Oral presentation

Strong indications for an excitonic insulator state in $Ta_2(Ni, Co)(Se, S)_5$: A combined study of the spatial and electronic structure

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The excitonic insulator (EI) is a coherent electronic phase which was predicted to be realized in narrow gap semiconductors and semimetals with small band overlap driven by poorly screened coulomb interaction between conduction band electrons and valence band holes [1, 2]. Ta_2NiSe_5 is one of the few promising candidates for an EI. The compound undergoes a second order structural phase transition from an orthorhombic (*Cmcm*) to a monoclinic (*C2/c*) phase at $T_C \sim 328$ K [3]. A semimetal-to-insulator (SI) transition is observed at T_C which is associated with an EI transition due to electronic correlations [4, 5]. The EI scenario in Ta_2NiSe_5 is supported by optical measurements [4], X-ray photoemission spectroscopy (XPS), angle resolved photoemission spectroscopy (ARPES) [6], and by band structure calculations [7].

With S substitution for Se in Ta_2NiSe_5, the transition temperature decreases and the magnitude of the band gap increases towards the BEC side of the phase diagram. On the other hand, for $Ta_2(Ni,Co)Se_5$, both the transition temperature and the magnitude of the semi-metallic band gap are reduced, suggesting that Co substitution drives the system towards the BCS side of the phase diagram [4, 8].

Here, we will present a combined study of (i) temperature-dependent SXRD and (ii) NEXAFS on Ta $_2NiSe_5$, Ta_2NiS_5, and $Ta_2Ni_{0.9}Co_{0.1}Se_5$. Our SXRD data show a clear 2nd order structural phase transition in Ta_2NiSe_5 and $Ta_2Ni_{0.9}Co_{0.1}Se_5$, while no structural change was found in Ta_2NiS_5 down to 80 K. The transition is accompanied with a shortening in the bond lengths which increases the hybridization between the relevant Ta, Ni, and Se orbitals. This agrees well with our NEXAFS data: a stronger hybridization is seen for Ta_2NiSe_5 and $Ta_2Ni_{0.9}Co_{0.1}Se_5$ together with a significant T-dependent change of the spectral weight and, thus, a T-dependent charge transfer between these orbitals is observed. In contrast, the in- and out-of-plane spectra of Ta_2NiS_5 are predominantly isotropic and thus, no T-dependent charge transfer between the corresponding orbitals is observed. The results from our SXRD and NEXAFS studies are fully consistent with and show strong indications for an excitonic insulator scenario in Ta_2NiSe_5 and $Ta_2Ni_{0.9}Co_{0.1}Se_5$ whereas this can be ruled out for Ta_2NiS_5.

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