Spontaneous emission of a moving atom in vacuum

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Spontaneous emission of an excited two-level nonrelativistic atom is examined in the Schrödinger picture by treating the atom's external and internal degrees of freedom on the same quantum footing. After all atomic transitions that lead to the evolution of this atom-vacuum system are analyzed, it is found that the atom's spontaneous emission rate is reduced by its center-of-mass motion. It is also found that, as a result of the entanglement between atomic and photonic states, the spontaneous emission process cannot be explained as a process that takes place in one coordinate system moving with the atom but is observed in another stationary system.

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I. INTRODUCTION

Although spontaneous emission of an excited atom is believed to have been first formulated by Dirac in 1927 [1], it still attracts a lot of research interest [1–15]. Usually the atom under study is assumed to be stationary for the purpose of simplicity. This assumption, however, is beset with problems. From the theoretical viewpoint, a stationary atom has its kinetic energy entirely undetermined according to Heisenberg's uncertainty relation, and is thus a system that requires an input of infinite energy. From the experimental viewpoint, even when the atom is trapped inside an optical lattice, its external motion still cannot be completely suppressed. Recently, it was found, in particular, that this stationary-atom model is too simple to be applied to applications for quantum information and atom chips, where the spontaneous emission of moving atoms near surfaces can no longer be ignored [13]. Clearly, in order to describe accurately the process of spontaneous emission, the atom's center-of-mass (c.m.) motion must be taken into consideration [3,4,6]. It is already recognized that the c.m. motion has non-negligible effects on many physical processes, such as the deflection of atoms by light [16] and entanglement of an atomic system [17]. This paper is devoted to an analysis of the spontaneous emission of an excited atom by treating the atom's external and internal degrees of freedom on the same quantum mechanical footing.

For simplicity, the atom, with mass M and position vector $\vec{R}$, is assumed to be in vacuum and to have two internal states: an excited state $|E\rangle$ with energy $\hbar\omega_E$ and the ground state $|G\rangle$ with energy $\hbar\omega_G$. The difference between ω_E and ω_G is known as the atomic transition frequency $\omega_0 \equiv \omega_E - \omega_G$. Inside the vacuum, each vacuum mode interacts with the atom through an operator V_I defined in the following relation:

$$V_I = \sum_\alpha (\vec{\mu}_{GE} \cdot \vec{g}_\alpha e^{-i\vec{k}_\alpha \cdot \vec{R}} a_\alpha^\dagger |G\rangle\langle E| + \vec{\mu}_{EG} \cdot \vec{g}_\alpha^* e^{i\vec{k}_\alpha \cdot \vec{R}} a_\alpha |E\rangle\langle G|), \tag{1}$$

where $\vec{\mu}_{GE}$ is the matrix element of the atom's electric dipole moment $\vec{\mu}$ between $|G\rangle$ and $|E\rangle$, and $\vec{\mu}_{EG}$ the complex conjugate of $\vec{\mu}_{GE}$. The frequency and the amplitude (containing the polarization unit vector $\vec{\epsilon}_\alpha$) of mode α are denoted, respectively, as ω_α and $\vec{g}_\alpha = i\sqrt{2\pi\hbar\omega_0^2/(L^3\omega_\alpha)}\vec{\epsilon}_\alpha$. The quantization volume is L^3. Also used in V_I are $\vec{k}_\alpha$, the wave vector of mode α ($|\vec{k}_\alpha| = \omega_\alpha / c$), and $a_\alpha^\dagger$ (a_α), the creation (annihilation) operator for the same mode. Throughout the paper, c is the speed of light in vacuum. Note that V_I is treated under the rotating-wave approximation, because the counter-rotating terms, if included, would give only a negligible contribution to the atom's spontaneous emission rate [11]. The Hamiltonian H of the atom-vacuum system is constructed by adding to V_I the unperturbed Hamiltonian H_0:

$$H = H_0 + V_I, \tag{2}$$

where $H_0 = \vec{P}^2/(2M) + \hbar\omega_E|E\rangle\langle E| + \hbar\omega_G|G\rangle\langle G| + \Sigma_\alpha \hbar\omega_\alpha a_\alpha^\dagger a_\alpha$. In H_0, the quantity $\vec{P}$ is the momentum operator of the atom, whose components and those of the atom's position operator $\vec{R}$ obey canonical commutation relations. Since it is constant and unimportant for atomic evolution, the zero-point energy of the vacuum is ignored in H_0. In its nonrelativistic form in Eq. (2), the Hamiltonian H is valid only when the speed v of the atom is negligible compared with c; in the present discussion, this condition is assumed to be satisfied, so that the Röntgen interaction [4,6], which is roughly of the order of $vc^{-1}V_I$, can be safely excluded from H.

Unlike the approach adopted in Refs. [3,4,6], which first derives the Hamiltonian of the atom-vacuum system and then uses Fermi's golden rule to compute directly the atom's spontaneous emission rate, in the present discussion, atomic transitions and translational motion, as well as the associated photon emission and absorption, are all examined. Such an examination is needed, because it will not only illustrate how the atom-vacuum system evolves as a result of light-atom interaction but also, more importantly, reveal that, in the evolution, atomic and photonic states actually become entangled.

If initially the external state of the atom is one eigenstate $|\vec{p}_0\rangle$ of the momentum operator, the internal state equal to the excited state $|E\rangle$, and no photons present, then the evolution of the atom-vacuum system starts from the state $|\psi(0)\rangle = |E\rangle \otimes |\vec{p}_0\rangle \otimes |0\rangle$. In this paper, the discussion is presented in the Schrödinger picture, so that the state $|\psi(t)\rangle$ of the system

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at time t is related to $|\psi(0)\rangle$ through an integral relation:

$$|\psi(t)\rangle = -\frac{1}{2\pi i}\int_{-\infty}^{\infty} dq \frac{e^{-iqt/\hbar}}{q-H}|\psi(0)\rangle, \quad (3)$$

where it is understood that the denominator in the integrand contains an imaginary component $i\eta(\eta\to 0^+)$. There are two advantages of working in the Schrödinger picture. First, in this picture, atomic and photonic operators commute, so that the common problem of how to order these operators no longer exists. Second, the Schrödinger picture allows a clear exhibition of atomic transitions and photon emission and absorption that finally lead to the evolution of the system. As in Ref. [11], the spontaneous emission is studied by computing the time-dependent probability $p_r(t)$ that the system remains in its initial state.

The rest part of this paper is organized as follows. In Sec. II, all relevant atomic transitions are studied, from which a general expression for the probability amplitude $A(t)$ is obtained. [Note that $p_r(t)=|A(t)|^2$.] In the following section, the spontaneous emission rate is derived for two cases, $\vec{p}_0=\vec{0}$ and $\vec{p}_0\neq\vec{0}$. The paper is summarized in Sec. IV.

II. EVOLUTION OF THE ATOM-VACUUM SYSTEM

To formulate the evolution of the system, which is driven by the Green function $1/(z-H)$ in Eq. (3), it is convenient to expand the function into a series of ascending powers of V_I [11]:

$$\frac{1}{q-H} = \frac{1}{q-H_0} + \frac{1}{q-H_0}V_I\frac{1}{q-H_0} + \frac{1}{q-H_0}V_I\frac{1}{q-H_0}V_I\frac{1}{q-H_0} + \cdots . \quad (4)$$

When the terms on the right-hand side (RHS) of the preceding equation operate on $|\psi(0)\rangle$, those atomic transitions that are responsible for the evolution of the system are found. Consider the first term $1/(q-H_0)$. Since this term does not contain V_I, it cannot change the state of the system:

$$\frac{1}{q-H_0}|\psi(0)\rangle = \frac{1}{q-p_0^2/(2M)-\hbar\omega_E}|\psi(0)\rangle. \quad (5)$$

On the other hand, under the action of the second term, the internal state of the atom is changed from $|E\rangle$ to $|G\rangle$, and one photon is emitted from the atom into any mode $|1_\alpha\rangle$:

$$\frac{1}{q-H_0}V_I\frac{1}{q-H_0}|\psi(0)\rangle = \frac{1}{q-p_0^2/(2M)-\hbar\omega_E} \times \sum_\alpha \frac{(\vec{\mu}_{GE}\cdot\vec{g}_\alpha)|G\rangle\otimes e^{-i\vec{k}_\alpha\cdot\vec{R}}|\vec{p}_0\rangle\otimes|1_\alpha\rangle}{q-(\vec{p}_0-\hbar\vec{k}_\alpha)^2/(2M)-\hbar\omega_G-\hbar\omega_\alpha}. \quad (6)$$

Due to the conservation of momentum, which is naturally taken into consideration in the present formulation, the photon emission subsequently causes the external state of the atom to transform from $|\vec{p}_0\rangle$ to $e^{-i\vec{k}_\alpha\cdot\vec{R}}|\vec{p}_0\rangle$; the latter state is still an eigenstate of $\vec{P}$ and has an eigenvalue of $\vec{p}_0-\hbar\vec{k}_\alpha$. As a result, in the denominator in Eq. (6), the kinetic energy of the atom becomes $(\vec{p}_0-\hbar\vec{k}_\alpha)^2/(2M)$.

The emitted photon can be absorbed by the atom through the left V_I operator in the third term:

$$\frac{1}{q-H_0}V_I\frac{1}{q-H_0}V_I\frac{1}{q-H_0}|\psi(0)\rangle = \frac{1}{[q-p_0^2/(2M)-\hbar\omega_E]^2}|E\rangle \otimes|\vec{p}_0\rangle\otimes|0\rangle\sum_\alpha\frac{|\vec{\mu}_{GE}\cdot\vec{g}_\alpha|^2}{q-(\vec{p}_0-\hbar\vec{k}_\alpha)^2/(2M)-\hbar\omega_G-\hbar\omega_\alpha}. \quad (7)$$

After such absorption, the internal state of the atom returns to $|E\rangle$, and the external state to $|\vec{p}_0\rangle$, still as a result of momentum conservation. Once in the excited state, the atom can resume photon emission and absorption repeatedly with the help of the remaining terms on the RHS of Eq. (4). These repeated photon emissions and absorptions by the same atom are known as the radiation reaction, which has been demonstrated to be the origin of spontaneous emission [11]. In the Heisenberg picture, on the other hand, depending on the ordering of atomic and photonic operators, spontaneous emission has been interpreted as resulting from either vacuum fluctuations, or radiation reaction, or both; see Ref. [18], for example. From Eqs. (6) and (7), it is evident that, since the atom's momentum also takes part in the radiation reactions, the atom must have its spontaneous emission influenced by its own c.m. motion.

When all the terms on the RHS of Eq. (4) are considered, it is found that, while those terms that contain odd numbers of V_I cause the system to reside in any state like $|G\rangle\otimes e^{-i\vec{k}_\alpha\cdot\vec{R}}|\vec{p}_0\rangle\otimes|1_\alpha\rangle$, the terms that contain even (including zero) numbers of V_I leave the system in its initial state $|\psi(0)\rangle$. Thus, in the evolution of the atom-vacuum system, the atom's internal and external states and the emitted photon are entangled; see also Refs. [19–21]. Since the present discussion is focused on the probability $p_r(t)$ that the system remains in $|\psi(0)\rangle$, it is convenient to write the final result simply as

$$\frac{1}{q-H}|\psi(0)\rangle = \frac{|\psi(0)\rangle}{q-p_0^2/(2M)-\hbar\omega_E-B} \quad (8)$$

and to leave out other terms irrelevant to the computation of the probability. With the help of Eq. (3), the probability amplitude $A(t)$ is first obtained:

$$A(t) = -\frac{1}{2\pi i}\int_{-\infty}^{\infty} dq\frac{e^{-iqt/\hbar}}{q-p_0^2/(2M)-\hbar\omega_E-B}. \quad (9)$$

In Eqs. (8) and (9), the quantity B is defined as

$$B = \sum_\alpha \frac{|\vec{\mu}_{GE} \cdot \vec{g}_\alpha|^2}{p_0^2/(2M) + \hbar\omega_0 - \hbar\omega_\alpha - (\vec{p}_0 - \hbar\vec{k}_\alpha)^2/(2M) + i\eta}. \quad (10)$$

The radiation reactions and momentum conservation, all needing to be addressed in the evolution of the system, are all contained in B. For example, $|\vec{\mu}_{GE}\cdot\vec{g}_\alpha|^2$ in Eq. (10) is noted to result from V_I and illustrates the fact that during the radiation reactions the emitted photon needs to interact with the atom when it is released and subsequently absorbed; see Eqs. (6) and (7). The denominator in Eq. (10), on the other hand, shows that the radiation reactions reach their maximum magnitudes when the emitted photon has a frequency ω_α that satisfies the following relation:

$$\omega_0 - \omega_\alpha + (\vec{p}_0 \cdot \vec{k}_\alpha)/M - \hbar k_\alpha^2/(2M) = 0. \quad (11)$$

The preceding relation is due to the c.m. motion of the atom and represents the familiar Doppler effect. See Refs. [22,23], respectively, for a discussion of the Doppler effect on light reflection from and propagation through an ensemble of atoms subject to thermal motion. Since the frequency ω_α in Eq. (11) is the dominant frequency of the emitted photon, it is denoted from now on as ω_D. In general, B is a complex number; in particular, while its real part Re B corresponds to the shift of the energy level of $|E\rangle$, which is not discussed in the present paper, its imaginary part Im B is directly associated with the spontaneous emission rate [see Eq. (9)]. For the convenience of subsequent discussion, the imaginary coefficient $i\eta$ is explicitly shown in Eq. (10).

In the next section, the spontaneous emission rate is derived by first computing $A(t)$. Note that the expression of $A(t)$ in Eq. (9) is valid at any time and is derived when every order of light-atom interaction is considered. One problem with the approach of using Fermi's golden rule [3,4,6] is that this rule considers the light-atom interaction only to the first order and is actually invalid unless the time is significantly large [24].

III. SPONTANEOUS EMISSION RATE Γ

The atom's initial momentum $\vec{p}_0$ certainly can be either equal to or different from zero. Consider first the case that the atom is initially at rest, that is, $\vec{p}_0 = \vec{0}$. Equation (10) then reduces to

$$B = \sum_\alpha \frac{|\vec{\mu}_{GE} \cdot \vec{g}_\alpha|^2}{\hbar\omega_0 - \hbar\omega_\alpha - \hbar^2\omega_\alpha^2/(2Mc^2) + i\eta}, \quad (12)$$

which shows through its denominator that, due to the atom's recoil, the dominant frequency ω_D of the emitted photon is approximately equal to $\omega_0[1-\hbar\omega_0/(Mc^2)]$, which is smaller than the transition frequency ω_0. As already pointed out in the last section, it is at ω_D that the radiation reactions reach their maximum magnitudes. By choosing the direction of $\vec{\mu}_{GE}$ as a reference direction, and by using the mode-continuum approximation [11], it is found from Eq. (12) that

$$\text{Im } B = -\frac{\Gamma_0\hbar}{2[\hbar\omega_0(Mc^2)^{-1} + 1]}, \quad (13)$$

where $\Gamma_0 = 4|\vec{\mu}_{GE}|^2\omega_0^3/(3\hbar c^3)$ is the spontaneous emission rate of an atom fixed in the vacuum. With the help of Eq. (9), the probability $p_r(t)$ is obtained:

$$p_r(t) = |A(t)|^2 = e^{-t\Gamma_0/[\hbar\omega_0(Mc^2)^{-1}+1]}, \quad (14)$$

from which the spontaneous emission rate of the atom that is initially at rest is immediately recognized to be

$$\Gamma = \frac{\Gamma_0}{\hbar\omega_0(Mc^2)^{-1} + 1}. \quad (15)$$

When $M\to\infty$, the emission rate Γ reproduces Γ_0, as expected. The relation in the preceding equation shows that in order to formulate the spontaneous emission of a moving atom accurately the mass of the atom cannot be ignored in principle. The relation also shows that the finite mass M actually decreases the atom's emission rate. To understand this latter observation, it is helpful to view the spontaneous emission as a process in which the atom is coupled to the vacuum and loses its energy to the vacuum via the emitted photon. Since the emitted photon in practice has a frequency close to ω_D, the larger ω_D is, the more efficient this energy transfer becomes. The spontaneous emission from colloidal nanocrystals is also found to have a rate that increases with the emission frequency [25]. So when the dominant frequency ω_D is reduced to be smaller than ω_0 as a result of the atom's c.m. motion, it is not a surprise to find $\Gamma < \Gamma_0$.

When $\vec{p}_0 \neq \vec{0}$, the expression of B becomes more complicated, because now, as Eq. (10) shows, it depends on the orientation of both $\vec{\mu}_{GE}$ and $\vec{p}_0$. But, since $\vec{\mu}_{GE}$ is never measured, its orientation can be averaged over. Consequently, $|\vec{\mu}_{GE}\cdot\vec{g}_\alpha|^2$ in Eq. (10) is approximately equal to $2|\vec{\mu}_{GE}|^2|\vec{g}_\alpha|^2/3$. This approximation is often used when the light-atom interaction is discussed [24]. In Refs. [3,6], a different approximation is adopted by assuming that $\vec{\mu}_{GE}$ has a fixed direction relative to $\vec{p}_0$. As in the previous case, the mode-continuum approximation is used again in Eq. (10), with the result

$$B \simeq -\frac{|\vec{\mu}_{GE}|^2\omega_0^2 M}{3\pi c p_0}\int_0^{\Omega/c} dk_\alpha \times \ln\frac{\hbar\omega_0 - \hbar c k_\alpha - \hbar p_0 k_\alpha M^{-1} - \hbar^2 k_\alpha^2(2M)^{-1} + i\eta}{\hbar\omega_0 - \hbar c k_\alpha + \hbar p_0 k_\alpha M^{-1} - \hbar^2 k_\alpha^2(2M)^{-1} + i\eta}, \quad (16)$$

where a cutoff frequency Ω is needed [26] to make the nonrelativistic Hamiltonian H applicable in the present discussion. Also in the preceding equation $p_0 = |\vec{p}_0|$. Note that, since the difference between the numerator and denominator of the integrand in Eq. (16) is $2\hbar p_0 k_\alpha M^{-1}$, which is in fact insignificant under the present assumption $p_0 M^{-1} \ll c$, it is valid to simplify B further to

$$B \simeq -\frac{2|\vec{\mu}_{GE}|^2\omega_0^2\hbar}{3\pi c}\int_0^{\Omega/c} dk_\alpha \times \frac{k_\alpha}{\hbar^2 k_\alpha^2(2M)^{-1} + (\hbar c + \hbar p_0 M^{-1})k_\alpha - \hbar\omega_0 - i\eta}. \tag{17}$$

After a straightforward calculation, the imaginary part of B is obtained:

$$\mathrm{Im}\, B = -\frac{\Gamma_0 M c^2}{2\omega_0}\left(1 - \frac{c + p_0 M^{-1}}{\sqrt{(c + p_0 M^{-1})^2 + 2\hbar\omega_0 M^{-1}}}\right). \tag{18}$$

The result in the preceding equation reproduces that in Eq. (13) when $\vec{p}_0 \to \vec{0}$. By following the same procedure as that in the previous case, the spontaneous emission rate Γ of the atom that has a nonzero initial momentum $\vec{p}_0$ is found to be

$$\Gamma = \frac{\Gamma_0 M c^2}{\hbar\omega_0}\left(1 - \frac{c + p_0 M^{-1}}{\sqrt{(c + p_0 M^{-1})^2 + 2\hbar\omega_0 M^{-1}}}\right), \tag{19}$$

which clearly demonstrates that, for a moving atom, its mass and momentum in principle all contribute to the spontaneous emission rate. Both the expressions in Eqs. (15) and (19) show that as a result of the c.m. motion the atom's spontaneous emission rate is reduced, no matter whether the atom is initially stationary or not. It is also worthwhile to note that, unlike the results reported in Refs. [3,6], the spontaneous emission rate Γ in Eq. (19) cannot be connected to Γ_0 through a relation like $\Gamma_0/\sqrt{1-(p_0 M^{-1} c^{-1})^2}$, which is a common expression in the theory of relativity and is needed when a phenomenon is observed in different coordinate systems. The reason is that, as demonstrated in Sec. II, during the evolution of the atom-vacuum system, the atom's internal and external states and the emitted photon are all entangled, so that the atom's spontaneous emission can no longer be viewed as a process that takes place in one coordinate system moving with the atom but is observed in another system fixed in space.

The two-level atom in the present discussion can be simulated, for example, by an excited helium atom in the metastable $2\,^3S_1$ state [19], because this state is connected to the $2\,^3P_2$ state through an optical transition at a wavelength of 1083 nm. If the speed of the present atom is additionally assumed to be 4000 m/s, in the range realized in the same reference, then it is found by using Eq. (19) that the c.m. motion can only reduce Γ insignificantly relative to Γ_0:

$$\frac{\Gamma_0 - \Gamma}{\Gamma_0} \sim 10^{-5}. \tag{20}$$

If the atom is initially at rest, the reduction in Γ is even smaller [see Eq. (15)]:

$$\frac{\Gamma_0 - \Gamma}{\Gamma_0} \sim 10^{-10}. \tag{21}$$

Although they might be difficult to observe experimentally, the results in Eqs. (20) and (21) are expected, because in the nonrelativistic region, where the Hamiltonian H in Eq. (2) is valid, the atom's c.m. motion is simply too slow in comparison with c.

IV. CONCLUSION

The spontaneous emission of an atom in vacuum is studied in the Schrödinger picture when the atom's internal and external degrees of freedom are all considered quantum mechanically. Through specific calculation, the atom's spontaneous emission rate is obtained for each of the two initial conditions where the atom is either at rest or moving.

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