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Trapped in a magnetic cage



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Nachrichten, Termine, Experten

Controlling how electrons interact in semiconductors is crucial for the development of electronic and optical devices. Quantum scientists have now discovered a surprisingly efficient new mechanism to adjust the interaction between electrons and even restrict their motion to a single dimension: magnetic order.

Next-generation electronics and optoelectronics depend on ultra-compact semiconductor devices. Atomically thin layers of so-called transition metal dichalcogenides, which can be exfoliated from bulk crystals using a simple adhesive tape, have thus been in the focus of solid state physics. As electrons in these ultrathin layers can only move in two dimensions and hardly avoid each other, they interact particularly strongly, which significantly influences the materials' optical properties. When light is absorbed in these solids, electrons can be excited to higher energy states, leaving behind a positively charged vacancy, known as a hole, at their original position. Due to their opposite charges, the electron can orbit the hole and form an atom-like bound state — an exciton. These particles give the material entirely new optical properties, which can be precisely tailored through structural modifications such as stacking different atomically thin layers — an exciting prospect for the development of ultrathin solar cells and LEDs. However, practical applications based on these materials are still difficult because, for example, inefficient exfoliation techniques are not compatible with production on an industrial scale.

Looking for alternatives to confine and control electrons, an international team of physicists from Regensburg, Ann Arbor, Prague and Dresden has found a new mechanism in the exceptional material chromium sulfide bromide (CrSBr) that does not rely on structural modifications. This material exhibits a layered structure in which the spins of the electrons - a quantum mechanical property that creates a magnetic moment – all point in the same direction within one layer, while the temperature dictates the relative orientation of the spins in the adjacent layers. An antiparallel alignment of these spins in adjacent layers could restrict the electron motion to a single layer, creating a "magnetic cage".

To test this hypothesis, the team of Prof. Rupert Huber in Regensburg deployed ultrafast laser pulses with a duration of only a few femtoseconds – a hundred trillion times faster than the blink of an eye. These incredibly short light flashes excited excitons in a CrSBr flake. A second ultrashort light pulse, in the mid-infrared region of the electromagnetic spectrum, interrogated the atom-like energy structure of these excitons by driving distinct transitions between different orbitals. This technique resembles a slow-motion camera for electrons, that precisely captures the behavior of excitons on their intrinsic timescales, providing insights into their binding energy, the spatial confinement, and the recombination dynamics.

In a close theory-experiment collaboration, the researchers found a way to control in-situ how tightly these excitons are bound together in a CrSBr crystal. These high-quality crystals required for this study were grown by the group of Prof. Zdeněk Sofer at the University of Chemistry and Technology Prague. By systematically varying the lattice temperature, the team observed a sudden shift in the excitonic binding energy that is directly linked to changes of the magnetic order of the material.

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A sophisticated quantum theory developed by the research group of Prof. Mackillo Kira at the University of Michigan provided microscopic insights into how the magnetic order affects the excitons in CrSBr. The team found that, in essence, the dimensionality of the exciton changes in unison with the magnetic phase of the crystal. As proposed, at low temperatures, the antiparallel spin alignment traps excitons within a single layer. Combined with the unusual crystal structure of CrSBr, the magnetic cage further restricts the motion of excitons within the plane. As a result, the excitons are essentially limited to a single dimension resulting in high binding energies - even in crystals hundreds of layers thick. As the temperature is increased, the spin alignment is lost, however, which effectively opens the magnetic cage. Consequently, the excitons are freed and can spread out in all spatial dimensions across several layers, which significantly reduces their binding energy while their lifetime is extended.

"It was fascinating to see how we could completely change the behavior of these excitons simply by lowering the temperature. To confirm that this behavior truly originates from the magnetic phase transition, we applied an external magnetic field. This allowed us to fine-tune the temperature at which the magnetic cage is opened", says Marlene Liebich, the lead author of the study. "The magnetic order is a new tuning knob for shaping excitons and their interactions. This could be a game changer for future electronics and information technology," adds Dr. Niloufar Nilforoushan, one of the corresponding authors of the study.

A second collaborative study with colleagues from Dresden, New York, and Prague, which appeared back-to-back in the scientific journal Nature Materials beautifully complements this picture. This work reports on the magnetic confinement of excitons by analyzing how light reflects off the material's surface. Dr. Florian Dirnberger, an author on both publications, expresses his excitement: "Surprisingly, the magnetic confinement is so effective that one can distinguish excitons in different atomically thin layers of the material." Indeed, the team found that the excitons on the surface exhibit noticeably different properties compared to those inside the material.

These discoveries open exciting opportunities for future spintronic and magneto-optical applications and the capability to tailor phase transitions on demand – a promising playground for next-gen¬er¬a¬tion information processing technologies. The identification of surface excitons further expands these possibilities, offering new functionalities for instance for sensing and optoelectronic devices.

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The antiparallel spin alignment in adjacent layers of the van der Waals crystal CrSBr confines Coulomb-bound electron-hole pairs (excitons) into one dimension with strongly different binding energies in the bulk and at the surface. Brad Baxley (parttowhole.com)