Vacuum Squeezing of Solids: Macroscopic Quantum States Driven by Light Pulses


Femtosecond laser pulses and coherent two-phonon Raman scattering were used to excite KTaO₃ into a squeezed state, nearly periodic in time, in which the variance of the atomic displacements dips below the standard quantum limit for half of a cycle. This nonclassical state involves a continuum of transverse acoustic modes that leads to oscillations in the refractive index associated with the frequency of a van Hove singularity in the phonon density of states.

Squeezing refers to a class of quantum mechanical states of the electromagnetic field and, more generally, of harmonic oscillators for which the fluctuations in two conjugate variables oscillate relative to each other. This provides a way for experimental measurements to overcome the standard quantum limit for noise imposed by vacuum fluctuations. As such, the generation of squeezed light with various nonlinear processes has attracted much attention as a means of reducing noise in optical interferometry and light communication networks (1).

Following the work on photons (1), a variety of intriguing proposals were put forward dealing with squeezed states of other bosons—particularly those associated with atomic vibrations in molecular (2) and condensed-matter systems (phonons) (3)—as well as polaritons (4). In addition, squeezed phonons were considered in variational approaches to the ground state of strongly correlated electron-phonon problems (5). Here, we report an experimental demonstration of phonon squeezing in a macroscopic system (6). We have generated a squeezed mechanical state by exciting a crystal, KTaO₃, with an ultrashort pulse of light. The measurements were performed with the standard pump-probe setup (Fig. 1). Second-order coupling of the photons with the lattice vibrations [specifically, transverse acoustic (TA) modes] amounts to an impulsive change in the phonon frequency that gives rise to squeezing; this mechanism is closely related to that used to generate two-photon coherent states in quantum optics (7). We monitored the squeezed state by measuring the transmission of a second (probe) pulse that is sensitive to changes in the refractive index arising from the modulations in the mean square displacement of the atomic positions. Our state comprises a continuum of modes, but the probe transmission is dominated by a single frequency associated with a van Hove singularity in the phonon density of states.

The Hamiltonian relevant to our problem is 

$$H = \sum \Omega_q (P_q^2 + U_q),$$

where 

$$U_q = -\frac{1}{4} \langle q \rangle F^2 Q_q^2$$

is the harmonic contribution to the lattice energy and (8, 9)

Here, \(Q_q\) is the amplitude of the phonon of frequency \(\omega_q\) and wave vector \(q\), \(P_q\) is the associated canonical momentum, \(F\) is the magnitude of the electric field, \(\mathcal{P} = \sum_q \mathcal{P}_{q} \mathcal{F}_{q} \mathcal{F}_{q} \mathcal{P}_{q}\) is the second-order polarizability tensor associated with Raman scattering (RS), and \(\mathbf{e} = \mathbf{F}/|\mathbf{F}|\) is a unit vector (for clarity, we omit the phonon branch index). Equation 1 describes an effective interaction between two phonons of opposite momenta and two photons and reflects the quadratic term in an expansion of the electronic susceptibility in powers of atomic displacements (10).

The generation of the squeezed state is best understood at temperature \(T = 0\). Let \(E\) denote the pump field, and consider the assumption, valid in our experiments, that the period of the relevant phonons is large compared with both the time it takes for the pulse to cross the sample and the optical pulse width \(\tau_\text{p}\) that is, we ignore the dependence of the field on position and approximate \(E^2 = E^2(t) = (4\pi/c\eta_\text{c})(\delta(t)\delta(t))\) in Eq. 1 \((\delta(t) = \text{the integrated intensity of the pulse, } \eta_\text{c} \text{ is the refractive index, } c \text{ is the speed of the light, and } \delta \text{ is the Dirac delta function.})\). Then, if \(\psi_+\) is the wave function (the ground state) of a given mode at \(t = 0^-\) immediately before the pulse strikes, integration of the Schrödinger equation gives the wave function at \(t = 0^+\)

$$\psi_+ = \exp \left( i \frac{\xi_q \Omega_q Q_q}{\hbar} \right) \psi_-$$

where \(\xi_q = (\pi l_0 |\mathcal{P}| c \eta_\text{c}) \Omega_q\) and \(\hbar\) is Planck's constant divided by \(2\pi\). It follows that \(\langle Q_q(t) \rangle = 0\) (the brackets denote expectation value). We use the equation of motion for \(Q_q\) and the initial conditions from Eq. 2 to obtain the variance.

![Fig. 1. Schematic snapshot diagram of the experiment (not to scale). The stronger pump pulse drives the sample into an excited time-varying state, which perturbs the weaker probe pulse that follows behind. Here, the signal of interest is the transmitted intensity of the probe beam as a function of the time delay \(\tau\), as measured by the relative distance between the two pulses.](image-url)
Here, $\psi_g(t)$ can be represented by a variance trajectory (Fig. 2A), which shows squeezing similar to that obtained for the electromagnetic field in two-photon coherent states (7). As in the latter case, the motion described by $\psi$ has no classical analog (11, 12). Parenthetically, we note that the same method can be used to reduce thermal noise, other than quantum fluctuations (Fig. 2B) (13). Classically, the equation of motion is \( \dot{Q}_g = \dot{Q}_0 \), whereas \( \dot{Q}_g = \dot{Q}_0 \) (for \( g = 0 \)). It can then be shown that the isotropic equilibrium distribution becomes an elliptical one that rotates at twice the harmonic frequency (13).

The previous discussion centered on the behavior of an excitation of a well-defined wave vector. However, our experiments concern themselves not with single phonon but with real-space squeezing involving an average over all of the modes. Here, we use the fact that the wave functions of the solid as a whole are $\Psi^- = \Pi_{\psi_g}^-(t < 0)$ and $\Psi^+ = \Pi_{\psi_g}^+(t > 0)$ to calculate the variance

$$\langle \Psi^2(t) \rangle = \sum_{l} \frac{M_l}{N} \langle \Psi_l \rangle = \sum_{q} \frac{Q_q^2(t)}{NM_r}$$

which provides a measure of the squeezing (14); $u_l$ and $M_l$ are, respectively, the deviation from equilibrium and the mass of the $l$th atom in the unit cell, $N$ is the number of unit cells, and $M_r = \Sigma M_l$. Because $Q_q^2$ depends only on the mode frequency, $u_l$ and $M_l$ are pertains to the phonon density of states, that is, the characteristic frequencies for $Q_q^2$ are those of van Hove singularities where the phonon density is large. Thus, the zero-temperature description is an accurate representation of experiments performed at temperatures for which $k_B T$ (the thermal energy) is small compared with the van Hove phonon energies.

The data were obtained from a $\sim 3$ mm x $10$ mm x $0.5$ mm single crystal of KTaO$_3$ oriented with the [001] axis perpendicular to the large face at 10 K (15). As a light source, we used a mode-locked Ti:sapphire laser providing pulses of full width at half-maximum of 70 fs centered at 810 nm at a repetition rate of 85 MHz and an average power of 60 mW focused to a 70-μm-diameter spot. Spontaneous RS measurements were recorded in the backscattering configuration with 30 mW of a continuous-wave Ti:sapphire laser also tuned to 810 nm, or an Ar laser operating at 514.5 nm (16).

The scattering of the probe pulse by the squeezed state relates to the nonlinear polarization $P_{nl} = \sum_{\alpha} \chi_{\alpha}^R \chi_{\alpha}^E$ (17). Here, $E'$ is the probe field, $\chi_{\alpha}^R = (2\nu^2) \frac{2}{\nu^2} \sum_{\alpha} \chi_{\alpha}^R \chi_{\alpha}^E$, and $V$ is the scattering volume. Consider a Gaussian-shaped probe pulse of width $\tau_p$ centered at frequency $\omega_o$, that is, $E(u) \sim \exp[-(u^2/\nu^2)]$, with $\nu = \omega_p/\tau_p$ and $\tau_p$ is the time delay between the pump and probe pulses. Using well-established results for coherent phonons (17) and in the limit $\omega_o \rightarrow 0$, which is relevant to the experiments, we obtain the $\tau$ dependence of the normalized change in the probe transmission at frequency $\omega$

$$\Delta T = \frac{\Delta T}{T} = \frac{4\pi\tau_p^2 \omega(o - \omega_o)}{\nu \exp[-(u^2/\nu^2)]} \sum_{\alpha} \chi_{\alpha}^R \chi_{\alpha}^E \exp(-2\nu^2\tau_p^2)$$

where $\Delta T = \sum_{\alpha} \chi_{\alpha}^R \chi_{\alpha}^E$ and $E' = E/\exp[-(u^2/\nu^2)]$. Using Eqs. 3 to 5 and neglecting the weak dependence of $\Delta T$ on $\nu$, we obtain the function $\varphi(\omega_o)$

$$\varphi(\omega_o) = \int \Delta T \chi(\omega_o) d\omega_o$$

Accordingly, the integral of $\Delta T$ probes the variance $\langle \Delta T \rangle$, which measures the strength of the squeezing. The proportionality constant as well as $\langle \Delta T \rangle$ can be unequivocally determined from our measurements.

Time-domain results are shown in Fig. 3A. The Fourier transform $F_{\omega} = \int \Delta T(\omega) \chi(\omega_o) d\omega_o$ (Fig. 3B) is dominated by a narrow peak, strongly dependent on temperature, that appears very close to the frequency of the TA mode at the X point of the Brillouin zone, as measured by neutron scattering (18). On the basis of this peak and the comparison between the time-domain (Fig. 3B) and the spontaneous RS measurements (Fig. 3C), we ascribe the structure to the 2TA overtone. The sharpness and strength of the 2TA peak reflects to some extent the flatness of the phonon dispersion near the zone boundary (18, 19). For a given irreducible component, it can be shown from previously derived expressions (8, 9) that $F_{\omega} = \varphi(\omega) \exp(-\Omega^2/2\nu^2) \exp(-2\nu^2\tau_p^2)$, where $\varphi(\omega)$ is the second-order RS cross section and $C(\omega) = (\nu/2\nu) [1 - \exp(-\nu^2\tau_p^2)]$.

**Fig. 3. (A) Normalized transmitted intensity of the probe pulse as a function of the delay for the $\Delta T \varphi$-symmetry configuration. (B) Fourier transform of the time-domain data. (C) Weighted second-order Raman cross section $\varphi(\omega) \exp(-\Omega^2/2\nu^2)$ obtained at 810.0 nm.**

**Fig. 4. Experimental time dependence of the squeezing factor $\varphi' = 1 - (\varphi^2/2)^{1/2}$ at integrated pulse intensity $I_o = 19 \mu\text{W/cm}^2$. (Inset) Amplitude of $\varphi'$ as a function of $I_o$.**
The comparison between Figs. 3B and 3C indicates that this theoretical prediction is in reasonable agreement with the experimental data. However, there are significant differences concerning the line shape that are not understood.

To further support our interpretation, as well as to provide a quantitative estimate of the variance (ΔT^2), we obtained the absolute RS cross-section by comparing KTaO_3 with the standard CaF_2 using the 514.5-nm laser line. From these measurements, if we ignore the dependence of the polarizability on the wave vector, we find that for the A_1g component, ΔT^2 = ΔT^2 = ΔT^2 = 6 (± 2) × 10^13 cm/g, which compares favorably with the value a = (4 ± 1) × 10^13 cm/g that we obtain from the pump-probe experiments using Eqs. 3 and 5. From these values, we determine the proportionality constant related to ΔT^2/ΔT, and from the spontaneous RS measurements (20), we obtain (ΔT^2/ΔT), which corresponds to the standard quantum limit ΔT^2 = (ΔT^2/ΔT)^1/2. Combining these results and integrating ΔT^2/ΔT, we get (ΔT^2/ΔT) = (ΔT^2/ΔT) = 1 (± 1), which is referred to as the squeezing factor (21). It is estimated that ε > 0. We notice that, for ε = 1, Eqs. 3 and 4 predict that ε (≤ ε) should be proportional to the pump energy density I_0. This prediction is well obeyed for densities in the range I_0 = 5 to 20 μJ/cm^2 (Fig. 4, inset).

REFERENCES AND NOTES


6. The generation of nonclassical states has been recently demonstrated for a trapped atom by D. M. Meekhof, C. F. Lo, B. E. King, W. M. Itano, and D. J. Wineland [Phys. Rev. Lett. 76, 1796 (1996)].


10. Such a phenomenological description relying on the Born-Oppenheimer approximation applies strictly only to systems for which it can be easily formalized.