CORRELATED ELECTRON SYSTEMS

Emitting electrons through phonons

Ultrashort laser pulses create strain waves that generate highly mobile charges at an oxide interface. These charges propagate into the oxide layer destroying its antiferromagnetic ordering and insulating properties, providing insight into the physics of metal–insulator transitions.

Valerio Scagnoli and Urs Staub

Metal–insulator transitions are of key importance for understanding the fundamental electronic interactions that determine the physical properties of strongly correlated materials. Understanding such interactions remains a major challenge in materials science. In metal–insulator transitions, the conduction electrons of the metallic state become localized and the material transforms into an insulator. The process can be electronically, structurally or magnetically driven, and is often associated with electronic ordering phenomena, although in many systems, the exact origin and the fundamental mechanism driving the transition remain controversial[1]. The advent of X-ray sources with sub-picosecond time resolution has provided unprecedented opportunities for studying metal–insulator transitions on a timescale comparable to that of the associated interactions, offering the possibility to disentangle the driving forces behind them. Recently, several experiments have investigated the dynamics of electronic, magnetic and structural modifications after exciting the electronic systems with very short near-visible optical pulses that destroy the insulating state and the underlying electronic ordering[2–5]. Now, writing in *Nature Materials*, Michael Först and colleagues[6] show that, just after the excitation of phonons in a substrate by mid-infrared radiation pulses, the magnetic ordering of a nickelate layer grown onto it is suppressed by a shockwave generated from the substrate. Using picosecond X-ray pulses, they studied the dynamics of the magnetic properties, gaining insight into the correlation between the magnetic ordering and the conduction properties in the nickelate, without directly and simultaneously modifying them. The main finding is the demonstration of an emission of electrons through phonons at an interface of two materials that represents a novel approach to manipulating materials properties on ultrashort timescales.

Först and colleagues used the nickelate $NdNiO_3$, a prototypical perovskite that exhibits a metal–insulator transition occurring concomitantly with a structural distortion and a non-collinear antiferromagnetic ordering[7,8]. This transition is susceptible to small perturbations of the lattice, for example, nickelates can be tuned from a metallic paramagnetic ground state to an insulating state via strain[9]. In the configuration studied by Först and co-workers, the $NdNiO_3$ thin film was grown on a $LaAlO_3$ substrate. Picosecond pulses of mid-infrared radiation were used to directly and selectively excite a phonon band of the $LaAlO_3$ substrate, leaving the nickelate film largely unaffected. Immediately after the lattice excitation of the substrate, the nickelate was characterized using ultrafast resonant magnetic soft X-ray diffraction, a technique that provides direct access to the dynamic electronic and magnetic ordering of the material, by collecting the width and intensity of the magnetic reflection as a function of time. In particular, they used sub-picosecond X-ray pulses from the free electron laser Linac Coherent Light Source at the Stanford Linear Accelerator Laboratory. The Bragg peaks of the diffraction pattern provide information about the long-range magnetic ordering. The reduction of the integrated intensity of the Bragg peak directly shows how strong and how fast the magnetism is suppressed, whereas the change of its width gives a direct measure of the extent of the magnetic ordered region, which shrinks as a function of time after the excitation. This is in contrast to the effects of raising the temperature or exciting the nickelate film directly above its electronic bandgap by using laser pulses[10]. In the latter case, the authors did not observe any change in width of the Bragg peak, although the intensity was similarly reduced due to thermal fluctuations of the spins, showing that the system does not simply break up into smaller domains when heated or exposed to a near-infrared optical pulse.

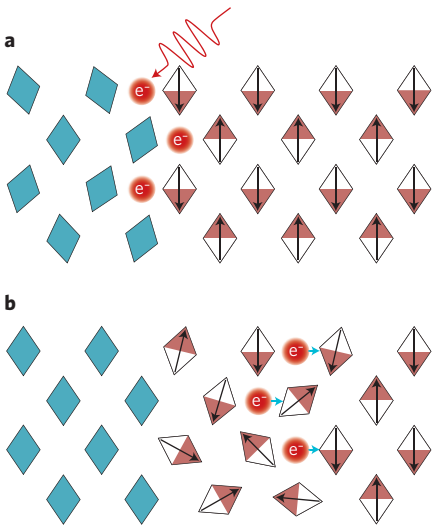

Figure 1 | Schematic representation of a shockwave excitation and evolution with time. **a**, A mid-infrared pulse (red wavy line) creates a strain wave in the $LaAlO_3$ substrate (blue diamonds) that releases a burst of electrons (red circles). Immediately during and after the excitation, the magnetic structure (black arrows) in the $NdNiO_3$ thin film (brown and white diamonds) is still mostly ordered. **b**, The electrons move as indicated by the blue arrows, scrambling the ordered arrays of magnetic moments in the nickelate thin film. One picosecond after the mid-infrared excitation, the system has undergone a transition to a highly magnetic disordered state close to the interface due to the supersonic mobility of the electrons.

The results reported by Först and colleagues indicate therefore that the effective thickness that remains magnetically ordered is shrinking on ultrafast timescales, while the antiferromagnetic order at the interface region is completely suppressed. Exciting the substrate phononic system and probing the film with terahertz pulses show that the insulating character is suppressed on a similar timescale. This confirms that the suppression of the magnetic order is indeed associated with the metal–insulator transition. Interestingly, the emitted wave travels faster than the speed of sound in the bulk nickelate material, which is generally believed to be the upper limit of heat and strain transport through a crystal.

Based on the supersonic speed of the wave, Först and colleagues argue that the emitted shockwave is of electronic origin; this process is shown in Fig. 1. The phonons excited at the $LaAlO_3$ substrate generate electrons at the oxide interface (Fig. 1a), which then propagate at supersonic speed into the $NdNiO_3$, destroying its antiferromagnetic ordering (Fig. 1b). The general instability of the metal–insulator transition in nickelates might be directly related to the pronounced detrimental effect of the fast-travelling electrons on the magnetic and electronic properties. It cannot be fully excluded that the wave travels with the speed of sound, since its value is not really known in the strained film. This alternative scenario of a strain wave emitted from the interface that creates the metal–insulator transition without transporting the heat would be equally intriguing.

This study underlines the importance of being able to selectively excite the lattice to control electronic properties, such as the electrical resistivity of a material or its magnetic state. Being able to indirectly manipulate the electronic properties through a substrate layer offers even more interesting options for possible applications. For example, one could imagine the creation of a two-dimensional electron gas at an interface on ultrafast timescales. On the other hand, understanding the microscopic origin of the emission of electrons that occurs at the interface remains experimentally and theoretically challenging. Whether such excitations through lattice electronic waves can be created at other interfaces and are strong enough to change properties of other interesting, strongly correlated materials remains an open question. ❐

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2D MATERIALS

Ultrafast exciton dynamics

Time-resolved measurements of the exciton dynamics in tungsten diselenide monolayers reveal ultrafast radiative recombination of the exciton ground state (~150 fs) and the interplay between optically bright and dark excitons.

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Two-dimensional semiconductor structures based on monolayer transition metal dichalcogenides (TMDCs) such as MoS_2 and WSe_2 show very strong light–matter interaction, with absorption on the order of 10% per monolayer in the visible region of the optical spectrum. This makes these 2D materials attractive for optoelectronics applications. Their optical properties are governed by robust excitons, electron–hole pairs tightly bound by Coulomb attraction. Although indispensable for designing future devices, a clear microscopic understanding of the exciton structure and recombination is still lacking.

Rupert Huber and colleagues report in *Nature Materials* a detailed investigation of the exciton characteristics in WSe_2 monolayers[1]. Their ultrafast measurements consist of two steps: first a femtosecond pump pulse in the visible range creates the exciton in its ground state; second, an infrared probe pulse induces a transition from the exciton ground state to an excited one. These experiments provide new information on both the exciton dynamics, such as its radiative lifetime, and on the internal structure of this two-particle complex.

In TMDC monolayers reduced screening of the Coulomb interaction and ideal 2D confinement lead to gigantic exciton binding energies E_b ~500 meV, ensuring that the excitons are stable at room temperature. The onset of optical absorption is given by E_b below the free-carrier bandgap. Analogous to two-particle complexes such as the hydrogen atom, excitons in TMDC monolayers possess a centre-of-mass momentum K and an internal quantum state that accounts for the motion of the electron and the hole relative to each other, with the ground state labelled 1*s* and the first excited states being 2*s* and 2*p*. In contrast, however, to the hydrogen atom, the spacing and order of the 1*s*, 2*s*, 2*p*, and so on, levels cannot be explained with simple Rydberg series, as recently shown by one- and two-photon optical spectroscopy experiments based only on interband transitions[2].

Despite providing a wealth of information, these optical interband measurements are restricted to a small subset of excitons due to symmetry and momentum conservation: the optically active excitons that lie in the so-called radiative cone with $K \sim 0$. Huber and co-workers investigate for the first time in TMDC monolayers intra-excitonic transitions between different exciton quantum states, as initially demonstrated in bulk germanium and silicon[3,4]. In contrast to interband absorption that can be used to measure the ability to generate bound electron–hole pairs, intra-excitonic absorption experiments probe existing excitons through transitions from the 1*s* ground state to higher relative-momentum states, such as 2*p*.

In the two-colour pump–probe scheme used by Huber and colleagues, the exciton 1*s* ground state is photogenerated by a femtosecond pulsed laser and the intra-excitonic transition is then probed with a phase-locked mid-infrared pulse. The measurements reveal a clear absorption resonance for an infrared photon energy of 165 meV. With this photon energy, the 1*s* exciton can be promoted to the 2*p* orbital exciton resonance, in agreement with the 1*s*–2*p* energy spacing measured by interband excitation spectroscopy[5].

Intra-excitonic transitions $1s \rightarrow 2p$ can be induced for all values of the centre-of-mass momentum K, not only for the $K \sim 0$ excitons within the light cone (bright states)[6]. Intra-excitonic resonances can thus provide access to the entire exciton population in momentum space, including