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Ultrafast Quasiparticle Dynamics in a Transition Metal Dichalcogenide and at Hybrid Organic/Inorganic Interfaces

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Abstract: Ta₂NiSe₅ is proposed to support an excitonic insulator phase below $T_C \approx 328$ K combined with a structural change. The former occurs in small gap semiconductors with strong electron-hole interaction where excitons form spontaneously and condense into a new insulating ground state. We study the ultrafast electron and lattice dynamics of Ta₂NiSe₅ by means of time- and angle- resolved photoemission spectroscopy¹ (trARPES) and time-resolved coherent optical phonon spectroscopy². We find that the low temperature structural phase persists even for high excitation densities and the photoinduced structural phase transition is hindered by absorption saturation at $FC = 0.2$ mJ cm⁻². Below FC, the band gap shrinks transiently due to photoenhanced screening of the Coulomb interaction. However, above FC, the band gap transiently widens at the Gamma point 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 density, persisting until interband carrier relaxation occurs. These results demonstrate the possibility to manipulate exciton condensates with light and gain ultrafast band gap control.

We also characterize the ultrafast dynamics of electrons and excitons at a model organic/inorganic interface (5-phenylpyridine/ZnO(10-10)). In particular, we populate the LUMO of the organic molecules by two different pathways: (i) Via photoexcitation from an interfacial hybrid state just below EF and (ii) by optical excitation of the organic dye. We thereby disentangle the two main contributions to interfacial charge separation, electronic coupling to the ZnO conduction band and electron-hole interaction in the organic molecule.

1 S. Mor et al. Phys. Rev. Lett. 119, 086401 (2017)

2 S. Mor et al. Phys. Rev. B (2018), accepted

