Abstract. A theoretical analysis of the fluctuation properties of optical phonons generated by
the optical excitation of a quantum dot is presented. In particular we study whether squeezed
states may be generated where either the fluctuations of the displacement or of the momentum
fall below the vacuum level. The calculations are based on a generating functions formalism
which provides analytical results for the case of excitation by ultrashort optical pulses. While
after a single pulse excitation no squeezing occurs we find that after an excitation with two
phase-locked pulses phonon states can be created where the uncertainties of displacement or
momentum temporarily fall below their vacuum values.

1. Introduction

Squeezed states of light have a long tradition in the field of quantum optics [1] and there are
various applications, e.g., in gravitational wave detection or quantum information processing
[2]. In analogy with photon states in quantum optics the generation and manipulation of
specific phonon states has become a major issue in solid state physics [3]. While in most cases
coherent phonon amplitudes and nonequilibrium phonon occupations have been investigated,
in recent years there has been a growing interest in the study of the fluctuation properties of
the generated phonons. The generation of squeezed phonons by second-order Raman scattering
has been predicted [4] and signatures of phonon squeezing have been observed by pump-probe
spectroscopy in transmission [5] or reflection [6] geometry as well as recently in femtosecond
X-ray diffraction [7].

The impulsive optical excitation of a quantum dot (QD) gives rise to the formation of a
lattice distortion in the region of the dot, i.e., to the creation of a polaron. This distortion is
characterized by a non-vanishing mean value of the lattice displacement. However, it is known
that only under specific excitation conditions the phonon system is in a fully coherent state [8], in
which the fluctuations of the displacement and the momentum are time-independent and agree
with the respective vacuum fluctuations. For other excitation conditions not much is known
about the fluctuation properties of the phonons. In this contribution we will study whether the
fluctuations of either the displacement or the momentum can be reduced below their vacuum
values, i.e., whether squeezed phonon states can be optically generated in a QD system.
2. Theoretical background

We consider a QD in the strong confinement limit excited by circularly polarized light, such that we can restrict ourselves to two electronic states, the ground state and the one-exciton state. Due to the different charge densities in ground and excited state, the exciton is coupled to longitudinal optical (LO) phonons with frequency $\omega_{LO}$ via the Fröhlich coupling matrix element $g_q$ [9]. Excitons can be created and manipulated in the system via ultra short laser pulses applied at times $t_j$ with pulse areas $A_j$ and phases $\phi_j$. For this system analytical formulas for all electron and phonon variables are provided by a generating function formalism [10].

For a phonon system the two conjugate variables are the lattice displacement $q$ and the momentum $\pi$, and their uncertainties are given by

$$\Delta q = \frac{\hbar}{\omega_{LO}}, \quad \Delta \pi = \frac{\hbar}{\omega_{LO}}$$

with $\omega_{LO}$ the LO phonon frequency. In a thermal state at temperature $T$ the uncertainties are given by

$$\Delta q = \frac{\hbar}{\omega_{LO}}, \quad \Delta \pi = \frac{\hbar}{\omega_{LO}}$$

In the case of LO phonons the corresponding operators are calculated from the creation (annihilation) operators $b_q^\dagger$ ($b_q$) for a phonon with wave vector $q$ according to

$$\hat{u}(r,t) = -\frac{i u_0}{\sqrt{N}} \sum_q \frac{q}{q} (b_q e^{iqr} - b_q^\dagger e^{-iqr})$$

$$\hat{\pi}(r,t) = -\frac{\pi_0}{\sqrt{N}} \sum_q \frac{q}{q} (b_q e^{iqr} + b_q^\dagger e^{-iqr})$$

with $u_0 = \sqrt{\hbar/(2M_w\omega_{LO})}$ and $\pi_0 = \sqrt{\hbar M_w \omega_{LO}/2}$; $M_w$ is the reduced mass of the lattice ions and $N$ is the number of unit cells. In a thermal state at temperature $T$ the uncertainties are given by

$$\Delta u_q(t)^2 = \frac{n_{LO} (\Delta \pi_q(t)^2) = \pi_0^2 (2n_{LO} + 1) + 1$$

and $\pi_0^2$ are thus the corresponding vacuum fluctuations. We assume sufficiently low temperatures such that $n_{LO}$ is negligible. It is convenient to define dimensionless fluctuations as $S_u = (\Delta u)^2/u_0^2 - 1$ and $S_\pi = (\Delta \pi)^2/\pi_0^2 - 1$, which vanish in the vacuum state. Negative values of one of these quantities thus correspond to squeezing of the corresponding variable $u$ or $\pi$. The Heisenberg uncertainty relation rewritten in these variables reads $S_u + S_\pi + S_u S_\pi \geq 0$. In our case, where typically $|S_u|, |S_\pi| \ll 1$, this effectively reduces to $S_u + S_\pi \geq 0$.

From the generating function approach we obtain closed but in general lengthy formulas for $\langle b_q \rangle$, $\langle b_q b_q^\dagger \rangle$, and $\langle b_q^\dagger b_q^\dagger \rangle$ [8, 10] and thus for $S_u$ and $S_\pi$. Because of the vanishing dispersion of LO phonons there is no spatial transport and the space dependence of both $S_u$ and $S_\pi$ is given by the time-independent function $|I_0(r)|^2$ with $I_0(r) = \sum_q \frac{q}{q} (g_q^* / \omega_{LO}) e^{iqr}$. In the Figures we therefore plot the space-independent quantities $\bar{S}_u = S_u/|I_0|^2$ and $\bar{S}_\pi = S_\pi/|I_0|^2$.

All calculations have been performed for a spherical GaAs quantum dot with diameter 5 nm.

3. Results

From the general formulas it is easily seen that after a single pulse excitation, although $\bar{S}_u$ and $\bar{S}_\pi$ are in general time-dependent, they never assume negative values. Thus, there is no squeezing in this case. Therefore we will concentrate on the case of excitation by two laser
pulses at $t_1 = -\tau$ and $t_2 = 0$. The fluctuation properties of the phonons generated by such an excitation depend on the pulse areas $A_{1, 2}$, the delay time $\tau$ between the pulses and the phase difference $\Phi = \phi_2 - \phi_1 - \bar{\omega} \tau$, $\bar{\omega}$ being the resonance frequency of the exciton.

Figure 1 shows the time-dependent fluctuations $S_u(t)$ and $S_\pi(t)$ for fixed pulse areas $A_{1, 2} = \pi/2$ and fixed phase difference $\Phi = \pi/2$ at different delay times $\tau = T_{LO}/4$, $T_{LO}/2$ and $3T_{LO}/4$, with $T_{LO} = 2\pi/\omega_{LO}$ being the phonon oscillation period. We find that the fluctuations may become negative in certain time windows, i.e., that now squeezing may occur. The uncertainty relation requires that at any time $S_u + S_\pi \geq 0$, which is indeed fulfilled. The fluctuation of the displacement $\tilde{S}_u$ is periodic with a period $T_{LO}$. For all delay times $\tilde{S}_u$ becomes negative in some time intervals, i.e., all phonon states generated by these pulse sequences are displacement-squeezed. For $\tau = T_{LO}/2$ the most pronounced negative values can be seen, while for $\tau = T_{LO}/4$ and $3T_{LO}/4$ the fluctuation only slightly goes below zero. In contrast, the fluctuation of the momentum $\tilde{S}_\pi$ oscillates with a period $T_{LO}/2$. Here the most negative values appear for $\tau = 3T_{LO}/4$. For $\tau = T_{LO}/2$ the fluctuation still becomes slightly negative, while for $\tau = T_{LO}/4$ it remains positive, thus there is no momentum squeezing in this case.

A more detailed analysis of the conditions for squeezing is shown in Fig. 2. Here we have plotted the minima of the time dependent fluctuations $(\tilde{S}_u(t))_{\text{min}}$ and $(\tilde{S}_\pi(t))_{\text{min}}$ as functions of the delay time $\tau$ and the phase difference $\Phi$. The pulse areas again are kept at $A_{1, 2} = \pi/2$. Blue areas indicate that the fluctuation has a minimum below zero, i.e., the quantity is squeezed. Red areas indicate fluctuations which always exceed the vacuum value. We notice that there is a symmetry when exchanging $\Phi \rightarrow 2\pi - \Phi$ and $\tau \rightarrow T_{LO} - \tau$. The most pronounced squeezing occurs for $\Phi = (2n - 1)\pi/2$. For the displacement the minimum values close to $T_{LO}/2$ are strongly negative, while for the momentum the most negative values appear close to $\tau = 3T_{LO}/4$ for $\Phi = \pi/2$ and $\tau = T_{LO}/4$ for $\Phi = 3\pi/2$. But not all generated states are squeezed; at $\tau = nT_{LO}$ or $\Phi = n\pi$ no squeezing occurs. For delay times around $T_{LO}/4$ and phase differences around $\pi/2$ or delay times around $3T_{LO}/4$ and phase differences around $3\pi/2$ the momentum fluctuations are enhanced compared to the vacuum level while the lattice displacement still exhibits squeezing.

The absolute strength of fluctuations is typically hard to measure in solid state systems and therefore a unique proof of squeezing is difficult. Therefore, the appearance of a Fourier component at a frequency $2\omega_{LO}$ in the spectrum of the fluctuations has been taken as an indication for the presence of a squeezed state [4, 5]. To analyze the spectral properties we have plotted in Fig. 3 the absolute value of the Fourier coefficients $\tilde{S}_u(\omega)$ and $\tilde{S}_\pi(\omega)$ at the
Figure 3. Absolute value of the Fourier coefficients (a) $|\tilde{S}_u(\omega)|$ and (b) $|\tilde{S}_\pi(\omega)|$ at the single ($\omega = \omega_{LO}$) and double ($\omega = 2\omega_{LO}$) phonon frequency as functions of the delay time $\tau$ for fixed pulse areas $A_{1,2} = \pi/2$ and phase difference $\Phi = \pi/2$.

single ($\omega = \omega_{LO}$) and double ($\omega = 2\omega_{LO}$) phonon frequency. Here the delay time $\tau$ has been varied while pulse areas are kept at $A_{1,2} = \pi/2$ and the phase difference is $\Phi = \pi/2$. In the case of $\tilde{S}_u$ the Fourier component at $\omega_{LO}$ is always stronger than the component at $2\omega_{LO}$. If we compare the Fourier coefficient with the minimum value $(\tilde{S}_u)_{\text{min}}$ shown in Fig. 2, we notice that the coefficient $2\omega_{LO}$ roughly follows the shape of the curve of the minima. The largest Fourier coefficient at $2\omega_{LO}$ corresponds to the most negative value of $(\tilde{S}_u)_{\text{min}}$. For the momentum no oscillation with the single phonon frequency $\omega_{LO}$ occurs and its Fourier coefficient is zero for all delay times $\tau$. The behavior of the component at $2\omega_{LO}$ is exactly the same as for $\tilde{S}_u$. When comparing this curve with $(\tilde{S}_\pi)_{\text{min}}$ shown in Fig. 2, however, we find that there is no correspondence between the strength of the Fourier coefficient and the appearance of squeezing.

4. Conclusions

We have studied the conditions for the generation of squeezed LO phonon states in a QD system excited optically with one or two laser pulses. While for single pulse excitation no squeezing occurs, two pulse excitations may lead to squeezing of the displacement and of the momentum. The details strongly depend on the delay time and the relative phase of the two laser pulses. However, we have found that there is no direct relation between the presence of squeezing and the appearance of a Fourier component in the fluctuations at twice the phonon frequency.

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