Contribution ID: **32** Type: **not specified**

Lattice Response to Femtosecond Laser Excitation Studied by Time Resolved Transmission Electron Diffraction

Lattice vibrations after femtosecond laser excitation can be launched by different processes such as the relaxation of hot electrons or the coupling of the laser pulses to the lattice via nonlinear-optical mechanisms. The pathway and time scale of energy transfer form photonic excitation to the phononic subsystem is of fundamental interest to understand various physical phenomena like e.g. laser-driven structural phase transitions. Time resolved transmission electron diffraction (TED) is an ideal tool for these investigations. The different excitation mechanisms produce distinct signatures in the time evolution of the electron diffraction pattern. The possibility of observing many diffraction spots simultaneously enables us to identify the signature the atomic motion. Here we report on experimental results of lattice excitation by time resolved electron diffraction and the analysis of disentangling disordered thermal motion and directional atomic displacement in Bismuth and Nickel.

The experiments were performed on free-standing single crystalline thin films of either 22 nm Bismuth with (001)surface orientation (hexagonal notation) or 20 nm Nickel with (001) surface orientation. The electron probe pulses had an energy of 30 keV, and the pulse duration was less than 700 fs. The samples were laser-excited using pulses of 800 nm at a repetition rate of 1 kHz with 2 or 4 mJ/cm^2 incident fluence for Bismuth or Nickel, respectively.

The diffraction patterns were recorded as function of delay time between optical pump and electron probe. The integrated intensity of all the diffraction spots decreased after laser excitation. The ultrafast lattice heating can be described by the transient Debye-Waller effect, which treats the lattice displacement as disordered thermal motion by fitting the decrease of the diffraction intensities to the Debye-Waller factor for corresponding diffraction orders. The extracted time constants for Bi (4 ps) and Ni (2 ps) indicated strong material dependence of the ultrafast lattice heating. In addition a directional displacement could be studied by analyzing the temporal evolution of proper diffraction orders under specified diffraction conditions. The directed motion as longitudinal elastic waves in direction of the surface normal was observed in Nickel and Bismuth. We discuss the diffraction patterns and the possible driving mechanisms.

We gratefully acknowledge the financial support by the Deutsche Forschungsgemeinschaft through SFB 616 “Energy dissipation at surfaces”.

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Session Classification: Molecular dynamics 1