Study demonstrates that Ta_2NiSe_5 is not an excitonic insulator

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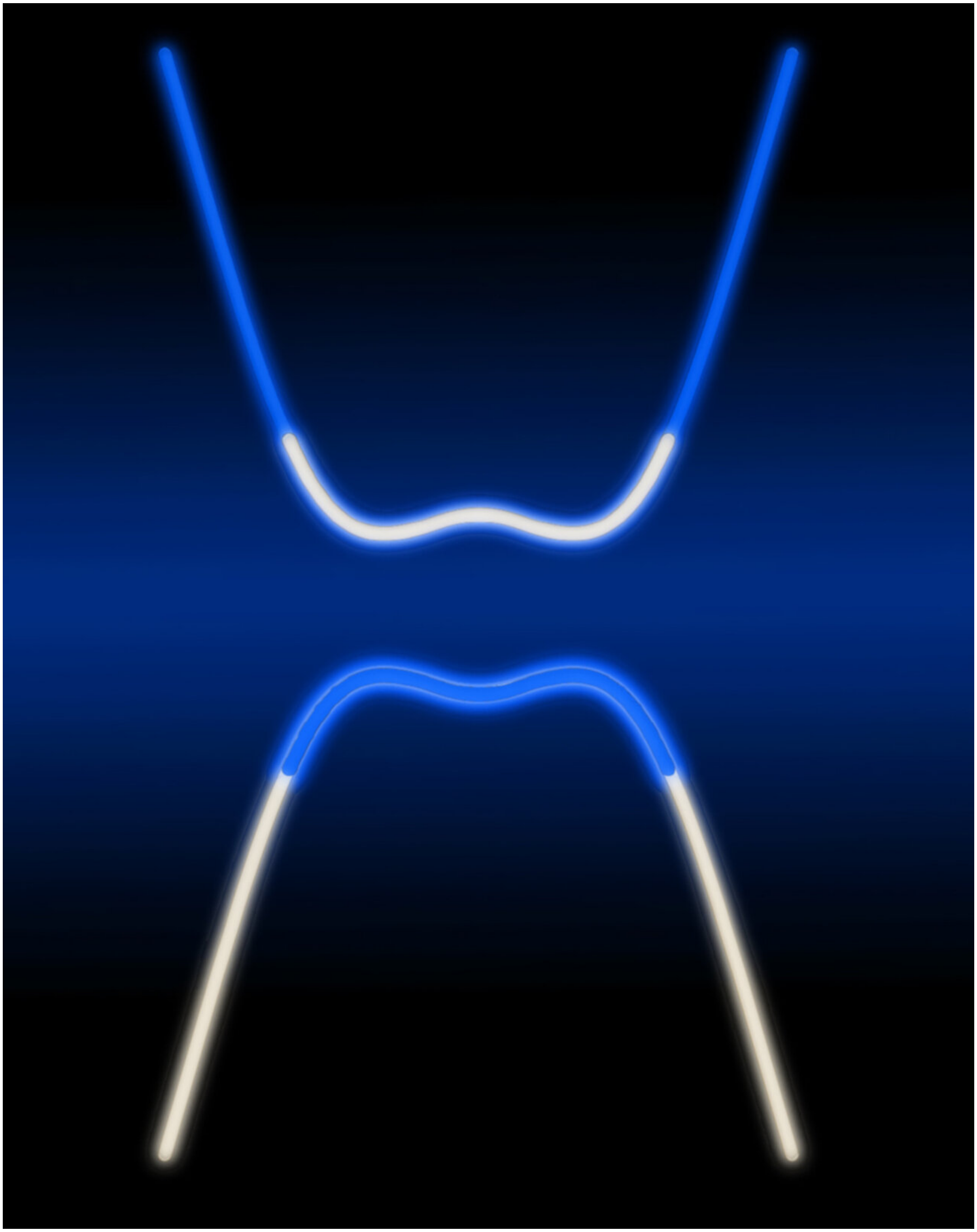

Characteristic electronic structure observed in candidate excitonic insulators. The shape of the electronic bands originates from the combined action of structural and electronic symmetry breaking. In Ta_2NiSe_5, the contribution of structural symmetry breaking is dominant and hinders any prospect of dissipationless energy transport. Credit: Jörg Harms, MPSD

The excitonic insulator is an electronically driven phase of matter that can occur in solids. Scientists are searching for ways to detect and stabilize this exotic order in candidate quantum materials because it could pave the way towards superfluid energy transport with no net charge (which is distinct from superconductivity). If realized, this phenomenon could lead to a new generation of devices where energy is transported at the nanoscale with high coherence and minimal dissipation.

However, spotting this phase in real solids has proven difficult so far. For the past two decades, it had been proposed that the quasi-two-dimensional solid Ta_2NiSe_5 may support an excitonic insulator phase above room temperature.

Above a critical temperature T_C = 328 K, this material crystallizes in a layered structure that consists of parallel Ta and Ni chains. At T_C, the system undergoes a semimetal-to-semiconductor transition, accompanied by a structural reorganization of the crystalline lattice. The scientific community has been engaged in an intense debate regarding whether this phase transition was induced by a purely electronic or a structural instability.

In a recently published study on *PNAS*, researchers in the U.S., Germany, and Japan probed the fundamental processes underpinning that transition via a joint experimental-theoretical approach.

Using an advanced experimental tool called time- and angle-resolved photoemission spectroscopy under highly controlled conditions, they exposed Ta_2NiSe_5 to a tailored laser pulse and recorded a real-time movie of the fundamental components of the excitons (i.e., electrons and holes) as well as the structural degrees of freedom. To resolve these microscopic phenomena, the movie had to achieve an ultrafast time resolution of less than a millionth of a billionth of a second.

Tracking the dynamics of the material's electronic and crystal structure after light excitation revealed spectroscopic fingerprints that are compatible only with a dominant order parameter of structural nature. This implies that the changes in the crystal structure actually hinder the development of electronic superfluidity in this quantum material.

"This work demonstrates that Ta_2NiSe_5 is not an excitonic insulator and that dissipationless energy transport is hampered by the prominent rearrangement of the crystal structure," says Nuh Gedik, Professor of Physics at the Massachusetts Institute of Technology (MIT), who coordinated the research.

"Our experiments provide a new approach to identifying the driving force behind spontaneous symmetry-breaking in a wide range of candidate excitonic insulators," adds lead author Edoardo Baldini, former postdoctoral fellow at MIT and now Assistant Professor of Physics at the University of Texas at Austin.

The findings were backed up by state-of-the-art calculations at several institutions who combined different theoretical techniques to understand the microscopic origin of these changes in Ta_2NiSe_5 with unprecedented accuracy.

"Confirming the microscopic mechanism driving this transition to be structural in nature required highly demanding and intertwined electronic and structural modeling that also provided relevant information on the impact of possible excitonic contributions," says Theory Director Angel Rubio from the Max Planck Institute for the Structure and Dynamics of Matter (MPSD) in Hamburg, Germany.

The groups of Eugene Demler at Harvard University, Andrew Millis at Columbia University, and Igor Mazin at George Mason University were partners in the theoretical collaboration. The experimental investigations were carried out at MIT, and the Ta_2NiSe_5 crystals used for this research were synthesized at the Max Planck Institute for Solid State Physics in Stuttgart, Germany, and at the University of Tokyo in Japan.

More information: Edoardo Baldini et al, The spontaneous symmetry breaking in Ta_2NiSe_5 is structural in nature, *Proceedings of the National Academy of Sciences* (2023). DOI: 10.1073/pnas.2221688120

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