Dynamics in Solids by Transient XUV Spectroscopy

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Ultrafast optical pulses allow to excite materials away from thermal equilibrium, where interactions between the degrees of freedom of solids can be explored. Electron-lattice interactions in particular determine numerous material functionalities, such as the fundamental limits on charge-carrier lifetimes and mobilities in photovoltaic and photoelectrochemical applications. Extreme ultraviolet light (XUV) is a compelling way to probe these dynamics, because of access to conduction and valence band information through core level transitions in solids.

Few-femtosecond XUV transient absorption spectroscopy reveals simultaneously intra- and interband carrier relaxation and light-induced (phonon) structural dynamics in layered 2H-$MoTe_2$ semiconductor material. By interrogating the valence electronic structure via localized Te 4d (39−46 eV) and Mo 4p (35−38 eV) core levels, the relaxation of the photoexcited hole distribution is directly measured in real time. Hole carrier-carrier thermalization and carrier-phonon cooling times of 15 ± 5 fs and 380 ± 90 fs are observed, respectively. Excitations of out-of-plane A_{1g} (5.1 THz) and in-plane E_{1g} (3.7 THz) lattice vibrations are visualized through coherent oscillations in the XUV absorption spectra. The spectral changes are mapped to real-space excited-state displacements of the lattice along the dominant A_{1g} coordinate.

Access to continuous core-to-conduction band absorption features and discrete core-exciton transitions in the same extreme ultraviolet spectral region in WS_2 semiconductor material provides a novel means to investigate the effect of carrier excitation on core-exciton dynamics. The core-level transient absorption spectra reveal that core-exciton transitions are strongly influenced by the photoexcited carriers, which is the dominant contributor to the spectral modifications of core-excitons, while direct laser-field-induced changes play a minor role.

In Peierls-distorted materials, photoexcitation leads to a strongly coupled transient response between structural and electronic degrees of freedom, always measured independently of each other. Transient reflectivity in the XUV is used to quantify both responses in photoexcited bismuth (Bi) in a single measurement. With the help of first-principles calculations based on density-functional theory (DFT) and time-dependent DFT, the real- space atomic motion and the temperature of both electrons and holes as a function of time are captured simultaneously. An anticorrelation between lattice expansion and carrier temperature is observed. The results reveal a coherent, bi-directional energy exchange between carriers and phonons which is a dynamical counterpart of the static Peierls-Jones distortion, providing first-time validation of previous theoretical predictions.

By directly and simultaneously probing excited carrier distribution dynamics in materials and their accompanying femtosecond lattice displacements, XUV transient absorption provides new benchmarks for understanding the interplay between electronic and structural dynamics in photoexcited materials.