Ultrafast electronic and structural dynamics in an excitonic insulator

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Strongly correlated systems often exhibit a rich phase diagram originating from the interaction between various degrees of freedom. Monitoring the electron and lattice dynamics after photoexcitation helps to disentangle the elementary processes associated with phase transitions and verify if a phase transition can be photoinduced. Here, we study the ultrafast electronic and structural dynamics of Ta_2NiSe_5 by means of time- and angle-resolved photoelectron spectroscopy (trARPES) and transient mid-IR reflectivity measurements. Ta_2NiSe_5 is proposed to support an excitonic insulator (EI) phase below $T_C \approx 328$ K, combined with a structural change. 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 which saturates at a critical fluence $F_C = 0.2$ mJ cm^{-2} due to optical absorption saturation. This limits the optical excitation density in the system below the threshold for a non-thermal *structural* phase transition. This effect is monitored by the transient mid-IR optical response below T_c, which also saturates and exhibits an *un*changed phonon spectrum above F_C. We thereby prove that the electronic subsystem of Ta_2NiSe_5 is capable to protect the structural phase from a photoinduced change.

TrARPES shows that below F_C the band gap transiently *shrinks* due to photoenhanced screening of the Coulomb interaction. However, above F_C the band gap transiently *widens* at Γ. Hartree-Fock calculations reveal that the band gap widening is due to photoenhancement of the EI condensate density until the system undergoes interband relaxation. Our work proves that it is possible to manipulate exciton condensates with light and to gain ultrafast control of semiconductor band gaps.

References

[1] S. Mor, et al, 2017 *Phys. Rev. Lett.* 119, 086401
[2] S. Mor, et al, 2018 *Phys. Rev. B Rapid Comm.* under review