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Infrared spectroscopy of collective excitations in quasi-one-dimensional Ta_2NiSe_5 and Ta_2NiS_5

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Cooper pairing of fermions is one of the most fundamental and successful concepts of condensed matter physics. In analogy to the superconducting state generated by pairing of electrons in metals, condensation of neutral electron-hole pairs in semimetals or narrow-band semiconductors has been predicted to generate a coherent quantum excitonic insulator state. There has been continued effort to look for an excitonic insulator in real materials.

Using spectroscopic ellipsometry, we have studied collective excitation modes in the quasi-one-dimensional chalcogenide Ta_2NiSe_5 , which exhibits a structural phase transition that has been attributed to exciton condensation, and in the isostructural Ta_2NiS_5 which does not exhibit such a transition [1-3]. We conclude that in Ta_2NiS_5 the exciton-phonon complexes are sparse and do not develop a long-range order. This local violation of symmetry allows the excitations to have a nonzero energy, which we see in the dielectric function as a soft localized excitation. In Ta_2NiSe_5 , on the other hand, the giant spectral weight of the exciton Fano resonances due to antenna emission of large exciton-phonon complexes implies that they are strongly overlapping and probably span the entire crystal, hence preserving the translational symmetry of the lattice. Due to this symmetry the internal excitations of the exciton-polaron aggregates may support propagating low-energy excitations akin to acoustic phonon modes. The observed difference reflects the different nature of the ground states of these two compounds, where extended and overlapping exciton-phonon complexes in Ta_2NiSe_5 lead to a lowering of the crystal symmetry, whereas sparse and localized complexes in Ta_2NiS_5 cause local distortions, which is in good agreement with the excitonic insulator hypothesis in the former, but not in the latter.

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