Quantum kinetics of squeezed lattice displacement generated by phonon down conversion

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We study the fluctuation properties of longitudinal acoustic (LA) phonons generated by the anharmonic decay of coherent longitudinal optical phonons in a bulk semiconductor and a quantum dot. This process is comparable to the down conversion of photons, which is well known to generate squeezed photons. We use a quantum kinetic model to calculate the fluctuations of lattice displacement and momentum. This allows us to analyze the strength and the spatial distribution of the fluctuations. It is shown that the fluctuations may fall below their vacuum value, i.e., squeezing occurs. The squeezing only persists for short times due to fast dephasing of the different LA phonon modes and the increasing LA phonon population. In the quantum dot case, it is found that although LA phonon wave packets travel out of the quantum dot, the squeezing does not leave the vicinity of the dot but remains confined.

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I. INTRODUCTION

Squeezed states of bosonic systems have attracted much interest due to their interesting quantum-mechanical properties. In addition to the case of photons, where the study of squeezed states has a long tradition,[1–4] other bosonic particles or quasiparticles have recently been investigated, such as atomic Bose-Einstein condensates,[5] surface plasmons,[6] magnons,[7] and in particular phonons in various systems.[8–16] From elementary quantum mechanics, it is known that the fluctuations of quantum-mechanical variables are governed by the Heisenberg uncertainty principle, which provides a lower limit for the product of the fluctuations of two conjugate variables. For a crystal lattice, two such conjugate variables are the lattice displacement and the corresponding momentum. Even in the phonon vacuum, i.e., when the lattice is in its ground state, the fluctuations are nonvanishing. In a squeezed state, the fluctuations of one of these variables is for some period of time reduced below its vacuum level at the cost of an increased uncertainty for the other variable.

Different approaches have been taken to investigate the fluctuations of phonon systems. In Ref. 8, the transmission change of a probe pulse was studied, which in that case, being caused by a Raman process of second order, was sensitive to the mean-square displacement of the atomic positions and therefore allowed the authors to draw conclusions about the atomic fluctuations. In Ref. 9, the intensity of the x-ray diffraction signal was measured, which is determined by the Debye-Waller factor and therefore again carries information about the mean-square displacement. In other studies, the atomic positions have been measured repeatedly by observing the reflection of a probe pulse,[11] and the fluctuation properties have been extracted from a statistical analysis.

In quantum optics, a well-established tool to create squeezed photons is parametric down conversion,[17] in which a nonlinear crystal converts a single incoming photon into an entangled pair of photons. By this process, the photon system evolves into a two-mode squeezed state. For phonons, a similar mechanism is the decay of longitudinal optical (LO) phonons into pairs of longitudinal acoustic (LA) phonons due to anharmonic contributions in the lattice potential. Indeed, in the case of a strictly energy-conserving decay into two fixed LA phonon modes and for a coherent LO phonon state that is unperturbed by the decay, it has been shown that this process may generate LA phonon states in which the two-mode quadrature operators are squeezed.[13] In this paper, we present quantum kinetic calculations of the anharmonic decay process based on a microscopic model, which allows us to analyze quantitatively the fluctuation properties of the lattice displacement itself. Two systems are discussed: a bulk semiconductor and a quantum dot structure. While in the bulk case the LO phonons and thus also the LA phonons are generated spatially homogeneously, in the quantum dot structure the LO phonons are generated inside the dot. The LA phonons that are created in the decay process have a nonvanishing group velocity and therefore may leave the dot region. The choice of these two cases will thus allow us to compare the purely temporal dynamics in the bulk system with the spatiotemporal dynamics in the quantum dot case.

The paper is organized as follows: In Sec. II, we start with a presentation of the theoretical background. We introduce the model Hamiltonian and briefly review the appearance of two-mode squeezing (Sec. II A), then we introduce the variables describing the fluctuations of the lattice displacement and the momentum (Sec. II B), and finally we derive the equations of motion for the relevant phonon variables (Sec. II C). In the following section, the results are presented both for the case of a bulk semiconductor (Sec. III A) and a quantum dot structure (Sec. III B). The paper finishes with some concluding remarks (Sec. IV).

II. THEORY

A. Squeezing induced by anharmonic decay

Lattice dynamics are typically described within a harmonic approximation of the binding potential. This allows for the introduction of phonons as quasiparticles and leads to the free phonon Hamiltonian H_0. Here we are particularly interested in LO and LA phonons. We denote the corresponding creation operators as $a_{\mathbf{k}}^{\dagger}$ and $b_{\mathbf{q}}^{\dagger}$, respectively, where $\mathbf{k}$ and $\mathbf{q}$ refer to the phonon wave vectors. Beyond this harmonic approximation, cubic terms in the phonon operators appear in the Hamiltonian, which give rise to phonon-phonon interactions, e.g., phonon scattering and decay of phonons into other modes. In this paper, we will concentrate on the latter process, in particular on the decay of an LO phonon into a pair of LA phonons.

The free phonon Hamiltonian and the Hamiltonian of the anharmonic decay read

$$H_0 = \sum_{\mathbf{k}} \hbar\omega_{\mathrm{LO}}(\mathbf{k}) a^\dagger_{\mathbf{k}} a_{\mathbf{k}} + \sum_{\mathbf{q}} \hbar\omega_{\mathrm{LA}}(\mathbf{q}) b^\dagger_{\mathbf{q}} b_{\mathbf{q}}, \tag{1}$$

$$H_{\mathrm{anh}} = \sum_{\mathbf{k},\mathbf{q}} (\lambda_{\mathbf{q},\mathbf{k}} a^\dagger_{\mathbf{k}} b_{\mathbf{k}-\mathbf{q}} b_{\mathbf{q}} + \lambda^*_{\mathbf{q},\mathbf{k}} a_{\mathbf{k}} b^\dagger_{\mathbf{k}-\mathbf{q}} b^\dagger_{\mathbf{q}}), \tag{2}$$

where $\lambda_{\mathbf{q},\mathbf{k}}$ is the phonon-phonon coupling matrix element while $\omega_{\mathrm{LO}}(\mathbf{k})$ and $\omega_{\mathrm{LA}}(\mathbf{q})$ are the dispersion relations of LO and LA phonons. The decay of LO phonons into pairs of LA phonons has been shown to be strong in a number of semiconductor crystals, such as GaAs, GaP, and InP.[18,19]

By means of an optical excitation with a short laser pulse, the LO phonons can be driven into a coherent state.[20] In a homogeneous system, for symmetry reasons only $\mathbf{k}=\mathbf{0}$ coherent phonons are excited.[21] Therefore, let us start by looking at the simple case in which initially the $\mathbf{k}=\mathbf{0}$ LO phonon mode is in a coherent state with $\langle a_0\rangle = \alpha_0(t)$. If the LO phonons are treated on a mean-field level and only a decay into a single pair of LA phonon modes with wave vectors $\pm\mathbf{q}_0$ is considered, we obtain the effective Hamiltonian

$$H'_{\mathrm{anh}} = \lambda_{\mathbf{q}_0,0}\alpha^*_0(t) b_{-\mathbf{q}_0} b_{\mathbf{q}_0} + \lambda^*_{\mathbf{q}_0,0}\alpha_0(t) b^\dagger_{-\mathbf{q}_0} b^\dagger_{\mathbf{q}_0}. \tag{3}$$

This Hamiltonian describes a decay similar to the photon down conversion.[17] It has been shown by Hu and Nori[13] that in this case the time evolution operator $U(t,t_0)$ for the LA phonon subsystem can be approximated by

$$\begin{aligned} U(t,t_0) &= \exp[-i(t-t_0)H_0/\hbar] \\ &\quad\times \exp[\rho^*(t,t_0) b_{-\mathbf{q}_0} b_{\mathbf{q}_0} - \rho(t,t_0) b^\dagger_{-\mathbf{q}_0} b^\dagger_{\mathbf{q}_0}], \\ \text{with } \rho(t,t_0) &= \frac{i\lambda_{\mathbf{q}_0,0}}{\hbar}\int_{t_0}^{t} \alpha_0(\tau) e^{i[\omega_{\mathrm{LA}}(\mathbf{q}_0)+\omega_{\mathrm{LA}}(-\mathbf{q}_0)]\tau} d\tau, \end{aligned} \tag{4}$$

which becomes exact either in the case of impulsive driving with $\alpha_0(t)\sim\delta(t)$ or if the phonon modes involved fulfill energy conservation with $\hbar\omega_{\mathrm{LO}} = \hbar\omega_{\mathrm{LA}}(\mathbf{q}_0) + \hbar\omega_{\mathrm{LA}}(-\mathbf{q}_0)$ and $\alpha_0(t)\sim e^{-i\omega_{\mathrm{LO}}t}$. The second term of $U(t,t_0)$ is formally equivalent to a two-mode squeeze operator.[17,22] Hence, if the LA subsystem is initially either in the vacuum state or in a coherent state, the expectation values of the conjugate two-mode quadrature operators of displacement $X_{\pm\mathbf{q}_0} = 2^{-3/2}(b_{\mathbf{q}_0} + b^\dagger_{\mathbf{q}_0} + b_{-\mathbf{q}_0} + b^\dagger_{-\mathbf{q}_0})$ and momentum $P_{\pm\mathbf{q}_0} = -i2^{-3/2}(b_{\mathbf{q}_0} - b^\dagger_{\mathbf{q}_0} + b_{-\mathbf{q}_0} - b^\dagger_{-\mathbf{q}_0})$ are squeezed, i.e., their variances fall below their vacuum values in turn.

Instead of studying the two-mode quadrature operators, in this paper we will investigate the fluctuation properties of the lattice displacement and lattice momentum themselves. The squeezing of the two-mode quadratures suggests that these variables could be squeezed as well, although there are crucial differences, as will become apparent. We present quantum-kinetic calculations based directly on the Hamiltonian H_{anh}. Therefore, decay processes into a quasicontinuum of LA phonon modes and effects of the decay on the dynamics of the LO phonon subsystem are included. Within our model, the approach yields quantitative results for the strength of the fluctuations as well as for their time dependence and, in the spatially inhomogeneous case, for their spatial distribution.

B. Fluctuations of lattice displacement and momentum

In the continuum limit, the operators for the lattice displacement $\mathbf{u}(\mathbf{r})$ and the lattice momentum $\boldsymbol{\pi}(\mathbf{r})$ caused by LA phonons can be expressed by

$$\mathbf{u}(\mathbf{r}) = i\sum_{\mathbf{q}} \sqrt{\frac{\hbar}{2MN\omega_{\mathrm{LA}}(\mathbf{q})}} (b_{-\mathbf{q}} + b^\dagger_{\mathbf{q}}) \frac{\mathbf{q}}{q} e^{-i\mathbf{q}\cdot\mathbf{r}}, \tag{5}$$

$$\boldsymbol{\pi}(\mathbf{r}) = \sum_{\mathbf{q}} \sqrt{\frac{\hbar M\omega_{\mathrm{LA}}(\mathbf{q})}{2N}} (b_{-\mathbf{q}} - b^\dagger_{\mathbf{q}}) \frac{\mathbf{q}}{q} e^{-i\mathbf{q}\cdot\mathbf{r}}, \tag{6}$$

where N is the number of unit cells in the system volume and M is the overall mass of the atoms in a cell. The operators describe the center-of-mass motion of the unit cells, as we study acoustic phonons.

The fluctuations of the displacement are

$$(\Delta\mathbf{u})^2 = \langle\mathbf{u}(\mathbf{r})\cdot\mathbf{u}(\mathbf{r})\rangle - \langle\mathbf{u}(\mathbf{r})\rangle^2, \tag{7}$$

and analogously for the momentum. This defines a scalar that gives the sum over the fluctuations in the different spatial directions, i.e., $(\Delta\mathbf{u})^2 = \sum_j (\Delta u_j)^2$. Displacement $\mathbf{u}(\mathbf{r})$ and momentum $\boldsymbol{\pi}(\mathbf{r})$ are conjugate variables and therefore have to satisfy the Heisenberg uncertainty relation

$$(\Delta\mathbf{u})^2(\Delta\boldsymbol{\pi})^2 \geqslant \frac{1}{4}\sum_{j,l} |\langle[u_j,\pi_l]\rangle|^2 = \frac{\hbar^2}{4}. \tag{8}$$

Note that the total minimum uncertainty product for the center-of-mass motion is in fact three times the value given here because the two transverse phonon branches each carry the same contribution to the minimum uncertainty as the longitudinal branch. However, in our case transverse phonons are not excited and therefore they only give rise to an additional, time-independent background of vacuum or thermal fluctuation and will not be considered here.

The vacuum uncertainties of the LA phonons are given by

$$(\Delta\mathbf{u})^2_{\mathrm{vac}} = \frac{\hbar}{2MN}\sum_{\mathbf{q}} \frac{1}{\omega_{\mathrm{LA}}(\mathbf{q})}, \tag{9a}$$

$$(\Delta\boldsymbol{\pi})^2_{\mathrm{vac}} = \frac{\hbar M}{2N}\sum_{\mathbf{q}} \omega_{\mathrm{LA}}(\mathbf{q}). \tag{9b}$$

For a constant dispersion relation, which is a good approximation for an optical branch, the product of the vacuum uncertainties reduces to the minimum value $\frac{\hbar^2}{4}$. In contrast, for the LA phonon branch assuming a linear isotropic dispersion relation $\omega_{\mathrm{LA}}(\mathbf{q}) = \omega_q = v_s q$ up to the Debye frequency with v_s being the speed of sound, the uncertainty product in the vacuum state is $(\Delta\mathbf{u})^2_{\mathrm{vac}}(\Delta\boldsymbol{\pi})^2_{\mathrm{vac}} = \frac{9}{8}\frac{\hbar^2}{4}$, thus exceeding the minimal uncertainty value.

In a squeezed state, the fluctuations of one of the conjugate variables, displacement $\mathbf{u}(\mathbf{r})$ or momentum $\boldsymbol{\pi}(\mathbf{r})$, are reduced below their vacuum value. Therefore, we introduce the dimensionless variables

$$S_{\mathbf{u}} = \frac{(\Delta\mathbf{u})^2 - (\Delta\mathbf{u})^2_{\text{vac}}}{(\Delta\mathbf{u})^2_{\text{vac}}}, \tag{10}$$

$$S_{\pi} = \frac{(\Delta\boldsymbol{\pi})^2 - (\Delta\boldsymbol{\pi})^2_{\text{vac}}}{(\Delta\boldsymbol{\pi})^2_{\text{vac}}}, \tag{11}$$

which become negative in the presence of squeezing. For the LA phonon branch in the absence of coherent phonons, i.e., $\langle b_{\mathbf{q}}\rangle = 0$, $S_{\mathbf{u}}$ can be expressed by

$$S_{\mathbf{u}}(\mathbf{r}) = \frac{\hbar}{MN(\Delta\mathbf{u})^2_{\text{vac}}} \sum_{\mathbf{q}_1,\mathbf{q}_2} \frac{1}{\sqrt{\omega_{q_1}\omega_{q_2}}} \frac{\mathbf{q}_1}{q_1}\cdot\frac{\mathbf{q}_2}{q_2} \times \text{Re}[(\langle b_{\mathbf{q}_1} b_{-\mathbf{q}_2}\rangle + \langle b^\dagger_{\mathbf{q}_1} b_{\mathbf{q}_2}\rangle)e^{i(\mathbf{q}_1-\mathbf{q}_2)\mathbf{r}}], \tag{12}$$

involving the generalized phonon occupation $\langle b^\dagger_{\mathbf{q}_1} b_{\mathbf{q}_2}\rangle$ and the two-phonon coherence $\langle b_{\mathbf{q}_1} b_{-\mathbf{q}_2}\rangle$. For S_π, a similar relation holds. The values of $S_{\mathbf{u}}$ can be best compared to the fluctuations of a thermal state, which are constant in time and independent of position. For example, for the GaAs parameters given in Sec. III, in a thermal state $S_{\mathbf{u}}$ has a value of 3.5×10^{-3} at 10 K and at room temperature it is of the order of 1. The vacuum value of the fluctuations of the displacement is $|(\Delta\mathbf{u})_{\text{vac}}| \approx 4 \times 10^{-3}a$, where a is the lattice constant.

C. Modeling the phonon dynamics

For a full modeling of the dynamics in the coupled LO-LA–phonon system, the excitation process of LO phonons has to be included. The complete model Hamiltonian is then given by

$$H = H_0 + H_{\text{anh}} + H_{\text{LOgen}}. \tag{13}$$

H_{LOgen} models an impulsive, optical excitation of the semiconductor, by which coherent LO phonons are generated. It depends on whether a bulk or a quantum dot system is considered and therefore will be discussed later.

The dynamics of the system is calculated within the density-matrix formalism. To determine the fluctuations of lattice displacement and momentum, the LA phonon variables $\langle b_{\mathbf{q}}\rangle, \langle b_{\mathbf{q}_1} b_{\mathbf{q}_2}\rangle$, and $\langle b^\dagger_{\mathbf{q}_1} b_{\mathbf{q}_2}\rangle$ are needed. If the system is initially in a state without a coherent LA amplitude $\langle b_{\mathbf{q}}\rangle$, this quantity remains zero as the model Hamiltonian only consists of terms with an even number of LA creation/annihilation operators. Therefore, only expectation values of terms with an even number of LA operators can become nonzero. We have already seen this characteristic in Sec. II A, where the system evolves into a two-mode squeezed vacuum state without a coherent amplitude. For the LA phonon variables $\langle b_{\mathbf{q}_1} b_{\mathbf{q}_2}\rangle$ and $\langle b^\dagger_{\mathbf{q}_1} b_{\mathbf{q}_2}\rangle$, the equations of motion can be derived from the Hamiltonian H. Due to the many-body nature of this problem, an infinite hierarchy of equations of motion builds up, which we truncate by the factorization of all density matrices that contain three or more phonon operators.[23] The equations of motion are then given by

$$i\hbar\frac{d}{dt}\langle b^\dagger_{\mathbf{q}_1} b_{\mathbf{q}_2}\rangle = \hbar(\omega_{q_2}-\omega_{q_1})\langle b^\dagger_{\mathbf{q}_1} b_{\mathbf{q}_2}\rangle + 2\sum_{\mathbf{k}}(\lambda_{\mathbf{q}_2,\mathbf{k}}\langle a_{\mathbf{k}}\rangle \times \langle b^\dagger_{\mathbf{q}_1} b^\dagger_{\mathbf{k}-\mathbf{q}_2}\rangle - \lambda_{\mathbf{q}_1,\mathbf{k}}\langle a^\dagger_{\mathbf{k}}\rangle\langle b_{\mathbf{q}_2} b_{\mathbf{k}-\mathbf{q}_1}\rangle), \tag{14}$$

$$i\hbar\frac{d}{dt}\langle b_{\mathbf{q}_1} b_{\mathbf{q}_2}\rangle = \hbar(\omega_{q_1}+\omega_{q_2})\langle b_{\mathbf{q}_1} b_{\mathbf{q}_2}\rangle + 2\sum_{\mathbf{k}}(\lambda_{\mathbf{q}_2,\mathbf{k}}\langle a_{\mathbf{k}}\rangle\langle b^\dagger_{\mathbf{k}-\mathbf{q}_2} b_{\mathbf{q}_1}\rangle + \lambda_{\mathbf{q}_1,\mathbf{k}}\langle a_{\mathbf{k}}\rangle\langle b^\dagger_{\mathbf{k}-\mathbf{q}_1} b_{\mathbf{q}_2}\rangle) + \lambda_{\mathbf{q}_1,\mathbf{q}_1+\mathbf{q}_2}\langle a_{\mathbf{q}_1+\mathbf{q}_2}\rangle. \tag{15}$$

Here the fact that $\langle b_{\mathbf{q}}\rangle \equiv 0$ has been used. We note that the density matrices $\langle b^\dagger_{\mathbf{q}_1} b_{\mathbf{q}_2}\rangle$ are only indirectly driven via the two-phonon coherences $\langle b_{\mathbf{q}_1} b_{\mathbf{q}_2}\rangle$, which in turn are directly driven by the coherent LO phonon amplitude $\langle a_{\mathbf{k}}\rangle$. For weak coupling, this means that if either the coupling constant or the LO phonon amplitude is multiplied by a constant factor, the latter matrices change linearly with this factor, whereas the former change quadratically.

III. RESULTS

In our calculations, we have used material parameters for GaAs. It is still under discussion whether the decay into two LA phonons is the dominant decay channel for LO phonons in this material;[18,19,24] however, our general findings are not affected by the specific choice of the material parameters and should be applicable to any similar system in which the decay into two LA phonons is strong, such as, e.g., GaP or InP. We restrict our model to the LA and LO phonon branches coupled by the anharmonic decay process. The dispersion relation of the LA phonons is assumed to be linear with $\omega_q = v_s q$ ($v_s = 5110$ m/s) up to the Debye frequency. The $\mathbf{k}$ dependence of the LO phonon energy is neglected and $\hbar\omega_{\text{LO}} = 36.4$ meV is taken. This is a good approximation near the center of the Brillouin zone, which is where the LO phonons will be generated. The coupling constant $\lambda_{\mathbf{q},\mathbf{k}}$ of the decay depends on the phonon wave vector involved and can be approximated by $\lambda_{\mathbf{q},\mathbf{k}+\mathbf{q}} = \frac{4\hbar}{\omega^2_{\text{LO}}}(\frac{\pi v_s^5}{\tau V})^{1/2}\sqrt{qk}$, where τ is the lifetime of the LO phonons.[25,26] The lifetime τ is about 7–10 ps at low temperatures in bulk GaAs,[27] and between 6 and 8 ps in GaAs QD's.[28] For better comparability, we have assumed a value of $\tau = 7$ ps in both cases.

A. Results: Bulk

We will first consider a GaAs bulk semiconductor. The LO phonons are indirectly generated by an optical excitation of the electronic degrees of freedom. The optical excitation creates electron-hole pairs, which then interact with the LO phonons via a polar or nonpolar electron-phonon interaction. Independent of the exact type of interaction, the coupling between carriers and LO phonons in a two-band model can be described by the Hamiltonian[23]

$$H_{\text{LOgen}} = \sum_{\mathbf{k}',\mathbf{k}}\left(M^e_{\mathbf{k}',\mathbf{k}} a_{\mathbf{k}} + M^{e*}_{\mathbf{k}',-\mathbf{k}} a^\dagger_{-\mathbf{k}}\right)c^\dagger_{\mathbf{k}'+\mathbf{k}} c_{\mathbf{k}'} - \sum_{\mathbf{k}',\mathbf{k}}\left(M^h_{\mathbf{k}',\mathbf{k}} a_{\mathbf{k}} + M^{h*}_{\mathbf{k}',-\mathbf{k}} a^\dagger_{-\mathbf{k}}\right)d^\dagger_{\mathbf{k}'+\mathbf{k}} d_{\mathbf{k}'}, \tag{16}$$

where $c^\dagger_{\mathbf{k}}$ and $c_{\mathbf{k}}$ ($d^\dagger_{\mathbf{k}}$ and $d_{\mathbf{k}}$) are the creation and annihilation operators for an electron (hole) with wave vector $\mathbf{k}$, and $M^{e/h}_{\mathbf{k},\mathbf{k}'}$ is the coupling matrix element. The equation of motion for the coherent LO phonon amplitude, including the optical excitation and the anharmonic decay, is

$$i\hbar \frac{d}{dt}\langle a_{\mathbf{k}}\rangle = \hbar\omega_{\mathrm{LO}}\langle a_{\mathbf{k}}\rangle + \sum_{\mathbf{q}} \lambda_{\mathbf{q},\mathbf{k}}\langle b_{\mathbf{q}} b_{\mathbf{k}-\mathbf{q}}\rangle + \sum_{\mathbf{k}'}\left(M^{e*}_{\mathbf{k}',\mathbf{k}}\langle c^{\dagger}_{\mathbf{k}'-\mathbf{k}} c_{\mathbf{k}'}\rangle - M^{h*}_{\mathbf{k}',\mathbf{k}}\langle d^{\dagger}_{\mathbf{k}'-\mathbf{k}} d_{\mathbf{k}'}\rangle\right). \tag{17}$$

We assume an optical excitation near the band gap with an ultrafast laser pulse and a pulse duration shorter than the time scale of phonon dynamics. Therefore, the δ-pulse limit with a steplike time dependence of the electronic density matrices is applicable, which leads to a displacive excitation of coherent phonons (DECP).[29,30] Furthermore, the matrices are diagonal due to spatial homogeneity,[21] which means that only LO phonons with $\mathbf{k} = \mathbf{0}$ are generated.

Coherent LO phonons correspond to an oscillation of the relative position of the ions in a unit cell. The magnitude of the excited carrier density and the coupling constants $M^{e/h}_{\mathbf{k},\mathbf{k}'}$ determine the initial amplitude A_{LO} of this oscillation. As only this amplitude is important for the phonon dynamics, we will use A_{LO} as a parameter.

Similar to Eq. (16), a direct coupling between carriers and LA phonons could be included, which in principle could cause a coherent LA phonon amplitude $\langle b_{\mathbf{q}}\rangle$. However, the same restriction to $\mathbf{q} = \mathbf{0}$ applies, which in the case of LA phonons would signify a translation of the crystal as a whole. Furthermore, the phonons that appear as final states of the decay of LO phonons are well separated in momentum space from phonons with $\mathbf{q} = \mathbf{0}$ and do not interfere with the latter. Therefore, direct couplings between carriers and LA phonons are not considered here as the phonons generated by the LO phonon decay are the focus of the present study.

The anharmonic decay process consumes LO phonons and thereby dampens the oscillation. Each LO phonon decays into a pair of LA phonons as sketched in Fig. 1(a) . In a semiclassical picture, momentum and energy conservation require that in each pair both LA phonons carry half of the LO phonon energy, $\hbar\omega_{\mathrm{LA}}(q_0) = \hbar\omega_{\mathrm{LO}}/2 = 18.2$ meV, and have opposite momenta $\pm\mathbf{q}_0$. A quantum kinetic calculation of the LA phonon population $\langle b^{\dagger}_{\mathbf{q}} b_{\mathbf{q}}\rangle$ resulting from the decay is shown in Fig. 1(b); due to the assumed isotropy of the system, $\langle b^{\dagger}_{\mathbf{q}} b_{\mathbf{q}}\rangle$ only depends on the absolute value q. We see that indeed with increasing time a peak at q_0 builds up. The peak is broadened by the energy-time uncertainty, which is also reflected in the small side peaks.

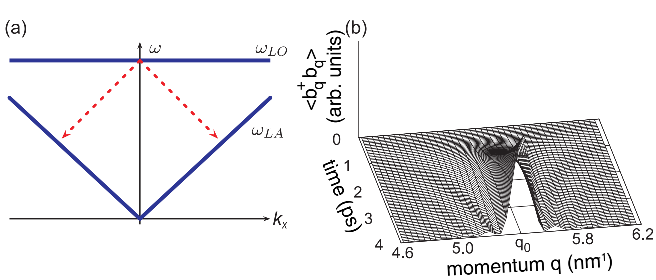

FIG. 1. (Color online) (a) Sketch of the phonon dispersions $\omega_j(k_x)$ and the anharmonic decay for the bulk system. (b) Resulting population of the LA phonons $\langle b^{\dagger}_{\mathbf{q}} b_{\mathbf{q}}\rangle$ as a function of time.

Let us now turn to the fluctuation properties of the generated LA phonons. In the bulk case, the fluctuations $S_{\mathbf{u}}$ can be simplified to

$$S_{\mathbf{u}} = \frac{\hbar}{MN(\Delta\mathbf{u})^2_{\mathrm{vac}}} \sum_{\mathbf{q}} \frac{1}{\omega_q}\mathrm{Re}(\langle b^{\dagger}_{\mathbf{q}} b_{\mathbf{q}}\rangle + \langle b_{\mathbf{q}} b_{-\mathbf{q}}\rangle). \tag{18}$$

Here each spatial direction contributes equally to the fluctuations. Due to spatial homogeneity, $S_{\mathbf{u}}$ does not depend on $\mathbf{r}$. While the phonon populations $\langle b^{\dagger}_{\mathbf{q}} b_{\mathbf{q}}\rangle$ are always positive, the real part of the coherences $\langle b_{\mathbf{q}} b_{-\mathbf{q}}\rangle$ can become negative and therefore may yield squeezing.

Figure 2(a) shows the fluctuations $S_{\mathbf{u}}$ as a function of time for an initial amplitude of the lattice oscillation of $A_{\mathrm{LO}} = 10^{-4}a$. Phonon squeezing is clearly visible, as $S_{\mathbf{u}}$ becomes negative for certain times. $S_{\mathbf{u}}$ oscillates with twice the LA phonon frequency $\omega_{\mathrm{LA}}(q_0)$, which is the same as the LO phonon frequency $2\omega_{\mathrm{LA}}(q_0) = \omega_{\mathrm{LO}}$. Note that a double frequency oscillation of the fluctuations is a necessary condition for a squeezed state, but it is not a sufficient one.[10,22,31] The fluctuations of the momentum $\boldsymbol{\pi}(\mathbf{r})$ behave analogously, but oscillate phase-shifted with respect to $S_{\mathbf{u}}$, and thus the Heisenberg uncertainty principle is satisfied for all times.

In contrast to the LA phonon population, which increases with time [cf. Fig. 1(b)], the amplitude of the oscillation of $S_{\mathbf{u}}$ is almost constant with time [Fig. 2(a)] and for longer times even decreases [Fig. 2(b)]. This is because the coherences $\langle b_{\mathbf{q}} b_{-\mathbf{q}}\rangle$ for phonon modes with different wave

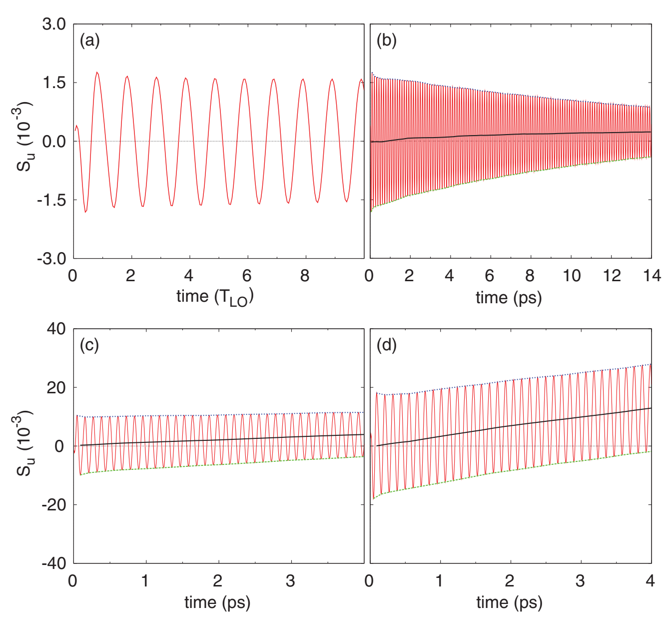

FIG. 2. (Color online) Fluctuations of the lattice displacement $S_{\mathbf{u}}$ as a function of time for $A_{\mathrm{LO}} = 10^{-4}a$ (a) for short times in units of $T_{\mathrm{LO}} \approx 0.11$ ps and (b) the long-time behavior in picoseconds. (c) $S_{\mathbf{u}}$ for $A_{\mathrm{LO}} \approx 5.7 \times 10^{-4}a$, which corresponds to a coupling $M^{e/h} = C^{e/h}\sqrt{\frac{\hbar}{2MN\omega_{\mathrm{LO}}}}$ with $C^h - C^e = 10^9$ eV/cm and an excitation density of 10^{19} cm^{-3}, cf. Ref. 21. (d) $S_{\mathbf{u}}$ for $A_{\mathrm{LO}} = 10^{-3}a$ in picoseconds. Also shown: envelope and center of the oscillation.

vectors $\mathbf{q}$ each oscillate with an individual frequency and therefore quickly acquire independent phases. The oscillation amplitude of $S_\mathbf{u}$, therefore, does not increase with time but follows adiabatically the strength of the LO oscillation. For longer times, the amplitude $\langle a_\mathbf{k}\rangle$ decays exponentially with a decay time of $2\tau = 14$ ps. Since this is the driving term for the two-phonon coherences $\langle b_\mathbf{q} b_{-\mathbf{q}}\rangle$, the squeezing is reduced in the same way. In Fig. 2(b), the rising LA phonon population also contributes noticeably to the fluctuations by shifting the oscillation upward. Therefore, also the product of the fluctuations of displacement and momentum $(\Delta\mathbf{u})(\Delta\boldsymbol{\pi})$, which equals the product of the vacuum uncertainties in the beginning, increases with time.

The different time dependencies of the contributions to the fluctuations resulting from the phonon populations $\langle b^\dagger_\mathbf{q} b_\mathbf{q}\rangle$ and the two-phonon coherences $\langle b_\mathbf{q} b_{-\mathbf{q}}\rangle$ are closely related to the energy-time uncertainty inherent in our quantum kinetic approach. The uncertainty causes a range of different phonon modes to be excited for both expectation values, which is getting narrower with time. Indeed, solving Eqs. (14) and (15) to the lowest order in the coupling matrix element $\lambda_{\mathbf{q},0}$, we obtain as dominant terms around the resonant transition for the modulus of the two-phonon coherence

$$|\langle b_\mathbf{q} b_{-\mathbf{q}}\rangle| \sim \left| \frac{\lambda_{\mathbf{q},0} A_{\mathrm{LO}}}{\hbar} \frac{\sin\frac{\Delta\omega_q t}{2}}{\Delta\omega_q} \right| \tag{19}$$

and for the phonon population

$$\langle b^\dagger_\mathbf{q} b_\mathbf{q}\rangle \sim \left| \frac{\lambda_{\mathbf{q},0} A_{\mathrm{LO}}}{\hbar} \right|^2 \frac{\sin^2\frac{\Delta\omega_q t}{2}}{(\Delta\omega_q)^2}, \tag{20}$$

where we have introduced the detuning of the transition from resonance $\Delta\omega_q = \omega_{\mathrm{LO}} - 2\omega_{\mathrm{LA}}(q)$. Thus, while the energy uncertainty, i.e., the width of the central peak, decreases in both cases $\sim 1/t$, the resonant phonon population rises quadratically while the modulus of the coherence grows only linearly with time. Therefore, their overall contributions to $S_\mathbf{u}$ [cf. Eq. (18)] are linearly rising and constant with time, respectively.

Interestingly, the behavior of $S_\mathbf{u}$ is completely different from previous findings for the two-mode quadrature operators $P_{\pm\mathbf{q}_0}$ and $X_{\pm\mathbf{q}_0}$ (cf. Sec. II A) that have been studied accounting only for energy-conserving decay processes and disregarding changes in the LO phonon system.[13] In these studies, the amplitudes of the fluctuations of $P_{\pm\mathbf{q}_0}$ and $X_{\pm\mathbf{q}_0}$ are rising constantly. There are two main reasons for this significant difference in the behavior: On the one hand, in the previous studies no dephasing occurs, as all the phonon operators that enter $P_{\pm\mathbf{q}_0}$ and $X_{\pm\mathbf{q}_0}$ develop in time with the same frequency $\omega_{\mathrm{LA}}(q_0)$. On the other hand, the coherent LO phonon amplitude does not decay with time, since the influence of the decay process on the LO phonon subsystem has been neglected.

The oscillation amplitude and the shift of $S_\mathbf{u}$ depend strongly on the initial LO phonon amplitude A_{LO}. This is demonstrated in Figs. 2(c) and 2(d), where $S_\mathbf{u}$ is plotted for larger values of A_{LO}. The oscillation amplitude of the fluctuations scales linearly with A_{LO}, because in lowest order the coherences $\langle b_\mathbf{q} b_{-\mathbf{q}}\rangle$ depend linearly on the coherent LO phonon amplitude $\langle a_\mathbf{q}\rangle$ [cf. Eq. (15)]. At the same time, the shift of $S_\mathbf{u}$ increases quadratically with A_{LO}, because the LA phonon population after a fixed time is proportional to the initial LO phonon population, which increases quadratically with A_{LO}. Thus for a larger initial amplitude A_{LO} the squeezing is more pronounced in the beginning, but vanishes faster.

B. Results: Quantum dot

Let us now turn to the case of a quantum dot structure. We consider a spherical GaAs QD with a parabolic confinement potential in the strong confinement limit. The dot has a diameter of 10 nm, where the diameter is defined as the full width at half-maximum of the electron density in the exciton ground state. The electronic states are modeled by a two-level system. As in the bulk case, we restrict ourselves to the coupling of the carriers to LO phonons, which is modeled here in terms of the Fröhlich interaction. It should be mentioned that in this case the direct coupling to acoustic phonons leads to the generation of coherent LA phonons with nonvanishing wave vectors. However, these coherent LA phonons involve modes with small wave vectors and are therefore well separated in reciprocal space from those created in the decay of LO phonons. Furthermore, for an excitation by a π pulse, as will be studied here, those phonons are in a purely coherent state[32] and therefore do not give rise to additional fluctuations.[17] Therefore, direct couplings between carriers and LA phonons can be disregarded here as in the bulk case.

The Hamiltonian describing the interaction of carriers and LO phonons is[23]

$$H_{\mathrm{LOgen}} = \sum_\mathbf{k} \left(g^e_\mathbf{k} a_\mathbf{k} + g^{e*}_\mathbf{k} a^\dagger_\mathbf{k}\right) c^\dagger c - \sum_\mathbf{k} \left(g^h_\mathbf{k} a_\mathbf{k} + g^{h*}_\mathbf{k} a^\dagger_\mathbf{k}\right) d^\dagger d\ , \tag{21}$$

where $c^\dagger c$ and $d^\dagger d$ are the electron and hole number operators of the lowest conduction- and highest valence-band state, respectively. The coupling matrix elements $g^{e/h}_\mathbf{k}$ depend on the localized wave functions of electron $\psi_e(\mathbf{r})$ and hole $\psi_h(\mathbf{r})$ of the exciton according to

$$g^{e/h}_\mathbf{k} = g_k \int \psi^*_{e/h}(\mathbf{r}) e^{i\mathbf{k}\cdot\mathbf{r}} \psi_{e/h}(\mathbf{r}) d^3r. \tag{22}$$

The prefactor is the bulk Fröhlich coupling constant given by $g_k = \frac{1}{k}\sqrt{\frac{e^2\hbar\omega_{\mathrm{LO}}}{2\epsilon_0 V}\left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_s}\right)}$, where ϵ_∞ and ϵ_s are the high- and low-frequency dielectric constants, respectively, and ϵ_0 is the vacuum permittivity. The overall interaction vanishes for identical carrier wave functions and is more effective for electron and hole wave functions, which are localized differently. The equation of motion for the coherent LO phonon amplitude is given by

$$i\hbar\frac{d}{dt}\langle a_\mathbf{k}\rangle = \hbar\omega_{\mathrm{LO}}\langle a_\mathbf{k}\rangle + \sum_\mathbf{q} \lambda_{\mathbf{q},\mathbf{k}} \langle b_\mathbf{q} b_{\mathbf{k}-\mathbf{q}}\rangle + g^{e*}_\mathbf{k}\langle c^\dagger c\ \rangle - g^{h*}_\mathbf{k}\langle d^\dagger d\ \rangle. \tag{23}$$

Again, the optical excitation is assumed to be on a shorter time scale than the phonon dynamics so that the δ-pulse limit is applicable.[32,33] A π pulse abruptly inverts the exciton system, i.e., it changes the electron and hole occupations from zero to

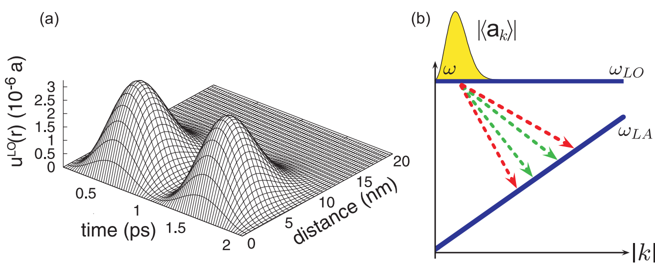

FIG. 3. (Color online) (a) Radial LO phonon lattice displacement $\mathbf{u}^{\mathrm{LO}}(\mathbf{r}) \cdot \mathbf{e}_r$ as a function of distance to the dot center and time. (b) Sketch of the phonon dispersions $\omega_j(k)$ and the decay for a QD system.

one. A radiative recombination of the exciton typically occurs on a time scale of nanoseconds and is therefore not taken into account. Here the electronic system is confined, which leads to a localized generation of LO phonons. Hence, the equations of motion for the coherent LO phonon amplitudes $\langle a_{\mathbf{k}} \rangle$ are similar to Eq. (17), but in contrast to the bulk case now a continuum of phonon modes with different wave vectors $\mathbf{k}$ is excited.

The localization of the LO phonons can be seen in Fig. 3(a) , where the radial LO phonon lattice displacement $\mathbf{u}^{\mathrm{LO}}(\mathbf{r}) \cdot \mathbf{e}_r$ is shown as a function of the distance to the dot center and time. In contrast to the acoustic phonons, here the lattice displacement describes a relative motion of the ions in a unit cell. As a function of the distance, the amplitude of the lattice displacement has a clear maximum at 5 nm, which coincides with the radius of the QD, and decreases rapidly for larger distances. In time, we find a sinusoidal oscillation of the lattice displacement with the LO phonon frequency.

The LO phonon amplitude in k space is sketched in the upper part of Fig. 3(b). Due to the spherical symmetry, only the absolute values of the wave vectors are important. Again, the arrows indicate which decay processes into pairs of LA phonons are allowed by energy and momentum conservation. Because now the LO phonon wave vector $\mathbf{k}$ is nonvanishing, the absolute value of the LA phonon wave vector has a range of possible values. The numerical results for the distribution of LA phonons in k space are shown in Fig. 4(a) . A single peak around q_0 appears, which indeed is broader than in the bulk case. A comparison for $t = 2$ ps is shown in the inset.

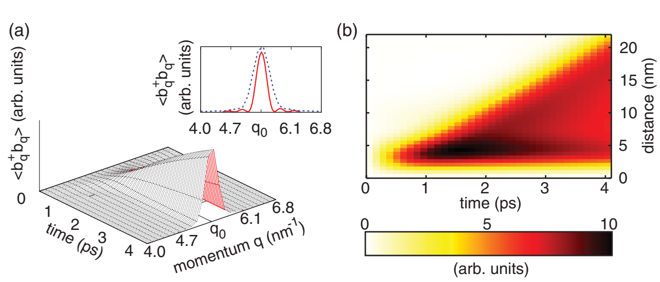

FIG. 4. (Color online) (a) Resulting population of the LA phonons $\langle b_{\mathbf{q}}^\dagger b_{\mathbf{q}} \rangle$ as a function of time. Inset: comparison of the distributions of a bulk (solid line) and a QD (dotted line) system at $t = 2$ ps. (b) Energy density $\rho_E(r,t)r^2$ vs. time and distance to the center of the QD time-averaged over the LO phonon period.

We now analyze the spatial evolution of the LA phonons in real space. For this purpose, we consider the energy density $\rho_E(\mathbf{r},t)$ of the LA phonons, which gives rise to the phonon contribution to heat transport, and is given by[34]

$$\rho_E(\mathbf{r},t) = \frac{\hbar}{2V} \sum_{\mathbf{q}_1,\mathbf{q}_2} \sqrt{\omega_{q_1}\omega_{q_2}} \left(1 + \frac{\mathbf{q}_1}{q_1} \cdot \frac{\mathbf{q}_2}{q_2}\right) \times \mathrm{Re}\left[\langle b_{\mathbf{q}_1}^\dagger b_{\mathbf{q}_2} \rangle e^{i(\mathbf{q}_1 - \mathbf{q}_2)\mathbf{r}} + \langle b_{\mathbf{q}_1} b_{\mathbf{q}_2} \rangle e^{i(\mathbf{q}_1 + \mathbf{q}_2)\mathbf{r}}\right]. \tag{24}$$

By integrating over the system volume V, this gives the total LA phonon energy $E_{\mathrm{LA}} = \sum_{\mathbf{q}} \hbar\omega_q \langle b_{\mathbf{q}}^\dagger b_{\mathbf{q}} \rangle$. In the present case, $\rho_E(\mathbf{r},t)$ only depends on the distance r to the quantum dot center due to symmetry. The QD acts as a source for LA phonons that travel out of the dot into the surrounding solid and thus provide heat transport. Superimposed on this spatial expansion are fast oscillations of ρ_E in time and space that result from the coherences $\langle b_{\mathbf{q}_1} b_{\mathbf{q}_2} \rangle$, which tend to dominate the behavior of ρ_E on a time scale shorter than the LO phonon period. In order to make the expansion more clearly visible, the energy density can be time-averaged over a full LO period. The resulting average of $\rho_E(\mathbf{r},t) \cdot r^2$ is shown in Fig. 4(b), where the factor r^2 compensates the spatial decay due to the spherical expansion of the wave packet. Here we see that the generation of the LA phonons takes place within the area of the quantum dot, to which the LO phonons are confined. Constantly new LA phonons are generated and propagate away from the dot with the sound velocity v_s.

The fluctuations $S_{\mathbf{u}}$ of the generated LA phonon displacement field are shown in Fig. 5(a) . We find that the lattice displacement is squeezed as $S_{\mathbf{u}}$ is negative for certain time intervals. As a function of time, the fluctuations oscillate around zero with the frequency $2\omega_{\mathrm{LA}}(q_0) = \omega_{\mathrm{LO}}$. As in the bulk case, the oscillation amplitude does not increase with time due to the fast dephasing of the two-phonon coherences. The long-time behavior is very similar to the bulk case and not explicitly shown here. The spatial distribution shows an interesting behavior. Even for longer times, $S_{\mathbf{u}}$ becomes negative only in the area of the dot. The squeezing does not travel out of the dot together with the LA phonon energy density; a squeezed lattice displacement is generated within the dot, but only the LA phonon population, which leads to an overall increase of the fluctuations, leaves the dot.

This effect can be explained by an analytically tractable approximation for $S_{\mathbf{u}}$. We evaluate the model only up to the lowest order of the phonon-phonon coupling $\lambda_{\mathbf{q},\mathbf{k}}$ (where $\mathbf{k}$ is the LO and $\mathbf{q}$ is the LA phonon wave vector). In this case, there is no LA phonon population and $S_{\mathbf{u}}$ is solely determined

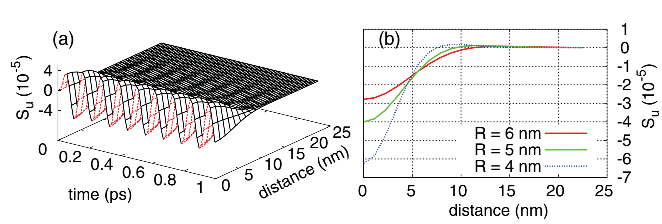

FIG. 5. (Color online) (a) Lattice fluctuation $S_{\mathbf{u}}$ vs time and distance to the center of the QD. (b) $S_{\mathbf{u}}$ at $t = 2T_{\mathrm{LO}}$ for different radii R of the QD.

by the coherences $\langle b_{\frac{\mathbf{k}}{2}+\mathbf{q}} b_{\frac{\mathbf{k}}{2}-\mathbf{q}}\rangle$, whose equations of motion can now be solved analytically. For the anharmonic decay processes considered in this paper, the LO phonon wave vector k is small compared to the wave vector of the generated LA phonons q. Therefore, we assume that $k^2/(4q^2)$ is negligibly small compared to 1. This leads to the analytical expression

$$S_{\mathbf{u}} = C\,\mathrm{Re}\left\{\int_0^{q_{\max}} \left(g_k^{e*} - g_k^{h*}\right) \frac{\sin(kr)}{r} k\, dk \times \int_0^{q_{\max}} \frac{\mathrm{e}^{-i\omega_{\mathrm{LO}}t} - \mathrm{e}^{-2i\omega_q t}}{\omega_{\mathrm{LO}} - 2\omega_q} q^2 dq\right\}, \tag{25}$$

with a real constant C and $q_{\max}$ being the maximum phonon wave vector. In this approximation of $S_{\mathbf{u}}$, we see that the spatial dependence of the fluctuations, described by the first integral, is time-independent and localized around $r = 0$. This means that the squeezing is confined to the dot for all times. The essential point in this approximation is the assumption $k^2/(4q^2) \ll 1$, which implies that phonon coherences $\langle b_{\mathbf{q}} b_{\mathbf{q}'}\rangle$ with $\mathbf{q} \approx -\mathbf{q}'$ are generated. If, in turn, coherences with $\mathbf{q} \approx \mathbf{q}'$ were generated, as in the degenerate parametric down conversion of photons, the fluctuations would be traveling like the phonon population.

Figure 5(b) demonstrates the effect of different dot sizes. Shown is the spatial dependence of the fluctuations for different sizes of the QD at a fixed time of $t = 2T_{\mathrm{LO}}$, when the oscillation has an extremum. As expected, the spatial confinement of $S_{\mathbf{u}}$ scales with the size of the QD. We also see that the oscillation amplitude is larger for smaller dots; this is because the coupling between carriers and LO phonons is stronger for smaller dots and therefore more LO phonons are generated.

IV. CONCLUSIONS

We have studied the fluctuation properties of LA phonons generated by the anharmonic decay of LO phonons in two different systems, a bulk semiconductor and a semiconductor quantum dot. In both systems, the time behavior of the fluctuations of the lattice displacement are similar. The fluctuations oscillate with twice the frequency of the resonantly generated LA phonons and for small times repeatedly fall below their vacuum value, i.e., the system is in a squeezed state with respect to the variables of lattice displacement and momentum. The squeezing disappears with increasing time due to the dephasing of the different LA phonon modes and the rising of the LA phonon population, which increases the fluctuations. Hence, a larger coherent LO amplitude initially causes stronger squeezing, but the effect vanishes faster. In the bulk system, the fluctuations of the lattice displacement are spatially homogeneous due to symmetry. In contrast, in the quantum dot case a spatial structure is imprinted on the system and the LA phonons are created locally at the dot. Although LA phonon wave packets leave the dot, the squeezing effect is confined to the vicinity of the dot. This is clearly different from the otherwise similar case of the degenerate parametric down conversion of photons. The difference is caused by the fact that by the anharmonic decay of phonons, predominantly coherences $\langle b_{\mathbf{q}} b_{\mathbf{q}'}\rangle$ with $\mathbf{q} \approx -\mathbf{q}'$ are generated, while in degenerate photon down conversion $\mathbf{q} \approx \mathbf{q}'$.

For our present calculations, we have assumed a decay of the LO phonons into a pair of LA phonons, the so called Klemens channel.[25] Since the appearance of two-mode squeezing is related to the structure of the anharmonic Hamiltonian, squeezing in the corresponding two-mode quadrature operators should be expected also for other decay channels.[24] Our results for the fluctuations of the lattice displacement and the momentum, however, depend on the specific modes and the dispersion relation of the phonons into which the decay occurs. Therefore, they cannot directly be transferred to other decay channels. Nevertheless, comparable effects seem likely due to the formal similarity of the Hamiltonian.

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