Coherent phonons in Bi$_2$Se$_3$ induced by a powerful THz pulse

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Abstract. Using the electro-optic detection method we have detected coherent phonons of $E^1_u$, $E^1_g$, $E^2_g$, and $A^2_{ig}$ symmetry generated in a crystalline film of Bi$_2$Se$_3$ by a powerful single-cycle THz pulse. Coherent excitation of Raman active modes is interpreted as a result of three- and four-phonon interactions in the anharmonic crystal lattice of Bi$_2$Se$_3$.

In the present work we studied the ultrafast structural response of a crystalline Bi$_2$Se$_3$ film to intense picosecond terahertz pulses. The sample of Bi$_2$Se$_3$ under study was an MBE-grown single-crystal film of high structural quality with a thickness of 24 nm on a (111)-oriented BaF$_2$ substrate covered by a 28 nm thick protective layer of BaF$_2$. The sample was excited by THz pulses with duration of about 1 ps and peak electric field strength reaching ~ 1 MV/cm. Spectral width of the pulses was ~ 2 THz and the central frequency ~ 1.5 THz. Thus, the IR active phonon mode $E^1_u$ of Bi$_2$Se$_3$ at 67 cm$^{-1}$ (2 THz) was resonantly excited. We detected anisotropic changes of the refractive index of the Bi$_2$Se$_3$ film induced by the electric field of the THz pulse. To do that we used a probe laser pulse at 800 nm with a duration of 30 fs. The intensities of its orthogonal polarization components were measured after the pulse propagated through the sample. We registered the dependence of the corresponding ratio $I_x/I_y$ on the delay time between pump and probe pulses. In Fig. 1(a) we show a typical ultrafast response (decay trace) of the Bi$_2$Se$_3$ film to the THz pulse. As follows from the FFT spectra of three decay traces measured in succession (Fig. 1(b)) the oscillating signal contains four frequencies of 1.1, 2.05, 4.05, and 5.3 THz (maximal error ~ 0.05 THz) that correspond to optical phonon modes of $E^1_g$, $E^1_u$, $E^2_g$, and $A^2_{ig}$ symmetry. These oscillations are caused by coherent vibrations of Bi$_2$Se$_3$ atoms (coherent phonons), that are induced by the pump THz pulse. The observed generation of coherent phonons cannot be explained using ISRS [1] or DECP [2] theories because the duration of the pump THz pulse is comparable to or larger than the periods of optical phonon modes in Bi$_2$Se$_3$. The frequencies of $E^2_g$ and $A^2_{ig}$ modes are situated outside the THz pulse spectrum and its direct action on them is excluded. Thus, to explain this unusual effect one should take into accounts higher order processes.

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Fig. 1. (a) Ultrafast response of Bi$_2$Se$_3$ to the THz pulse. (b) FFT spectra of three decay traces measured in succession (vertical axis is in the logarithmic scale).

Recently similar experiments were described in [3] and [4], where the generation of $E^2_g$ mode was interpreted as a result of vibrationally induced symmetry breaking in the bulk and sum frequency Raman scattering respectively. In contrast to those works we argue here that the coherent excitation of Raman active phonons in Bi$_2$Se$_3$ by the THz pulse is the result of crystal lattice anharmonicity and multiphonon processes somewhat similar to “ionic” Raman scattering proposed in [5]. Thus, the generation of $E^2_g$ phonons is explained as resonant excitation of the Raman active mode by the second harmonic of nonlinear $E^1_u$ oscillator, the fundamental frequency of which is approximately half that of the $E^2_g$ mode. In this case the nonlinearity is the consequence of the large amplitude of $E^1_u$ vibrations and the cubic lattice anharmonicity (three-phonon processes with two $E^1_u$ phonons and one $E^2_g$ phonon). At the same time the generation of $E^1_g$ and $A^2_g$ phonons can be caused by quartic lattice anharmonicity that provides the frequency combination $2\Omega(E^1_u) = \Omega(A^2_g) - \Omega(E^1_g)$. The proposed interpretation was further supported by experiments with nonresonant radiation and by experiments, in which the dependence of the signal on THz field strength and sample orientation was measured.

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References

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