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Novel Lattice Instability in Ultrafast Photoexcited SnSe

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There has been growing interest in using ultrafast light pulses to drive materials into nonequilibrium states with novel properties. Using time-resolved X ray scattering, I demonstrated that SnSe, one of the IV-VI resonantly bonded compounds, hosts a novel photo-induced lattice instability associated with an orthorhombic distortion of the rock-salt structure with space group Immm [1]. The new lattice instability is accompanied by a drastic softening of the lowest-frequency Ag phonon (TO phonon), which has previously been identified as the soft mode in the thermally driven phase transition to a Cmcm structure. Therefore, we provide a counterexample of the conventional wisdom that laser pump pulse serves as a heat dump. Density functional theory calculations reveal that the photoinduced Immm lattice instability arises from electron excitation from the Se 4p- and Sn 5s-derived bands (the lone pair orbitals) deep below the Fermi level that cannot be excited thermally.
Furthermore, I show results of non-zone-center measurements of time-resolved X-ray scattering, from which I extracted interatomic force constants in the photoexcited states. and identify a certain bond that is largely overlapped with the lone pair orbital as responsible for the observed photoinduced lattice instability. The conclusion is in contrary to the consensus that in thermal equilibrium, the resonant bonding network of chalcogen p orbitals is the main origin for lattice instability and a soft TO mode. And indeed, the photoexcited phonon modes have a significantly longer lifetime, which means less anharmonicity of the lattice, than those in thermal equilibrium, consistent with the observation that interatomic interaction driving the photoinduced lattice instability is different from the thermal.
The results have implications for optical control of the functional properties of monochalcogenides and other resonantly bonded materials. More generally, the results emphasize the importance ultrafast structural probes that reveal distinct atomic-scale dynamics too subtle for conventional spectroscopies.
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[1] Y. Huang et al., Phys. Rev. X, 12, 011029 (2021)

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