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Efficient single-photon absorption by a trapped moving atom

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The influence of the center-of-mass motion of a trapped two-level system on efficient resonant single-photon absorption is investigated. It is shown that this absorption process depends strongly on the ratio between the characteristic time scales of spontaneous photon emission and of the two-level system's center-of-mass motion. In particular, if the spontaneous photon emission process occurs almost instantaneously on the time scale of the center-of-mass motion, coherent control of the center-of-mass motion offers interesting perspectives for optimizing single-photon absorption. It is demonstrated that time-dependent modulation of a harmonic trapping frequency allows to squeeze the two-level system's center-of-mass motion so strongly that high efficient single-photon absorption is possible even in cases of weak confinement by a trapping potential.

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I. INTRODUCTION

Recent technological advances in the area of resonant photon-matter interaction have opened new exciting experimental possibilities [1–3]. One line of research in this direction focuses on the development of efficient means for coupling a single elementary quantum system, such as a trapped atom or ion, to properly engineered multimode radiation fields in order to achieve a controlled and almost perfect transfer of excitation between few-photon multimode states and material quantum systems [4,5]. Aside from being of fundamental quantum optical interest, these investigations are also driven by the desire to explore new possibilities for realizing efficient ways of transferring quantum information between material elementary two-level systems (stationary qubits) and photons (flying qubits) [6].

Recently, it has been demonstrated experimentally that by trapping a single elementary quantum system in the center of a parabolic mirror, an efficient coupling to optical photonic multimode states can be achieved [4,5,7]. Thereby, a parabolic mirror constitutes a convenient tool for redirecting and focusing an asymptotically incoming (almost) plane wave containing a few photons onto an elementary material quantum system trapped in the parabola's focus. For focal lengths and distances large in comparison with the wavelengths of optical photons, the quantized radiation field dominantly coupling to such a trapped material quantum system looks like a dipole field in free space whose center is located in the focus of the parabola. Guided by characteristic features of the spontaneous photon emission process, it has been demonstrated theoretically that even in free space optimal single-photon wave-packet states exist which are capable of exciting an elementary two-level system at a fixed position in space almost perfectly. However, the center-of-mass motion of an absorbing material quantum system complicates the situation considerably because, in general, single-photon absorption and the subsequent spontaneous photon emission process together with the resulting recoil effects [8] entangle the center of mass and the photonic degrees of freedom in an intricate way. On the basis of these general features it appears that achieving almost perfect single-photon excitation in free space or with the help of a parabolic mirror in the presence of center-of-mass motion requires preparation of a highly entangled quantum state of the center of mass and the photonic degrees of freedom. Even by nowadays technological capabilities, the controlled preparation of such entangled quantum states constitutes a major technological obstacle and appears unrealistic.

Motivated by these developments, we explore the influence of the center-of-mass motion of a trapped material two-level system on single-photon absorption in free space or equivalently in a parabolic cavity with large focal length. In particular, we explore possibilities for optimizing this process with the help of the particular single-photon wave packet [9] which would achieve almost perfect absorption in the absence of any center-of-mass motion. At first sight, one may be tempted to conclude that almost perfect excitation of a trapped two-level system by such an optimal single-photon wave packet is only achievable in sufficiently strongly confining traps with large trap frequencies. However, our investigations demonstrate that almost perfect photon absorption from such an optimal single-photon wave packet is possible even in weakly confining traps and for initially prepared thermal states of the center-of-mass motion provided the trap frequency is modulated periodically in an appropriate way. Our analysis exhibits also the crucial role played by the characteristic dynamical parameters, namely, the spontaneous photon emission rate of the electronic transition involved and the trap frequency. The investigation of the impact of the center-of-mass motion on atom field interactions for high-finesse cavities has recently also attracted attention in the literature (see [10] for example).

This paper is organized as follows. In Sec. II, the quantum electrodynamical model describing single-photon absorption by a trapped moving two-level system is presented. Based on the dipole and rotating-wave approximations, the dynamics of the single-photon excitation process and its relation to the relevant field correlation function is discussed in Sec. III. In Sec. IV, characteristic features of the excitation probability and its deviation from the ideal motionless case are investigated for tightly confining trapping potentials. In the subsequent Sec. V, these results are generalized to weakly confining trapping potentials with particular emphasis on the experimentally interesting dynamical regime of spontaneous photon emission rates large in comparison with relevant trapping frequencies. Section VI finally explores possibilities to optimize the excitation probability by periodic modulation of the trap frequency of a harmonic trap.

II. A QUANTUM ELECTRODYNAMICAL MODEL

We consider a single trapped atom or ion, whose internal electronic dynamics is modeled by a two-level system resonantly coupled to an optical single-photon radiation field and explore its capability of absorbing this single-photon almost perfectly. Inspired by recent experiments [4,5,7] we assume that the center of the trap is positioned in the focal point of a large parabolic mirror which is capable of focusing a well-directed asymptotically incoming single-photon radiation field towards the two-level system trapped close to the focal point of the parabola. This excitation process is depicted in Fig. 1 schematically.

In order to describe the dynamics of this single-photon excitation process, we take advantage of the fact that in the optical frequency regime, typical wavelengths are large in comparison with the Bohr radius characterizing spatial extensions of atoms or ions in energetically low-lying bound electronic states. Therefore, the interaction between the two-level system and the radiation field can be described in the dipole approximation. Furthermore, we assume that the time scale of typical optical transitions is orders of magnitude smaller than all other interaction-induced time scales, i.e., the optical transitions frequency ω_{eg} is large in comparison to the spontaneous decay rate in free space Γ. For atoms or ions interacting with the radiation field in the optical frequency regime, this is well satisfied (see for example the parameters of the experiment described in [7]) (with $\Gamma = 1.2 \times 10^8\ \mathrm{s}^{-1}$ and $\omega_{eg} = 5.1 \times 10^{15}\ \mathrm{s}^{-1}$). In this regime, the rotating-wave approximation is applicable. In addition, the velocity of the two-level system's center-of-mass motion is assumed to be negligible in comparison with the speed of light in vacuum c so that the dynamics of the center-of-mass motion can be described in the nonrelativistic approximation. Under these conditions, the total Hamiltonian governing the dynamics of the quantum electrodynamical interaction between the moving two-level system and the radiation field is given by

$$\hat{H} = \hat{H}_{\mathrm{E}} + \hat{H}_{\mathrm{T}} + \hat{H}_{\mathrm{F}} + \hat{H}_{\mathrm{int}}. \tag{2.1}$$

The free evolution of the radiation field is described by the Hamiltonian $\hat{H}_{\mathrm{F}} = \hbar \sum_i \omega_i \hat{a}_i^\dagger \hat{a}_i$ with the photonic destruction and creation operators $\hat{a}_i$ and $\hat{a}_i \dagger$ and with i indexing the modes with frequency ω_i of the transverse radiation field. The Hamiltonians $\hat{H}_{\mathrm{E}} = E_g|g\rangle\langle g| + E_e|e\rangle\langle e|$ and $\hat{H}_{\mathrm{T}} = \hat{\mathbf{p}}^2/2m + V_{\mathrm{T}}(\hat{\mathbf{x}})$ describe the dynamics of the electronic degrees of freedom and of the center of mass of the trapped two-level system. The frequency of the relevant electronic transition is denoted $\omega_{eg} = (E_e - E_g)/\hbar$ and $\hat{\mathbf{x}}$, $\hat{\mathbf{p}}$, and m are position operator, momentum operator, and mass of the two-level system's center-of-mass degrees of freedom. The Hamiltonian $\hat{H}_{\mathrm{int}} = -\hat{\mathbf{E}}^{(+)}(\hat{\mathbf{x}}) \cdot \mathbf{d}|e\rangle\langle g| + \mathrm{H.c.}$ describes the interaction between the two-level system and the transverse radiation field in the dipole and rotating-wave approximations with the two-level system's dipole matrix element $\mathbf{d} = \langle e|\hat{\mathbf{d}}|g\rangle$ and its dipole operator $\hat{\mathbf{d}}$. The mode decomposition of the positive part of the transverse electric field operator is given by

$$\hat{\mathbf{E}}^{(+)}(\mathbf{x}) = \sum_j i\sqrt{\frac{\hbar\omega_j}{2\epsilon_0}}\mathbf{g}_j(\mathbf{x})\hat{a}_j. \tag{2.2}$$

The orthonormal transverse mode functions $\mathbf{g}_i(\mathbf{x})$ are solutions of the Helmholtz equation with appropriate boundary conditions and with unit normalization per field mode. (ϵ_0 is the dielectric constant of the vacuum.)

In order to describe the dynamics of the absorption by a single photon, we have to solve the time-dependent Schrödinger equation with Hamiltonian (2.1). In particular, we are interested in its solution with a separable pure state of the form

$$|\psi_0\rangle = |g\rangle\,|\psi_{\mathrm{T}}\rangle\,|\psi_{\mathrm{F}}\rangle \tag{2.3}$$

prepared initially at time $t = t_0$. Thereby, $|\psi_{\mathrm{T}}\rangle$ describes the pure initial state of the center-of-mass degree of freedom and $|\psi_{\mathrm{F}}\rangle$ denotes the initial one-photon state of the radiation field. In the following, we are particularly interested in the center-of-mass induced dynamics of this single-photon excitation process for a photon state which is capable of exciting a two-level system located at the fixed position $\mathbf{x} = 0$, such as the focal point of a parabolic mirror, almost perfectly.

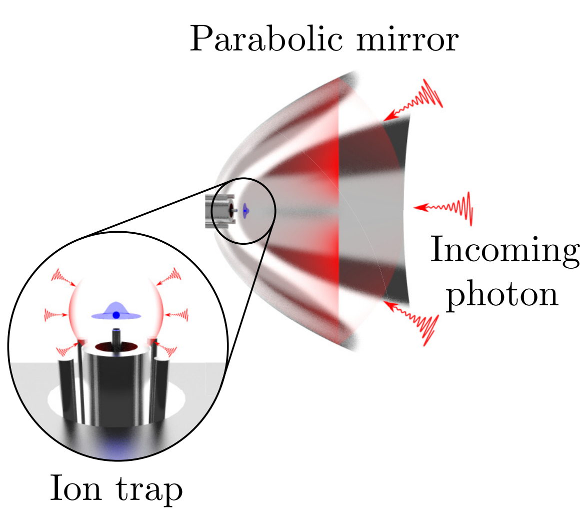

FIG. 1. Single-photon absorption process in a parabolic cavity: a two-level system trapped close to the focal point $\mathbf{x} = 0$ of a parabolic cavity is excited by an asymptotically incoming single-photon wave packet. This wave packet couples to the two-level system in the same way as a single-photon wave packet capable of exciting this two-level system almost perfectly in free space [see Eq. (3.4)]. A suitable setup with a trapped ion at the focal point of the parabolic mirror has been described in [4,7,11].

III. DYNAMICS OF SINGLE-PHOTON ABSORPTION

In this section, we explore the dynamics of optimal resonant single-photon absorption (in free space) by a moving trapped two-level system whose Hamiltonian is given by Eq. (2.1).

In view of the rotating-wave approximation, the Hamiltonian (2.1) conserves the numbers of excitations. Therefore, for an initial state of the form of Eq. (2.3) the general solution of the Schrödinger equation is given by a pure quantum state which is a superposition of the photonic vacuum state correlated with the excited two-level system and of a single-photon multimode state correlated with the two-level system in its ground state and with its center-of-mass degrees of freedom generally entangled with the field modes. As the center-of-mass motion is assumed to be nonrelativistic, the resulting modification of the spontaneous photon emission process from the excited two-level system is negligible so that it is still described by the spontaneous decay rate Γ in free space, i.e.,

$$\Gamma = \frac{\omega_{eg}^3 |\mathbf{d}|^2}{3\pi\epsilon_0\hbar c^3}. \tag{3.1}$$

Thus, from the time-dependent Schrödinger equation we find that the probability of detecting at time t the two-level system in its excited state $|e\rangle$ is given by

$$P_e(t) = \int_{t_0}^{t} dt_1 e^{i(\omega_{eg}+i\Gamma/2)(t-t_1)} \int_{t_0}^{t} dt_2 e^{-i(\omega_{eg}-i\Gamma/2)(t-t_2)} \times \frac{\mathbf{d}^*}{\hbar}\cdot\langle\psi_T|\mathbf{G}^{(1)}(\hat{\mathbf{x}}_I(t_1),t_1,\hat{\mathbf{x}}_I(t_2),t_2))|\psi_T\rangle\cdot\frac{\mathbf{d}}{\hbar}. \tag{3.2}$$

This excitation probability is determined by the mean value of the normally ordered field correlation tensor of first order, i.e.,

$$\mathbf{G}^{(1)}(\hat{\mathbf{x}}_I(t_1),t_1,\hat{\mathbf{x}}_I(t_2),t_2) = \langle\psi_F|\hat{\mathbf{E}}^{-}(\hat{\mathbf{x}}_I(t_1),t_1)\otimes\hat{\mathbf{E}}^{+}(\hat{\mathbf{x}}_I(t_2),t_2)|\psi_F\rangle,$$

averaged over the two-level system's initially prepared center-of-mass state $|\psi_T\rangle$. Thereby, the position operator

$$\hat{\mathbf{x}}_I(t) = \exp[i\hat{H}_T(t-t_0)/\hbar]\hat{\mathbf{x}}\exp[-i\hat{H}_T(t-t_0)/\hbar]$$

denotes the time evolution of the center-of-mass position of the two-level system in the interaction picture. In this interaction picture, the transverse electric field operators $\hat{\mathbf{E}}^{(+)}(\mathbf{x}_I(t),t)$ and $\hat{\mathbf{E}}^{(-)}(\mathbf{x}_I(t),t) = [\hat{\mathbf{E}}^{(+)}(\mathbf{x}_I(t),t)]^\dagger$ are defined by

$$\hat{\mathbf{E}}^{(+)}(\mathbf{x},t) = \sum_j i\sqrt{\frac{\hbar\omega_j}{2\epsilon_0}}\mathbf{g}_j(\mathbf{x})e^{-i\omega_j(t-t_0)}\hat{a}_j. \tag{3.3}$$

Let us now concentrate on an initially prepared one-photon state $|\psi_F\rangle$ which achieves almost perfect excitation of a two-level system positioned at the fixed position $\mathbf{x}=0$ in free space. It has been demonstrated [9] that the pure single-photon state

$$|\psi_F\rangle = \sum_j \hat{a}_j^\dagger|0\rangle\frac{1}{\hbar}\sqrt{\frac{\hbar\omega_j}{2\epsilon_0}}\frac{\mathbf{d}^*\cdot\mathbf{g}_j(\mathbf{x}=0)}{\omega_j-\omega_{eg}-i\Gamma/2}e^{i\omega_j(t_{\text{out}}-t_0)} \tag{3.4}$$

prepared at time t_0 with $\Gamma(t_{\text{out}}-t_0)\gg 1$ achieves such an almost perfect excitation at time t_{out} in the absence of any center-of-mass motion. The frequencies ω_i of this one-photon state are distributed according to a Lorentzian spectrum centered resonantly around the two-level system's transitions frequency ω_{eg}. The relative phases between these modes are determined by the parameter t_{out} which describes the time at which a two-level system at the fixed position $\mathbf{x}=0$ is excited almost perfectly. If the two-level system were positioned in the center of a spherically symmetric cavity of radius R with ideally conducting walls, for example, the discrete orthonormal mode functions coupling to the two-level system in the dipole approximation would be given by

$$\mathbf{g}_n(\mathbf{x}) = \sqrt{\frac{1}{R}}\nabla\times[j_1(\omega_n r/c)\mathbf{x}\times\nabla Y_1^0(\theta,\varphi)] = -\sqrt{\frac{3}{4\pi R}}\frac{\omega_n}{c}\left[e^{i\omega_n r/c}\mathbf{g}_{\omega_n}^{(+)}(\mathbf{x}) + e^{-i\omega_n r/c}\mathbf{g}_{\omega_n}^{(-)}(\mathbf{x})\right] \tag{3.5}$$

with

$$\mathbf{g}_\omega^{(+)}(\mathbf{x}) = -\mathbf{e}_r\cos\theta\left[\frac{1}{(\omega r/c)^2}+\frac{i}{(\omega r/c)^3}\right] - \mathbf{e}_\theta\frac{\sin\theta}{2}\left\{\frac{1}{(\omega r/c)^2} - i\left[\frac{1}{(\omega r/c)} - \frac{1}{(\omega r/c)^3}\right]\right\} \tag{3.6}$$

and with the angular momentum eigenfunction $Y_1^0(\theta,\varphi)$ [12]. For real-valued frequencies ω the relation $\mathbf{g}_\omega^{(-)}(\mathbf{x}) = [\mathbf{g}_\omega^{(+)}(\mathbf{x})]^*$ applies. Here, $j_1(u) = \sin u/u^2 - \cos u/u$ denotes the spherical Bessel function of fractional order [12] and $r = |\mathbf{x}|$. The angle between the direction of the dipole matrix element $\mathbf{d}$ and $\mathbf{x}$ is denoted θ. Furthermore, $\mathbf{e}_r$ and $\mathbf{e}_\theta$ are the spherical coordinate unit vectors in the r and θ directions with the z direction oriented parallel to the dipole vector $\mathbf{d}$. In the continuum limit of large cavity sizes, i.e., $\omega_{eg}R/c \gg 1$, the mode frequencies are given approximately by $\omega_n = c\pi(n+1)/R$ with $n \geqslant 0$ being integer.

In accordance with current experimental activities [4,5,7] and with the scenario depicted in Fig. 1, this excitation process can be realized with the help of a parabolic mirror with large focal length f and with the two-level system's trap centered in the parabola's focal point $\mathbf{x}=0$. In the geometric optical limit in which f is large in comparison with the photon's wavelengths, the optimal single-photon state of Eq. (3.4) can be prepared by focusing an appropriately polarized (almost) plane-wave single-photon state which asymptotically enters the parabola along the direction of its symmetry axis [13].

For the pure one-photon state $|\psi_F\rangle$, the first-order correlation tensor factorizes, i.e.,

$$\mathbf{G}^{(1)}(\hat{\mathbf{x}}_I(t_1),t_1,\hat{\mathbf{x}}_I(t_2),t_2) = \mathbf{F}^*(\hat{\mathbf{x}}_I(t_1),t_1)\otimes\mathbf{F}(\hat{\mathbf{x}}_I(t_2),t_2)$$

with the effective one-photon operator $\mathbf{F}(\hat{\mathbf{x}}_I(t),t) = \langle 0|\hat{\mathbf{E}}^{(+)}(\hat{\mathbf{x}}_I(t),t)|\psi_F\rangle$. For our initial state, we get

$$\mathbf{F}(\mathbf{x},t) = \frac{i}{2\epsilon_0}\sum_n \omega_n\mathbf{g}_n(\mathbf{x})e^{-i\omega_n(t-t_0)} \times\frac{\mathbf{d}^*\cdot\mathbf{g}_n(\mathbf{x}=0)}{\omega_n-\omega_{eg}-i\Gamma/2}e^{i\omega_n(t_{\text{out}}-t_0)} = \frac{i}{\epsilon_0}\sum_n\frac{\omega_n^3}{4\pi Rc^2}\left[e^{-i\omega_n t_+}\mathbf{g}_{\omega_n}^{(+)}(\mathbf{x}) + e^{-i\omega_n t_-}\mathbf{g}_{\omega_n}^{(-)}(\mathbf{x})\right] \times\frac{\mathbf{d}^*\cdot e_z}{\omega_n-\omega_{eg}-i\Gamma/2} \tag{3.7}$$

with $t_\pm = t - t_{\text{out}} \mp r/c$. In the continuum limit, i.e., $R \to \infty$, we can replace the sum by an integral. We obtain

$$\mathbf{F}(\mathbf{x},t) \underset{R\to\infty}{=} \frac{i}{4c^3\epsilon_0\pi^2}\int_0^\infty d\omega[e^{-i\omega t_+}\mathbf{g}_\omega^{(+)}(\mathbf{x}) + e^{-i\omega t_-}\mathbf{g}_\omega^{(-)}(\mathbf{x})] \times \frac{\omega^3\mathbf{d}^*\cdot e_z}{\omega - \omega_{eg} - i\Gamma/2}. \tag{3.8}$$

By extending the region of integration to $-\infty$ (which is well justified for $\omega_{eg} \gg \Gamma$) and by applying Cauchy's residue theorem, we obtain the following analytical expression:

$$\mathbf{F}(\mathbf{x},t) = -\frac{\hbar\Gamma 3}{\mathbf{d}\cdot\mathbf{e}_z 2}[\mathbf{g}_\omega^{(+)}(\mathbf{x})\Theta(-t_+)e^{-i\omega t_+} + \mathbf{g}_\omega^{(-)}(\mathbf{x})\Theta(-t_-)e^{-i\omega t_-}]|_{\omega=\omega_{eg}+i\Gamma/2}. \tag{3.9}$$

Close to the ideal position $\mathbf{x} = 0$ of the two-level system, i.e., for $\eta = \omega_{eg}r/c \ll 1$, this one-photon operator simplifies to

$$\mathbf{F}(\mathbf{x},t) = -\frac{\hbar\Gamma}{\mathbf{d}\cdot\mathbf{e}_z}e^{i(\omega_{eg}+i\Gamma/2)(t_{\text{out}}-t)}\Theta(t_{\text{out}}-t) \times \left[\mathbf{e}_z\left(1-\frac{\eta^2}{10}\right) + \mathbf{e}_\theta\sin\theta\frac{\eta^2}{10} + O(\eta^4)\right]. \tag{3.10}$$

Inserting Eq. (3.9) into Eq. (3.2) yields the time dependence of the excitation probability $P_e(t)$. For a two-level system fixed at position $\mathbf{x} = 0$ and for times $t \geqslant t_0$, this one-photon excitation probability reduces to

$$P_{id}(t) = e^{-\Gamma|t_{\text{out}}-t|}(1 - e^{-\Gamma(\tau - t_0)})^2 \tag{3.11}$$

with $\tau = t\Theta(t_{\text{out}} - t) + t_{\text{out}}\Theta(t - t_{\text{out}})$. Thus, for large interaction times, i.e., $\Gamma(\tau - t_0) \gg 1$, the single-photon state of Eq. (3.4) achieves almost perfect excitation of the two-level system at time t_{out} apart from terms exponentially small in the parameter $\Gamma(\tau - t_0) \gg 1$.

Depending on the ratio between the characteristic time scale of the center-of-mass motion and of the single-photon absorption and spontaneous emission process, two extreme dynamical cases can be distinguished. If the trap frequency ω_T of a harmonically trapped two-level system is much larger than the spontaneous decay rate Γ, the spontaneous decay process is so slow that details of the center-of-mass motion are averaged out in the time integrals of Eq. (3.2). Consequently, the excitation probability dominantly depends on the time-averaged center-of-mass motion. In the opposite limit, i.e., $\omega_T \ll \Gamma$, the spontaneous photon emission process occurs almost instantaneously on the time scale of the center-of-mass motion. Consequently, the time evolution of the excitation probability depends on details of the two-level system's center-of-mass motion in the trap.

IV. SINGLE-PHOTON ABSORPTION AND STRONG CONFINEMENT OF THE CENTER-OF-MASS MOTION

Inserting Eq. (3.10) into Eq. (3.2), a systematic understanding of the influence of the two-level system's center-of-mass motion on the single-photon absorption process can be obtained in cases in which this motion is confined to a region close to the ideal position $\mathbf{x} = 0$ in the sense that $\eta = \omega_{eg}r/c \ll 1$. From Eqs. (3.10) and (3.2), we obtain the result

$$P_e(t) = \Gamma^2\int_{t_0}^t dt_1\int_{t_0}^t dt_2\Theta(t_{\text{out}}-t_1)\Theta(t_{\text{out}}-t_2)e^{-\Gamma(t+t_{\text{out}}-t_1-t_2)} \times \left\{1 - 2\frac{\omega_{eg}^2}{10c^2}\langle\psi_T|\left[\hat{z}_I^2(t_2) + 2\hat{x}_I^2(t_2) + 2\hat{y}_I^2(t_2)\right]|\psi_T\rangle + O(\eta^4)\right\} \tag{4.1}$$

with $\hat{x}_I(t)$, $\hat{y}_I(t)$, $\hat{z}_I(t)$ denoting the time-dependent Cartesian x, y, z components of the position operator of the center-of-mass degrees of freedom in the interaction picture. Thereby, the z direction is oriented parallel to the dipole vector $\mathbf{d}$.

Let us investigate the center-of-mass motion in an anisotropic harmonic trapping potential of the form

$$V_T(\mathbf{x}) = m\omega_z^2z^2/2 + m\omega_x^2x^2/2 + m\omega_y^2y^2/2 \tag{4.2}$$

in more detail. In the interaction picture, the resulting dynamics of the z component of the center-of-mass position is given by

$$\hat{z}_I(t) = \hat{z}\cos[\omega_z(t-t_0)] + \frac{\hat{p}_z}{m\omega_z}\sin[\omega_z(t-t_0)] \tag{4.3}$$

with analogous expressions for the other Cartesian components. Position and momentum operators in the Schrödinger picture are denoted by $\hat{z}$ and $\hat{p}_z$, etc. Inserting these position operators into Eq. (4.1) yields the result

$$\frac{P_e(t)}{P_{id}(t)} = 1 - \frac{\omega_{eg}^2}{5c^2}\left[\frac{\hbar A_z(\tau)}{2m\omega_z} + 2\sum_{j=x,y}\frac{\hbar A_j(\tau)}{2m\omega_j}\right] \tag{4.4}$$

with $\tau = t\Theta(t_{\text{out}} - t) + t_{\text{out}}\Theta(t - t_{\text{out}})$ and with

$$A_j(\tau) = \langle\psi_T|\hat{b_j}^{\dagger 2}|\psi_T\rangle\frac{\Gamma e^{2i\omega_j(\tau-t_0)}}{\Gamma + 2i\omega_j}\frac{1 - e^{-(\Gamma+2i\omega_j)(\tau-t_0)}}{1 - e^{-\Gamma(\tau-t_0)}} + \langle\psi_T|(\hat{b_j}^\dagger\hat{b_j} + 1/2)|\psi_T\rangle + \text{c.c.} \tag{4.5}$$

for $j = x,y,z$. The creation and destruction operators of the harmonic oscillators in the Cartesian directions $j = x,y,z$ are denoted by $\hat{b}_j^\dagger$ and $\hat{b}_j$.

According to Eq. (4.4) for long interaction times, i.e., $\tau - t_0 \gg 1/\Gamma$, and for small spontaneous photon emission rates, i.e., $\Gamma \ll \omega_j$, the excitation probability $P_e(t)$ is determined by the time-averaged center-of-mass motion. In this dynamical regime we have $A_j(\tau) = 2\langle\psi_T|(\hat{b_j}^\dagger\hat{b_j} + 1/2)|\psi_T\rangle + O(\Gamma/\omega_j)$ so that the deviation of the excitation probability $P_e(t)$ from its ideal value $P_{id}(t)$ is proportional to the mean energy of the center-of-mass degrees of freedom in the harmonic trap, i.e.,

$$\frac{P_e(t)}{P_{id}(t)} = 1 - \frac{\omega_{eg}^2}{5c^2}\left[\frac{\hbar\omega_z\langle\psi_T|(\hat{b_z}^\dagger\hat{b_z} + 1/2)|\psi_T\rangle}{m\omega_z^2} + 2\sum_{j=x,y}\frac{\hbar\omega_j\langle\psi_T|(\hat{b_j}^\dagger\hat{b_j} + 1/2)|\psi_T\rangle}{m\omega_j^2}\right]. \tag{4.6}$$

As the mean energy of a harmonic oscillator is lower bounded by its zero-point energy in this dynamical regime, the excitation probability $P_e(t_{\rm out})$ assumes its maximal value if the center-of-mass degrees of freedom are prepared in the ground state of the harmonic trap so that $\hat{b}_j|\psi_T\rangle = 0$ for $j = x,y,z$.

According to Eq. (4.4) in the opposite limit of long interaction times, i.e., $\tau - t_0 \gg 1/\Gamma$, but large spontaneous photon emission rates, i.e., $\Gamma \gg \omega_j$, the excitation probability $P_e(t)$ is determined by the center-of-mass motion at time $\tau = t\Theta(t_{\rm out} - t) + t_{\rm out}\Theta(t - t_{\rm out})$. In this dynamical regime we obtain the approximate result

$$A_j(\tau) = \langle\psi_T|(\hat{b}_j^\dagger e^{i\omega_j(\tau-t_0)} + \hat{b}_j e^{-i\omega_j(\tau-t_0)})^2|\psi_T\rangle \quad (4.7)$$

by neglecting terms of the order of $O(\omega_j/\Gamma)$. The resulting strong dependence of the excitation probability $P_e(t)$ on the center-of-mass motion at time τ can be exploited for minimizing the deviations of the excitation probability from its ideal values $P_{id}(t)$. This can be achieved by preparing appropriate initial states $|\psi_T\rangle$ which minimize the quantities $A_j(\tau)$. For this purpose, squeezed vacuum states [14] of the center-of-mass motion of the form

$$|\psi_T\rangle = \hat{S}(\xi_z)\hat{S}(\xi_x)\hat{S}(\xi_y)|0_T\rangle \quad (4.8)$$

offer interesting possibilities. In Eq. (4.8), $|0_T\rangle$ denotes the ground state of the harmonic trap, i.e., $\hat{b}_j|0_T\rangle = 0$ for $j = x,y,z$ and

$$\hat{S}(\xi_j) = e^{\xi_j^*\hat{b}_j^2/2 - \xi_j\hat{b}_j^{\dagger 2}/2} \quad (4.9)$$

are the squeezing operators for the Cartesian components j. For complex-valued squeezing parameters of the form $\xi_j = r_j\exp(2i\varphi)$ with $r_j > 0$, for example, we obtain the result

$$\frac{P_e(t)}{P_{id}(t)} = 1 - \frac{\omega_{eg}^2}{5c^2}\left(\Delta z_0^2 e^{-2r_z} + 2\Delta x_0^2 e^{-2r_x} + 2\Delta y_0^2 e^{-2r_y}\right) \quad (4.10)$$

at all times t for which $\sin[\omega_j(\tau - t_0) - \varphi] = 0$. Thereby, the quantities $\Delta z_0 = \sqrt{\hbar/(2m\omega_z)}$, etc., denote the extensions of the ground state of the harmonic trap along the corresponding Cartesian directions. In contrast, at times for which $\cos[\omega_j(\tau - t_0) - \varphi] = 0$ these extensions are enhanced periodically by corresponding factors of e^{2r_z}, etc. Thus, strong squeezing of the initial state of Eq. (4.8) in all directions, i.e., $r_j \gg 1$, implies a significant increase of the excitation probability at all interaction times t with $\sin[\omega_j(\tau - t_0)] = 0$ and may lead to significantly more efficient single-photon excitation than achievable in cases of small spontaneous decay rates.

V. SINGLE-PHOTON ABSORPTION AND WEAK CONFINEMENT OF THE CENTER-OF-MASS MOTION

If the center-of-mass motion is not confined to a region around the ideal position $\mathbf{x} = 0$ small in comparison with the wavelength of the resonantly absorbed photon, a Taylor expansion of the mode functions $\mathbf{g}_n(\mathbf{x})$ around $\mathbf{x} = 0$ is no longer adequate. In this case, we start from the general form of the one-photon operator as given by Eq. (3.9). Inserting this expression into Eq. (3.2) yields the excitation probability.

For typical spontaneous decay times $1/\Gamma$ of the order of nanoseconds and extensions of center-of-mass wave packets Δx small in comparison with a few meters so that $\Gamma\Delta x/c \ll 1$, this one-photon operator can be further simplified to the expression

$$\mathbf{F}(\mathbf{x},t) = -\frac{3\hbar\Gamma}{2\mathbf{d}\cdot\mathbf{e}_z}\Theta(t_{\rm out} - t)e^{-i(\omega_{eg}+i\Gamma/2)(t-t_{\rm out})} \times [\mathbf{g}_\omega^{(+)}(\mathbf{x})e^{i\omega r/c} + \mathbf{g}_\omega^{(-)}(\mathbf{x})e^{-i\omega r/c}]\big|_{\omega=\omega_{eg}}. \quad (5.1)$$

In many trapping experiments, typical spontaneous photon emission rates of electronic transitions Γ are large in comparison with typical trap frequencies, i.e., $\Gamma \gg \omega_T$. In this dynamical regime, the single-photon absorption process takes place almost instantaneously on the characteristic time scale induced by the trapping potential. In particular, we demand that for the typical velocities $\Delta v(t) = \sqrt{\langle\psi_T|[\frac{d}{dt}\hat{\mathbf{x}}_I(t)]^2|\psi_T\rangle}$ the condition

$$\Delta v(t)/(\lambda_{eg}\Gamma) \ll 1 \quad (5.2)$$

is fulfilled within a time interval of the order of a few lifetimes of the excited state, i.e., $1/\Gamma$, around $t_{\rm out}$ (λ_{eg} is the wavelength of the optical transition $|e\rangle \to |g\rangle$.). For a thermal state of the harmonic trap, we obtain

$$\Delta v(t) = \sqrt{\langle\hat{H}_j\rangle/m} \text{ for } j \in \{x,y,z\} \quad (5.3)$$

with $\hat{H}_j$ denoting the Hamiltonian describing the dynamics of the center-of-mass motion along the j axis. By using this result, we get for the ground state

$$\Delta v(t) = \sqrt{\omega_j\hbar/(2m)} \text{ for } j \in \{x,y,z\}. \quad (5.4)$$

By using the experimental parameters given in [7] (trapping of ^{174}Yb, $\omega_T = 2\pi \times 480$ kHz, $\Gamma = 1.2 \times 10^8$ s^{-1}, and $\lambda_{eq} = 369$ nm), we obtain $\Delta v(t)/(\lambda_{eg}\Gamma) = 5.3 \times 10^{-4} \ll 1$ for the ground state and $\Delta v(t)/(\lambda_{eg}\Gamma) = 3.3 \times 10^{-3} \ll 1$ for the thermal state in the Doppler limit. Hence, the condition is well satisfied for typical experimental parameters. Condition (5.2) can also be seen as a condition for a negligible Doppler shift in comparison with the spontaneous decay rate of the atomic transition. Thus, the integration over times t_1 and t_2 appearing in Eq. (3.2) can be performed with the help of partial integration [15] and the excitation probability at time t is determined dominantly by the position operators $\hat{\mathbf{x}}_I(t_1)$ and $\hat{\mathbf{x}}_I(t_2)$ at time $\tau = t\Theta(t_{\rm out} - t) + t_{\rm out}\Theta(t - t_{\rm out})$ and at the initial time t_0. Consequently, for large interaction times, i.e., $\Gamma(\tau - t_0) \gg 1$, and neglecting terms exponentially small in this parameter, Eq. (3.2) simplifies to

$$\frac{P_e(t)}{P_{id}(t)} = \frac{9}{4}\langle\psi_T|\{\mathbf{e}_z\cdot\mathbf{g}_\omega^{(+)}[\hat{\mathbf{x}}_I(\tau)]e^{i\omega|\hat{\mathbf{x}}_I(\tau)|/c} + \mathbf{e}_z\cdot\mathbf{g}_\omega^{(-)}[\hat{\mathbf{x}}_I(\tau)] \times e^{-i\omega|\hat{\mathbf{x}}_I(\tau)|/c}\}^2|_{\omega=\omega_{eg}}|\psi_T\rangle. \quad (5.5)$$

For a spherically symmetric trapping potential, a simple analytical relation can be obtained from Eq. (5.5) if at the observation time t the center-of-mass degrees of freedom can be described by an isotropic Gaussian state $\hat{\rho}_T(\tau)$ centered around $\mathbf{x} = 0$ with spatial variance $\mathrm{Tr}[\hat{\rho}_T(\tau)\hat{\mathbf{x}}^2] \equiv \mathrm{Tr}[\hat{\rho}_T\,\hat{\mathbf{x}}_I^2(\tau)]$, i.e.,

$$\frac{P_e(t)}{P_{id}(t)} = \frac{3e^{-2\eta_0^2}}{10\eta_0^6}\left[-2\eta_0^4 - \eta_0^2 + \left(2\eta_0^4 - \eta_0^2 + 1\right)e^{2\eta_0^2} - 1\right] \quad (5.6)$$

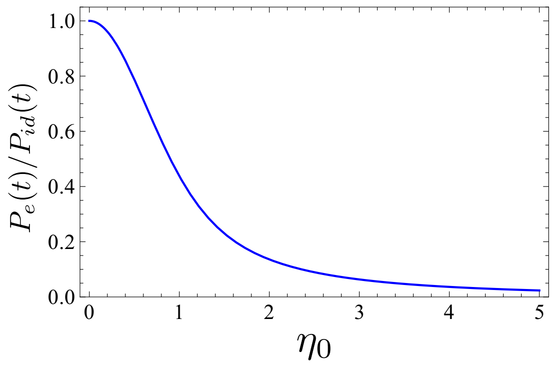

FIG. 2. Probability $P_e(t)$ of exciting a two-level system in a spherically symmetric trap and its dependence on the effective Lamb-Dicke parameter η_0 [see Eq. (5.7)]: the center-of-mass state $\hat{\rho}_T(\tau)$ is assumed to be an isotropic Gaussian state. The spontaneous decay rate is assumed to be large compared to the trap frequency, i.e., $\Gamma \gg \omega_{\text{trap}}$.

with the effective (time-dependent) Lamb-Dicke parameter

$$\eta_0 = \frac{\omega_{eg}}{c_0}\sqrt{\text{Tr}\big[\hat{\rho}_T\ \hat{\mathbf{x}}_I^2(\tau)\big]/3}. \tag{5.7}$$

This result can be applied to a large class of center-of-mass states including squeezed vacuum and thermal states (with respect to the isotropic trapping potential). The dependence of the excitation probability $P_e(t)/P_{id}(t)$ on the effective Lamb-Dicke parameter η_0 is depicted in Fig. 2 for $\Gamma \gg \omega_{\text{T}}$. The corresponding time dependence of the excitation probability $P_e(t)$ is illustrated in Fig. 3 for several values of η_0. It is apparent that for small effective Lamb-Dicke parameters η_0, almost perfect excitation is achievable at time t_{out}. For all effective Lamb-Dicke parameters, almost instantaneous excitation with a large spontaneous decay rate, i.e., $\Gamma \gg \omega_T$, is more effective than excitation with a small spontaneous decay rate, i.e., $\Gamma \ll \omega_T$.

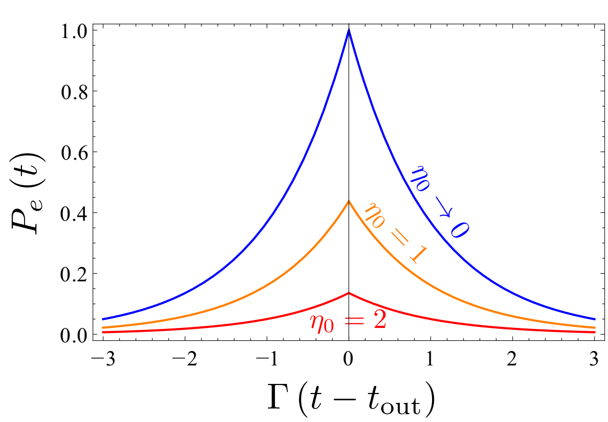

FIG. 3. Time dependence of the excitation probability $P_e(t)$ in a spherical symmetric trap for an isotropic Gaussian center-of-mass state $\hat{\rho}_T(\tau)$: the parameter η_0 characterizing the center-of-mass state is chosen to be $\eta_0 \to 0$ (blue), $\eta_0 = 1$ (orange), and $\eta_0 = 2$ (red). The trapping frequency is small, i.e., $\Gamma \gg \omega_T$, so that the time dependence of η_0 can be neglected. The interaction time is large, i.e., $\Gamma(\tau - t_0) \gg 1$.

VI. OPTIMIZING SINGLE-PHOTON ABSORPTION BY COHERENT CONTROL OF THE CENTER-OF-MASS MOTION

As discussed in Sec. V, the maximum probability for exciting the two-level system in the regime $\Gamma \gg \omega_{\text{trap}}$ is mainly limited by the spatial width of the center-of-mass state during the short time of the order $1/\Gamma$ in which the absorption process takes place. This limitation is of particular significance in cases in which no sub-Doppler cooling techniques are applied and in which this spatial width is not sufficiently small. A straightforward strategy to overcome this hurdle is to increase the depth of the trap and thus increase the confinement of the center-of-mass degrees of freedom. This procedure, however, is typically limited by experimental constraints and therefore cannot constitute an ultimate solution. As discussed in Sec. IV, even if the relevant trapping frequency becomes larger than the spontaneous photon emission rate, the achievable spatial confinement of the center-of-mass state is limited ultimately by the zero-point fluctuations in the trap. However, as discussed in Sec. V, if the photon absorption process takes place almost instantaneously, it is possible to increase the excitation probability by preparing a center-of-mass state whose width is sufficiently small during the photon absorption process, such as a squeezed state, for example. The squeezing of the center-of-mass state in ion traps has already been demonstrated in experiment [16,17]. One method for achieving significant squeezing is to modulate the trapping frequency with twice the trapping frequency [18,19]. This way, highly efficient one-photon excitation can be achieved even in a weakly confining harmonic trapping potential.

For this purpose, let us consider the dynamics of the center-of-mass motion of a nonrelativistic particle of mass m in a periodically modulated spherically symmetric harmonic trapping potential of the form

$$V_T(\mathbf{x},t) = \tfrac{1}{2}m\omega^2(t)\mathbf{x}^2,$$
$$\omega^2(t) = \omega_T^2 + \omega_T^2\delta\sin[\omega_M(t - t_0)]. \tag{6.1}$$

The trapping potential above corresponds to the well-studied problem of a parametric oscillator. The time evolution of the classical as well as the quantum mechanical problem can be expressed by using solutions of the Mathieu differential equation [12]. It is well known that the solutions of the Mathieu equation become unstable in the region $\omega_M \approx 2\omega_T$ [12]. This phenomenon of parametric resonance can be used to achieve significant squeezing of the center-of-mass state.

If the modulation strength of the trapping potentials is small, i.e., $|\delta| \ll 1$, and $\omega_M = 2\omega_T$ the dynamics of the center-of-mass motion can be determined perturbatively. The unperturbed dynamics is defined by the modulation strength $\delta = 0$ and by the corresponding explicitly time-independent Hamiltonian $\hat{H}_0 = \hat{\mathbf{p}}^2/(2m) + m\omega_T^2\mathbf{x}^2/2$. The resulting time evolution of an initially prepared pure state $|\psi_T\rangle$ is given by

$$|\psi_T(t)\rangle = \hat{S}(\xi_x(t))\hat{S}(\xi_y(t))\hat{S}(\xi_z(t))e^{-i\hat{H}_0(t-t_0)/\hbar}|\psi_T\rangle \tag{6.2}$$

with the time-dependent squeezing parameters $\xi_x(t) = \xi_y(t) = \xi_z(t) = r(t)e^{-2i\varphi(t)}$ being approximately determined by

$$r(t) = \omega_T\delta(t - t_0)/4,\ \varphi(t) = \omega_T(t - t_0) - \pi/2\ . \tag{6.3}$$

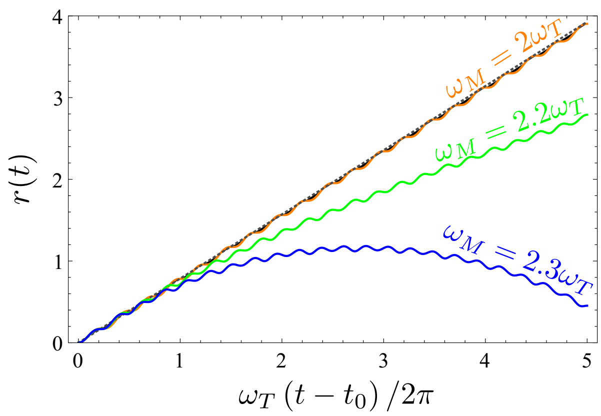

FIG. 4. Time evolution of the squeezing parameter $r(t)$ for $\delta = 0.5$: numerically exact results for modulation frequencies $\omega_M = 2\omega_T$ (orange solid line), $\omega_M = 2.2\omega_T$ (green solid line), $\omega_M = 2.3\omega_T$ (blue solid line), and the associated approximation (dotted line).

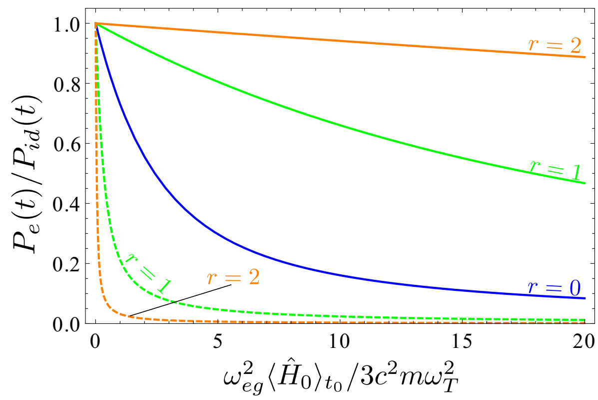

FIG. 5. Influence of squeezing of initially prepared thermal center-of-mass states on excitation probability $P_e(t)$: the parameters are $r = 0$ (blue), $r = 1$ (green), and $r = 2$ (orange). The solid lines correspond to $\sin^2[\varphi(t)] = 0$ and the dashed lines correspond to $\sin^2[\varphi(t)] = 1$ during the short time the absorption of the photon takes place. For the experimental parameters given in [7] (trapping of ^{174}Yb, $\omega_T = 2\pi \times 480$ kHz, $\Gamma = 1.2 \times 10^8$ s^{-1}, and $\omega_{eg} = 5.1 \times 10^{15}$ s^{-1}), we obtain $\omega_{eg}^2\langle\hat{H}_0\rangle_{t_0}/3c^2m\omega_T^2 = 1.7 \times 10^{-2}$ (ground state) and $\omega_{eg}^2\langle\hat{H}_0\rangle_{t_0}/3c^2m\omega_T^2 = 0.7$ (thermal state in Doppler limit).

The time evolution of the squeezing parameter $r(t)$ is depicted in the numerical results of Fig. 4 for several scenarios. The plot illustrates that for $\omega_M = 2\omega_T$ the numerical results are in excellent agreement with the approximate analytical expression of Eq. (6.3) even for moderately large modulation amplitudes. Even if the condition $\omega_M = 2\omega_T$ is violated, significant squeezing can be achieved. For larger deviations of ω_M form $2\omega_T$ [roughly $|\omega_M^2 - (2\omega_T)^2| \gtrsim 2\delta\omega_T^2$] a transition from an unstable solution of the Mathieu equation to a stable solution takes place. Squeezing can also be achieved in the stable region, but in this case the value of the squeezing parameter is bounded from above (see solid blue line in Fig. 4).

For an initially prepared energy eigenstate of the unperturbed trapping Hamiltonian $\hat{H}_0$, the corresponding mean values and variances of the position operator are given by

$$\langle\hat{\mathbf{x}}_I(t)\rangle = 0,$$
$$\langle\hat{\mathbf{x}}_I^2(t)\rangle = \frac{\langle\psi_T|\hat{H}_0|\psi_T\rangle}{m\omega_T^2}[e^{-2r(t)}\cos^2\varphi(t) + e^{2r(t)}\sin^2\varphi(t)]. \tag{6.4}$$

This implies that also for any incoherent mixture of energy eigenstates, such as a thermal state, for $\delta > 0$ at times t with $\sin[\varphi(t)] = 0$ the position uncertainties are squeezed significantly. For a thermal state, the mean energy of the unperturbed isotropic harmonic motion in the trap at temperature T is given by $\langle\hat{H}_0\rangle = 3\hbar\omega_T(1/2 + 1/\{\exp[\hbar\omega_T/(kT)] - 1\})$. The squeezing $r(t)$ induced by the periodic modulation of the trapping frequency with twice the trapping frequency ω_T increases linearly with the interaction time $(t - t_0)$. Thus, it is capable of reducing the uncertainty around the mean position $\langle\hat{\mathbf{x}}_I(t)\rangle = 0$ significantly even for an initially prepared thermal state. Consequently, even if the center-of-mass motion is confined only weakly by a trapping potential, the excitation probability can achieve values very close to the ideal motionless case.

The influence of squeezing of initially prepared thermal center-of-mass states on the excitation probability $P_e(t)$ is illustrated in Fig. 5 for several scenarios. These results were derived under the assumption that the condition stated in Eq. (5.2) is satisfied. If the degree of squeezing becomes too large, this condition may be violated and our results no longer apply. We can take this into account by using the results from Sec. IV. By using Eq. (4.4), we obtain the following probability for exciting the atom for a squeezed thermal center-of-mass state with $\sin^2[\phi(t_{\rm out})] = 0$:

$$P_e(t_{\rm out}) = 1 - \frac{\omega_{eg}^2}{5c^2}\Big\{\frac{2}{\Gamma^2 m^2}[\langle\hat{p}_z^2(t_{\rm out})\rangle + 2\langle\hat{p}_x^2(t_{\rm out})\rangle + 2\langle\hat{p}_y^2(t_{\rm out})\rangle] - \frac{2}{\Gamma^2}[\omega_z^2\langle\hat{z}_I^2(t_{\rm out})\rangle + 2\omega_x^2\langle\hat{x}_I^2(t_{\rm out})\rangle + 2\omega_y^2\langle\hat{y}_I^2(t_{\rm out})\rangle]$$
$$+ \langle\hat{z}_I^2(t_{\rm out})\rangle + 2\langle\hat{x}_I^2(t_{\rm out})\rangle + 2\langle\hat{y}_I^2(t_{\rm out})\rangle\Big\} + O\left(\frac{\omega_{x,y,z}^3}{\Gamma^3}\right) \tag{6.5}$$

for $(t_{\rm out} - t_0)\Gamma \gg 1$. For a spherically symmetric trapping potential, this simplifies to

$$P_e(t_{\rm out}) = 1 - \frac{\omega_{eg}^2\langle\psi_T|\hat{H}_0|\psi_T\rangle}{3mc^2}\left[\left(1 - \frac{2\omega_T^2}{\Gamma^2}\right)\frac{1}{\omega_T^2}e^{-2r(t_{\rm out})} + \frac{2}{\Gamma^2}e^{2r(t_{\rm out})}\right] + O\left(\frac{\omega_T^3}{\Gamma^3}\right)$$
$$\approx 1 - \frac{\omega_{eg}^2\langle\psi_T|\hat{H}_0|\psi_T\rangle}{3mc^2}\left[\frac{1}{\omega_T^2}e^{-2r(t_{\rm out})} + \frac{2}{\Gamma^2}e^{2r(t_{\rm out})}\right]. \tag{6.6}$$

By using the above expression, we find that squeezing is increasing the probability for absorbing the photon as long as

$$r \leqslant \ln[\Gamma/(\sqrt{2}\omega_T)]/2. \tag{6.7}$$

For the parameters of the experiment described in [7] ($\omega_T = 2\pi \times 480$ kHz, $\Gamma = 1.2 \times 10^8$ s^{-1}), we obtain

$$r \leqslant 1.7.$$

For smaller trapping frequencies or higher decay rates, even larger squeezing parameters r are still beneficial. Hence, for typical experimental parameters a significant increase of the excitation probability can be achieved.

Although our discussion has concentrated on a spherically symmetric harmonic trapping potential, generalizations to anisotropic cases are straightforward. They lead to different degrees of squeezing in different directions.

VII. CONCLUSION

We have investigated the influence of the center-of-mass motion of a trapped two-level system on resonant single-photon absorption. In particular, we have concentrated on single-photon excitation by an optimal photon wave packet which is capable of exciting a two-level system at a fixed position almost perfectly.

It has been demonstrated that the achievable excitation probability depends crucially on the ratio between the time scales of spontaneous photon emission and absorption on the one hand and of the center-of-mass motion in the trap on the other hand. If single-photon absorption and emission takes place on a time scale long in comparison with the characteristic time scale of the center-of-mass motion in the trap, it is the time-averaged center-of-mass motion which determines and limits the achievable single-photon excitation probability. In the opposite limit of fast spontaneous photon emission and absorption, it is the spatial width of the center-of-mass wave packet at the absorption time which limits the achievable single-photon excitation probability. This latter dependence can be exploited for increasing the achievable excitation probabilities significantly by squeezing the spatial width of the center-of-mass wave packet. By modulating the harmonic trapping frequency appropriately, such a significant squeezing at particular times during the periodic center-of-mass motion can be achieved. This way, the single-photon wave packets considered can achieve highly efficient excitation of a two-level system in free space even if the center-of-mass motion is only weakly confined and prepared in a thermal state initially.

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