Ultrafast spectroscopy uncovers single steps of phase transitions

Photoemission spectroscopy with ultrashort X-ray pulses reveals a so far unknown microscopic mechanism of an ultrafast insulator-metal phase transition. Through a self-amplified melting process - induced by photoexcited electrons - the insulating state disappears within a few femtoseconds.

Phase transitions are modifications in the properties of materials that occur, for example, through temperature and pressure changes. The best known phase transition is the transition from liquid water to water vapor. Phase transitions occur, nevertheless, also in other materials, for example, when they change from a superconducting to a normally conducting state, or from an insulator to an electrical conductor. In all these phase transitions the modifications of the macroscopic properties is based on several microscopic processes. In the case of superconductivity this is the formation of new states of two electrons and in the case of a transition from an insulator to metal this is the increase of free charge carriers upon heating. Until a few years ago, phase transitions could not be detected "fast enough" in order to reveal the timeframe of these microscopic processes. In recent studies, these limitations could be overcome by means of ultrashort optical and X-ray light pulses.

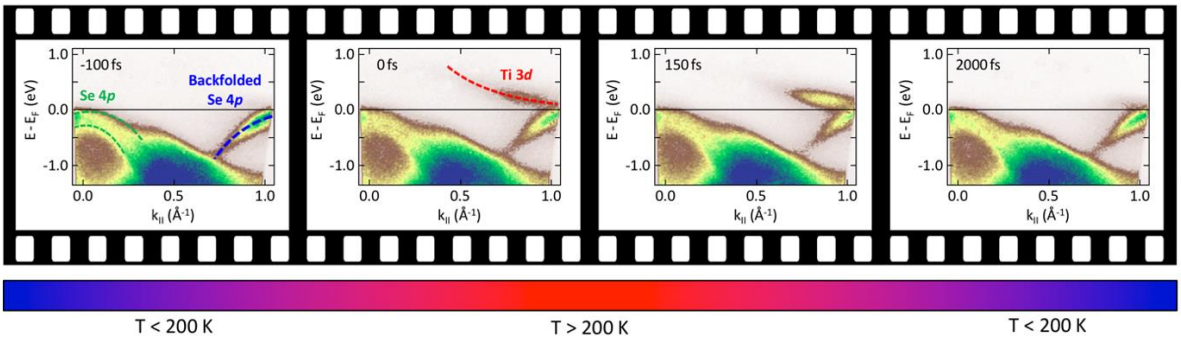
An international team of physicists from Kaiserslautern, Göttingen, Kiel, and Boulder (Colorado, USA) has now investigated a phase transition with a very high time resolution. For the material that was investigated, titanium diselenide ($TiSe_2$), an insulator-metal phase transition at a temperature of about 200 Kelvin is known. The team now reports in the prestigious journal Nature Communications how they monitor in real-time the microscopic processes during a phase transition that was initiated by an ultrashort optical excitation. Using time-resolved photoemission spectroscopy with ultrashort X-ray pulses, they observed the occupation of electronic energy states on time scales of femtoseconds (10^{-15} s), and followed how the energy gap between occupied and unoccupied electronic states was reduced by the dynamics of the excited electrons.

The central research result is that an optical excitation of the material induces a self-amplifiying melting process, which accelerates the transition from the insulating to the metallic state. After excitation of electrons across the electronic band gap a further very strong carrier multiplication process occurs that is driven by energy loss processes of the excited electrons. The researchers were able to demonstrate that these additional charge carriers further reduce the energy band gap, which on the other hand amplifies the multiplication process. Using a theoretical model that illustrates key aspects of the electronic dynamics, this self-amplifying effect could be identified as the underlying principle of the ultrafast phase transition.

The results obtained here are interesting from two other perspectives. Firstly, titanium diselenide is a complex material that neither complies with a description as a pure metal nor as a pure insulator/semiconductor. Interest in complex materials has increased in recent decades and they are therefore also intensively studied in basic research. Titanium diselenide is complex, since the electronic and lattice degrees of freedom are coupled in a complicated manner. At low temperatures, this material is in a so-called charge density wave insulator state in which the crystal structure and the electronic structure are different compared to the structure at room temperature. On the other hand the research on phase transitions shows

that by using ultrafast excitations, new non-equilibrium states of matter can be generated, with potential novel functionalities. Since the mechanism found here is considered to be universal for a large number of phase transition in materials, the research results open up the prospect to optically control and manipulate the individual steps of the phase transition.

Central parts of the research were carried out within the framework of two collaborative research centers of the German Research Foundation: SFB 1073 (Control of energy conversion on atomic scales) and SFB/TRR 173 (Spin+X), as well as the State Research Center OPTIMAS of the University of Kaiserslautern. The participating teams are from the University of Kaiserslautern, the University of Göttingen, the University of Kiel, as well as the University of Colorado and the National Institute of Standards in Boulder (Colorado, USA).



Photoemission spectroscopy of titan diselenide with high time resolution (femtoseconds, 10^{-15} s) allows the microscopic characterization of the electron dynamics during an optically induced phase transition. Ultrafast carrier multiplication was observed in real-time, which reveals how the insulating state disappears through a self-amplifying process. (Source: Stefan Mathias, Georg-August-Universität Göttingen)

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Self-amplified photo-induced gap quenching in a correlated electron material
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