} \prod_1^{2n} (\rho_{0i}' V_i)^{-1/2}. \tag{3.6}$$

The density ρ_0' introduced into (3.6) for each incident photon in order to satisfy the normalization and dimensionality can be defined as

$$\rho_0' = \frac{j_0'}{c} \approx 2 \int_{\omega_0 - \Gamma/2}^{\omega_0 + \Gamma/2} \frac{dk}{(2\pi)^3} = \frac{\omega^2 \Gamma}{\pi^2 c^3}. \tag{3.7}$$

With allowance for (3.6) and (3.7), the equality (3.5) can be rewritten for the reaction rates (1.4) in the form

$$\sum_{m \neq n} R_{nm} = -2(j_0/j_0')^n \,\mathrm{Im}\, T_{nn} \equiv 2(j/j_0')^n \,\mathrm{Re}(1 - S_n). \tag{3.8}$$

We shall examine the structure of the sum in (3.8); at the resonance frequencies, the only important terms in it are $R_{n,n+1}$ and $R_{n,n-1}$, which for large n are close. Consequently, taking (3.3) into account, we can write the approximate equality

$$R(\omega, j) \equiv \sum_0^\infty R_{n,n+1} \approx \sum \left(\frac{j}{j_0'}\right)^n \mathrm{Re}(1 - S_n)$$

$$= \frac{v' \cos 2\delta(\omega) - 1}{1 - 2v' \cos 2\delta + v'^2} + \frac{1}{1 - v'} \tag{3.9}$$

or, for $|\omega - \omega_0| \ll \Gamma/2$,

$$R(\omega_0, j) = 2(j/j_0')[1 - (j/j_0')]^{-2}. \tag{3.10}$$

The formulas (2.8) and (3.10) are very close, although different types of approximation were made in their derivation and, apart from this, the transition rate (3.10) takes account of chains of processes omitted in the calculation of (2.8).

4. PHYSICAL MEANING OF THE EXISTENCE OF A LIMITING PHOTON FLUX DENSITY

The presence of an essential singularity of R(j) at $j \to j_0$ has been demonstrated above. Mathematically, this divergence is caused by the fact that, close to resonance, the expansion parameter for the S-matrix is effectively different from $e^2/\hbar c$, since each term in the expansion contains within itself an infinite summation over the virtual interactions of the electron in the atom.

For a physical interpretation, we can approximate (2.8) by a Lorentz distribution with an "effective width" for the radiation flux:

$$R = f(\Gamma, \omega_0, j)\gamma[(\omega-\omega_0)^2+\gamma^2/4]^{-1}, \qquad (4.1)$$

$$\frac{\gamma^2}{4} = \frac{(\omega-\omega_0)^4+(\Gamma^2/4)^2(1-j/j_0)^2}{(\Gamma^2/2)(1-j/j_0)} \approx \frac{\Gamma^2}{8}\left(1-\frac{j}{j_0}\right). \qquad (4.2)$$

For $\omega = \omega_0$ and $j \to j_0$, we obtain $\gamma \to 0$, i.e., the radiation flux becomes ideally monochromatic and its density corresponds to the presence of one photon in each quantum state.

The question of the nature of the interactions when $j > j_0$ arises naturally here. It is physically obvious that these interactions cannot accelerate the stimulated emission process, which is occurring with probability unity, and must consist only of absorption and re-emission of a photon by an electron without a transition to a real level, i.e., they must introduce factors of the type (2.11) into the transition probability. During n resonance transitions, there will be, in all, $p = [(j/j_0 - 1)n]$ such non-resonance transitions with $k = 1$, and, therefore, we shall have $q = p/n = j/j_0 - 1$ asymptotically in (2.12). Hence we obtain for the reaction rate when $j > j_0$

$$R'(j) = R(j_0) + \pi\Gamma(j/j_0)\bar{v}(1-\bar{v})^{-2} \approx R(j_0), \qquad (4.3)$$

where

$$\bar{v} = (j/j_0)^{j/j_0}(\Gamma/4\omega_0)^{2(j/j_0-1)} = (j\tau_{NR}^{(1)}/j_0\tau_R^{(1)})^{j/j_0}(\tau_R^{(1)}/\tau_{NR}^{(1)}), \qquad (4.4)$$

$$\tau_{NR}^{(1)} = (\Gamma/2)[(\omega+\omega_0)^2+\Gamma^2/4]^{-1} \approx \Gamma/4\omega_0^2 \approx (\Gamma^2/8\omega_0^2)\tau_R^{(1)}, \qquad (4.5)$$

$\tau_{NR}^{(1)}$ is the duration of a non-resonance transition.

The relation (4.3) shows that for $j > j_0$ the function R(j) decreases slowly but monotonically with increase of j (this behavior of R(j) has been noted by a number of authors, e.g., in[11], for other processes when $j \to \infty$). Our analysis shows that these singularities are connected with the opening up of new reaction channels, which is thus connected not with increase in the energy of an individual particle being scattered but with increase of j[4] (saturation of the single-photon scattering at the upper level of the system when $j \sim j_1$ opens up the two-photon scattering channel, and so on; these singularities of R(j) are close to each other).

Comparison with (4.2) of the constancy of R(j) (from (4.3)) in a large part of the interval (j_0, j_1) shows that a superluminescence process occurs in matter when $j < j_0$, and generation when $j_0 < j < j_1$. This conclusion does not contradict experimental data, since, e.g., for a neodymium laser, $j_0 \sim 5 \times 10^{15}$ quanta/cm^{2}sec (the power $P_0 \sim 10^{-3}$ W/cm^2), and $j_1 \sim 8 \times 10^{30}$ quanta/cm^{2}sec ($P_1 \sim 2 \times 10^{12}$ W/cm^2 is much greater than any values obtained up to now).

We note that, if the flux interacts with the system for a period of time t, then, since the reaction rate and the width (4.2) have the form ($N = t/\tau \gg 1$)

$$R(j,t) \approx \pi\Gamma\sum_1^N nv^n, \quad \gamma^2(t) \approx \Gamma^2[N + j_0^2/(j_0^2 - j^2)]^{-1}, \qquad (4.6)$$

questions of convergence are not important; nevertheless, all the statements made above remain in force.

In conclusion, we remark that the quantum approach to resonance interactions of an intense photon beam with atomic electrons enables us to expose a number of features which it would be interesting to check experimentally.

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