

Exchange magnetostriction in two-dimensional antiferromagnets

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Magnetostriction, coupling between the mechanical and magnetic degrees of freedom, finds a variety of applications in magnetic actuation, transduction and sensing^{1,2}. The discovery of two-dimensional (2D) layered magnetic materials^{3,4,5,6,7,8} presents a new platform to explore the magnetostriction effects in ultrathin solids. Here we demonstrate an exchange-driven magnetostriction effect in mechanical resonators made of 2D antiferromagnetic CrI₃. The mechanical resonance frequency is found to depend on the magnetic state of the material. We quantify the relative importance of the exchange and anisotropy magnetostriction by measuring the resonance frequency under a magnetic field parallel and perpendicular to the easy axis, respectively. Furthermore, we show efficient strain-tuning of the internal magnetic interactions in 2D CrI₃ as a result of inverse magnetostriction. Our results establish the basis for mechanical detection and control of magnetic states and magnetic phase transitions in 2D layered materials.

Two-dimensional (2D) layered magnetic materials are attractive systems for exploring magnetostrictive phenomena and applications: while they share high mechanical stiffness and strength and low mass with other 2D materials, they are magnetically active^{1,2,3,6,7,8}. Among the large class of newly emerged 2D layered magnetic materials, of particular interest is few-layer CrI₃, whose magnetic ground state consists of antiferromagnetically coupled ferromagnetic (FM) monolayers with out-of-plane easy axis^{3,5}. The interlayer exchange interaction is relatively weak. A magnetic field on the order of 0.5 T in the out-of-plane ($\hat{z}$) direction can induce a spin-flip transition in bilayer CrI₃. Remarkable phenomena and device concepts based on detecting and controlling the interlayer magnetic state have been recently demonstrated, including spin-filter giant magnetoresistance^{9,10,11,12}, magnetic switching by electric field¹³ or electrostatic doping^{14,15,16,17}, and spin transistors¹⁸. The coupling between the magnetic and mechanical degrees of freedom in the material, however, remains elusive.

Here we demonstrate exchange magnetostriction and its inverse effect in mechanical resonators made of 2D CrI₃ drumhead membranes (Fig. 1a). We focus on even-layer CrI₃ for maximum effect (see Supplementary Fig. S2 for an odd-layer case). We encapsulate

CrI₃ (an air sensitive insulator) within two stable 2D materials, few-layer graphene (below) and monolayer WSe₂ (above) (Fig. 1b). In addition to protect CrI₃ from degradation under ambient conditions, few-layer graphene acts as a conducting electrode, while monolayer WSe₂ as a strain gauge as we discuss below. The 2D heterostructure is first assembled and then transferred on prefabricated circular microtrenches (2 – 3 μm in radius) with patterned Au electrodes and Si back gate. The optical image of a sample device is shown in Fig. 1c. We drive mechanical resonances in the membrane electrically in the linear regime using a small r.f. gate voltage (~ 1 mV). It is superposed on a DC gate voltage V_g that applies static tension to the membrane by pulling it towards the gate. We detect the mechanical resonances interferometrically at 633 nm using a network analyzer. We also measure the out-of-plane magnetization of the membrane using magnetic circular dichroism (MCD) at 633 nm. Unless otherwise specified, all measurements were performed at 4 K in a helium exchange gas environment (<10⁻⁵ Torr). (See Methods for details on device fabrication and measurement techniques.)

Figure 1d shows the fundamental resonance mode of a bilayer CrI₃ membrane at $V_g = 0$ (the imperfect symmetry in the response to $V_g > 0$ and $V_g < 0$ gives rise to a net force at the r.f. driving frequency). It has a Lorentzian lineshape (solid line) with a peak frequency f around 25.5 MHz and a width around 10 kHz (corresponding to a quality factor of about 2500). Figure 1e shows the gate dependence of the resonance frequency. It is well described by the continuum model for a tensioned membrane with negligible bending stiffness and fully clamped boundary: $f = \frac{\xi}{2\pi R} \sqrt{\frac{\sigma}{\rho}}$ (red solid line). Here $\xi \approx 2.405$ is the first root of the zeroth order Bessel function. The resonance frequency scales inversely with the drumhead radius R , and is determined by stress on the membrane σ and its 2D mass density ρ . Near $V_g = 0$, the resonance frequency is set by the built-in stress σ_0 , whereas at higher V_g the gate-induced stress $3Y_{eff}\epsilon(V_g)$ causes the frequency to increase roughly as V_g^4 . Here Y_{eff} is the effective 2D Young's modulus of the membrane, and $\epsilon(V_g)$ is the gate-induced strain. This behavior is fully consistent with other 2D mechanical resonators, including graphene and MoS₂ [Ref. ^{19,20,21}]. We calibrate $\epsilon(V_g)$ from the exciton peak shift of the WSe₂ layer (63 meV per percent of biaxial strain ²²), and determine the parameters of the resonator $\sigma_0 \approx 0.5$ N/m, $Y_{eff} \approx 600$ N/m and $\rho \approx 3 \times 10^{-5}$ kg/m² for the example shown in Fig. 1 [Ref. ^{19,20,21}] (see Methods).

We investigate the effect of magnetic field on CrI₃ resonators at $V_g = 0$. The mechanical resonance of the bilayer CrI₃ device is measured while the field is swept from 1 T to -1 T back to 1 T along the easy axis $\hat{z}$ (Fig. 2a). The resonance frequency f is independent of field except an abrupt redshift of ~ 0.06% when the field magnitude exceeds ~ 0.5 T. The linewidth and amplitude basically do not change (Supplementary Fig. S1). We correlate the field dependence of f (Fig. 2b) with MCD (Fig. 2c). MCD of the membrane is consistent with the reported results for bilayer CrI₃ [Ref. ¹⁴]. The material is antiferromagnetic (AF) under small fields (the finite MCD signal is caused by layer

asymmetry in the heterostructure) and undergoes a first-order AF-FM spin-flip transition (with hysteresis) around ± 0.5 T. The mechanical resonance frequency is thus correlated with the sample's magnetic state with $f_{AF} > f_{FM}$, where f_{AF} and f_{FM} denote the resonance frequency in the AF and FM state, respectively. In contrast to MCD, f is insensitive to the magnetization direction.

Similar behavior is observed in all CrI₃ resonators (6 in total) that were investigated in this study. Figure 2d - 2f shows the corresponding result for a 6-layer CrI₃ resonator under an out-of-plane field up to 2.3 T (see Supplementary Fig. S2 for the other devices). Compared to bilayer CrI₃, there are now two spin-flip transitions around 0.9 T and 1.8 T, which correspond to spin flip in the surface layer and the interior layers, respectively^{9,18}. The resonance frequency redshifts at each spin-flip transition. The total redshift from f_{AF} (~ 0 T) to f_{FM} (> 1.8 T) is about 0.23%. This is nearly 5 times larger than in bilayer CrI₃.

The behavior of the resonators under an in-plane magnetic field is different. Figure 3a shows the mechanical resonance of the same 6-layer CrI₃ device as in Fig. 2d - 2f while the magnetic field is swept from 8 T to -8 T to 8 T along the in-plane direction. The field dependence of f is shown in Fig. 3b after subtracting a small linear drift due to stress relaxation and/or slow temperature drift. Sharp transition and hysteresis that are characteristic of the first-order spin-flip transition are absent. Instead, f redshifts smoothly with increasing field and saturates beyond ~ 6 T. The total change in f ($\sim 0.13\%$) is about half of the value observed under the out-of-plane field. Consistent behavior is observed in a second 6-layer device (Supplementary Fig. S3). The smooth redshift is correlated with the spin-canting effect observed in 2D CrI₃ [Ref. ^{9,18}]. Because of the large out-of-plane magnetic anisotropy, the spins under an in-plane field are canted continuously from $\hat{z}$ until reaching the saturation field, beyond which they are fully aligned with the field. The observed saturation field (~ 6 T) agrees with the value reported for multilayer CrI₃²³. It is higher than in bilayer CrI₃, in consistence with the higher magnetic transition temperatures in thicker samples.

The resonance frequency of a mechanical resonator at $V_g = 0$ is determined by the built-in stress σ_0 . The observed correlation between the resonance frequency and the magnetic state of 2D CrI₃ suggests that magnetostriction is a result of competition between minimizing the elastic energy and the internal magnetic interactions. Other effects such as the one arisen from the magnetostatic energy cannot explain the experimental observations (see Methods). The elastic energy of a membrane per unit area can be expressed as $U_{el} = \frac{3}{2}Y_{eff}\epsilon^2$ [Ref. ^{19,20,21}]. In our devices Y_{eff} is dominated by that of few-layer graphene and is independent of magnetic field (Methods); strain $\epsilon = (a - a_0)/a_0$ is defined as a fractional change of the in-plane lattice constant a from its equilibrium value a_0 . For fully clamped membranes with no vertical displacement, the boundary/substrate does work $U_b = -\sigma_0\epsilon = -2U_{el}$ to keep the area or in-plane lattice constant a of the membrane fixed. Here $\sigma_0 = 3Y_{eff}\epsilon$ is the stress. If we neglect the strain

dependence of the demagnetization energy and leave out the intralayer exchange interactions (which do not play a role in the interlayer metamagnetic transition), the part of free energy that is strain dependent can be expressed to the lowest relevant order as

$$F = -J_{\perp}(\hat{\mathbf{S}}_t \cdot \hat{\mathbf{S}}_b) + K_{eff}(|\hat{\mathbf{S}}_t \times \hat{\mathbf{z}}|^2 + |\hat{\mathbf{S}}_b \times \hat{\mathbf{z}}|^2), \quad (1)$$

for bilayer CrI₃. Here $\hat{\mathbf{S}}_t$ and $\hat{\mathbf{S}}_b$ denote the spin unit vector of the top and bottom CrI₃ layers, respectively. The two terms describe, respectively, the interlayer exchange coupling with energy per unit area $J_{\perp}$ (> 0 for FM and < 0 for AF ordering) and anisotropy with effective energy per unit area per layer K_{eff} (> 0). For simplicity we only consider the effect of bond length change on $J_{\perp}$ and K_{eff} . The equilibrium lattice constant of the strained membrane in each magnetic state can be found by minimizing the total energy ($U_{tot} = -U_{el} + F$) with respect to a_0 , or equivalently strain ϵ , from which stress and mechanical resonance frequency can be evaluated²⁴ (see Methods).

For spin-flip transitions, the anisotropy energy is not relevant because spins are along the easy axis^{24,25,26}. Minimizing U_{tot} for bilayer CrI₃ yields a change in strain ($\frac{2}{3Y_{eff}} \frac{\partial J_{\perp}}{\partial \epsilon}$) and a fractional change in the resonance frequency ($\frac{1}{3\sigma_0} \frac{\partial J_{\perp}}{\partial \epsilon}$) for the AF-FM transition. We estimate $\frac{\partial J_{\perp}}{\partial \epsilon} A_u \approx +2$ meV from experiment, where A_u (≈ 0.41 nm²) is the unit cell area. The positive sign indicates that strain weakens AF ordering. We also estimate the saturation magnetostriction from the AF-FM transition induced strain change to be $\sim 10^{-6}$. It corresponds to intrinsic saturation magnetostriction of $\sim 10^{-5}$ in bilayer CrI₃ because the Young's modulus of CrI₃ is about an order of magnitude smaller than Y_{eff} of the heterostructure. The intrinsic value is comparable to that of elemental ferromagnetic metals²⁷. The physical interpretation above is further supported by the nearly 5-time stronger magnetostrictive response in 6-layer CrI₃ than in bilayer CrI₃. The interlayer exchange energy is 5 times larger in 6-layer CrI₃ since now there are 5 interfaces.

Under an in-plane field, both interlayer exchange and anisotropy contribute to the free energy. A similar energy minimization scheme can be applied to yield $\frac{1}{3\sigma_0} \left(5 \frac{\partial J_{\perp}}{\partial \epsilon} - 6 \frac{\partial K_{eff}}{\partial \epsilon} \right) \left(\frac{H_{\parallel}}{H_S} \right)^2$ (for $H_{\parallel} \leq H_S$) for the fractional resonance frequency shift in 6-layer CrI₃. Here $H_{\parallel}$ and H_S are the applied and saturation field, respectively. The quadratic dependence of the resonance frequency shift on $H_{\parallel}$ is in good agreement with experiment (dashed line, Fig. 3b). The discrepancy at small fields likely reflects a non-perfect alignment of the field with the sample plane. The observed $\sim 50\%$ smaller frequency shift under saturation in Fig. 3b compared to Fig. 2e suggests that $\frac{\partial K_{eff}}{\partial \epsilon} \approx \frac{1}{2} \frac{\partial J_{\perp}}{\partial \epsilon}$ in the 6-layer CrI₃ device. The stronger dependence on in-plane strain of the interlayer exchange than

the intralayer anisotropy is somewhat counterintuitive. Future studies are required to better understand this observation.

Finally, we use the mechanical device platform to demonstrate strain-tuning of the magnetic interactions through the inverse magnetostriction effect. We probe the spin-flip transition in the same bilayer CrI_3 device as in Fig. 2a-c at different V_g 's by MCD (see Supplementary Fig. S4 for the study on a 6-layer device). The application of V_g can induce tension on the suspended membrane by an electrostatic force. It can also induce electrostatic doping into CrI_3 through the graphene layer because of its large quantum capacitance from the flat 3d bands^{14,15}. Figure 4a and 4b compare the behavior of a suspended and substrate-supported region of the membrane. Only the MCD data near the spin-flip transition for the positive field sweeping direction is shown for clarity. For the substrate-supported region, only the electrostatic doping effect is relevant. The spin-flip transition field H_C varies linearly with V_g (red symbols, Fig. 4c), which is in good agreement with previous studies^{14,15}. In contrast, the behavior of the suspended region is almost symmetric about $V_g = 0$; H_C decreases nonlinearly with V_g (blue symbols, Fig. 4c). We conclude from the comparison that electrostatic doping into suspended CrI_3 is not significant because of the low gate dielectric constant (vacuum) and the change in H_C is caused primarily by strain.

Figure 4d shows H_C as a function of gate-induced strain calibrated by the WSe_2 gauge. It decreases linearly with strain by as much as 32 mT. Quantitatively, H_C can be related to the interlayer exchange coupling $J_\perp$ through $\mu_0 H_C = \frac{-J_\perp + \mu_0 M_0^2/t}{M_0}$ [Ref. 14], where μ_0 , M_0 and t denote the vacuum permeability, the saturation magnetization per CrI_3 monolayer, and the interlayer distance in bilayer CrI_3 , respectively (see Methods). We obtain $\frac{\partial J_\perp}{\partial \epsilon} A_u \approx +4$ meV from the slope in Fig. 4d and M_0 (assuming that each Cr^{3+} cation carries a magnetic moment of 3 Bohr magnetons). The value agrees well with that estimated from the mechanical resonance measurement. The discrepancy is largely due to uncertainty in the resonator parameters (such as initial stress and mass). Nevertheless, the good agreement between two independent measurements illustrates the importance of interlayer exchange magnetostriction in 2D CrI_3 .

In conclusion, we have demonstrated an exchange magnetostriction effect in 2D antiferromagnetic CrI_3 . An inverse magnetostriction effect has also been demonstrated, which allows continuous strain-tuning of the internal magnetic interactions. Our results have put in place the groundwork for potential applications of these devices, including magnetic actuation and sensing, and for mechanical detection of emerging magnetic states and phase transitions in 2D layered magnetic materials²⁸.

Methods

Device fabrication

We used the layer-by-layer dry transfer method to fabricate drumhead resonators made of atomically thin CrI₃ fully encapsulated by few-layer graphene and single-layer WSe₂ as shown in Fig. 1b. Atomically thin flakes of CrI₃, graphene and WSe₂ were mechanically exfoliated onto silicon substrates with a 285-nm oxide layer from the corresponding bulk synthetic crystals (from HQ Graphene). A polymer stamp made of a thin layer of polycarbonate (PC) on polydimethylsiloxane (PDMS) was used to pick up the desired flakes to form the heterostructure. The complete heterostructure was first released onto a new PDMS substrate so that the residual PC film on the sample can be removed by dissolving it in N-Methyl-2-pyrrolidone. The sample was then transferred onto a circular microtrench of 2 – 3 μm in radius and 600 nm in depth on a silicon substrate with prepatterned Ti/Au electrodes. A small amount of PDMS residual was left untreated. Samples of CrI₃ were handled inside a nitrogen-filled glovebox with oxygen and water less than 1 part per million (ppm) to avoid degradation. The thickness of atomically thin materials was first estimated by their optical reflection contrast and then measured by atomic force microscopy (AFM). The thickness of WSe₂ flakes was verified by optical reflection spectroscopy. The thickness of CrI₃ flakes was further verified by the magnetization versus out-of-plane magnetic field measurement. Six devices (two bilayers, one trilayer and three 6-layers) were studied. Devices with 4-layer and 5-layer CrI₃ samples were not investigated. Their behaviors are expected to be similar to the ones covered in this study.

Mechanical resonance detection

An optical interferometric technique was applied to detect the out-of-plane displacement of the resonators^{19,21,29} (Fig. 1a). The resonators were mounted in a closed-cycle cryostat (Attocube, attoDry1000). The output of a HeNe laser at 632.8 nm was focused onto the center of the suspended membrane using a high numerical aperture (N.A. = 0.8) objective. The beam size on the device was on the order of 1 μm and the total power was kept below 1 μW to minimize the laser heating effect. The reflected beam was collected by the same objective and detected by a fast photodetector. Motion of the membrane was actuated capacitively by applying a small r.f. voltage (~ 1 mV) between the membrane and the back gate. As the membrane moves in the out-of-plane direction, the optical cavity formed between the membrane and the trenched substrate modulates the reflectance. The amplitude of the motion as a function of driving frequency was measured by a network analyzer (Agilent E5061A), which provided the r.f. voltage and measured the fast photodetector response. The amplitude of the motion reaches its maximum when the driving frequency matches the natural frequency of the resonator. The signal-to-noise ratio (SNR) for typical measurements is around 100. It gives rise to a strain detection sensitivity of $\delta\epsilon \approx \frac{2}{Q \cdot \text{SNR}} \epsilon \sim 10^{-8}$, where the mechanical quality factor Q is ~ 2000 and the initial strain ϵ is $\sim 10^{-3}$. This value is comparable to the performance of high-strain-sensitivity magneto-strain sensors³⁰.

Magnetic circular dichroism (MCD) measurements

MCD was employed to characterize the membranes' magnetic properties. A nearly identical optical setup as the one used for the mechanical resonance detection was employed. The polarization of the optical excitation at 632.8 nm was modulated between left and right circular polarization by a photoelastic modulator at 50.1 kHz. Both the AC and DC components of the reflected beam were detected by a photodiode in combination with a lock-in amplifier and multimeter, respectively. MCD was determined as the ratio of the AC to DC component.

Strain calibration

It is challenging to determine the gate-induced strain $\epsilon(V_g)$ in the suspended membranes from the gate dependence of the resonance frequency (Fig. 1e) using the continuum model described in the text. Many parameters of the resonator are involved and cannot be determined independently. Instead, we determine $\epsilon(V_g)$ by using monolayer WSe₂ as a strain gauge assuming no relative sliding between WSe₂ and CrI₃ layers. It relies on the fact that the fundamental exciton resonance energy in monolayer WSe₂ redshifts linearly with strain (63 meV/%) for a relatively large range of biaxial strain²². The fundamental exciton energy in monolayer WSe₂ was determined as a function of V_g by optical reflection spectroscopy. In this measurement, broadband radiation from a single-mode fiber-coupled halogen lamp was employed as the source. A spectrometer equipped with a charge-coupled-device (CCD) camera was used for detection. The excitation power on the device was kept well below 0.1 μ W to minimize the laser heating effect. The gate dependence of the exciton resonance energy and the calibrated gate-induced strain is shown in Supplementary Fig. S5.

Characterization of resonator parameters

By minimizing the sum of the elastic energy and the electrostatic energy with respect to strain (i.e. $\frac{\partial}{\partial \epsilon} \left[\frac{3}{2} Y_{eff} \epsilon^2 - \frac{1}{2} C_g V_g^2 \right] = 0$), we obtain the gate-induced strain $\epsilon(V_g) \approx \frac{1}{96} \left(\frac{R}{D} \right)^2 \left(\frac{\epsilon_0 V_g^2}{D \sigma_0} \right)^2 \propto V_g^4$ for $\epsilon(V_g) \ll \epsilon_0$ (the built-in strain). Here the back gate capacitance C_g is strain or gate dependent because of the gate-induced vertical displacement z of the membrane, D is the vertical separation between the membrane and the back gate at $V_g = 0$, R is the drumhead radius, and ϵ_0 is the vacuum permittivity. The built-in stress σ_0 can be obtained from the slope of $\epsilon(V_g)$ as a function of V_g^4 (Supplementary Fig. S5c). The effective Young's modulus Y_{eff} and the 2D mass density ρ of the membrane can be obtained by fitting the experimental gate dependence of the resonance frequency using $f = \frac{\xi}{2\pi R} \sqrt{\frac{\sigma}{\rho}}$ with $\sigma = \frac{R^2}{4} \frac{\partial^2}{\partial z^2} \left[\frac{3}{2} Y_{eff} \epsilon^2 - \frac{1}{2} C_g V_g^2 \right]$ [Ref. ^{19,20,21}]. The extracted values for bilayer CrI₃ resonator 1 are $\sigma_0 \approx 0.5 \text{ Nm}^{-1}$, $\rho \approx 3 \times 10^{-5} \text{ kgm}^{-2}$, and $Y_{eff} \approx 600 \text{ Nm}^{-1}$. The mass density is 1.9 times of the expected mass density of the heterostructure presumably due to the presence of polymer residues and other adsorbates on the membrane. The effective Young's modulus is also consistent with the expected value. We estimate the effective Young's modulus of 2D heterostructures as the total contribution of constitute layers, $Y_{eff} = \sum_i n_i Y_{2D,i}$, where n_i and $Y_{2D,i}$ are the layer number and the 2D elastic stiffness per layer of the i -th material. We obtain $Y_{eff} \approx$

1200 Nm^{-1} for bilayer device 1 using the reported value for $Y_{2D,g} = 340 \text{ } Nm^{-1}$, $Y_{2D,CrI_3} = 25 \text{ } Nm^{-1}$ and $Y_{2D,WSe_2} = 112 \text{ } Nm^{-1}$ [Ref. ^{31,32,33,34}]. The Young's modulus is dominated by the contribution of graphene in most devices studied in this work, which typically consist of 3 layers of graphene, monolayer WSe_2 , and 2-6 layers of CrI_3 .

Mechanisms for mechanical resonance shift in 2D CrI_3 under a magnetic field

In the main text, we have assigned the competition between the internal magnetic interactions and elastic energy as the major mechanism for the observed mechanical resonance shift in 2D CrI_3 resonators under a magnetic field at $V_g = 0$. Other effects could potentially also give rise to a mechanical resonance shift in 2D CrI_3 . One such possibility is the magnetostatic pressure from the gradient of the magnetostatic energy in the $\hat{z}$ direction, $\mathbf{M} \cdot \frac{\partial \mathbf{B}}{\partial z}$. Here $\mathbf{M}$ is the sample magnetization and $\mathbf{B}$ is the magnetic field at the sample. However, for a nanometer thick sample with a lateral size of a few microns the gradient is expected to be negligible. This is supported by the absence of field dependence for the mechanical resonance up to high fields except at the spin-flip transitions (Supplementary Fig. S6).

Model for exchange magnetostriction in bilayer CrI_3

We consider bilayer CrI_3 under an out-of-plane magnetic field $\mu_0 H_\perp$. The free energy per unit area for a free membrane in the AF and FM state in the zero-temperature limit can be expressed as

$$F_{AF} = 2F_0 + J_\perp, \quad (S1)$$

$$F_{FM} = 2F_0 - J_\perp - 2\mu_0 M_0 \left(H_\perp - \frac{M_0}{t} \right). \quad (S2)$$

Here F_0 , M_0 , $J_\perp$ and t denote the free energy of each monolayer, saturation magnetization of each layer, interlayer exchange interaction, and interlayer distance, respectively. The spin-flip field $\mu_0 H_C$ can be evaluated by requiring $F_{AF} = F_{FM}$ [Ref. ¹³]. In the presence of fixed boundary, strain is developed with an elastic energy of $U_{el} = \frac{3}{2} Y_{eff} \epsilon^2$, where $\epsilon = (a - a_0)/a_0$ is the fractional change of the in-plane lattice constant a (which is fixed by the boundary) from its equilibrium value a_0 . To determine the equilibrium lattice constants a_{AF} and a_{FM} for the two states at the spin-flip transition, we consider the total energy $U_{tot} = U_{el} + F + U_b$. The last term $U_b = -\sigma_0 \epsilon = -2U_{el}$ takes into account the work done by the boundary/substrate to keep the area of the membrane fixed, *i.e.* in-plane lattice constant a fixed. By taking the derivative of the total energy with respect to strain, we obtain

$$\frac{\partial(-U_{el}+F_{AF})}{\partial \epsilon} = 2 \frac{\partial F_0}{\partial \epsilon} + \frac{\partial J_\perp}{\partial \epsilon} - 3Y_{eff} \epsilon_{AF} = 0, \quad (S3)$$

$$\frac{\partial(-U_{el}+F_{FM})}{\partial \epsilon} = 2 \frac{\partial F_0}{\partial \epsilon} - \frac{\partial J_\perp}{\partial \epsilon} - 3Y_{eff} \epsilon_{FM} = 0. \quad (S4)$$

The strain dependence of M_0 is ignored. The change of strain in the AF and FM states ϵ_{AF} and ϵ_{FM} can be found by subtracting (S3) from (S4)

$$\epsilon_{AF} - \epsilon_{FM} = \frac{2}{3Y_{eff}} \frac{\partial J_{\perp}}{\partial \epsilon}. \quad (S5)$$

The fractional change in the resonance frequency can be expressed as

$$\frac{f_{AF} - f_{FM}}{f_{AF}} \approx \frac{\sigma_{AF} - \sigma_{FM}}{2\sigma_{AF}} = \frac{1}{3\sigma_0} \frac{\partial J_{\perp}}{\partial \epsilon}, \quad (S6)$$

since $f \propto \sqrt{\sigma}$. Here we have taken the initial state to be AF. From experiment, we obtain $\frac{\partial J_{\perp}}{\partial \epsilon} > 0$, which corresponds to $\epsilon_{AF} - \epsilon_{FM} > 0$ according to Eq. (S5). The lattice constant of CrI₃ in the AF state is smaller than in the FM state.

For in-plane magnetic fields, minimizing U_{tot} with respect to strain [with F given by Eqn. (1)] yields

$$\frac{f_{AF} - f_{FM}}{f_{AF}} \approx \frac{1}{3\sigma_0} \left(\frac{\partial J_{\perp}}{\partial \epsilon} - 2 \frac{\partial K_{eff}}{\partial \epsilon} \right) \sin^2 \theta. \quad (S7)$$

Here θ is the spin canting angle, which is related to the applied and saturation magnetic fields as $\sin \theta = \frac{H_{\parallel}}{H_S}$ for $H_{\parallel} \leq H_S$. The result can be generalized to 6-layer CrI₃ as presented in the main text.

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Data availability

The data that support the findings of this study are available within the paper and its Supplementary Information. Additional data are available from the corresponding authors upon request.

Competing interests

The authors declare no competing interests.

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Contributions

S.J., K.F.M., J.S. and H.X. designed the study, discussed the results and co-wrote the manuscript. H.X. and S.J. developed the experimental setup and fabricated the devices. S.J. performed the bulk of the measurements and data analysis, assisted by H.X.

Figure captions

Fig. 1: 2D CrI₃ mechanical resonators. **a**, Schematic of the measurement system. The resonator (suspended 2D membrane on a Si trench of depth D) is actuated by an r.f. voltage from a vector network analyzer (VNA) through a bias tee. A DC voltage V_g is superimposed to apply static tension to the membrane. The motion is detected interferometrically by a HeNe laser, which is focused onto the center of the resonator. BS: beam splitter; PD: photodetector. **b**, Schematic of a bilayer CrI₃ resonator with

antiferromagnetic CrI₃ encapsulated by few-layer graphene and monolayer WSe₂. Filled spheres and arrows denote Cr atoms and spins in the top and bottom CrI₃ layers. **c**, Optical microscope image of a bilayer CrI₃ device suspended over a circular trench. Dashed line shows the boundary of the CrI₃ flake. Scale bar is 4 μm . **d**, Vibration amplitude (symbols) of bilayer CrI₃ resonator 1 (radius 2 μm) vs. driving frequency and a Lorentzian fit of the resonance spectrum (solid line). **e**, Gate dependence of the measured resonance frequency (symbols) and fit to the continuum model (solid line) with initial stress $\sigma_0 \approx 0.5 \text{ Nm}^{-1}$, mass density $\rho \approx 3 \times 10^{-5} \text{ kgm}^{-2}$ and effective Young's modulus $Y_{eff} \approx 600 \text{ Nm}^{-1}$.

Fig. 2: Mechanical detection of the spin-flip transition in 2D CrI₃. **a**, Normalized vibration amplitude of bilayer CrI₃ resonator 1 vs. driving frequency under an out-of-plane magnetic field ($\mu_0 H_{\perp}$) that sweeps from 1 T to -1 T to 1 T. **b**, Resonance frequency extracted from **a** as a function of magnetic field. **c**, Magnetic circular dichroism (MCD) of the membrane as a function of magnetic field. The red (blue) lines in **b** and **c** correspond to the measurement for the positive (negative) sweeping direction of the field. **d-f**, Same measurements as in **a-c** for 6-layer CrI₃ resonator 1 (radius 3 μm).

Fig. 3: Mechanical detection of spin canting in 2D CrI₃. **a**, Normalized vibration amplitude vs. driving frequency under an in-plane magnetic field ($\mu_0 H_{\parallel}$) that sweeps from 8 T to -8 T to 8 T for 6-layer CrI₃ resonator 1. **b**, Resonance frequency extracted from **a** as a function of magnetic field. Red (blue) symbols correspond to the measurement for the positive (negative) sweeping direction of the field. Dashed line is the expected quadratic dependence before saturation.

Fig. 4: Strain-tuning of the spin-flip transition in 2D CrI₃. **a, b**, Normalized Magnetic circular dichroism (MCD) as a function of out-of-plane magnetic field ($\mu_0 H_{\perp}$) that sweeps from -1 T to 1 T (only -0.6 T to 0.6 T is shown for clarity) at different gate voltages for a suspended (**a**) and a substrate-supported region (**b**) of bilayer CrI₃ resonator 1. **c**, Spin-flip transition field vs. gate voltage for the suspended (blue) and substrate-supported (red) region of the membrane. The lines are drawn to guide the eye. **d**, Spin-flip transition field as a function of gate-induced strain (symbols). The solid line is a linear fit.

Extended data figure captions

Extended Data Fig. 1: Mechanical resonance of 2D CrI₃ under an out-of-plane magnetic field. **a, b**, Field dependence of the amplitude (**a**) and linewidth (**b**) of the fundamental resonance of bilayer CrI₃ resonator 1. The field dependence of the resonance frequency is shown in Fig. 2b. The red (blue) symbols correspond to the measurement for the positive (negative) field sweeping direction. **c**, Field dependence of the reflectance at 633 nm normalized by the reflectance at zero field.

Extended Data Fig. 2: Summary of 6 CrI₃-resonators under an out-of-plane magnetic field. For each device, the contour plot shows the vibration amplitude vs. driving frequency under an out-of-plane field that sweeps from the top to the bottom value. The other two panels compare the field dependence of the resonance frequency (upper panel) and MCD (lower panel) of the membrane. The two colors denote the two field sweeping directions. **a-c**, Bilayer CrI₃ resonator 1 (radius 2 μm), same as Fig. 2a-c. **d-f**, Bilayer CrI₃ resonator 2 (radius 2 μm). **g-i**, Trilayer CrI₃ resonator (radius 3 μm). **j-l**, 6-layer CrI₃ resonator 1 (radius 3 μm), same as Fig. 2d-f. **m-o**, 6-layer CrI₃ resonator 2 (radius 3 μm). **p-r**, 6-layer CrI₃ resonator 3 (radius 3 μm). MCD of the trilayer resonator shows a ferromagnetic hysteresis loop centered at zero magnetic field (**i**), to which the mechanical resonance is not sensitive (**h**). The insets in **i** show the spin configuration of the system at given fields.

Extended Data Fig. 3: Summary of 2 CrI₃-resonators under an in-plane magnetic field. For each device, the contour plot shows the vibration amplitude vs. driving frequency under an in-plane field that sweeps from the top to the bottom value. The other panel is the field dependence of the resonance frequency. The two colors denote the two field sweeping directions. **a,b**, 6-layer CrI₃ resonator 1 (same as Fig. 3). **c,d**, 6-layer CrI₃ resonator 3.

Extended Data Fig. 4: Inverse magnetostriction in 6-layer CrI₃ resonator 2. **a**, Normalized MCD vs. out-of-plane magnetic field that sweeps from 2.2 T to -2.2 T (only 2 T to 0.5 T and -0.5 T to -2 T is shown for clarity) at different V_g 's. **b, c**, Strain dependence of the two spin-flip transition fields (symbols). The error bars correspond to the spin-flip transition widths. The solid lines are linear fits.

Extended Data Fig. 5: Calibration of gate-induced strain and determination of resonator parameters. **a**, Reflection contrast of bilayer CrI₃ resonator 1 from 1.6 – 1.9 eV as a function of gate voltage. The main feature is a dip (around 1.75 eV at $V_g = 0$ V), which corresponds to the fundamental exciton resonance of monolayer WSe₂. The feature redshifts slightly with V_g up to about 40 V followed by a larger redshift with further increase of V_g . **b**, Representative spectra at selected V_g 's. **c**, Exciton resonance energy extracted from **a** (left axis) and gate-induced strain calibrated from the exciton resonance energy (right axis) as a function of V_g^4 for V_g up to 39 V. The solid line is a linear fit. The built-in stress σ_0 was determined from the slope.

Extended Data Fig. 6: 2D CrI₃ resonators under high out-of-plane fields. Normalized vibration amplitude vs. driving frequency under an out-of-plane magnetic field up to 5 T for 6-layer CrI₃ resonator 2. The resonance frequency, amplitude and linewidth basically do not change up to 5 T except at the spin-flip transitions.

Figure 1

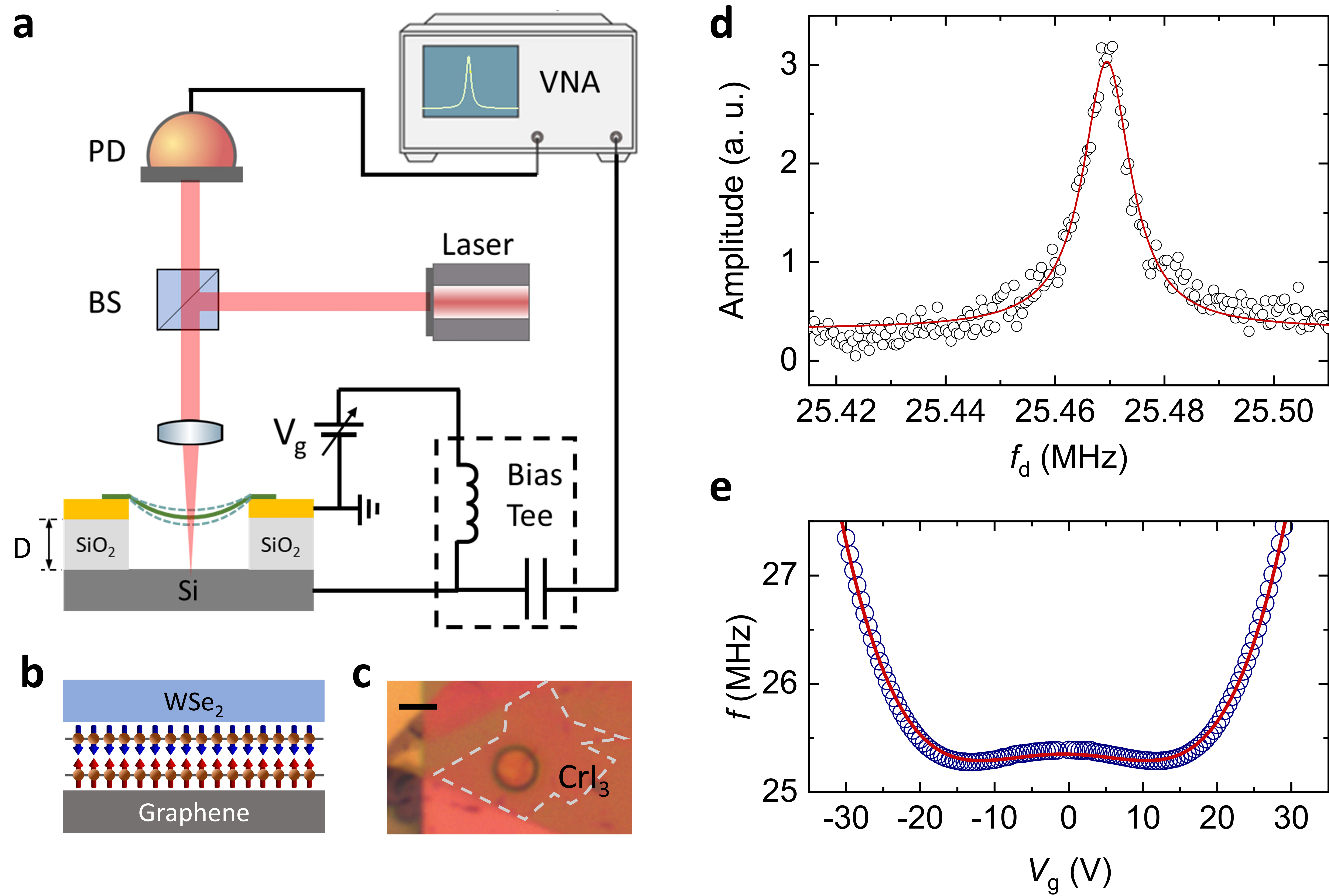


Figure 2

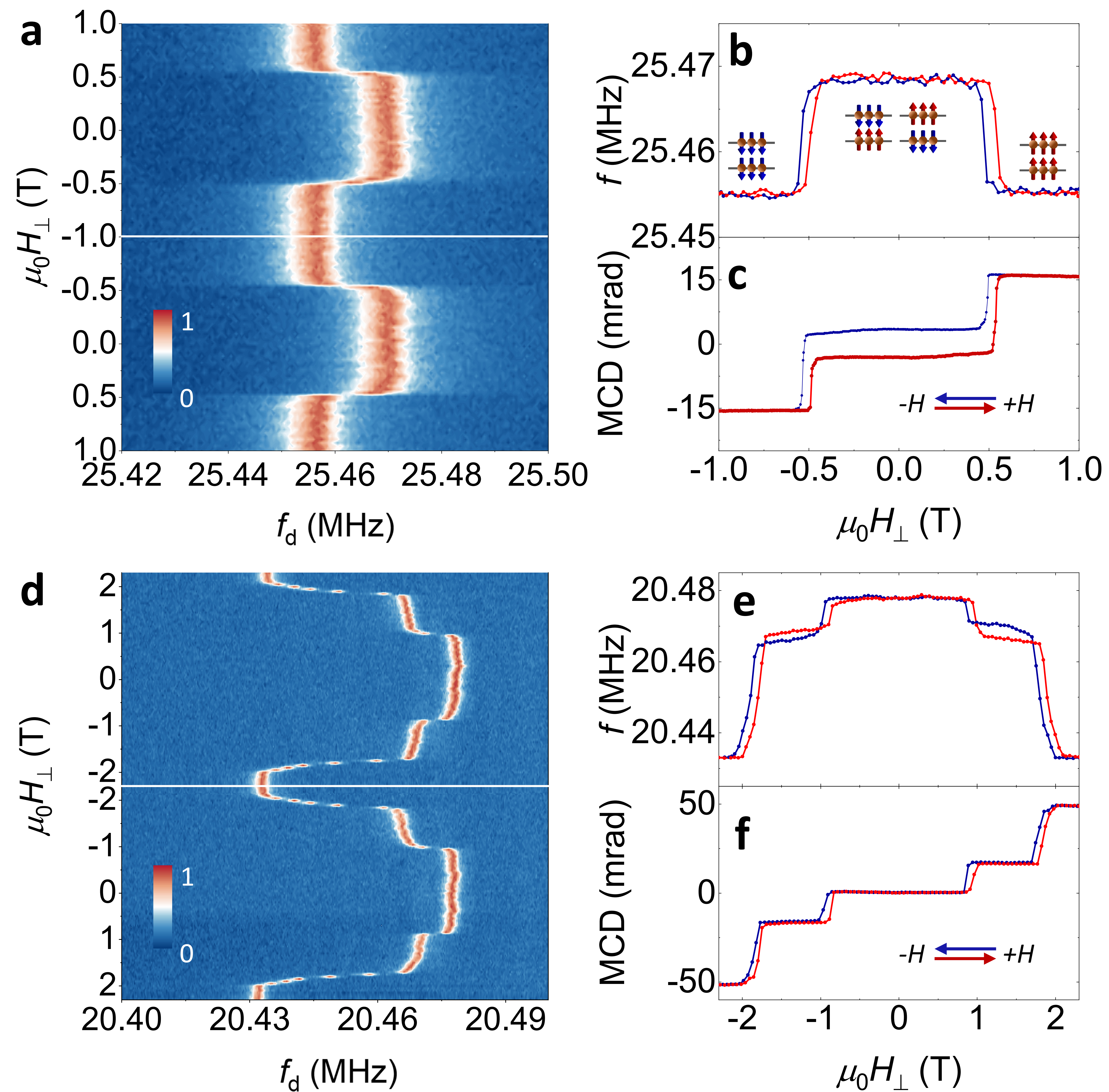


Figure 3

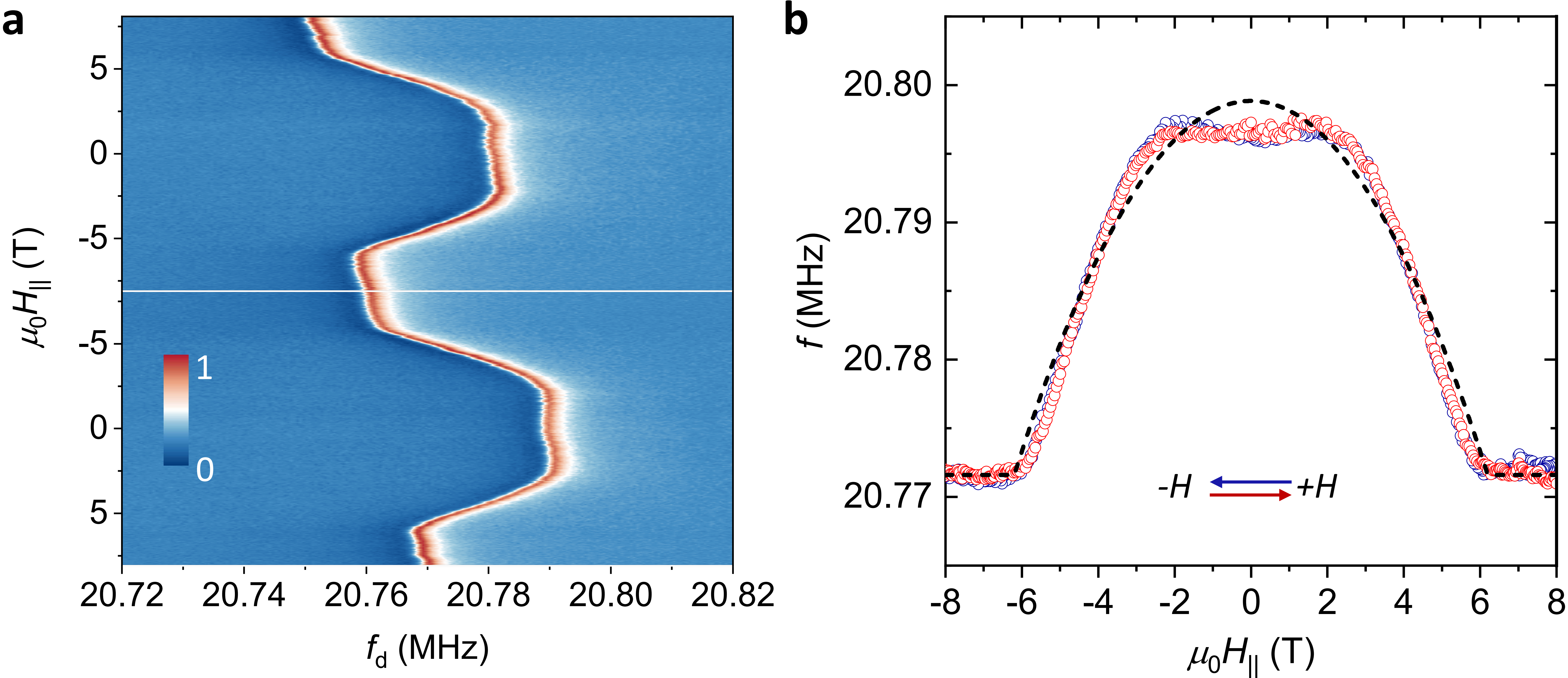


Figure 4

