Inducing ferromagnetism with light

Light-induced ferromagnetism in moiré superlattices
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Recommended with a Commentary by Liang Fu, Massachusetts Institute of Technology

Imagine the ability to transform matter from one state to another with light. This could lead to useful devices and powerful technologies, such as ultrafast photoconductive switches and energy-efficient laser writing. In quantum materials, it is often observed that optical excitation weakens or destroys pre-existing orders in the equilibrium state, as seen in light-induced demagnetization. Now, the highlighted work reports an extraordinary phenomenon in a 2D semiconductor heterostructure: light induces ferromagnetic behavior that is absent in equilibrium.

The platform under study is a WSe_2/WS_2 heterobilayer, where recent experiments found a plethora of novel electronic states in equilibrium [1, 2, 3]. In this system, the interesting physics comes from the moiré superlattice formed due to the lattice mismatch between the two atomic layers, see Fig (a). The moiré structure introduces a slowly varying periodic potential for doped charges and thus leads to minibands. At nearly zero-degree twist angle, the system is essentially an array of quantum dots forming a triangular lattice, with a lattice spacing of 7.5nm. Electron hopping between adjacent sites $t \sim 1$meV is small compared to Coulomb energy on the moire lattice scale. Thus, WSe_2/WS_2 is an excellent realization of the extended Hubbard model on an emergent lattice [4, 5]. Its phase diagram as a function of filling can be mapped out by tuning the doping density with the gate voltage.

In line with the Hubbard model physics, recent experiments on WSe_2/WS_2 found a Mott insulator at the filling of $n = 1$ hole per moiré unit cell, as well as generalized Wigner crystals at various commensurate fractional fillings [1, 3]. In the latter case, holes on the triangular lattice self-organize into a periodic superstructure to minimize the long-range Coulomb repulsion. For example, a $\sqrt{3} \times \sqrt{3}$ charge order is formed at $n = \frac{1}{3}$, as observed by STM [6]. The existence of such a variety of correlated insulators shows the richness of many-body physics on the moiré superlattice.

The highlighted work by Wang *et al* finds a new surprise in WSe_2/WS_2 when the system is driven out of equilibrium by optical excitation. The authors perform optical reflection measurements with both left and right circularly polarized lights, at photon energy near the WSe_2 1s-exciton resonance. A difference in reflection of left and right circularly polarized

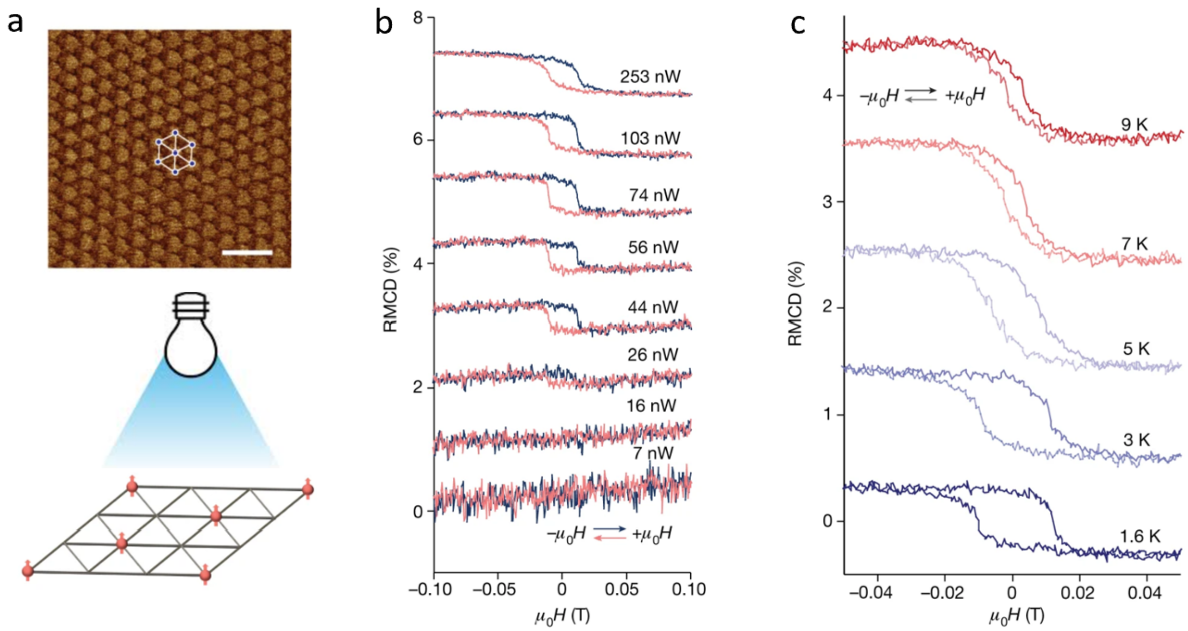

Figure 1: Adapted from the highlighted work. The piezoresponse force microscopy image in (a) shows the moiré superlattice of WSe_2/WS_2. At $n = \frac{1}{3}$ filling of the moiré unit cell, the ground state is a generalized Wigner crystal with $\sqrt{3} \times \sqrt{3}$ charge order. Optical reflection measurements reveal the emergence of magnetic hysteresis loop above a threshold excitation power as shown in (b) and (c), indicating light-induced ferromagnetism.

light is induced by the presence of an external magnetic field or spontaneous magnetization, a phenomenon known as magnetic circular dichroism (MCD). In a previous work from Cornell group [2], the optical response of WSe_2/WS_2 is measured as a function of magnetic field H, hole density and temperature. The results are insightfully analyzed and reveal a wealth of information about the magnetic property of strongly interacting holes on the moire superlattice. No MCD signal is observed at $H = 0$, showing the absence of ferromagnetism down to 1.6K.

Wang *et al* goes further to study the dependence of MCD signal on optical excitation power. Figure (b) shows the MCD signal versus the magnetic field H for different excitation powers near the filling $n = \frac{1}{3}$. Remarkably, as the power increases above 16nW, a small hysteresis loop appears in the MCD$-H$ curve, with a width about 20mT and an amplitude of about 0.5%. Magnetic hysteresis loop is the typical behavior of a ferromagnet and demonstrates its ability to retain magnetization after the field is removed. The appearance of hysteresis loop due to optical excitation is strong evidence of light-induced ferromagnetic ordering in WSe_2/WS_2.

The observed phenomenon is as surprising as it is fascinating. How could it be that the photoexcited state is ferromagnetic when the equilibrium state is not? The authors propose that optically injected interlayer excitons may mediate a ferromagnetic exchange interaction between holes. There are many open questions. Why is the width of the hysteresis loop—while strongly temperature dependent—nearly independent of excitation power? Why does the amplitude of the remnant MCD signal change non-monotonously with temperature? With the hope of better understanding, I suggest below new experiments that may provide useful information.

In the current experiment, the incident light is used to both excite and probe the system. The laser beam is modulated at 50.1kHz to produce an alternating left and right circularly polarized light wave, and the MCD signal is detected by lock-in technique at this frequency. In future studies, it is highly desirable to perform pump-probe measurements with independently controlled pump and probe light beams. With this technique one can study the dependence of light-induced ferromagnetism on the frequency, polarization, and power of the pump light, while keeping the probe beam at exciton resonance with low power for high-sensitivity detection. By varying the time delay between pump and probe pulses, one can gain information about the dynamics of the ferromagnetic state.

An intriguing question is the relationship between light-induced ferromagnetism and correlated insulating states at fractional fillings. While the magnetic hysteresis is observed throughout the low doping regime (not just near $n = \frac{1}{3}$), the MCD signal is enhanced and survives to much higher temperature at $n = \frac{1}{7}$, where the equilibrium state is a generalized Wigner crystal with a larger lattice constant and thus weaker Coulomb interaction energy than $n = \frac{1}{3}$. Is charge order still present in the light-induced ferromagnetic state?

The discovery of light-induced ferromagnetism in WSe_2/WS_2 is likely just the tip of the iceberg. Thanks to the narrow bandwidth, strong interaction and topological effect [7, 8], semiconductor moire materials may host a multitude of non-equilibrium states that can be optically controlled, waiting to be uncovered and understood. Throw light on them!

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References

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