Collapse and revival of large-amplitude coherent phonons: polarization interference versus quantum beats

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Abstract: We report femtosecond time-resolved measurements of lattice dynamics in Bi made at helium temperature over a wide range of excitation levels. We demonstrate that the collapse/revival of large-amplitude A_{1g} coherent phonons is a polarization interference arising due to laterally inhomogeneous excitation

1. Introduction

Ultrafast laser pulses, with durations shorter than typical nuclear vibrations, are widely used to prepare the specific condensed matter states that would be otherwise inaccessible. Moreover, the same pulses can be used to study the evolution of the created state in real time. One of the effects of a ultrashort laser pulse incident on a crystal is to induce nonstationary lattice states (phonon coherence) [1]. Bismuth, the most studied crystal in the time-domain, exhibits pronounced oscillations of two symmetries (A_{1g} and E_g) in transient optical reflectivity following ultrashort pulse laser excitation [1]. The amplitude of the oscillations, and presumably the atomic displacements, increases with excitation strength. It should be noted that phenomena under high laser fluence excitation have become a popular topic in recent years due to the capability of intense laser radiation to produce high density plasma and large amplitude lattice distortions, both capable to trigger photo-induced thermodinamical (or quantum) phase transitions [2]. Increasing the excitation strength in Bi towards the Lindemann stability limit, it was found that at helium temperatures the amplitude of coherent phonon oscillations is fading away and, at a later time, can reappear [3]. This phenomenon, initially explained as a quantum mechanical effect, had been referred to as "amplitude collapse and revival" [3] and later was detected in Sb (A_{1g} and E_g modes) and graphite (E_g mode) [1]. The observation of this phenomenon started a discussion as to whether the coherent phonons behave classically as usually assumed, or some quantum effects might contribute to detected signals [4]. However, the results of quantum dynamical simulation, carried out as a function of system size, suggested that the observed collapse/revival in photoexcited bismuth [3] cannot be ascribed to a quantum mechanical effect, but is most likely of classical origin. As an alternative explanation, the classical interference between signals reflected from different parts of the crystal (polarization beats) was suggested [3]. This mechanism of beats requires that the crystal is excited inhomogeneously, thereby providing the regions with different atomic amplitudes or electronic densities. In this communication we have studied the effect of inhomogeneous excitation for large-amplitude fully-symmetric phonons in Bi at helium temperature and concluded that lateral inhomogeneity combined with phonon chirp can explain the collapse and revival phenomenon rendering it to polarization interference instead of quantum beating.

2. Experimental

The pump-probe experiments were performed in a degenerate pump-probe setup utilizing a 250-kHz regenerative Ti:sapphire amplifier, delivering 50-fs pulses at λ=800 nm (1.55 eV). The induced changes in reflectivity (ΔR) were recorded by a fast-scan technique. Crystal was mounted in an optical helium flow cryostat with a silver paste and all measurements were carried out at T=5K. The pump laser beam was polarized along the trigonal axis of Bi in order to avoid the excitation of doubly degenerate phonons, while the probe beam was always polarized along the bisectrix axis, both the pump and probe beam entering the sample at near normal incidence. The pump beam diameter was fixed at 100 μm, while the probe diameter was varied between 50 and 100 μm, depending on experiment type.

3. Results and discussion

At liquid helium temperature and for pump fluence below F=0.1÷0.15 mJ/cm2, the amplitude of fully symmetric oscillations in bismuth is small and increases linearly with excitation strength [1]. In this excitation range, the frequency and lifetime of oscillations are independent of the pump fluence and coincide with the frequency-domain

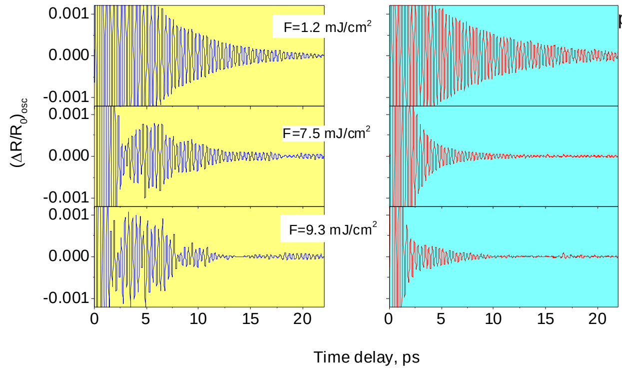

Fig.1. Oscillatory parts of isotropic reflectivity at different non-uniform (pump and probe diameters are 100μm, left panel) and uniform (pump diameter = 100μm, probe diameter = 50μm, right panel) excitation

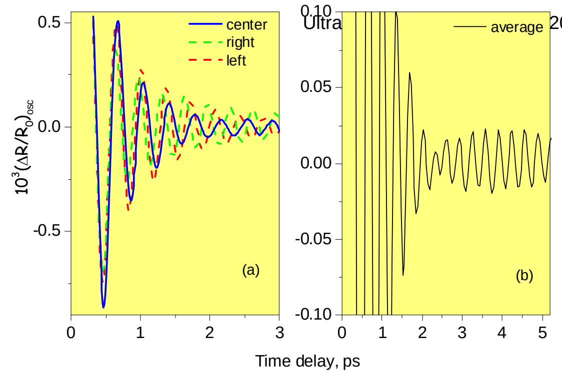

Fig.2. The oscillations detected for three different positions of the probe beam (ø=50μm) within the pumped area ø =100μm (a) with F=8.0 mJ/cm^2 and their average (b)

data obtained by spontaneous Raman scattering. In the time-domain, a harmonic oscillator model fits well the data for the delays up to 100 ps. For the pulses of the regenerative amplifier, the increase in the amplitude rapidly becomes nonlinear and the frequency and lifetime of the oscillations begin to depend on time [1]. Here, a harmonic oscillator model fits the data only for the first few cycles while it gradually goes out of phase with later oscillations. For larger initial displacements, the oscillatory frequency effectively becomes time dependent, starting at lower frequencies and evolving toward higher frequencies as the oscillations damp. The fits in real time present such a signal with a low frequency and short lifetime component for the early time delays and with a longer-lived higher frequency component for the signal at later times [1]. At low temperature above the fluence threshold (around 7÷8 mJ/cm^2) the oscillation pattern changes spectacularly: the oscillations die out and then, at some later time, revive, as shown in on the left panel of Fig.1, exhibiting an collapse and revival phenomenon.

In attempting to explain the complicated time-resolved response of the crystal, we have to bear in mind that both optically induced lattice distortion and *e-h* plasma density may be strongly inhomogeneous in real space due to non-uniform excitation conditions. To minimize the lateral real space inhomogeneity arising due to the excitation with a Gaussian beam profile it is enough to use the probe spot smaller than the pump spot. Right panel of Fig.1 depicts large-amplitude coherent phonon oscillations detected in Bi for approximately the same fluencies as in the left panel. However, the right panel data were recorded using a twice smaller probe spot diameter than that of pump in order to study more uniform excitation conditions in the center of the pumped area. By comparing these data with those obtained for a less uniform case (pump and probe sizes are equal), we can easily see that the collapse-and revival pattern disappears for more uniform excitation. Furthermore, we can scan the probe spot across the pumped area along the bisectrix axis, thus detecting differently excited areas. Even though each trace does not exhibit the collapse and revival, the average of these traces does demonstrate a clear beating pattern arising due to different phases of the oscillations, see Fig.2. This phase difference stems from various phonon chirps that are larger for the central part and smaller at the peripheries of the pumped area. These results demonstrate that the beating observed at high fluence excitation stems from different contributions within the laterally inhomogeneous pumped area. As the initial amplitude, decay rate and chirp increase while the averaged frequency decreases with fluence, the parameters of coherent oscillations become dependent on the observation spot. In the center of the pump spot the amplitude and chirp are maximal, whereas for detection spots away from the center the chirp decreases and with time a plane phase front of atomic displacements evolves into a curved one. It should be mentioned that despite the fact that the collapse and revival vanishes for a more uniform excitation, biexponential decay, phonon softening and chirp are still present in the data even for the uniform exciatation.

To summarize, we have demonstrated that for the fluencies near the Lindemann stability limit the collapse and revival pattern appears as a result of laterally inhomogeneous excitation. In this case the beats arise from the interference of polarizations, which do not have a common level, and are different from the quantum beats, which necessarily have a common level.

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