# III-H Coherent Phonon Dynamics in Crystals

## III-H-1 Femtosecond Pump-Probe Study of Coherent Soft Phonon in Ferroelectric Materilas

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Structural phase transitions in solids have been investigated with X-ray- and neutron-scatterings, and especially through the "*soft mode*" dynamics using Raman scattering spectroscopy. However, such the soft mode is usually located at extremely low frequency or shifts to "zero" frequency when the lattice temperature approaches to the critical temperature $T_c$, which makes frequency-domain spectroscopic study difficult. For this point of view the time-domain spectroscopy method has advantage to study such low frequency phonon modes. The time-domain study provides also possibility of observing nonthermal phase transitions under high density photoexcitation above mJ/cm$^2$ pump fluences, which is inaccessible by frequency-domain techniques. Motivated by the observation of the ultrafast dynamics of structural phase transition in ferroelectric materials under the high-density photoexcitation, in this work, we have investigated the coherent soft mode in $Pb_{1-x}Ge_xTe$ ($x$ = 0.07) using a pump-probe technique with amplified femtosecond laser pulses. As the pump fluence increases, the frequency of the coherent $A_1$ phonon becomes soften and at higher fluence it reaches saturation due to the screening effect in the electron-phonon interaction, while the large damping rate dose not show similar saturation due to the dominant anharmonic phonon-phonon coupling. The linear increase of the amplitude of the coherent phonon is ascribed to the linear increase of photo-excited carrier density under the DECP mechanism. The saturation behavior of the frequency of the soft mode implies that it will be difficult to realize the laser induced structural phase transition using single pump in $Pb_{1-x}Ge_xTe$, and multiple-pump excitation will provide a possibility for the future studies.

**Reference**

1) R. Lu, M. Hase, M. Kitajima, S. Nakashima and S. Sugai, *J. Lumin.* **378**, 119–120 (2006).

## III-H-2 Temperature Dependence of Coherent $A_{1g}$ and $E_g$ Phonons of Bismuth

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Bismuth has been a model material to study femtosecond dynamics of coherent lattice oscillations. The generation mechanism was considered to be displacive for the totally symmetric $A_{1g}$ mode, which was the only coherent mode observed in the conventional (isotropic) detection. The absence of the other Raman active mode, $E_g$, has not been fully explained, but was phenomenologically attributed to the exclusive coupling of the hot electrons at $k < 0$ and high symmetry phonons. In the present study, we demonstrate that both $A_{1g}$ and $E_g$ modes are coherently excited at comparable amplitudes at low temperature, and thus proved that the coherent phonons are generated basically *via* Raman process. We found a puzzling $\pi/2$ difference in the initial phases of the two coherent oscillations, which suggests that the initial phase is not a clear-cut indication for the generation mechanism of the coherent phonons in absorbing media.

**Reference**

1) K. Ishioka, M. Kitajima and O. V. Misochko, *J. Appl. Phys.* **100**, 093501 (6 pages) (2006).