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Ultrafast dynamics of crystalline bismuth studied by femtosecond pulses in visible and near-infrared range

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Ultrafast dynamics of laser excited bismuth were studied by means of wavelength-resolved femtosecond pump-probe technique. Several values of excitation wavelengths from 400 to 2300 nm and a probe femtosecond continuum in 400-900 nm range were used. The analysis of decay traces revealed three processes with relaxation time values of 1 ps, 7 ps and ˜ 1 ns ascribed to groups of photoexcited electrons with essentially different coupling to the lattice. A conclusion was made that the fastest process along with the relaxation of the instantaneous frequency shift equally represent the return of the displaced equilibrium position of bismuth lattice to its unperturbed value. An intermediate 7-ps process was attributed to the crystal heating. The nanosecond component is believed to be due to electrons near the Fermi level. Spectral amplitude of A1g oscillations was also measured. The analysis of its specific shape shows that fully symmetric coherent atomic oscillations affect reflectance of bismuth crystal through the modulation of conductivity. Using different excitation wavelengths we found a sharp decrease (up to 5 times) of the coherent phonon amplitude at excitation photon energies less than 0.7 eV. Relying on this fact we conclude that only a portion of excited electrons (probably in states near the T6+ point of the Brillouin zone) alter the potential surface of bismuth sufficiently and cause the shift of A1g mode equilibrium position.

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