Disentangling electronic and lattice instabilities in an excitonic insulator candidate Ta₂NiSe₅ by pump-probe Raman and luminescence spectroscopy

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<u>Kota Katsumi</u>^{1*}, Alexandr Alekhin¹, Sofia Michaela Souliou², Michael Merz^{2,3}, Amir-Abbas Haghighirad², Matthieu Le Tacon², Sarah Houver¹, Maximilien Cazayous¹, Alain Sacuto¹, and Yann Gallais¹

¹Matériaux et Phénomènes Quantiques, Université Paris Cité,Paris, France ²Institute for Quantum Materials and Technologies, Karlsruhe Institute of Technology Karlsruhe, Germany

³Karlsruhe Nano Micro Facility (KNMFi), Karlsruhe Institute of Technology, Eggenstein-Leopoldshafen, Germany

*Currently at Center for Quantum Phenomena, Department of Physics, New York University, New York, USA

In narrow gap semiconductors or semimetals, the Coulomb interaction between electrons and holes may lead to a spontaneous formation of excitons. These excitons are expected to condense and give rise to an unconventional insulating ground state called excitonic insulator [1]. Among the various exitonic insulator candidate materials, Ta₂NiSe₅ is a prototypical example because it has a direct band gap and has no instability at finite wave-vector, such as charge density wave order [2]. As the temperature is lowered, Ta₂NiSe₅ displays a semiconductor-to-insulator (SI) transition below the transition temperature $T_c = 326$ K, which is associated with an excitonic insulator transition due to electronic correlations [2,3,4]. However, the origin of the SI transition has been still elusive because it is accompanied by a structural transition from orthorhombic to monoclinic symmetry [5]. To investigate whether the SI transition is lattice or electronic-driven, pump-probe optical spectroscopy is a promising technique because it can track the electronic and lattice responses separately.

In this study, we first performed equilibrium emission spectroscopy for Ta_2NiSe_5 [6]. In addition to low-energy electronic and phononic Raman excitations, we found significant photoluminescence (PL) signal whose intensity increases below T_c , which we attribute to enhanced electron-hole interactions in the insulating phase. We further investigated the pump-probe Raman and PL intensity after photoexcitation, where the Raman and PL signals serve as reporters of the lattice symmetry and the insulating gap, respectively. In this talk, we will discuss the results of the pump-probe measurements in light of the lattice-driven versus electronic-driven scenarios for the SI transition.

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