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Exploring atomically thin van der Waals metals for spintronics

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14. ABSTRACT
In the first year of the project period, our lab has focused on developing new experimental techniques to control the valley magnetization in 2D transition metal dichalcogenide (TMD) semiconductors. Although control of the electron valley degree of freedom in 2D TMD semiconductors by light and magnetic fields is now routine, practical valleytronics requires pure electrical control of the electron valley. Also, the demonstrated magnetic control of the electron valley needs very high magnetic fields and is very inefficient. To overcome these issues we have developed techniques to fabricate both high electron mobility and strain tunable monolayer TMD transistor devices. The strain-engineered transistor devices have allowed us to demonstrate a new valley magnetoelectric effect, i.e. the pure electrical generation of valley magnetization in monolayer TMDs (published in Nature Materials). The resultant valley magnetization has been probed and imaged by magneto-optical microscopy techniques...

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Below I summarize my major achievements under the support of the AFOSR YIP award.

(1) Demonstration of valley magnetoelectricity in monolayer MoS_2

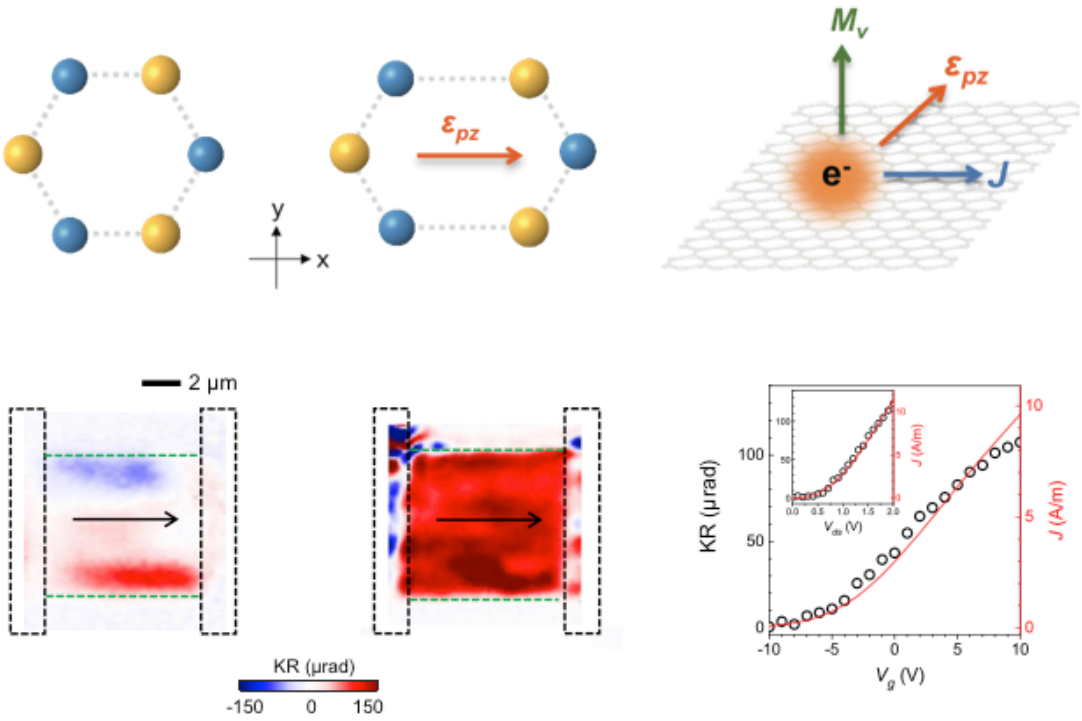

Figure 1. Valley magnetoelectricity in monolayer MoS_2: Top: crystal structure of unstrained (left) and uniaxial strained (middle) monolayer MoS_2. Schematic illustration of the valley magnetoelectric effect (right): an in-plane bias current perpendicular to the strain-induced polar axis generates an out-of-plane valley magnetization. Bottom: Kerr rotation image of unstrained (left) and uniaxial strained (right) monolayer MoS_2 transistor (the arrow shows the bias current direction). The right figure shows the gate voltage dependence of the Kerr rotation signal and the corresponding bias current density in the uniaxial strained device. The inset shows the bias voltage dependence of the Kerr rotation signal and the current density at zero gate voltage.

Magnetoelectric effect, the control of material magnetization by electric fields and vice versa, plays an important role in applications of magnetic switching and spin-charge conversions. Experimental studies on the magnetoelectric effect have so far focused on the generation and control of electron *spin* magnetizations. Electrons in monolayer MoS_2 possess a new *valley* degree of freedom, which is associated with the K and K' points in the Brillouin zone. The electron valley carries an extra orbital magnetic moment (valley magnetic moment), which can be controlled by light polarizations and magnetic fields. Practical valleytronics devices, however, require efficient control of the *valley* magnetization by pure electric fields. Such valley magnetoelectricity is, however, forbidden in materials with three-fold rotation symmetry, such as monolayer MoS_2. To achieve valley magnetoelectricity in monolayer MoS_2, we break the three-fold rotation by application of in-plane uniaxial strain along high symmetry axes (figure 1), which induces an in-plane polar axis. Current biasing perpendicular to the polar axis then generates a net out-of-plane valley magnetization. To image the spatial distribution of the

current-induced valley magnetization, we employ magneto-optical Kerr effect (MOKE) microscopy, in which the Kerr rotation signal is directly proportional to the magnetization. Figure 1 compares the Kerr rotation image of unstrained and uniaxial strained monolayer MoS_2 transistors under current biasing. While finite Kerr rotation signal can only be seen on the device edges for the unstrained case (due to the valley Hall effect), that for the strained case is uniformly distributed throughout the entire device channel, indicating the electrical generation of a uniform sample magnetization. A unique feature of valley magnetoelectricity in a 2D semiconductor is its gate tuneability. Figure 1 shows that the valley magnetization, similar to the channel current, can be effectively turned on and off by a back gate voltage. Our experimental study opens up the possibility of valley-charge conversion in valleytronics devices. This study was conducted in collaboration with Jie Shan's group at Cornell University. The result has been published in *Nature Materials*.

(2) Electric field switching of magnetic states in bilayer CrI_3

Controlling magnetism by purely electrical means is a key challenge to better information technology. A variety of material systems, including ferromagnetic metals, ferromagnetic semiconductors, multiferroics and magnetoelectric materials, have been explored for the electric-field control of magnetism. The recent discovery of two-dimensional (2D) van der Waals magnets has opened a new door for the electrical control of magnetism at the nanometre scale through a van der Waals heterostructure device platform. In 2D CrI_3, each individual monolayer material is a ferromagnet with the Cr^{3+} spins all lined up in the out-of-plane direction. The monolayers stack up vertically with an antiferromagnetic arrangement of spins. As a result, the spatial inversion and time reversal symmetries are both broken, allowing control of its magnetic order by the magnetoelectric effect. By fabricating dual-gated bilayer CrI_3 field effect devices, which allows independent tuning of the vertical electric field and the sample doping density, we have been able to efficiently generate an out-of-plane magnetization in the materials purely by a vertical electric field (figure 2). In contrast, no electric field-induced magnetization can be created when the sample is in the ferromagnetic state, which is consistent with the restored inversion symmetry of the material. Furthermore, the electric field-induced magnetization in the antiferromagnetic state helps the material gain Zeeman energy under a constant magnetic bias. As a result, the material can be switched from an antiferromagnet to a ferromagnet, and vice versa, when the external electric field exceeds a critical value. Indeed, robust and repeatable electric field switching of the magnetic orders in bilayer CrI_3 has been demonstrated, opening up great potentials to realize novel non-volatile memory device concepts. This study was conducted in collaboration with Jie Shan's group at Cornell University. The result has been published in *Nature Materials*.

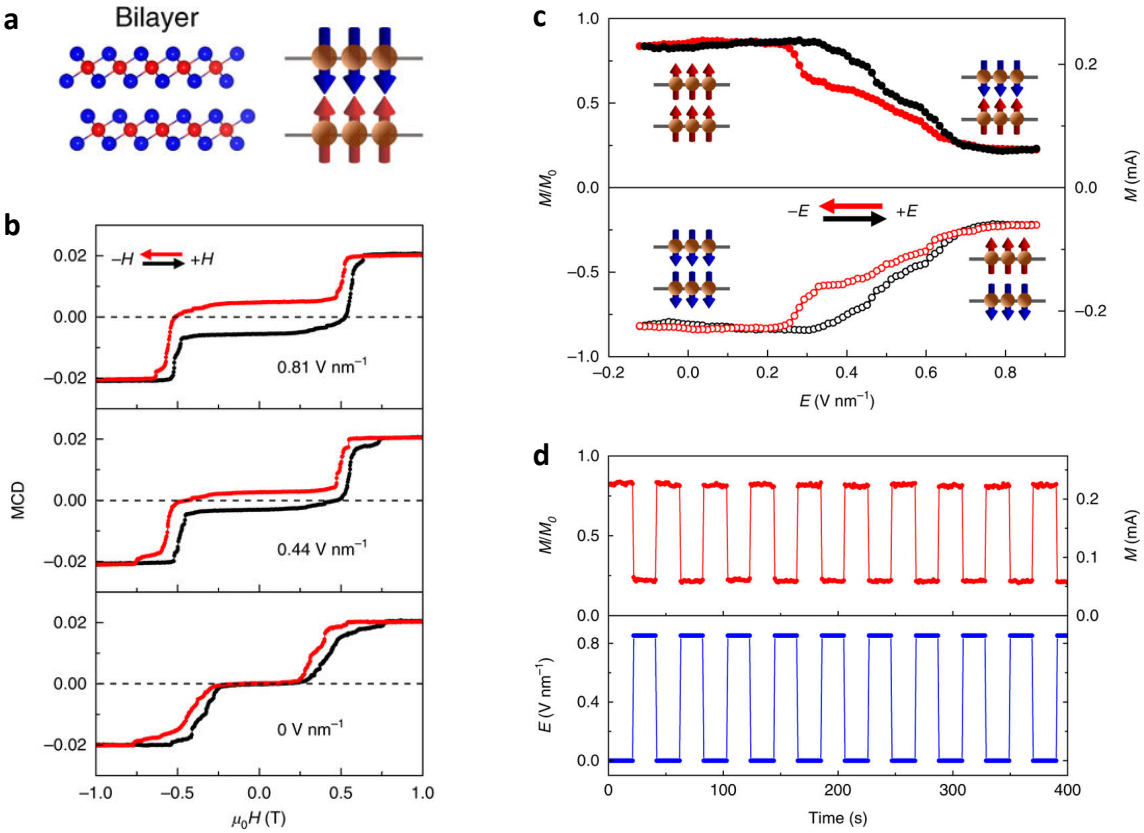

Figure 2. Electric field switching of magnetic orders in bilayer CrI_3: **a**) Crystal structure and interlayer antiferromagnetic ground state of bilayer CrI_3. **b**) Sample magnetization versus magnetic field under different vertical electric fields. An electric field-induced magnetization in the antiferromagnetic state is seen. **c**) Reversible electrical switching between the antiferromagnetic and the ferromagnetic states in bilayer CrI_3 under constant magnetic bias. Top and bottom panels are for positive and negative magnetic biases, respectively. **d**) Demonstration of robust and repeatable electrical switching of magnetic orders in bilayer CrI_3. Top and bottom panels are the time dependences of sample magnetization and of the applied vertical electric field, respectively.

(3) Switching of magnetic orders in 2D CrI_3 by electrostatic doping

The atomic thickness of 2D materials provides a unique opportunity to control their properties and to drive electronic phase transitions by electrostatic doping. Although our experiment above has demonstrated control of the magnetic order in bilayer CrI_3 by electric fields, this approach is limited to non-centrosymmetric materials magnetically biased near the antiferromagnet–ferromagnet transition. Practical applications desire electrical switching of magnetism under *zero* external magnetic fields. Recently, we have been able to achieve this goal in bilayer CrI_3 by electrostatic doping via gating. This has been made possible by the device design shown in figure 3. The device consists of bilayer CrI_3 sandwiched between graphene electrodes, which enable efficient interlayer charge transfer for introduction of doping, and with top and bottom gates. Applications of symmetric top and bottom gate voltages can introduce high doping density into the bilayer CrI_3. Furthermore, we can monitor the conductance of the device as a function of gate voltage, in which a tunnel field effect transistor action with high on-off ratio is seen.

Under high electron doping density (i.e. high positive gate voltage), the material can be switched from an interlayer antiferromagnetic ground state to a paramagnetic ground state under *zero* magnetic field. Such doping controlled magnetism provides a more general approach for switch magnetic orders in 2D CrI_3 by electric field effects. This study was conducted in collaboration with Jie Shan's group at Cornell University. The result has been published in *Nature Nanotechnology*.

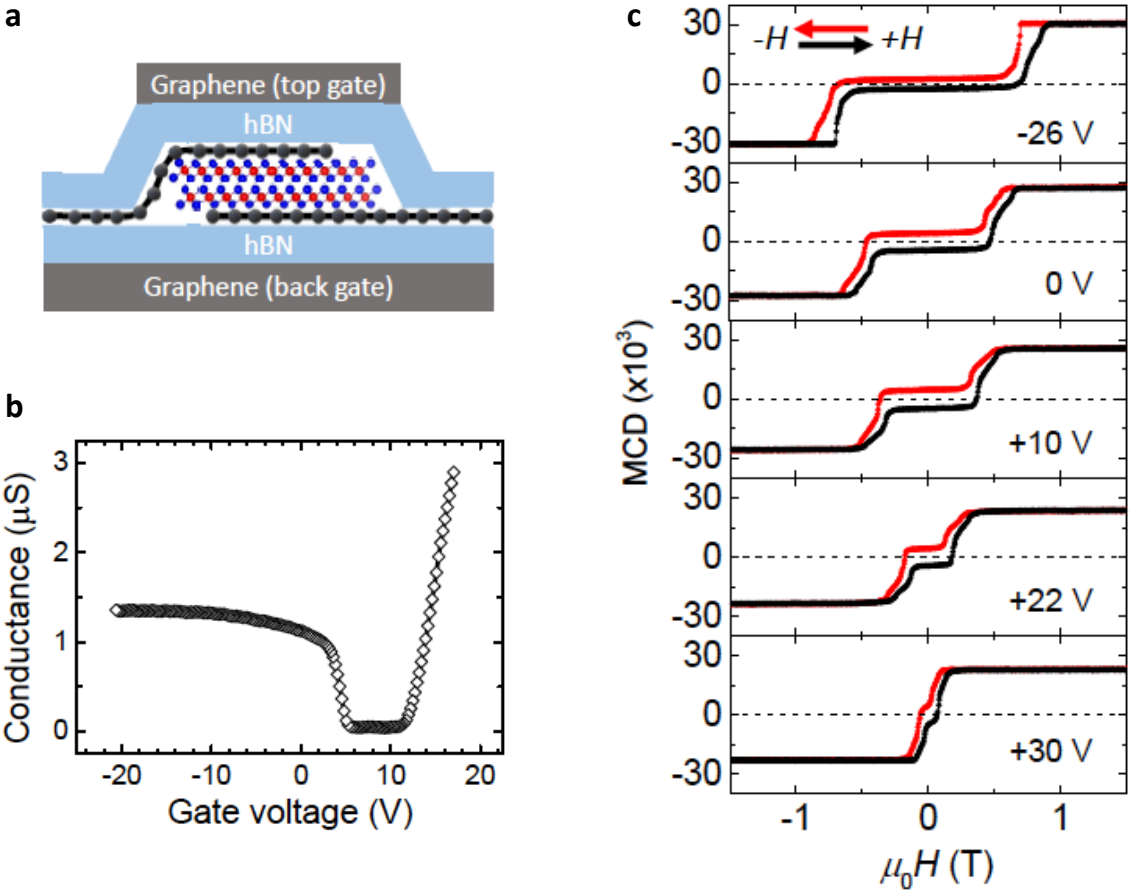

Figure 3. Switching of magnetic orders in bilayer CrI_3 by electrostatic doping: **a**) Schematics of a tunnel field effect transistor of bilayer CrI_3 with top and bottom gates. **b**) Tunnel conductance versus gate voltage demonstrating tunnel field effect transistor action with high on-off ratio. **c**) Sample magnetization versus magnetic field under different gate voltages corresponding to different doping levels. Electrical switching from the antiferromagnetic to the paramagnetic state under zero magnetic field and high electron doping density is seen.

(4) Strongly interaction-enhanced valley magnetic response

Electrons in monolayer transition metal dichalcogenides with a honeycomb lattice structure possess a twofold valley degree of freedom, corresponding to the K and K' points of the Brillouin zone. Because of the strong spin-orbit interaction, the bands at the two valleys are spin split with the valley and spin locked to satisfy the time reversal

symmetry. Similar to the spin, the valley carries a magnetic moment and has been proposed as a new type of information carrier. Magnetic control of this valley degree of freedom is particularly important for the development of valleytronics. We have studied the valley magnetic response of monolayer WSe_2 (a member in the transition metal dichalcogenide family) under varying doping densities so to identify the regime where the valley magnetic response is the most efficient. By fabricating high-quality dual-gated transistors of monolayer WSe_2 and by use of magneto-optical spectroscopy, we have observed a significant enhancement in the valley magnetic response induced by electrostatic doping (figure 4). Such an enhancement is originated from the strong exchange interactions, which favor "alignment" of electrons to a particular valley, and therefore significantly lower the external magnetic field required to valley polarize the material. The physical picture is further supported by the appearance of a nonlinear magnetic field dependence of the valley Zeeman splitting. Furthermore, the valley magnetic response shows non-saturated enhancement as temperature decreases, suggesting the possibility of a valley ferromagnetic ground state and efficient electrical control of the valley magnetic moments. This study was conducted in collaboration with Jie Shan's group at Cornell University. The result has been published in *Physical Review Letters*.

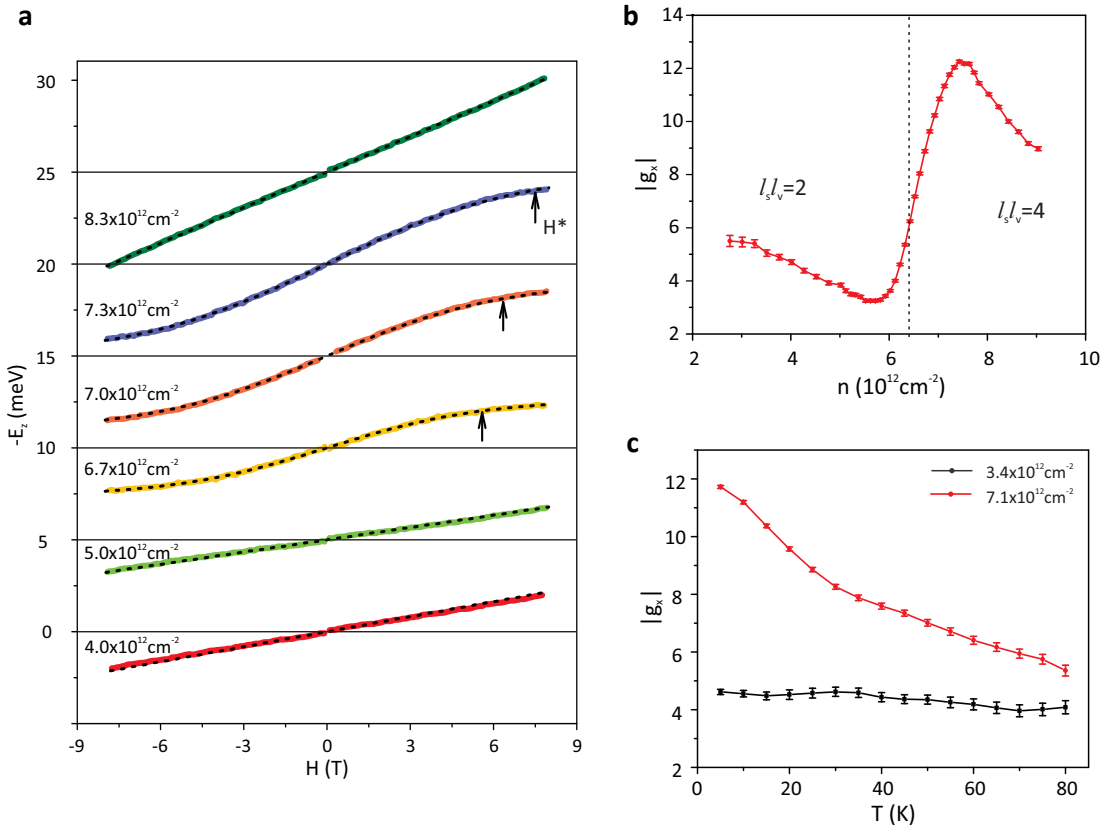

Figure 4. Interaction enhanced valley magnetic response in monolayer WSe_2: **a**) Magnetic field dependence of the valley Zeeman splitting at several electron-doping densities in monolayer WSe_2. **b**) The doping density dependence of the valley g-factor. Significant doping-induced enhancement in the g-factor is seen

because of the strongly enhanced exchange interactions. **c**) The temperature dependence of the valley g-factor at two doping densities.

(5) Helicity-sensitive optical bistability in monolayer WSe_2

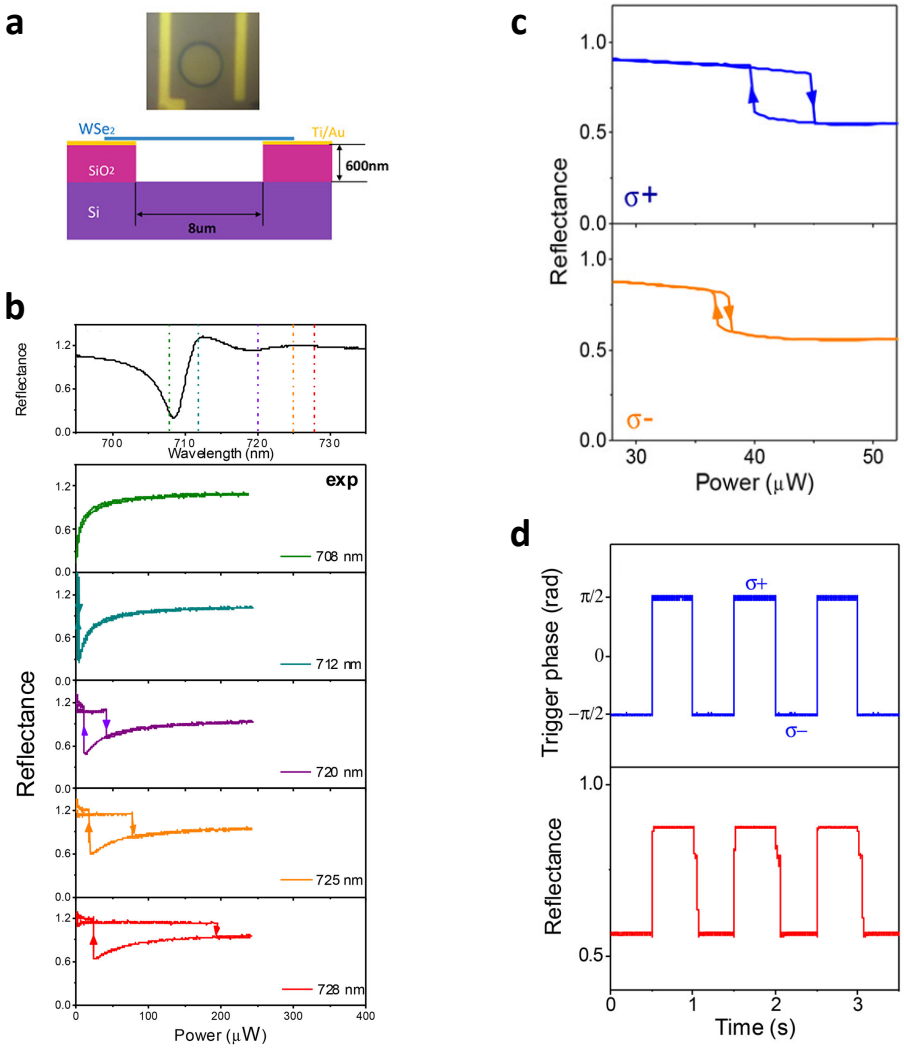

Figure 5. Optical bistability in suspended monolayer WSe_2: **a**) Optical image and schematics of fully suspended monolayer WSe_2 over silicon substrates. **b**) Optical bistability induced by optical pumping at selected pump wavelengths near the exciton resonance of the material. Optical memory has been demonstrated. **c**) Helicity dependent optical bistability induced by optical pumping under a finite magnetic field. Such a dependence on the handedness of the incident light allows switching of the reflectance state of the material by pure modulation of photon spins, as demonstrated in **d**). The top and bottom panels show the time dependence of the incident light handedness and the corresponding sample reflectance, respectively.

The extremely strong light–matter interaction and the presence of a new valley degree of freedom in monolayer transition metal dichalcogenide semiconductors have attracted much interest in both fundamental science and photonic and optoelectronic applications. Monolayer transition metal dichalcogenide semiconductors are direct band

gap materials. Excitons, optically excited bound electron–hole pairs, are tightly bound in these materials due to the weak dielectric screening in two dimensions, which gives rise to extremely strong light–matter interaction near the exciton resonance. Indeed, a single layer of $MoSe_2$ encapsulated by hexagonal boron nitride (hBN) has been shown to reflect over 80% of incident light at the fundamental excitonic resonance. The material has also exhibited optical bistability at a high optical pump intensity level of 10^6 W/cm^2. Moreover, monolayer TMD semiconductors possess two direct energy gaps located at the K and K′ valleys of the Brillouin zone, which exclusively couple to light of opposite helicity. Combining these two properties can, in principle, achieve the valley-selective optical nonlinearity or even bistability in monolayer TMDs. We have developed techniques to prepare fully suspended monolayer WSe_2, in which a back gate voltage can pull down and strain the atomic membrane (figure 5). The suspended structure and the low thermal conductance of WSe_2 allow us to realize optical bistability and optical memory with moderate optical pumping through the photothermal effect. Furthermore, under an external magnetic field, the bistability becomes dependent on the helicity of the incident light, allowing us to control the sample reflectivity purely by the photon spins without relying on intensity modulations. The ability to control light purely by its polarization state will find potential applications in quantum optics and photonics. This study was conducted in collaboration with Jie Shan's group at Cornell University. The result has been published in *Nano Letters*.

(6) Two-dimensional magnetostrictive NEMS

NEMS (nanoelectromechanical systems) are small-scale mechanical devices that find many important applications in nanoscale actuators, accelerometers and sensors. Atomically thin 2D materials, which are both mechanically flexible and strong, and possess functional electronic and optical properties, have attracted much attention for realizing novel NEMS devices. The recent discovery of a large class of 2D magnetic materials, many of which may have strong spin-mechanical coupling, has provided a promising avenue to realize a new class of magnetic NEMS. We have demonstrated magnetically active 2D CrI_3 NEMS devices, whose mechanical resonance frequency is highly sensitive to the magnetic state of the material (figure 6). A significant redshift in the resonance frequency driven by exchange magnetostriction (change in sample strain by spin reorientation) is observed when the material undergoes an antiferromagnetic-paramagnetic transition. A strong inverse magnetostriction is also observed, which allows efficient strain tuning of the spin-flip transition field by simple electrostatic gating. Furthermore, the relative importance of exchange and anisotropy magnetostrictions is quantified. Our results demonstrate a new type of magnetostrictive NEMS with potential applications in spin-phonon transduction as well as magnetic and force sensing. This study was conducted in collaboration with Jie Shan's group at Cornell University. The result is being written up for publication.

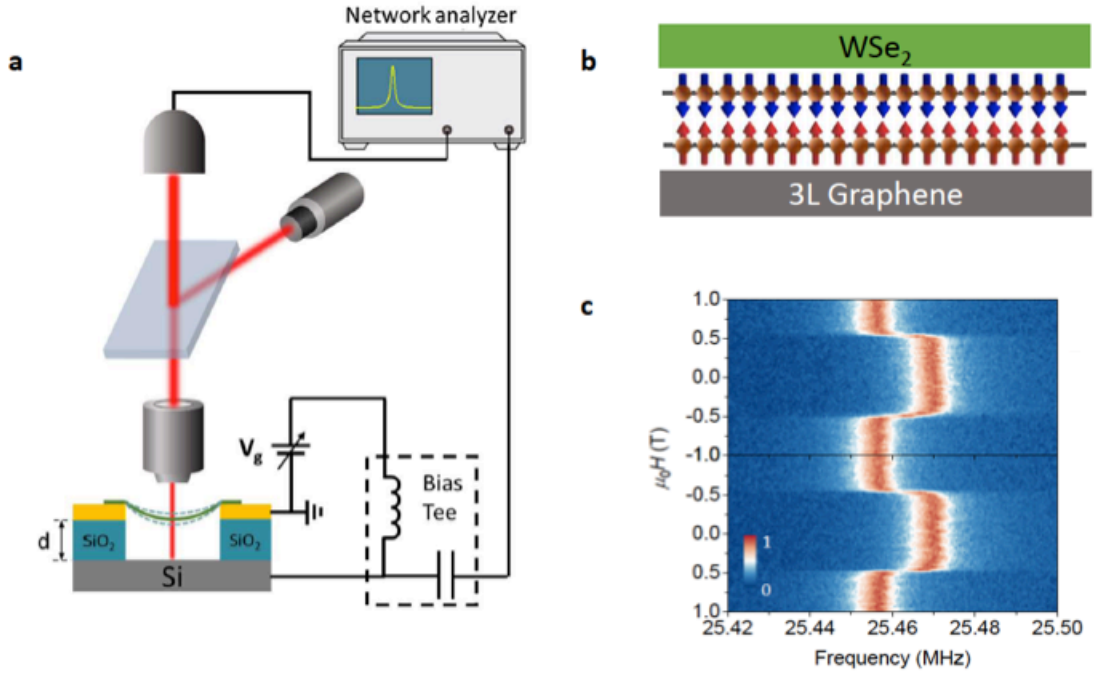

Figure 6. 2D magnetostrictive NEMS: **a**) Schematic illustration of optical detection of the mechanical resonance of suspended bilayer CrI_3. **b**) Side view of the bilayer CrI_3 vertical heterotructure encapsulated by graphene and monolayer WSe_2. **c**) Contour plot of the magnetic field dependence of the mechanical resonance. A clear shift in the resonance is seen when the material undergoes an antiferromagnetic to paramagnetic phase transition.