Quantum radiation from a shaken two-level atom in vacuum

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We present a nonrelativistic theory of quantum radiation generated by shaking a two-level atom in vacuum. Such radiation has the same origin of photon emission in dynamical Casimir effect. By performing a time-dependent "dressing" transformation to the Hamiltonian, we derive an interaction term that governs the radiation. In particular, we show that photon pairs can be generated, not only by shaking the position of the atom, but also by changing the internal states of the atom. As applications of our theory, we calculate the emission rate from an oscillating atom and the multiphoton state generated in a single-photon scattering process.

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I. INTRODUCTION

In quantum mechanics, fluctuations in vacuum fields can result in a variety of observable physical phenomena [1]. An interesting example is the dynamical Casimir effect (DCE) [2,3], which converts vacuum fluctuations into radiation by modulating the system with time-dependent parameters. Traditionally, DCE is studied in macroscopic systems such as a moving mirror [4] or a cavity with a time-varying length [5], and DCE by modulation of boundary conditions was observed in superconducting circuits [6]. At the microscopic level, DCE occurs when an atom moves nonuniformly in vacuum [7], and such a problem has also been discussed in the context of Unruh radiation [8,9]. Apart from moving an atom, we note that a change of internal states of a rest atom may also distort the vacuum nonadiabatically and emit photons [10]. This is understood because the vacuum field can interact differently with different electronic states.

In this paper we present a microscopic Hamiltonian which governs the generation of photons when an atom is subjected to time-dependent changes in its external or internal states. This would provide a microscopic picture of DCE and other similar parametric amplification processes of the quantum vacuum. Mathematically, the presence of counter-rotating terms in atom-field interactions is responsible for the radiation. In stationary systems these counter-rotating terms determine how an atom is dressed by virtual photons, and a useful technique of handling counter-rotating terms is the "dressing" transformation [11]. Such a transformation can significantly simplify the description because the Hamiltonian is represented in a suitable photon-atom dressed basis, in a way that virtual transitions between dressed states appear only as higher-order processes. The transformation has been applied to the studies of quantum Rabi model [12,13], spin-boson model [14–16], effects of counter-rotating terms on spontaneous decay [17–19], control of Lamb shift [20], and quantum Zeno and anti-Zeno effects [21–23]. Here we generalize this transformation to time-dependent systems and discover an interaction term directly connected to DCE or Unruh radiation. By treating this term as a perturbation, we employ time-dependent perturbation theory to calculate the two-photon emission rate from an oscillating atom and the three-photon amplitude generated in a single-photon scattering process. The multiphoton state in the latter serves as a basic example of quantum radiation triggered by a change of internal states during a quantum process.

II. MODEL HAMILTONIAN

We begin with a Hamiltonian of a two-level atom interacting with a quantized electromagnetic field:

$$H = \frac{\omega_e}{2}\sigma_z + \sum_k \omega_k a_k^\dagger a_k + \sum_k [g_k^*(t)a_k^\dagger + g_k(t)a_k]\sigma_x, \quad (1)$$

where $\sigma_z = |e\rangle\langle e| - |g\rangle\langle g|$ and $\sigma_x = |e\rangle\langle g| + |g\rangle\langle e|$ are Pauli matrices describing the two-level atom with an excited state $|e\rangle$ and a ground state $|g\rangle$. The atomic transition frequency is denoted as ω_e, and a_k and $a_k^\dagger$ are annihilation and creation operators associated with the field mode k of frequency ω_k. Note that the mode index k used here is a general label for a normal mode of the field. For example, in free space, k corresponds to a wave vector $\mathbf{k}$. The time-dependence of coupling strengths $g_k(t)$ can be realized by various settings, for example, changing the position of the atom, and the specific expression is determined by the form of interaction. Throughout this paper, we assume that $g_k(t)$ changes slowly in the time scale of ω_e^{-1}.

A. Time-dependent dressing transformation

We consider a time-dependent unitary operator defined by

$$T(t) \equiv \exp[\sigma_x X(t)], \quad (2)$$

where

$$X(t) \equiv \sum_k [\xi_k^*(t)a_k^\dagger - \xi_k(t)a_k] \quad (3)$$

with $\xi_k(t)$'s being some small time-dependent parameters to be determined later. Let $|\psi(t)\rangle \equiv T(t)|\Psi(t)\rangle$ be the state in the transformed frame, where $|\Psi(t)\rangle$ is the state in the original frame; then the evolution of $|\psi(t)\rangle$ is governed by the transformed Hamiltonian $H' = THT^\dagger - iT\frac{dT^\dagger}{dt}$. In Appendix A, we show that H' up to second order in ξ is given by

$$\begin{aligned} H' &= \frac{\omega_e}{2}\sigma_z + \sum_k \omega_k a_k^\dagger a_k \\ &\quad + \sum_k \sigma_+ a_k[(\omega_e - \omega_k)\xi_k + g_k - i\dot{\xi}_k] + \text{H.c.} \\ &\quad + \sum_k \sigma_- a_k[(-\omega_e - \omega_k)\xi_k + g_k - i\dot{\xi}_k] + \text{H.c.} \\ &\quad + \omega_e\left[\sum_k (\xi_k^* a_k^\dagger - \xi_k a_k)\right]^2 \sigma_z + E(t), \end{aligned} \tag{4}$$

with $\dot{\xi}_k = \frac{d}{dt}\xi_k$. The last term $E(t) \equiv \sum_k [\frac{i}{2}\xi_k^*(t)\dot{\xi}_k(t) - g_k(t)\xi_k^*(t) + \text{c.c.} + \omega_k|\xi_k(t)|^2]$ is a time-dependent real number which only contributes a phase to the overall state and can be ignored.

The aim of the transformation is to eliminate counter-rotating terms in the third line of Eq. (4) with a suitable set of $\{\xi_k(t)\}$. This is done by setting the coefficients of the counter-rotating terms to vanish, i.e.,

$$(-\omega_e - \omega_k)\xi_k(t) + g_k(t) - i\dot{\xi}_k(t) = 0, \tag{5}$$

which has the solution

$$\xi_k(t) = \xi_k(0)e^{i(\omega_k+\omega_e)t} - i\int_0^t dt' g_k(t')e^{i(\omega_k+\omega_e)(t-t')}. \tag{6}$$

We are free to choose the initial condition $\xi_k(0)$. If we choose $\xi_k(0) = \frac{g_k(0)}{\omega_k+\omega_e}$ and make use of the assumption that $g_k(t)$ varies slowly in the time scale of ω_e^{-1}, then $\xi_k(t)$ is approximated by

$$\xi_k(t) \approx \frac{g_k(t)}{\omega_k + \omega_e}. \tag{7}$$

The Hamiltonian H' then becomes

$$\begin{aligned} H' &= \frac{\omega_e}{2}\sigma_z + \sum_k \omega_k a_k^\dagger a_k + \sum_k (\eta_k a_k \sigma_+ + \eta_k^* a_k^\dagger \sigma_-) \\ &\quad + \frac{1}{4\omega_e}\sum_{j,k}(\eta_j^* a_j^\dagger - \eta_j a_j)(\eta_k^* a_k^\dagger - \eta_k a_k)\sigma_z, \end{aligned} \tag{8}$$

where the new corotating coupling strength $\eta_k(t) = 2\omega_e\xi_k(t)$ is defined.

The second line of Eq. (8) can be put into normal order, and this yields a time-dependent c-number multiplying σ_z which corresponds to a time-dependent shift of transition frequency between the two atomic levels. To properly account for the shift in a natural atom in nonrelativistic theory, one can impose a frequency cut-off ω_c and subtract the relevant self-energy terms as in the standard treatment of the Lamb shift problem [24]. For a multilevel atom in three-dimensional free space with constant g_k's, it has been demonstrated that the dressing transformation and the mass renormalization procedure can lead to a standard expression of the Lamb shift [17,21]. Here, for time-dependent systems, since $\eta_k(t)$ follows $g_k(t)$ adiabatically according to Eq. (7), the renormalized shift can be interpreted as a generalized (time-dependent) Lamb shift. For convenience we shall use ω_e' to denote the shifted transition frequency of the atom.

Finally, the Hamiltonian reads

$$H' = H_0 + H_1 + \sigma_z\Gamma, \tag{9}$$

where

$$H_0 = \frac{\omega_e'}{2}\sigma_z + \sum_k \omega_k a_k^\dagger a_k, \tag{10}$$

$$H_1 = \sum_k (\eta_k a_k \sigma_+ + \text{H.c.}), \tag{11}$$

$$\Gamma(t) = \frac{1}{4\omega_e}\sum_{j,k}(\eta_j^*\eta_k^* a_j^\dagger a_k^\dagger - \eta_j^*\eta_k a_j^\dagger a_k + \text{H.c.}) \tag{12}$$

are defined. Note that the counter-rotating terms $a_k^+\sigma_+$ and $a_k^-\sigma_-$ have been eliminated without invoking any rotating wave approximation. The transformation T has taken care of most of the virtual transitions or dressing effects due to counter-rotating terms, leaving $\sigma_z\Gamma$ as a small correction. If $\sigma_z\Gamma$ can be ignored, then the ground state is simply $|g\rangle|0\rangle$ (where $|0\rangle$ is the vacuum state in the transformed frame). Such a state in the original frame reads as $T^\dagger|g\rangle|0\rangle$, a dressed state in which the atom and virtual photons are entangled.

We point out that $\sigma_z\Gamma$ governs the generation of radiation via the pair creation operators $a_j^\dagger a_k^\dagger$. Such a term has often been neglected in stationary systems because it is second order in g and off resonance. However, when the atom is subjected to time-dependent modulation, $\sigma_z\Gamma$ could lead to a resonant generation of photons. As a remark, we note that our Hamiltonian is different from the one derived from the atomic polarizability approach [7]. The theoretical framework provided here allows us to study the quantum radiation process in dressed basis and, by keeping track of the internal degrees of freedom, DCE due to time-dependent perturbation of internal states can be addressed.

B. Ground state at $t = 0$

Assuming the coupling strengths $g_k(t)$ start changing only for positive times $t > 0$, the ground state defined at $t = 0$ can serve as an initial state to study the quantum dynamics. It should be noted that $|g\rangle|0\rangle$ mentioned above is not the true ground state of the system because of the presence of the $\sigma_z\Gamma$ term. If we take $|g\rangle|0\rangle$ as an initial state, there will be additional radiative effects due to self-dressing of the system [25,26], which would obscure the quantum radiation we are interested in.

We construct the ground state $|\phi_0\rangle$ of H' approximately as

$$|\phi_0\rangle \approx |g\rangle|0'\rangle, \tag{13}$$

where $|0'\rangle$ is the lowest state of the following quadratic field Hamiltonian H_B:

$$H_B \equiv \sum_k \omega_k a_k^\dagger a_k - \Gamma(0). \tag{14}$$

By perturbation theory up to first order in Γ, we have

$$|0'\rangle \approx |0\rangle + \sum_{kk'}\frac{\Lambda_{kk'}(0)}{\omega_k + \omega_{k'}}a_k^\dagger a_{k'}^\dagger|0\rangle, \tag{15}$$

where

$$\Lambda_{kk'}(t) = \frac{\eta_k^*(t)\eta_{k'}^*(t)}{4\omega_e'(t)} \tag{16}$$

is defined. Note that $|g\rangle|0'\rangle$ is the ground state of $H_0 + \sigma_z\Gamma(0)$, and it can be used to approximate the ground state of the full Hamiltonian $H' = H_0 + \sigma_z\Gamma(0) + H_1$ at $t = 0$ because H_1 would bring higher-order corrections only.

It is worth noting that although $|\phi_0\rangle$ contains some photon pairs, they are virtual photons not contributing to radiation. This is understood because $|\phi_0\rangle$ is a photon-atom bound state, and the corresponding photon density is localized around the atom as a part of the dressing.

III. QUANTUM RADIATION

In this section we treat $\sigma_z\Gamma$ as a weak perturbation and examine the evolution of the system. By first-order time-dependent perturbation theory, the system state $|\psi(t)\rangle$ in the Schrödinger picture is given by

$$|\psi(t)\rangle \approx U(t)|\psi(0)\rangle - i\int_0^t d\tau\, U(t-\tau)\sigma_z \times \Gamma(\tau)U(\tau)|\psi(0)\rangle, \tag{17}$$

where $U(t)$ is the evolution operator generated by $H_0 + H_1$, and the integral containing $\Gamma(\tau)$ determines the amplitude of photons generated during the evolution. We point out that although the emitted photons described by $|\psi(t)\rangle$ are defined in the transformed frame, they are also photons in the original frame. This is because $\Gamma(\tau)$ is already second order in ξ. As long as we keep the accuracy to this order consistently, the inverse transformation $T^\dagger$ should be taken as an identity operator when operating on the second term in Eq. (17).

In the following we examine two cases of photon production. The photons generated in these two cases can be considered as quantum radiation because they originate from nonadiabatic perturbations to the quantum vacuum as in the photon emission in DCE or Unruh radiation. To facilitate the calculation, η_k is assumed to be a broad function of frequency as the ones given in Eq. (B8) and Eq. (24). In addition, since the Lamb shifts are typically a tiny fraction of ω_e, we shall approximate $\omega_e' \approx \omega_e$ as a constant. In this way we can write $U(t) \approx e^{-i(H_0+H_1)t}$ with ω_e' replaced by ω_e in H_0.

A. Shaking the atom's position

In this case the initial state is assumed to be the ground state $|\phi_0\rangle$ obtained in Eq. (13), and the atom is shaken so that its position $\mathbf{r}_A(t)$ is a function of time. This leads to a time-dependent coupling $g_k(t)$, whose explicit form in three-dimensional free space is given in Appendix B. By using Eqs. (13) and (15), and keeping terms to first order in Γ, Eq. (17) becomes

$$|\psi(t)\rangle \approx U(t)|\phi_0\rangle + i\int_0^t d\tau\, U(t-\tau)\Gamma(\tau)|g\rangle|0\rangle. \tag{18}$$

Note that we have replaced $|\phi_0\rangle$ by $|g\rangle|0\rangle$ in the second term because the two-photon part in Eq. (15) is first order in Γ. In addition, we have used $\sigma_z U(t)|g\rangle|0\rangle = -|g\rangle|0\rangle$.

A further approximation can be made by observing that H_1 has little effect on the photons in the dressed ground state $|\phi_0\rangle$. This is because real transitions described by H_1 are only significant for photons at frequencies within several linewidths around the atomic transition frequency. However, we note that, due to our assumption of η_k, photon pairs in $|\phi_0\rangle$ spread out very broadly in frequency space (over many linewidths), and the fraction of near resonance photons is very small. Consequently, we can write $U(t)|\phi_0\rangle \approx U_0(t)|\phi_0\rangle$, where $U_0(t) \equiv e^{-iH_0t}$ is the free evolution operator. The same argument can also be applied to the integrand of Eq. (18), where most of the photons generated by Γ are of the same far off-resonance nature, so that it is justified to make the approximation $U(t-\tau)\Gamma(\tau)|g,0\rangle \approx U_0(t-\tau)\Gamma(\tau)|g,0\rangle$. With these approximations, the perturbed state becomes

$$\begin{aligned}|\psi(t)\rangle &\approx U_0(t)|\phi_0\rangle - i\int_0^t d\tau\, U_0(t-\tau)\Gamma(\tau)|g,0\rangle \\ &= |g\rangle|0\rangle + |g\rangle\sum_{kk'} C_{kk'}(t)a_k^\dagger a_{k'}^\dagger|0\rangle,\end{aligned} \tag{19}$$

where

$$C_{kk'}(t) = \frac{\Lambda_{kk'}(0)}{\omega_k + \omega_{k'}}e^{-i(\omega_k+\omega_{k'})t} + i\int_0^t d\tau\, \Lambda_{kk'}(\tau)e^{-i(\omega_k+\omega_{k'})(t-\tau)}. \tag{20}$$

If the couplings, and hence $\Lambda_{kk'}$, are time independent, then the freely propagating terms in the first line of Eq. (20) due to $U_0(t)|\phi_0\rangle$ will be exactly cancelled by the lower limit of the time integral in the second line, and $C_{kk'}$ is simply the dressing given in Eq. (15) without producing any freely propagating photons.

However, if the couplings are time dependent, then the propagating terms are no longer canceled, resulting in DCE or Unruh type radiation. As an example, consider the following coupling:

$$\eta_k(t) = \eta_k^0 + ik_m r_m(e^{i\omega_m t}\eta_k^+ + e^{-i\omega_m t}\eta_k^-), \tag{21}$$

where k_m, r_m, η_k^0, and $\eta_k^\pm$ are real, time-independent numbers. In Appendix B we show that this is the form of coupling taken by an electromagnetic field interacting with a two-level atom moving in an externally prescribed nonrelativistic simple harmonic motion, with frequency $\omega_m = ck_m$ and amplitude r_m under the long-wavelength approximation $k_m r_m \ll 1$. Taking the continuum limit, the coupling $\eta_k(t)$ in Eq. (21) leads to a two-photon emission rate given by Fermi's golden rule:

$$\mathcal{R} = \frac{\pi(k_m r_m)^2}{4\omega_e^2}\int d^Dk\int d^Dk'\rho(k)\rho(k') \times \left(\eta_k^+\eta_{k'}^0 + \eta_{k'}^+\eta_k^0\right)^2\delta(\omega_k + \omega_{k'} - \omega_m), \tag{22}$$

where D is the dimension of k space and $\rho(k)$ is the corresponding density of states of the field modes. The sum of frequencies of the emitted photon pairs concentrates at ω_m.

Specializing to $D = 3$ free space and using the definitions of η_k^0 and $\eta_k^\pm$ in Appendix B, we find that the emission rate is given by

$$\mathcal{R} \approx C(k_m r_m)^2 \left(\frac{\gamma}{\omega_e}\right)\left(\frac{\omega_m}{\omega_e}\right)^7 \gamma, \tag{23}$$

where γ is the spontaneous decay rate of the atom and C is a proportionality constant of order about 10^{-2} [27]. The scaling dependence of the system parameters in Eq. (23) was also found in [7] with a different approach.

By Eq. (23), we note that $\mathcal{R}$ is extremely small because ω_m and γ are much lower than ω_e in general. In particular, since emitted photons are limited to frequencies below ω_m, and the density of state scales as $\rho(k) \propto k^2$ (for $D = 3$), the emission rate is strongly suppressed when ω_m is low. If we consider the setup in $D = 1$ space where $\rho(k)$ is uniform, for example, in a one-dimensional waveguide, then $\mathcal{R}$ will scale as $(\frac{\omega_m}{\omega_e})^3$ instead of $(\frac{\omega_m}{\omega_e})^7$ above.

B. Shaking the atom's internal state

In this case, g_k's are time independent but the atom is shaken internally such that there is a time-dependent population difference between the two atomic levels. This would generate radiation through σ_z in the interaction term $\sigma_z\Gamma$. Such radiation should be distinguished from the usual dipole radiation interaction, because the latter is governed by atomic coherence σ_+ or σ_- instead of σ_z. A simple way of changing the atomic population is by absorption and reemission of a photon. Here we show that a single photon scattering is always accompanied by emission of multiple photons.

For simplicity, we consider the scattering in a one-dimensional waveguide of length L (which will be taken to infinity in the continuum limit) and cross-section area A, in which the normal modes are labeled by k. A positive (negative) k corresponds to a right- (left-) propagating mode of frequency $\omega_k = c|k|$. Assuming $x = 0$ is the position of the atom, the coupling is given by

$$\eta_k = \frac{2\omega_e}{\omega_e + \omega_k}\sqrt{\frac{\omega_k}{2\epsilon_0 \hbar A L}}d, \tag{24}$$

where d is the electric dipole matrix element. We have suppressed the polarization label of the field modes because the dipole is assumed to be in parallel with one of the orthogonal polarizations.

At $t = 0$, there is an incident single-photon wave packet at a far distance from the atom, such that the atom prepared in the dressed ground state $|\phi_0\rangle$ would not experience the incident photon initially. The initial state of the system in the transformed frame is given by

$$|\psi(0)\rangle = W^\dagger|\phi_0\rangle, \tag{25}$$

where $W^\dagger$ is a creation operator of the single-photon wave packet defined by

$$W^\dagger = \sum_k W_k a_k^\dagger. \tag{26}$$

Here W_k are coefficients determining the shape of the wave packet. Noting that the dressing transformation T only modifies the field in the neighborhood of the atom; the transformation does not affect the initial photon as long as the wave packet is sufficiently far away from the atom. Mathematically, this corresponds to the condition $[W^\dagger, T] = [W^\dagger, T^\dagger] = 0$, so that the initial state in the original frame is $T^\dagger|\psi(0)\rangle = T^\dagger W^\dagger|\phi_0\rangle = W^\dagger T^\dagger|\phi_0\rangle$.

Specifically, let us consider the following Lorentzian photon wave packet defined by

$$W_k = \sqrt{\frac{\gamma'}{cL}}\frac{e^{-i(k-k_e)x_0}}{-i(k-k_e) + \frac{\gamma'}{2c}}, \tag{27}$$

where $x = x_0 < 0$ is the position of the front edge of the packet on the left of the atom and $\gamma' \ll \omega_e$ is a positive real number characterizing the spectral width of the packet. In addition, we consider $ck_e = \omega_e$ so that the incident photon is in resonance with the atom and travels to the right. Note that $W^\dagger$ commutes with T and $T^\dagger$ as $|x_0| \to \infty$.

The perturbed state given by Eq. (17) can be evaluated approximately. Together with the incident photon, there can be three freely propagating photons in the final state after the scattering is completed. The calculation is presented in Appendix C; we find that the three-photon amplitude in the long-time limit is approximately given by

$$|\psi_3\rangle \approx \sum_{jkl} C_{jkl} e^{-i(\omega_{jkl} - \frac{\omega_e}{2})(t-t_0)} a_j^\dagger a_k^\dagger a_l^\dagger |0\rangle, \tag{28}$$

where

$$C_{jkl} = \frac{\sqrt{\gamma'\gamma}}{2\omega_e}\frac{\eta_j^*\eta_k^*\eta_l^*}{\left(i\Delta_l - \frac{\gamma}{2}\right)\left(i\Delta_{jkl} - \frac{\gamma}{2}\right)\left(i\Delta_{jkl} - \frac{\gamma'}{2}\right)}. \tag{29}$$

Here we have defined $\Delta_l \equiv \omega_l - \omega_e$; $\Delta_{jkl} \equiv \omega_j + \omega_k + \omega_l - \omega_e$, and $t_0 \equiv \frac{|x_0|}{c}$ is the time needed for the photon wave packet travel from its initial position to the atom's.

Equation (29) indicates that C_{jkl} is significant when the frequency sum of the three photons is near the atomic transition frequency ω_e, i.e., $\omega_j + \omega_k + \omega_l \approx \omega_e$. Since the numerator $\eta_k^*\eta_j^*\eta_l^* \propto \sqrt{\omega_k\omega_j\omega_l}$, the three-photon amplitude on the $\omega_j + \omega_k + \omega_l \approx \omega_e$ surface is not sensitive to the single-photon resonance associated with the $i\Delta_l - \frac{\gamma}{2}$ denominator. For example, in the case of $\gamma \approx \gamma'$ and $\omega_j = \omega_k$, $C_{jkl} \sim c^{3/2}\gamma^{1/2}/\omega_e^2 L^{3/2}$ over the entire range $\omega_l \in (\gamma, \omega_e - \gamma)$.

IV. CONCLUSION

To conclude, we have developed a Hamiltonian for the study of DCE or Unruh type radiation at the microscopic level. Through the time-dependent dressing transformation T, we are able to identify the interaction term $\sigma_z\Gamma$ which governs photon pair generation when modulations are applied to atom-field couplings or the atom's internal states. As demonstrated by the examples in Sec. III, the radiation is extremely weak in natural systems because the atomic transition frequency ω_e is generally much higher than the spontaneous emission rate γ and mechanical modulation frequency ω_m. However, recent progress of realizing ultrastrong coupling in artificial systems could be an important step towards the observation of such radiation [28,29]. In particular, the value of γ can be a significant fraction of ω_e in waveguide QED [30]. We also point out that related quantum radiation effects based on various modulation schemes [10,29,31–34] and photon scattering [16,35] in ultrastrong coupling regime have been reported recently. In the future, we hope to explore applications of the time-dependent dressing transformation in ultrastrong coupling problems and quantum radiation with multiple atoms.

APPENDIX A: DERIVATION OF THE TRANSFORMED HAMILTONIAN

The derivation is similar to that in stationary systems (see, for example, Ref. [15]); the main difference is the time dependence of $\xi_k(t)$ which generates extra terms in the Hamiltonian:

$$H' = THT^\dagger - iT\frac{\partial}{\partial t}T^\dagger. \tag{A1}$$

By $T = e^{\sigma_x X} = \cosh(X)I + \sinh(X)\sigma_x$, we have

$$T\sigma_z T^\dagger = \cosh(2X)\sigma_z - i\,\sinh(2X)\sigma_y. \tag{A2}$$

We can also consider T as a spin-dependent displacement operator, with $Ta_kT^\dagger = a_k - \sigma_x\xi_k^*$. As such,

$$\begin{aligned} T\left[\sum_k \omega_k a_k^\dagger a_k + \sum_k (g_k^* a_k^\dagger + g_k a_k)\sigma_x\right]T^\dagger &= \sum_k \omega_k (a_k^\dagger - \sigma_x\xi_k)(a_k - \sigma_x\xi_k^*) \\ &\quad + \sum_k [g_k^*(a_k^\dagger - \sigma_x\xi_k) + g_k(a_k - \sigma_x\xi_k^*)]\sigma_x. \end{aligned} \tag{A3}$$

Next we employ the expansion,

$$e^S\frac{\partial}{\partial t}e^{-S} = -\dot{S} - \frac{1}{2}[S,\dot{S}] - \frac{1}{6}[S,[S,\dot{S}]] - \cdots, \tag{A4}$$

which gives

$$\begin{aligned} T\frac{\partial}{\partial t}T^\dagger &= -\sigma_x\dot{X} - \frac{1}{2}[X,\dot{X}] - \cdots \\ &= -\sigma_x\sum_k(\dot{\xi}_k^* a_k^\dagger - \dot{\xi}_k a_k) - \frac{1}{2}\sum_k(\xi_k^*\dot{\xi}_k - \xi_k\dot{\xi}_k{}^*), \end{aligned} \tag{A5}$$

where $\dot{\xi}_k = \frac{d}{dt}\xi_k$. This is exact since the second-order nested commutator is a c number, causing all higher-order nested commutators to vanish. The transformed Hamiltonian is therefore

$$\begin{aligned} H' &= \sum_k \omega_k(a_k^\dagger - \sigma_x\xi_k)(a_k - \sigma_x\xi_k^*) \\ &\quad + \sum_k [g_k^*(a_k^\dagger - \sigma_x\xi_k) + g_k(a_k - \sigma_x\xi_k^*)]\sigma_x \\ &\quad + \frac{\omega_e}{2}\left\{\cosh\left[2\sum_k(\xi_k^* a_k^\dagger - \xi_k a_k)\right]\sigma_z \right. \\ &\quad \left. - i\,\sinh\left[2\sum_k(\xi_k^* a_k^\dagger - \xi_k a_k)\right]\sigma_y\right\} \\ &\quad + i\sigma_x\sum_k(\dot{\xi}_k^* a_k^\dagger - \dot{\xi}_k a_k) + \frac{i}{2}\sum_k(\xi_k^*\dot{\xi}_k - \xi_k\dot{\xi}_k^*). \end{aligned} \tag{A6}$$

We expand $\cosh(2X)$ and $\sinh(2X)$ in powers of ξ. By keeping terms up to ξ^2, we obtain the form of H' given in Eq. (4). Note that for quantum states near the vacuum considered in this paper, ξ^3 and higher power terms in the expansion can be neglected provided that $\sum_k |\xi_k|^2 \ll 1$.

APPENDIX B: COUPLING η_k OF A MOVING ATOM IN FREE SPACE

The interaction between a moving atom and the electromagnetic field takes the following form under the dipole approximation:

$$\begin{aligned} H_{\rm int} = &-\mathbf{d}\cdot\mathbf{E}^\perp(\mathbf{r}_A) + \frac{1}{2m_A}\{\mathbf{p}_A\cdot[\mathbf{d}\times\mathbf{B}(\mathbf{r}_A)] \\ &+ [\mathbf{d}\times\mathbf{B}(\mathbf{r}_A)]\cdot\mathbf{p}_A\}, \end{aligned} \tag{B1}$$

where $\mathbf{d}$ is the electric dipole of the atom, $\mathbf{E}^\perp(\mathbf{r}_A)$ and $\mathbf{B}(\mathbf{r}_A)$ are the transverse electric and magnetic fields at the position of the atom, $\mathbf{r}_A$, $\mathbf{p}_A$ is the canonical momentum of the center of mass of the atom, and m_A is the mass of the atom. The second term is known as the Röntgen term [36,37], which arises from the magnetic field interacting with the magnetic dipole moment due to the motion of the electric dipole.

By expanding the field operators in plane-wave modes in free space, the interaction Hamiltonian can be written as

$$H_{\rm int} = \sum_{\mathbf{k},s}[g_{\mathbf{k},s}(t)a_{\mathbf{k},s} + g_{\mathbf{k},s}^*(t)a_{\mathbf{k},s}^\dagger]\sigma_x, \tag{B2}$$

where

$$\begin{aligned} g_{\mathbf{k},s} = \chi_k e^{i\mathbf{k}\cdot\mathbf{r}_A(t)}\{&\hat{d}\cdot\hat{\epsilon}_{\mathbf{k},s} + \boldsymbol{\beta}(t) \\ &\cdot[\hat{\epsilon}_{\mathbf{k},s}(\hat{d}\cdot\hat{k}) - \hat{k}(\hat{d}\cdot\hat{\epsilon}_{\mathbf{k},s})]\}, \end{aligned} \tag{B3}$$

with

$$\chi_k = \sqrt{\frac{\omega_k}{2\epsilon_0\hbar V}}d, \tag{B4}$$

$$\mathbf{d} = \langle e|e\mathbf{r}|g\rangle = d\hat{d}, \tag{B5}$$

$$\mathbf{k} = k\hat{k}, \tag{B6}$$

$$\boldsymbol{\beta}(t) = \frac{\dot{\mathbf{r}}_A(t)}{c}. \tag{B7}$$

Here V is the quantization volume and $\epsilon_{\mathbf{k},s}$ is the polarization unit vector of mode $\mathbf{k}$ with s polarization. All hatted quantities are unit vectors.

Next we consider the trajectory of the atom given by $\mathbf{r}_A(t) = r_m\cos\omega_m t\,\hat{r}_m$ with $r_m\omega_m \ll c$ in the nonrelativistic regime. In addition, we assume the long-wavelength condition $kr_m \ll 1$ for field modes that are effectively involved in the two-photon emission. This condition is consistent with the fact that the two photons emitted have their sum of frequencies around ω_m for nonrelativistic motion. Consequently, we take the approximation $e^{i\mathbf{k}\cdot\mathbf{r}_A(t)} \approx 1 + i\mathbf{k}\cdot\mathbf{r}_A(t)$, and obtain

$$\eta_{\mathbf{k},s}(t) = \eta_{\mathbf{k},s}^0 + ik_m r_m(e^{i\omega_m t}\eta_{\mathbf{k},s}^+ + e^{-i\omega_m t}\eta_{\mathbf{k},s}^-), \tag{B8}$$

where $k_m = \omega_m/c$, and we have defined the following real quantities:

$$\eta_{\mathbf{k},s}^0 \equiv \frac{\chi_k}{1+\frac{\omega_k}{\omega_e}}2(\hat{d}\cdot\hat{\epsilon}_{\mathbf{k},s}), \tag{B9}$$

$$\eta^{\pm}_{\mathbf{k},s} \equiv \frac{\chi_k}{1+\frac{\omega_k}{\omega_e}}\left\{\left(\frac{\mathbf{k}}{k_m}\cdot\hat{r}_m\right)(\hat{d}\cdot\hat{\epsilon}_{\mathbf{k},s}) \pm \hat{r}_m\cdot[\hat{\epsilon}_{\mathbf{k},s}(\hat{d}\cdot\hat{k}) - \hat{k}(\hat{d}\cdot\hat{\epsilon}_{\mathbf{k},s})]\right\}. \quad \text{(B10)}$$

APPENDIX C: CALCULATION OF THE THREE-PHOTON AMPLITUDE GENERATED BY SINGLE-PHOTON SCATTERING

We start with the initial state Eq. (25),

$$|\psi(0)\rangle = \left(1+\sum_{kk'}\frac{\Lambda_{kk'}}{\omega_{kk'}}a_k^\dagger a_{k'}^\dagger\right)\sum_k W_k a_k^\dagger|g,0\rangle, \quad \text{(C1)}$$

where $\Lambda_{kk'}$ are time independent.

The first-order time-dependent state is given by the second term in Eq. (17). To evaluate the integral, we note that the dressed photon pairs have a very broad spectrum, so they barely interact with the atom through $U(t)$. Hence we can approximate their evolution under $U(t)$ as free, i.e.,

$$U(t)\sum_{kk'}\frac{\Lambda_{kk'}}{\omega_{kk'}}a_k^\dagger a_{k'}^\dagger U^\dagger(t) \approx \sum_{kk'}\frac{\Lambda_{kk'}}{\omega_{kk'}}a_k^\dagger a_{k'}^\dagger e^{-i(\omega_k+\omega_{k'})t}. \quad \text{(C2)}$$

Hence for the first term in Eq. (17),

$$U(t)|\psi(0)\rangle \approx \left(1+\sum_{kk'}\frac{\Lambda_{kk'}}{\omega_{kk'}}a_k^\dagger a_{k'}^\dagger e^{-i(\omega_k+\omega_{k'})t}\right) \times U(t)\sum_{k''}W_{k''}a_{k''}^\dagger|g,0\rangle. \quad \text{(C3)}$$

As we shall see, the last term in Eq. (C3) does not contribute to three-photon emission because its propagating part will be canceled by the second term in Eq. (17).

For the second term in Eq. (17), we focus on the part that corresponds to three photons in the final state, which is

$$-i\int_0^t d\tau\, U(t-\tau)\sum_{kk'}\Lambda_{kk'}a_k^\dagger a_{k'}^\dagger\sigma_z U(\tau)W^\dagger|g,0\rangle,$$

where we have dropped the dressing terms in $|\psi(0)\rangle$ since their contributions are of higher order. Next we insert $I = U^\dagger(t-\tau)U(t-\tau)$ after $a_k^\dagger a_{k'}^\dagger$ and make approximations similar to Eq. (C2); this gives

$$\begin{aligned} &-i\int_0^t d\tau\, U(t-\tau)\sum_{kk'}\Lambda_{kk'}a_k^\dagger a_{k'}^\dagger\sigma_z U(\tau)W^\dagger|g,0\rangle \\ &\approx +i\int_0^t d\tau\sum_{kk'}\Lambda_{kk'}a_k^\dagger a_{k'}^\dagger e^{-i(\omega_k+\omega_{k'})(t-\tau)}U(t)W^\dagger|g,0\rangle \\ &\quad -2i\int_0^t d\tau\sum_{kk'}\Lambda_{kk'}a_k^\dagger a_{k'}^\dagger e^{-i(\omega_k+\omega_{k'})(t-\tau)} \\ &\quad \times U(t-\tau)|e\rangle\langle e|U(\tau)W^\dagger|g,0\rangle, \end{aligned} \quad \text{(C4)}$$

where we have replaced σ_z by $2|e\rangle\langle e| - 1$. In the first integral of Eq. (C4), the lower limit cancels the propagating term in Eq. (C3), leaving the upper limit as the original dressed photon pair which is bounded to the atom after the scattering.

The second integral of Eq. (C4) contains the photon pair production terms dependent on population in the atomic excited state. It is this integral that determines the three-photon emission.

To evaluate $U(t-\tau)|e\rangle\langle e|U(\tau)W^\dagger|g,0\rangle$, we note that

$$U(t)|e\rangle = e^{(-\gamma-i\omega_e)t/2}|e\rangle + \sum_k \frac{\eta_k^* e^{-i\omega_e t/2}}{\frac{\gamma}{2}-i\Delta_k}\left(e^{-i\Delta_k t} - e^{-\gamma t/2}\right)|g,k\rangle \quad \text{(C5)}$$

is the well-known solution to spontaneous atomic decay. In addition, if we choose W_k to take the Lorentzian form given by Eq. (27), then the single-photon scattering excited-state amplitude is

$$\langle e|U(\tau)W^\dagger|g,0\rangle = \frac{2i\sqrt{\gamma'\gamma}}{\gamma-\gamma'}\left(e^{-\frac{\gamma}{2}\tau} - e^{-\frac{\gamma'}{2}\tau}\right)e^{-i\frac{\omega_e}{2}\tau}. \quad \text{(C6)}$$

By using Eqs. (C5)- and (C6), and working out the second integral of Eq. (C4), we obtain the freely propagating three-photon amplitude given by Eqs. (28)- and (29) in the long-time limit.

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