Dynamics of the Dielectric Function in fs-Laser Excited Bismuth

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Abstract: Time-resolved study of the dielectric function of femtosecond laser excited bismuth demonstrates that excitation of coherent phonons leads to a solid-plasma phase transition, and into a quasi-stable excited state lasting up to 4 ns.

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1. Introduction

Studies of non-equilibrium, non-thermal states hold the promise of discovering new transient phases, metastable states, and chemical reaction pathways. The absence of thermal equilibrium provides a way to observe the dynamics of the electronic and atomic subsystems, which determine the basic electronic, magnetic, and optical properties of the materials. Observation of coherent displacement and oscillation of phonons, which appear as oscillations of reflected light after excitation by an ultrashort laser pulse, enables investigation of lattice dynamics in solids through the electron-phonon energy coupling.[1,2]

Semi-metallic bismuth, with a small overlap in the energy of the conduction band and valence bands, is one of the most studied elements for unique electrical and thermal properties and their applications. Ultrafast laser-induced coherent lattice vibrations in bismuth, changes in long-range order, as well as direct measurements of the atomic positions within the unit cell, have been established using time-resolved x-ray diffraction[2-5]. However, the role of excitation electrons and oscillation of optical phonons in the solid-melt phase transition, and in particularly non-thermal melting such as found in tetrahedral bonded semicionductors,[6,7] is still obscure. Density functional theory (DFT) calculations suggested that bismuth undergoes a structural phase transition to a higher symmetry state, and it was not clear the level of excitation that could be achieved without thermal melting of the material.[4]

A direct way to test if the transient state in Bi is an intermediate state between the solid and the liquid or whether it is a transient solid-state phase is to measure the real and imaginary parts of the dielectric function. Here we present the results of time-resolved measurements of the dielectric constant of fs-laser excited Bi crystal.

2. Experimental

We performed dual-probe reflectivity measurements at 800 nm to measure the changes in the reflectivity of bismuth after excitation with femtosecond laser pulses. The dielectric constant was recovered from the time-resolved reflectivities of two optical probes, measured with an accuracy of $\Delta R = 10^{-5}$, employing dual-angle reflectometry with 40 fs time resolution. We have chosen *p*-polarisation for the pump for effective absorption and thus effective excitation of coherent oscillation of the A_{1g} phonon mode along the *c*-axis in Bi; subsequently the probes had to be *s*-polarised on the target surface.

The pump was directed to the crystal surface at a close to normal angle of incidence, and focused to a 125 μm spot size (FWHM), while the two probes were focused down to 40 μm spots and the angles of incidence were 19.5° and 34.5°; at these angles the geometrical time resolution was kept ~40 fs. The pump beam spot size was much larger than that of the probe beam in order to probe a central excited part of the sample. Spatial overlapping of two probes was double-checked by time synchronisation using an initial negative drop in reflectivity of the excited crystal. This negative dip in reflectivity relates to a coherent displacement of atoms by the polarisation force during the pulse,[8] and hence the drop was shorter than the excitation pulse. The laser fluence was much lower than the damage threshold, which was confirmed by reflectivity recovery to the initial value, and by observations of the crystal surface in the visualisation system.

The pump pulse was chopped at 500 Hz and the probe reflectivity measured using two digital lock-in amplifiers, which allowed us to improve the signal-to-noise ratio and to observe reflectivity changes, ΔR with accuracy $\Delta R/R_0 < 10^{-5}$. Fig.1 shows the recorded reflectivity behaviour at the excitation fluence 6.9 mJ/cm^2, and the dynamics of real and imaginary parts of the dielectric function are shown in Fig. 2.

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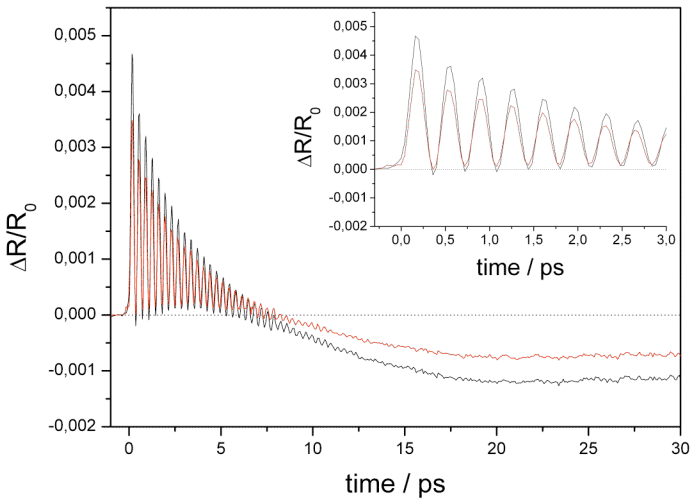

Fig.1. Transient reflectivity changes at 19.5° and 34.5° in the first 32 ps after the excitation. The inset shows the first negative dip during the pulse and the following coherent oscillations in the first 5 ps.

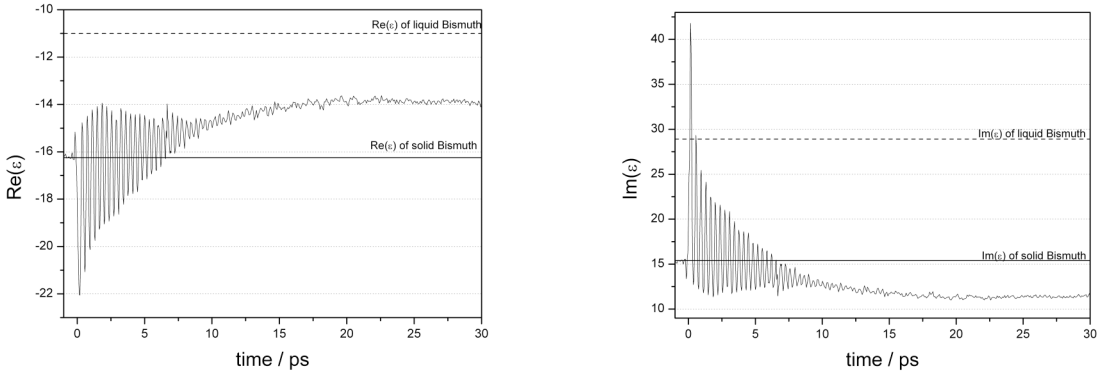

Fig. 2. Dynamics of real and imaginary parts of the dielectric function. Values for solid (solid line) and liquid (dotted line) are also shown.

3. Conclusions

Our findings show that the laser-induced oscillations of coherent phonons followed by a transition to a new quasi-equilibrium state of Bi lasts up to 4 ns. While the real part of the dielectric function does move toward the liquid value, the imaginary part moves to lower values, away from the range defined by the solid and the liquid dielectric constants. There was no single moment in time when both, the real and the imaginary parts had values within the solid-liquid gap. The value of the complex dielectric constant of the transient state $\varepsilon^{tr} = 17.9 \pm 0.2$ at 800 nm was out of the solid-liquid gap between the $\varepsilon^{sol} = 22.4$ and $\varepsilon^{liq} = 30.9$. This suggests that the excitation of coherent oscillations in Bi is followed by a solid-plasma transition into a quasi-steady state. The transient state does not lead to melting even at the absorbed excitation laser fluence as high as 4 mJ/cm^2, supplying the energy into the 28-nm skin layer far above the energy density required for the equilibrium melting.

[1] T. K. Cheng, et. al., "Impulsive excitation of phonons observed in reflection in bismuth and antimony", Appl. Phys. Lett. **57**, 1004 (1990).

[2] A. Rousse, et. al., "Non-thermal melting in semiconductors measured at femtosecond resolution", Nature **410**, 65 (2001).

[3] K. Sokolowski-Tinten, et. al., "Femtosecond x-ray measurements of coherent lattice vibrations", Nature **422**, 287 (2003).

[4] D. M. Fritz, et.al., "Ultrafast bond softening in bismuth:Mapping the solid's interatomic potential with x-rays", Science **315**, 633 (2007).

[5] P. Beaud, et.al., "Spatiotemporal stability of a femtosecond hard x-ray undulator source", Phys. Rev. Lett. **99**, 174801 (2007).

[6] A. M. Linderberg, et. al., "Atomic-scale visualisation of inertial dynamics",Science **308**, 392 (2005).

[7] V. Recoules, et. al., "Effect of Intense Laser Irradiation on the Lattice Stability", Phys. Rev. Lett. **96**, 055503 (2006).

[8] D. Boschetto, et. al., "Fine atomic motion recorded by reflectivity of fs-laser excited Bismuth", accepted in Phys. Rev. Lett., 14 Nov. 2007.