Nonlocal effects in single-photon superradiance

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We consider influence of nonlocal (retardation) effects caused by the finite value of the speed of light on collective emission of a single photon by atomic ensembles. Using a fully quantum mechanical description of light and atoms we obtain an evolution equation for the atomic system that takes into account retardation. We found an exact analytical solution of this equation for the atomic slab geometry that yields insight on how the crossover between local and nonlocal dynamics occurs. In particular, it shows that initially nonlocal evolution, accompanied by collective oscillations of atomic population, becomes local at large time. In addition, atomic excitation in some parts of the sample rises above its initial value. We propose an experiment in which the transition between local and nonlocal regimes can be observed by increasing the size of the atomic sample or atomic density.

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I. INTRODUCTION

Collective spontaneous emission from a cloud of N atoms has been a subject of long-standing interest since the 1954 pioneering work of Dicke [1]. If an atom is excited, it can spontaneously emit a photon and go to the ground state. Such single-atom spontaneous emission usually occurs on a nanosecond time scale. However, if there are N atoms confined in a small volume, they can spontaneously emit light N times faster than an isolated atom. Such radiation speed-up is known as superradiance. Correlation between atoms is the key issue here. Thermally excited atoms emit light randomly, and the emitted intensity goes as the number of atoms N. However, when atoms are coherently radiating in phase with each other, the net field is proportional to N, and the emitted intensity goes as N^2. As a result, the system radiates its energy N times faster than a single atom.

Radiation speed-up occurs even if one photon is stored in the atomic cloud (it is shared among many atoms), which prepares a system in an entangled state with no macroscopic dipole moment [2]. Recent studies focus on collective and virtual effects in such systems [3–16]. Cooperative emission can provide insights into quantum electrodynamics and is important for various applications of the entangled atomic ensembles and generated quantum states of light for optical quantum-state storage [17], quantum cryptography [18,19], quantum communication [9,20,21], and quantum information [9,11].

Virtual transitions are fascinating feature of quantum electrodynamics. An atom can jump into an excited state and a virtual photon is emitted, then the atom quickly jumps back to the ground state and absorbs a photon. Such processes do not conserve energy and occur on a time scale governed by the uncertainty principle. Virtual transitions have real effects—they shift energy levels of emitting atoms, which is known as the Lamb shift. In 1947 Willis Lamb together with his graduate student Robert Retherford measured splitting between the $2S_{1/2}$ and $2P_{1/2}$ states of the hydrogen atom caused by virtual transitions [22]. The measured value of the level splitting (1058 MHz) has provided solid experimental foundation for development of renormalizable quantum field theory.

Apart from influence on a single atom, virtual transitions modify the evolution equation for atomic ensembles. Let us consider N two-level (a and b, $E_a - E_b = \hbar\omega$) atoms, which are prepared in a collective state with only one atom excited (such a state has zero dipole moment). The initial excitation is distributed among the atoms with a probability amplitude $\beta(\mathbf{r})$, which depends on the atom position $\mathbf{r}$. If we disregard virtual transitions, then for a dense cloud of volume V evolution of the atomic system in the scalar photon theory is described by integral equation with sin kernel [23]

$$\frac{\partial\beta(t,\mathbf{r})}{\partial t} = -\gamma\frac{N}{V}\int d\mathbf{r}'\frac{\sin(k_0|\mathbf{r}-\mathbf{r}'|)}{k_0|\mathbf{r}-\mathbf{r}'|}\beta(t,\mathbf{r}'), \qquad (1)$$

where $\beta(t,\mathbf{r})$ is the probability amplitude to find an atom at position $\mathbf{r}$ excited at time t, γ is the single atom decay rate, $k_0 = \omega/c$, and the integral is taken over the volume of the atomic sample. However, inclusion of virtual processes yields an equation with exp kernel [6,7,15,24]

$$\frac{\partial\beta(t,\mathbf{r})}{\partial t} = i\gamma\frac{N}{V}\int d\mathbf{r}'\frac{\exp(ik_0|\mathbf{r}-\mathbf{r}'|)}{k_0|\mathbf{r}-\mathbf{r}'|}\beta(t,\mathbf{r}'). \qquad (2)$$

The continuous-density approximation is valid when there are many atoms in the volume λ^3, where $\lambda = 2\pi/k_0$ is the wavelength of the atomic transition.

The evolution equation (1) was the subject of investigation several decades ago [23,25,26], while Eq. (2) has been "rediscovered" and studied in detail only recently [6–8,12–15,27,28]. Recent analysis shows that virtual transitions have an interesting effect on collective emission of atoms [12,14,15]. In particular, if the initial atomic state is superradiant, the virtual transitions partially transfer population into slowly decaying states, which results in a trapping of atomic excitation [Fig. 1(a)]. On the other hand, for slowly decaying states virtual processes yield additional decay channels, which leads to a slow decay of the otherwise trapped states [Fig. 1(b)]. Collective frequency (Lamb) shift produced by virtual processes is another fascinating subject of recent theoretical [27–33] and experimental investigation [34].

In this paper we go further and, apart from virtual transitions, include nonlocal effects in consideration. Equations (1)

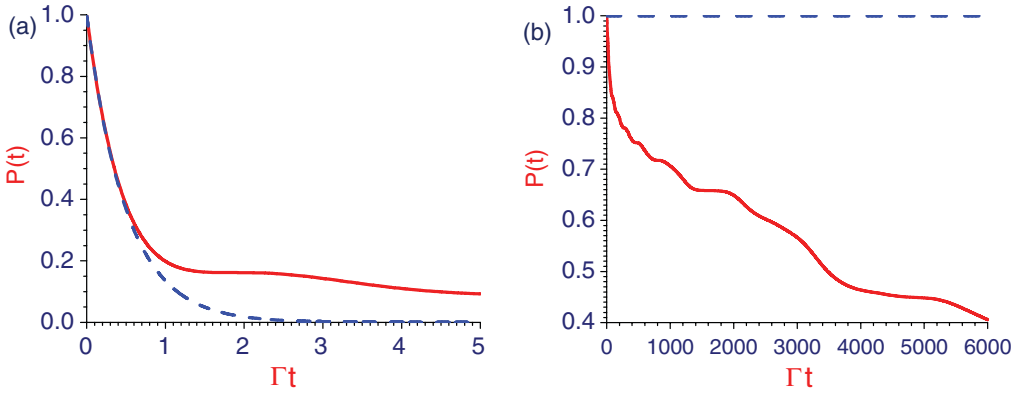

FIG. 1. (Color online) Probability that atoms are excited $P(t)$ as a function of time t for extended spherical atomic cloud of radius R calculated with (solid lines) and without (dashed lines) virtual transitions. Initially atoms are prepared in a timed state $\beta(0,\mathbf{r}) = \sin(k_0 r)/k_0 r$ (a) or symmetric state $\beta(0,\mathbf{r}) = 1$ (b). For the timed state, the atoms, although spread out over a large distance relative to the wavelength, undergo superradiant decay with the rate $\Gamma = 3N\gamma/2(k_0R)^2$. However, at long times, some population remains trapped because of the virtual interactions. For the symmetric state, the opposite takes place. The atoms are trapped in the initial subradiant state. However, virtual transitions disrupt the highly correlated subradiant state, and radiation gradually escapes. Plots are taken from Ref. [15].

and (2) disregard retardation caused by finite value of the speed of light and assume that evolution of the system at time t depends only on the state of the system at this moment of time (local or Markov approximation). This assumption is valid if the atomic system evolves slowly so that during propagation of the signal through the sample the atomic state does not change substantially. However, if size of the sample is large enough, the local approximation breaks down and system's dynamics becomes nonlocal in time. Now evolution of the system at time t will depend on the history, that is, on the states of atoms in the previous moments of time.

II. NONLOCAL EVOLUTION EQUATION AND ITS EXACT SOLUTION

For simplicity of presentation, we skip here the details of tedious calculations and present only the final results. We find that nonlocal effects modify the evolution equation (2) as follows:

$$\frac{\partial\beta(t,\mathbf{r})}{\partial t} = i\gamma\frac{N}{V}\int d\mathbf{r}'\frac{\exp(ik_0|\mathbf{r}-\mathbf{r}'|)}{k_0|\mathbf{r}-\mathbf{r}'|}\beta\left(t-\frac{|\mathbf{r}-\mathbf{r}'|}{c},\mathbf{r}'\right). \tag{3}$$

Equation (3) shows that evolution of an atom at point $\mathbf{r}$ at time t is influenced by the state of the atom at point $\mathbf{r}'$ at the previous moment of time $t' = t - |\mathbf{r}-\mathbf{r}'|/c$. The time difference equals the time of photon flight from $\mathbf{r}'$ to $\mathbf{r}$. If the speed of light c would be infinitely large, then $t' = t$ and the system evolution is local. A finite value of c results in interesting effects, which we discuss next.

We consider an atomic sample having a slab geometry shown in [Fig. 2(a)]. Slab thickness is $R \gg \lambda$, where λ is the wavelength of the atomic transition. We assume that initially the sample is excited by a plane wave photon so that

$$\beta(0,\mathbf{r}) = e^{ik_0 z}, \tag{4}$$

where z is the coordinate perpendicular to the slab plane [Fig. 2(a)]. For such an initial condition we find the following exact analytical solution of Eq. (3):

$$\beta(t,\mathbf{r}) = e^{ik_0 z}\left\{\cos(\Omega t) + \theta(ct-z)\frac{\Omega}{c}\int_z^{ct}\sqrt{\frac{ct-z'}{z'}} \times J_1\left[\frac{2\Omega}{c}\sqrt{z'(ct-z')}\right]dz'\right\}, \tag{5}$$

where $J_1(x)$ is the Bessel function, $\theta(z)$ is the Heaviside step function, and Ω is the collective Rabi frequency

$$\Omega = \sqrt{\frac{n\lambda^2 c\gamma}{2\pi}}, \tag{6}$$

which is proportional to the square root of the atomic density $n = N/V$ and analogous to the plasma frequency in classical electrodynamics. On the other hand, Eq. (2), which omits retardation, yields the following answer:

$$\beta(t,\mathbf{r}) = J_0\left(\frac{2\Omega}{c}\sqrt{ctz}\right)e^{ik_0 z}. \tag{7}$$

Collective frequency Ω determines the characteristic size of the atomic slab R_0 for which nonlocal effects become substantial:

$$R_0 = \frac{c}{\Omega}. \tag{8}$$

If $R \lesssim R_0$, the atomic evolution is local and $\beta(t,\mathbf{r})$ is accurately described by Eq. (7). However, for $R \gg R_0$ the nonlocal effects dramatically modify evolution of the system, yielding oscillations of atomic population with collective frequency Ω.

As a demonstration, in Figs. 2(b) and 2(c) (left side) we compare solution (5) of the nonlocal equation (3) with those obtained omitting retardation effects (7). Namely, we plot the probability of finding atoms excited $P(t)$ as a function of time t for a thin sample $R = 0.5R_0$ [Fig. 2(b)] and a thick sample $R = 5R_0$ [Fig. 2(c)]. Dashed lines are obtained in the

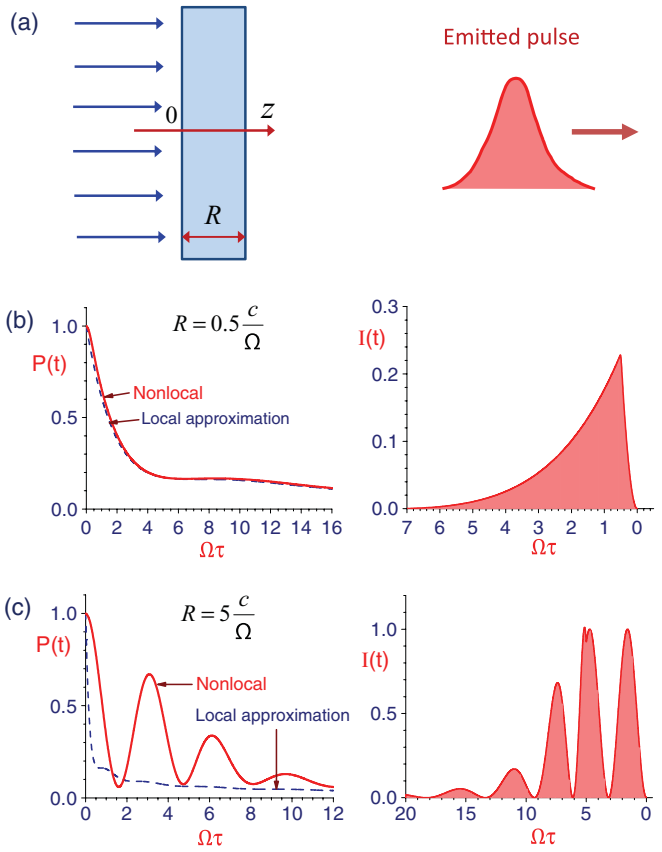

FIG. 2. (Color online) (a) Plane-wave photon prepares atomic slab in the collectively excited state $e^{ik_0 z}$. An excited atomic system undergoes superradiant decay and emits an electromagnetic pulse in the direction of the incident photon. (b) and (c) Probability $P(t)$ of finding atoms excited as a function of time t and shape of the emitted pulse $I(t)$ (arbitrary units) for different thicknesses of the atomic slab $R = 0.5c/\Omega$ (b) and $R = 5c/\Omega$ (c). Dashed lines are obtained in the local approximation, and solid lines represent the exact solution including nonlocal effects.

local approximation, while solid curves represent the exact solution including nonlocal effects. For a thin sample, atoms monotonically decay, and nonlocal effects are not important. However, a fraction of the atomic population is trapped due to virtual transitions. For a thick sample, decay of the atomic population is nonmonotonic. Nonlocal effects produce oscillations of the atomic population. Such oscillations indicate that a photon is emitted and then reabsorbed several times before it leaves the sample.

The analytical solution (5) yields new insights on nonlocal dynamics. For example, for a thick sample the initial evolution (at $t \leqslant R/c$) is nonlocal, and the atomic system undergoes collective oscillations. However, for $t \gg R/c$ Eq. (5) yields

$$\beta(t,\mathbf{r}) \approx \frac{(ct-z)}{(ct-2z)} J_0\left[\frac{2\Omega}{c}\sqrt{z(ct-z)}\right] e^{ik_0 z} \approx J_0\left(\frac{2\Omega}{c}\sqrt{ctz}\right) e^{ik_0 z}, \tag{9}$$

which coincides with the answer (7) obtained omitting retardation. Thus, at large time the system's behavior changes to the local one.

This transition can be understood as follows. The initial atomic state (4) is not an eigenstate, but rather a superposition of many eigenstates. Rapidly decaying states contribute to the nonlocal system's dynamics at early time. At $t \gg R/c$ the fast decaying states have already decayed, and atomic evolution becomes slow. However, for slowly evolving states retardation is no longer important, and the local approximation becomes valid. Thus, at large time, when evolution becomes slow, the system's behavior must change to the local one. Such a transition from the nonlocal to local atomic evolution is properly described by Eq. (5).

III. DISCUSSION

One should mention that collective oscillations of the atomic system with frequency Ω in the nonlocal limit have been discussed in our previous publications for spherical [35] and cylindrical [36] geometries. However, the previous results are obtained in the simple limiting case and are unable to describe a rich system's dynamics in the transition region. For example, for a large spherical atomic cloud of radius R we previously found that the initial atomic state (4) evolves as [35]

$$\beta(t,\mathbf{r}) \approx \cos(\Omega t)\exp\left(-\frac{3c}{8R}t\right) e^{ik_0 z}. \tag{10}$$

Solution (10) was obtained as the first-order perturbation correction to the $R \to \infty$ limit and expected to be valid for $t \ll R/c$. As a consequence, Eq. (10) is unable to describe evolution at $t \gtrsim R/c$, and, in particular, it fails to predict crossover into local dynamics at large time.

The exact analytical solution (5) found here for the slab geometry is valid for any t and R. This solution provides, for the first time, an example of a complete description of nonlocal dynamics of the single-photon emission. In addition, solution (5) shows that such dynamics is rich and cannot be treated by perturbation. Moreover, using the Slowly Varying Envelope Approximation in Space and the Eikonal Approximation one can obtain an approximate closed-form analytical result for the atomic evolution in other geometries from the known answer for the slab. This approach is accurate for large samples when diffraction is not important and was implemented for the single-photon superradiance by Friedberg and Manassah [37]. In such treatment the atomic sample is divided into strips parallel to the z axis, and each strip is treated independently as in a one-dimensional slab geometry problem. In particular, for a spherical cloud of radius R atoms located along the z axis passing through the center of the sphere evolve the same way as atoms in a slab of thickness $2R$. Thus, after simple modifications, the exact solution found here for the slab can also be applied for a sphere.

Figure 3 compares the probability amplitude $\beta_A(t)$ to find atoms at the edge point A of the spherical sample of radius $R = 7c/\Omega$ excited and obtained using the exact Eq. (5) for $z = 2R$ (solid line) and the perturbation solution (10) (dashed line). The two curves are dramatically different apart from short time $t \ll R/c$. In particular, Eq. (5) shows that $\beta_A(t)$ oscillates with constant amplitude for $t < 2R/c$, while for $t \gg R/c$ the oscillation period increases with time and amplitude undergoes power-law decay. In contrast, the

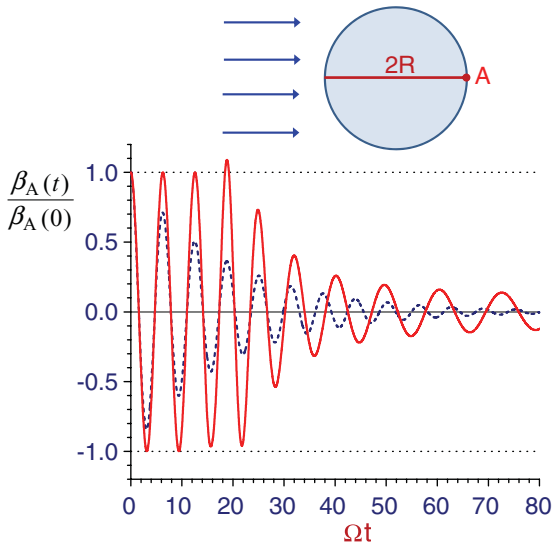

FIG. 3. (Color online) Probability amplitude β_A that atoms at the edge point A of the spherical sample of radius $R = 7c/\Omega$ are excited as a function of time. Solid line is the exact solution (5) with $z = 2R$, while the perturbation answer (10) is shown as dashed line.

perturbation solution (10) yields that the oscillation period remains the same and amplitude exponentially decays at all t. Equation (5) also shows an interesting feature in the transition region. Namely, the atomic excitation at point A increases above its initial value. For the parameters of Fig. 3 the probability $P_A(t) = |\beta_A(t)|^2$ of finding atoms at point A excited is 20% higher at $t = 18.8/\Omega = 1.34(2R/c)$ than $P_A(0)$. This peculiarity is not present in the local dynamics.

Nonlocal effects also dramatically modify shape of the electromagnetic pulse emitted by atoms. Namely, we find that intensity I of the pulse emitted by atomic slab (at $z = R$) goes as

$$I \propto \left\{\sin(\Omega t) - \theta(ct - R)\frac{\Omega}{c}\int_R^{ct} J_0\left[\frac{2\Omega}{c}\sqrt{z(ct-z)}\right]dz\right\}^2, \tag{11}$$

which is different from the local approximation result:

$$I \propto \frac{R}{ct}J_1^2\left(\frac{2\Omega}{c}\sqrt{ctR}\right). \tag{12}$$

In Figs. 2(b) and 2(c) (right side) we plot the intensity of the emitted pulse $I(t)$ as a function of time for thin and thick samples. Nonlocal effects yield modulation of $I(t)$ with the collective frequency Ω.

This feature can be used for experimental observation of crossover between local and nonlocal behavior. For instance, an experiment can be realized for the following parameters. One can prepare the initial atomic state $\beta(0,\mathbf{r}) = e^{ik_0 z}$ in a gas of density $n = 10^{14}$ cm^{-3} and transition wavelength $\lambda = 500$ nm. Then for $\gamma = 10^7$ s^{-1} we obtain the collective frequency $\Omega = 1.3 \times 10^{11}$ s^{-1}, and the characteristic size of the sample for which nonlocal effects become important is $R_0 \approx 2.2$ mm. By measuring the shape of the emitted pulse (e.g., using a streak camera) as a function of the sample size R one can observe crossover from the local to the nonlocal regime when $R \sim R_0$. Appearance of modulation of the emitted pulse intensity at $R \gtrsim R_0$ corresponds to the onset of nonlocal dynamics. Alternatively one can fix the size of the sample and vary atomic density.

Preparation of the initial state $\beta(0,\mathbf{r}) = e^{ik_0 z}$ is tricky. The point is that if we send a plane wave resonant with the dipole allowed atomic transition the wave will be absorbed at the front edge of the slab and atoms in the bulk of the sample will not be excited. To prepare uniform excitation one can send a long laser pulse that is multiphoton resonant to the atomic transition. In this case absorption length of the incident pulse

$$l \approx \frac{4\pi}{3n\lambda^2}\frac{\gamma_{\rm tot}}{\gamma}\left(\frac{\omega}{\Omega_{\rm pulse}}\right)^{m-1} \tag{13}$$

can be much larger than the sample size, which prepares weak uniform excitation of the whole atomic medium. For example, if the medium is excited by a three-photon resonant laser pulse ($m = 3$), then for the pulse Rabi frequency $\Omega_{\rm pulse} = 10^{-3}\omega$ Eq. (13) yields absorption length of $l = 100$ m. Such multiphoton absorption prepares weak excitation of the medium in the state $\beta(0,\mathbf{r}) = e^{ik_0 z}$, where $k_0 = \omega/c$ is the wave number of the atomic transition. An atomic sample will emit light at the atomic frequency ω even though the excitation pulse frequency is ω/m. In such a scheme the medium is optically thick upon emission but optically thin upon excitation.

One should mention that quantum mechanical analysis presented in this paper assumes that only one atom is initially excited, but excitation is distributed among many atoms. However, the results obtained here remain valid if more than one atom is excited. Namely, equations correctly describe atomic evolution and light emission for any weak excitation of the medium (when probability to find each atom excited is much smaller than one).

In summary, we study nonlocal (retardation) effects on collective superradiant emission of atomic ensembles prepared by absorption of a single photon. Both light and atoms are treated quantum mechanically. We obtain an evolution equation for the atomic system that takes into account retardation and solve it analytically for the atomic slab geometry. The obtained exact solution provides new insights into the nonlocal dynamics of the single-photon emission. In particular, it shows that initially nonlocal evolution, accompanied by collective oscillations of the atomic population, becomes local at large time. In addition, during nonlocal evolution, atomic excitation in some parts of the sample rises above its initial value. Nonlocal effects also lead to intensity modulation of the emitted pulse. Transition from the local to the nonlocal regime can be observed experimentally by increasing the size of the sample or changing the atomic density.

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