Transient band gap enhancement of the photoexcited excitonic insulator phase in quasi-1D Ta2NiSe5

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Abstract

Electronic correlations can lead to exotic electronic properties that a single-particle theory cannot explain. Among these is the strong electron-hole interaction in small gap semiconductors and semimetals, which may lead to a spontaneous formation of excitons resulting in a phase transition (PT) to an excitonic insulator (EI) [1]. The ternary chalcogenide Ta2NiSe5 (TNS) has been proposed as a possible candidate for such semiconductor-to-EI transition occurring at Tc ≈ 328 K in combination with a structural change from orthorhombic to monoclinic symmetry [2]. In order to unveil how the monoclinic/EI phase stabilizes in TNS, we investigate the photoinduced dynamics by means of time-resolved photoemission and optical spectroscopy and we interpret the complex relaxation processes in the framework of strong electron-hole correlations in EI systems.

Time-resolved photoemission in the monoclinic/EI phase shows a strong depletion of the valence band as a function of the excitation density, until a saturation of the PE intensity occurs above a critical fluence Fsat. This effect is reflected in a saturation threshold of the overall transient optical response in the near-IR, resonantly probing the energy gap of TNS. A coherent phonon at 4 THz, which is characteristic of the monoclinic/EI phase, persists above Fsat, indicative of a hindered photoinduced phase transition. Excitation density dependence at different temperatures in the EI phase shows that Fsat decreases as the sample temperature approaches Tc. This effect is interpreted as an enhanced resonance condition for the pump photon energy, which is consistent with the thermally-driven shifting of the valence band maximum towards lower binding energy observed by ARPES [3]. Time-resolved photoemission in the monoclinic/EI phase reveals a band gap shrinking below Fsat, which we assign to free-carrier-enhanced screening of Coulomb interaction. We note that this process persists away from G point of the Brillouin zone even above Fsat. However, at G and for strong excitation above Fsat, these dynamics compete with a delayed band gap widening that we attribute to further localization of the photoexcited excitons due to repulsive interaction, as expected in a weakly interacting Bose gas. After ˜ 1.5 ps, excess energy is transferred to the lattice and the band gap shrinking is driven quasi thermally. Thus, time-resolved photoemission of the occupied band structure proves that the saturation in the optical response is due to a depopulation of the valence band with consequent photobleaching of pump pulse absorption that precludes the possibility of a photoinduced gap closing in TNS. Furthermore, under strong excitation the band structure at G is likely affected by dynamical change of the exciton binding energy that compete with free-carrier screening of electron-hole Coulomb interaction, such that the excitonic band gap can be transiently enhanced. This supports the key role of exciton correlation as origin of the EI phase in TNS.

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