Ultrafast optical excitation of a combined coherent-squeezed phonon field in $SrTiO_3$

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Abstract: We have simultaneously excited a coherent and a squeezed phonon field in $SrTiO_3$ using femtosecond laser pulses and stimulated Raman scattering. The frequency of the coherent state (~ 1.3 THz) is that of the A_{1g}-component of the soft mode responsible for the cubic-tetragonal phase transformation at ≈ 110 K. The squeezed field involves a continuum of transverse acoustic phonons dominated by a narrow peak in the density of states at ~ 6.9 THz.

OCIS codes: (320.7130) Ultrafast processes in condensed matter, including semiconductors; (290.5910) Scattering, stimulated Raman; (270.1670) Coherent optical effects

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1. Introduction

The excitation of THz coherent optical phonons using femtosecond laser pulses has been demonstrated by several groups during the past decade [1]. More recently, ultrafast pulses were also used to generate a squeezed phonon field in $KTaO_3$ [2]. As in the electromagnetic case [3], phonon squeezing provides a way for experimental measurements to overcome the standard quantum limit for noise imposed by vacuum fluctuations. In this letter, we report on the *simultaneous* excitation of coherent and squeezed phonon fields in $SrTiO_3$. These fields are associated with, respectively, a low-frequency phonon of symmetry A_{1g} and a continuum of transverse acoustic (TA) modes. The nature of these fields can be understood by referring to the two-atom cell shown in Fig. 1 and the accompanying movies. Because the wavelength of visible light is commonly much larger than the lattice parameter, the relevant wavevectors of laser-induced phonon fields are considerably smaller than the size of the Brillouin zone. Hence, every unit cell behaves almost the same way.

Let $\mathbf{r}$ denote the averaged relative position between the atoms and $\mathbf{u}$ the instantaneous deviation from the equilibrium position for each atom. Consider also the variance $\langle \mathbf{u}^2 \rangle$. In the coherent state, $\mathbf{r}$ oscillates in time but the variances remain constant while squeezed fields show constant $\mathbf{r}$ and varying $\langle \mathbf{u}^2 \rangle$ (notice that the two frequencies are, generally, unrelated). Finally, both the relative position and the variances oscillate in the combined situation applying to $SrTiO_3$. A significant difference between coherent and squeezed lattice fields created with light pulses is that the former are monochromatic (more precisely, their frequency spectrum is discrete) while the latter are not, even in the harmonic approximation. However, squeezed fields may exhibit nearly discrete behavior in situations where the continuum spectrum is dominated by frequencies associated with peaks in the phonon density of states [2].

2. Theory of phonon field generation

The Hamiltonian relevant to our problem is $H=H_0-|E(t)|^2\sum_{\mathbf{q}}\chi(\mathbf{q})$. Here, $H_0=\sum_{\mathbf{q}}(P_{\mathbf{q}}^2+\Omega_{\mathbf{q}}^2Q_{\mathbf{q}}^2)/2$ is the harmonic contribution to the lattice energy, $E(t)$ is the magnitude of the pump electric field, and $Q_{\mathbf{q}}$ and $P_{\mathbf{q}}$ are the amplitude and the momentum of the phonon with frequency $\Omega_{\mathbf{q}}$ and wave vector $\mathbf{q}$. To second-order in the atom displacements, $\chi(\mathbf{q})=\chi_1\delta_{\mathbf{q},\mathbf{0}}+\chi_2(\mathbf{q})$ where [1,2]

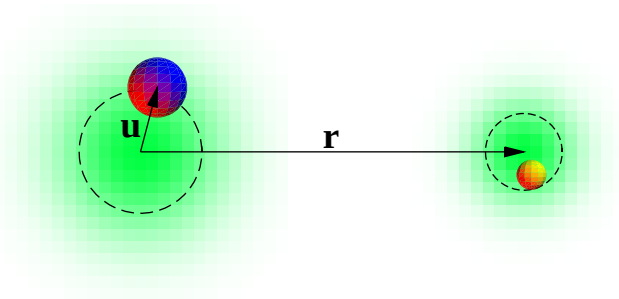

Fig. 1. Schematic diagram showing a unitary cell containing two atoms. The averaged relative position between the atoms is labeled $\mathbf{r}$, the instantaneous displacement with respect to the equilibrium position is indicated by $\mathbf{u}$, and the dashed circles represent the square root of the variance $\langle \mathbf{u}^2 \rangle$. The following links show animations illustrating motion associated with coherent, squeezed, and combined coherent-squeezed fields.

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$$\mathcal{H}_1 = \frac{1}{2}\left(\frac{\partial \chi}{\partial Q_{\mathbf{0}}}\right) Q_{\mathbf{0}} \tag{1}$$

$$\mathcal{H}_2(\mathbf{q}) = \frac{1}{4}\left(\frac{\partial^2 \chi}{\partial Q_{\mathbf{q}}^2}\right) Q_{\mathbf{q}}^2 . \tag{2}$$

Here, χ is an effective electronic susceptibility, which includes factors depending on the experimental geometry. Its derivatives, $\partial\chi/\partial Q$ and $\partial^2\chi/\partial Q^2$, are proportional to linear combinations of components of the polarizability tensors associated with first- and second-order Raman scattering [1,2]. Since $\mathcal{H}_1 \propto Q_{\mathbf{q}\equiv\mathbf{0}}$ (optical modes), Eq. (1) gives a force density F acting on $Q_{\mathbf{0}}$ which is proportional to the electric field intensity. This interaction is associated with coherent states [1]. If we ignore phonon dispersion and dissipation, the quantum-mechanical equation of motion for the expectation value of the phonon amplitude, $\langle Q_{\mathbf{0}} \rangle$, is the same as the classical expression and given by

$$\frac{d^2\langle Q_{\mathbf{0}}\rangle}{dt^2} + \Omega_{\mathbf{0}}^2 \langle Q_{\mathbf{0}}\rangle = \frac{1}{2}\left(\frac{\partial \chi}{\partial Q_{\mathbf{0}}}\right)\left|E(t)\right|^2 = F . \tag{3}$$

As discussed in [2], $\mathcal{H}_2(\mathbf{q})$ is responsible for squeezing. This term, reflecting contributions of pairs of modes at $\pm\mathbf{q}$, represents a change in the phonon frequency of $\Delta\Omega_{\mathbf{q}} \approx \nu^2/(2\Omega_{\mathbf{q}})$ with

$$\nu^2 = -\frac{1}{2}\left(\frac{\partial^2 \chi}{\partial Q_{\mathbf{0}}}\right)\left|E(t)\right|^2 . \tag{4}$$

The varying frequency leads to a time-dependent variance, $\langle Q_{\mathbf{q}}^2 \rangle$, but it does not affect $\langle Q_{\mathbf{q}} \rangle$ unless the latter quantity is different than zero. The variance satisfies the equation

$$\frac{d^3\langle Q_{\mathbf{q}}^2\rangle}{dt^3} + 4\left[\Omega_{\mathbf{q}}^2 + \nu^2\right]\frac{d\langle Q_{\mathbf{q}}^2\rangle}{dt} = -2\frac{d(\nu^2)}{dt}\langle Q_{\mathbf{q}}^2\rangle . \tag{5}$$

Consider now an optical pulse of width τ_0 such that $\tau_0 \ll 2\pi/\Omega_{\mathbf{q}}$. Then, we can approximate $E^2 = (4\pi I_0/nc)\delta(t)$ and the solutions to Eq. (3) and Eq. (5) are

$$\langle Q_{\mathbf{0}}\rangle = 2W_{\mathbf{0}}\left(\frac{\partial \chi}{\partial Q_{\mathbf{0}}}\right)\sin(\Omega_{\mathbf{0}} t) \tag{6}$$

and, to lowest order in I_0,

$$\langle Q_{\mathbf{q}}^2(t)\rangle \approx \langle Q_{\mathbf{q}}^2(0)\rangle\left[1 + 2W_{\mathbf{q}}\left(\frac{\partial^2 \chi}{\partial Q_{\mathbf{q}}^2}\right)\sin(2\Omega_{\mathbf{q}} t)\right] . \tag{7}$$

Here I_0 is the integrated intensity of the pulse, n is the refractive index and $W_{\mathbf{q}} = \pi I_0/(nc\Omega_{\mathbf{q}})$. Provided that the factor multiplying $\sin(2\Omega_{\mathbf{q}}t)$ is sufficiently large so as to overcome the thermal contribution, the variance dips below the standard quantum limit, $\sigma_{\mathbf{q}}^2 = \hbar/(2\Omega_{\mathbf{q}})$, for some fraction of the cycle. Specifically, the conditions for quantum-squeezing at low intensities are $W_{\mathbf{q}}(\partial^2\chi/\partial Q^2) > n_{\mathbf{q}}$ and $n_{\mathbf{q}} \ll 1$; $n_{\mathbf{q}}$ is the Bose factor. Thus, a situation may arise in an experiment where the high- but not the low-frequency modes become squeezed below $\hbar/(2\Omega_{\mathbf{q}})$.

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The approximation $E^2 \propto \delta(t)$ leads to a simple expression for the lattice wavefunction. Let Ψ^- be the wavefunction at $t=0^-$ immediately before the pulse strikes. Integration of the Schrödinger equation gives the wavefunction at $t=0^+$

$$\Psi^+ = \exp\left(\sum_{\mathbf{q}} \frac{2iW_{\mathbf{q}}}{\sigma_{\mathbf{q}}^2} \left[\chi_1 \delta_{\mathbf{q},\mathbf{0}} + \chi_2(\mathbf{q}) \right] \right) \Psi^- . \tag{8}$$

For $\Psi^- = |0\rangle$ and $\chi_2 \equiv 0$ ($|0\rangle$ is the harmonic oscillator ground state), this wavefunction describes a Glauber coherent state while, for $\chi_1 \equiv 0$, it shows squeezing similar to that obtained for the electromagnetic field in two-photon coherent states [3]. We should emphasize the fact that the spectrum of coherent phonon fields generated impulsively is discrete, containing a finite set of δ-function peaks [1], whereas squeezed fields involve a *continuum* of modes throughout the Brillouin zone. The continua in $KTaO_3$ [2] and $SrTiO_3$ (see later) are quasi-monochromatic for they are dominated by frequencies associated with van Hove singularities in the phonon density of states [2]

3. $SrTiO_3$

Strontium titanate undergoes an antiferro-distortive phase transition at $T_C \approx 110$ K from a cubic perovskite, point group O_h, to a low-temperature tetragonal structure of symmetry D_{4h} [4]. With decreasing T, the dielectric constant shows a dramatic increase, reaching a plateau of $\sim 10^4$ below 3 K. This plateau reflects quantum fluctuations which suppress the transition into the ferroelectric state [5] (for electric-field- and stress-induced ferroelectricity, see [7-9]). This behavior, referred to as *quantum paraelectricity* [5], has been extensively discussed in the literature [6]. Here, we are interested primarily in the change of symmetry associated with the transformation at 110 K. Second-order scattering is observed in $SrTiO_3$, *i. e.*, $\chi_2(\mathbf{q}) \neq 0$ for both $T > T_C$ and $T < T_C$ [10]. However, there are no first-order Raman-allowed modes [4] or, alternatively, $\chi_1 \equiv 0$ for $T > T_C$ (note that, in $KTaO_3$, $\chi_1 \equiv 0$ at all temperatures). In the tetragonal phase, $T < T_C$, group theory predicts $\chi_1 \neq 0$ for phonons of various symmetries [4]. This applies in particular to the A_{1g}-mode at 48 cm^{-1} and the E_g-mode at 15 cm^{-1} which are the *soft* phonons associated with the phase transition [4]. It follows that below T_C, $SrTiO_3$ meets the conditions required for the impulsive excitation of a combined coherent-squeezed field.

4. Experiments

The details of the experiment are described in the earlier work on $KTaO_3$ [2]. Fig. 2(a) shows time-domain results using a standard pump-probe configuration in the transmission geometry. The data were obtained at 7 K from a $\sim 5 \times 5 \times 0.5$ mm^3 single crystal of $SrTiO_3$ oriented with the cubic [001] axis perpendicular to the large face. We used a mode-locked Ti-sapphire laser providing pulses of full width ≈ 75 fs centered at 810.0 nm. The oscillator had a repetition rate of 80 MHz giving an average power of ~ 80 mW for the pump and ~ 30 mW for the probe pulse which were focused to a 70-μm-diameter spot. The polarizations of the pump and probe beam were along the cubic [010] and [100] directions, respectively.

The Fourier transform in Fig. 2(b) shows peaks at ~1.3 and ~6.9 THz. Based on a comparison with spontaneous Raman spectra [10] we ascribe them to the soft A_{1g}-phonon and the 2TA overtone. Consistent with the spontaneous results [10], the second-order feature is dominated by a sharp peak very close to twice the frequency of TA modes at the X and M points of the Brillouin zone [11]. The observation of the A_{1g}-mode and the 2TA continuum conclusively proves that terms of both χ_1 [Eq. (1)] and $\chi_2(\mathbf{q})$ [Eq. (2)] character participate in the excitation process and, therefore, that the overall coherence is that of a combined coherent-squeezed field. It should be noted that these results reveal no evidence of interaction

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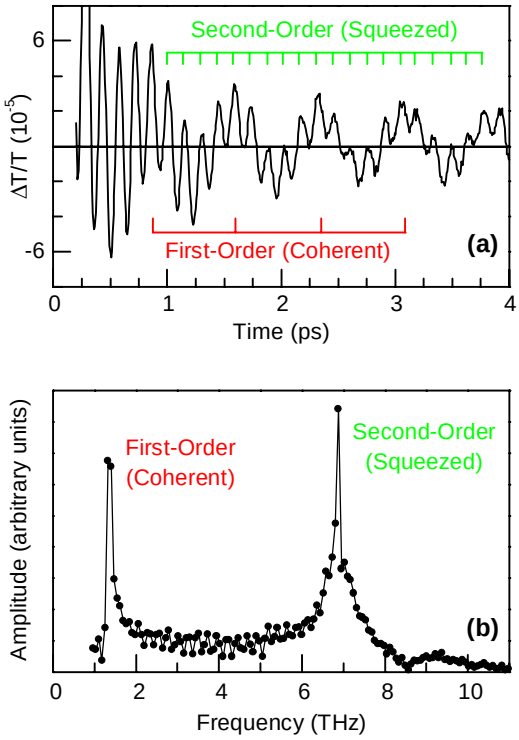

Fig. 2. (a) Normalized transmitted intensity of the probe pulse as a function of the delay for the A_{1g}-symmetry configuration. (b) Fourier transform of the time-domain data.

between the coherent and the squeezed modes, i.e., the two fields are excited independently by the pump.

In closing, we note that, because of domain structure [4], the tetragonal axis of the crystal, $\hat{c}$, has no unique direction in the laboratory and, therefore, the selection rules are not known *a priori* below T_C. However, symmetry considerations indicate that E_g-modes are only allowed if the polarizations have components parallel to $\hat{c}$. Since the data in Fig. 2 show a single first-order feature of symmetry A_{1g}, we conclude that the tetragonal axis in this case is along the cubic [001]. Other results (not shown) exhibit additional oscillations associated with the E_g-phonon at 4.3 THz [10] which, accordingly, correspond to domains for which $\hat{c}$ is perpendicular to [001].

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