Stationary Phonon Squeezing by Optical Polaron Excitation

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We demonstrate that a stationary squeezed phonon state can be prepared by a pulsed optical excitation of a semiconductor quantum well. Unlike previously discussed scenarios for generating squeezed phonons, the corresponding uncertainties become stationary after the excitation and do not oscillate in time. The effect is caused by two-phonon correlations within the excited polaron. We demonstrate by quantum kinetic simulations and by a perturbation analysis that the energetically lowest polaron state comprises two-phonon correlations which, after the pulse, result in an uncertainty of the lattice momentum that is continuously lower than in the ground state of the semiconductor. The simulations show the dynamics of the polaron formation process and the resulting time-dependent lattice uncertainties.

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In a squeezed state the uncertainty of one measurable variable is reduced below its zero-point value, while the uncertainty of its conjugate variable is increased. Squeezed states are of interest because they are genuinely quantum mechanical in nature, but also because the reduced variance allows measurements with unprecedented precision. In particular, squeezed light is employed for high-precision interferometric measurements [1–3], which may be used, e.g., for the detection of gravitational waves [2,4,5]. While squeezing was first demonstrated for photons [6], it has become a focus of research also for other bosonic systems, such as magnons, where a reduction of the spin noise below the vacuum level has been realized [7,8].

Recently, squeezing has been reported even for micrometer-scale mechanical resonators [9–11] opening new perspectives for quantum engineering of states of matter at macroscopic length scales and the realization of ultrasensitive sensing of force and motion. Another mechanical realization of squeezing can be found for lattice vibrations in a crystal, i.e., phonons. Here, position and momentum provide a natural choice for the two conjugate variables. With this choice, a squeezed phonon state is a state in which the uncertainties of either the positions or the momenta of the lattice nuclei are reduced to values smaller than those at absolute zero temperature. Phonon squeezing continues to attract much attention both in theory and experiment [3,12–22]. The corresponding uncertainties are measured usually either optically in a pump-probe setup or by ultrafast x-ray diffraction [12,17–19].

The uncertainties of the two conjugate variables in a squeezed state usually oscillate in time; they alternate between being reduced and being inflated compared to their zero-point value. If there is a characteristic phonon frequency, for example, because primarily phonons with a certain energy are excited, the oscillation frequency is twice this value [23]. This double-frequency oscillation has often been interpreted as a clear indication of a squeezed phonon state. Although it has been shown that the oscillation does not necessarily indicate that the uncertainties actually fall below their zero-point level [16], the double-frequency oscillation was found for all preparation schemes for squeezed states discussed so far. For free systems like photons in vacuum it is obvious that squeezing leads to the double-frequency oscillation because it requires two-boson correlations which oscillate with this frequency. As will be shown in this Letter, this does not necessarily hold true for the preparation of a squeezed state in a solid.

We present quantum kinetic simulations of a semiconductor quantum well that is optically excited on its lowest transition line. A special emphasis lies on the dynamics of the lattice uncertainties. In this way, we can monitor how the excitation leads to a new kind of squeezed phonon state in which the uncertainty of the lattice momentum is continuously smaller than in the ground state.

The physics behind this effect is the optical excitation of a polaron whose phonon component is squeezed. Squeezed states and, more generally, states with two-phonon correlations have been employed previously in variational approaches to describe polarons [24–27]. In these calculations the inclusion of two-phonon correlations led to lower energies. Here, we shall show that a stable polaron state comprising the two-phonon correlations required for squeezing can be optically excited. That it is indeed the polaron which contains the squeezed phonons is further substantiated by a perturbation theoretic analysis.

We employ a microscopic model of a GaAs/AlAs quantum well with two electron and three heavy-hole subbands, similar to the one used in an earlier publication [28]. The subbands are calculated within the envelope-function framework for a square potential with a width of 20.34 nm. A sketch of the subband structure is shown in Fig. 1.

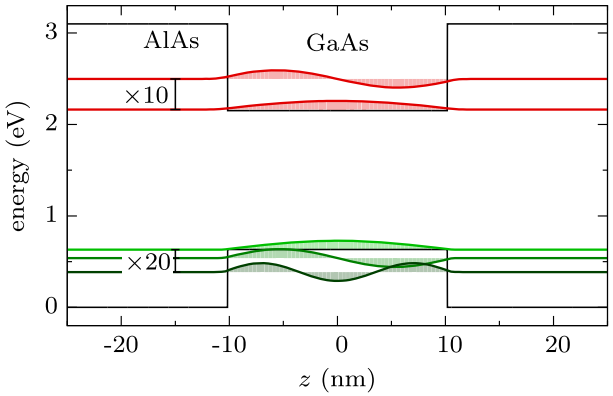

FIG. 1. Electronic structure of the GaAs/AlAs quantum well. Two electron and three heavy-hole subbands are considered. The spacing between the electron and hole subbands is exaggerated by a factor of 10 and 20, respectively. Thus, the well appears shallower than it really is.

The charge carriers are coupled to the lattice via the Fröhlich Hamiltonian [29]

$$H_{\mathrm{Fr}} = \sum_{\substack{i_1 i_2 \\ \boldsymbol{k_1 k_2} q_z}} \left[g_{\boldsymbol{q}}^{e i_1 i_2} c^{\dagger}_{i_1 \boldsymbol{k_2}} c_{i_2 \boldsymbol{k_1}} b_{\boldsymbol{q}} + g_{\boldsymbol{q}}^{e i_1 i_2} c^{\dagger}_{i_2 \boldsymbol{k_1}} c_{i_1 \boldsymbol{k_2}} b^{\dagger}_{\boldsymbol{q}} \right] - \sum_{\substack{j_1 j_2 \\ \boldsymbol{k_1 k_2} q_z}} \left[g_{\boldsymbol{q}}^{h j_2 j_1} d^{\dagger}_{j_1 \boldsymbol{k_2}} d_{j_2 \boldsymbol{k_1}} b_{\boldsymbol{q}} + g_{\boldsymbol{q}}^{h * j_2 j_1} d^{\dagger}_{j_2 \boldsymbol{k_1}} d_{j_1 \boldsymbol{k_2}} b^{\dagger}_{\boldsymbol{q}} \right]. \tag{1}$$

Here, $g^{e/h}$ is the Fröhlich coupling matrix and $c^{(\dagger)}$, $d^{(\dagger)}$, and $b^{(\dagger)}$ are Fock annihilation (creation) operators for electrons, holes, and phonons. $\boldsymbol{k_1}$ and $\boldsymbol{k_2}$ are in-plane wave vectors, $\boldsymbol{q}$ is the three-dimensional phonon wave vector with $\boldsymbol{q} = \boldsymbol{k_2} - \boldsymbol{k_1} + q_z \boldsymbol{e}_z$, and $i_{1,2}, j_{1,2}$ are the subband indices.

We consider only the LO phonon branch, to which electronic excitations couple most strongly; positions and momenta and their uncertainties therefore refer to the relative motion of the lattice nuclei. In principle, the coupling of the electronic subsystem to phonons can be enhanced by the application of an external electric field, which increases the charge separation in excited states. While this allows a more efficient driving of coherent phonons [30], in the present case it proved to lessen the squeezing within the excited polaron.

The model also includes the Coulomb interaction between charge carriers and the electric dipole coupling to the driving laser pulse. In particular, this accounts for excitons visible as discrete lines in the absorption spectrum. The dynamics are calculated with a method that traces the order of all dynamical variables in the strength of the driving field E and the phonon coupling element g, and neglects all contributions above a certain order in E or g. More details about the model and the simulation method can be found in earlier publications [28,30].

In order to get realistic values for the strength of the squeezing, the spatial averaging inherent in any measurement has to be taken into account [28]. Otherwise, the fluctuations of too many phonon modes would contribute. We therefore take a Gaussian average over the relative displacement $U_z(t)$ of the lattice and the corresponding momentum $\Pi_z(t)$. The averaging profile has a standard deviation of $\sigma_{x,y} = 5\ \mu$m in the in-plane directions and $\sigma_z = 20$ nm in growth direction. The extent in the z direction is chosen equal to the width of the quantum well because a larger value would obviously degrade the relative contribution of the well in favor of the barrier, in which no dynamics are excited. Such a small value could experimentally be achieved by a measurement technique that reacts to the well material only, for example, an optical probe pulse that energetically lies between the energy gaps of well and barrier material.

Throughout this Letter, we make use of dimensionless units for lattice displacement and momentum. The factors connecting the dimensionless variables to the real values are $\sqrt{\hbar/(2m\omega)} = 4.0$ pm for the lattice displacement and $\sqrt{\hbar m\omega/2} = 1.3 \times 10^{-23}$ kg m/s for the momentum. Here, ω is the frequency and m is the reduced mass of the LO lattice motion; the numerical values are for GaAs. By definition, the uncertainties of the dimensionless lattice variables are equal to one at absolute zero temperature. The spatial averaging reduces those values to $(\Delta U_z)_0^2 = (\Delta \Pi_z)_0^2 = a^3/(32\pi^{\frac{3}{2}}\sigma_x\sigma_y\sigma_z) = 2.0 \times 10^{-12}$, where a is the lattice constant.

In a squeezed state, one of the uncertainties is reduced below its zero-point value. The strength of the squeezing is quantified by the squeezing factors, which are defined as the change of the uncertainties relative to their zero-point level; for example, the positional squeezing factor is defined as

$$S_U = \frac{(\Delta U_z)^2 - (\Delta U_z)_0^2}{(\Delta U_z)_0^2}. \tag{2}$$

A negative value of either S_U or S_Π signifies squeezing.

In terms of the phonon operators, the squeezing is determined by the number of incoherent phonons $\delta\langle b^{\dagger}_{\boldsymbol{q}} b_{\boldsymbol{q}'}\rangle = \langle b^{\dagger}_{\boldsymbol{q}} b_{\boldsymbol{q}'}\rangle - \langle b^{\dagger}_{\boldsymbol{q}}\rangle\langle b_{\boldsymbol{q}'}\rangle$ and the two-phonon correlations $\delta\langle b_{\boldsymbol{q}} b_{\boldsymbol{q}'}\rangle = \langle b_{\boldsymbol{q}} b_{\boldsymbol{q}'}\rangle - \langle b_{\boldsymbol{q}}\rangle\langle b_{\boldsymbol{q}'}\rangle$. Explicitly, we have

$$S_{U/\Pi} = \frac{1}{(\Delta U_z)_0^2}\frac{1}{N}\sum_{\boldsymbol{q},\boldsymbol{q}'} \frac{q_z q_z'}{q q'} e^{-\frac{1}{2}\sigma_x^2(q_x^2+q_x'^2)-\frac{1}{2}\sigma_y^2(q_y^2+q_y'^2)} \times e^{-\frac{1}{2}\sigma_z^2(q_z^2+q_z'^2)} 2\mathrm{Re}(\delta\langle b^{\dagger}_{\boldsymbol{q}} b_{\boldsymbol{q}'}\rangle \pm \delta\langle b_{\boldsymbol{q}} b_{\boldsymbol{q}'}\rangle), \tag{3}$$

where the upper sign refers to S_U and the lower to S_Π and N denotes the number of primitive unit cells in the system volume.

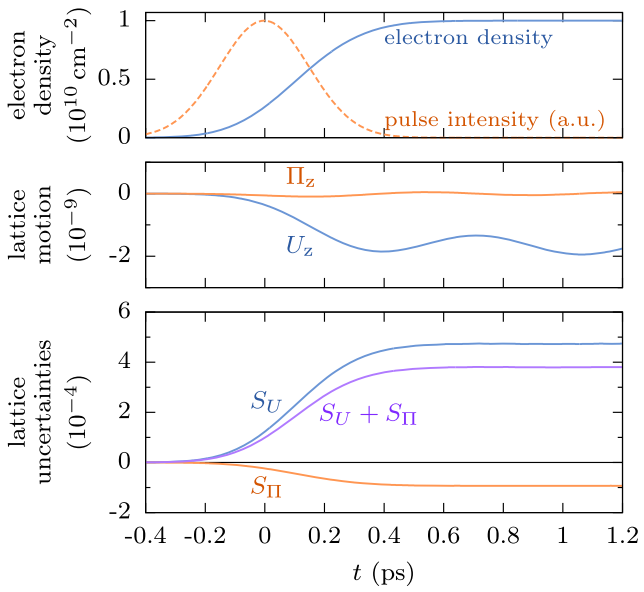

FIG. 2. Formation of the squeezed polaron state. The upper part shows the sheet density of electron-hole pairs, in the center the dimensionless lattice variables are depicted, and the lower part shows the uncertainties relative to their zero-point values. The quantum well is optically excited around $t = 0$ by a Gaussian pulse resonant with the lowest exciton (1.522 eV, full width at half maximum 354 fs).

Because the number of incoherent phonons cannot be negative, a squeezed state depends crucially on the two-phonon correlations. They also are the origin of the double-frequency oscillation of the uncertainties that is so characteristic for many squeezed states. In our case, they arise as part of the polaron. Within this stationary compound state, their free energy oscillation with twice the phonon frequency is suppressed.

In our quantum kinetic simulations we excite the quantum well with an optical pulse with a full width at half maximum of its intensity of 354 fs, which is long compared with the phonon oscillation period of 114 fs. The laser is resonant with the lowest exciton, which for this quantum well lies at 1.522 eV. A short pulse within the band gap has been shown previously to impulsively drive coherent phonons, which yields a more typical squeezed state in which the uncertainties oscillate [28]. In this case, the long excitation time serves to selectively excite the polaron while keeping all other excitations as small as possible. A further increase of the pulse duration therefore leads to very similar results.

Figure 2 shows the dynamics of the quantum well under the excitation conditions specified above and the resulting formation of a polaron. The pulse slowly excites electron-hole pairs up to a sheet density of 10^{10} cm^{-2} (upper plot). As is seen from the central plot, associated with the optical excitation a mean displacement of the lattice ions builds up. Since the pulse duration is much longer than the phonon oscillation period, this buildup occurs almost adiabatically. A small oscillation remains. This is not the free oscillation of the lattice, but driven by a quantum beat between higher electronic transitions that have also been coherently excited and now drag the lattice into a slow oscillation. It disappears when the upper hole subbands are artificially switched off in the calculations (not shown).

The lower part of the figure displays the squeezing factors. During the pulse, the uncertainty of the lattice displacement is increased, whereas the momentum uncertainty clearly falls below its zero-point level. Both uncertainties remain constant after the pulse. As explained above, the uncertainties in a squeezed state would normally oscillate with twice the phonon frequency. In the present case, we have excited a polaron state, which is stable on these time scales; within the polaron state, two-phonon correlations are present and form a squeezed state. In other words, the changes in the electronic subsystem sustain a state of the phonon subsystem in which the lattice momentum is permanently squeezed.

The state is not an ideal squeezed state in the sense that the uncertainty product is larger than the minimum value mandated by the uncertainty principle. For small values of the squeezing factors, the uncertainty principle means that the sum of the two squeezing factors must be equal to or larger than zero. In this particular case, S_U rises much more than S_Π falls because incoherent phonons are created.

The change in the uncertainties is relatively small compared to their zero-point value. In experiments, even smaller changes have been measured [19]. In addition, the perturbative calculations below will show that the squeezing factors are proportional to the square of the electron-phonon coupling g, so a larger effect can be expected in materials in which the coupling is stronger.

In our model the squeezed state does not decay because we assume an infinite lifetime for the polaron. This approximation is justified because even for coherent phonons in GaAs at low temperatures measurements of the decoherence time led to the result $T_2/2 \approx 9$ ps [31], which should be a lower limit for the lifetime of phonons bound in a polaron. In comparison, we simulate a maximum time of 1.6 ps, while the double-frequency oscillation of the uncertainties in a common squeezed state in this system lies at 57 fs.

The quantum kinetic calculations indicate that we have excited a polaron whose phonon component is squeezed. In order to gain insight into the phonon component of the polaron and to exclude the possibility that the creation of the squeezed lattice state and of the polaron are only coincidental, we calculate the polaron state perturbatively in a simplified model.

Because only phonon modes close to the Brillouin zone center noticeably contribute to the uncertainty of the spatially averaged lattice variables, we restrict the Fröhlich Hamiltonian (1) to the phonon mode with wave vector $q = 0$.

Some care must be taken in the limit $q \to 0$: In the continuum limit, the coupling element g_q diverges as q approaches zero, but due to the overall charge neutrality the diverging contributions from electrons and holes cancel each other and a finite value remains. The reduced Hamiltonian reads

$$\tilde{H}_{\mathrm{Fr}} = \sum_{i_1 i_2} \left[g^{e i_1 i_2} c^\dagger_{i_1} c_{i_2} b + g^{e i_1 i_2} c^\dagger_{i_2} c_{i_1} b^\dagger \right] - \sum_{j_1 j_2} \left[g^{h j_2 j_1} d^\dagger_{j_1} d_{j_2} b + g^{h * j_2 j_1} d^\dagger_{j_2} d_{j_1} b^\dagger \right], \quad (4)$$

where we have left out a sum over the in-plane wave vector of the electronic states and all formerly q-dependent variables implicitly refer to the limit $q \to 0$.

As we have seen in Eq. (3), the lattice uncertainties are determined by the number of incoherent phonons $\delta\langle b^\dagger b\rangle = \langle b^\dagger b\rangle - |\langle b\rangle|^2$ and the two-phonon correlations $\delta\langle bb\rangle = \langle bb\rangle - \langle b\rangle^2$, via

$$S_U \propto \mathrm{Re}(\delta\langle b^\dagger b\rangle + \delta\langle bb\rangle), \quad (5a)$$

$$S_\Pi \propto \mathrm{Re}(\delta\langle b^\dagger b\rangle - \delta\langle bb\rangle). \quad (5b)$$

Incoherent phonons increase both uncertainties, while the two-phonon correlations increase either the displacement or the momentum uncertainty and decrease the other one, depending on the sign of their real part. The proportionality factor is the same in both equations.

We calculate these quantities up to second order in the phonon coupling g with the help of time-independent perturbation theory. The unperturbed system is given by

$$\tilde{H}_0 = \sum_i \varepsilon^e_i c^\dagger_i c_i + \sum_j \varepsilon^h_j d^\dagger_j d_j + \hbar\omega b^\dagger b, \quad (6)$$

where the Coulomb interaction has been neglected. The electron-phonon coupling $\tilde{H}_{\mathrm{Fr}}$ acts as the perturbation. We are interested in the first and second order corrections to the lowest pair state $c^\dagger_1 d^\dagger_1 |0\rangle$, which is the state reached by the dipole coupling starting from the crystal ground state.

The first order correction is given by

$$|\Delta\psi^{(1)}\rangle = \sum_{i,j} -\frac{g^{*e1,i} - g^{*hj,1}}{\Delta\varepsilon^e_i + \Delta\varepsilon^h_j + \hbar\omega} |i, j, 1\rangle, \quad (7)$$

with $\Delta\varepsilon^{e/h}_n = \varepsilon^{e/h}_n - \varepsilon^{e/h}_1$ and the eigenstate of the unperturbed Hamiltonian $|i, j, n\rangle = (n!)^{-\frac{1}{2}} c^\dagger_i d^\dagger_j (b^\dagger)^n |0\rangle$. The first order correction contains the coherent phonons, i.e., the shift in the equilibrium position of the lattice, and also allows us to determine the number of incoherent phonons, which is of second order in g:

$$\delta\langle b^\dagger b\rangle = \sum_{(i,j)\neq(1,1)} \frac{|g^{e1,i} - g^{hj,1}|^2}{(\Delta\varepsilon^e_i + \Delta\varepsilon^h_j + \hbar\omega)^2}. \quad (8)$$

In the same way, the second order correction yields the two phonon correlations. We make use of the fact that $g^{e/hn_1n_2} = (g^{e/hn_2n_1})^*$ when the divergent term is removed in the limit $\boldsymbol{q} = q_z \boldsymbol{e}_z, q_z \to 0^+$ and thereby obtain

$$\delta\langle bb\rangle = \sum_{(i,j)\neq(1,1)} \frac{|g^{e1,i} - g^{hj,1}|^2}{\hbar\omega(\Delta\varepsilon^e_i + \Delta\varepsilon^h_j + \hbar\omega)}. \quad (9)$$

As we look at the lowest optical excitation, the electronic energy differences $\Delta\varepsilon^{e/h}_n$ are positive. This means that the two-phonon correlations are larger than the number of incoherent phonons and, consequently, the lattice momentum is squeezed, just as we have seen in the full quantum kinetic calculations. This result also implies that at least one higher electronic subband is required, and indeed the dynamical calculations do not show any squeezing if only one electron and one hole subband are present.

Momentum and position enter symmetrically in the uncertainty relation as well as in the harmonic oscillator Hamiltonian. However, our results reveal a striking asymmetry as it is always the lattice momentum that is squeezed and not the lattice position. The cause is the electron-phonon coupling, which is not symmetric in position and momentum. It connects the effective potential of electrons with the lattice displacement and thus involves only the lattice position operator.

It is also worth noting that the two-phonon correlations and, therefore, also the squeezing effect depend on the combination of resonant and off-resonant terms in the electron-phonon interaction, Eq. (4). Applying the rotating wave approximation (RWA) to this interaction destroys the squeezing effect. The RWA is a standard approximation for modeling systems with, e.g., photons at optical frequencies, but in this case it would prevent the stationary squeezing.

In conclusion, we have shown by quantum kinetic simulations of a semiconductor quantum well that it is possible to prepare a permanently squeezed phonon state by a pulsed optical excitation of the polaron. The squeezing is atypical in the sense that in this case the uncertainties do not exhibit oscillations with twice the phonon frequency. For, e.g., squeezed photons in free space such a permanent reduction of only one uncertainty would not be possible, but in a crystal the electronic component of the polaron stabilizes the change in the lattice uncertainties.

Under quite general circumstances the uncertainty of the lattice momentum is reduced, while the positional uncertainty is increased. This can be understood with the help of a perturbation theory calculation in a simplified model. The perturbative calculation clearly reveals that it is indeed the excitation of a polaron that creates the squeezed phonons.

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