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Nonclassical states of lattice excitations: squeezed and entangled phonons

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Abstract. We consider theoretical and experimental evidence for nonclassical states of a crystal lattice. Our primary goal is the critical analysis of recent experiments in which squeezed and entangled phonons are created and investigated. The quantum aspects of elastic deformation fields similar to nonclassical light fields are of interest for potential use in applied and fundamental research, including quantum computing and testing various hypotheses in quantum physics.

1. Introduction

From the very beginning, physics has tended toward describing the motion of increasingly smaller objects over increasingly shorter periods of time. Atoms in solids move at rifle bullet speeds of about 1 km s^{-1}, and it takes tens of femtoseconds for an atom to slightly displace in a crystal. It therefore has been and is a challenge for solid state physics to understand the nature of how atoms move on their natural angstrom–femtosecond scale. Even as recently as a few decades ago, few believed that objects this small and this fast could be observed with the naked eye. But with the technological advances made and the laser pulse duration reduced to femtoseconds [1–3], the real-time study of solid-state excitations has become possible and indeed is currently one of the fastest developing areas in solid-state physics, where new, specially developed, technologically promising materials are being investigated along with model systems [4].

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Studies of the response of a condensed medium to an ultrashort laser pulse (see Refs [2, 4–8] for reviews) reveal subpicosecond oscillations in the relaxation of a pump pulse-produced state. The fact that the period of these oscillations is equal to the inverse frequency of Raman-active phonons identifies them as crystal lattice excitations (which, parenthetically, are called coherent phonons (CPs) because their phase is well-defined for excitation pulses that are short compared to the inverse frequency of the crystal lattice mode). Because optical studies tend to deal with (long-wavelength) phonons from the Γ point of the Brillouin zone, oscillations due to ultrashort laser pulses allow visualizing the motion of atoms in an individual unit cell.

It is important (but often ignored) that ultrashort pulse pump–probe techniques are the realm of active spectroscopy, where a crystal state to be investigated is specifically created by light, whereas most spectroscopic studies deal with equilibrium states that are determined by thermal excitations of the crystal. The pump–probe method using femtosecond laser pulses deals with a superposition of crystal states, as opposed to a mixture of lattice excitations, usually dealt with by standard spectroscopy. It is the active nature of dynamic spectroscopy that allows creating and studying highly nonequilibrium crystal states often inaccessible under thermodynamic equilibrium conditions (the states are nonequilibrium because the excitation pulse is much shorter than the phonon lifetime).

Explaining experiments on the influence of lattice excitations on solid-state properties does not necessarily require a quantum description; lattice dynamics can be nicely treated classically within the Newton–Boltzmann framework. Clearly, quantum theory is more general than classical theory, but is it worth invoking the former where the latter does well? To answer this question, we note that in our simple customary way of thinking, quantum fluctuations start to determine the properties of a macroscopic system only at low temperatures, whereas the Schrödinger equation and quantum statistics become necessary when objects such as quantum crystals [9], superfluid liquids [10], superconduc-

tors [11], and Bose–Einstein condensates [12] are investigated. Indeed, classical behavior dominates at temperatures $k_B T \geqslant \hbar\omega/2$, where quantum fluctuations are small compared to thermal ones. Less obvious is the fact that on a time scale less than that for the interaction of phonons among themselves and with other elementary excitations, quantum fluctuations may dominate regardless of the system temperature.

The following argument illustrates that quantum fluctuations may indeed be dominant on short timescales, implying that coherent phonons may be regarded as a quantum, rather than a classical object. The inequality $k_B T \geqslant \hbar\omega/2$ holds only for measurements averaged over times $\tau \gg \tau^*$, where τ^* is the characteristic relaxation time. However, for the femtosecond pulse methods of dynamic spectroscopy, usually $\tau \ll \tau^*$. During this short time interval, the energy exchange between the lattice mode under study and the heat bath is much less than $k_B T$, and the condition for quantum behavior to manifest itself is modified to $k_B T \leqslant \hbar\omega(\tau^*/\tau)$ [13], an inequality that holds in most femtosecond laser pulse experiments. The sensitivity to quantum fluctuations dominant over short times has enabled essentially nonclassical phonon states to be generated and studied in some femtosecond laser pulse experiments.

2. Thermal and coherent phonons

A phonon (from the Greek $\varphi\omega\nu\acute{\eta}$, sound) is a quantum of vibration that propagates in a discrete medium in the form of waves owing to the interaction between the atoms [14, 15]. We can associate a quasiparticle, a phonon, having the energy $E = \hbar\omega$ and quasimomentum $p = \hbar k$ with each such wave. Phonons in a solid arise from the quantization of the elastic deformation field, when a spatially ordered system of atoms considered as a set of *coupled* oscillators is reduced to a set of *independent* oscillators, each of which includes the displacements of all the atoms involved. Although phonons and photons obey the same commutation relations, there are numerous differences between them. Photons, sometimes called simple bosons, are elementary excitations of a continuous field. Phonons, on the contrary, are collective excitations of a discrete medium, classified as composite bosons [16]. There are two categories of the latter: bosons comprising an even number of fermions (helium atoms or Cooper pairs), and most of the collective excitations (magnons, excitons, phonons).

Differences in dispersion relations, in the ability to interact among themselves and with other excitations, the possible existence of a high-frequency limit of the spectrum, the boundedness or unboundedness of the amplitude of vacuum fluctuations — all these are factors that determine the specific behavior of phonons and photons and, in some cases, are the reason why the physics of phonons does not reduce to the physics of photons described in terms of quantum electrodynamics [17].

In the harmonic approximation, independently moving phonons behave like an ideal gas, except that the number of phonons is not conserved. The average energy of each of the independent (collective) oscillators at a temperature T is the sum of the zero-point vibration energy and the energy of the quantum times the Bose–Einstein factor $(\exp(\hbar\omega/k_B T - 1))^{-1}$ (the average number of phonons in the mode). Thermal phonons, which always exist in discrete media, are noncoherent (i.e., not correlated in any way), differ in energy, wavelength, and propagation direction, and, when superposing themselves, cause only a slight chaotic displacement of individual atoms. But if we create a large number of phonons of the same wavelength, the same frequency, and the same phase, a monochromatic deformation wave would result. Such waves, created in solids by coherent phonons, can be monitored in real time using femtosecond laser pulses [4].

The term 'phonon' is most often used simply to refer to a certain portion of the crystal lattice energy and is therefore equivalent to 'lattice excitation' or 'sound', similarly to how 'photon' is synonymous with 'light'. The field of elastic crystal deformations is in this case treated purely classically, without any hint of the discrete nature of the mode spectrum (the 'first quantization' is not of use for processes that change the number of particles), i.e., a quantum oscillator is replaced by a classical one.

However, what undoubtedly distinguishes a quantum from a classical oscillator is that the former can move in regions forbidden to classical motion (tunneling, over-barrier reflection) and that, importantly, its spectrum exhibits zero-point (vacuum) vibrations of the energy $\hbar\omega/2$, which correspond to the quantum number $n = 0$ and whose wave function has no zeros for finite displacements. A nonzero minimum energy is a consequence of the uncertainty relation [18], which prevents a quantum system from being at rest.

Zero-point vibrations play a major role in physics; in particular, they prevent liquid helium from crystallizing at normal pressure, even at the absolute zero of temperature [10], and determine the 'superproperties' of quantum crystals [9]. This energy is usually neglected when passing from a quantum description to a classical one, and the zero (vacuum) level is taken as the reference point This approximation is not always innocuous, however: a quantum potential can be reduced to a classical one if and only if all energy levels are taken into account. It is easily shown that the removal of the zero level does not violate the equidistant nature of the spectrum, meaning that the potential remains a quantum harmonic one. Still, the period of quantum oscillations in this potential turns out to be energy-dependent [19], i.e., the classical analog of this potential does not have the property of being isochronous or, in other words, is not a classical harmonic potential [20].

Attempts to find a combination of quantum (discrete) states that obeys Newton's mechanics has led to the concept of a 'coherent state' (CS) [21, 22]. Clearly, applying a classical force cannot bring an oscillator from a vacuum state to a Fock state with a definite number of quanta [23]. In that state, the average coordinate and the average momentum are both identically zero, whereas for a quantum oscillator subject to an external force, these average quantities vary in a harmonic fashion, as is the case of a classical oscillator.

Coherent states, which approximate the classical description best, can be obtained 1) as minimum-uncertainty states, 2) as eigenstates of the annihilation operator, or 3) by using the displacement operator. In particular, a CS $|\alpha\rangle$ can be obtained from the vacuum state $|0\rangle$ by applying the displacement operator, giving $|\alpha\rangle = \hat{D}(\alpha)|0\rangle$. The unitary operator $\hat{D}(\alpha) = \exp(\alpha\hat{a}^\dagger - \alpha^*\hat{a})$, linear in the ladder operators $\hat{a}^\dagger$ and $\hat{a}$, displaces the vacuum uncertainty contour from the coordinate origin of the phase plane to the point $(\mathrm{Re}\,\alpha, \mathrm{Im}\,\alpha)$, i.e., displaces the mean value of the coordinate by an amount proportional to the real part of α and the mean value of the momentum, by an amount proportional to the imaginary

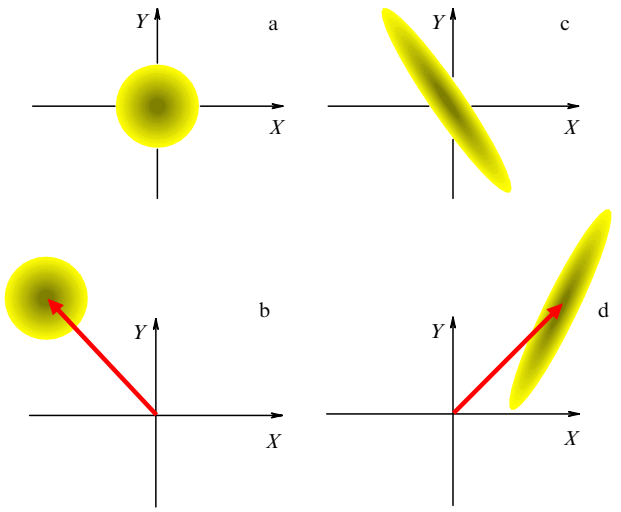

Figure 1. Schematic representation of (a) the vacuum, (b) a coherent state, (c) a squeezed vacuum, and (d) a generalized coherent state. States in (a) and (b) have their quadratures uncorrelated, leading to an uncertainty contour circular in shape. The presence of correlations in the states in (c) and (d) results in elliptically shaped uncertainty contours.

part of α. We note that this does not change the shape or area of the uncertainty contour, because the classical force acting on the oscillator changes only the first moments, i.e., the means, and leaves the second moments, the dispersions, unchanged. This indicates that a CS can be represented as a classical state with added vacuum noise, as shown schematically in Fig. 1b, and this also explains why this state is sometimes called displaced.

Because the canonical variables of the oscillator have different dimensions, the dimensionless quadrature operators $\hat{X} = (m\omega/2\hbar)^{1/2}\,\hat{q}$ and $\hat{Y} = (2\hbar m\omega)^{-1/2}\,\hat{p}$ are introduced to simplify the description, allowing a lattice mode to be written as

$$A = \frac{\sigma_0}{\sqrt{2}}\,(\hat{X}\cos\omega t + \hat{Y}\sin\omega t)\,,$$

where

$$\frac{\sigma_0}{\sqrt{2}} = \sqrt{\frac{\hbar}{2m\omega}}$$

is the vibration amplitude. Passing from the phase space (q, p) to the space (X, Y) deforms the oscillator orbit from an ellipse to a circle. The dispersions of quadrature operators for the CS are $\langle\Delta\hat{X}^2\rangle = \langle\Delta\hat{Y}^2\rangle = 1/4$, which saturates the Heisenberg uncertainty relation $\langle\Delta\hat{X}^2\rangle\,\langle\Delta\hat{Y}^2\rangle \geqslant 1/16$, whereas the circular shape of the uncertainty contour indicates that both quadratures have the same noise. Moreover, $\langle\Delta\hat{X}^2\rangle + \langle\Delta\hat{Y}^2\rangle = 1/2$, i.e., the quadratures of a CS are independent and uncorrelated. A CS is the most classical of all possible quantum states, but it still remains quantum. More precisely, a coherent state is a boundary between the sets of classical and essentially quantum states of a field.

3. Nonclassical excitations of a crystal lattice

A classical force acting on the vacuum state can produce a coherent state because such a force changes only the mean values of the coordinates and momenta while leaving their dispersion relations unchanged. If the force changes the uncertainties (which is possible if the interaction is nonlinear), we obtain states that have little in common with classical physics (for example, with mean square deviations larger than the mean values, something quite unusual for nonnegative quantities such as the kinetic or potential energies).

Whether nonclassical excitations exist and what their properties are has been and still is the subject of many studies concerned with the light field [25, 26]. It is photons for which, for the first time, the uncertainty contour was squeezed [27–29] and entangled [30], and other nonclassical electromagnetic field states were obtained. Subsequent theoretical studies used analogies with the light field to examine the prospects for creating squeezed phonons (SPs) and their specific properties. There are two groups into which these studies can be classified. The first treats the phonon as an element of a complex object (a phonon polariton [31, 32] or a polaron [33]); in the second group, the phonon is regarded as an independent excitation, with fluctuation squeezing represented as resulting from the interaction of phonons between themselves or with photons [17, 34–46]. Some of these studies laid the necessary foundation for the experimental implementation of squeezed vibrational states (in molecules [47]) and phonon (in crystals states [42, 48–54]).

If the phonon is considered a quantum object rather than a classical wave, the repeated measurements of any of the dynamical variables needed for its description yield the mean value whose accuracy is limited by noise. We know, indeed, that each normal lattice mode quantized into phonons can be described mathematically by a wave equation whose solution is a function of coordinates and time. The uncertainty (or noise) of this function corresponds to the 'thickness' of the phase space line, and this thickness reflects the quantum uncertainty in the positions of the atoms whose motion gives rise to the phonons. This uncertainty, while usually small, can be modulated in a periodic fashion by squeezing fluctuations. Franco Nori's site [55] offers vivid animations of various 'squeezed' phonon states and their evolution.

Squeezed states (SSs), which are a certain generalization of coherent states, were introduced under a variety of names, including pulsed [56], generalized coherent [57], two-mode vacuum [40], new quantum [58], twisted [59], and correlated coherent [60] states. The well-established term *squeezed states* comes from Hollenhorst [61]. Squeezed states were first introduced by Kennard [62] in his analysis of the evolution of wave packets with a time-dependent width and were reinvented nearly thirty years later by Takahashi [63] in a parametric amplification analysis using the theory of wave functions. A detailed history of the subject and a nearly complete bibliography can be found in [64]. In Russian, papers [28, 29] and monograph [65] are representative reviews of the physics of SSs.

The physics of SSs is best illustrated using the Schrödinger picture. We recall that when introducing the CS, in order to obtain the minimum-uncertainty state, we require the width of the Gaussian function to be equal to that of the vacuum level. To see what happens if we lift this requirement, we represent the width of the Gaussian function in the form $\sigma = s\sigma_0$, where s is a real number called the squeezing factor,

$$\psi_{ss}(q) = \frac{1}{\sqrt{2\pi\sigma^2}}\exp\left[-\frac{(q-q_0)^2}{2\sigma^2} + \frac{\mathrm{i}}{\hbar}\,p_0 q\right].$$

Introducing the factor s either squeezes (for $s < 1$) or stretches (for $s > 1$) the Gaussian wave packet (in the momentum representation, squeezing and stretching are interchanged). The fundamental difference between a CS and an SS is that for $t \neq 0$, an SS is no longer a minimum-uncertainty state. To see what happens as this state evolves, we calculate the mean values and the squared mean values of the SS coordinate and momentum, which gives their dispersions

$$\left(\Delta q(t)\right)^2 = \sigma_0^2 \left(s^2 \cos^2 \omega t + \frac{1}{s^2} \sin^2 \omega t \right),$$

$$\left(\Delta p(t)\right)^2 = \frac{\hbar^2}{4\sigma_0^2} \left(\frac{1}{s^2} \cos^2 \omega t + s^2 \sin^2 \omega t \right).$$

Because shifting the argument by $\pi/2$ changes sine to cosine, the coordinate and momentum dispersions oscillate in counterphase. Their product

$$\left(\Delta q(t)\right)^2 \left(\Delta p(t)\right)^2 = \frac{\hbar^2}{4} \left(1 + \frac{1}{4} \left(s^2 - \frac{1}{s^2} \right)^2 \sin^2 2\omega t \right)$$

demonstrates that for $s = 1$, we obtain a CS for which the product of uncertainties is independent of time. But for an SS with $s \neq 1$, this product oscillates with time at the doubled frequency, meaning that the time variation of the shape of the uncertainty contour is of a quadrupole nature. Thus, the dimensionless dispersion relations for noncommuting dynamic variables are different for an SS and identical for a CS — with the result that as the Gaussian wave packet corresponding to a CS evolves, its width is not constant but oscillates. Because of their specific properties, squeezed states have proved very useful in solving a range of problems in the physics of quantum optics, gravitational wave experiments, and quantum nondemolition measurements [13].

It is generally possible to create a phonon field for which the quadrature dispersions are different. Physically, this means that a correlation between the quadratures begins to emerge; in mathematical terms, this is achieved through the introduction, for a field with an annihilation operator $\hat{a}$, of a new operator $\hat{b} = \mu\hat{a} + v\hat{a}^\dagger$, with complex numbers μ and v satisfying the relation $|\mu|^2 - |v|^2 = 1$.

We note that the introduction of a new operator is in fact analogous to the Bogoliubov transformation [66] necessary in describing superfluidity and superconductivity. In performing this transformation, we no longer require the width of the wave packet composed of discrete states to be equal to that of the vacuum state, but instead allow it to be arbitrary.

Experimentally, an SS is implemented by parametric excitation, a process that changes not only the mean values but also the second central moments of the quadratures. It is the presence of correlations induced by parametric excitation described by the squeezing operator that results in the unitary squeezing operator

$$\hat{S}(\zeta) = \exp\left(\frac{\zeta(\hat{a}^\dagger)^2 - \zeta^*(\hat{a})^2}{2} \right)$$

(which is quadratic in the ladder operators) results in the elliptical shape of the SS uncertainty contour. The complex parameter $\zeta = s\exp(i\theta)$ specifies both the amount s and the phase θ of squeezing, its phase being equal to that of the parametric pumping. We note that one of the quadratures exponentially decreases (with the means and dispersions decreasing proportionally), whereas the other increases. From the standpoint of an observer at rest relative to the coherent amplitude, noise becomes time dependent, with its variation frequency 2ω being twice that of the coherent amplitude.

Unlike CSs, SSs are sometimes very far from being classical, because the quadrature uncertainties may become arbitrarily large for squeezing factors tending to zero or infinity. Lattice states with a definite coordinate or a definite quasimomentum correspond to these limit states. For lattice excitations, various SPs can be implemented depending on the type of correlation that occurs. Thus, quadrature squeezed states correspond to a quadrature anticorrelation in which the major axis of the uncertainty ellipse is parallel to one of the quadratures. If the major axis of the uncertainty contour is parallel (perpendicular) to the coherent amplitude, we are dealing with phase-squeezed (amplitude-squeezed) phonons. These phonons correspond to lattice excitations squeezed with respect to fluctuations in the number of phonons, i.e., the uncertainty in the number of phonons becomes less than that in a CS (Poisson distributed fluctuations) — something possible only for sub-Poisson statistics. For an uncertainty ellipse oriented arbitrarily with respect to the quadrature unit vectors, we are dealing with 'rotated' SPs [65] (Fig. 1d).

In the foregoing discussion, squeezing and the correlations responsible for it are not related in any quantitative way. But if we consider a phonon for which quadrature fluctuations are different due to correlations between the quadratures, then we easily see that the degree of correlation and the degree of squeezing are no longer independent variables [60]. To show this, we consider the Schrödinger–Robertson uncertainty relation

$$\sigma_X \sigma_Y \geqslant \frac{1}{2} \left(\left\langle \left| [X, Y] \right| \right\rangle + \left\langle \left| \{X, Y\} \right| \right\rangle \right),$$

where the square (curly) brackets denote the commutator (anticommutator) and the angular brackets denote the mean [67, 68]. In the case where the mean of the anticommutator is expressed in terms of the correlator $r = \sigma_{XY}/\sqrt{\sigma_X \sigma_Y}$, this becomes

$$\sigma_X \sigma_Y \geqslant \frac{\left\langle [X, Y] \right\rangle}{2(1 - r^2)}.$$

Because the mean of the quadrature commutator is unity, determining the mean square deviation for one of the quadratures, σ_X, yields the second one as [60]

$$\sigma_Y = \frac{1}{2\sigma_X} (1 - r^2)^{-1}.$$

With the lattice function squeezing parameter introduced as $s = \sigma_X/\sigma_Y$, the mean square deviation of the quadrature s can be written as

$$\sigma_X = \sqrt{\frac{s}{2\sqrt{1 - r^2}}},$$

clearly indicating that the fluctuations depend on both squeezing and correlations. Introduced in this way, the parameters s and r characterize the eccentricity and orientation of the uncertainty contour defined in the intervals $0 \leqslant s \leqslant \infty$ and $-1 \leqslant r \leqslant 1$ [60].

Quantum correlations are stronger than classical ones and can lead to entanglement, a phenomenon with no classical

analogs, in which the motions of lattice atoms are related not via usual interactions limited by finite propagation speed but due to nonlocal quantum correlations. The concept of entanglement, closely related to the Einstein–Podolsky–Rosen paradox [69], is, in the words of Schrödinger, the essence of quantum mechanics [70]. For any quantum system composed of a number of parts, entanglement is defined as the nonfactorizability of the total wave function and reduces to the presence of quantum correlations between the parts [71].

4. Experimental details

Coherent photons, i.e., coordinated vibrations in which the atoms of each crystal unit cell move in unison, are studied (similarly to SPs) by the femtosecond pump–probe technique. In one of its simplest forms, the excitation method measures transmission or reflection [4–8], for which a beamsplitter transforms a train of laser pulses into two beams, with the probe beam one to three orders of magnitude lower in power than the pump beam.

By introducing a controllable probe–pump time delay, the evolution of a lattice state prepared by a pump pulse can be traced. The way to realize this is by detecting the normalized differential reflection/transmission, i.e., the change in the corresponding characteristic of the probe pulse produced by the pump beam normalized to the same probe characteristic in the absence of pumping. This change is proportional to either atomic displacements or their dispersions, depending on whether one-phonon or two-phonon excitations are generated [4, 6].

As an example, Fig. 2 shows a typical reflection relaxation response observed in a bismuth single crystal by the pump–probe method using femtosecond laser pulses [8]. It is clearly seen that the excited state evolves toward equilibrium and that superposed on this relaxation are oscillations produced by the CP generation process. At liquid helium temperatures, CPs are sufficiently long-lived: for example, bismuth atoms perform hundreds of oscillations before the lattice subsystem loses its coherence [72].

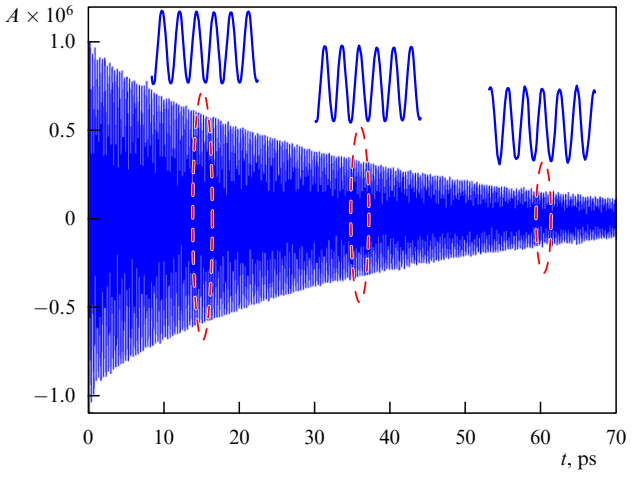

Figure 2. Coherent part of the time-resolved reflection $\Delta R/R_0 = A$ in a bismuth single crystal at liquid helium temperature for weak excitation by 30 fs laser pulses.

5. Evidence for squeezed phonon states

There is currently abundant evidence that the excitation of a crystal by ultrashort laser pulses causes its phonon subsystem to behave nonclassically, supposedly due to the squeezing and to the noncorrelated nature of lattice modes. Indeed, such pulses allow not only preparing lattice excitations in a coherent or a squeezed state but also 'entangling' macroscopic objects by exciting the crystal to entangled states. We discuss these experiments in what follows.

Squeezed phonons, i.e., lattice excitations fluctuating differently in different quadratures, were first created and registered by Roberto Merlin's group at Michigan State University [42] in what was seemingly an ordinary dynamic spectroscopy experiment on a potassium tantalate ($KTaO_3$) crystal at a sufficiently low temperature of 10 K. The researchers came to recognize, however, that in order to detect a squeezed phonon state, it is desirable to eliminate the coherent amplitude, whose large value makes vacuum fluctuations difficult to observe. Because the coherent amplitude behaves classically, destructive interference can be used to suppress it to zero. The way the group achieved this was by coherently exciting wave-vector-anticorrelated pairs of transverse acoustic (TA) phonons from the X point of the Brillouin zone. Because of the wide frequency range of acoustic phonons in crystals, their produced periodic squeezing of atomic fluctuations occurs in various time intervals, thus preventing the experimental separation of a squeezed state from multiple signals due to various lattice excitations. But in $KTaO_3$ crystals, because of the presence of a van Hove singularity at the boundary of the Brillouin zone, the density of acoustic phonon states is large for a certain frequency. Because of the in-tune nature of all these phonons, their squeezed states produce a very distinct signature, a regular change in the refractive index of the material detected by the probe pulse. The cubic O_h symmetry of $KTaO_3$ and the selection rules prevent the excitation of one-phonon states, further facilitating the identification of squeezing of the phonon subsystem. The Michigan group used pulse durations of 70 fs (the repetition frequency 85 MHz, the wavelength 810 nm) to obtain a sufficient time resolution for creating and tracing the evolution of a squeezed lattice state. Each pump pulse was followed by a (time delayed) probe pulse, whose transmission depends on the crystal refractive index, which is in turn sensitive to the mean square of atomic displacements.

The change of the $KTaO_3$ transmission T under the influence of an ultrashort laser pulse is caused by second-order Raman scattering,

$$\Delta T \equiv T - T_0 = \sum_q \frac{\partial^2 T}{\partial Q_q \, \partial Q_{q'}} \langle Q_q Q_{q'} \rangle ,$$

where Q is the normal vector of the acoustic wave and the averaging is over the phonon states for which the total wave vector is zero [42]. The averaging yields $\langle Q_q Q_{-q} \rangle = \langle \Delta u^2(\pm q, t) \rangle$, where u is the atomic displacement in the acoustic mode. We note that what was measured in the experiment exciting a pair of acoustic 2.7 THz phonons was precisely the dispersion of atomic displacement, because the coherent amplitudes of those modes responsible for the modulation of interatomic distances are anticorrelated, and therefore their sum vanishes. A schematic of this situation is shown in Fig. 3, which presents a unit cell of a noncentrosymmetric crystal.

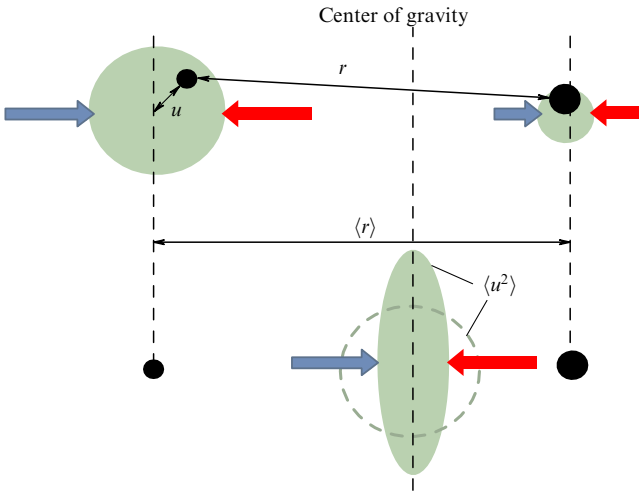

Figure 3. (Color online) Unit cell of a nonsymmetric crystal. Blue and red arrows represent the normal vectors of acoustic phonons with anti-correlated wave vectors. Atoms are shown as black dots with diameters proportional to their masses. $\langle r\rangle$ is the average atomic separation, r is the instantaneous atomic separation, and u is the instantaneous displacement from the equilibrium position. The green dots and the ellipse indicate the dispersions $\langle u^2\rangle = \langle r^2\rangle - \langle r\rangle^2$. Excitation of a pair of phonons leaves the average atomic separations $\langle r\rangle$ unchanged, with only their dispersion $\langle u^2\rangle$ changing as shown at the bottom.

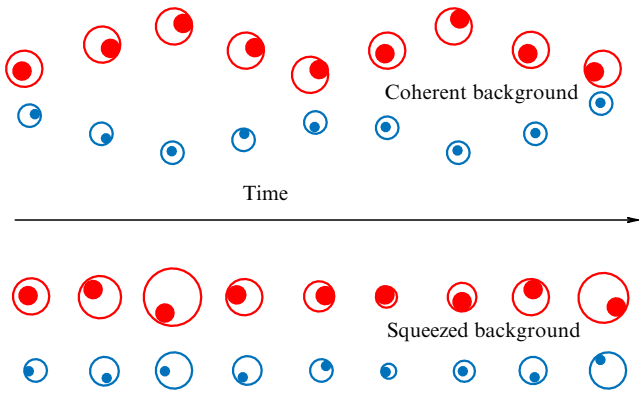

Figure 4. Coherent and squeezed phonons are shown schematically to demonstrate their difference. The evolution of a coherent state is accompanied by a change in the average atomic separation $\langle r\rangle$; the evolution of a squeezed state leaves $\langle r\rangle$ unchanged and affects only $\langle u^2\rangle$.

As shown in Fig. 4, the squeezed phonons observed in Ref. [42] modulate just the dispersions and leave $\langle r\rangle$ unchanged — in contrast to CPs, which modulate the average atomic separation $\langle r\rangle$ and leave dispersions $\langle u^2\rangle$ unchanged. Using this fact, Merlin's group measured the time dependence of the dispersion of atomic displacement and experimentally demonstrated its oscillatory nature. The 2.7 THz oscillations that were associated with SPs showed the symmetry $\Gamma_1(A_g)$, the same as that of the crystal. Despite the clear and definite signal obtained, the researchers' estimate of the squeezing was moderate. By estimating the amplitude of vacuum displacements from the measurements of the absolute Raman scattering cross section, the uncertainty in the amplitude distribution was found to decrease by one millionth. Following this experiment, similar implementations of squeezed acoustic phonons in $SrTiO_3$ [48] and ZnTe [54] were reported.

For strontium titanate, which, unlike potassium tantalate, undergoes a cubic-to-tetragonal $O_h \to D_{4h}$ phase transition at 110 K, together with a squeezing of the pairs of acoustic phonons from the X and M points of the Brillouin zone, the coherent excitation of fully symmetric optical A_{1g} phonons of the 'soft' 1.3 THz mode was demonstrated [48]. That is, following ultrafast excitation at 7 K, the soft-mode phonons turn out be in a coherent (Glauber) state and the acoustic phonons are in a two-mode squeezed state with an energy of 6.9 THz. The soft-mode phonons, following the excitation by an ultrashort (70 fs) pulse (the repetition frequency 80 MHz and the wavelength 810 nm), experience a sharp change in the average quasimomentum $\langle p\rangle$, whereas a sharp change in $\langle p^2\rangle$ occurs for acoustic phonons. For the coherent case, the average quasimomentum $\langle p\rangle$ oscillates, as it should according to the classical equation of motion, whereas $\langle p^2\rangle$ remains unchanged. For acoustic phonons, on the contrary, $\langle p\rangle = 0$, whereas $\langle p^2\rangle$ oscillates at twice the frequency of the acoustic mode, remaining less than the mean square vacuum state throughout half the cycle. A point to note is that both the $KTaO_3$ and $SrTiO_3$ crystals exhibit the phenomenon of paraelectricity [73–75]. The squeezing of lattice excitations detected in these crystals possibly reflects the onset of the transition to the ferroelectric phase, frustrated by large quantum mechanical fluctuations.

Unlike the perovskite paraelectrics $KTaO_3$ and $SrTiO_3$, a typical II–VI compound of zinc telluride (ZnTe) is a direct-band 2.34 eV gap semiconductor crystallizing in the zinc blende type structure ($T_d(\bar{4}3m)$ space group). Due to its high photosensitivity, ZnTe has applications in efficiently converting solar energy and in visualizing images in the teraherz range. Its first-order Raman scattering spectrum consists of two modes, the 6.3 THz longitudinal optical (LO) and the 5.3 THZ transverse optical (TO) ones. Two-phonon scattering from the Brillouin zone X point is described by the direct product of three irreducible representations, $X^2 = \Gamma_1 \otimes \Gamma_{12} \otimes \Gamma_{15}$, in which the fully symmetric component dominates with a frequency of 3.24 THz (108 cm^{-1}) [76].

The generation of CPs in ZnTe (001) excited by 30 fs 800 nm laser pulses was reported in [77]. This experiment, in which, because of its geometry, the time-resolved transmission response was dominated by the 6.3 THz LO phonon along with the acoustic overtone, is in many respects analogous to the experiment on $SrTiO_3$ in Ref. [58]. The authors of Ref. [77] only mention the possibility of squeezing acoustic modes and focus their attention on polarization measurements aimed at establishing the symmetry of the phonon modes observed. Changing the geometry of the experiment in [54] allowed eliminating the one-phonon contribution and separating the second-order scattering, thus leading to a situation similar to that in the experiment with $KTaO_3$ [42]. In Ref. [54], this was implemented by using a ZnTe crystal with a different, (110), orientation, for which scattering on an LO phonon is forbidden by selection rules (TO scattering is in both cases weak due to resonance conditions). A number of features identified in the same paper by a detailed study of two-phonon scattering suggest the presence of squeezing and entanglement in the acoustic subsystem.

The evolution of squeezed ZnTe phonons created by an fs pump pulse is shown in Fig. 5. From the signal Fourier spectrum shown in the figure, it can be seen that vibrations at the fundamental acoustic phonon frequency (1.62 THz) are absent; there are only changes in the dispersions of the coherent amplitudes of two acoustic modes occurring at

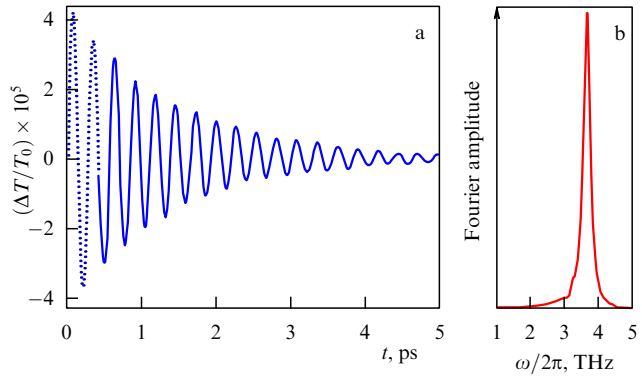

Figure 5. (a) Squeezed state evolution in ZnTe as a change in the normalized differential transmission $\Delta T/T_0$ (dotted line extrapolates to the zero delay, demonstrating the onset of oscillations). (b) Fourier transform of the oscillations.

nearly a double frequency. The frequency of the oscillations (3.67 THz) is somewhat higher than that of the acoustic overtone (3.24 THz), identifying the oscillations as an acoustic biphonon [54]; polarization measurements identified the symmetry of the biphonon excitations as the unit representation with the full symmetry of the point group. That is, an ultrashort pulse excites two acoustic modes in ZnTe, with the same frequency and magnitude but with vectors $|q_+\rangle$ and $|q_-\rangle$ opposite in sign. Just as with the perovskite paraelectrics $KTaO_3$ and $SrTiO_3$, the high density of acoustic phonon states (due to the van Hove singularities) allows energy to be pumped effectively into a ZnTe crystal. What possibly distinguishes ZnTe from the perovskite structures is that the residual phonon–phonon interaction is responsible for the energy shift that places the biphonon above the acoustic continuum band. The dispersion of the acoustic biphonon at the center of the Brillouin zone depends on the wave vector, which corresponds to a negative mass, whereas at the edges of the Brillouin zone, the dispersion is linear [78]. For the ultrafast excitation of ZnTe, the biphonon components defined by the annihilation operators $\hat{a}_+$ and $\hat{a}_-$ satisfy the relation $2\left|\langle\hat{a}_+\hat{a}_-\rangle\right| > \langle\hat{a}_+^\dagger\hat{a}_+\rangle + \langle\hat{a}_-^\dagger\hat{a}_-\rangle$, and we therefore have a typical case of two-mode squeezing [54].

A toy model provides a qualitative understanding of the experimental results in Refs [42, 48, 54]. We can think of lattice atoms as pendulums, each of which is in its particular (and totally random) phase of vibration, some in full swing, some close to equilibrium, etc. The laser pulse has the effect of a 'vertical blow' on all of the pendulums. The pendulums near the equilibrium and those at the maximum height are respectively least and most affected. That is, the momentum a pendulum receives along its path of motion is proportional to the deviation from equilibrium, with the ultimate result that the motion of the system of pendulums changes from chaotic to ordered (or coordinated), and the pendulums make the transition to synchronous motion, i.e., come to an ordered state that is implemented by the squeezing of the wings of the distribution of instant displacements and which 'squeezes' noise from the motion of the system of pendulums. Importantly, such an action creates a correlation between the displacement and the momentum for each of the pendulums. Under the assumption of an elastic impact, each pendulum has its frequency unchanged, and the only property that does change is its impedance, an aspect ratio that characterizes the phase space orbit of the pendulum [79]. This may point to the nonadiabatic nature of the influence, for which the phonon mode has one of the quadratures exponentially decreasing, $\Delta X \propto \exp(-s)$, and the other exponentially increasing, $\Delta Y \propto \exp(s)$. Motion in this case occurs such that the kinetic and potential energies fluctuate in an anticorrelated manner, which is possible only if the kinetic and potential energy dispersions exceed their mean values [16]. Physically, this corresponds to the situation where the oscillator is most of the time near the equilibrium rather than far from it as in classical motion, where the time needed for an object to pass a given portion of its path is inversely proportional to its velocity. We note that the atoms whose motion forms a phonon spend most of their time on lattice sites.

Although the first observation of CPs [42] received wide media news coverage at the time, things are not entirely smooth with this interpretation. Shortly after the publications by the Michigan group, their conclusions were questioned by two theoretical physicists, who came up with critical comments [17]; the criticism was primarily concerned with the interpretation of the experiments and did not consider the setup and the results. Comparing the thermal noise and the degree of squeezing, the authors of [17] conclude that the magnitude of modulation does not suggest vacuum squeezing. Importantly, however, the experiments in Ref. [42] used a differential technique, thus excluding the contribution from thermal, and indeed from any time-independent, noise. Essentially, the critics argue that the noise modulated in the experiments in Ref. [42] was that of the anticommutator, not of the commutator, because the experimental conditions used corresponded to the case where $\left\langle\left|\{\Delta\hat{X}, \Delta\hat{Y}\}\right|\right\rangle \geqslant \left\langle\left|[\hat{X}, \hat{Y}]\right|\right\rangle$. Such a situation occurs in the semiclassical limit, in which, as $\hbar \to 0$, the commutator contributes nothing to the Schrödinger–Robertson relations, whereas the contribution from the anticommutator tends to the correlator of classical quantities and is nonzero if there is a statistical dependence between them.

The possibility of such a situation is evidenced indirectly by a detailed study of the temperature dependence of the ultrafast dynamics of potassium tantalate [51]. By shortening the exciting pulse to 25 fs, the authors of [51] were able, in the time range, to register oscillations of the sum $(TO_{1,2} + TA, TO_4 + TA)$ and of the difference $(TO_{1,2} - TA, TO_4 - TA)$ of phonon modes. Decreasing the temperature considerably decreased the contribution from the latter, although at room temperature the oscillation spectrum was practically the same as that of spontaneous Raman scattering [80].

A number of discrepancies between the results of spontaneous Raman scattering and time-resolved measurements were also noted in [17]. An example is the difference in spectral line shapes between an acoustic overtone observed in the frequency domain and the squeezed phonon state registered in the temporal domain. This shows up, in particular, in the narrower line of the squeezed state, indicating that the lifetime of a lattice excitation is longer for an ultrashort laser excitation than for continuous laser exposure.

We must note, however, that the difference in lifetimes between the coherent and incoherent excitations is a widespread, although not yet understood phenomenon [8]. According to the generation mechanism due to SP Raman scattering [17, 42–44], the presence of second-order scattering in the time-resolved coherent response necessarily indicates the squeezing of the phonon subsystem, whereas alternative mechanisms [46] require several pump pulses (i.e., coherent

control) to implement the vacuum squeezing of lattice excitations.

Attempts have also been made to register the fluctuation properties of optical CPs to detect the squeezing of the phonon subsystem. For this, statistical properties of coherent optical modes were investigated in bismuth, antimony, gallium arsenide, and a number of high-temperature superconductors [49, 50, 52]. The study of the fluctuation properties of phonons produced by ultrashort laser pulses required a modification of the measurement scheme [8], because the traditional scheme for the study of CPs provides only the time dependence of the mean value of the coherent amplitude. Investigating fluctuations requires determining how the coherent amplitude changes with time at a fixed phase determined by the time delay; hence, the measurement should be conducted so as to lose no information about the individual implementations of the lattice state. Although a standard experimental configuration cannot measure a single realization of the lattice state, it is possible to compare statistical samples obtained by applying about 10^5-10^6 laser pulses at a fixed time delay. For this, a part of the oscillating response $A(t)$ was measured repeatedly, such that for each time delay value, along with the first moment $\mu = \langle A \rangle$, the central second-order moment $\sigma^2 = \langle A^2 \rangle - \mu^2$ could be computed.

A typical result for a Bi single crystal is shown in Fig. 6, from which it is obvious that the average amplitude obtained with this scheme is identical to that from a traditional experiment. But there is also additional information contained in the coherent-amplitude dispersion calculated for each time instant. For all the crystals studied, the dispersion (or the noise characteristic) of CPs turns out to be time dependent, with its variation frequency twice the CP frequency (Fig. 6a). We note that for a system in equilibrium, noise is time independent, as is illustrated by the behavior of dispersion at negative time delays. Typical noise and coherent-amplitude power spectra obtained by the Fourier transformation are shown in Figs 6a and 6b. It is clear that these two spectra are different and that the noise spectrum contains a component with twice the CP frequency. As the pumping power increases, the noise intensity increases linearly, whereas the spectrum remains virtually unchanged.

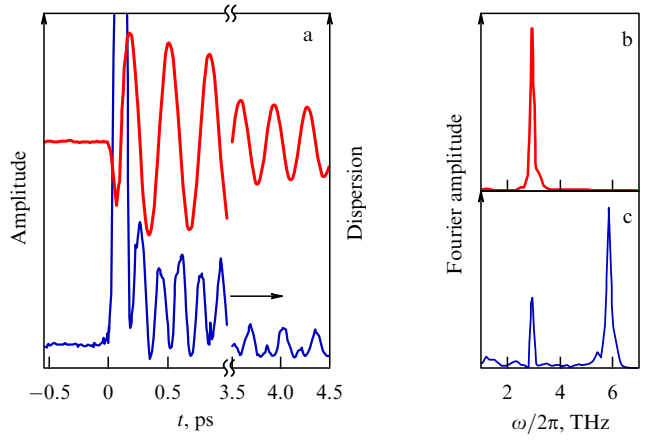

Figure 6. (a) Coherent amplitude and its dispersion for a Bi single crystal as a function of the phase angle, which is proportional to the time delay between the exciting and probe laser pulses. Panels (b) and (c) respectively show the Fourier transforms of the coherent amplitude and of its dispersion.

We note, however, that the noise spectrum components change somewhat in intensity, and the intensity ratio between the high-frequency and low-frequency components tends to increase [8].

The results of studying the statistical properties have not escaped criticism, either [81]; to summarize, it centers on the following: CPs, by whatever physical mechanism they are produced, cannot lead to the phenomenon of squeezing, which is always due to the nonlinear nature of the system and which is involved in none of the CP generation mechanisms. Indeed, the critics argued, quite correctly, that the experiments in Refs [49, 50, 52] say nothing of the zero fluctuation level, whose knowledge is needed to confirm the actuality of vacuum squeezing. Furthermore, they applied a somewhat modified scheme to measure noise [81] in Bi and GaAs, which led them to the discovery of time-independent noise. We must note, however, that the noise measurements in Ref. [81] used not the isotropic detection scheme but rather an anisotropic one, which does not involve the subtraction of noise at negative time delays. Moreover, the measurements for bismuth were made on a polycrystalline sample (this is the only possible explanation for the dominance of fully symmetric oscillations in the time-resolved response in the anisotropic detection approach) and, hence, along with the (identical) xx and yy polarization tensor components; the zz component, which greatly exceeds the basis components in magnitude, was also investigated. The presence of the second harmonic in the noise spectrum of the crystals studied was attributed in Ref. [81] to the uncontrollable deviation of the laser pulse repetition frequency, also known as jitter (this explanation had, in fact, already been proposed in Ref. [8]). It was shown, moreover, that the intensity of the second harmonic is proportional to the integration time of the synchronous detector. To find out definitely whether squeezed fluctuations exist in the case of the coherent one-phonon excitation, it is necessary to substantially modify the measurement scheme so as to gain access to a correlation function like $\langle A(t)\, A(t+\tau)\rangle$ instead of $\langle A(t)\rangle\langle A(t+\tau)\rangle$, i.e., to measure the coherent amplitude induced by the same laser pulse at two different time delays.

The experiments described above used the degenerate scheme, in which the excitation and probing light frequencies are the same. The nondegenerate scheme, with the carrier frequency of the probe pulse different from the pump frequency, provides additional information on lattice dynamics, especially in the case where the pump carrier frequency is in the X-ray range. Indeed, the X-ray radiation wavelength is about the size of the unit cell, which allows the monitoring and quantitative assessment of how optical pump pulses affect the interatomic distances.

There has been an obvious boom in experiments on ultrafast X-ray optics over the past decade [82], in large part due to the development of bright radiation sources and to the reduced duration of the X-ray pulse and its time jitter. Time-resolved X-ray experiments have allowed studying coherent lattice dynamics (in terms of both acoustic and optical modes) for a number of crystals with a signal-to-noise ratio close to the typical value for an optical probe [83]. A feature of the experiments that revealed the existence of squeezed phonon modes is the observation of the time evolution of the Debye–Waller factor for a number of reflexes in a bismuth single crystal. It is known that increasing the temperature decreases the peak intensity of Bragg diffraction, with the peak position and width remaining virtually unchanged. This decrease in

intensity, known as the Debye–Waller effect, depends exponentially on the mean square atomic displacements and determines the effect of phonons on the probability of processes that transfer momentum to the crystal as a whole without changing the state of the lattice. In a typical experiment, this results in a time-averaged reduction in the integral intensity of Bragg diffraction, which becomes more pronounced with increasing the temperature and for higher diffraction orders [84].

The group at the Paul Scherer Institute, Switzerland, used the Debye–Waller effect to measure the time dependence of the mean square displacements of bismuth atoms due to an ultrashort laser pulse [53]. In bismuth, an intensive ultrafast excitation is well known to generate coherent fully symmetric (A_g) and doubly degenerate (E_g) optical phonon modes [85, 86] (the higher the pumping intensity is, the lower the phonon frequency [85]). The laser wavelength used by the Swiss team to excite bismuth was larger than the lattice constant, and therefore an optical excitation transferred a small momentum to the crystal lattice [53]. The registration of the time response of the $[10\bar{1}]$ and $[11\bar{2}]$ Bragg reflexes revealed a strongly damped oscillation with a period of about 750 fs. It was also found that the oscillation amplitude for $[11\bar{2}]$ was about three times that for $[10\bar{1}]$ and that cooling the crystal to 170 K reduced the amplitude of the $[11\bar{2}]$ reflex by half. Because the reflexes under study correspond to the atomic displacements orthogonal to the trigonal [111] axis, the contribution from the fully symmetric phonons to the 750 fs oscillation was excluded. The idea of identifying the observed oscillations as doubly degenerate phonons was dismissed based on a comparison of frequencies: for an E_g phonon, the oscillation frequency at this excitation level corresponds to 1.6 THz (the period $T = 625$ fs).

The authors of Ref. [53] explain the observed response by assuming that the strongly damped oscillations are due to the acoustic modes softening instantly and uniformly over the entire Brillouin zone. Indeed, in the experiment, a laser took less than the period of the fastest lattice vibrations to promote a considerable fraction of electrons from the valence to the conduction band, thus instantly modifying the interatomic forces and hence changing the potential in which the atoms are located. As a result, the atoms start moving so as to be on average either somewhat closer to or farther from the equilibrium positions (with a broader or a squeezed momentum distribution, respectively), which corresponds to squeezed acoustic phonons. In the view of the authors of Ref. [53], the occurrence of squeezing is a consequence of the uniform acoustic mode softening (over all points of the Brillouin zone) predicted by the density functional theory simulations. A point of note here is that experiments on the ultrafast disordering of an InSb crystal [87] were also interpreted in the context of squeezed phonons due to a uniform softening of Debye phonons over the Brillouin zone; but no evidence for the oscillating Debye–Waller factor was observed in these experiments. It was reported, however, that when measuring the fluctuation properties of coherent optical phonons in an InSb crystal [88], its spectral mode with twice the SP frequency was found to exhibit noise.

The results of experiments that relied on the nondegenerate method and which used X-ray probe pulses to detect squeezed phonons have also been subjected to criticism [89], centered on the interpretation aspects and on deficiencies of the model used in the density functional method. It was noticed first [89] that the computation model in Ref. [52] used different chemical potentials for electrons and holes, thereby assuming that although the two types of carriers thermalize fast, there is no equilibrium between them. Then, after using the density functional method with a common chemical potential for the charge carriers, the authors of Ref. [89] came to the conclusion that the softening of acoustic modes occurs only in the vicinity of the X and M points of the Brillouin zone and cannot therefore be interpreted as evidence of the squeezing of lattice excitations. It should also be noted that earlier two-chemical-potential calculations of the coherent dynamics in bismuth [90] had not revealed the softening of optical modes, although optical mode softening observed in intensive pumping experiments was perfectly reproduced.

To summarize, based on the experimental data obtained and on the criticism presented, it is currently difficult to conclude which of the opposing sides is right, i.e., whether the vacuum squeezing of lattice excitations has in fact been, or is yet to be, implemented. There are many factors that have to be understood when seeking an interpretation, such as the generation mechanism of squeezed states, the interaction of these states among themselves and with the thermal bath, and the role of the electronic subsystem of the crystal; the individual contributions of those factors are very difficult to discern. Only further experiments and a detailed theoretical understanding of the effects mentioned above will provide final answers to these questions.

6. Coherent control of biphonons as a way to reveal nonclassical correlations. Entangled phonons

Entanglement, a phenomenon with no classical analog, was first demonstrated for electromagnetic field quanta [91]. Somewhat later, quantum correlations violating Bell's inequality [92] were found to exist for biphotons created in the spontaneous parametric scattering of light [93]. For crystal lattice excitations, the analog of a biphoton is a biphonon [94], a correlated quantum of the field of elastic deformations. It has been proved experimentally that a biphonon state can be produced by an ultrashort laser pulse in a crystal with a van Hove singularity in the density of lattice states: ZnTe (110), a typical crystal with this property, was reported in [95] to exhibit an acoustic biphonon when exposed to a 40 fs laser pulse. In Fig. 7, which shows both the coherent part of the lattice response and its Fourier transform, we see that the laser pulse creates a biphonon state in ZnTe, which has a somewhat higher energy than that of the acoustic overtone and which, for a single pulse excitation, decays with a typical time of 1.4 ps. Each component of the biphonon is in the superposition of the vacuum state and all the excited states of the one-phonon mode. The conservation principles that control the generation of lattice excitations require that these phonons interfere destructively, thus leading to two-phonon coherence. The phase correlation of pairs of phonons with equal but oppositely directed wave vectors can, by analogy with superconductivity, be called pairing — with the reservation, however, that unlike in superconductivity, the physical reason for pairing is here the pump pulse, which separates phonon pairs with the maximum instability increment from the phase chaos.

A biphonon state in ZnTe can be regarded as the superposition of the vacuum state and the first excited state that results from 'phonon localization' [96]. The variable that

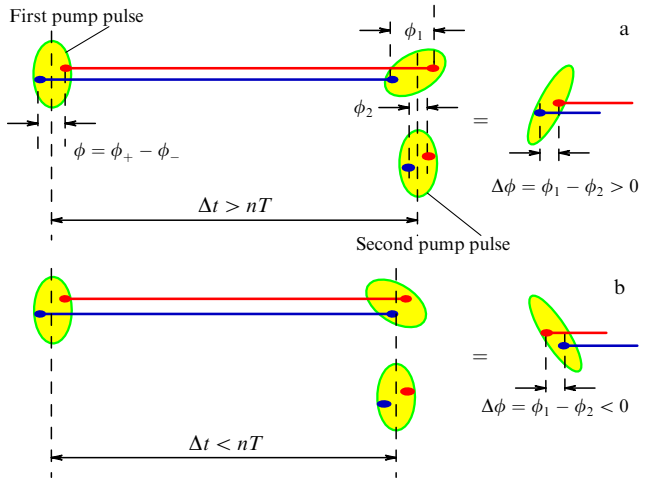

Figure 7. (Color online). Schematic representation of two-phonon interference and the definition of the internal biphonon phase in the cases where the control parameter is (a) larger or (b) smaller than a multiple of the biphonon period T ($\Delta t > nT$ or $\Delta t < nT$). Large green ellipses correspond to the phonon uncertainty contours; their orientation determines the biphonon phase. Small red and blue ellipses correspond to the 'halves' of the biphonon, each of which has its own phase. The difference between these phases corresponds to the internal phase ϕ.

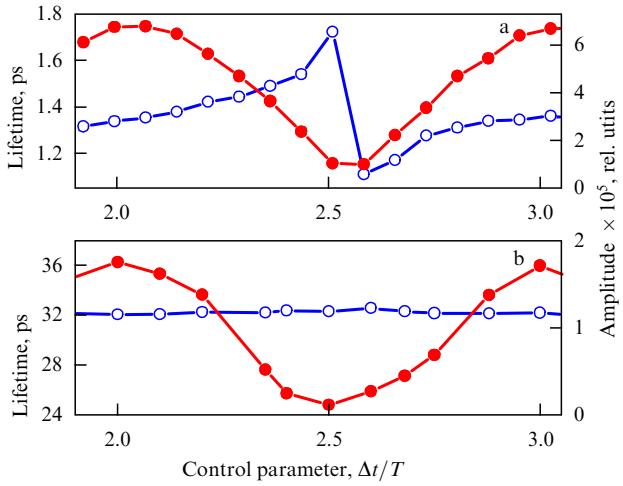

Figure 8. Results on the coherent control of (a) biphonons in ZnTe at room temperature and (b) coherent phonons in Bi at liquid helium temperature. Lines of dark dots (left scale): life time variation. Lines of light dots (right scale): oscillation amplitude as a function of the dimensionless control parameter $\Delta t/T$, where T is the period of the lattice excitation being studied.

localizes in such a transition is the 'distance' between phonons, and a biphonon can in this case be written as a two-mode squeezed state of the lattice [54],

$$|\alpha_+, \alpha_-, \zeta\rangle = \hat{D}_+(\alpha_+)\,\hat{D}_-(\alpha_-)\,\hat{S}_{+-}(\zeta)|\,\text{vacuum}\rangle\,.$$

Squeezing leads to correlations between the orthogonal quadratures of individual phonon modes, mixing the annihilation operators $\hat{a}_\pm$ of one mode with the creation operators $\hat{a}_\mp^\dagger$ of the other. Such a squeezed two-mode state is in many ways analogous to a thermal state, because both have the same relation between the number of phonons and the probability of finding n phonons in *any* of these two modes.

The well-defined phase of lattice excitations produced by ultrashort laser pulses provides the opportunity to influence the system in a number of ways to create a definite quantum state. The way to implement such influences is usually by the method of coherent control, whose key idea is to create superpositions of lattice states with certain phases. Varying these phases leads to either constructive or destructive interference, thereby enabling the control of the final state of the lattice. Such control in the case of CPs is implemented by two-pulse excitation in a modified pump–probe method [4].

Unlike the coherent control of one-phonon excitations, whose results are easily explained within the paradigm of classical interference [4, 97], controlling biphonons raises paradoxes that require a quantum mechanical treatment for their resolution [54]: because biphonons have two phases, one for a biphonon as a whole and the other for its components, the coherent control of biphonons is an acoustic analog of two-photon interference [98–102].

This was demonstrated in two-pump-pulse experiments [54] that investigated the superposition of two ensembles of biphonons produced at various time instants, as shown in Fig. 7. By varying the pump-to-pump time delay, it is possible to create a state with both its modes simultaneously excited or a vacuum state with both its acoustic modes deactivated. Such a two-phonon vacuum corresponds to the situation where lattice atoms are in their equilibrium positions (atomic displacement distribution is very narrow), but their kinetic energy is maximum, corresponding to a broad velocity distribution. The situation corresponding to a biphonon state is, on the contrary, where the atoms are at rest, are located at classical return points, and have a narrow velocity distribution (with their displacement distribution washed out).

A double pulse excitation can, in the first approximation, be represented as the sum of two interfering biphonon ensembles created at different time instants, with the control parameter, the pump-to-pump time delay, determining whether the interference is constructive or destructive. It can be seen from Fig. 8a that as this parameter varies, the phonon amplitude increases and decreases at the frequency of the biphonon. This amplitude modulation, while easily explained within the paradigm of classical interference, is accompanied by modulation of the excitation lifetime, an effect that is impossible for classical interference.

Experiments show that the biphonon lifetime can vary in either a correlated or uncorrelated manner with the biphonon amplitude. Furthermore, the process of decreasing the lifetime (with the decrease factor reaching $1.5 \pm 0.2 \approx \sqrt{2}$) is faster than the increasing process, which is more monotonic and involves a larger change in the control parameter. This sharp increase is highly reminiscent of 'entanglement sudden death', an effect that was predicted relatively recently [103] and soon implemented experimentally [104].

The variation of the biphonon lifetime under coherent control is in sharp contrast to the control-parameter-independent behavior of the lifetime of single-phonon coherent excitations. For fully symmetric phonons in bismuth in the linear excitation regime, when the lifetime is independent of the pumping power [97, 105, 106], varying the control parameter over a wide range leaves the CP lifetime unchanged, although the resulting amplitude is modulated with the frequency of the phonon, as shown in Fig. 8b.

If the individual acoustic phonons that form biphonons in ZnTe are assumed to have lifetimes independent of the control parameter, then it is clear that the way the correla-

tions weaken with time is determined not so much by the initial degree of correlation (or entanglement) as by the specific type of the state. The amplitude of the biphonon is proportional to the real part of the squeezing parameter, where its lifetime determines how long the components of the biphonon state remain correlated: the longer the lifetime is, the stronger the correlation. It is currently unclear whether these correlations in ZnTe are classical or quantum, i.e., whether Bell's inequalities are violated.

The interpretation of the coherent control of the biphonon is complicated by the fact that, experimentally, interference occurs between identical, spatially inseparable lattice excitations, making it impossible to find whether nonlocal correlations exist that lead to 'action at a distance'. In principle, an experiment can be suggested to separate classical from quantum correlations. By modifying a double-pulse excitation such that the first pump pulse with a wide cross section creates a biphonon and the second, narrow one performs coherent control and focusing the narrow probe pulse into a crystal region not subject to the second pump pulse, it is easy to distinguish entanglement from classical correlations. Here, in the first case, we would see the result of coherent control (action at a distance) at once, whereas in the second case, the oscillations would not be perturbed by the second pump pulse (or the result of coherent control would manifest itself after some time delay).

We take a more detailed look at the way coherent control is involved in the experiment in Ref. [54]. The first pump pulse creates a biphonon state, which evolves freely, until after a time Δt it is overlapped by a second state, due to the repeated pump pulse. The phase difference between these two composite states is determined by the pulse separation Δt. The internal phase ϕ_i of each of the biphonons, which is defined as the phase difference between its components, has no classical analog. For $\Delta t = nT$, where T is the biphonon period, the difference of the internal phases is equal to zero, $\Delta\phi = \phi_1 - \phi_2 \equiv 0$, i.e., any pump pulse creates a biphonon with the same internal phase. This corresponds to the interference of biphonons with the same orientation of uncertainty contours. However, for $\Delta t \neq nT$, with the uncertainty ellipses oriented differently, the internal phase difference $\Delta\phi = \Delta t - nT$ can be positive or negative, depending on whether n is even or odd. This internal phase asymmetry, determined by the sign of the phase difference, $\operatorname{sgn}\Delta\phi$, is responsible for the sharp change in the lifetime for $\Delta t/T = (2n-1)/2$. For $\Delta t/T < (2n-1)/2$, lattice excitations behave as a set of independent anharmonic oscillators, for each of which the lifetime anticorrelates with the biphonon amplitude. For $\Delta t/T > (2n-1)/2$, lattice excitations can be reasonably regarded as a set of coupled harmonic oscillators with lifetimes controlled by the strength of the coupling (dispersion), which is in turn proportional to the phonon amplitude. That is, for control parameter values $\Delta t/T < (2n-1)/2$ and $\Delta t/T > (2n-1)/2$, the lattice system is respectively described by the Einstein and Debye models. If we were able to measure the statistics of biphonon components, i.e., the statistics of each of its halves, then such a measurement would demonstrate a bunching for $\Delta t/T < (2n-1)/2$ and an antibunching for $\Delta t/T > (2n-1)/2$, each of which should change to Poisson's statistic (for respectively even and odd $\Delta t/T$). Thus, for $\Delta t/T < (2n-1)/2$, when the energy is localized on lattice atoms, lattice excitations behave as a system of particles (Einstein's model), whereas for $\Delta t/T > (2n-1)/2$, when energy is delocalized, they demonstrate wave properties (Debye model). We note that in the cases $\Delta t/T < (2n-1)/2$ and $\Delta t/T > (2n-1)/2$, the coupling between the biphonon components is respectively controlled by amplitude and phase fluctuations. Only at the boundary, where the amplitude and phase fluctuations act together, does the lifetime show a sharp change because the fluctuations influence the localized and distributed coherences differently.

It can be conjectured that a higher amount of phonon squeezing should lead to a stronger correlation between the phonons. However, it can be seen from Fig. 8a that the lifetime has its extrema near the minima of the biphonon amplitude, where the squeezing is maximum. This feature of the biphonon lifetime reflects the particle–wave duality and, together with the regular behavior of the biphonon amplitude, shows that the relation between biphonon squeezing and entanglement may be quite complex, because these effects are of different orders. Squeezing is a second-order effect determined by amplitude–amplitude correlations (quadratic in ladder operators) that are controlled by single-phonon interference; entanglement is a fourth-order effect dependent on intensity–intensity correlations (biquadratic in ladder operators).

The difference between the behavior of the biphonon amplitude and lifetime can be understood by noting that squeezing influences the diagonal elements of the individual acoustic phonon mode, $\langle \hat{a}_{\pm}^{\dagger}\hat{a}_{\pm}\rangle = |\alpha_{\pm}|^2 + \sinh^2 s$ and the nondiagonal intermode elements $\langle \hat{a}_{+}\hat{a}_{-}\rangle = \langle \hat{a}_{-}\hat{a}_{+}\rangle = \alpha_{+}\alpha_{-} - \exp(\mathrm{i}\theta)\sinh s\cosh s$ [105]. Therefore, at large degrees of squeezing, the population of each of the biphonon 'halves' can only increase, whereas their entanglement can increase or decrease. Roughly speaking, two-phonon interference modulates both the phase (θ) and the amplitude (s) of the squeezing factor, whereas single-phonon interference is only responsible for the modulation of the amplitude. Thus, similarly to the case of photons [100–103], one-phonon interference and two-phonon interference are respectively controlled by coherence and entanglement. Importantly, one-phonon interference (i.e., that of the halves) tends to localize phonons in the phase space, whereas two-phonon interference (that of biphonons) affects the overlapping of the wave functions of the 'halves'.

Depending on the value of the control parameter, coherent control demonstrates the possibility of changing from regime to regime because two-phonon interference oscillates at twice the frequency of the one-phonon interference. This time dependence difference is convincingly supported by the fact [54] that for control parameter values that are odd multiples of the biphonon period, the oscillation spectrum, along with the biphonon line, has an acoustic overtone from a noncoupled pair of acoustic phonons. This is illustrated in Fig. 9, which shows the wavelet-transformed time-resolved response and in which we clearly see the contribution of 3.2 THz oscillations, which corresponds to the coherent excitation of an uncoupled pair of acoustic phonons, i.e., to an acoustic overtone. It is clear that for control parameter values $\Delta t = (n + 1/2)\,T$, two-phonon interference is destructive (minimum biphonon amplitude), and one-phonon interference is, on the contrary, constructive. We note that it is totally untenable to argue that the overtone also exists for other values of the control parameter and is not seen only because it is masked by the large value of the biphonon amplitude; even for a small biphonon amplitude, realized in the case of a weak one-

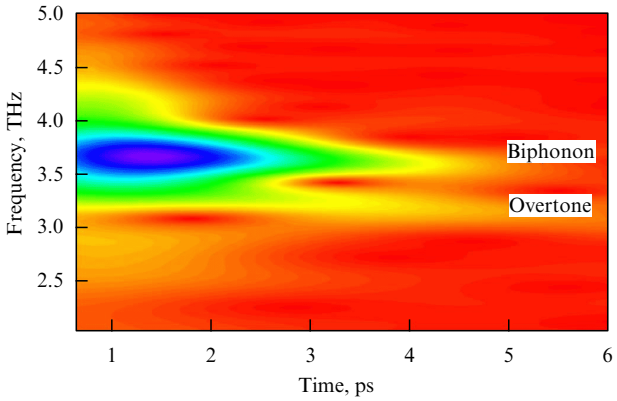

Figure 9. (Color online). Wavelet-transformed results of coherent control in Zn Te with the control parameter $\Delta t = 2.5T$, where T is the biphonon period. The figure illustrates the coexistence of coupled and uncoupled pairs of acoustic phonons. Blue (red) color corresponds to the maximum (minimum) signal amplitude.

pulse excitation, uncoupled pairs of acoustic phonons are not registered.

As the phase-space distribution broadens under coherent control, the one-phonon coherence decreases and entanglement, on the contrary, increases. Hence, the one- and two-phonon interferences respectively decrease and increase in intensity. Because coherence and entanglement depend in opposite senses on the state separability, the distribution width acts oppositely in determining the intensities of one- and two-phonon interferences. Whereas a broad distribution corresponds to strongly entangled, weakly coherent phonons, a narrow distribution corresponds to highly coherent, weakly entangled phonons.

We note that in their experiments discussed above, the authors of Ref. [54] failed to measure squeezing and entanglement quantitatively, and what they did at that stage was simply to suggest that the squeezing is proportional to the amplitude and entanglement is proportional to the lifetime of the biphonon.

Nevertheless, the attempt to describe a (squeezed and correlated [54]) biphonon excitation state of the ZnTe lattice as resulting from the excitation of two classical transverse waves is doomed. The waves turn out to be entangled with respect to the polarization variable. Thus, every transverse phonon has two polarization eigenstates, a vertical $|\mathrm{V}\rangle$ or a horizontal $|\mathrm{H}\rangle$ one. Because orthogonally polarized phonons cannot interfere, the absence of oscillations with the frequency of a TA phonon in the time-resolved response indicates that the biphonon components have the same polarization. Hence, the polarization state of the excited lattice created by an ultrashort laser pulse can be written in the form $|\varphi\rangle_{\pm} = 1/\sqrt{2}\big(|\mathrm{VV}\rangle \pm |\mathrm{HH}\rangle\big)$. It is easy to show that $|\varphi\rangle_{\pm} \neq |\varphi\rangle_{+}|\varphi\rangle_{-}$ for any wave functions of the vertically ($|\mathrm{V}\rangle$) and horizontally ($|\mathrm{H}\rangle$) polarized states, i.e., the biphonon wave function does not factor. Because phonons are generated in pairs simultaneously, the population numbers of the polarization modes are fully correlated (i.e., one of the Stokes parameters is zero [97]), whereas their phases are anticorrelated. The absence of the polarization basis for each of the acoustic modes suggests that the sound produced by an ultrashort laser pulse is either 'scalar' or has a hidden polarization. In this context, biphonons are in many respects analogous to nonclassical states of the electromagnetic field, for which it is known that two-photon light in maximally entangled Bell states is nonpolarized from the standpoint of classical optics [107]. According to the theory in [108], 'scalar polarization light' exists, a nonclassical state with an even number of photons, which is characterized by two-photon squeezing with respect to any pair of orthogonal polarization modes. Indeed, such scalar polarization light has already been realized experimentally [109].

An international research group led by Ian Walmsley at Oxford was the first to provide experimental evidence that a lattice can be in a state violating the Bell inequalities. In the experiment, phonons excited in the process of spontaneous Raman scattering in two spatially separated diamonds were brought to an entangled state [110]. The optical phonons that exhibited entanglement were of the symmetry $\Gamma_{25'}(\mathrm{F_{2g}})$ and had a frequency of 40 THz. To prove entanglement, one of its quantitative characteristics, concurrence, was used. The entanglement of $\Gamma_{25'}$ phonons followed from the positive sign of concurrence; the value of this characteristic, $(5.2 \pm 2.6) \times 10^{-6}$, was established with a reliability above 98%. Because phonon counting detectors are currently nonexistent, the authors of Ref. [111] assumed that phonon creation/annihilation events are local and inferred the entanglement of lattice states from the entanglement of Stokes and anti-Stokes photons, whose interference pattern visibility was close to that of maximally entangled Bell states.

The crystals used in the Oxford experiment (to which we refer as 'upper' and 'lower') were 3 mm in size and 15 cm apart; the experiment was conducted at room temperature. The high energy of the phonons studied (about 40 THz, or 1332 cm^{-1}) ensured that the lattice of both diamonds was in the ground (vacuum) state prior to excitation. The pump laser pulses (the duration 60 fs, the energy 1.5 eV, the repetition rate 82 MHz) passed through a symmetric beamsplitter and arrived at the two crystals via different optical paths, and then, after passing through the crystals, merged into a single pulse, thereby 'erasing' information about the path the photon took (Fig. 10a). Each of the pump photons could arrive at either the upper or the lower crystal after leaving the beamsplitter. According to the laws of quantum mechanics, it is impossible to predict before the measurement which way the photon takes, the reason being that it is in a superposition of its two possible states. If photons obeyed classical laws, then, after leaving the beamsplitter, they would move up or down, but by no means in both directions simultaneously. When, upon leaving the beamsplitter, a photon enters the diamond, part of its energy can be absorbed to produce a phonon in the crystal lattice. Because phonons also behave as quasiparticles, it follows that the two diamonds that have absorbed a photon coming from the beamsplitter share this single phonon, and are therefore entangled. According to classical thinking, the phonon is in the upper or the lower diamond, whereas according to quantum mechanics, it is 'smeared out' over the two. In the process of stimulated Raman scattering, the absorbed photon is re-emitted at a lower frequency, and this 'reddened' Stokes photon signals the achievement of an entangled state. However, because the output response of a single photon detector does not identify the exact crystal through which the photon passed, the excited phonons in one of the crystals are quantum-correlated with those in the second. In other words, once the photon is absorbed, the atoms of *both* crystals are set into motion. To see this, a probe pulse polarized orthogonally to the pump pulse was used, which again was sent with a delay of 350 fs

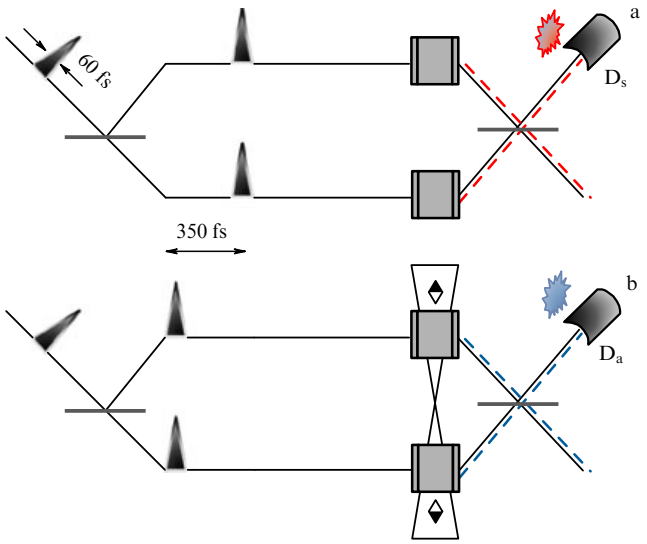

Figure 10. Schematic experiment to (a) create and (b) register the entanglement of diamond phonons. Panel (a): on passing through the beamsplitter, a photon creates a phonon in an entangled state in one of the diamonds (which is marked by a rectangular box), which is signaled by a Stokes photon in detector D_s. Then a (350-fs-delayed) orthogonally polarized pulse [panel (b)] eliminates the photon and the anti-Stokes photon is registered by detector D_s. The manifestation of an entangled state is the visibility of the interference pattern of anti-Stokes photons during the registration of coincidences in detecting Stokes and anti-Stokes photons, the visibility being measured by rotating the polarization of these photons.

through a symmetric beamsplitter onto two diamonds at once. When encountering a phonon, such a probe photon increases in energy (transforms into a 'blue' anti-Stokes photon) and is directed to two detectors via a system of beamsplitters (quarter- and half-wavelength plates). The fact that it is not known in which diamond the phonon is located allows distinguishing the classical state of the two crystals from their quantum (entangled) state. Classically, after sending the probe pulse, the appearance of the blue anti-Stokes photon at the output of the system should be registered with the same probability by both detectors; but if an object is described by the laws of the quantum world, the photon should be registered by one and only one designated detector, because the appearance of a blue anti-Stokes photon should correlate with the appearance of a red Stokes phonon.

It is precisely this nonclassical correlation that the Oxford physicists observed. When a phonon is annihilated and hence atomic motion ceases in one of the crystals, the atoms in the other also suddenly cease moving, although the crystals are spatially apart and cannot interact. And although such entanglement creation and registration experiments lasted a mere 0.35 ps (phonons in diamond are short lived at room temperature), this suffices, if not to store quantum data, then at least to perform quantum computations.

Thus, the existence of quantum entanglement is confirmed by measuring correlations between the polarization states of Stokes and anti-Stokes Raman scattering photons. The fact that spatially separated diamonds were entangled at room temperature is important because it indicates that such an entangled state can persist in ordinary environments in macroscopic solids, which, according to the authors of Ref. [110], makes these objects the basis for developing cryogen-free quantum computers. The THz-fast read/write cycle using a diamond optical phonon [111] amply illustrates the potential of the field.

It may seem at first glance that a phonon, which is an indivisible quantum of lattice vibrations, cannot be in an entangled state, because entanglement presupposes the existence of at least two quantum mechanical objects. This follows from the fact that in the secondary quantization representation, the state $|\phi\rangle_{A,B} = |0\rangle_A|1\rangle_B + |1\rangle_A|0\rangle_B$, where $|0\rangle_{A,B}$ is a state without a phonon and $|1\rangle_{A,B}$ is a state with one phonon in modes A and B, is factorable, $|\phi\rangle_{A,B} = (\hat{a}_A^\dagger + \hat{a}_B^\dagger)\,|0,0\rangle$, and hence is not entangled. However, it is shown theoretically [112] that when the modes A and B are spatially separated, the state $|\phi\rangle_{A,B}$ does exhibit entanglement features. Unlike the biphonon state entanglement, which can be realized between two phonons and the vacuum, entanglement for the state $|\phi\rangle_{A,B}$ is realized for two spatially separated modes, A and B. Such entanglement was also realized for neodymium ions implanted into two yttrium orthosilicate crystals [113].

A point of special note is that the phonons studied in Ref. [110], unlike those in Refs [42, 48, 54], were not in the Glauber state, because the 20 fs pulses have a spectrum of insufficient width to create superpositions of states 40 THz apart in energy. Because of the short vibration period of diamond carbon atoms ($T = 25$ fs), much shorter laser pulses are needed to cause the lattice atoms to move coherently. This was previously demonstrated in [114] using 3.2 eV, 10 fs laser pulses.

7. Conclusion

Two decades of theoretical research have provided evidence that under certain conditions, it is possible to prepare ensembles of phonons in essentially nonclassical states. It has been reliably established by the methods of dynamic spectroscopy that coherent and, possibly, squeezed and entangled states of a crystal lattice can be created using ultrashort laser pulses. Importantly, although in most cases coherent phonons do not require a quantum mechanical treatment for their description, crystal lattice states that have no classical analog nevertheless exist. They are made possible by the quantum nature of the phonon, which is simultaneously a particle and a wave. Although there are currently various quasiparticles for which crystal excitations have been realized [115–117], it is experiments with lattice excitations that have developed most.

Questions concerning squeezed or entangled crystal states are only beginning to be addressed in detail, both experimentally and theoretically. With lattices in such nonclassical states, crystals may exhibit some special features, both in their properties and in the way they interact with an electromagnetic field, which can significantly change the physics of acoustic and optical phonon modes. Due to their nonclassical properties, squeezed and entangled lattice states will undoubtedly become a focus of intense future research, both fundamentally theoretical (quantum theory as a description of physical reality) and experimental and applied (the prospect of creating the element base for quantum computers and of solving information transmission/processing problems).

We note that the increased interest in nonclassical fields is in part due to their unique correlation properties and is also stimulated by attempts to use them for applied purposes (for example, in applications of quantum information theory).

From the standpoint of their potential in this particular field, optical fields are not the best choice, because the available materials are only slightly nonlinear. It is nonlinearity that underlies the interaction fundamentally necessary for performing quantum logic operations, and the high nonlinearity of elastic deformation fields is simply bound to result in a variety of applications for them.

Future research will undoubtedly show that the small magnitude of quantum correlations in our macroscopic world is an illusory reason for neglecting them and that, very importantly, these correlations are so unusual in their properties that under certain conditions they can 'outweigh' even the strongest classical correlations.

A potentially very important fact is that it may be possible to control exactly how much a phonon is coherent, squeezed, or entangled. To do this effectively requires, as a top priority for joint efforts by experimenters and theoreticians, finding (i) the microscopic mechanism for generating squeezed and entangled phonons and (ii) how both the properties of the electronic (and other) subsystem and the parameters of the ultrashort laser pulse (chirp, duration, energy, etc.) affect the lattice state created by the pulse. Particular theoretical attention should be given to establishing a relation between the squeezing, coherence, and entanglement of phonon modes.

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