

**Boschetto *et al.* Reply:** The Comment from Misochko *et al.* [1] questions our interpretation of the initial negative dip in reflectivity as competition between the Plazceck effect, responsible for the negative change in reflectivity, and the positive contribution due to electron excitation [2]. They suggest that it is due to interference between the pump and the probe beams as observed in their experiments. We can exclude their interpretation on the following grounds. First, we observed the initial negative dip only with short pulses  $< 40$  fs. Any interference effect should also be present using longer pulses, as in [1] and Refs. [2–4] of the Comment. With long pulses the negative polarization-driven intensity change disappears. Moreover, interference effect gives rise to both increase and decrease of the signal, whereas we only observe a decrease. Second, the amplitude of the negative dip qualitatively followed our theoretical interpretation, namely, the higher the pump intensity, the larger the polarization, and thus the deeper the dip in reflectivity [3]. It, also, cannot be related to any phonon mode of particular symmetry, as suggested in the Comment, because the effect appears in a time about 30 times shorter than the phonon oscillation period. The polarization certainly depends on the anisotropy of the dielectric function, but it does not depend on the asymmetry of phonon modes mentioned in [1]. The excitation of phonons by the electronic pressure gradient given in [2,3] is a further development of the displacive model (DECP) [4]. We established how the atomic motion enters into the dielectric function and into the reflectivity through the electron-phonon momentum exchange rate and through the field-induced polarization. The force proportional to the gradient of the electronic pressure initiates fast coherent displacement of the atoms, which produces a negative change in reflectivity, while the electronic excitation induces a positive contribution. Subsequently, the laser-induced forces generate coherent atomic vibrations at the phonon frequency. These are major differences in our theory when compared with the previous approaches where the atomic motion had been inserted into the reflectivity in an *ad hoc* manner. The electron-electron collision rate is based on thorough studies of the electron-electron scattering in the degenerate and nondegenerate gases summarized in [5]. We treat the decrease in the phonon amplitude as due to nonlinear phonon-phonon interaction intensified by the lattice heating in the nonequilibrium conditions of fs-laser excitation, while the Comment's statement that "heating Bi above the melting temperature leads to a decrease of reflectivity" [1] and their estimates of the lattice temperature were derived for the equilibrium case. We calculated the maximum lattice temperature directly from the conservation of energy taking the measured energy density absorbed in the skin layer [2,3] and omitting the electrons diffusion, because it is negligible. The electron-lattice energy transfer time is much shorter than the lattice cooling time due to high electron collision frequency in Bi of

$\geq 2 \times 10^{15} \text{ s}^{-1}$  above room temperature [3]. The reference to the experiments presented in Fig. 1 in the Comment is not relevant to our experiments [2,3] at room temperature as the electron density of free electrons in Bi at 8 K in [1] is lower by several orders of magnitude [6,7]. It has been demonstrated that ice [8], gallium [9], and aluminum [10,11], excited by ultrashort pulses to the state where the absorbed energy  $E_{\text{abs}}$  is higher than the equilibrium enthalpy of melting  $H_{\text{melt}}$ , namely, to  $E_{\text{abs}}/H_{\text{melt}} = 1.073, 2.67, 9.4$  correspondingly, do not melt for time periods much longer than the electron-lattice equilibration time when melting should have occurred from the naïve point of view. In fact, we have recently measured the transient dielectric function of fs-excited Bi [12], and found that the transient state of Bi is very different from both the solid and the liquid states but shows similar behavior as in [8–11]. The transient state does not correspond to the melt even at absorbed excitation laser fluences far above the energy density required for equilibrium melting. Further experimental and theoretical studies are needed in order to reveal the unusual electronic and lattice properties of transient state of Bi excited by powerful ultrashort laser pulses.

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