Bulletin of the American Physical Society

APS March Meeting 2022
Monday–Friday, March 14–18, 2022; Chicago

Session G15: Coupled Lattice and Electronic Phase Transitions: The Case of TaNiSe5
11:30 AM–1:54 PM, Tuesday, March 15, 2022
Room: McCormick Place W-183C

Sponsoring Units: DCMP DMP
Chair: Antoine Georges, College de France

Abstract: G15.00002 : Critical excitonic mode interacting with phonons in excitonic insulator $Ta_2Ni(Se_{1-x}S_x)_5$*
12:06 PM–12:42 PM

Abstract

Presenter:
Mai Ye
(Rutgers University)

Authors:
Mai Ye
(Rutgers University)

Pavel A Volkov
(Rutgers University)

Himanshu Lohani
(Technion-Israel Institute of Technology)

Irena Feldman
(Technion-Israel Institute of Technology)

Amit Kanigel
(Technion - Israel Institute of Technolog)

Girsh E Blumberg
(Rutgers University, New Brunswick)

Excitonic insulator is a quantum coherent phase resulting from formation of a macroscopic population of electron-hole pairs. For a semimetal with narrow overlap of the conduction and valence bands, a finite exciton binding energy could lead to excitonic instability. Candidate material Ta_2NiSe_5 shows a second-order structural phase transition at T_c=328K, with two mirror symmetries broken.

Using polarization-resolved Raman spectroscopy, we demonstrate that Ta_2NiSe_5 is an excitonic insulator below T_c [npj Quantum Mater. 6, 52 (2021)]. The order parameter of the structural transition is of quadrupolar symmetry. In this symmetry channel, we observe strongly coupled excitonic and optical phonon modes at low energy. The resulting Fano lineshape can be decomposed to reveal an overdamped exciton mode that exhibits critical softening on cooling towards T_c. In contrast, the energy of the bare optical phonons increases on cooling, which indicates that the phase transition does not result from lattice instability. We conclude that the transition is driven by the critical excitonic mode, and the transition temperature is enhanced by the coupling of this mode to the lattice modes of the same symmetry.

We further show that for $Ta_2Ni(Se_{1-x}S_x)_5$ the excitonic instability, the transition temperature T_c, and the magnitude of the structural change across T_c are suppressed with increasing sulfur content x [PRB 104, 045102 (2021); arXiv 2104.07032 (2021)]. The Fano lineshape at low energy persists up to x=0.67, up to which point $Ta_2Ni(Se_{1-x}S_x)_5$ has a semimetallic high-temperature phase. However, Ta_2NiS_5 is a semiconductor with more than 0.3 eV gap; the asymmetric lineshape is absent because the exciton mode appears at high energy. As both the exciton and optical phonons of Ta_2NiS_5 do not show critical softening, we conclude that its phase transition is driven by ferroelastic instability.

*Spectroscopic work by NSF DMR-1709161; sample growth and characterization by ISF 320/17.