Electron-phonon dynamics in an ensemble of nearly isolated nanoparticles

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We investigate the electron population dynamics in an ensemble of nearly isolated insulating nanoparticles, each nanoparticle modeled as an electronic two-level system coupled to a single vibrational mode. We find that at short times the ensemble-averaged excited-state population oscillates but has a decaying envelope. At long times, the oscillations become purely sinusoidal about a ''plateau'' population, with a frequency determined by the electron-phonon interaction strength and with an envelope that decays algebraically as $t^{-1/2}$. We use this theory to predict electron-phonon dynamics in an ensemble of $Y_2O_3:Eu^{3+}$ nanoparticles.

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I. INTRODUCTION

Nanocrystals exist in a size regime which lies between that of atomic and bulk matter, thus making them ideal for the study of extreme quantum effects in condensed matter systems. In particular, the vibrational properties of nanoparticles are strikingly different than their bulk counterparts: A spherical nanoparticle of diameter d cannot support internal vibrations at frequencies less than about $2\pi v/d$, where v is a characteristic bulk sound velocity. Any property of the nanoparticle that depends on the vibrational spectrum, such as its thermodynamic properties or electron-phonon dynamics, will be very different at low energies than in bulk crystals. This will be especially true for nanoparticles—for example, in powder form—only weakly coupled to their surroundings.

One way to probe the vibrational spectrum of a nanoparticle is to optically excite an electron-hole pair and study the intraband electronic energy relaxation prior to radiative recombination.[1,2] However, the excitonic states, being only weakly localized, will suffer significant quantum-confinement effects in the nanoparticle, making comparison with bulk relaxation rather indirect. An alternative probe of the vibrational spectrum is provided by well-localized electronic impurity states in a doped nanocrystal.[3,4] The impurity states can be used to probe both energy relaxation by phonon emission[3] and phonon-induced dephasing.[4] In these cases, the difference between the nanocrystal and bulk behavior is almost entirely a consequence of their differing vibrational modes.

In a recent experiment, Yang *et al.*[3] measured the phonon emission rate in Eu-doped Y_2O_3 nanoparticles between two electronic states separated by 3 cm^{-1} in energy. The experiment was performed on a powder of nanoparticles, prepared by gas-phase condensation, with a mean diameter of 13 nm. Regarding the nanoparticles as isotropic elastic spheres,[5] the lowest internal vibrational mode (a fivefold degenerate torsional mode) has a frequency of about 11 cm^{-1}. The excited-state population was found to decay exponentially with a lifetime of 27 μs, more than two orders of magnitude longer than that between the same levels in the bulk (about 220 ns). Although there is no quantitative theory available yet to explain their results, the inhibited phonon emission is consistent with a large suppression of the low-frequency vibrational density of states (DOS) expected for these small crystals.

Experiments such as the one by Yang *et al.*[3] present an exciting opportunity to study the crossover of phonon dynamics from bulk to nanoscale systems. However, the following question immediately arises: Given the large apparent modification of the DOS by finite-size effects, is it still correct to expect exponential relaxation and to use perturbation theory (Fermi's golden rule in this case) to relate the relaxation rate to the phonon DOS? After all, the energy in a nanoparticle that is completely isolated from its surroundings would have to be exchanged between the electron and vibrational degrees of freedom in a Rabi-like manner, and no relaxation would be observed. Indeed, an isolated nanoparticle may be regarded as a phonon analog of a two-level atom in a cavity, which is known to exhibit oscillatory population dynamics.[6]

It should be possible to detect oscillatory population dynamics experimentally using the techniques of Ref. 3, but what would be the effect of the unavoidable *distribution* of nanoparticle sizes and corresponding vibrational-mode frequencies? In the limit where each nanoparticle has a two-level system interacting with only the lowest-frequency vibrational mode, called the Lamb mode, each nanoparticle would exhibit vacuum Rabi oscillations.[7] However, the Rabi frequency, which is a function of the electron-phonon interaction strength and the energy mismatch between the two-level system and Lamb mode, would vary from nanoparticle to nanoparticle.

The purpose of this paper is to investigate the electron-phonon dynamics of an ensemble of nearly isolated nanoparticles. We find that at short times the ensemble-averaged excited-state population oscillates in a fashion that depends on the detailed size distribution of the nanoparticles, but that at long times the oscillations become purely sinusoidal about a ''plateau'' population fraction, with the frequency of the oscillation determined by the electron-phonon interaction strength alone and with an envelope that decays algebraically as $t^{-1/2}$. In the infinite-time limit, the excited-state population approaches the constant, finite plateau value. (Of course, the small damping of the Lamb mode, produced by the weak but nonzero interaction of a nanoparticle with its environment, will eventually cause the electrons to relax irreversibly.)

In the next section we derive a general expression for the average excited-state population in an ensemble of nanoparticles at zero temperature and study its short-time and long-time behavior. The rotating-wave approximation (RWA) we use there limits the application of our results to situations where the detuning energies (the energy mismatch between the two-level system and Lamb mode) are not much larger than the electron-phonon interaction strength. In Sec. III we apply this theory to an ensemble of $Y_2O_3:Eu^{3+}$ nanoparticles similar to that of Yang *et al.*[3] and give quantitative predictions for the average excited-state population as a function of the mean nanoparticle size and standard deviation. Section IV contains our conclusions and a discussion of questions for future investigation.

II. ENSEMBLE-AVERAGED POPULATION DYNAMICS

To begin, we consider a single doped nanoparticle. We assume this nanoparticle to be an isotropic elastic sphere with a single localized electronic two-level system embedded within. We limit our investigation to small detuning energies, thus allowing for the use of the RWA, which discards non-energy-conserving terms in the Hamiltonian.[6] In addition, we will neglect the fivefold degeneracy of the lowest vibrational mode that would be present in a perfectly spherical nanoparticle[5] and will assume a single nondegenerate Lamb mode. We shall return to this point below in Sec. IV.

Dissipation terms in the Hamiltonian are neglected as well. By dissipation terms we mean interactions of the nanoparticle with its surroundings that allow vibrational energy to be carried away irreversibly. For weak dissipation, these terms would cause an exponential decay of the population at large times, causing the ''plateau'' population to fall exponentially to zero. This effect would be negligible at small times and therefore would have no impact on the short-time calculations given below. For large dissipation, the exponential decay would be noticeable at all times and would therefore render our results invalid.

We assume that there is only one vibrational mode available for the electronic system to couple to—the next vibrational mode being so high in energy as to make the effect of its coupling negligible. Therefore, as stated, our doped nanoparticle system consists of a two-level atom coupled to single vibrational mode. The Hamiltonian, in units where $\hbar=1$, is given by

$$H=\sum_{\alpha}\ \epsilon_{\alpha}c_{\alpha}^{\dagger}c_{\alpha}+\omega_0 a^{\dagger}a+\sum_{\alpha\alpha'}\ g_{\alpha\alpha'}c_{\alpha}^{\dagger}c_{\alpha'}(a+a^{\dagger}), \quad (1)$$

where $\alpha=1,2$. The first term in H is the Hamiltonian for a noninteracting two-level system with energies ϵ_1 and ϵ_2 and fermionic creation and annihilation operators $c_{\alpha}^{\dagger}$ and c_{α}. The second term is the Hamiltonian for a vibrational mode with frequency ω_0 and phonon creation and annihilation operators $a^{\dagger}$ and a. The third term is the ordinary first-order interaction between the two-level system and the vibrational mode. $g_{\alpha\alpha'}$ is the electron-phonon interaction energy, with $g_{12}=g_{21}=g$, and the other terms equal to zero.

The wave function can be written as a superposition of the two electronic states, together with all possible populations of the single vibrational mode,

$$|\psi(t)\rangle=\sum_{\alpha n}\ C_{\alpha n}(t)e^{-i(\epsilon_{\alpha}+n\omega_0)t}|\alpha n\rangle, \quad (2)$$

where

$$|\alpha n\rangle\equiv\frac{1}{\sqrt{n!}}(a^{\dagger})^{n}c_{\alpha}^{\dagger}|0\rangle. \quad (3)$$

The coefficients $C_{\alpha n}(t)$ satisfy the coupled equations,

$$\partial_t C_{1n}(t)+ig\sqrt{n+1}\,e^{-i(\omega_0+\Delta\epsilon)t}C_{2,n+1}(t)+ig\sqrt{n}e^{i(\omega_0-\Delta\epsilon)t}C_{2,n-1}(t)=0 \quad (4)$$

and

$$\partial_t C_{2n}(t)+ig\sqrt{n+1}\,e^{-i(\omega_0-\Delta\epsilon)t}C_{1,n+1}(t)+ig\sqrt{n}e^{i(\omega_0+\Delta\epsilon)t}C_{1,n-1}(t)=0, \quad (5)$$

where $\Delta\epsilon\equiv\epsilon_2-\epsilon_1$ is the electronic energy-level separation.

In the RWA, which is valid near resonance (defined by the condition $\omega_0=\Delta\epsilon$), these coupled differential equations reduce to

$$\partial_t C_{1n}(t)+ig\sqrt{n}\,e^{i\nu t}C_{2,n-1}(t)=0 \quad (6)$$

and

$$\partial_t C_{2n}(t)+ig\sqrt{n+1}\,e^{-i\nu t}C_{1,n+1}(t)=0, \quad (7)$$

where

$$\nu\equiv\omega_0-\Delta\epsilon \quad (8)$$

is the detuning frequency.

These equations can be solved by Laplace transformation using the boundary condition $C_{\alpha n}(0)=\delta_{\alpha 2}\delta_{n0}$. The amplitude for the upper state is

$$C_{2n}(t)=\delta_{n0}\left[\cos\left(\frac{\Omega}{2}t\right)+i\frac{\nu}{\Omega}\sin\left(\frac{\Omega}{2}t\right)\right]e^{-i\nu t/2}, \quad (9)$$

where

$$\Omega\equiv\sqrt{\nu^2+4g^2} \quad (10)$$

plays the role of the Rabi frequency in this problem.[6]

The probability that the electron is in state α, irrespective of the number of phonons present, is

$$N_{\alpha}(t)\equiv\sum_{n}\ |C_{\alpha n}(t)|^2. \quad (11)$$

Then $N_2(t)=|C_{20}(t)|^2$ is given by

$$N_2(t)=1-\left(\frac{\Omega^2-\nu^2}{\Omega^2}\right)\sin^2\left(\frac{\Omega}{2}t\right) \quad (12)$$

and

$$N_1(t) = 1 - N_2(t). \tag{13}$$

The dependence of $N_1(t)$ and $N_2(t)$ on ν will be suppressed for simplicity.

In an ensemble of such nanoparticles, a variation in diameter yields a similar variation in detuning frequency ν. We assume this to be a Gaussian distribution. To obtain values for the mean detuning $\overline{\nu}$ and standard deviation in detuning, σ, we assume each nanoparticle to be an isotropic elastic continuum with stress-free boundary conditions. Then, as shown by Lamb,[5] the lowest vibrational frequency is

$$\omega_0(d) \approx \frac{2\pi v_t}{d}, \tag{14}$$

where v_t is the bulk transverse sound velocity and d is the nanoparticle diameter. The mean detuning is then

$$\overline{\nu} = \omega_0(\overline{d}) - \Delta\epsilon, \tag{15}$$

where $\overline{d}$ is the mean particle diameter. Using these relationships, we assume a distribution in detuning frequency given by

$$P(\nu) \equiv \frac{e^{-(\nu-\overline{\nu})^2/\sigma^2}}{\sqrt{\pi}\sigma}. \tag{16}$$

The ensemble-averaged population of electronic state $|\alpha\rangle$ is then given by

$$\overline{N}_\alpha(t) \equiv \int_{-\infty}^{\infty} d\nu\, P(\nu) N_\alpha(t). \tag{17}$$

The behavior of $\overline{N}_2(t)$ at short times is sinusoidal with a decaying envelope dependent on the specifics of the particle size distribution. We will give examples of the short-time dynamics below.

The long-time behavior of $\overline{N}_2(t)$ can be obtained analytically by an asymptotic expansion. We begin by writing Eq. (17) as

$$\overline{N}_2(t) = \overline{N}_2(\infty) + \frac{\Omega_{\rm res}^2}{2\sqrt{\pi}\sigma} \mathrm{Re}\, I(t), \tag{18}$$

where

$$\overline{N}_2(\infty) \equiv 1 - \frac{\Omega_{\rm res}^2}{2\sqrt{\pi}\sigma} \int_{-\infty}^{\infty} d\nu \frac{e^{-(\nu-\overline{\nu})^2/\sigma^2}}{\Omega_{\rm res}^2 + \nu^2} \tag{19}$$

is a constant independent of time and

$$I(t) \equiv \int_{-\infty}^{\infty} d\nu \frac{e^{-(\nu-\overline{\nu})^2/\sigma^2}}{\Omega_{\rm res}^2 + \nu^2} e^{it\sqrt{\Omega_{\rm res}^2 + \nu^2}}, \tag{20}$$

where

$$\Omega_{\rm res} \equiv 2g \tag{21}$$

TABLE I. Plateau poplation values for $\overline{N}_2(t)$ as $t \to \infty$. Here $\Omega_{\rm res} = 2g$ is the resonant Rabi frequency.

		$\overline{\nu}/\Omega_{\rm res}$		
		0.0	1.0	2.0
$\sigma/\Omega_{\rm res}$	0.1	0.50	0.74	0.90
	0.5	0.54	0.73	0.89
	1.0	0.62	0.73	0.87

is the *resonant* Rabi frequency. It will turn out that the constant $\overline{N}_2(\infty)$ is simply the value of $\overline{N}_2(t)$ in the $t \to \infty$ limit. Values of this plateau population are given in Table I.

The integral $I(t)$ can be evaluated at large times by analytically continuing ν into the complex plane and expanding around the saddle point at $\nu = 0$. This leads to the asymptotic result

$$I(t) \approx e^{-\overline{\nu}^2/\sigma^2} \sqrt{\frac{2\pi}{t\Omega_{\rm res}^3}} e^{i(t\Omega_{\rm res} + \pi/4)}, \tag{22}$$

and, hence,

$$\overline{N}_2(t) \approx \overline{N}_2(\infty) + \sqrt{\frac{1}{2}} \frac{e^{-\overline{\nu}^2/\sigma^2}}{\sigma/\Omega_{\rm res}} \frac{\cos(t\Omega_{\rm res} + \pi/4)}{\sqrt{t\Omega_{\rm res}}} \quad (t \to \infty). \tag{23}$$

Note that at long times the population oscillates at the *resonant* Rabi frequency, independent of the mean detuning $\overline{\nu}$. This occurs because the higher-frequency components tend to average out faster. However, the amplitude of the asymptotic oscillations decreases with $\overline{\nu}$.

To illustrate the short-time behavior of $\overline{N}_2(t)$ as a function of $\overline{\nu}$ and σ, we present several plots showing $\overline{N}_2(t)$ as a function of $t\Omega_{\rm res}$. In choosing values for $\overline{\nu}$ and σ, we attempted to cover the entire range of these parameters which could be reasonably addressed in the RWA.

In Fig. 1, we study the resonant case where $\overline{\nu} = 0$. By choosing three values of σ we are able to directly observe the effects that it has on the envelope function. As σ in-

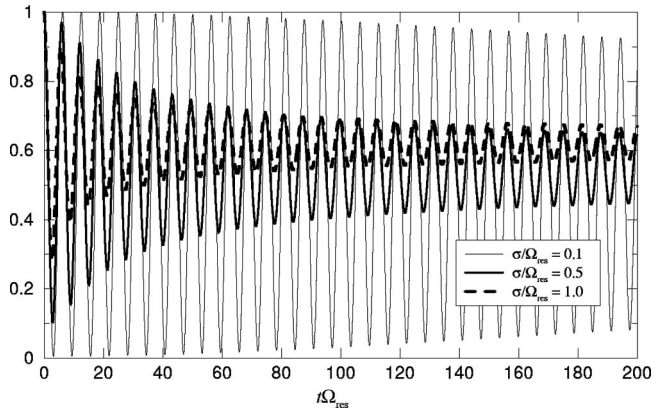

FIG. 1. Electronic population in state $|2\rangle$ for $\overline{\nu} = 0.0$.

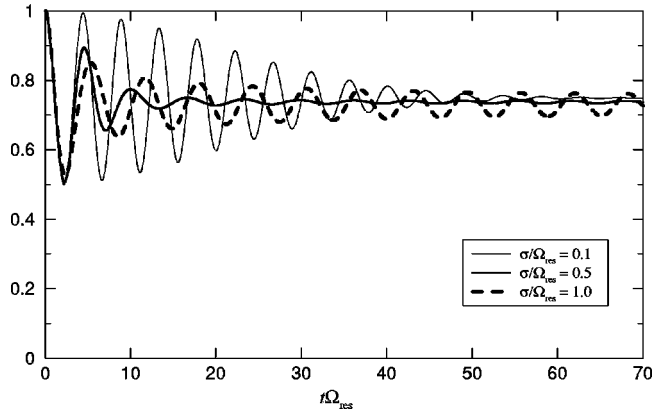

FIG. 2. Electronic population in state $|2\rangle$ for $\overline{\nu}=\Omega_{\rm res}$.

creases, the envelope function decays faster and the time at which asymptotic behavior becomes observable decreases. In this case of $\overline{\nu}=0$, changing σ has little effect on the frequency at which $\overline{N}_2(t)$ oscillates.

Figure 2, where $\overline{\nu}=\Omega_{\rm res}$, illustrates the effects of an intermediate value of $\overline{\nu}$. The short-time envelope function decays faster and the plateau population is increased. As in Fig. 1, increasing σ increases the rate at which the short-time envelope function decreases, but here it can be seen to lower the frequency of oscillation in $\overline{N}_2(t)$. Unlike Fig. 1, Fig. 2 clearly shows how increasing σ up to $\Omega_{\rm res}$ increases the amplitude of oscillations at large times [see Eq. (23)].

The last plot, Fig. 3, illustrates the limits of the RWA. The effects of increasing $\overline{\nu}$ are now taken to the extreme: the short-time envelope functions decay very rapidly, making asymptotic behavior apparent at early times, and the plateau population approaches unity. Altering σ has similar effects as in Figs. 1 and 2, only more pronounced. [$\overline{N}_2(t)$ for $\sigma/\Omega_{\rm res}=0.1$ levels off to its plateau population shortly past $t\Omega_{\rm res}=50$.]

III. POPULATION DYNAMICS IN $Y_2O_3:Eu^{3+}$ NANOPARTICLES

Here we use the results of Sec. II to address future experiments. We examine the case of $Y_2O_3:Eu^{3+}$ nanoparticles, specifically the electron dynamics between two crystal-field split $^5D_1$ levels separated by $\Delta\epsilon=3\ \ {\rm cm}^{-1}$, such as in the experiment by Yang *et al.*[3] To make quantitative predictions for such nanoparticles, the only parameters that need to be specified are the mean detuning frequency $\overline{\nu}$ and the standard deviation in detuning, σ. We also need the electron-phonon interaction strength g. Our treatment requires that $\overline{\nu}$ be not much greater than $\Omega_{\rm res}$.

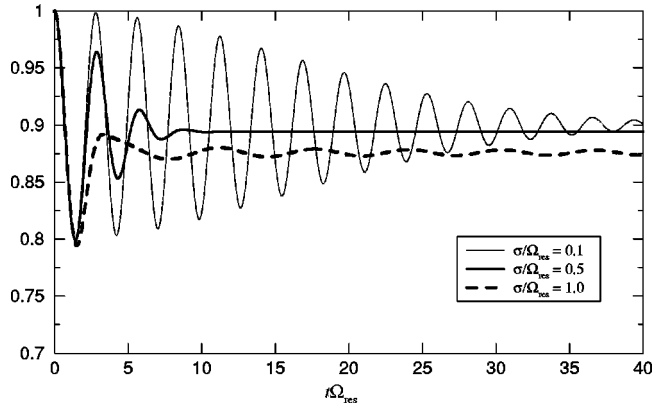

FIG. 3. Electronic population in state $|2\rangle$ for $\overline{\nu}/\Omega_{\rm res}=2.0$.

In the case of $Y_2O_3:Eu^{3+}$ nanoparticles, we can estimate the electron-phonon interaction strength by using the experimentally observed bulk phonon emission rate.[3] We obtain a value for $g_{\rm micron}$, the electron-phonon interaction strength in micron-size particles, and then scale this value to get g for the nanoparticles. The scaling is achieved by assuming that g varies with energy and system volume as it does in a bulk crystal. In principle, this method is only correct between two systems with continuous vibrational spectra. However, using this technique should provide a reasonable estimate of g in the nanoparticle.

In the micron-scale crystal, Fermi's golden rule states that for an electron-phonon interaction of the form given in Eq. (1), the phonon emission rate is

$$\tau^{-1}=\frac{2\pi}{\hbar}g^2\Gamma(\Delta\epsilon)V, \tag{24}$$

where V is the crystal volume and $\Delta\epsilon$ is the electronic level spacing. $\Gamma(\epsilon)$, the phonon DOS (per volume) as a function of phonon energy ϵ, is defined by

$$\Gamma(\epsilon)\equiv\frac{1}{V}\sum_n\ \delta(\epsilon-\hbar\omega_n), \tag{25}$$

where ω_n are the phonon frequencies and, in a bulk crystal at low energies,

$$\Gamma(\epsilon)=\frac{3\epsilon^2}{2\pi^2\hbar^3\overline{v}^3}, \tag{26}$$

where

$$\overline{v}\equiv\left(\frac{1}{3}\sum_\lambda\frac{1}{v_\lambda^3}\right)^{-1/3} \tag{27}$$

is a branch-averaged sound velocity.

The lifetime in the micron-sized crystal was observed to be $\tau=220$ ns,[3] and in Y_2O_3 the sound velocities are approximately,[8]

$$v_{\rm l}=6.7\times10^5\ \ {\rm cm\ s}^{-1}, \tag{28}$$

$$v_{\rm t}=4.3\times10^5\ \ {\rm cm\,s}^{-1}, \tag{29}$$

and

$$\overline{v}=4.7\times10^5\ \ {\rm cm\,s}^{-1}. \tag{30}$$

g is then given by

$$g = \frac{\pi \hbar^2 \overline{v}^{3/2}}{\Delta\epsilon} \sqrt{\frac{2\tau^{-1}}{3V}}. \tag{31}$$

The mean diameter of these nanoparticles was about 5 μm, leading to $g_{\text{micron}} = 2.04 \times 10^{-6}$ cm^{-1}.

In a bulk crystal (with deformation-potential coupling), g varies as $(\epsilon/V)^{1/2}$, so we scale g_{micron} in ϵ and V to get a value for g_{nano}, the electron-phonon interaction strength in the nanoparticle:

$$g_{\text{nano}} = \left(\frac{10 \text{ cm}^{-1}}{3 \text{ cm}^{-1}} \right)^{1/2} \left(\frac{13 \text{ nm}}{5 \ \mu\text{m}} \right)^{-3/2} g_{\text{micron}} \tag{32}$$

$$= 2.8 \times 10^{-2} \text{ cm}^{-1}. \tag{33}$$

Then the resonant Rabi frequency, as defined in Eq. (21), is about

$$\Omega_{\text{res}} = 5.6 \times 10^{-2} \text{ cm}^{-1}. \tag{34}$$

In the experiment of Yang *et al.*, the mean particle diameter was 13 nm and the standard deviation in particle size was 5 nm.[3] By Eqs. (14) and (15), these values yield $\overline{\nu} = 71\Omega_{\text{res}}$ and $\sigma = 44\Omega_{\text{res}}$. This detuning is large enough that higher-energy vibrational modes can no longer be ignored, making our single-mode treatment inapplicable. Furthermore, these values are too far off resonance to be reliably addressed by the RWA.

To observe the behavior predicted in Sec. II for $Y_2O_3:Eu^{3+}$ nanoparticles, the mean diameter should be about 46 nm and the standard deviation in diameter no more than about 3 nm [see Eqs. (14) and (15)].

IV. DISCUSSION

We have shown that the unavoidable size dispersion in a collection of nanoparticles can effect the ensemble-averaged electronic population dynamics considerably, affecting even the oscillation frequency at long times. However, our analysis has been restricted to near-resonant conditions in an idealized single-mode nanocrystal. A comprehensive theory of electron-phonon dynamics in nearly isolated nanoparticles will have to address a number of additional issues.

First, the vibrational modes of a nanoparticle will be broadened due to intrinsic mechanisms and from interaction with the environment. For example, anharmonicity will broaden the Lamb mode at finite temperature,[9] as will mechanical coupling to a substrate or to a cluster of other nanoparticles.[10] This broadening will cause the envelope of the population oscillations to decay exponentially at long times. For weak damping, however, we do not expect any qualitative changes in the short-time dynamics.

Second, nanoparticles will generally have large detuning frequencies, making the inclusion of higher-frequency modes necessary and making the RWA invalid. One way to include these effects would be to do exact-diagonalization studies in models with a truncated Hilbert space—for example, having bounded phonon occupation numbers for one or more modes.

Finally, it will be necessary to understand the effects of vibrational-mode degeneracy, which has been neglected here. The presence of degeneracy means that the Lamb mode has a vectorlike internal degree of freedom, the components of the vector describing the phonon amplitude in each branch. Although there has been work done in the quantum optics literature on the multimode generalizations of our Hamiltonian (1), that work has focused on the two-photon resonance case where $\Delta\epsilon = 2\omega_0$.[11,12] One way to approach the degenerate case nonperturbatively would be to assume N degenerate modes in the limit of large N. By analogy with other quantum systems with internal degrees of freedom, like particles with N spin components, it is reasonable to expect that quantum effects will be diminished in the large-N limit. In a nanoparticle this would suggest a suppression of the Rabi oscillations.

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[1] U. Woggon, *Optical Properties of Semiconductor Quantum Dots* (Springer-Verlag, Berlin, 1997).
[2] S. V. Gaponenko, *Optical Properties of Semiconductor Nanocrystals* (Cambridge University Press, Cambridge, England, 1998).
[3] H.S. Yang, S.P. Feofilov, D.K. Williams, J.C. Milora, B.M. Tissue, R.S. Meltzer, and W.M. Dennis, Physica B **263**, 476 (1999).
[4] R.S. Meltzer and K.S. Hong, Phys. Rev. B **61**, 3396 (2000).
[5] H. Lamb, Proc. London Math. Soc. **13**, 189 (1882).
[6] M. O. Scully and M. S. Zubairy, *Quantum Optics* (Cambridge University Press, Cambridge, England, 1997).
[7] Vacuum refers to the case where no phonons, thermal or otherwise, are present in the initial electron-phonon state.
[8] C. Proust *et al.*, Solid State Commun. **93**, 729 (1995).
[9] V.A. Markel and M.R. Geller, J. Phys.: Condens. Matter **12**, 7569 (2000).
[10] K. R. Patton and M. R. Geller, cond-mat/0106544 (unpublished).
[11] C.M. Caves and B.L. Schumaker, Phys. Rev. A **31**, 3068 (1985).
[12] S.C. Gou, Phys. Rev. A **40**, 5116 (1989).