Ultrafast electronic band gap control and inhibition of the photoinduced structural phase transition in an excitonic insulator

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Ultrafast control of matter phases is of both fundamental and technological interest. Here, we study the ultrafast dynamics of Ta_2NiSe_5 by means of time- and angle-resolved photoelectron spectroscopy (trARPES) [1] and transient reflectivity measurements [2]. Ta_2NiSe_5 is proposed to support an excitonic insulator (EI) phase below $T_C \approx 328$ K, combined with a structural change from orthorombic to monoclinic symmetry. Such an EI phase is expected to occur in small gap semiconductors with strong electron-hole interaction as excitons can form spontaneously and condense into a ground state. Below T_C, trARPES around Γ shows a strong fluence-dependent valence band depopulation, until absorption saturates at a critical fluence F_{sat} = 0.2 mJ cm^{-2}. This is reflected in a saturation of the mid-IR optical response at F_{sat}. A coherent phonon at 4 THz, marker of the EI/monoclinic phase, persists above T_C, indicating that the photoinduced *structural* transition is hindered by pump absorption saturation. trARPES shows that below F_{sat} the band gap *shrinks* transiently due to photoenhanced screening of the Coulomb interaction, while above F_{sat} it *widens* and recovers to its equilibrium value after ~1.5 ps. Hartree-Fock calculations reveal that the band gap widening is due to photoenhancement of the exciton condensate, persisting until the system undergoes interband relaxation. Our results prove it is possible to manipulate exciton condensates optically and gain ultrafast control of semiconductor band gaps.

[1] Mor et al., Phys. Rev. Lett. 119, 086401 (2017).
[2] Mor et al., Phys. Rev. B 97, 115154 (2018)