Phonon Squeezed States Generated by Second-Order Raman Scattering

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(Received 28 August 1997)

We study squeezed states of phonons, which allow a reduction in the quantum fluctuations of the atomic displacements to below the zero-point quantum noise level of coherent phonon states. We investigate the generation of squeezed phonon states using a second-order Raman scattering process. We calculate the expectation values and fluctuations of both the atomic displacement and the lattice amplitude operators, as well as the effects of the phonon squeezed states on macroscopically measurable quantities, such as changes in the dielectric constant. These results are compared with recent experiments. [S0031-9007(97)04745-5]

PACS numbers: 63.20.–e, 42.50.Dv, 42.50.Lc, 42.65.Dr

Nonclassical photon states such as various forms of squeezed states have attracted much attention during the past decade [1]. These novel states are attractive because they have new statistical and quantum mechanical properties. For instance, some of these states can achieve lower quantum noise than the zero-point fluctuations of the vacuum or coherent states. Thus they provide a way of manipulating quantum fluctuations and have a promising future in different applications ranging from optical communications to gravitational wave detection [1]. In recent years, squeezed states are also being explored in a variety of non-quantum-optics systems, including ion-motion and classical squeezing [2], molecular vibrations [3], polaritons [4,5], and phonons in crystals [6–8]. References [7,8] propose a second-order Raman scattering (SORS) process for phonon squeezing: if the two incident light beams are in coherent states, the phonons generated by the SORS are in a two-mode squeezed state. Here we consider both the continuous wave case studied in [7] and the impulsive case studied in [9]. The experimental realization of squeezed phonons [9] via a SORS process has brought attention to the subject of squeezed phonon states [10].

Regarding detection methods, Refs. [7,8] proposed that if the first-order Raman scattering is either very weak or prohibited, the second-order stimulated Raman scattering process can be used to generate two-mode phonon quadrature squeezed states. Moreover, squeezed phonons could be detected by measuring the intensity of the reflected probe light [7,8]. This method has been used to detect phonon amplitudes, since the reflectivity is closely related to the atomic displacements in a crystal. The same argument applies for the transmittivity. Measuring a transmitted probe light pulse, Ref. [9] observed squeezed phonons produced by an impulsive SORS. The intensity of the cw SORS signal for many materials might be too weak to be detected with current techniques, but might be accessible in the future.

The SORS process originates from the quadratic term in the polarizability change $\delta P_{\alpha\beta}$. The photon-phonon interaction V that leads to the SORS process is [11] $V = -\frac{1}{4}\sum_{\alpha\beta}\sum_{\mathbf{q}}^{N}\sum_{jj'} P_{\alpha\beta}^{\mathbf{q}j,-\mathbf{q}j'} Q_{\mathbf{q}j} Q_{-\mathbf{q}j'} E_{1\alpha} E_{2\beta}$. Here, $E_{1\alpha}$ and $E_{2\beta}$ are electric field amplitudes along α and β directions with frequencies ω_1 and ω_2. The second-order polarizability tensor $P_{\alpha\beta}^{\mathbf{q}j,-\mathbf{q}j'}$ satisfies $P_{\alpha\beta}^{\mathbf{q}j,-\mathbf{q}j'} = P_{\alpha\beta}^{-\mathbf{q}j',\mathbf{q}j} = P_{\alpha\beta}^{-\mathbf{q}j,\mathbf{q}j'}$. Recall that the complex normal mode operator $Q_{\mathbf{q}j}$ of the phonons is related to the phonon creation $b_{-\mathbf{q}j}^{\dagger}$ and annihilation $b_{\mathbf{q}j}$ operators by $Q_{\mathbf{q}j} = b_{\mathbf{q}j} + b_{-\mathbf{q}j}^{\dagger}$. If the incident photon fields are not attenuated we can treat the optical fields as classical waves, and also consider the different pairs of $\pm\mathbf{q}$ modes as independent, and treat them separately. Thus, for one particular pair of $\pm\mathbf{q}$ modes, the complete Hamiltonian for the two phonon modes involved in the SORS process has the form [11] $\mathcal{H}_{\mathbf{q}} = H_{\mathbf{q}} - \{4^{-1}\sum_{\alpha\beta} P_{\alpha\beta}^{\mathbf{q},-\mathbf{q}} E_{1\alpha} E_{2\beta}\} Q_{\mathbf{q}} Q_{-\mathbf{q}}$, where $H_{\mathbf{q}} = \hbar\omega_{\mathbf{q}}\{b_{\mathbf{q}}^{\dagger} b_{\mathbf{q}} + b_{-\mathbf{q}}^{\dagger} b_{-\mathbf{q}}\}$ is the free phonon Hamiltonian for the modes $\mathbf{q}$ and $-\mathbf{q}$, $\omega_{\mathbf{q}} = (\omega_1 - \omega_2)/2$, and the branch labels j and j' have been dropped.

Here we consider two different cases. The first is when the incident photons are in two monochromatic beams [7]; i.e., with electric fields $E_j = \mathcal{E}_j \cos(\omega_j t + \phi_j)$; $j = 1, 2$. In the second case the incident photons are in an ultrashort pulse whose duration is much shorter than the phonon period [9].

Squeezed phonons via continuous wave SORS.—Let us now first consider the continuous wave (cw) case. Because the photons are monochromatic, we can take a rotating wave approximation [12] and keep only the on-resonance terms in the Hamiltonian. The off-resonance terms contribute only to virtual processes [13] at higher orders. This approximation is appropriate for times much longer than the phonon period. The simplified Hamiltonian has the form

$$\mathcal{H}_{\mathbf{q}}^{(\mathrm{cw})} = H_{\mathbf{q}} - \lambda_{\mathbf{q}}\{b_{\mathbf{q}} b_{-\mathbf{q}}\, e^{2i\omega_{\mathbf{q}}t + i\phi_{12}} + \mathrm{c.c.}\},$$
$$\lambda_{\mathbf{q}} = \frac{1}{16}\left|\sum_{\alpha\beta} P_{\alpha\beta}^{\mathbf{q},-\mathbf{q}} \mathcal{E}_{1\alpha}\mathcal{E}_{2\beta}\right|, \qquad (1)$$

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where ϕ_{12} and $\lambda_{\mathbf{q}}$ refer to the overall phase and amplitude, respectively, of the product of the second-order polarizability and the incident electric fields. Recall that $P_{\alpha\beta}^{\mathbf{q},-\mathbf{q}}$ is real; therefore the phase ϕ_{12} has no $\mathbf{q}$ dependence. It originates solely from the two photon modes. The Schrödinger equation for the $\pm\mathbf{q}$-mode phonons is $i\hbar\partial_t|\psi_{\mathbf{q}}(t)\rangle = \mathcal{H}_{\mathbf{q}}^{(\mathrm{cw})}(t)\,|\psi_{\mathbf{q}}(t)\rangle$, and its time-evolution operator can be solved by a transformation into the interaction picture. The result can be expressed as [7]

$$|\psi_{\mathbf{q}}(t)\rangle = e^{\{H_{\mathbf{q}}t/i\hbar\}}\, e^{\{\zeta_{\mathbf{q}}^* b_{\mathbf{q}} b_{-\mathbf{q}} - \zeta_{\mathbf{q}} b_{\mathbf{q}}^\dagger b_{-\mathbf{q}}^\dagger\}}|\psi_{\mathbf{q}}(0)\rangle, \quad (2)$$

where $\zeta_{\mathbf{q}} = -i\lambda_{\mathbf{q}}\, t\, e^{-i\phi_{12}}/\hbar$. Notice that the second factor in the time-evolution operator is a two-mode quadrature squeezing operator [14].

In the cw case considered here, the amplitude of the squeezing factor $\zeta_{\mathbf{q}}$ grows linearly with time. However, this initial linear growth will be eventually curbed by subsequent phonon-phonon scattering and optical pump depletion. In other words, the expression for the squeezing factor $\zeta_{\mathbf{q}}$ is valid for times much larger than one phonon period, but much smaller than phonon lifetimes (because this treatment considers nondecaying phonons). Indeed, if this growth rate is not fast enough compared to the phonon decay rate, the squeezing effect may never reveal itself in an experiment. In addition, the phase of the squeezing factor is determined by the phase difference of the two incoming light waves. If the $\pm\mathbf{q}$ phonon modes are initially in a vacuum state or in a coherent state, the SORS will drive them into a two-mode quadrature squeezed state [7].

The time evolution operator of *all* the phonon mode pairs (instead of just one pair of $\pm\mathbf{q}$ modes) that are involved in this SORS process has the form $U(t) = \prod_{\mathbf{q}} U_{\mathbf{q}}(t)$. Therefore, as long as the photon depletion is negligible, all the phonon modes that are involved in a SORS process are driven into two-mode quadrature squeezed states. In other words, squeezing can be achieved in a continuum of phonon modes by a cw stimulated SORS process.

Squeezed phonons via impulsive SORS.—Recently, an impulsive SORS process has been used to experimentally generate phonon squeezing [9]. Here we treat the problem expressing the time evolution operator of the system in terms of a product of the two-mode quadrature squeezing operator and the free rotation operators [15]. Since the incident photons are now in an ultrashort pulse, the complete Hamiltonian can be solved exactly in the limit when the optical field can be represented by a δ function. Such an approximation is usually considered when the optical pulse duration is much shorter than the optical phonon period, which is experimentally feasible with femtosecond laser pulses. The Hamiltonian for the SORS can now be written as $\mathcal{H}' = \sum_{\mathbf{q}}\{H_{\mathbf{q}} - \lambda'_{\mathbf{q}}\delta(t)Q_{\mathbf{q}}Q_{-\mathbf{q}}\}$, where $\lambda'_{\mathbf{q}}$ carries the information on the amplitudes of the incoming optical fields and the electronic polarizability. Notice that the light-phonon coupling strength $\lambda_{\mathbf{q}}$ in the cw case has units of energy, while $\lambda'_{\mathbf{q}}$ here has units of $\hbar$. To further simplify the problem, we assume that only $\pm\mathbf{q}$ modes are involved in the process. Such a simplification is possible when the photon depletion and the phonon anharmonic interaction are negligible, so that different pairs of phonon modes are independent from each other. The Hamiltonian is now

$$\mathcal{H}'_{\mathbf{q}} = H_{\mathbf{q}} - \lambda'_{\mathbf{q}}\delta(t)Q_{\mathbf{q}}Q_{-\mathbf{q}}, \quad (3)$$

and the Schrödinger equation for these two phonon modes is $i\hbar\partial_t|\psi_{\mathbf{q}}(t)\rangle = \mathcal{H}'_{\mathbf{q}}|\psi_{\mathbf{q}}(t)\rangle$. This equation can be solved by separating the free oscillator terms and the two-phonon creation and annihilation terms. The resulting time-dependent wave function is

$$|\psi_{\mathbf{q}}(t)\rangle = \exp\left\{\frac{tH_{\mathbf{q}}}{i\hbar}\right\}\exp\left\{\frac{i\lambda'_{\mathbf{q}}H_{\mathbf{q}}}{\hbar^2\omega_{\mathbf{q}}}\right\} \times \exp\{\zeta'^*_{\mathbf{q}}b_{\mathbf{q}}b_{-\mathbf{q}} - \zeta'_{\mathbf{q}}b_{\mathbf{q}}^\dagger b_{-\mathbf{q}}^\dagger\}\,|\psi_{\mathbf{q}}(0^-)\rangle. \quad (4)$$

Here $\zeta'_{\mathbf{q}} = -i\lambda'_{\mathbf{q}}\, e^{-i\lambda'_{\mathbf{q}}/\hbar}/\hbar$. Hence the effect of the optical pulse is clear: it first applies a two-mode quadrature squeezing operator on the initial state, then rotates the state by changing its phase [15]. The state will then freely evolve after $t = 0^+$. This result is consistent with Ref. [9] where the time-evolution operator is expressed in terms of real phonon normal mode operators [11], instead of the complex ones used in this paper. Notice that, in contrast to the cw SORS, the phase of the squeezing factor ζ' for the impulsive case is fixed by the intensity of the light pulse.

Macroscopic implications.—Now that we have obtained the phonon states for both the cw and pulsed cases, let us consider the macroscopic implications of these states. An experimentally observable quantity O which is related to the atomic displacements in the crystal can generally be expressed in terms of $Q_{\mathbf{q}}$: $O = O(0) + \sum_{\mathbf{q}}(\partial O/\partial Q_{\mathbf{q}})Q_{\mathbf{q}} + \cdots = O_0 + O_1 + O_2 + \cdots$, where the first term $O_0 = O(0)$ is the operator O when all $Q_{\mathbf{q}}$'s vanish. An example of an experimentally observable quantity O is the change in the crystal dielectric constant $\delta\epsilon$ due to the atomic displacement produced by the incident electric fields. To first order in $Q_{\mathbf{q}}$, $\delta\epsilon = \delta\epsilon_1 = \sum_{q_x>0}|\frac{\partial(\delta\epsilon)}{\partial Q_{\mathbf{q}}}|\sqrt{\hbar/2\omega_{\mathbf{q}}}[(b_{\mathbf{q}} + b_{-\mathbf{q}}^\dagger)e^{i\Psi_{\mathbf{q}}} + (b_{-\mathbf{q}} + b_{\mathbf{q}}^\dagger)e^{-i\Psi_{\mathbf{q}}}]$. Here $\Psi_{\mathbf{q}}$ is the phase of $\partial O/\partial Q_{\mathbf{q}} = \partial(\delta\epsilon)/\partial Q_{\mathbf{q}}$. Indeed, a widely used method to track the phases of coherent phonons in the time domain [16] is based on the observation of the reflectivity (or transmission) modulation δR (δT) of the sample, which is linearly related to $\delta\epsilon$—the change in the dielectric constant due to lattice vibrations. The above equation for $\delta\epsilon$ indicates that we can define a generalized [7] lattice amplitude operator [5,8]: $u_g(\pm\mathbf{q}) = (b_{\mathbf{q}} + b_{-\mathbf{q}}^\dagger)e^{i\Psi_{\mathbf{q}}} + (b_{-\mathbf{q}} + b_{\mathbf{q}}^\dagger)e^{-i\Psi_{\mathbf{q}}}$. This generalized lattice amplitude $u_g(\pm\mathbf{q}) = 2\,\mathrm{Re}\{Q_{\mathbf{q}}e^{i\Psi_{\mathbf{q}}}\}$

is the underlying microscopic quantity related to an observed reflectivity or transmission modulation when the linear term in $Q_{\mathbf{q}}$, $\delta\epsilon_1$, exists.

Since different pairs of $\pm\mathbf{q}$ phonon modes are uncorrelated to one another, the fluctuation of $O_1(=\delta\epsilon_1)$ can be expressed as $\langle(\Delta\delta\epsilon_1)^2\rangle = \sum_{q_x>0}(\hbar/2\omega_{\mathbf{q}})|\partial(\delta\epsilon)/\partial Q_{\mathbf{q}}|^2\langle\Delta u_g^2(\pm\mathbf{q})\rangle$. Here the state is $|\psi(t)\rangle = U(t)|\psi(0)\rangle = \prod_{\mathbf{q}} U_{\mathbf{q}}(t)|\psi_{\mathbf{q}}(0)\rangle$ in either the cw or the impulsive case. We can again focus on a single pair of $\pm\mathbf{q}$ modes. In the cw case, using Eq. (2), the fluctuation is

$$\langle\Delta u_g^2(\pm\mathbf{q})\rangle^{(\mathrm{cw})} = 2\{e^{-2r_{\mathbf{q}}}\cos^2[\Omega_{\mathbf{q}}(t) + \phi_{12}/2] + e^{2r_{\mathbf{q}}}\sin^2[\Omega_{\mathbf{q}}(t) + \phi_{12}/2]\}, \quad (5)$$

where $r_{\mathbf{q}} = |\zeta_{\mathbf{q}}| = \lambda_{\mathbf{q}}t/\hbar$, $\Omega_{\mathbf{q}}(t) = \omega_{\mathbf{q}}t + \pi/4$, and hereafter $\langle\cdots\rangle$ denotes an expectation value on squeezed states, unless stated otherwise. Therefore, at certain times, the fluctuation $\langle\Delta u_g^2(\pm\mathbf{q})\rangle^{(\mathrm{cw})}$ can be smaller than 2, which is the vacuum fluctuation level. Furthermore, all the pairs of phonon modes that are driven by the stimulated SORS process share the same frequency: $\omega_{\mathbf{q}} = (\omega_1 - \omega_2)/2$. Therefore, all the fluctuations $\langle\Delta u_g^2(\pm\mathbf{q})\rangle^{(\mathrm{cw})}$ evolve with the same $\omega_{\mathbf{q}}$. Notice that there is no dependence on $\Psi_{\mathbf{q}}$ in the final expression of $\langle\Delta u_g^2(\pm\mathbf{q})\rangle^{(\mathrm{cw})}$, and the squeezing factor phase $\phi_{12}/2$ has no $\mathbf{q}$ dependence; all the pairs of modes involved through the SORS share the same phase in their fluctuations. Therefore there can be squeezing in the overall fluctuation $\langle(\Delta\delta\epsilon_1)^2\rangle^{(\mathrm{cw})}$. Furthermore, the phase of this overall fluctuation can be adjusted by tuning the phase difference of the two incoming light beams.

In the impulsive case [9,16], if the $\pm\mathbf{q}$-mode phonons are driven into a squeezed vacuum state, the fluctuation in $u_g(\pm\mathbf{q})$ is

$$\langle\Delta u_g^2(\pm\mathbf{q})\rangle' = 2\{e^{-2r'_{\mathbf{q}}}\cos^2\Omega'_{\mathbf{q}}(t) + e^{2r'_{\mathbf{q}}}\sin^2\Omega'_{\mathbf{q}}(t)\}, \quad (6)$$

where $r'_{\mathbf{q}} = |\zeta'_{\mathbf{q}}| = \lambda'_{\mathbf{q}}/\hbar$, and $\Omega'_{\mathbf{q}}(t) = \Omega_{\mathbf{q}}(t) - r'_{\mathbf{q}}$. Again, the squeezing will reveal itself through oscillations in $\langle[\Delta(\delta\epsilon_1)]^2(\mathbf{q})\rangle'$ which is proportional to $\langle\Delta u_g^2(\pm\mathbf{q})\rangle'$. Note that these oscillations are essentially the same as the ones obtained in the cw case. However, now the squeezing factor is time independent. Also, the $t = 0$ phase $\pi/4 - r'_{\mathbf{q}}$ in Eq. (6) is $\mathbf{q}$ dependent. Equation (6) can be rewritten as $\langle\Delta u_g^2(\pm\mathbf{q})\rangle' = 2\{\cosh 2r'_{\mathbf{q}} + \sinh 2r'_{\mathbf{q}}\sin(2\omega_{\mathbf{q}}t - r'_{\mathbf{q}})\}$. For small $r'_{\mathbf{q}}$, this becomes $\langle\Delta u_g^2(\pm\mathbf{q})\rangle' = 2\{1 + 2r'^2_{\mathbf{q}} + 2r'_{\mathbf{q}} \times \sin(2\omega_{\mathbf{q}}t - r'_{\mathbf{q}})\}$. This expression has essentially the same form as the one obtained in [9]: $\langle Q^2_{\mathbf{q}}(t)\rangle = \langle Q^2_{\mathbf{q}}(0)\rangle\{1 + 2\xi^2_{\mathbf{q}} + 2\xi_{\mathbf{q}}\sin(2\omega_{\mathbf{q}}t + \varphi_{\mathbf{q}})\}$. The small phase term $\varphi_{\mathbf{q}}$ is neglected in [9] when computing transmission changes. The difference in phases, $r'_{\mathbf{q}}$ versus $\varphi_{\mathbf{q}}$, is negligible in the limit of very small squeezing factor, and originates from the different interaction Hamiltonians used here and in [9]. The interaction term in [9] is proportional to $u_g^2(\pm\mathbf{q})$ with $\Psi_{\mathbf{q}} = 0$ (notice that their $Q_{\mathbf{q}}$ is real and based on standing wave quantization [11]). Therefore, the interaction Hamiltonian in [9] is (in our notation) $V \propto u_g^2(\pm\mathbf{q}) \propto 2Q_{\mathbf{q}}Q_{-\mathbf{q}} + Q^2_{\mathbf{q}} + Q^2_{-\mathbf{q}}$. However, the last two terms in this expression do not satisfy momentum conservation; we thus did not include them and kept only $Q_{\mathbf{q}}Q_{-\mathbf{q}}$ in our interaction term (this form is also used by Ref. [11]).

If the linear perturbation $\delta\epsilon_1$ due to phonons is negligible, such as in [9], then the second-order correction $O_2(=\delta\epsilon_2)$ must be considered. When the phonon states are modulated by a SORS, so that the $\pm\mathbf{q}$ modes are the only ones which are correlated, then $\delta\epsilon_2 = \sum_{\mathbf{q}}\frac{\partial^2(\delta\epsilon)}{\partial Q_{\mathbf{q}}\partial Q_{-\mathbf{q}}}Q_{\mathbf{q}}Q_{-\mathbf{q}}$. Let us first focus on one pair of $\pm\mathbf{q}$ modes in the cw case. In a vacuum state, $\langle 0|Q_{\mathbf{q}}Q_{-\mathbf{q}}|0\rangle = 1$; while in a squeezed vacuum state $|0\rangle_{\mathrm{sq}}$, ${}_{\mathrm{sq}}\langle 0|Q_{\mathbf{q}}Q_{-\mathbf{q}}|0\rangle_{\mathrm{sq}} = \langle\Delta u_g^2(\pm\mathbf{q})\rangle^{(\mathrm{cw})}/2$, with the right hand side given in Eq. (5). Therefore, the expectation value of $Q_{\mathbf{q}}Q_{-\mathbf{q}}$ in a squeezed vacuum state is periodically smaller than its vacuum state value. Let us now include all the phonon modes that contribute to $\delta\epsilon_2$. In a vacuum state, $\langle 0|\delta\epsilon_2|0\rangle = \sum_{\mathbf{q}}\partial^2\delta\epsilon/(\partial Q_{\mathbf{q}}\,\partial Q_{-\mathbf{q}})$. On the other hand, in a squeezed vacuum state,

$$\langle\delta\epsilon_2\rangle = \frac{1}{2}\sum_{\mathbf{q}}\frac{\partial^2(\delta\epsilon)}{\partial Q_{\mathbf{q}}\,\partial Q_{-\mathbf{q}}}\langle\Delta u_g^2(\pm\mathbf{q})\rangle^{(\mathrm{cw})}. \quad (7)$$

Since the phase $\phi_{12}/2$ has no $\mathbf{q}$ dependence, contributions from the phonon modes sharing the same frequency add up constructively. It is thus possible that $\langle\delta\epsilon_2\rangle$ is periodically smaller than its vacuum state value. Similarly, in the impulsive case, $\langle\delta\epsilon_2\rangle' = 2^{-1}\sum_{\mathbf{q}}\frac{\partial^2(\delta\epsilon)}{\partial Q_{\mathbf{q}}\partial Q_{-\mathbf{q}}}\langle\Delta u_g^2(\pm\mathbf{q})\rangle'$; however, the phase factor in $\langle\delta\epsilon_2\rangle'$ has a $\mathbf{q}$ dependence through $r'_{\mathbf{q}}$, so that all the phonon modes with the same $\omega_{\mathbf{q}}$ do *not* contribute to $\langle\delta\epsilon_2\rangle'$ synchronously. In the cw SORS and in the very-small-$r'_{\mathbf{q}}$ limit impulsive SORS the phase of the expectation value $\langle Q_{\mathbf{q}}Q_{-\mathbf{q}}\rangle$ does not depend on $\mathbf{q}$; this is crucial to the experimental observation of modulations in the dielectric constant, because this $\mathbf{q}$ insensitivity leads to constructive summations of all the $\mathbf{q}$ pairs involved. Also, at a Van Hove singularity a large number of modes contribute to $\delta\epsilon_2$ with the same frequency and phase; thus their effect is larger and easier to observe [9].

Squeezed phonons via a finite-width SORS.—Of course, real light pulses are not δ functions. Therefore, we have also considered a SORS pumped by a light pulse with a finite width (smaller than the phonon period T) instead of a δ function. For a fixed peak height I, we find [17] that the optimal pulse width $T_{\mathrm{p}}^{\mathrm{opt}}$ that maximizes the squeezing effect satisfies $T_{\mathrm{p}}^{\mathrm{opt}} \approx T/4.4$. This calculation indicates that the experiments [9] used a pulse width which is nearby the optimal value ($T/4.4 \approx 300/4.4$ fs ≈ 68 fs $\approx T_p$). The calculation [17] can be summarized as follows. First, in the impulsive

Hamiltonian we replace the δ function by a Gaussian with its width T_p as a variational parameter. Since now the Hamiltonian is time dependent in the interaction picture, we cannot directly integrate the Schrödinger equation. Instead, we use the Magnus method to obtain the time-evolution operator and keep only the dominant first term. This approximation is valid when the pulse duration is shorter than the phonon period. We then calculate the width T_p^{opt} of the Gaussian that maximizes the squeezing factor. For a constant peak intensity, a pulse that is too narrow does not contain enough photons, while it can be proven that a pulse which is too long (i.e., with a width comparable to T) attenuates the squeezing effect.

Phonon squeezing mechanism.—What is the mechanism of phonon squeezing in the SORS processes? For the cw case, the Hamiltonian is the same as an optical two-mode parametric process [12], with the low frequency interference of the combined photon modes as the pump, the two phonon modes as the signal and idler. The frequencies of these modes satisfy $\omega_{\mathbf{q}} + \omega_{-\mathbf{q}} = \omega_1 - \omega_2$. The impulsive case is slightly different. Although the Hamiltonian is similar to a parametric process, the energy transfer from the photons to the two phonon modes is instantaneous. The resulting phonon state is a two-mode quadrature squeezed vacuum state. Indeed, a regular parametric process pumps energy into the signal and idler modes gradually, while the impulsive SORS does it suddenly. The correlation between the two phonon modes, and thus the squeezing effect, is also introduced instantaneously. Notice that this mechanism is reminiscent of the frequency-jump mechanism proposed in [3]. In the impulsive SORS, the frequency of the phonon modes has an "infinite" δ-peak change at $t = 0$, while the frequency-jump mechanism has finite frequency changes, and squeezing there can be intensified by repeated frequency jumps at appropriate times. However, as has been pointed out in [3], a finite frequency jump up immediately followed by an equal jump down results in no squeezing at all.

In conclusion, we have studied theoretically the generation of phonon squeezing [18] using a stimulated SORS process. In particular, we calculated the time-evolution operators of the phonons in two different cases: when the incident photons are in monochromatic continuous waves, and when they are in an ultrashort pulse. The amplitude of the squeezing factor initially increases with time and then saturates in the cw SORS case, while it remains constant in the pulsed SORS case. In addition, the $t = 0$ phase of the squeezing factor in the cw SORS, ϕ_{12}, can be continuously adjusted by tuning the relative phase of the two incoming monochromatic photon beams, while for the pulsed SORS the phase ($\propto \lambda'_{\mathbf{q}}$) of the squeezing factor is determined by the amplitude of the incoming light pulse. For both cases we calculated the quantum fluctuations of a generalized lattice amplitude operator and the second-order contribution to the change in dielectric constant, which is measurable. For the finite-width impulsive case, we computed the optimal pulse width, in terms of the phonon period, that maximizes the squeezing effect.

We acknowledge useful conversations with S. Hebboul, R. Merlin, S. Tamura, and H. Wang. One of us (X. H.) acknowledges support from the U.S. Army Research Office.

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