The exciting story of TiSe2

One of the most beautiful theories of condensed matter is certainly the theory of conventional superconductivity. Called the BCS theory after the names of their creators (Bardeen, Cooper and Schrieffer), it explains within an effective model the zero resistance appearing in particular materials when temperature approaches the absolute zero. In this theory, electrons are bound together by some attractive interaction and form pairs which will condensate in a macroscopic quantum state at low temperature. Since breaking such a pair costs energy, it is somehow easier for the system to let them flow without losses, giving rise to conductivity with zero resistance.
Recently, in Fribourg, in collaboration with the PSI [1], we unveiled a similar mechanism in the layered compound TiSe2: here such a pairing is also at work, but between particles of different types. This material was intriguing scientists for a long time since the mystery of the origin of a charge density wave (CDW) at low temperature hold out against their assaults. In a CDW phase, electrons reorganize themselves and deviate from their initial highly uniform density, displaying a new periodicity different than that of the original atomic lattice. In some cases, this rearrangement is accompanied by a periodic lattice deformation, so that the total energy in the system is lowered. The key point for a CDW transition is the nature of its driving force. In the case of TiSe2, this issue was under discussion for a long time and now strong evidence has been found for the realisation of an exotic ground state, the excitonic insulator phase [2].

TiSe2 has an electronic structure near the Fermi energy comparable to that of a semiconductor, but its valence and conduction bands are slightly overlapping through an indirect gap. In the case of the excitonic insulator phase, the Coulomb interaction, weakly screened due to the low free carrier density, is strong enough to bind together an electron in the conduction band and a hole in the valence band. This bound pair is called an exciton. But this does not take into account temperature. Temperature kills the excitons by giving them enough energy to separate their components.

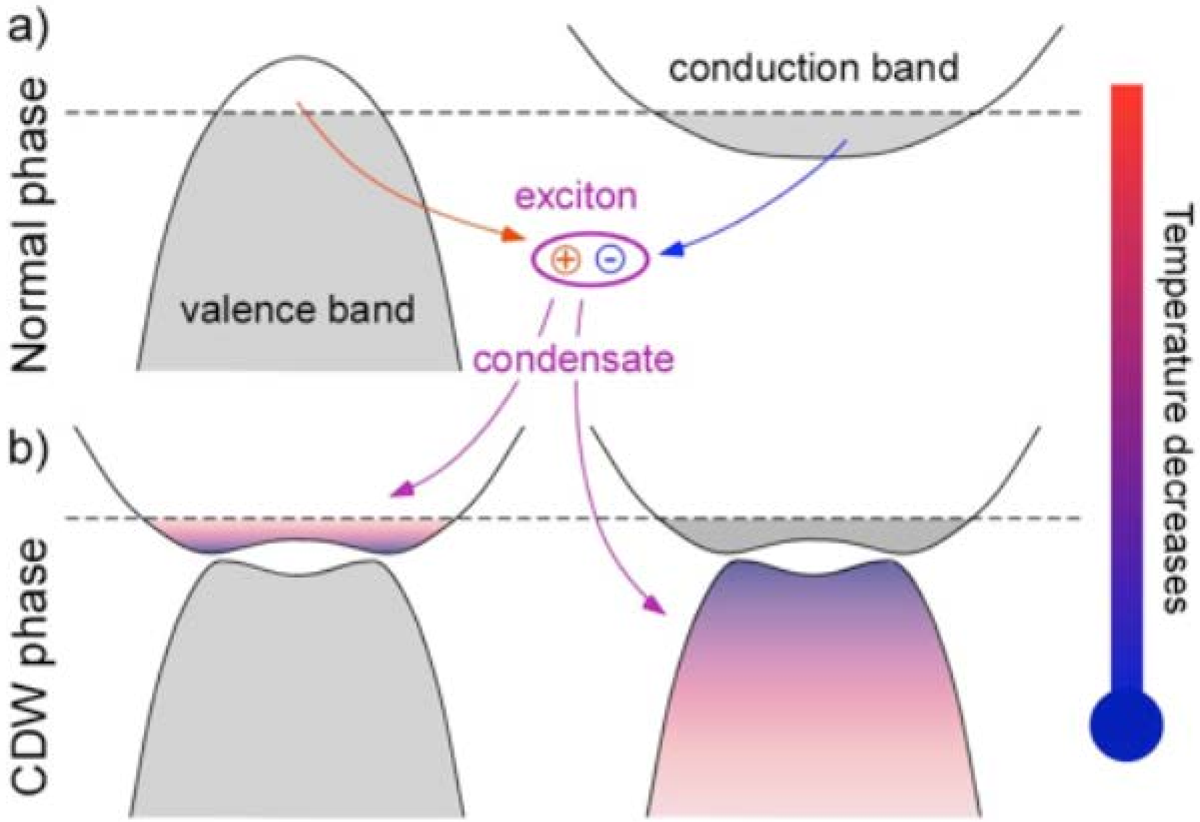

Figure 1: Schematic picture of the CDW phase transition in the excitonic insulator phase model. At high temperature (a), the electronic structure of TiSe2 near the Fermi energy consists of a valence band and a conduction which overlaps slightly and indirectly. At low temperature (b), excitons, consisting of holes in the valence band and electrons in the conduction band, condensate in a macroscopic quantum state, leading to the CDW phase.

However, once temperature decreases, a scenario similar to BCS superconductivity settles in, as illustrated schematically in figure 1. Since the valence and the conduction bands are indirectly overlapping, excitons will form spontaneously and under the pressure of their growing population, they will condense into a macroscopic state. Due to the nature of excitons, this state induces a new periodicity in the system arising naturally as the distance between the valence and the conduction bands. In other words, exciton condensation can lead the system towards a CDW phase with a purely electronic origin.

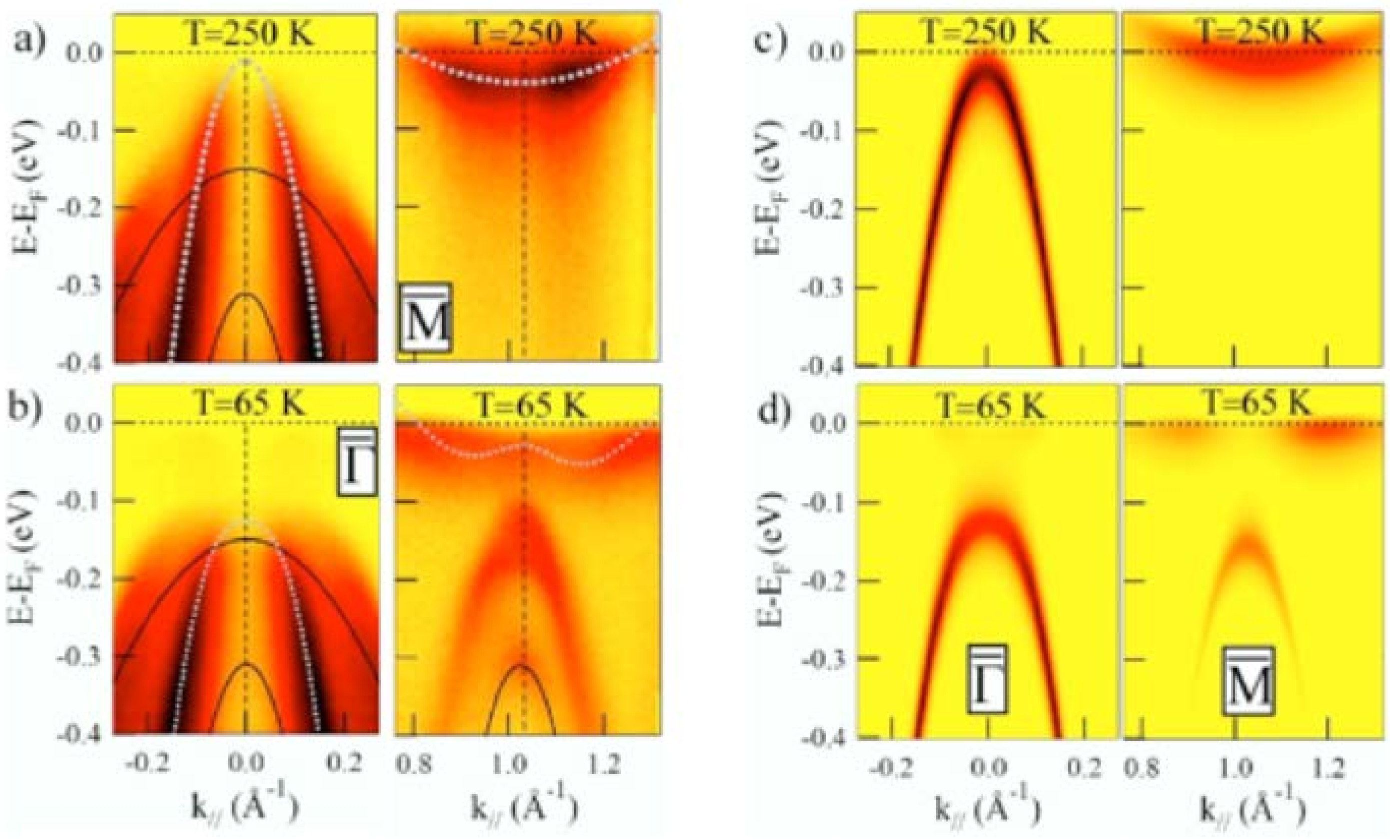

Figure 2: Figure (a) and (b) show the angle-resolved photoemission spectroscopy data taken at the X09LA beamline of the SLS on 1T-TiSe2 samples. At the temperature of 250K (a), the system is in the normal phase, above the transition. At 65K (b), it is in the CDW phase where one clearly sees at the border of the Brillouin zone (the so-called M point) replica of the bands originally situated in the center of the Brillouin Zone (the ? point). The right part of the figure shows the simulated intensity maps derived from the excitonic insulator model, above (c) and below (d) the CDW transition. The very good agreement between the experiment and the simulations gives strong evidence for the excitonic insulator phase as the low temperature ground state of 1T-TiSe2.

So far, so good. But how can one prove the existence of such a mechanism in TiSe2? At this point, photoemission comes into play. This experimental technique uses the photoelectric effect, which tells that the photons can kick off electrons from a solid. By collecting these electrons and measuring their velocity, one can infer precious information about the electronic structure of the solid. In the case of TiSe2, excitons leave a particular trace on the spectra, which can be compared to theory by computing the corresponding spectral function, a quantity intimately related to photoemission. Based on the excitonic insulator model, we simulated photoemission spectra exhibiting a very good agreement with experiment and giving consequently strong evidence for an excitonic origin in the CDW phase of TiSe2. Photoemission data together with their corresponding simulations are reproduced on figure 2.

More details on this work can be found here [3,4].

[1] H. Cercellier et al., Phys. Rev. Lett. 99, 146403 (2007)
[2] D. Jérome et al., Phys. Rev. 158, 462 (1967)
[3] C. Monney et al., CondMat/0809.1930v1.pdf
[4] C. Monney et al., CondMat/0809.1936v1.pdf