Coherent phonons in bismuth under high-density excitation

Muneaki Hase[1], Masahiro Kitajima[1], Kohji Mizoguchi[2] and Shin-ichi Nakashima[3]

[1]National Institute for Materials Science, 1-2-1 Sengen, Tsukuba, 305-0047 Ibaraki, Japan
[2]Department of Applied Physics, Osaka City University, 3-3-138 Sugimoto, Sumiyoshi-ku, Osaka 558-8585, Japan
[3]Department of Electrical and Electric Engineering, Miyazaki University, 1-1 Gakuen-kibanadai-nishi, Miyazaki 889-2192, Japan

High-density excitation of condensed matters with amplified femtosecond pulses has recently provided a noble method to study noticeable physical phenomena such as femtosecond laser ablation and laser-induced phase transition. In order to understand these phenomena, ultrafast dynamics of photo-excited carriers as well as nonequilibrium phonons should be investigated. A transient red-shift of the frequency of the coherent A_{1g} phonon was observed for Ti_2O_3. The origin of the red-shift was attributed to an ionic screening by the photo-excited carriers [1]. Hunsche *et al.* have reported that the coherent A_{1g} mode frequency in Te was decreased linearly with increasing pump power densities [2]. Up to now, however, systematic study on the coherent phonon dynamics using amplified pulses has been performed only for Te, and detailed investigation of coherent phonons is required for wide materials. In the present work, we report on the systematic study of coherent optical phonons in semimetal Bi under the condition of the high-density excitation using amplified femtosecond pulses with a pulse duration of 120 fs. The coherent oscillation in Bi films prepared by vacuum deposition was observed by using a reflection-type pump-probe technique.

The time domain signals observed in the Bi films reveal the coherent A_{1g} vibration, which dramatically changes with the pump power density. As the pump power density is increased, the amplitude and the period of the coherent A_{1g} phonon are increased and the decay time of the oscillation is decreased. We found the fact that under the excitation with 7.6 mJ/cm^2 the period of the A_{1g} mode gradually becomes longer with increasing the time delay. In order to analyze the coherent oscillations under the high-density excitation, we have performed Fourier transformation (FT) of the time-domain signals. The FT spectra show that a line shape of the A_{1g} mode becomes asymmetric and the phonon frequency shifts from 2.89 to 2.67 THz with increasing the pump power density. The frequency change of the A_{1g} mode with the time delay observed at the excitation with 7.6 mJ/cm^2 was analyzed by using a time partitioning FT technique. Time-resolved analysis of the coherent oscillation indicates that the asymmetry of the phonon band is caused by a transient red-shift of the phonon frequency originated from the anharmonic phonon oscillation.

Reference:

[1] T. K. Cheng, L. H. Acioli, J. Vidal, H. J. Zeiger, G. Gresselhaus, and M. S. Dresselhaus, Appl. Phys. Lett. **62**, (1993) 1901.
[2] S. Hunsche, K. Wienecke, T. Dekorsy, and H. Kurz, Phys. Rev. Lett. **75**, (1995) 1815.