# Optical excitation of squeezed longitudinal optical phonon states in an electrically biased quantum well

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We present quantum kinetic calculations showing how squeezed phonon states can be generated in a biased quantum well by optical driving. In such states the uncertainty of the lattice displacement or momentum is reduced below its zero-point value. It is shown that quantitative results that are meaningful for real observations require accounting for the inevitably limited spatial resolution. Our simulations yield results for the strength of the squeezing and predict under which conditions it can occur: Optical excitations on the lowest absorption line need at least two pulses to generate squeezing, while a single pulse below the band gap may produce a squeezed state which has an almost minimum-value uncertainty product.

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## I. INTRODUCTION

The generation of squeezed photon states is one of the showpieces demonstrating nonclassical behavior of light.[1] In addition, these states allow for measurements with unprecedented precision as needed, e.g., for the detection of gravitational waves.[2] While in quantum optics it is well established how squeezed states can be prepared, the excitation and detection of such states in a mechanical system, i.e., squeezed phonons, is still challenging. Squeezing for a mechanical system means that the uncertainties of either momentum or position reach values below their vacuum level while the product of these uncertainties is of course still above the limit provided by the Heisenberg uncertainty principle. Substantial efforts have been made to realize and measure squeezed states in a crystal lattice. The techniques to detect phonon squeezing can be roughly divided into two groups. First, the uncertainty of the lattice displacement $u$ can be directly determined by measuring a quantity that is proportional to $u^2$. For that purpose a second-order Raman scattering signal has been used in $KTaO_3$ and $SrTiO_3$ crystals[3–5] and, more recently, in ZnTe.[6] Johnson *et al*. have used femtosecond x-ray diffraction in which the intensity of a reflection peak is directly related to $u^2$ by the Debye-Waller factor.[7] Second, the uncertainty can be observed as the variance of an ensemble of measurements. Misochko *et al*. have in this way seen oscillations of the uncertainty of the lattice displacement in a variety of different crystals;[8–10] however, these results are still under debate.[11]

On the theoretical side, second-order Raman scattering[12,13] predicts the generation of squeezed states after excitation with either ultrafast pulses or monochromatic waves. In contrast to the Raman model it has been found within an exactly solvable quantum dot model that a single resonant ultrafast excitation of the dot does not lead to squeezed longitudinal optical (LO) phonons.[14] Furthermore the decay of LO phonons into acoustic phonons, which is analogous to the parametric down conversion process in quantum optics, leads to acoustic phonon squeezing.[15] For quantum dots this squeezing stays localized in striking contrast to the situation in quantum optics.[16]

In this paper we present quantum kinetic simulations of the carrier-phonon dynamics in an optically driven quantum well. We demonstrate two ways in which a squeezed phonon state can be reached: by a two-pulse excitation on the lowest transition line, which exhibits similarities with the behavior of the quantum dot model,[14] and by a single-pulse below-band-gap excitation, which yields a state close to a prototypical squeezed state. We also illuminate the important role of spatial averaging. The microscopic model adopted in this work has been shown to reproduce quantitatively experimental observations devoted to the generation of coherent phonons in quantum wells.[17–20]

## II. OBSERVABLES AND SPATIAL AVERAGING

A quantitative description of lattice uncertainties requires taking into account spatial averaging. Spatial averaging is experimentally unavoidable, but it also is the reason why fluctuations do not completely dominate the signals from lattice dynamics. The measurement techniques, be it an optical pump-probe setup,[3,17] detection of emitted terahertz radiation,[18] or x-ray scattering,[7,21] are not sensitive to the position of individual nuclei, but average over a comparatively large volume of the lattice. This affects the lattice uncertainties because it excludes modes with higher $q$ vectors.

Without averaging, even at zero temperature the zero-point fluctuations of every phonon mode contribute to the lattice displacement uncertainty. In a typical crystal, even relatively strong lattice vibrations have an amplitude smaller than this zero-temperature uncertainty. The same effect is known from the quantized electromagnetic field: Without averaging, it has infinite uncertainties.[22]

We therefore start by defining spatially averaged observables. In an experiment, the region averaged over is defined by, e.g., the spot size of the probe beam. In realistic situations, it is so large that only phonon modes very close the Brillouin zone center contribute to the observables. We assume that the averaged observables are completely determined by the $q = 0$ mode, which reduces the numerical complexity of the problem and allows us to give an explicit equation for the dependence on the size of the averaging volume. However, all other phonon modes still contribute indirectly to the dynamics.

We focus on optical phonons, which in a polar crystal are the quanta of the relative motion of the two oppositely charged

 

sublattices. LO phonons have the most efficient coupling to the electronic subsystem and dominate in optical signals, but to quantify the vacuum uncertainties of the relative lattice motion also the transverse optical (TO) phonon branches need to be taken into account. We denote the phonon operators as $b_{\boldsymbol{q}}^{\alpha}$, where $\boldsymbol{q}$ is the phonon wave vector and the branch index $\alpha$ differentiates between the LO and the two TO branches. The operator of the displacement of the two sublattices in the continuum limit is called $\hat{\boldsymbol{u}}(\boldsymbol{r})$; the corresponding momentum operator is $\hat{\boldsymbol{\pi}}(\boldsymbol{r})$. Assuming a flat dispersion relation and neglecting LO-TO splitting[29] we define those as dimensionless quantities by

$$\hat{\boldsymbol{u}}(\boldsymbol{r}) = \frac{1}{\sqrt{N}} \sum_{\alpha,\boldsymbol{q}} \boldsymbol{e}_{\boldsymbol{q}}^{\alpha} \left( b_{\boldsymbol{q}}^{\alpha\dagger} e^{-i\boldsymbol{q}\boldsymbol{r}} + b_{\boldsymbol{q}}^{\alpha} e^{i\boldsymbol{q}\boldsymbol{r}} \right), \tag{1}$$

$$\hat{\boldsymbol{\pi}}(\boldsymbol{r}) = \frac{i}{\sqrt{N}} \sum_{\alpha,\boldsymbol{q}} \boldsymbol{e}_{\boldsymbol{q}}^{\alpha} \left( b_{\boldsymbol{q}}^{\alpha\dagger} e^{-i\boldsymbol{q}\boldsymbol{r}} - b_{\boldsymbol{q}}^{\alpha} e^{i\boldsymbol{q}\boldsymbol{r}} \right). \tag{2}$$

Here $N$ is the number of unit cells in the system volume and $\boldsymbol{e}_{\boldsymbol{q}}^{\alpha}$ is the mode unit vector; in particular, $\boldsymbol{e}_{\boldsymbol{q}}^{\mathrm{LO}} = \boldsymbol{q}/q$. To get the dimensioned quantities, $\hat{\boldsymbol{u}}$ has to be multiplied with $\sqrt{\hbar/(2m\omega)}$ and $\hat{\boldsymbol{\pi}}$ with $\sqrt{\hbar m\omega/2}$, where $\omega$ is the optical phonon frequency and $m$ is the reduced mass of the lattice ions.[23,24] As we assume homogeneity in the $x$-$y$ plane, the lattice can only be displaced in the $z$ direction. Hence we will focus on $\hat{u}_z$ and $\hat{\pi}_z$.

The spatial averages of $\hat{u}_z(\boldsymbol{r})$ and $\hat{\pi}_z(\boldsymbol{r})$ are introduced as Gaussian-weighted integrals over these quantities with width $\sigma_z$ in growth direction and $\sigma_{\parallel}$ in in-plane directions:

$$\hat{U}_z = \int G_{\sigma_{\parallel}}(x) G_{\sigma_{\parallel}}(y) G_{\sigma_z}(z)\, \hat{u}_z(\boldsymbol{r})\, d^3r \tag{3}$$

with $G_{\sigma}(x) = (\sqrt{2\pi}\sigma)^{-1} \exp[-x^2/(2\sigma^2)]$; the average $\hat{\Pi}_z$ is defined accordingly.

An important effect of the spatial averaging is that it reduces the vacuum uncertainties, i.e., the uncertainties which are present even at zero temperature. Without averaging we have $(\Delta u_z)_0^2 = (\Delta \pi_z)_0^2 = 1$. For GaAs this means a position uncertainty of more than a thousandth of the lattice constant, which is large compared to typical lattice displacements.[25] The averages have a vacuum uncertainty of $(\Delta U_z)_0^2 = (\Delta \Pi_z)_0^2 = a^3/(32\sqrt{\pi}^3 \sigma_{\parallel}^2 \sigma_z)$, where $a$ is the lattice constant. In an experiment the averaging will in general extend over many unit cells, and this value will be much smaller than one.

The uncertainties in an arbitrary state of the system are determined by the phonon correlations

$$\delta\langle b_{\boldsymbol{q}}^{\dagger} b_{\boldsymbol{q}'} \rangle = \langle b_{\boldsymbol{q}}^{\dagger} b_{\boldsymbol{q}'} \rangle - \langle b_{\boldsymbol{q}}^{\dagger} \rangle \langle b_{\boldsymbol{q}'} \rangle, \tag{4}$$

$$\delta\langle b_{\boldsymbol{q}} b_{\boldsymbol{q}'} \rangle = \langle b_{\boldsymbol{q}} b_{\boldsymbol{q}'} \rangle - \langle b_{\boldsymbol{q}} \rangle \langle b_{\boldsymbol{q}'} \rangle. \tag{5}$$

From here on the branch index $\alpha$ is left out and all phonon operators implicitly refer to the LO branch. In our model the coupling to TO phonons is neglected, and therefore the transverse branches contribute to the vacuum uncertainties only. Explicitly, we have

$$(\Delta U_z)^2 = (\Delta U_z)_0^2 + \frac{1}{N} \sum_{\boldsymbol{q},\boldsymbol{q}'} \frac{q_z}{q} \frac{q_z'}{q'} e^{-(1/2)\sigma_{\parallel}^2 (q_x^2 + q_y^2 + q_x'^2 + q_y'^2)} \times e^{-(1/2)\sigma_z^2 (q_z^2 + q_z'^2)} \cdot 2\mathrm{Re}(\delta\langle b_{\boldsymbol{q}}^{\dagger} b_{\boldsymbol{q}'} \rangle + \delta\langle b_{\boldsymbol{q}} b_{\boldsymbol{q}'} \rangle). \tag{6}$$

For $(\Delta \Pi_z)^2$ the equation is almost the same with only the rightmost plus sign changed into a minus. We define $S_U$ and $S_{\Pi}$ as the relative change in the lattice uncertainties, e.g., $S_U = (\Delta U_z)^2/(\Delta U_z)_0^2 - 1$. Negative values mean that the uncertainty is smaller than the vacuum uncertainty and thus signify squeezing.

In reciprocal space the spatial averaging means that only small $q$ vectors contribute. For sufficiently large $\sigma_{\parallel}$ and $\sigma_z$ we can then approximate the phonon correlations by their values at $q, q' = 0$. With the appropriately defined limits

$$\delta n_0 = \lim_{q_z, q_z' \to 0} \mathrm{sgn}(q_z)\mathrm{sgn}(q_z')\, \delta\langle b_{q_z \boldsymbol{e}_z}^{\dagger} b_{q_z' \boldsymbol{e}_z} \rangle, \tag{7}$$

$$\delta b_0 = \lim_{q_z, q_z' \to 0} \mathrm{sgn}(q_z)\mathrm{sgn}(q_z')\, \delta\langle b_{q_z \boldsymbol{e}_z} b_{q_z' \boldsymbol{e}_z} \rangle, \tag{8}$$

where $\boldsymbol{e}_z$ is the unit vector in $z$ direction, and taking into account the correct asymptotic behavior of the phonon correlations, we then arrive at

$$S_{U,\Pi} = \frac{a}{\sigma_z} F\left(\frac{\sigma_{\parallel}}{\sigma_z}\right) \left(\frac{N}{4}\right)^{1/3} \cdot 2\mathrm{Re}\,(\delta n_0 \pm \delta b_0) \quad \text{with}$$
$$F(r) = \frac{r^2 \left[ r^2 - 1 + \frac{\pi}{2} - \frac{2r^2}{\sqrt{2r^2-1}} \arctan(\sqrt{2r^2-1}) \right]}{\pi^{1/2} (r^2-1)^2}. \tag{9}$$

$F(r)$ is a monotonic function with upper limit $\pi^{-1/2}$. Hence the modulation of the lattice uncertainties is strongest if $\sigma_z$ is small and $\sigma_{\parallel}$ is large. Averaging over only a small extent in $z$ direction could be achieved by using a measurement that is sensitive only to the well material, but not to the barrier. $\sigma_z$ still has to be larger than the width of the quantum well as otherwise the approximation of including only terms with $q_z \approx 0$ becomes invalid.

In what follows we will leave out the prefactor $aF/\sigma_z$ that depends only on the extent of the averaging and call the remaining quantities $\tilde{S}_{U,\Pi}$. As an example, with the GaAs lattice constant $a = 0.565$ nm, $\sigma_z = 20$ nm, and $\sigma_{\parallel} = 5\ \mu$m, this prefactor has a value of 0.016. In the same way we define the extent-independent quantities $\tilde{U}_z$ and $\tilde{\Pi}_z$ by excluding the prefactor $(8\pi)^{-1/2} a/\sigma_z$, which for the same parameters is $5.6 \times 10^{-3}$.

## III. QUANTUM KINETIC CALCULATIONS

We make use of a microscopic model of a GaAs/AlAs quantum well with a width of 11.3 nm under an electric bias field in growth direction of 170 kV/cm. The electric field leads to a charge separation in excited states and thereby increases the electron-phonon coupling. The envelope function description is used to calculate subband energies and wave functions. We are concerned with optical excitation on the lowest exciton line and below and therefore limit the model to the lowest electron and heavy hole subband. LO phonons are coupled via

Fröhlich interaction to the electronic subsystem. Anharmonic contributions to the lattice potential can be neglected on the time scales considered here.[26] Coulomb interaction between the carriers accounts for excitonic effects. The light field is treated classically and is coupled to the electronic subsystem in the usual dipole and rotating wave approximation. Further details of the model can be found in Refs. 27 and 19, where almost the same model has been used to study coherent phonon generation.

The dynamics are calculated using a variant of the density matrix theory. The infinite hierarchy of equations of motion is truncated in this way: The dynamical variables are expanded in orders of the phonon coupling $g$ and the laser field strength $E$, and for each order the equations of motion are set up separately; subsequently, all contributions to the variables exceeding a certain order in $E$ or $g$ are neglected. If the initial state of the system is a pure state, this is equivalent to directly solving the Schrödinger equation while keeping contributions to the state vector strictly separated by their order in $E$ and $g$. The relevant density matrices can then be derived from the state vector, again neglecting all contributions above a certain order.[30]

In this work, the initial state always is the ground state, and the latter approach has been used because its numerical complexity is smaller. We have calculated the uncertainties $S_{U,\Pi}$ up to an order of $E^2\,g^4$. Because the next higher orders vanish, the lowest order corrections not included are of order $E^4$ or $g^6$. For resonant driving conditions as discussed in Ref. 19, the order $g^6$ might still be important; however, this case is of no interest for the purpose of generating squeezed states because the generation of incoherent phonons by carrier relaxation is also resonant and inflates the lattice uncertainties. For the conditions considered here, it turns out that even calculations of maximal order $g^2$ are sufficient, as will be shown below. In this paper moderately low excitation densities are used where contributions of orders beyond $E^2$ are not important. We have verified this by performing standard correlation expansion calculations of the electronic dynamics, in which higher orders in $E$ are accounted for.

## IV. SIMULATION RESULTS

With a single-pulse excitation on the lowest exciton line or above our simulations reveal that no squeezed state is created. A similar result has been found for a microscopic model of a quantum dot, where it has been shown analytically that a single excitation with an ultrashort laser pulse resonant with the lowest transition line cannot produce a squeezed phonon state.[14] Nevertheless, we will see that in both cases a two-pulse excitation can produce a squeezed state, and that the conditions necessary are quite similar. These general similarities occur despite the obvious differences between the two systems in their electronic structure, which also affect the phonon coupling: In a quantum dot in the strong confinement limit, phonon effects are limited to pure dephasing, while in a quantum well there are many more electronic states, allowing real phonon absorption and emission processes to take place.

Figure 1 shows the minimum value of the uncertainties $\tilde{S}_U$ and $\tilde{S}_\Pi$ after excitation with two equally strong pulses resonant on the lowest exciton line. The laser field envelope of each

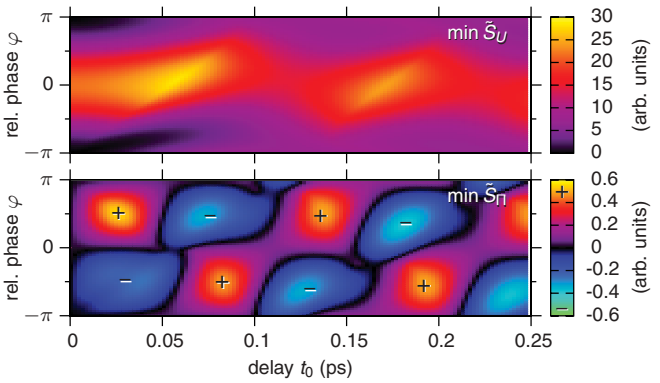


FIG. 1. (Color online) Minimum values of $\tilde{S}_U(t)$ and $\tilde{S}_\Pi(t)$ in the real-time interval 0.4 . . . 0.6 ps for an excitation with two equally strong laser pulses resonant with the lowest exciton line and a FWHM of 70 fs. The plot shows the dependence on the delay time and on the relative phase between the two pulses.

pulse is a Gaussian with a full width at half maximum (FWHM) of 70 fs. The horizontal and vertical axes, respectively, give the delay time $t_0$ and the relative phase $\varphi = \varphi_2 - \varphi_1 - \omega_0 t_0$, where $\omega_0$ is the central frequency of the laser and $\varphi_{1,2}$ are the phases of the pulses. For certain values of $t_0$ and $\varphi$ the lattice momentum uncertainty $\tilde{S}_\Pi$ is clearly squeezed. We observe a checkered area of regions with reduced and increased uncertainty, with the delay time dependence being approximately periodic with the optical phonon oscillation period of 114 fs. This behavior is very similar to the quantum dot case.[14,28] However, in contrast to that system, the lattice displacement here is never squeezed and $\tilde{S}_U$ is generally much larger than $\tilde{S}_\Pi$.

A situation in which momentum squeezing is particularly strong with $t_0 = 126$ fs and $\varphi = -0.61\pi$ is further detailed in Fig. 2. The upper panel shows the density of electron-hole pairs generated by the laser pulses. The pulse intensities have been adjusted so that a density of $10^{10}$ cm$^{-2}$ is reached. The new

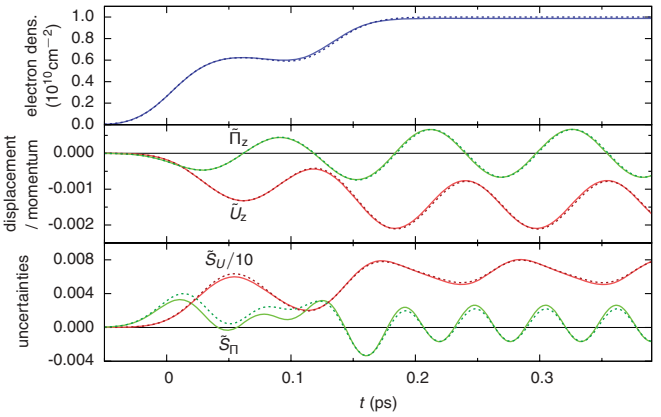


FIG. 2. (Color online) Effects of two 70-fs pulses resonant with the lowest exciton line centered at $t = 0$ and $t = 126$ fs with a relative phase of $\varphi = -0.61\pi$. The plot shows the electron density (upper panel), the mean value of lattice displacement and momentum (center panel), and their uncertainties (lower panel). The dashed lines are from a calculation of higher maximal order ($g^4$), where the phase has been adjusted to $\varphi' = -0.82\pi$ in order to account for energy renormalization due to phonons.

charge distribution after the excitation leads to a shift of the lattice equilibrium position. This induces a coherent oscillation of the lattice displacement around the new equilibrium (center panel). In the lower panel we see that a squeezed state has been created, as $\tilde{S}_\Pi$ acquires negative values. After the second pulse, it oscillates with twice the phonon frequency around a value close to zero. $\tilde{S}_U$ is much larger and has been scaled down by a factor of 10 to make it fit better into the same plot. The lattice displacement uncertainty is strongly increased and its period of oscillation corresponds essentially to the single phonon frequency, although a Fourier transformation reveals a double-frequency component, too.

The plot shows calculations with maximal order of both $g^2$ (solid lines) and $g^4$ (dashed lines). For the higher order calculation there is a small shift of the exciton line due to the phonon interaction, which has been accounted for by readjusting the relative phase to $\varphi' = -0.82\pi$ for this calculation. The two calculations are in good agreement and hence the chosen maximal order is confirmed to be sufficient.

There is quite a difference between the uncertainties of lattice displacement and momentum, which might come as a surprise, considering their apparent symmetry in the theory and the fact that in the quantum dot case their behavior is completely analogical. But there is one respect in which lattice displacement and momentum are different: The displacement suffers a shift of its equilibrium position, whereas the momentum always oscillates around zero. In the quantum dot model, this is also true, but there the shift happened instantaneously. This means that the differences should go away if there is no residual electron-hole population after the pulse which shifts the lattice equilibrium position.

This indeed happens, and it is why an off-resonant excitation with a single laser pulse might be even better suited for an experimental demonstration of a squeezed phonon state. Figure 3 shows the effects of a 50-fs below-band-gap excitation. The optical pulse has a central energy of 1.410 eV, whereas the lowest exciton line lies at 1.487 eV. Its strength has been chosen so that a peak density of $10^{10}$ cm$^{-2}$ is reached; under off-resonant conditions this corresponds to a rather high laser intensity. The generated electron density is transient and

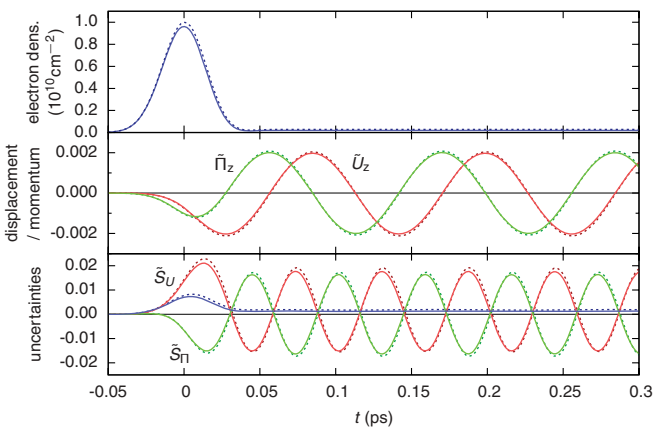


FIG. 3. (Color online) Generation of a squeezed state by a 50-fs below-band-gap excitation. The unlabeled line in the lower panel is the sum $\tilde{S}_U + \tilde{S}_\Pi$. Dashed lines are from a calculation of maximal order $g^4$.

after the pulse only a small fraction remains (upper panel). As a side effect, this could reduce stray influences in experiments because a measurement of the phononic variables often also is sensitive to changes in the electronic state. The fast change in the carrier density exerts a force on the lattice. This creates an oscillation of the mean lattice displacement with the LO phonon frequency (center panel).

The lower panel shows that the state is squeezed: $\tilde{S}_U$ and $\tilde{S}_\Pi$ alternatingly fall below zero with a frequency of twice the phonon frequency. The Heisenberg uncertainty principle, which for our variables reads $(S_U + 1)(S_\Pi + 1) \geqslant 1$, can for small values of $S_U$ and $S_\Pi$ be reduced to $\tilde{S}_U + \tilde{S}_\Pi \geqslant 0$. This sum is also plotted and indicates how much the uncertainty relation is overfulfilled. After the pulse it is small compared to the oscillation amplitude of $\tilde{S}_U$ and $\tilde{S}_\Pi$, which means that a state very close to a prototypical squeezed state has been created.[1] In contrast, in the resonant two-pulse excitation discussed above, the uncertainty $\tilde{S}_U$ strongly dominates and the sum of uncertainties is almost equal to this value. Additionally, the squeezing effect now is larger by an order of magnitude and affects both lattice displacement and momentum. For the exemplary prefactor given above the oscillation amplitude of $S_U$ and $S_\Pi$ is about $2.6 \times 10^{-4}$; for comparison, in the experiment of Ref. 3 on a bulk $KTaO_3$ crystal a value of roughly $10^{-5}$ has been reported. Obviously, the reduction of the uncertainties relative to the standard quantum limit is rather small. Stronger squeezing could be achieved by more intense optical excitation, limited of course by phase-space filling, or by using a different well material such as ZnSe, in which electron-phonon coupling is stronger.

## V. CONCLUSIONS

We have shown simulations of the generation of squeezed phonon states in quantum wells, focusing on the experimentally relevant quantities, i.e., the spatially averaged lattice variables. Our calculations pave the way towards quantitative comparisons with real experiments by providing absolute values for the strength of the squeezing, which depends on the extent of the averaging. A single-pulse excitation resonant on the lowest exciton line leads to a steplike time evolution of the electronic density and does not produce squeezed states, but certain two-pulse excitations yield squeezed lattice momenta. A single pulse below the lowest exciton line generates a transient electron density that impulsively exerts a force on the lattice. This results in squeezing of both the lattice displacement and the momentum. The latter method is particularly promising for two reasons: First, the state generated is very close to an ideal squeezed state in that its uncertainty product is close to its minimum value; and second, the electron density after the pulse is small, which could otherwise interfere with the measurement of the lattice variables.

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[29]The only effect of including LO-TO splitting would be a correction to the absolute value of the vacuum uncertainties that for GaAs parameters would be very small.
[30]This truncation scheme ensures that all integrals of motion are exactly conserved, which in this special case is important because it allows certain divergent terms in the equations of motion to cancel each other. Correlation expansion approximately includes contributions of arbitrarily high order; in many cases this partial summation is an advantage, but here it also disrupts the cancellations.