Interaction of a single-photon wave packet with an excited atom

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The interaction of a single-photon wave packet with an initially excited two-level atom in free space is studied in semiclassical and quantum approaches. It is shown that the final state of the field does not contain doubly occupied modes. The process of the atom's transition to the ground state may be accelerated, decelerated, or even reversed by the incoming photon, depending on parameters. The spectrum of emitted radiation is close to the sum of the spectrum of the incoming single-photon wave packet and the natural line shape, with small and complicated deviations.

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I. INTRODUCTION

The main purpose of this paper is a calculation of the kinetics of population of atomic levels and of the spectrum of light emitted as the result of the interaction of a single-photon wave packet with an excited atom. The problem concerns an elementary process of interaction of light with matter that plays an important role in laser theory. The experiments with one-atom, one-photon setup can be traced back as far as 1985 [1]; with the techniques of today it seems possible to study the process in the experiment directly.

In Sec. II the simple classical model of the packet is introduced and used along with the quantum model of the atom to obtain the estimate of the difference of population induced by the photon. This difference comes to be very small (in typical situations) and can change its sign. In Sec. III the equations of a fully quantum model are derived. They are solved in the approximation that starts with the assumption of the negligibility of the photons' influence on the atomic evolution. The solutions, which are obtained in the form of quadratures, describe the kinetics of probabilities of the atomic states and the spectrum of the emitted radiation. In Sec. IV these data are visualized with the detailed study of a one-dimensional numerical example. The discussion of discrepancies between the semiclassical solution and the simplest approximation of the quantum approach is also given in this section. The established properties of the interaction of a single-photon wave packet with an initially excited two-level atom in free space are discussed in Sec. V in the context of the ongoing discussions of the nature of the stimulated emission. Section VI gives a summary of the obtained results.

II. SEMICLASSICAL APPROACH

The classical model that represents a photon (more precisely, a single-photon wave packet) as a limited in space pulse of the quasimonochromatic classical radiation is well known [2,3]. The studies of the spatial distribution of the energy of the electromagnetic field of a photon in the framework of quantum theory started long ago [4] and have been carried out actively in recent years [5–7].

From the energy density considerations one can equate the total energy of the field of the pulse to the energy of the photon $\hbar\omega$, where $\hbar$ is the Planck's constant and ω is the photon frequency [4,5]:

$$\frac{1}{8\pi}\int [\mathbf{E}(\mathbf{r},t)^2 + \mathbf{H}(\mathbf{r},t)^2]d\mathbf{r} = \hbar\omega. \quad (1)$$

Here $\mathbf{E}(\mathbf{r},t)$ and $\mathbf{H}(\mathbf{r},t)$ are the vectors of the electric and magnetic fields, respectively.

In this section we treat the linearly polarized pulse of the Gaussian shape, propagating in the positive direction of the OX axis:

$$\mathbf{E}(\mathbf{r},t) = \mathbf{e}\mathcal{E}_0 \exp\left[-\frac{(x-ct)^2}{4l^2}\right]\cos\left[k(x-\Lambda)-\omega t\right], \quad (2)$$

where $\mathbf{e}$ is the polarization vector, $\mathcal{E}_0$ is the pulse amplitude (maximal value of the field), l is the length of the pulse, Λ is the initial displacement of the packet's center, and c is the speed of light. We assume that the field is restricted in the transverse directions to some domain with a section area S; the exact form of the transverse structure will not be needed.

From Eq. (1) we have

$$\mathcal{E}_0 = (32\pi)^{1/4}\sqrt{\frac{\hbar\omega}{lS}}. \quad (3)$$

We study the action of this packet on a two-level atom, located at the origin of the coordinate system. The atom is represented by a two-level system with the transition frequency ω_0 and the dipole transition matrix element d along the field direction. The evolution of this system under the influence of the harmonic external field with the carrier frequency ω and the envelope $\mathcal{E}(t)$ [$\mathcal{E}_0 = \max\mathcal{E}(t)$] can be described by the equations for the components of the Bloch vector [in the rotating wave approximation (RWA) and in the rotating frame of reference], which have in general the following form [8]:

$$\dot{u} + \Gamma_2 u = -\Delta v, \quad (4)$$

$$\dot{v} + \Gamma_2 v = \Delta u + \Omega w, \quad (5)$$

$$\dot{w} + \Gamma_1(w - w_0) = -\Omega v. \quad (6)$$

Here u and v are the transverse components and the population difference w is the longitudinal component of the Bloch vector, w_0 is the population difference at equilibrium, Γ_2 and Γ_1 are the rates of the transverse and longitudinal relaxations,

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correspondingly, $\Delta = \omega - \omega_0$ is the frequency detuning, and the Rabi frequency is

$$\Omega(t) = \frac{d\mathcal{E}(t)}{\hbar}. \tag{7}$$

We limit ourselves to the resonant case $\Delta = 0$. In this case the system reduces to two last equations, which for our problem should be written as

$$\dot{v} + \gamma v = \Omega(t) w, \tag{8}$$

$$\dot{w} + 2\gamma (w + 1) = -\Omega(t) v. \tag{9}$$

We have taken into account that for the spontaneous emission $2\Gamma_2 = \Gamma_1 = 2\gamma$, and the equilibrium corresponds to the system being in a ground state (no thermal noise). The time-dependent Rabi frequency follows the pulse envelope Eq. (2), and its form can be written as a Gaussian,

$$\Omega(t) = \Omega_0 \exp\left[-\frac{(t-T)^2}{4\tau^2}\right], \tag{10}$$

where $\Omega_0 = \max \Omega(t)$, $\tau = lc$ is the pulse duration, and $T = -\Lambda c$ is the arrival time of the pulse peak to the point of location of the atom. The spectrum of this pulse has the width $\delta = (2\tau)^{-1}$. The initial conditions for Eqs. (8) and (9) are $v(0) = 0$ and $w(0) = 1$, that is, at the initial moment the atom is in the excited state.

To find the proper approximation we turn to the numerical estimates for a typical situation. For the atomic parameters we choose the transition frequency $\omega_0 = 3.54 \times 10^{15}\ \mathrm{s}^{-1}$ and the atomic unit of the dipole moment $d = ea_0 = 2.42 \times 10^{-18}$ CGS units. For these parameters the rate of the spontaneous emission is

$$\Gamma_1 = 2\gamma = \frac{4d^2\omega_0^3}{3\hbar c^3} = 1.34 \times 10^7\ \mathrm{s}^{-1}. \tag{11}$$

We take as the duration of the pulse $\tau = 1$ ns and as the area of the transverse section of the pulse $S = 5 \times 10^{-3}\ \mathrm{cm}^2$. Then for the maximal amplitude of the field we have $\mathcal{E}_0 = 1.58 \times 10^{-5}$ G, and for the maximal Rabi frequency we have $\Omega_0 = d\mathcal{E}_0/\hbar = 3.62 \times 10^4\ \mathrm{s}^{-1} = 2.70 \times 10^{-3}\Gamma_1$. The Rabi frequency is small in comparison with the relaxation rate, and the system is overdamped.

Since the influence of the pulse on the atom is small ($\Omega_0 \ll \gamma$), does not last long ($\gamma\tau \ll 1$), and commences around the arrival moment T, we replace in the right-hand side of Eq. (8) the function $w(t)$ by its value at the arrival time $w(T)$. Then for the change of the population difference under the influence of the pulse we have

$$\Delta w = -\frac{w(T)}{2}\left(\int_0^\infty \Omega(t)\, dt\right)^2. \tag{12}$$

The absence of γ in the right-hand side of Eq. (12) along with the presence of Ω may seem surprising, since we are dealing with the case $\gamma \gg \Omega$. However the longitudinal component $w \sim 1$ has no time to change its value during the pulse owing to relaxation, since $\gamma\tau \ll 1$. On the other side, before the pulse the transverse component $v = 0$, and during the pulse it increases its value up to $v \sim \Omega w \tau$. Thus the relaxation term in the left-hand side of Eq. (8) remains much smaller than that in the right-hand side, $\gamma v \ll \Omega w$, due to the same inequality $\gamma\tau \ll 1$.

Since the arrival time T considerably exceeds the pulse duration τ, we can shift the lower limit in the integral in Eq. (12) to $-\infty$. Then we find that during a short interval around the arrival time the incoming pulse changes the rate of the transition of the atom from the excited to the ground state, and this rapid change results in an induced shift of the probability of the excited state by $\Delta P_+ = \Delta w/2$, or

$$\Delta P_+ = -\pi\left(\frac{d\mathcal{E}_0 l}{\hbar c}\right)^2 w(T). \tag{13}$$

This quantity can serve as a measure of influence of the irradiation by one photon on the process of spontaneous emission.

By using Eqs. (3) and (11), Eq. (13) could be cast into the form

$$\Delta P_+ = -\sqrt{\frac{\pi}{2}}\left(\frac{\sigma_0}{S}\right)\left(\frac{\gamma}{\delta}\right) w(T), \tag{14}$$

where

$$\sigma_0 = \frac{3}{2\pi}\lambda_0^2 \tag{15}$$

is the maximal cross section of the resonant fluorescence of the two-level atom ($\lambda_0 = 2\pi c/\omega_0$ is the wavelength of the resonant radiation).

Equation (14) shows that the change in the populations of a two-level atom under the influence of a pulse of the classical electromagnetic field that is equivalent to a single-photon wave packet is proportional to the fraction of the transverse section of the pulse that is covered by the cross section of the resonance fluorescence and to the fraction of the spectral density of the incoming radiation that gets into the band of the resonant interaction, that is, the natural linewidth. This result intuitively seems obvious.

With the parameters chosen above we have $\sigma_0 = 1.35 \times 10^{-9}\ \mathrm{cm}^2$, $S = 5 \times 10^{-3}\ \mathrm{cm}^2$, $\gamma = 6.70 \times 10^6\ \mathrm{s}^{-1}$, $\delta = 5 \times 10^8\ \mathrm{s}^{-1}$, and $\max|\Delta P_+| = 4.53 \times 10^{-9}$, a very small quantity.

III. QUANTUM APPROACH

Now we turn to study the interaction of the atom with the one-photon packet of the quantized electromagnetic field. For the atom we use the same two-level model with the excited state $|+\rangle$ and the ground state $|-\rangle$, which are connected by the electrical dipole transition with the matrix element of the dipole moment $\mathbf{d}$, and the atomic transition frequency ω_0. The electromagnetic field is described by the modes of the quantization cube, an imaginary cube with the edge length L with conditions of periodicity imposed on the field on its faces [9]. The mode μ is characterized by its wave vector $\mathbf{k}_\mu$ and its polarization vector $\mathbf{e}_\mu$, which obey the condition $\mathbf{k}_\mu \mathbf{e}_\mu = 0$, and the mode frequency is $\omega_\mu = c\left|\mathbf{k}_\mu\right|$.

The Hamiltonian of the system we take in the form

$$\hat{H} = \frac{\hbar\omega_0}{2}\hat{\sigma}_z + \sum_\mu \hbar\omega_\mu \hat{a}_\mu^\dagger \hat{a}_\mu + i\hbar\sum_\mu g_\mu(\hat{\sigma}_+\hat{a}_\mu - \hat{a}_\mu^\dagger\hat{\sigma}_-), \tag{16}$$

where $\hat{\sigma}_i$ are the Pauli matrices, $\hat{\sigma}_\pm = \left(\hat{\sigma}_x \pm i\hat{\sigma}_y\right)/2$, and $\hat{a}_\mu^\dagger$ and $\hat{a}_\mu$ are the operators of creation and annihilation of

photons in the mode μ. Three terms in Eq. (16) represent the Hamiltonian of the atom $\hat{H}_a$, the Hamiltonian of the field $\hat{H}_f$, and the interaction term $\hat{V}$, correspondingly. The interaction parameter

$$g_\mu = -\sqrt{\frac{2\pi\omega_\mu}{\hbar\mathcal{V}}}\mathbf{d}\mathbf{e}_\mu, \tag{17}$$

where $\mathcal{V} = L^3$ is the volume of the quantization cube. The structure of the interaction term assumes the RWA.

The state vector of the system can be expanded as

$$|\Psi(t)\rangle = \sum_\mu a_\mu|+\rangle|1_\mu\rangle + \sum_\mu b_\mu|-\rangle|2_\mu\rangle + \frac{1}{2}\sum_{\mu,\nu} c_{\mu\nu}|-\rangle|1_\mu\rangle|1_\nu\rangle, \tag{18}$$

where $|N_\mu\rangle$ is the N-photon Fock state of the mode μ. The indices in the last sum must not take the equal values. The vector $|\Psi\rangle$ is normalized by the condition

$$\|\Psi\|^2 = \sum_\mu |a_\mu|^2 + \sum_\mu |b_\mu|^2 + \frac{1}{2}\sum_{\mu,\nu}|c_{\mu\nu}|^2 = 1. \tag{19}$$

The vectors of the basis are taken in the interaction picture: $|+(t)\rangle = |+(0)\rangle \exp -i(\omega_0 t/2)$, $|-(t)\rangle = |-(0)\rangle \exp i(\omega_0 t/2)$, $|1_\mu(t)\rangle = |1_\mu(0)\rangle \exp -i\omega_\mu t$, etc. After the substitution of the expansion Eq. (18) in the Schrodinger equation and projection on the basis of the initial vectors ($\langle+(0)|\langle 1_\mu(0)|$, etc.), we obtain the system of equations for the probability amplitudes

$$\frac{da_\mu}{dt} = -\sqrt{2}g_\mu b_\mu e^{i\Delta_\mu t} - \sum_\nu g_\nu c_{\mu\nu} e^{i\Delta_\nu t}, \tag{20}$$

$$\frac{db_\mu}{dt} = \sqrt{2}g_\mu a_\mu e^{-i\Delta_\mu t}, \tag{21}$$

$$\frac{dc_{\mu\nu}}{dt} = g_\mu a_\nu e^{-i\Delta_\mu t} + g_\nu a_\mu e^{-i\Delta_\nu t}. \tag{22}$$

Here the frequency detuning is the difference between the transition frequency and the mode frequency, $\Delta_\alpha = \omega_0 - \omega_\alpha$.

We take the initial state of the field in the form of the normalized single-photon wave packet

$$|\Phi(0)\rangle = \sum_\mu \phi_\mu|1_\mu\rangle. \tag{23}$$

Then the initial conditions for the system Eqs. (20)–(22) are

$$a_\mu(0) = \phi_\mu, \quad b_\mu(0) = 0, \quad c_{\mu\nu}(0) = 0. \tag{24}$$

From the results of the previous section we can conclude that for the typical values of parameters the influence of the incoming photon on the process of the spontaneous emission will be very small and the process will mainly evolve in the same way as in the unperturbed atom. Then, following the approach of Weisskopf and Wigner [10], we can assume that all amplitudes of the one-photon states decrease with time by the same exponential law with the rate $\gamma = \Gamma/2$, which is one-half of the rate of the spontaneous decay given by Eq. (11):

$$a_\mu(t) = \phi_\mu \exp(-\gamma t). \tag{25}$$

By substitution of the ansatz Eq. (25) in Eq. (21) and the integration with the initial conditions Eq. (24) we have

$$b_\mu(t) = \sqrt{2}g_\mu\phi_\mu\frac{1 - e^{-\gamma t - i\Delta_\mu t}}{\gamma + i\Delta_\mu}. \tag{26}$$

Analogously, from Eq. (22) we have

$$c_{\mu\nu}(t) = g_\mu\phi_\nu\frac{1 - e^{-\gamma t - i\Delta_\mu t}}{\gamma + i\Delta_\mu} + g_\nu\phi_\mu\frac{1 - e^{-\gamma t - i\Delta_\nu t}}{\gamma + i\Delta_\nu}. \tag{27}$$

In the following we refer to Eqs. (25), (26), and (27) as the basic approximation.

The values of the initial amplitudes depend on the quantization volume. This dependence can be seen from the normalization condition Eq. (19). For simplification we temporarily ignore the polarization and consider that the value of $\mathbf{k}_\mu$ defines completely the mode μ. Let the initial state of the packet be given by the probability amplitude in the k space $\psi(\mathbf{k})$ that is normalized by the condition

$$\int |\psi(\mathbf{k})|^2 d\mathbf{k} = 1. \tag{28}$$

For large enough volume $\mathcal{V}$ the summation over μ could be replaced by the integration over $\mathbf{k}$:

$$\sum_\mu |\phi_\mu|^2 = \sum_\mu |\phi(\mathbf{k}_\mu)|^2 = \int |\phi(\mathbf{k})|^2\rho(\mathbf{k})d\mathbf{k}, \tag{29}$$

where $\rho(\mathbf{k})$ is the density of permissible discrete values of the vector $\mathbf{k}_\mu$ in the k space,

$$\rho(\mathbf{k}) = \frac{\mathcal{V}}{(2\pi)^3}. \tag{30}$$

Thus from comparison of Eqs. (28) and (29) the discrete amplitudes will have the form

$$\phi_\mu = \sqrt{\frac{8\pi^3}{\mathcal{V}}}\psi(\mathbf{k}_\mu). \tag{31}$$

The summary probability of the doubly occupied field modes,

$$P_2 = \sum_\mu |b_\mu|^2 \propto g^2\phi^2\rho \propto \mathcal{V}^{-1}, \tag{32}$$

in the limit $\mathcal{V} \to \infty$ turns to zero. Thus the probability of creation of the doubly occupied state in the process of interaction of a single-photon wave packet with the excited atom *in the free space* is zero. This result is quite general: it is not limited by the applicability of the ansatz Eq. (25), since one can substitute Eqs. (26) and (27) into Eq. (20), find a new approximation for a_μ, and continue the iterations, and on every step the scaling relation Eq. (32) will hold.

The summary probability of pairs of singly occupied states,

$$P_{11} = \sum_{\mu,\nu} |c_{\mu,\nu}|^2 \propto g^2\phi^2\rho^2 \propto \mathcal{V}^0, \tag{33}$$

does not depend on the volume of quantization and thus gives a finite value for the process in a free space.

IV. NUMERICAL EXAMPLE

In principle the expressions Eqs. (26) and (27) along with the conversion rule Eq. (31) present the approximate answer to

the problem of evolution of the system in quadratures. However the numerical integration over two copies of the k space in the three-dimensional case happens to be too demanding on the computer resources. To demonstrate the properties of the solution we limit ourselves by a one-dimensional case that still has some direct physical interest.

Let's assume that the field is located on the interval from $-L/2$ to $L/2$ with periodic boundary conditions. In the following we use the system of units in which the average wave number of the incoming packet $K = 1$; we also take $\hbar = 1$ and $c = 1$. We replace the interaction parameters g_μ by the constant value g; that is relatively unimportant since only components around the transition frequency take active part in the interaction. We also limit ourselves to a single polarization. The connection between the interaction constant g and the decay rate γ follows from the Fermi golden rule: $\gamma = g^2 L$.

We take the packet of the Gaussian form

$$\phi_\mu = \phi(k_\mu) = \frac{(2\pi)^{14}}{\sqrt{\kappa L}} \exp\left[-\frac{(k_\mu - 1)^2}{4\kappa^2} - ik_\mu \Lambda\right]. \quad (34)$$

From Eqs. (27) and (34), using the one-dimensional analogs of Eqs. (30) and (31), we obtain the expression for the time-dependent probability density of photons in the k space:

$$C(k_1,k_2;t) = \frac{1}{\sqrt{8\pi^3}} \frac{\gamma}{\kappa} |\xi(k_1)\chi(k_2,t) + \xi(k_2)\chi(k_1,t)|^2, \quad (35)$$

where

$$\xi(z) = \exp\left[-\frac{(z-1)^2}{4\kappa^2} - iz\Lambda\right], \quad (36)$$

$$\chi(z,t) = \frac{1 - \exp[-\gamma t - i(1-|z|)t]}{\gamma + i(1-|z|)}. \quad (37)$$

The total probability of finding the atom in the ground state and two photons in the field is given by the twofold integral,

$$P_-(t) = \frac{1}{2}\int\int C(k_1,k_2;t)dk_1 dk_2, \quad (38)$$

where the integration is carried out over all values of k_1 and k_2. In the basic approximation we used the estimate $P_+(t) = 1 - P_-(t)$.

For the numerical calculations we choose the values of parameters $\gamma = 0.0125$ and $\kappa = 0.25$. This value of the wave-number distribution width corresponds to the single-photon packet with the spatial length $l = \kappa^{-1} = 4$ and the spectral width $\delta = c\kappa = 20\gamma$. For the dependence $P_+(t)$ with a given value of T we define formally the induced probability shift $\Delta P_+(T)$ as the value of the function $f(t) = P_+(t) - \exp(-2\gamma t)$ taken at the point where $|f(t)|$ has the absolute maximum. This function behaves discontinuously, but the rupture is purely formal: the acceleration of the transition smoothly turns into deceleration. If $0 < w(T) \ll 1$, then the first part of the photon accelerates the decay, whereas the second decelerates it.

The numerically found dependence of $P_-(t)$ is shown in Fig. 1. For the negative displacement $\Lambda = -20$ around the arrival time $T = 20$, the growth of the probability P_- accelerates for a short time in accordance with the semiclassical description given in Sec. II. For comparison the results for the positive displacement $\Lambda = 20$ are shown. In the latter case the packet passed the atom before it became excited and never hit it. Naturally, the dependence shows the behavior appropriate to the spontaneous emission of the unperturbed atom. The vertical distance between the curves in the right part of the graph in Fig. 1 represents the induced probability shift; its calculated value is $\Delta P_+(20) = -\Delta P_-(20) = -0.076$.

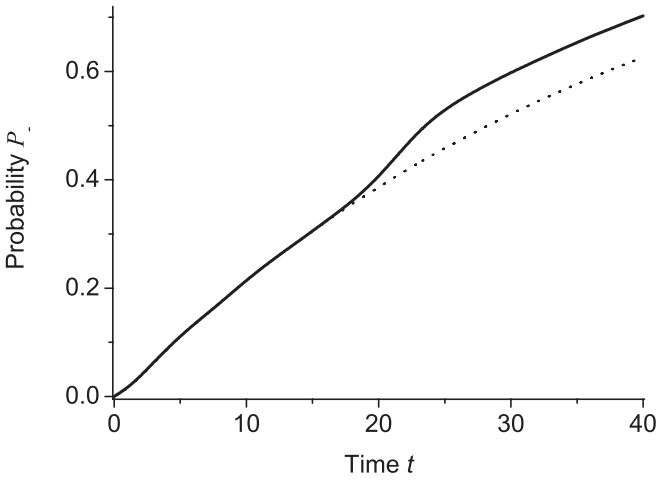

FIG. 1. The dependence of the probability P_- of finding the atom in the ground state on time t [in units of $(Kc)^{-1}$] for values of the initial displacement (in units of K^{-1}) $\Lambda = -20$ (solid line) and $\Lambda = 20$ (dash-dotted line).

The time-dependent rate of the downward transition can be defined as

$$\Gamma(t) = -\frac{1}{P_+}\frac{dP_+}{dt}, \quad (39)$$

where $P_+(t)$ is the probability of finding the atom in the excited state. The behavior of $\Gamma(t)$ is shown in Fig. 2. The maximum of the decay rate is reached at $t = 21.6$, which is somewhat later than the arrival time $T = 20$. The maximal value of Γ exceeds the rate of transition due to the spontaneous emission by the factor 1.94.

If we apply the semiclassical approximation presented in Sec. I, then for the one-dimensional case [cf. Eq. (12)] we obtain for the induced shift of the probability

$$\Delta P_+ = -\sqrt{2\pi}\frac{\gamma}{\delta} w(T). \quad (40)$$

In our numerical example this formula yields $\Delta P_+(20) = -0.027$, which is about one-third of the value given by the quantum calculation. What is more important is that Eq. (40) predicts the change of sign of the induced probability shift for $T \geqslant (\ln 2/2)\gamma^{-1}$, that is, slowing down of the process of spontaneous radiation. The quantum calculation shows that the incoming photon always accelerates the downward transition irrespectively of the sign of the population difference $w(T)$; instead of Eq. (40) it yields

$$\Delta P_+ = -\sqrt{2\pi}\frac{\gamma}{\delta} P_+(T). \quad (41)$$

The reason for the proportionality of the quantum value of ΔP_+ to the population of the upper state at the time of the arrival $P_+(T)$ is clear: with the ansatz Eq. (25) the behavior

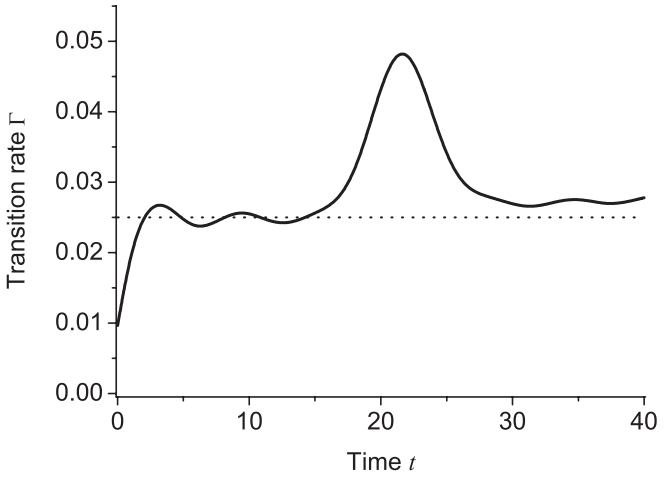

FIG. 2. The dependence of the temporal rate of the downward transition Γ (in units of Kc), defined by Eq. (39), on time t [in units of $(Kc)^{-1}$] for the initial displacement (in units K^{-1}) $\Lambda = -20$ (solid line). The dashed line shows the value of $\Gamma = 2\gamma = 0.025$ for the unperturbed atom.

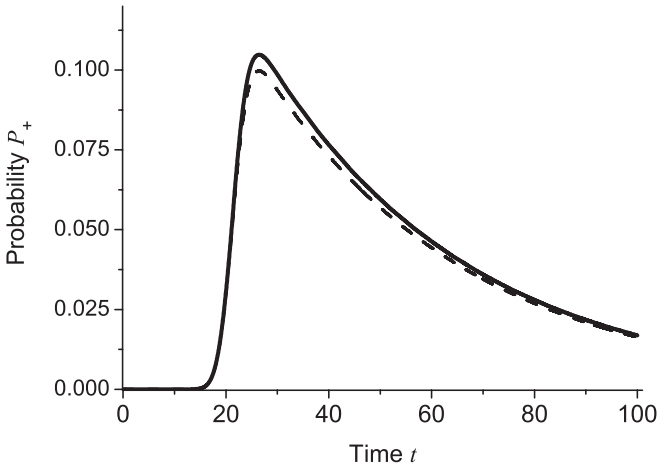

FIG. 3. The dependence of the probability of the excited state P_+ on time t [in units of $(Kc)^{-1}$] for the case of the one-photon packet Eq. (34) with the initial displacement (in units of K^{-1}) $\Lambda = -20$ scattered on the atom in the ground state. Solid line: Quantum model, numerical integration of the system Eq. (43). Dashed line: Semiclassical model, numerical integration of Eqs. (8) and (9).

of the amplitudes $a_\mu(t)$ does not depend on the parameters of the wave packet at all. The choice of positive T just uniformly decreases the values of all amplitudes by multiplying them by the factor $\exp(-\gamma T)$.

At this point we have to admit the superiority of the semiclassical theory over the simplest approximate solution of the quantum model. Let's consider the case $2\gamma T \gg 1$, in which the packet arrives at the atom, when it is almost exactly in the ground state. In this case the action of the photon on the atom will not depend on T. The incoming photon will excite the atom with some probability $\Delta P_+(\infty)$ (just because there is no other way of evolution from the state with $w = -1$), then the process of spontaneous emission will be renewed from the initial value $\Delta P_+(\infty)$. This qualitative picture is in agreement with the semiclassical result Eq. (40).

The quantity $\Delta P_+(\infty)$ can be calculated from a purely quantum model. Taking the expansion of the state vector in the form

$$|\Psi(t)\rangle = A|+\rangle|\mathrm{vac}\rangle + \sum_\mu B_\mu|-\rangle|1_\mu\rangle \tag{42}$$

and its substitution in the Schrodinger equation with the Hamiltonian Eq. (16) leads to the system of equations for the probability amplitudes

$$\frac{dA}{dt} = -g\sum_\mu B_\mu e^{i\Delta_\mu t}, \qquad \frac{dB_\mu}{dt} = gAe^{-i\Delta_\mu t}. \tag{43}$$

The numerical solution of this system with the chosen values of κ and γ has been carried out for the length of the interval of quantization $L = 251.32$ with the account of $\mathcal{N} = 159$ modes with minimal $|k_\mu|$. When tested, this approximation has produced the value of the spontaneous decay rate that differed from the theoretical value by less than 1%. The calculated dependence of $P_+(t) = |A(t)|^2$ is shown in Fig. 3. This calculation is direct and does not depend on the ansatz Eq. (25).

The numerically found quantum value $\Delta P_+(\infty) = 0.105$ is quite close to the semiclassical value $\Delta P_+(\infty) = 0.125$ that follows from Eq. (40). The numerical integration of the Bloch Eqs. (8) and (9) gives even a better agreement with the quantum theory: then $\Delta P_+(\infty) = 0.100$. The functions $P_+(t)$ calculated in semiclassical and quantum models are compared in Fig. 3.

The constancy of the negative sign of $\Delta P_+(T)$ that follows from the simplest approximate quantum solution can be improved by the iteration process. By substitution of the solution Eq. (27) in Eq. (20) [the first term can be neglected in accordance with Eq. (32)], we obtain the formula for the amplitudes in the approximation of the first order,

$$a(k_\mu,t) = \phi(k_\mu) - \frac{\gamma}{2\pi}\int_{-\infty}^{k_\mu} F(k_\mu,k_\nu;t)\,dk_\nu, \tag{44}$$

where $\phi(z)$ is given by Eq. (34), and

$$\begin{aligned} F(x,y;t) &= \frac{\phi(x)}{\gamma + i(1-|x|)}\{G[i(1-|y|)] - G[-\gamma + i(|x|-|y|)]\} \\ &\quad + \frac{\phi(y)}{\gamma + i(1-|y|)}\{G[i(1-|y|)] - G[-\gamma]\}, \end{aligned} \tag{45}$$

$$G(z) = \frac{e^z - 1}{z}. \tag{46}$$

These expressions permit one to calculate directly the kinetics of the probability of the excited state:

$$P_+(t) = \frac{L}{2\pi}\int_{-\infty}^{\infty} |a(k,t)|^2 dk. \tag{47}$$

In the first approximation $\Delta P_+(20) = -0.059$, which is closer to the semiclassical value than the result of the zeroth approximation. The value of the ΔP_+ calculated from the expression Eq. (47) for large T becomes positive [e.g., $\Delta P_+(130) = 3 \times 10^{-3}$], although it remains much smaller

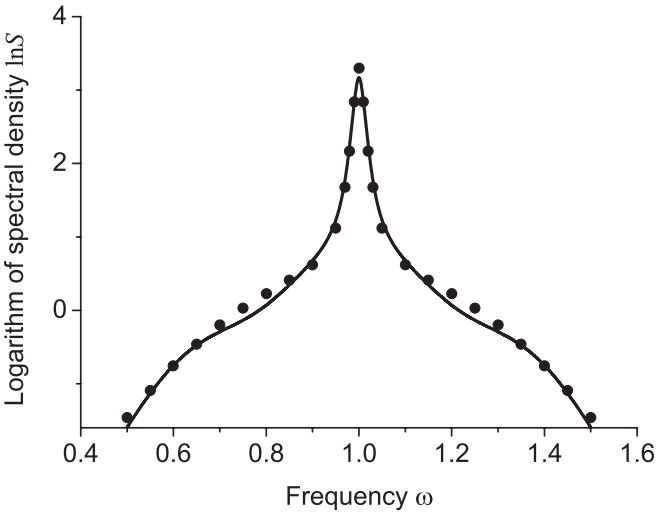

FIG. 4. The logarithm of the spectral density as a function of frequency ω (in units of Kc). The solid line shows the calculated numerically values of S for the initial displacement (in units of K^{-1}) $\Lambda = -20$. The dots shows the summary spectrum S_0.

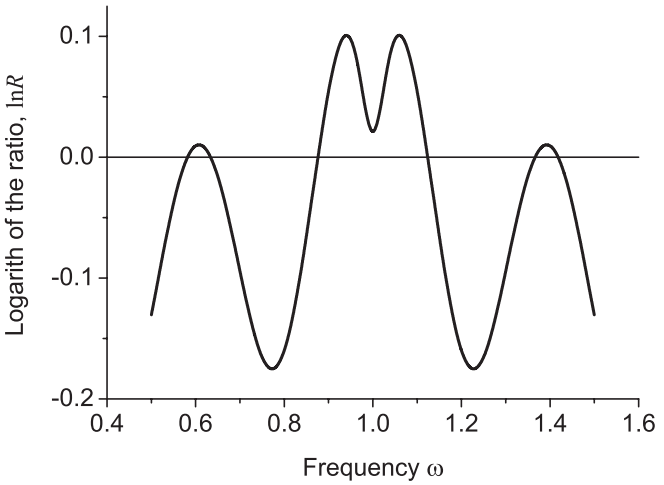

FIG. 5. The logarithm of the ratio of the real spectral density [found numerically for the initial displacement (in units of K^{-1})] to the summary one, $R = S/S_0$, as a function of frequency ω (in units of Kc).

than the numerically found limiting value $\Delta P_+(\infty) = 0.105$. One may hope that the higher approximations will improve the agreement.

Now we discuss the spectral properties of the photons in the final state. The spectral density of the final photons $S(\omega)$ can be expressed through the limiting value of the double k-space density given by Eq. (35), $C_f(x,y) = C(x,y;\infty)$. The expression has the form

$$S(\omega) = \int_0^\infty [C_f(\omega,\omega') + C_f(-\omega,\omega') + C_f(\omega,-\omega') + C_f(-\omega,-\omega')]d\omega'. \quad (48)$$

Since the interaction of the photon with the atom is weak, it is natural to assume that the spectrum is close to the sum of two relevant spectra, for $\delta \gg \gamma$—the wide Gaussian form of the incoming packet and the narrow Lorentzian form of the spontaneous radiation of the isolated atom,

$$S_0(\omega) = \frac{1}{\sqrt{2\pi}\delta}\exp\left[-\frac{(\omega-1)^2}{2\delta^2}\right] + \frac{1}{\pi}\frac{\gamma}{\gamma^2+(1-\omega)^2}. \quad (49)$$

We call this expression the summary spectrum. The numerical calculations show that the final spectrum of photons is very close to this form indeed (see Fig. 4).

It is difficult from this picture to see the deviations of the real spectrum from the summary one that are created by the interaction of the single-photon wave packet with the excited atom. The ratio of spectral densities that is shown in Fig. 5 is more instructive.

The interaction somewhere increases the spectral density and somewhere decreases it. We define the spectral with $\Delta\omega$ as a minimal interval that contains half of the total spectral density. With this definition we find that the net effect of the interaction produces small, 11%, broadening of the spectrum.

V. DISCUSSION

Einstein in his groundbreaking papers of 1916 and 1917 [11–13] treated both radiation induced processes—that of absorption and that of acceleration of the downward transition—on equal footing. He named them "Zustandsänderungen durch Einstrahlung," changes of state due to irradiation. The specific term for the downward process, "negative Einstrahlung," negative irradiation, came in use by 1923 [14]. In 1924 Tolman [15] introduced the term "negative absorption," and in the same year van Vleck launched the parallel term "induced emission" [16]. Finally, the predominant at the present time term "stimulated emission" was brought in by Dirac [17]. This chain of renamings has shifted the focus of attention from the evolution of the atomic (molecular) system to that of the radiation. Nevertheless, it is the acceleration of the downward transition in the atomic system induced by irradiation that remains the determinative indicator of the stimulated emission.

Dirac established the theory of interaction of atoms with the quantized electromagnetic field and derived Einstein's phenomenological equations from this theory [17]. In the course of the derivation it became clear, that the specific property of the operator of creation of photons,

$$\hat{a}^\dagger|N\rangle = \sqrt{N+1}|N+1\rangle, \quad (50)$$

is responsible for the acceleration. Dirac used the approximation that is known now as the Fermi golden rule: the rate of the atomic transition is proportional to the square of modulus of the matrix elements summed over different modes. On the other hand, Eq. (50) can be interpreted as a statement that the stimulated photons are radiated to the same mode (definitely with the same frequency ω_μ, and if the modes are taken in the form of traveling plane waves—then with the same wave vector $\mathbf{k}_\mu$ and the polarization $\mathbf{e}_\mu$), making the exact copies of the initial, stimulating photon(s).

In Dirac's approximation the acceleration of the transition, that is, the stimulated emission, is due to the process of emitting photons into the occupied modes, which can be interpreted as a process of radiation of exact copies of the initial photons. In the

course of history the fine print was frequently neglected, and the previous (correct) statement has been sometimes reduced to assertion that "stimulated emission is a process of radiation of exact copies of the initial photons." This statement may be still correct, if we make it a definition and apply the term "stimulated emission" only to vectors of multiply occupied modes. Some authors do that [18,19]. But one must remember that this definition is different from the historically developed one.

Acceleration of the transition can occur without copying of photons. Drobny, Havukainen, and Buzek [7] studied the scattering of a single-photon wave packet on an atom in the *ground* state. In this situation all radiation is emitted into vacant modes, since in the RWA, which was used by the authors, with the given initial conditions the creation of the doubly occupied mode is strictly forbidden. However, the temporal rate of the transition from the upper state [see Eq. (39)] in their studies reached values about an order of magnitude larger than that of the spontaneous emission of the unperturbed excited atom. In the problem studied in the present paper, that of scattering of a single-photon wave packet on an atom in the *excited* state, the situation is similar: the acceleration may be present (see Fig. 2), whereas the copying is absent [see Eq. (32)], albeit not due to the limitations of the RWA.

The idea of the universal identity of the acceleration of the atomic transition by the radiation field and the emission of photons in the occupied modes is so widespread and deeply rooted that the relation Eq. (32) is sometimes considered as a threat to the operability of lasers. Therefore some additional explanations are due.

The decrease of the energy of the system under the influence of the radiation is possible even in *classical* models. This fact served as a guiding analogy to Einstein in his construction of the kinetic equations for quantum systems, and it is properly noted in his papers [11–13]. The classical theory of the stimulated emission has been developed for a long time (see, e.g., Refs. [20–22]).

There are several *semiclassical* theories of lasers and masers that describe the radiating atoms by quantum models with the discrete energy spectrum and use the classical description of the electromagnetic field. The examples are the perturbation-theory-based approach used by Gordon, Zeiger, and Townes [23], the rate equations derived by Statz and deMars [24], and Lamb's theory of an optical maser [25]. These theories do not use either the model of the quantized electromagnetic field or Dirac's theory. Consequently, they are completely independent from the presence or absence of the multiply occupied modes of the electromagnetic field—but still they are theories of maser and laser operation that have proven themselves to be viable and useful.

Our results have no direct influence on the theory of lasers, even those without the resonators [26]. The model of the ensemble of separate noninteracting atoms at rest (the distance between the atoms must be much larger than the wavelength of the resonant radiation λ_0) that are interacting with the sparse ensemble of nonoverlapping photons (for the numerical values chosen in Sec. II the intensity must be much less than $10^{-7}\,\mathrm{W\,cm^{-2}}$), this model, to which the results of the calculations given in previous sections could be applied more or less straightforwardly, does not seem to be adequate for a real laser.

However, the results presented above could be verified experimentally. A single atom in a trap could be excited by a short π pulse right before the arrival of a single-photon wave packet. The most favorable conditions for spectral studies correspond to the case in which the spectral width of the packet δ is somewhat larger than the natural linewidth γ, but not too much (for instance, $\delta = 20\gamma$ as in the numerical example of Sec. IV). Then the accuracy of measurement of the spectral density of about 1% will be enough to observe the deviations from the summary spectrum shown in Fig. 4. Furthermore, at present the methods of experimental study of multiple occupancy of modes are known [27], and the absence of the multiply occupied modes could be confirmed directly.

The problem solved in this paper may be of some interest for studies of the cloning of photons [18,28], especially of their nonpolarizational degrees of freedom.

Finally we mention the problem of the radiation of two adjacent atoms separated by a distance that is much smaller than (or comparable to) the radiation wavelength. The situation in which both atoms are initially excited is somewhat similar to the problem studied in this paper: the radiation of an atom occurs under the influence of the electromagnetic field created by its neighbor, akin to a photon coupled to its source. The influence of the adjacent atom on the kinetics of radiation is strong: the energy decay rate of the system, defined by the logarithmic derivative analogously to Eq. (39), tends to zero for $t \to 0$ and for $t \to \infty$ tends to $2\Gamma_1$, the double rate of the single atom radiation decay (see, e.g., Ref. [29]). The nature of this radiation was described in different ways. In the pioneering paper by Dicke [30] it was defined as a "spontaneous coherent radiation." Ernst and Stehle [31] have found that the properties of this radiation "cannot be explained in conventional terms of spontaneous or stimulated emission of radiation." Lehmberg [29] concluded that the acceleration of the decay "can be regarded, at least in part, as due to stimulated emission." The kinetic definition of the stimulated emission, which we gave at the beginning of this section, makes the last point of view preferable.

Despite the active studies of many aspects of the problem of two adjacent radiating atoms carried out for several decades, the question about the weight P_2 of the doubly occupied modes in the final state of the radiation field nonetheless lacks a definite answer. Still the opinion that for a small number of neighboring radiating atoms the contribution of the doubly occupied modes is negligible can be found in the literature [32]. The calculation of P_2 remains as an interesting problem for further studies.

VI. CONCLUSION

The main results of this paper describe the kinetics of probabilities of the excited atom *in the free space* under the influence of the incoming photon and the power spectrum of the final state of the radiation field. The photon is represented by a single-photon wave packet with a finite spectral width δ and a variable time of arrival of the center of the packet to the atom T.

The atom is described by the model of the two-level system, which at the initial moment is turned into the excited state. For the description of the photon two models are used.

The first is a classical one in which the packet is described by the quasimonochromatic pulse of the radiation with the carrier frequency equal to that of the atomic transition ω_0, and the total energy of the pulse equals that of the photon [see Eq. (1)]. The second approach is based on Dirac's theory of interaction of the atomic system with the quantized electromagnetic field, where the packet is described by the initial set of probability amplitudes in the space of wave vectors.

Both approaches agree in their results, which shows that the photon with the bandwidth δ much larger than the half-rate of the spontaneous emission γ ($\delta \gg \gamma$) influences the kinetics of the atomic transition only in a short interval of time with the duration $\tau \sim \delta^{-1}$ around the arrival time T, and the amount of this influence, measured by the change of the probability of the excited state P_+ during the pulse duration, is small, $\Delta P_+ \propto \gamma/\delta$. If at the arrival time the atom is in the initial stage of the spontaneous emission ($\gamma T \ll 1$), then the photon accelerates its transition to the ground state (see Figs. 1 and 2), $\Delta P_+ < 0$. In the opposite case, when the spontaneous decay is nearly completed by the time of arrival ($\gamma T \gg 1$), the photon for a short time reverses the direction of the evolution of the P_+ ($\Delta P_+ > 0$), then the spontaneous emission starts anew (see Fig. 3).

Since the influence of the photon on the atomic evolution is small, the spectral distribution of the photons in the final state is close to the sum of the spectrum of the initial single-photon wave packet and the Lorentzian natural line shape of the spontaneous emission [see Eq. (49) and Fig. 4]. The deviations of the spectral distribution, which are due to the interaction of the incoming photon with the atom, from this summary spectrum have a complicated form (see Fig. 5); they are relatively small but could be observed if the parameter $\gamma\delta$ is not too small.

Finally, in the frame of the quantum model it is shown that the final state of the radiation field consists of the vectors of pairs of singly occupied modes, $|\Psi_f\rangle = \sum c_{\mu\nu}|1_\mu\rangle|1_\nu\rangle$, and the contribution of the doubly occupied modes, $\sum b_\mu|2_\mu\rangle$ to the final state of the radiation field of the excited atom *in the free space* under the influence of a single-photon wave packet is zero. This result demonstrates that the acceleration of the atomic transition is not necessarily due to the emission of photons into the occupied modes. This fact was established earlier in a different physical situation [7]. It does not threaten the operability of masers and lasers, since several semiclassical theories of these devices, which reflect the influence of the radiation field on the rate of the transition, but make no notion on the multiple occupancy of modes of the quantized field, are known as successful.

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