EPJ Web of Conferences **161**, 01009 (2017) DOI: 10.1051/epjconf/201716101009
PECS-2017

Coherent phonons in Bi_2Se_3 induced by a powerful THz pulse

Alexey Melnikov[1,*], *Kirill* Boldyrev[1] , *Yuri* Selivanov[2], *Victor* Martovitskii[2], *Sergey* Chekalin[1], and *Evgeni* Ryabov[1]

[1]Institute of Spectroscopy RAS, 108840 Troitsk, Moscow, Russia
[2]P.N. Lebedev Physics Institute RAS, 119991 Moscow, Russia

Abstract. Using the electro-optic detection method we have detected coherent phonons of E_g^1, E_u^1, E_g^2, and A_{1g}^2 symmetry generated in a crystalline film of Bi_2Se_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_2Se_3.

In the present work we studied the ultrafast structural response of a crystalline Bi_2Se_3 film to intense picosecond terahertz pulses. The sample of Bi_2Se_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_u^1 of Bi_2Se_3 at 67 cm^{-1} (2 THz) was resonantly excited. We detected anisotropic changes of the refractive index of the Bi_2Se_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_2Se_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_g^1, E_u^1, E_g^2, and A_{1g}^2 symmetry. These oscillations are caused by coherent vibrations of Bi_2Se_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_2Se_3. The frequencies of E_g^2 and A_{1g}^2 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.

* Corresponding author: melnikov@isan.troitsk.ru

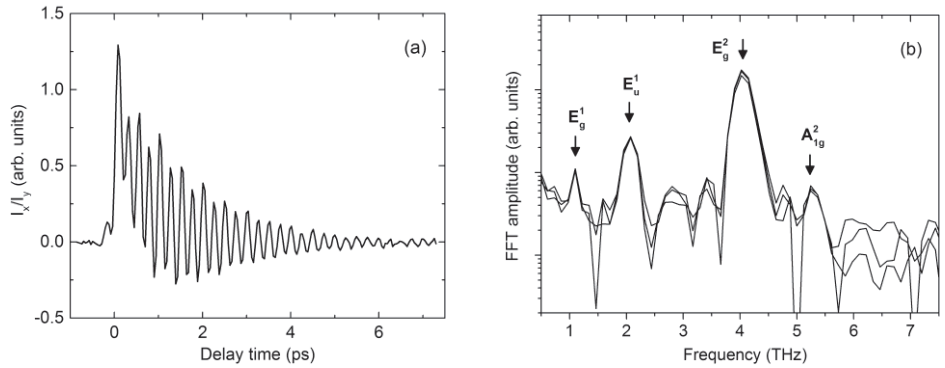

Fig. 1. (a) - Ultrafast response of Bi_2Se_3 to the THz pulse. (b) - FFT spectra of three decay traces measured in succession (vertical axis is in the logarithmic scale).

Recently similar experiments were described in [3] and [4], where the generation of E_g^2 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_2Se_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_g^2 phonons is explained as resonant excitation of the Raman active mode by the second harmonic of nonlinear E_u^1 oscillator, the fundamental frequency of which is approximately half that of the E_g^2 mode. In this case the nonlinearity is the consequence of the large amplitude of E_u^1 vibrations and the cubic lattice anharmonicity (three-phonon processes with two E_u^1 phonons and one E_g^2 phonon). At the same time the generation of E_g^1 and A_{1g}^2 phonons can be caused by quartic lattice anharmonicity that provides the frequency combination $2\Omega(E_u^1) = \Omega(A_{1g}^2) - \Omega(E_g^1)$. 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.

This work was supported by the Ministry of Education and Science of the Russian Federation (project # RFMEFI61316X0054).

References

[1] L. Dhar, J. A. Rogers, K. A. Nelson, Chem. Rev. **94**, 157 (1994)

[2] H.J. Zeiger, J. Vidal, T.K. Cheng, E.P. Ippen, G. Dresselhaus, M.S. Dresselhaus, Phys. Rev. B **45**, 768 (1992)

[3] P. Bowlan, J. Bowlan, S.A. Trugman, J.-X. Zhu, N.J. Hur, A.J. Taylor, D.A. Yarotski, and R.P. Prasankumar, Optica **4**, 383 (2017)

[4] L. Braun, *Electron and phonon dynamics in topological insulators at THz frequencies* (Freie Universität Berlin, Germany, 2016)

[5] M. Först, C. Manzoni, S. Kaiser, Y. Tomioka, Y. Tokura, R. Merlin, and A. Cavalleri, Nature Phys. **7**, 854 (2011)