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Excitonic effects on the optical response of quasi-one-dimensional Ta2NiSe5 and Ta2NiS5

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Cooper pairing of fermions is one of the most fundamental and successful concepts of condensed matter physics. The electron pair state is responsible for superconductivity, whereas the condensation of neutral electron-hole composite bosons is predicted to lead to an excitonic insulator state. Unlike superconductivity, the latter state does not exhibit a striking macroscopic manifestation of the quantum phase coherence and has no unequivocal experimental confirmation. Recent ARPES studies have considered quasi one-dimensional Ta2NiSe5 as a candidate for an excitonic insulator, which exhibits flattening of the valence band top below the transition temperature at 325 K [1]. In order to explore the excitonic states we perform a comparative optical study of the closely related ternary chalcogenides Ta2NiSe5 and Ta2NiS5. By means of wide-band spectroscopic ellipsometry we directly measured the complex dielectric function and unambiguously identified the excitonic doublet in both the compounds at low temperatures. Many-body interactions in these systems manifest themselves as a Fano interference of the discrete excitonic states with a band continuum. The determined exciton binding energy decreases with the increase in size of the chalcogenide atom and remains comparable with the optical gap energy. A gradual closing of the optical gap is observed in Ta2NiSe5 as the transition temperature is approached. The optical absorption spectra above the energy scale of the excitonic Fano resonances are dominated by a series of sharp interband transitions. Their steep temperature dependence corroborates strong electron-phonon interaction with a modification of the peak energies and widths proportional to the filling factor of optical phonon modes. The electron-phonon coupling may significantly reduce the effect of the local Coulomb attraction and stabilize the exciton condensation in the system.

[1] Y. Wakisaka et al., Phys. Rev. Lett. 103, 026402 (2009).

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