Electrically Tunable, High Curie Temperature 2D Ferromagnetism in Van der Waals Layered Crystals

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Abstract:
Identifying intrinsic low-dimensional ferromagnets with high transition temperature and electrically tunable magnetism is crucial for the development of miniaturized spintronics and magnetoelectrics. Recently long-range 2D ferromagnetism was observed in van der Waals crystals CrI\(_3\) and Cr\(_2\)Ge\(_2\)Te\(_6\), however their Curie temperature is significantly lowered when reducing down to monolayer/few layers. Herein, using renormalized spin-wave theory and first-principles electronic structure theory, we predict electrically tunable 2D ferromagnetism in van der Waals layered CrSBr and CrSeBr semiconductors with high Curie temperature of \(~150\)K and sizable band gap. High transition temperature is attributed to strong anion-mediated superexchange interaction and a sizable spin-wave excitation gap due to large exchange and single-ion anisotropy. Remarkably, hole and electron doping can switch magnetization easy axis from in-plane to out-of-plane direction, allowing for the realization of spin field effect transistor in monolayer. A microscopic mechanism is provided based on perturbation theory. These unique characteristics establish monolayer CrSBr and CrSeBr as promising platform for realizing 2D spintronics and magnetoelectrics such as spin field effect transistor with ultimate thickness down to 1 nm.
Introduction
Achieving long-range magnetism at low dimensions and high temperature is of both fundamental and technological importance (1-3). In particular, intrinsic low-dimensional semiconducting ferromagnets with high Curie temperature $T_c$, large band gap, and high carrier mobility will help go beyond dilute magnetic semiconductor (1) and pave the way for the development of next-generation ultra-miniaturized, highly integrated spintronics and magneto-optoelectronics (2, 3). However, the coexistence of ferromagnetic and semiconducting characteristics in a single material is generally difficult (3, 4), whereas achieving long-range magnetic ordering is even harder.

According to the Mermin-Wagner theorem (5) 2D ferromagnetic / antiferromagnetic order is strongly prohibited by thermal fluctuations within isotropic Heisenberg model with continuous SU(2) symmetry. Finite magnetic anisotropy such as exchange anisotropy and single-ion anisotropy becomes critical for establishing long-range magnetic order. Long-range 2D FM order has not been observed until the very recent studies in van der Waals crystals CrI$_3$ (6) and Cr$_2$Ge$_2$Te$_6$ (7). However, Curie temperature is markedly lowered with decreasing atomic layers. The corresponding $T_c$ in monolayer and few layers was only $\sim$45K for CrI$_3$ and 25K in Cr$_2$Ge$_2$Te$_6$. High $T_c$ semiconducting ferromagnets are thus highly desirable.

Herein, based on a combined spin-wave theory and first-principles density-functional theory (8, 9), we predict that van der Waals layered 2D semiconductors, chromium sulfur bromide (CrSBr) and chromium selenium bromide (CrSeBr), first synthesized 50 years ago (10, 11), possess electrically tunable magnetic ordering and a high $T_c$ of $\sim$150K. The combination of large spin-wave excitation gap and exchange constants leads to high Curie temperature. Remarkably, the magnetization easy axis can be tuned from in-plane to out-of-plane by electrostatic doping. A microscopic mechanism is provided for the origin of high $T_c$ and electrically controllable magnetism in monolayer CrSBr, which not only offers rational design rules for novel high-temperature ferromagnetic semiconductors, but also allows for realizing spin field effect transistor (spin FET) in monolayer 2D material (12).

Results
Atomistic and electronic structures of monolayer CrXBr
Bulk CrXBr (X=S, Se) are van der Waals layered crystals with Pmmn orthorhombic space group, and their monolayer has Pmmm space group (Figs. 1A-D). The optimized structural parameters listed in Table S1 agree well with experiment. The cleavage energy of CrSBr and CrSeBr is $\sim$0.3 J/m$^2$, which is less than 0.465 J/m$^2$ of graphene with the same van der Waals correlation functional (see Fig. 1E). Their dynamical stability is verified by phonon dispersion displayed in Fig. 1F and Fig. S1 for monolayer CrSBr and CrSeBr, respectively (see calculation details in Supporting Information). These results suggest that monolayer CrSBr and CrSeBr can be exfoliated from their bulk counterpart, which indeed agrees with the experimental observation of soft, pliable, and cleavable nature in bulk CrSBr (11).

Monolayer CrSBr and CrSeBr exhibit highly anisotropic electronic structure with a semiconducting band gap of 1.66 eV and 0.78 eV, respectively. Figure 2A shows the band structure for monolayer CrSBr using hybrid HSE06 exchange-correlation functional (see additional results in Figs. S2 and S3). The stronger dispersion along $b$ axis indicates large anisotropy. This is supported by small effective hole mass of only $\sim$0.11$m_0$ and the associated large hole mobility of
\( \sim 720 \text{ cm}^2\text{V}^{-1}\text{s}^{-1} \) calculated by using a phonon-limited scattering approach (see Table S2) \((13, 14)\). Moreover, bulk CrSBr is stable in air with black color \((1I)\), which is consistent with the 1.66 eV gap in its monolayer as interlayer van der Waals interaction will bring it down. Therefore, monolayer CrSBr and CrSeBr are promising alternatives to dilute magnetic semiconductors and suitable for 2D spintronic applications.

**Magnetic properties of monolayer CrXBr**

The local geometrical environment of Cr\(^{3+}\) can be viewed as a distorted octahedron with each Cr\(^{3+}\) surrounded by four S\(^2\text{-}^2\) (Se\(^2\text{-}^2\)) and two Br\(^-\), as illustrated in Fig. 2B. In an ideal octahedron, the crystal field splitting breaks five-fold degenerate \(d\) orbitals into two groups: double-degenerate \(e_g\) and three-fold degenerate \(t_{2g}\) orbitals. The presence of two disparate types of anions further reduces it to \(C_2v\) and lifts the degenerate \(e_g\) and \(t_{2g}\) orbitals. Because spin pairing energy \(U_p\) for transition from parallel spins on two orbitals to antiparallel spins on a single orbital is greater than crystal field splitting energy \(\Delta_c\), parallel high spin state and hence ferromagnetism should be favored.

To verify the ferromagnetic ground state, we investigate the relative stability of ferromagnetic and antiferromagnetic configurations using a \(2\times2\times1\) supercell (Fig. S4). Ferromagnetic coupling is found to be more stable than antiferromagnetic coupling. Spin density in Fig. 2C-D reveals that ferromagnetism mainly comes from Cr with \(\sim 3\mu_B/\text{Cr}\), consistent with the high spin state of Cr\(^{3+}\) with three unpaired electrons. In contrast, S atoms carry small opposite spin moment and Br atoms are hardly magnetized. The ferromagnetic coupling between Cr atoms originates from superexchange interaction mediated by S (Se) and Br. To illustrate this, we classify the linking geometry between Cr\(^{3+}\) ions into two types. In the first type, the neighboring octahedra have two common edge atoms S(Se) and Br where the Cr-S(Se)-Cr and Cr-Br-Cr angle is \(~90^\circ\), e.g. \(\alpha\) along \(ab\) diagonal, and \(\beta\) and \(\gamma\) along \(a\) axis (Figs. 1B-C). In the second type, the neighboring octahedra along the \(b\) axis have one common corner S (Se) with a Cr-S(Se)-Cr angle \(\delta\) of \(~160^\circ\) (Fig. 1D). According to Goodenough-Kanamori-Anderson rules \((15, 16)\), ferromagnetic coupling is favored for \(90^\circ\) superexchange interaction between two magnetic ions with partially filled \(d\) shells, while antiferromagnetic coupling is preferred for \(180^\circ\) superexchange interaction. As \(\delta\) significantly deviates from \(90^\circ\) and \(180^\circ\), the superexchange interaction along \(b\) exhibits competing ferromagnetic and antiferromagnetic coupling. In contrast, ferromagnetic coupling along \(a\) axis and \(ab\) diagonal is strongly favored due to \(~90^\circ\) \(\alpha\), \(\beta\) and \(\gamma\), establishing ferromagnetic ground state.

Magnetic anisotropy is crucial for establishing long-range 2D ferromagnetic order. Single-ion anisotropy and exchange anisotropy are two important sources. Single-ion anisotropy, also known as magnetocrystalline anisotropy energy (MAE), determines easy/hard magnetization axis. Here we estimate the MAE by calculating total energy as function of magnetization direction with spin-orbit coupling (SOC) taken into account. Figure 2E shows the angular dependent MAE in monolayer CrSBr (see Fig. S5 for CrSeBr). It demonstrates high magnetic anisotropy with easy axis along \(a\) (i.e. \(x\) direction) in the 2D plane, distinct from Cr\(_3\) \((6)\) and Cr\(_2\)Ge\(_2\)Te\(_6\) \((7)\) with out-of-plane easy axis. The MAE as listed in Table S1 reveals the easy and hard axis along the \(a\) and \(b\) axis, respectively.

**Heisenberg model and spin-wave theory of Curie temperature in monolayer CrXBr**


Having known the easy axis along $a$ axis (or, $x$ direction), we build the corresponding spin Hamiltonian using Heisenberg XZZ model (17) including both single-ion anisotropy and Heisenberg exchange anisotropy,

$$H = -\sum_{\langle ij \rangle} J_1 \vec{S}_i \cdot \vec{S}_j - \sum_{\langle \langle ij \rangle \rangle} J_2 \vec{S}_i \cdot \vec{S}_j - \sum_{\langle \langle \langle ij \rangle \rangle \rangle} J_3 \vec{S}_i \cdot \vec{S}_j - \sum_i D (S_i^x)^2 - \sum_{\langle ij \rangle} \lambda_1 S_i^z S_j^z - \sum_{\langle \langle ij \rangle \rangle} \lambda_2 S_i^z S_j^z - \sum_{\langle \langle \langle ij \rangle \rangle \rangle} \lambda_3 S_i^z S_j^z. \quad (1)$$

We calculate the exchange coupling constants $J_{1,2,3}$, single-ion anisotropy constant $D$, and exchange anisotropy constant $\lambda_{1,2,3}$ by mapping magnetic configurations (Fig. S4) to the Hamiltonian in Eq. 1. The results are listed in Table S1. Under linear spin-wave approximation, we obtain a second quantization representation using Holstein-Primakoff transformation (18),

$$H_{\text{spin-wave}} = \sum_i \varepsilon_0 b_i^+ b_i - J_1 S \sum_{\langle ij \rangle} b_i^+ b_j - J_2 S \sum_{\langle \langle ij \rangle \rangle} b_i^+ b_j - J_3 S \sum_{\langle \langle \langle ij \rangle \rangle \rangle} b_i^+ b_j. \quad (2)$$

where $\varepsilon_0 = 2DS + 2S(J_1 + 2J_2 + J_3 + \lambda_1 + 2\lambda_2 + \lambda_3)$. The resulted spin-wave excitation gap at $\Gamma$ point from Eq. 2 is given by $\Delta_0 = 2S(D + \lambda_1 + 2\lambda_2 + \lambda_3)$. It yields $\Delta_0$ of 0.07 meV for CrSBr and 0.31 meV for CrSeBr. Their 2D magnon dispersion is shown in Fig. 3A and Fig. S6A. We then estimate the Curie temperature $T_c$ based on renormalized spin-wave theory (RSWT),

$$M(T) = S - \frac{1}{n N_k} \sum_k \left[ \exp \left( \frac{M(T)E(k_x,k_y)}{S k_B T} \right) - 1 \right]^{-1} \quad (3)$$

where $n$ and $N_k$ refer to number of spins per unit cell and number of $k$ points sampled in the first Brillouin zone. Curie temperature $T_c$ is subsequently determined by requiring $M(T)|_{T=T_c} = \frac{M_0}{2}$ using Eq. 3, as shown in Fig. 3B and Fig. S6B-C.

The calculated $T_c$ from RSWT are 150K and 152K for monolayer CrSBr and CrSeBr, respectively, significantly higher than that of 2D CrI$_3$ (6) (45K) and Cr$_2$Ge$_2$Te$_6$ (7) (20K) discovered in recent experiments. Furthermore, as shown in Fig. 3C, Fig. S6D and Table S3, $T_c$ from RSWT is close to that obtained from Monte Carlo simulation with Heisenberg model (172K for CrSBr and 145K for CrSeBr), but, as expected, lower than the estimates from linear spin-wave theory (518K for both) and simple Ising model (590K and 520K). High $T_c$ 2D ferromagnetism (~150K) beyond liquid nitrogen boiling point of 77K opens up many exciting opportunities, among which electric control of 2D ferromagnetic ordering is an important one we will address next.

**Electric control of 2D ferromagnetic ordering in monolayer CrXBr for 2D spin-FET**

Externally controlled magnetism is highly desirable for magnetoelectrics. Recent works have shown that carrier doping can be introduced into monolayers by electric gating to control magnetism (19, 20). In addition, van der Waals gap in 2D magnetic layers induces giant tunneling magnetoresistance (21, 22). Here we show that carrier doping can drastically change MAE in 2D CrSBr and switch the magnetization easy axis. Figure 4A shows total energy as function of carrier concentration $n$ with different magnetization directions. Under electron doping ($n<0$), the energy difference between out-of-plane and in-plane magnetization decreases slowly with increasing electron doping. The out-of-plane magnetization becomes favored beyond a critical electron doping of $-2 \times 10^{13}$/cm$^2$. In contrast, hole doping ($n>0$) has a much bigger influence, and a hole doping above $4 \times 10^{12}$/cm$^2$ will stabilize the out-of-plane FM ordering. Experimentally, it is
possible to achieve carrier concentration of up to $10^{13}$-$10^{14}$/cm² in 2D materials, therefore carrier doping is a feasible strategy to control magnetization ordering in monolayer CrSBr.

Doping induced tunability of MAE can be understood from a perturbation theory analysis (23). Given a pair of valence ($v$) eigenstate $\psi_v$ and conduction ($c$) eigenstate $\psi_c$, their contribution to MAE is given by

$$\Delta E_{vc} = \frac{1}{\Delta_{vc}} (|H_{vc}^{soc}(\vec{x})|^2 - |H_{vc}^{soc}(\vec{y})|^2)$$

where $H_{vc}^{soc}(\vec{n}) = \langle \psi_v | H^{soc}(\vec{n}) | \psi_c \rangle$ is the SOC matrix element and $\Delta_{vc} = \varepsilon_v - \varepsilon_c$. $H^{soc}(\vec{n}) = \xi \vec{\sigma} \cdot \vec{L}$, where $\vec{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$ are the 2×2 Pauli matrices, $\vec{L}$ is orbital angular momentum operator, and $\xi$ is the SOC strength. The spin and magnetic quantum number of orbital characters determine the sign of $\Delta E_{vc}$ (Figure 4B), while the sum of $\Delta E_{vc}$ in Eq. 4 over all valence-conduction pairs determines the MAE and the easy axis. Cr contributes to both in-plane and out-of-plane magnetization, while the contribution of S and Br to MAE favors the in-plane easy axis. Due to higher atomic number of Br, its SOC strength is much larger (about 3 times of Cr and 30 times of S), hence Br has stronger influence on MAE than Cr and S. Consequently, in-plane easy axis is preferred in pristine 2D CrSBr. Upon electron doping, Cr-$d_{x^2-y^2}$ favors in-plane magnetization. Increasing occupation in Cr-$d_{x^2-y^2}$ from electron doping will therefore reduce the stability of in-plane magnetization. Upon hole doping S-$p_z$ and Br-$p_z$ in the highest valence band become unoccupied, thereby reducing the stability of in-plane magnetization since $\Delta E_{vc}$ between valence Br-$p_z$ and conduction Br-$p_z$ favors in-plane magnetization. Due to stronger SOC in Br, hole doping has larger impact on MAE than electron doping, reflected in the stiffer slope upon hole doping (see Fig. 4A). Hence, the relative stability of in-plane/out-of-plane magnetization can be tuned by carrier doping at a critical concentration of $4 \times 10^{12}$/cm² (hole) and $-2 \times 10^{13}$/cm² (electron).

The doping-modulated easy axis allows for realizing 2D spin FET (12). A schematic of such magnetoelectric device is proposed in Fig. 4C, where monolayer CrSBr is double-gated by top and bottom electrodes for carrier doping with two dielectric layers (e.g. hexagon BN) to prevent direct tunneling. Carrier concentration and easy axis are controlled by the double gate, while the sourcedrain voltage drives spin-dependent transport. The easy axis switches between in-plane and out-of-plane direction upon critical doping, leading to the emergence of in-plane FM/out-of-plane FM interface between the doped and undoped region. This hetero-magnetic interface gives a high resistance state due to strong interface scattering. In contrast, homo-magnetization below critical doping maintains a low resistance state, realizing 2D spin FET.

**Discussion**

In summary, we reported here a class 2D intrinsic ferromagnetic semiconductors CrSBr and CrSeBr. Both possess ferromagnetic ground state with high $T_c$ of 150 K and sizable band gap of 1.66 eV and 0.78 eV, respectively. Carrier mobility in CrSBr is hole-dominating and highly anisotropic in the 2D plane, up to 720 cm²/V•S. Remarkably, magnetization easy axis can be tuned from in-plane to out-of-plane by electrostatic doping. The microscopic mechanisms of both high $T_c$ and electric tunability provide rational principles for designing novel 2D ferromagnetic
semiconductors. These characteristics, together with the fact that bulk CrSBr was synthesized three decades ago, suggest that monolayer CrSBr and CrSeBr ferromagnets not only give long-desired promising alternatives to dilute magnetic semiconductors, but also offer unprecedented opportunities for 2D spintronic and magneto-optoelectronic applications such as spin FET.

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Acknowledgments:

Funding: H.W. and X.Q. acknowledge the support by NSF under award number DMR-1753054. Portions of this research were conducted with the advanced computing resources provided by Texas A&M High Performance Research Computing. J.Q. acknowledges the financial support from the National Natural Science Foundation of China (Projects No. 11674132) and PAPD.

Author contributions: X.Q. and J.Q. conceived the idea and supervised the project. J.Q., H.W. and X.Q. performed the calculations and analyzed the results. All authors participated in discussions and manuscript writing. J.Q. and H.W. contribute equally to this work.

Competing interests: The authors declare that they have no competing interests.
Fig. 1. Monolayer CrXBr and their structural properties. (A) Overview of van der Waals layered crystals CrXBr (X=S, Se) in their monolayer form. Bulk CrXBr has orthorhombic symmetry group $Pmmm$. (B-D) Top and side views of monolayer CrXBr. $J_1$, $J_2$, and $J_3$ refer to isotropic Heisenberg exchange coupling constants between the first, second, and third nearest-neighbor spins, respectively. (E) Cleavage energy of CrSBr and CrSeBr with graphene included for comparison. The low cleavage energy suggests that they can be easily exfoliated from their van der Waals layered bulk. (F) Phonon dispersion of monolayer CrSBr. The absence of imaginary modes demonstrates that CrXBr monolayers are dynamically stable against thermal fluctuations.
Fig. 2. Electronic structure and ferromagnetism in monolayer CrSBr. (A) Atom-projected electronic band structure of CrSBr. Red (blue) indicate spin up (down). (B) Crystal field splitting from an ideal octahedron with $O_h$ symmetry to a distorted octahedron with $C_{2v}$ symmetry. The distorted octahedron is composed of one Cr, four S and two Br atoms. (C-D) Spin density in monolayer CrSBr, indicating that ferromagnetism is mainly localized around Cr atom. For illustration, the isovalue for spin down (blue) is chosen to be half of spin up (green). (E) Angle-dependent magnetocrystalline anisotropy energy (MAE).
Fig. 3. Ferromagnetism in monolayer CrSBr. (A) Magnon dispersion in monolayer CrSBr with spin-wave excitation gap located at the Γ point of the first Brillouin zone. Δ₀ refers to spin-wave excitation gap. (B) Normalized magnetic moment as function of temperature and spin-wave excitation gap in monolayer CrSBr. The red line indicates the Curie temperature $T_c$ as function of spin-wave excitation gap, where the white dot indicates the corresponding $T_c$ in monolayer CrSBr. (C) Temperature dependent normalized magnetization in monolayer CrSBr using different theoretical models, including Ising model and fitting, Heisenberg model and fitting, linearized spin-wave theory (LSWT), and renormalized spin-wave theory (RSWT).
Fig. 4. Electrical control of 2D ferromagnetism in monolayer CrSBr. (A) Energy of FM configurations with different magnetization direction as function of carrier concentration $n$. Under moderate electron doping ($n>0$), the in-plane magnetization remains favored over the out-of-plane magnetization, although the energy difference decreases slowly with increasing electron doping concentration. In contrast, hole doping ($n<0$) has a much larger impact on the relative stability of different magnetization directions. (B) Contribution of each valence and conduction state pair to the in-plane/out-of-plane magnetization determined by the spin and magnetic quantum number of orbital characters in the valence and conduction states. (C) A schematic of 2D magnetoelectric device that can realize giant magnetoresistance effect that is controlled by electrostatic doping rather than by external magnetic field. 2D ferromagnetic CrSBr/CrSeBr is doubly gated, and two dielectric layers such as hexagon BN to avoid direct tunneling. The gate voltage controls carrier concentration and subsequently changes the corresponding easy axis direction upon hole doping, while the longitudinal in-plane source-drain voltage drives spin-dependent quantum transport. Upon critical hole doping, the easy axis will switch from in-plane to out-of-plane direction. As a result, an in-plane FM/out-of-plane FM interface will emerge between the hole-doped region and undoped region. Strong scattering is expected to take place at this hetero-magnetic interface due to different magnetization direction, resulting in high resistance state, while homo-magnetization below critical doping corresponds to low-resistance state, thereby realizing electrically-controlled GMR effect by dynamic electrostatic doping.