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$_2$NiSe$_5$ by means of time- and angle-resolved photoelectron spectroscopy (trARPES) and transient mid-IR reflectivity measurements. Ta$_2$NiSe$_5$ 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 $\Gamma$ 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 unchanged phonon spectrum above $F_C$. We thereby prove that the electronic subsystem of Ta$_2$NiSe$_5$ 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 $\Gamma$. 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