Lattice Fluctuations at a Double Phonon Frequency with and without Squeezing: An Exactly Solvable Model of an Optically Excited Quantum Dot

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Time-dependent lattice fluctuations of an optically excited strongly confined quantum dot are investigated with the aim to analyze the characteristics commonly used for identifying the presence of squeezed phonon states. It is demonstrated that the appearance of fluctuations oscillating with twice the phonon frequency, commonly regarded as a clear indication of squeezed states, cannot be considered as such. The source of the discrepancy with earlier investigations is discussed. Conditions for generating a squeezed state by using a two-pulse excitation are analyzed.

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Fluctuation properties of quantized systems strongly deviate from those of classical systems. On the one hand, Heisenberg's uncertainty relation establishes a lower limit on the product of fluctuations of conjugate variables such as position and momentum or the quadrature components of an electromagnetic field. On the other hand there are quantum states—the squeezed states—in which the fluctuations of one of these variables may be considerably reduced below their zero-point value. Squeezed states of light have been extensively studied in quantum optics both experimentally and theoretically [1–3]. They are attractive for applications such as quantum teleportation [4] or quantum-enhanced gravitational wave detection [5]. The concept of squeezed states is, however, not restricted to photons and squeezing has been studied also in other systems like atomic Bose-Einstein condensates [6], surface plasmons [7], or magnons [8]. In addition, there has been a growing interest in squeezing in the case of quantized lattice vibrations, i.e., phonons. To this end fluctuations of lattice displacements (FLDs) have been analyzed experimentally in many solid state systems including metals and semimetals like Bi [9,10] and Sb [11], perovskite materials like $KTaO_3$ [12,13] or $SrTiO_3$ [14], semiconductors like GaAs [9], as well as high-T_C superconductors [15]. Although these systems are very different it is commonly found that the FLDs exhibit modulations with both the single and the double phonon frequency. A characteristic phonon frequency may arise, e.g., from the confinement of an acoustic mode, from a van Hove singularity in the phonon density of states, or from the excitation of a nearly dispersionless longitudinal optical (LO) phonon mode. Note that no matter which types of phonons are involved the FLDs are time independent both for a thermal as well as for a coherent state. In contrast, a system in a squeezed state fluctuates at twice the phonon frequency. Thus, the observation of double phonon frequency oscillations in the FLDs has often been taken as an evidence for phonon squeezing [9–11,14].

In this Letter we study theoretically the fluctuation properties of lattice displacements caused by ultrafast optical excitations of a quantum dot (QD). From the experiments in Refs. [9–15] it is evident that the occurrence of single and double phonon frequency modulations of FLDs is a generic feature which does not critically depend on details of the electronic system. This suggests that also a simple electronic system, such as the QD studied here, should be able to capture the essential aspects of the fluctuation dynamics. We can thus benefit from the fact that for ultrafast excitations all dynamical variables may be evaluated analytically in our model without further approximations [16]. Our main conclusion will be that in contrast to a widespread belief the occurrence of double phonon frequency modulations of the fluctuations is not a sufficient criterion to unambiguously indicate the presence of squeezing. Squeezed states appear only in special cases, in particular, when a sequence of ultrashort pulses with selected phases excites the dot.

We consider a resonant ultrafast excitation of the lowest QD exciton. Assuming circularly polarized laser pulses and a sufficiently strong confinement of the QD states the electronic degrees of freedom may be restricted to an effective two-level system consisting of the crystal ground state $|g\rangle$ and the single exciton state $|x\rangle$. The coupling to the light field is described within the usual dipole and rotating wave approximation. Since phonon-induced transitions to higher states or across the band gap can be neglected because of the strong energy mismatch, the coupling to phonons is typically dominated by pure dephasing type interactions as described by the independent Boson model. Corresponding calculations accounting for acoustic phonons were indeed able to quantitatively reproduce the measured polarization decay on short times [17]. The coupling to optical phonons gives rise to discrete phonon side bands in absorption or luminescence spectra [18]. Here we will concentrate on the quantum state of LO phonons coupled to the exciton via the Fröhlich interaction. The corresponding Hamiltonian reads

$$\hat{H} = \hbar\Big[\Omega + \sum_{\mathbf{q}}(g_{\mathbf{q}}\hat{b}_{\mathbf{q}} + g^*_{\mathbf{q}}\hat{b}^\dagger_{\mathbf{q}})\Big]|x\rangle\langle x| + \hbar\omega_{\rm LO}\sum_{\mathbf{q}}\hat{b}^\dagger_{\mathbf{q}}\hat{b}_{\mathbf{q}} - \hat{\mathbf{P}}\cdot\mathbf{E}, \tag{1}$$

where $\hbar\Omega$ is the exciton energy, $\hat{b}_{\mathbf{q}}$ ($\hat{b}^\dagger_{\mathbf{q}}$) are annihilation (creation) operators of a LO phonon with wave vector $\mathbf{q}$, $g_{\mathbf{q}}$ describes the exciton-phonon coupling, $\mathbf{E}$ is the laser field, and $\hat{\mathbf{P}} = \mathbf{M}_0|x\rangle\langle g| + \mathbf{M}_0^*|g\rangle\langle x|$ is the operator for the electronic polarization with the transition dipole matrix element $\mathbf{M}_0$. We use parameters typical for a spherical InGaAs QD of 5 nm diameter and parametrize the couplings $g_{\mathbf{q}}$ as in Ref. [16]. Although the pulse spectra should be sufficiently narrow to avoid excitations of higher electronic states, they still can be broad enough to cover the relevant phonon sidebands. This is the limit of ultrafast excitation, in which the calculations can be simplified by modeling the laser field as a sequence of δ-shaped pulses with pulse areas A_j and phases ϕ_j arriving at times t_j.

The operator for the displacement associated with LO phonons is given by

$$\hat{\mathbf{u}}(\mathbf{r}, t) = -i\frac{u_0}{\sqrt{N}}\sum_{\mathbf{q}}\frac{\mathbf{q}}{q}(\hat{b}_{\mathbf{q}}e^{i\mathbf{q}\cdot\mathbf{r}} - \hat{b}^\dagger_{\mathbf{q}}e^{-i\mathbf{q}\cdot\mathbf{r}}), \tag{2}$$

where $u_0 = \sqrt{\hbar/(2M_r\omega_{\rm LO})}$, M_r is the reduced mass of the lattice ions, and N denotes the number of unit cells. FLDs are described by the variance $(\Delta\mathbf{u}(\mathbf{r}, t))^2 = \langle\hat{\mathbf{u}}(\mathbf{r}, t)\cdot\hat{\mathbf{u}}(\mathbf{r}, t)\rangle - \langle\hat{\mathbf{u}}(\mathbf{r}, t)\rangle^2$. In a thermal state at temperature T this variance is given by $(\Delta\mathbf{u})^2_{\rm th} = u_0^2(2n_{\rm LO} + 1)$, where $n_{\rm LO} = [\exp(\hbar\omega_{\rm LO}/k_BT) - 1]^{-1}$ is the Bose distribution. The results are most easily interpreted by considering the normalized deviation from the thermal value, $S_u = [(\Delta\mathbf{u})^2 - (\Delta\mathbf{u})^2_{\rm th}]/u_0^2$. In the literature often a reduction below the thermal limit, corresponding to negative values of S_u, is referred to as squeezing. Vacuum squeezing refers to a reduction below the vacuum limit. The evaluation of S_u under nonequilibrium conditions requires the evaluation of the time-dependent values of $\langle b_{\mathbf{q}}\rangle$, $\langle b^\dagger_{\mathbf{q}}b_{\mathbf{q}'}\rangle$, and $\langle b_{\mathbf{q}}b_{\mathbf{q}'}\rangle$. For a QD driven by ultrafast pulses all these quantities can be obtained analytically by using a generating function formalism [16,19].

We start by considering a QD excited by a single pulse arriving at time $t_1 = 0$ with pulse area $A_1 = A$. This case is most instructive due to its simplicity and yields

$$S_u = \theta(t)|I(r)|^2[\cos(\omega_{\rm LO}t) - 1]^2\sin^2(A) \tag{3}$$

with $I(r) = -\frac{i}{\sqrt{N}}\sum_{\mathbf{q}}\frac{\mathbf{q}\cdot\mathbf{r}}{qr}\frac{g^*_{\mathbf{q}}}{\omega_{\rm LO}}e^{i\mathbf{q}\cdot\mathbf{r}}$. Obviously, S_u vanishes if $A = n\pi$, n being an integer. For all other pulse areas S_u is always positive, which implies that the fluctuations never fall below the thermal limit. This also holds for the fluctuations of the momentum. Thus, within our model a single pulse excitation never gives rise to phonon squeezing. Nevertheless, the fluctuations exhibit oscillations at frequencies $\omega_{\rm LO}$ and also at $2\omega_{\rm LO}$. This demonstrates that a $2\omega_{\rm LO}$ modulation is clearly not sufficient to conclude that the system is in a squeezed state.

These results can be understood by looking at the quantum states of the phonons. The electron-phonon coupling in Eq. (1) describes the fact that the equilibrium position of the ions is shifted when the QD is in the excited state. A sudden excitation by a pulse with $A = (2n + 1)\pi$ drives the QD into the upper state transforming the phonon ground state into a coherent state oscillating around the shifted equilibrium position. For a pulse with $A = 2n\pi$ there is no excitation at all and the lattice remains in its ground state. In both cases $S_u = 0$. For all other pulse areas exciton and phonons become entangled. For the phonon system this corresponds to a statistical mixture of the ground state and a coherent state oscillating around a shifted equilibrium. The FLDs of such a mixture are time dependent, but never fall below those of the ground or the coherent state alone [20].

It is interesting to compare these findings with results derived from Raman tensor models, which are widely used to interpret temporal modulations of FLDs [12,14,22,23]. There the electronic degrees of freedom are effectively eliminated by performing a twofold expansion of the optical polarization [24]: First it is linearized with respect to the laser field; second it is expanded with respect to the lattice displacements usually up to second order. The result is an effective Hamiltonian for the light-induced phonon dynamics characterized by a direct instantaneous coupling of the field intensity to linear and quadratic contributions in the phonon operators. For an impulsive excitation by a single ultrafast pulse such Raman tensor models predict the generation of a squeezed phonon state whenever the second order Raman tensor is nonvanishing [22,23]. Likewise, FLDs oscillate with the frequency $2\omega_{\rm LO}$ only when the second order Raman tensor is nonzero. Thus, in clear contrast to our result, within a second order Raman tensor model there can never arise a situation where FLDs are modulated with twice the phonon frequency without simultaneously exhibiting squeezing. On the contrary, in the model defined in Eq. (1) the laser field couples only indirectly to phonons: first an exciton is excited which then couples to the phonons. This indirect coupling leads to memory effects and results in an entanglement between exciton and phonon systems. The neglect of memory and entanglement effects—implicitly made in the derivation of the Raman tensor model—however, becomes questionable in the regime of ultrafast dynamics. Indeed, we find that in this regime our model, where such approximations have not been made, behaves qualitatively different from a corresponding Raman tensor model.

The situation becomes much richer when the QD is excited by a sequence of two ultrashort pulses arriving at times $t_1 = -\tau$ and $t_2 = 0$. Also in this case we obtain explicit analytical expressions for S_u. It turns out that S_u again can be written in the form $S_u = |I(r)|^2 \tilde{S}_u$, where all spatial dependencies are contained in the factor $|I(r)|^2$ defined above. The general expressions for $\tilde{S}_u$ are rather lengthy and will not be given here. Instead we consider the special case of two pulses with equal pulse areas $A_1 = A_2 = \pi/2$ and a delay time $\tau = t_{\rm LO}/2 = \pi/\omega_{\rm LO}$, for which the general expression simplifies considerably. We find at temperature $T = 0$

$$\tilde{S}_u = 4\sin^4\left(\frac{\omega_{\rm LO}t}{2}\right)[1 - B^2\cos^2(\Phi)] + 4\cos^2(\omega_{\rm LO}t) + 8B\sin(\Phi)\sin^2\left(\frac{\omega_{\rm LO}t}{2}\right)\sin(\omega_{\rm LO}t), \quad (4)$$

where $\Phi = \phi_2 - \phi_1 - \tilde{\Omega}\tau$ is the total phase difference between the pulses, $\tilde{\Omega}$ is the polaron-shifted exciton energy, and $B = \exp(-2\sum_{\mathbf{q}}|g_{\mathbf{q}}/\omega_{\rm LO}|^2) < 1$. We note that the sum of the first two terms in Eq. (4) is non-negative and thus there is again no squeezing when the last term vanishes. This is the case for $\Phi = n\pi$. However, as in the single pulse case, the second harmonic is always present.

In contrast to the single pulse case now negative values of $\tilde{S}_u$ are possible. From Eq. (4) we expect the largest negative contributions around $\Phi = (2n + 1)\pi/2$. Figure 1 shows the time dependence of $\tilde{S}_u$ at $\Phi = 0$ and $\Phi = \pi/2$. The inset shows the spectral weights of the first and second harmonics. At $\Phi = 0$ the function $\tilde{S}_u$ indeed oscillates essentially only at twice the phonon frequency and is non-negative for all times indicating the absence of squeezing. In contrast, at $\Phi = \pi/2$ oscillations with both the single and double phonon frequency occur with similar weights. Even more important, now squeezing takes place as can be seen from the negative values of $\tilde{S}_u$ during specific time intervals.

Let us look again at the quantum states created by the pulses. As discussed above, the first pulse creates an entangled equal superposition of $|g\rangle$ with phonon vacuum and $|x\rangle$ with phonons in coherent states. Starting from each of these states the second pulse again creates superpositions between the QD states $|g\rangle$ and $|x\rangle$. This leads to an entangled electron-phonon state where in general both in the QD ground and excited state subspace the phonons are in a superposition of two coherent states with relative phases depending on the delay time τ and phase difference Φ of the pulses. Such superpositions of coherent states are generally called "cat states" and it is known from quantum optics that indeed such cat states may exhibit squeezing [25].

To explore the conditions which are favorable for squeezing we have plotted in Fig. 2(a) the minimum $\tilde{S}_{\rm min}$ of $\tilde{S}_u$ with respect to the real time t as a function of Φ and τ at fixed pulse areas $A_{1,2} = \pi/2$. Indeed, from the full analytical formula it can be deduced that no squeezing is possible if at least one of the pulses has a pulse area of $n\pi$, since in this case no cat states are produced, and that the maximal squeezing is expected for $A_{1,2} = (n + \frac{1}{2})\pi$. Figure 2(a) shows that $S_{\rm min}$ is minimal for $\Phi = (2n + 1)\pi/2$ and at $\tau \approx t_{\rm LO}/2$. Thus, the simplified expression Eq. (4) obtained for $\tau = t_{\rm LO}/2$ corresponds to a parameter range which is highly favorable for squeezing.

So far we have considered values of $g_{\mathbf{q}}$ typical for self-assembled InGaAs-type QDs. To learn more about the influence of the exciton-phonon coupling strength on squeezing we have scaled $g_{\mathbf{q}}$ by a factor f. Stronger couplings appear either in QDs made of stronger polar materials or in structures with a stronger separation between electron and hole wave functions such as QDs in the presence of an electric field [26] or in core-shell QDs [27]. The prefactor $|I(r)|^2$ of S_u scales with f^2 and thus the normalized variance S_u scales as $f^2\tilde{S}_u$. Therefore, in Fig. 2(b) we have plotted $f^2\tilde{S}_{\rm min}$ as a function of τ and f at $A_1 = A_2 = \pi/2$ and $\Phi = \pi/2$. We find that with increasing exciton-phonon coupling the amount of squeezing first increases, then reaches a maximum around $f \approx 20$, and for higher coupling strengths decreases again. The initial increase is related to the fact that for $f = 0$ there is no excitation of phonons at all and thus $S_u = 0$. With increasing coupling phonons are generated and, in agreement with our previous findings, these phonons exhibit squeezing

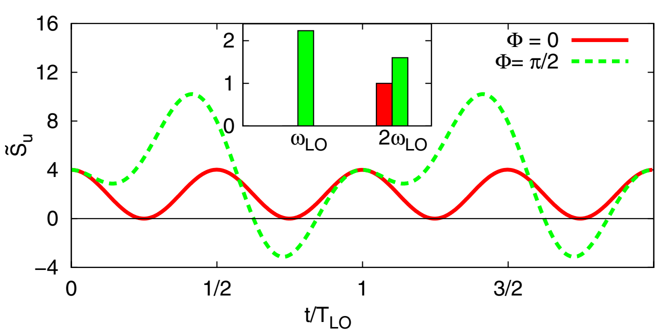

FIG. 1 (color online). Scaled deviation of lattice fluctuations from their thermal value $\tilde{S}_u$ (see text) at $T = 0$ of a QD excited by two pulses with phase difference $\Phi = 0$ or $\Phi = \pi/2$ and delay time $\tau = \pi/\omega_{\rm LO}$. The inset shows the spectral weights of the oscillations with $\omega = \omega_{\rm LO}$ and $\omega = 2\omega_{\rm LO}$.

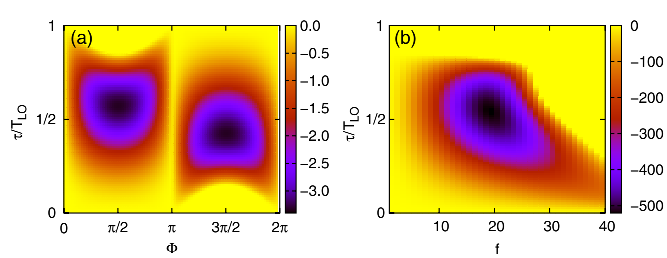

FIG. 2 (color online). (a) Minimum of $\tilde{S}_u$ vs phase Φ and delay time τ at $A_1 = A_2 = \pi/2$; (b) Minimum of $f^2\tilde{S}_u$ vs delay time τ and the strength of the exciton-phonon coupling (i.e., the factor f that scales $g_{\mathbf{q}}$), at $A_{1,2} = \pi/2$ and $\Phi = \pi/2$.

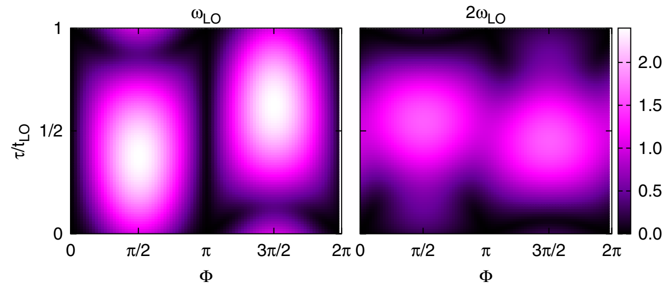

FIG. 3 (color online). Spectral weight of the oscillations at $\omega = \omega_{\mathrm{LO}}$ and $\omega = 2\omega_{\mathrm{LO}}$ in the scaled fluctuations $\tilde{S}_u$ plotted vs phase Φ and delay time τ for pulse areas $A_{1,2} = \pi/2$.

which is most pronounced at delay times $\tau \approx t_{\mathrm{LO}}/2$. The subsequent decrease of the squeezing results from the decrease of the prefactor B entering in the last term of Eq. (4) which is the only term that may give a negative contribution to S_u.

To further elucidate the relation between squeezing and spectral properties of FLDs we have plotted in Fig. 3 the spectral weights of the ω_{LO} and $2\omega_{\mathrm{LO}}$ components of S_u versus Φ and τ at $A_1 = A_2 = \pi/2$. It is seen that the $2\omega_{\mathrm{LO}}$ component is strongest slightly above $\tau = t_{\mathrm{LO}}/2$ for $\Phi = \pi/2$ and slightly below $\tau = t_{\mathrm{LO}}/2$ for $\Phi = 3\pi/2$, which coincides with parameter ranges where the squeezing is strongest [cf. Fig. 2(a)]. In contrast, the single phonon component is largest below $\tau = t_{\mathrm{LO}}/2$ for $\Phi = \pi/2$ and above $\tau = t_{\mathrm{LO}}/2$ for $\Phi = 3\pi/2$. Even though the occurrence of double phonon frequency oscillations in FLDs is not sufficient to indicate squeezing we do find a correlation between these two properties: the squeezing effect is typically strongest when the $2\omega_{\mathrm{LO}}$ oscillations of the FLDs have the largest amplitudes.

Up to now we have considered the case of zero temperature. At finite temperature T the thermal background increases above the vacuum level. Furthermore, in the two-pulse case there is an additional temperature dependence in $\tilde{S}_u$ which, however, at low T is of minor importance. To determine the T range where vacuum squeezing is possible we have estimated the temperature where the increase in thermal fluctuations compensates the squeezing of the generated phonons. We find that for our parameters this occurs at about 20 K. Thus, we expect that below this temperature the FLDs indeed may exhibit vacuum squeezing.

In conclusion, we have analyzed the dynamical and spectral properties of nonequilibrium lattice fluctuations using a model of an optically excited QD coupled to LO phonons. The fluctuations exhibit quite generally oscillations with both the single and double phonon frequency, which is a most prominent feature that is found almost universally in experiments performed on large classes of materials. For an excitation with a single ultrafast pulse the fluctuations never fall below their thermal values demonstrating that there is no strict relation between the appearance of fluctuations with double phonon frequencies and squeezing. This finding is in contrast to calculations using Raman tensor models which suggest that the detection of double phonon frequency fluctuations is clear evidence for squeezing. For two-pulse excitations squeezing is found under specific excitation conditions due to the generation of cat states. The squeezing is indeed strongest for parameters where the fluctuations oscillate strongly with twice the phonon frequency. However, also in this case there is no one-to-one correspondence between squeezing and the occurrence of double phonon frequencies in the lattice fluctuations. Our studies should inspire new research towards conclusive experiments to demonstrate squeezing in a mechanical system like lattice vibrations.

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