Phase-dependent noise in femtosecond pump-probe experiments on Bi and GaAs

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Femtosecond laser pulses generate in semimetal Bi and semiconductor GaAs the phonon field that, in addition to being coherent, demonstrates phase-dependent fluctuation properties. The variance of the coherent phonon amplitude, representative of fluctuation properties, is a function of time delay, and the oscillations of the phonon amplitude and its variance occur at different frequencies.

Any quantum field fluctuates in such a way that the observables describing the field are subject to a stochastic indeterminacy. This quantum indeterminacy sanctioned by the Heisenberg uncertainty principle puts severe limitations on the precision of any measurement. The Heisenberg uncertainty principle has, however, a built-in degree of freedom. One can squeeze the variance of one observable provided that the variance of the other becomes stretched. The squeezing for a photon field is a mature field with many experimental results and theoretical models.[1] More recently, various schemes to squeeze material waves have been proposed.[2-5]

The fact that the solids subjected to femtosecond laser pulses can exhibit lattice vibrations with a high degree of temporal and spatial coherence is well established.[6] These lattice vibrations, usually referred to as coherent phonons, have been reported in a great variety of solids, and increasing attention is now being turned towards not only the mere observation but also the understanding of physics that occurs at this short time scale. In an attempt to make clear that lattice coherence is not the only consequence of the excitation with ultrashort pulses, the pump-probe experiments carried out on potassium tantalate and strontium titanate have provided the example of the squeezing for crystal vibrations.[7] Before that, the vibrational squeezing had been reported for a sodium dimer.[8] Guided by these developments and noticing that a unique signature of squeezing is the phase dependence of noise properties,[1,9] a feature that can be experimentally explored, we have decided to approach the problem of phonon squeezing in ultrafast pump-probe experiment. Since the magnitude of femtosecond reflectivity change is largest for semimetal Bi and semiconductor GaAs,[10,11] we have performed the pump-probe experiments on the two systems, both in the form of optically thick films, to investigate *fluctuation properties* of the state created by femtosecond pulse.

For the excitation of coherent phonons we used an all-solid state laser system. The system comprised a frequency-doubled Nd:vanadate laser (''Verdi'') and a Ti:sapphire pulse mode-locked laser (''Mira-seed''). The first provided single-frequency green output at 532 nm to pump the second laser operating at 800 nm. The mode-locked laser produced a 78-MHz train of 30-fs pulses. The pulses were divided into the high-intensity pump and low-intensity probe pulses polarized perpendicular to each other to reduce the scattering of pump into the detector. The average power of the pump and probe pulses was, respectively, 60 and 1.2 mW, if not stated otherwise. Both the pump and probe beams were focused by a single lens into a spot diameter of 50 μm. By varying the time delay between the pump and probe, we were able to trace the transient reflectivity. For detection we employed a phase-sensitive scheme modulating the pump beam at 550 Hz with a shaker. All measurements were performed at room temperature.

Figure 1(a) shows a typical pump-probe result for bismuth. A pronounced oscillatory feature occurs at 2.93 THz as follows from the Fourier transform shown in the inset. The phonon responsible for the oscillations is fully symmetric.[10] The coherent amplitude measured either directly from the oscillatory signal (not shown) or from the parameters of the Fourier transform depends linearly on pump power, whereas the decay time is independent of the pump laser intensity, see Fig. 1(b). Now we address the question of how the coherent amplitude fluctuates at different time delays. To measure these fluctuations, we repeatedly ($\sim$100 times) recorded a part of the oscillatory waveform shown in Fig. 1(a) for the time delays ranging from -1 to 6 ps. From the data set we calculated the mean, $m=\langle A\rangle$, and the variance, $\sigma^2=\langle(A-m)^2\rangle$, of the oscillatory signal A at each time delay (the brackets in the expressions denote ensemble average). The obtained results are summarized in Fig. 2. A similar set of results was also obtained on GaAs for which the oscillatory signal and its variance at various delay times as well as their Fourier transforms are shown in Fig. 3. The coherent oscillations in transient reflectivity now occur with the frequency 8.54 THz and are due to longitudinal optical phonon.[11]

There are two striking features to the data presented in Figs. 2 and 3. First, the fluctuations of coherent vibrations

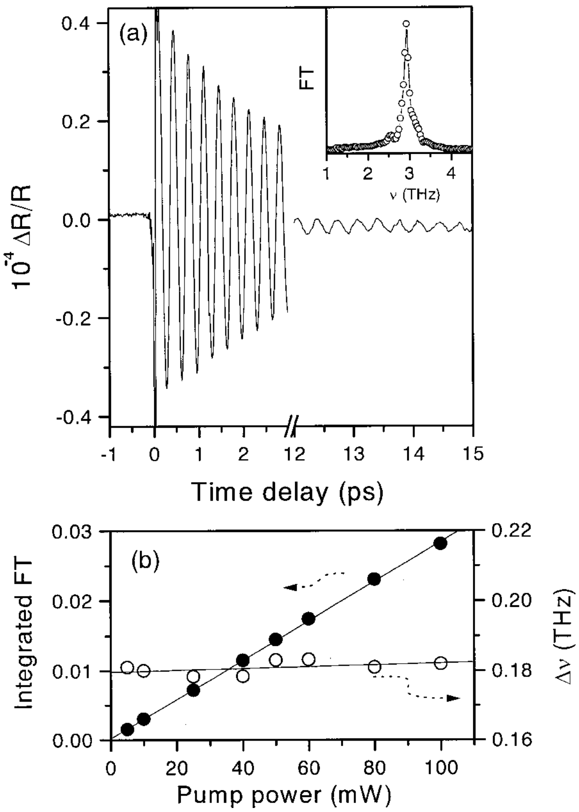

FIG. 1. (a) Transient fractional reflectivity vs time delay obtained for bismuth. The inset shows Fourier transform of the oscillatory trace. (b) The pump dependence of coherent amplitude (closed circles) and its decay time (open circles).

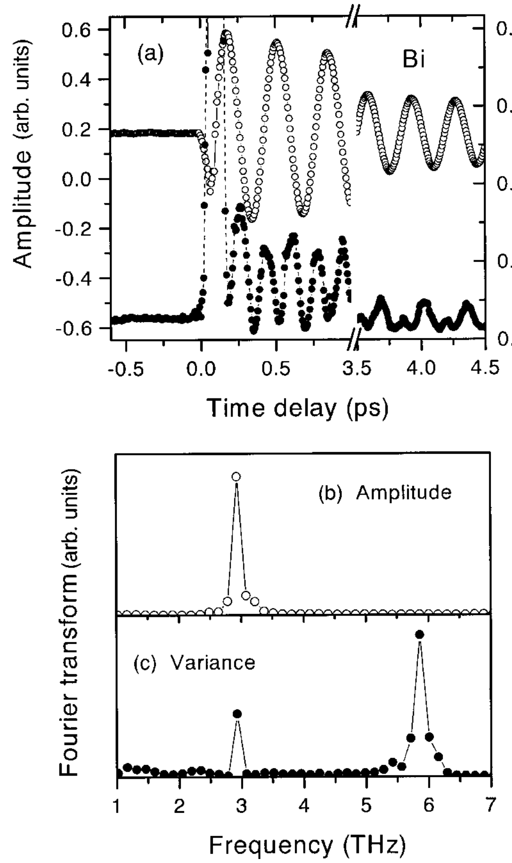

FIG. 2. (a) The mean (open circles, left-hand scale) and the variance (dashed line, right-hand scale) of oscillatory waveform in bismuth as a function of time delay. (b) Fourier transforms of the mean and variance.

are phase dependent as evidenced by the variance plotted against delay time. This phase dependence occurs for positive time delays only, whereas the variance for negative delays is independent of time. Second, the frequencies of the Fourier transform of coherent amplitude and its variance are different; the latter oscillates at doubled frequency as compared to the coherent amplitude.

We attach special importance to the first feature since the variance being a measure of noise is usually not phase-dependent for any of the classical or quantum states except the squeezed one.[9] The possible explanation for the phase-dependent noise that oscillates with doubled frequency of the phonon can be found provided the phonon field we are dealing with is coherent, that is, the product of variances for two conjugate variables satisfies the equality sign in the Heisenberg relation. However, our technique lacks the reference level associated with zero-point fluctuation and we have to use another property of coherent states to check the coherence. We will make use of the following property: for the coherent state, the ratio of variance to amplitude is a constant independent of the amplitude magnitude. This is the property of Poisson distribution with equal mean and variance and physically means the fluctuations are independent of excitation level.[9] The excitation level in pump-probe experiment is proportional to pump power, and its measure is the integrated intensity of the Fourier transform of coherent amplitude. On the other hand, the fluctuations of coherent amplitude can be related to its decay time and estimated from the linewidth of the Fourier transform. Figure 1(b) shows that the linewidth is independent of excitation level. This linewidth independence proves the coherence of phonon field. Given the coherent state of the lattice, the phase-dependent noise is suggestive of squeezing for the phonons created in pump-probe experiment.

To get a physical understanding of what is actually measured in our experiment and what are the implications, let us recall that the coherent state is usually taken as a minimum-uncertainty state, and its fluctuation properties are akin to those of a vacuum state. Let us emphasize that this is a crucial point for our conclusion since our technique lacks a readily defined benchmark reference level corresponding to the usual shot noise and therefore we have to rely on qualitative features that distinguish a thermal, coherent, and squeezed state. To observe the phonon squeezing we must select active phonon quadrature from the complex oscillation, which is done by our phase-sensitive detection. As shown in Fig. 4, the detection axis remains still in the Schrödinger picture, but it rotates in the phasor plane for the Heisenberg picture. Actually, the phase angle of our detector is fixed, but we measure the coherent state with different phase chosen by time delay. Analysis of the detection scheme shows that only the states with a well-defined phase (coherent and/or squeezed) contribute to the signal measured in time domain. For a coherent state the wave packet oscillates with the same shape, therefore the projection onto the detection axis yields a coherent amplitude fluctuating identically for any phase angle. Thermal, number, and vacuum

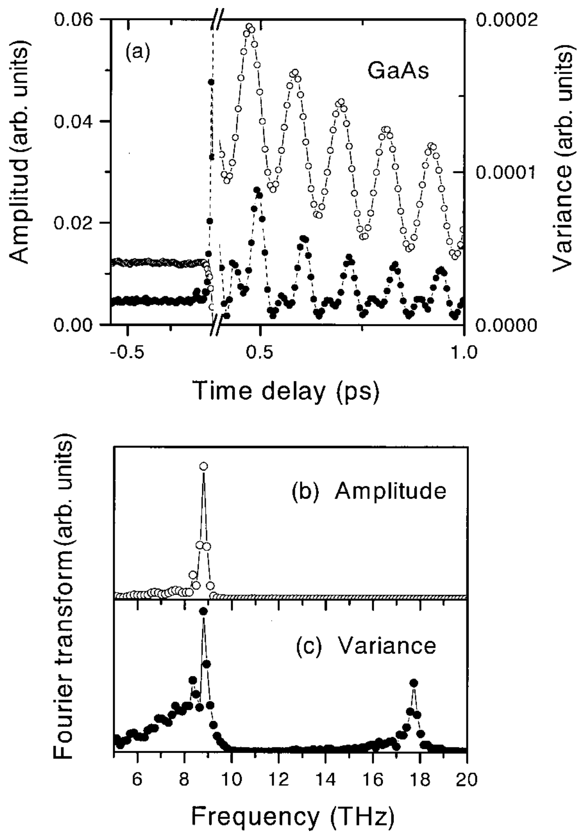

FIG. 3. (a) The mean (open circles, left-hand scale) and the variance (dashed line, right-hand scale) of oscillatory waveform in GaAs as a function of time delay. (b) Fourier transforms of the mean and variance.

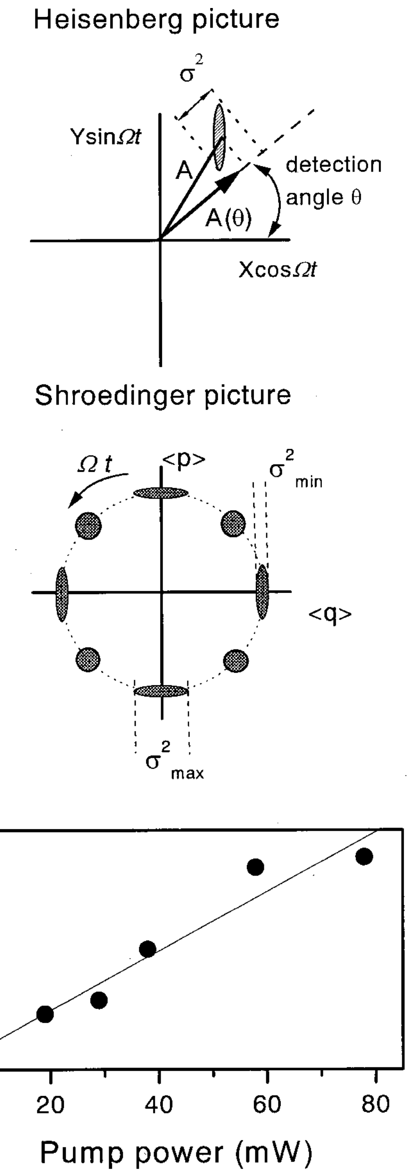

FIG. 4. Schematic diagram of the uncertainty area for squeezed states and of measuring scheme in the Heisenberg and Schrödinger pictures. The bottom panel depicts the pump dependence of variance in Bi film.

states exhibit similar time-independent noise properties, therefore their contribution to the Fourier-transformed variance is limited by the zero-frequency contribution. The dynamics of the squeezed state is different and better illustrated in the Schrödinger picture. The squeezed wave packet in Fig. 4 follows the large circle with angular frequency ω, but the shape oscillates with angular frequency 2ω as the wave packet has the same shape when it has gone one-half period. In this case the uncertainty contour changes shape with a quadrupole motion, and the noise measured as a projection on a detection line is phase-dependent with the frequency twice the frequency of the coherent amplitude.

A very characteristic feature of the variance observed in our experiments should be mentioned. For every cycle of the coherent amplitude there are two peaks with *distinct* heights in the variance. This additional modulation is present in both samples, and is a function of pump power. For a lower pump power, the additional modulation was more pronounced and the ratio between the spectral weight at doubled frequency and that at fundamental frequency was smaller. By comparing with other materials where the noise properties of coherent phonons have been studied,[12,13] we observe that the modulation depends on the coherent amplitude, which is also supported by pump dependence, and on the coherent phonon frequency (the larger the frequency, the stronger the additional modulation for all other experimental parameters fixed). Note that the theoretical study of generalized geometric states has predicted a similar anisotropy for the Wigner function in phasor space.[14]

For both Bi and GaAs, the increase of pump power does not affect the variance at negative time delays but leads to more pronounced oscillations at positive delays. To be more precise, the oscillations in variance depend almost linearly on pump power, which can be inferred for Bi from the parameter $\gamma=(\sigma^2_{\max}-\sigma^2_{\min})/(\sigma^2_{\max}+\sigma^2_{\min})$ as a function of pump power plotted in Fig. 4. It is important that the oscillations in variance depend on coherent amplitude magnitude and not on overall signal level, which was checked by unbalancing the detection scheme. For Bi film, the phase-dependent pattern was observed when the temperature was decreased to liquid-helium temperature and as well as when the shaker was changed to a chopper modulating the pump beam at 2 KHz.

As far as the mechanism of phonon squeezing is concerned, we would like to point out that the so far suggested mechanisms could not be straightforwardly applied to our case. It is clear that our approach differs significantly from that of Garrett *et al.*,[7] who use stimulated two-phonon scattering and, thus, measure a coherent amplitude supposedly directly proportional to the noise. Most promising seems to be the suggestions made in Refs. 2–4, but they too will require some modification. We conjecture that the mechanism responsible for the generation of coherent phonons can be related to that for the squeezing. Both coherence and squeezing can be described if the coherent phonons are generated through a (stimulated) Raman-like process since the Raman effect is a parametric process that, as we know from optics, is capable of generating squeezed states.[1,3] On the other hand, if coherent phonons are generated by a displacive excitation mechanism,[10] the phase-dependent noise is suggestive of minimum uncertainty squeezing, since the phonon state in this case is the displaced vacuum that satisfies the equality sign in the Heisenberg relation. The phonon state created by the displacive mechanism can be squeezed because the ground-state wave function of an oscillator placed in a potential with a lower frequency is a squeezed state.[15] If the mechanism for coherent phonons is Bose-Einstein condensation, as suggested for GaAs,[16] the squeezing can probably be found since the same Bogolubov transformation is technically responsible for superfluidity and squeezing.[17,18] We also note that similar phase-dependent noise has been observed in high-T_c superconductors $YBa_2Cu_3O_{7-x}$, semimetal Sb, and semiconductor InSb.[12,13] This suggests that such behavior may be intrinsic for coherent phonons and require a better understanding.

In conclusion, we have presented the experimental data on fluctuation properties of the coherent phonons created in pump-probe experiments on Bi and GaAs. We suggest that the phase-dependent noise observed in the experiments indicates the elliptical shape for the uncertainty contour.

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