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Title: Critical dynamics and quantum coherence in excitonic insulator Ta_2NiSe_5

Abstract:

Excitonic insulator is a quantum coherent phase resulting from the formation of a macroscopic population of electron-hole pairs. Because crystal structural symmetries are broken at the transition temperature T_c, it is difficult to determine whether a particular transition is of excitonic or structural origin. The nature of transition for the candidate material Ta_2NiSe_5 (T_c=328K) is currently under debate, with both excitonic [1] and structural [2] explanations proposed.

We report Raman-scattering results on Ta_2NiSe_5 to explore its critical excitonic fluctuations above T_c and emergent coherence below T_c [3]. The overdamped excitonic mode in the quadrupolar symmetry channel softens above T_c, while the optical phonon modes show no softening behavior. Moreover, the softening of the acoustic phonon mode [4] can be accounted for by its coupling to the excitonic mode, i.e. intrinsic ferroelastic instability is absent. On cooling, coherent superposition of band states at the gap edge gradually emerges. From these results, we demonstrate that the phase transition of Ta_2NiSe_5 is of excitonic nature. We further show that sulfur doping suppresses the excitonic contribution to ordering [5]. For Ta_2NiS_5, we identify a phase transition induced by ferroelastic instability at 120K, and a sharp in-gap exciton mode.

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