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Critical charge fluctuations and emergent coherence in a strongly correlated excitonic insulator¹

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Attraction between electrons and holes in semiconductors or semimetals can drive a transition to a macroscopically coherent state, characterized by a proliferation of particle-hole pairs – the excitonic insulator (EI). With only a few candidate materials known, formation of the EI breaks lattice symmetries, which makes it challenging to distinguish it from a structural transition. Recently, the attention has been attracted to the possible EI transition in the candidate material Ta₂NiSe₅ [1]; however, a structural origin of the transition has been also proposed [2]. I will present the results of an experimental study of Ta₂NiSe₅ by means of polarization-resolved Raman scattering, which allows to selectively probe the excitations with the symmetry of the order parameter, yielding access to the critical soft mode of the transition. We reveal an overdamped electronic collective mode, consistent with excitonic fluctuations in a semimetal, that softens close to the transition temperature. At the same time, the optical phonons do not soften, ruling out their role in the transition. Furthermore, on cooling, an emergence of a many-body gap is observed with signatures of coherent superpositions of band states at the gap edge. Its temperature dependence shows strong departures from mean-field theory, bearing analogy with that observed in strongly coupled fermionic superfluids. Finally, I will present the results on the evolution of the transition in the presence of gradual chemical substitution of Se by S, which allows to probe the effect of band structure changes on the EI. [1] Y. Wakisaka et al., Phys. Rev. Lett. 103, 026402 (2009); Y.F. Lu et al., Nat. Commun. 8, 14408 (2017) [2] E. Baldini et al., arXiv:2007.02909; A. Subedi, Phys. Rev. Materials 4, 083601 (2020). [3] P. A. Volkov et al., arXiv:2007.07344

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