

Supporting Information

Coexistence of Magnetic Orders in Two-Dimensional Magnet CrI_3

Ben Niu,^{1,2†} Tang Su,^{3,2†} Brian A. Francisco,² Subhajit Ghosh,⁴ Fariborz Kargar,⁴ Xiong Huang,⁵ Mark Lohmann,² Junxue Li,² Yadong Xu,² Takashi Taniguchi,⁶ Kenji Watanabe,⁶ Di Wu,¹ Alexander Balandin,⁴ Jing Shi,^{2*} Yong-Tao Cui^{2*}

1. National Laboratory of Solid State Microstructures, Department of Materials Science and Engineering, Jiangsu Key Laboratory for Artificial Functional Materials, and Collaborative Innovation Center of Advanced Microstructures, Nanjing University, Nanjing 210093, China
2. Department of Physics and Astronomy, University of California, Riverside, CA 92521, USA
3. International Center for Quantum Materials, School of Physics, Peking University, Beijing 100871, P. R. China
4. Department of Electrical and Computer Engineering, University of California, Riverside, CA 92521, USA
5. Department of Materials Science and Engineering, University of California, Riverside, CA 92521, USA
6. National Institute for Materials Science, 1-1 Namiki, Tsukuba, 305-0044, Japan

† These authors contribute equally to this work.

* Email: jing.shi@ucr.edu, yongtao.cui@ucr.edu

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Section I. Sample fabrication

CrI₃ flakes are exfoliated inside an argon-filled glovebox. A suitable CrI₃ flake is dry transferred and released either directly on top of a thin hBN flake or on a Pt (5 nm) covered hBN flake. The Pt film helps to screen the electrostatic potential from the SiO₂ substrate. Another hBN flake is then transferred on top of the CrI₃ to protect it from degradation. With the hBN capping, the sample can then be taken out of the glovebox for MFM measurement. The CrI₃ quality is confirmed with Raman spectroscopy (see Fig. S1).

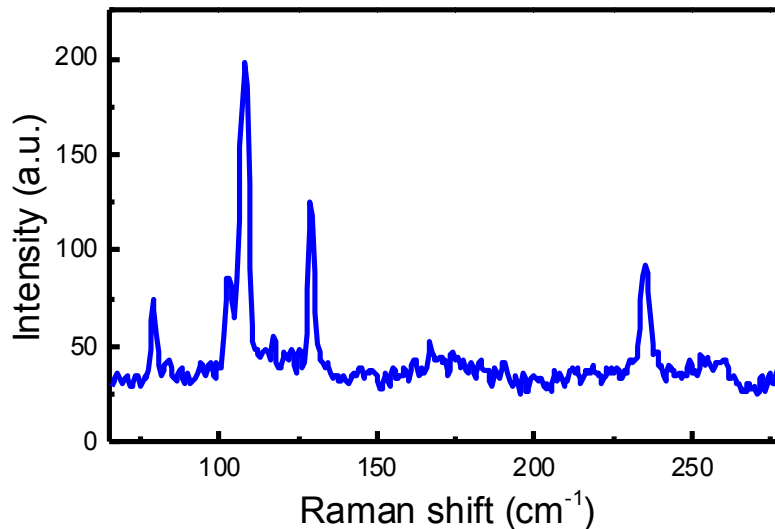


Figure S1. A typical Raman spectrum of the CrI₃ flake taken at room temperature.

Section II. Experimental setup

MFM measurements are performed in a home-built low temperature scanning probe microscope with Co-Cr coated commercial cantilever probes. The probe driven at its resonance (~ 75 kHz) by an AC excitation is raster scanned over the sample surface at constant heights to avoid topography induced artifacts. Phase locked loop (PLL) is used to obtain MFM signal, the change in the probe's mechanical resonant frequency.

1. MFM measurement simultaneously with electrostatic force microscope (EFM)

The change in the probe's mechanical resonance can be caused not only by the long-range magnetic force but also by the long-range electrostatic force. CrI₃ is a semiconductor with a bandgap of 1.2 eV, thus charge disorders possibly introduced during the fabrication process could lead to potential variation on the sample surface which will generate long-range electrostatic force. To nullify the effect of the potential disorder, we simultaneously perform EFM to compensate the electrostatic force. As illustrated in Fig. S2, at 2 T, when domain pattern appears during the reversal

process, EFM assisted MFM clearly uncovers magnetic domains (Fig. S2 (b)) which were dominated by the potential disorder (dark blue regions in Fig. S2 (a)), extracting the electrical potential distribution in Fig. S2 (c).

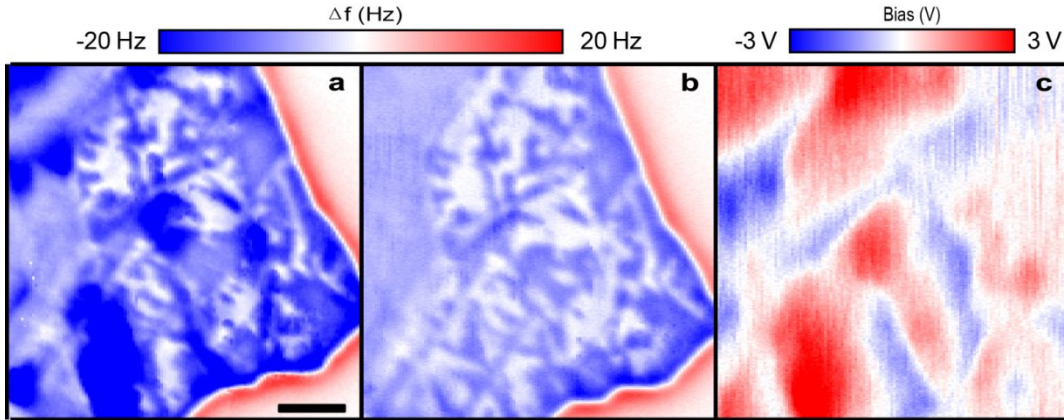


Figure S2. Performance of EFM-assisted MFM. MFM imaging of a 200 nm flake at 10 K and 2 T simultaneously with EFM (b) and without EFM (a). (c) demonstrates the potential distribution on the sample surface. Scale bar is 2 μm .

2. Quantitative characterization of the MFM signal as a function of magnetic field

In case of laterally uniform magnetization, MFM signal is proportional to the area density of moment integrated over the sample thickness. (See Section IV for further discussions on this property) To probe the magnetic moment more quantitatively, we perform MFM measurement in areas with fewer domains. Note that some of the contrast patterns seen in the MFM image at high field where the moments are expected to be fully polarized are due to regions of different thickness, which can be correlated well with the topography of the flake show in Fig. S3 (a). The tip is then scanned across the sample edge repeatedly along a single line as illustrated by the dashed arrow in Fig. S3 (b) while the field along c axis is varied from +5 T to -5 T. An example scan profile measured at 5 T is shown in Fig. S3 (c) in which a pair of maxima and minima indicates a dramatic change in the derivative of the magnetic field across the sample edge. There is also a peak in the sample region which is due to a topography edge between regions of different thicknesses. Away from that, the steady signal inside the sample bulk indicates a laterally uniform magnetization fully polarized by the applied field. The background signal corresponding to the non-magnetic substrate is further subtracted from each line. The resulting MFM signal is spatially averaged inside the sample bulk at each field and then plotted as a function of the field, which generates the MFM vs B plots. Importantly, sample position drifts should be corrected when sweeping the field.

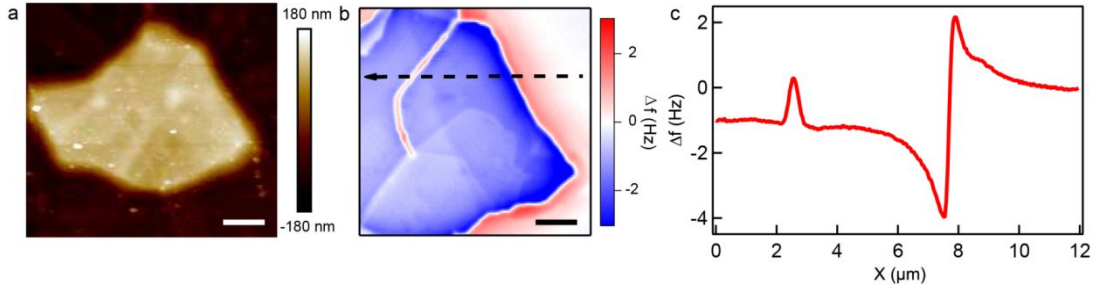


Figure S3. Scan profile at 5 T. (a) Topography imaging of the 200 nm flake. Scale bar is 5 μm (b) MFM imaging at 10 K and 5 T and a scanning line across the sample edge. Scale bar is 2 μm (c) Scan profile along the line. The signal difference between sample and non-magnetic substrate is extracted as the MFM signal.

Section III. MFM images

Figure S4 supplements MFM images of the flake in Fig. 1(b) at detailed fields above -0.1 T (Fig. S4 (a)) and below -0.1 T (Fig. S4 (b)). The domains due to the partial reversal are most pronounced at 2.05 T. Below -0.1 T, similarly to the trend of the positive field side, the average MFM signal in the bulk changes rapidly from -0.15 T to -0.7 T, then the domain configuration persists as the field is further reduced to -5 T except within a narrow range between -1.95 T and -2.1 T where magnetic domains appear.

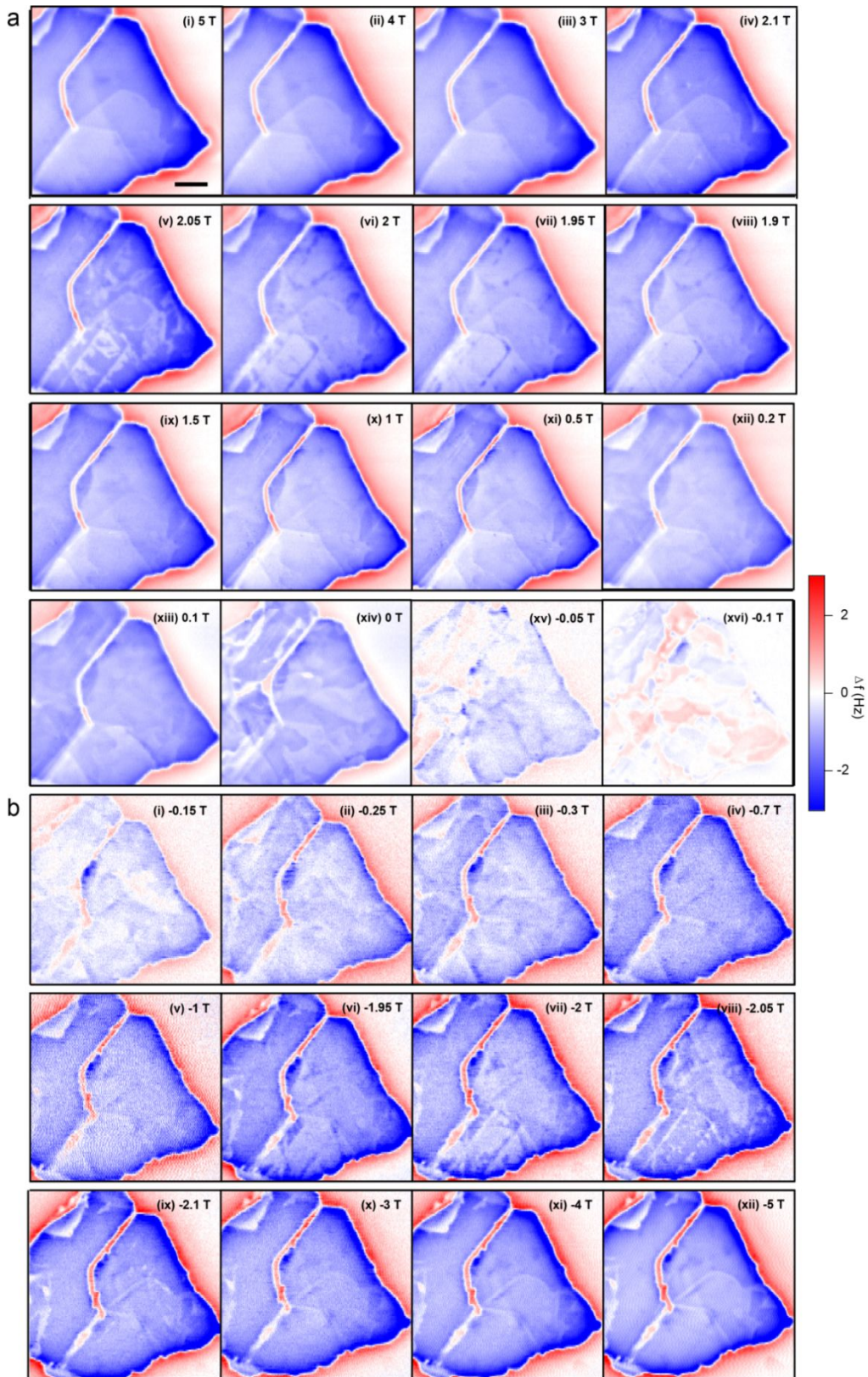


Figure S4. Field dependent MFM images at 10 K. At fields above (a) and below -0.1 T (b). Scale bar is 2 μm .

Section IV. Determination of the hard layer thickness

We calculated the thickness of the hard layer by multiplying the total layer thickness with the percentage fraction of the MFM signal change at 2 T which corresponds to the hard layer switching. This method assumes that the MFM signal is proportional to the thickness of the ferromagnetic layers that exhibit a net vertical moment when it is in a uniform magnetic state, i.e., without magnetic domains. We justify this assumption with finite element simulation, as shown in Fig. S5. The simulated MFM signal is the derivative of the z -component of the magnetic field with respect to z , dB_z/dz , evaluated at a fixed height (150 nm) above the surface of the magnetic flake. The thickness of the fully aligned magnetic flake is varied from 0 to 1000 nm hence the net vertical moment increases linearly. In the small thickness range relevant for our analysis (<200 nm), dB_z/dz is approximately linear, and it deviates from linear behavior more significantly at large thicknesses. The linear relation between MFM signal and flake thickness in the range of interest (<200 nm) indicates that surface layers and inner layers contribute approximately equally to the MFM signal, therefore the MFM signal change can be used to extract the thickness change of the magnetically aligned layers.

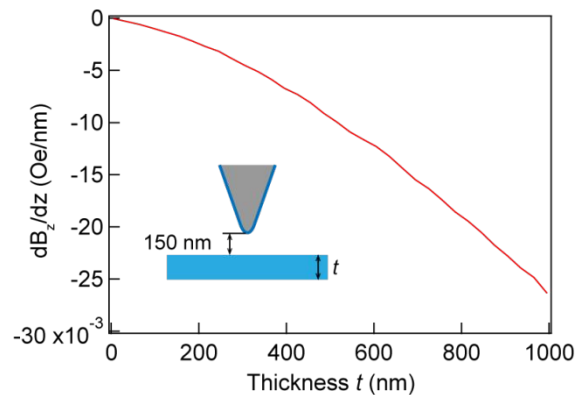


Figure S5. Simulated dB_z/dz as a function of the flake thickness. The derivative is evaluated at a fixed height (150 nm) from the top surface of the flake.

The physics for such behavior is the following. The magnetic field configuration is sensitive to the lateral geometry of the sample. When the magnetic state is uniform in a relative wide but thin magnetic flake, the magnetic field near the flake surface decays very slowly, which applies to our case where the tip-sample distance (100 - 200 nm) is much smaller than the flake's lateral size (several μm). (Recall that in the extreme case of an infinitely large magnetic thin film, the magnetic field would be perfectly perpendicular to the film and uniform in the entire space, not decaying at all.) While B_z itself already decays slowly over distance due to the large sample size relative to tip-sample distance, dB_z/dz changes even more slowly near the surface. Therefore, the contributions from surface layers and inner layers at the tip location are roughly the same, leading to the linear dependence of MFM signal (proportional to dB_z/dz) as a function of the flake thickness. We expect such behavior will break down as the tip is moved further away from the

surface. In particular, when the tip-sample distance is comparable to the lateral size of the sample ($\sim \mu\text{m}$), the magnetic field will behave more like that from a dipole magnet than that from a thin film magnet.

Based on this property, the MFM signal can be used to evaluate the relative change of the magnetic moment in a thin film magnet, given the following conditions: 1) “thin film geometry”: the sample thickness should be much smaller than the sample’s lateral size so that the magnetic field near the sample surface decays gently; 2) “near surface probe”: the tip-sample distance should also be much smaller than the sample’s lateral size; 3) “uniformly magnetized state”: the entire thin film should be in a uniform magnetic state without domain formation. Meanwhile, we note that the MFM signal cannot be used to obtain an absolute measurement of the magnetization because the field derivative depends sensitively on the lateral size of the sample. In our CrI₃ measurement, switching of the magnetic moments in different layers only causes the effective thickness of the magnetic layer to change, and all the conditions mentioned above are satisfied. Therefore, our analysis based on the relative change of the MFM signal within the same flake provides a good estimation on the thicknesses of the hard and soft layers.

The thickness of the top hBN is in the range of 20 – 40 nm, which varies from sample to sample. The tip distance from the actual surface of the CrI₃ flake will be slightly larger than the tip distance from the top hBN surface, but the total distance is still small compared to the sample size and will not change the roughly linear dependence of the MFM signal on the effective thickness of the magnetic flake.

Section V. Thickness dependence

Figure S6 plots MFM signal as a function of magnetic field for flakes of 110 nm, 100 nm, 80 nm, 30 nm and 25 nm at 10 K. The two-stage switching behavior is commonly observed in all samples except the 25 nm flake. The group of hard layers switches at around 2 T while the soft layers switch at low fields. Blue and red curves represent the data for increasing and decreasing the field. In general, the soft switching has a larger hysteresis than the hard switching at 2 T.

We would like to note that the absolute value of the MFM signal from samples of different lateral geometry cannot be compared directly. For example, in Fig. 2 of the main text, the MFM signal in the 200 nm flake is actually slightly smaller than that from the 110 nm flake, which is due to their different lateral size. As discussed in Section IV, the derivative of the magnetic field, dB_z/dz , is very sensitive to the lateral geometry of a thin flake. The 200 nm flake is $\sim 17 \mu\text{m}$ wide while the 110 nm flake is $\sim 6 \mu\text{m}$ wide (Fig. S7a). The larger width of the 200 nm flake will decrease the MFM signal inside the flake. As an illustration, we simulated the magnetic field derivative, dB_z/dz , in both a $17 \mu\text{m}$ wide and $6 \mu\text{m}$ wide stripe (the other side is infinitely long). The value of dB_z/dz at 150 nm height from the flake surface is plotted in Fig. S7b. The signal in the $17 \mu\text{m}$ wide / 200 nm thick flake is almost 10 times lower than the $6 \mu\text{m}$ wide / 110 nm thick flake. In reality, considering the irregular shape of the flakes and the fact that the MFM tip has a thin magnetic

coating over the entire tip body (not only at the tip apex), the measured MFM signal will be a complicated convolution of the dB_z/dz values in space. As a result, the absolute value of the MFM signal cannot be compared across samples of different lateral geometry.

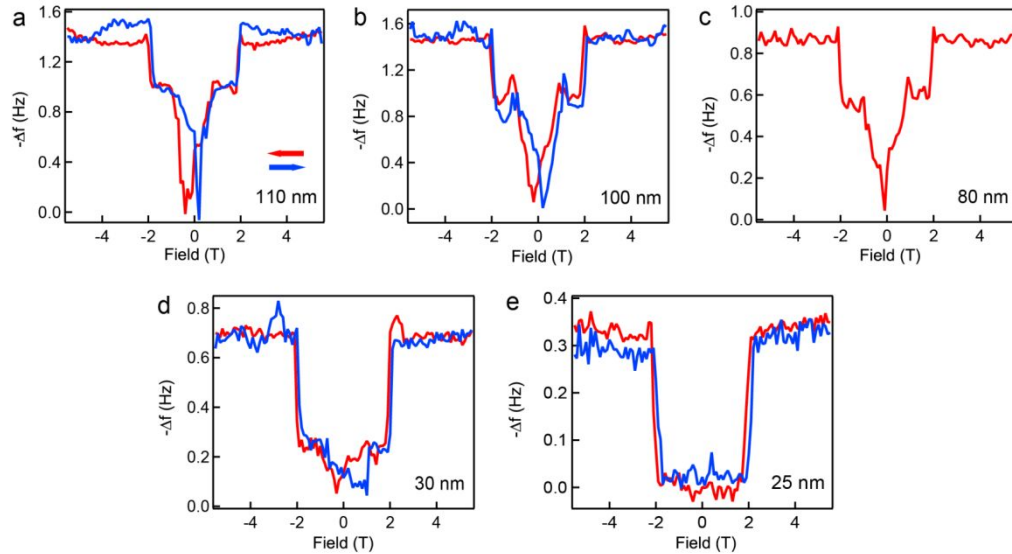


Figure S6. Two groups coexist in samples of various thicknesses. (a) 110 nm (b) 100 nm (c) 80 nm (d) 30 nm (e) 25 nm. Blue and red arrows indicate the direction of increasing and decreasing field.

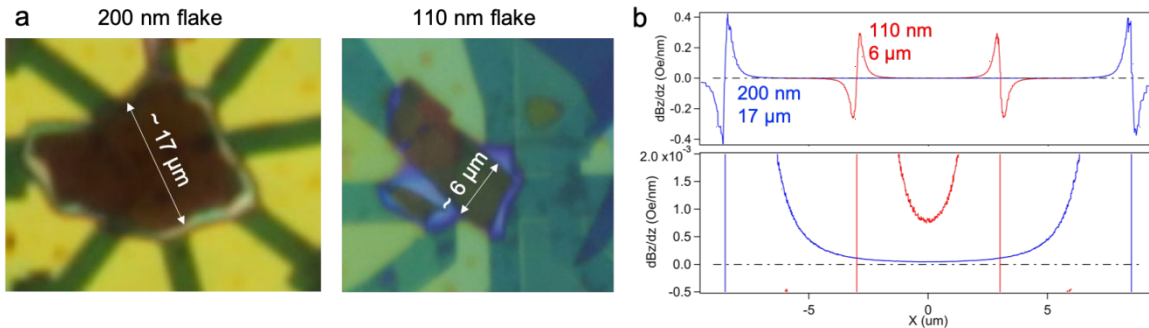


Figure S7. Simulated MFM signal in 200 nm and 110 nm flakes (a) Optical images of the 200 nm flake and 110 nm flake. (b) Simulated dB_z/dz signals across a $17\ \mu\text{m}$ wide / 200 nm thick (blue) and $6\ \mu\text{m}$ wide / 110 nm thick (red) flake, evaluated at 150 nm from the top surface.

Section VI. Temperature dependence

For samples of various thicknesses, MFM signal vs B curves at temperature from 10 K to 70 K are plotted in Figure S8. Through all thicknesses, the hard switching vanishes at ~ 45 K while the soft switching persists up to 60 K, resembling the behaviors in CrI_3 thin layers and bulk crystals, respectively.

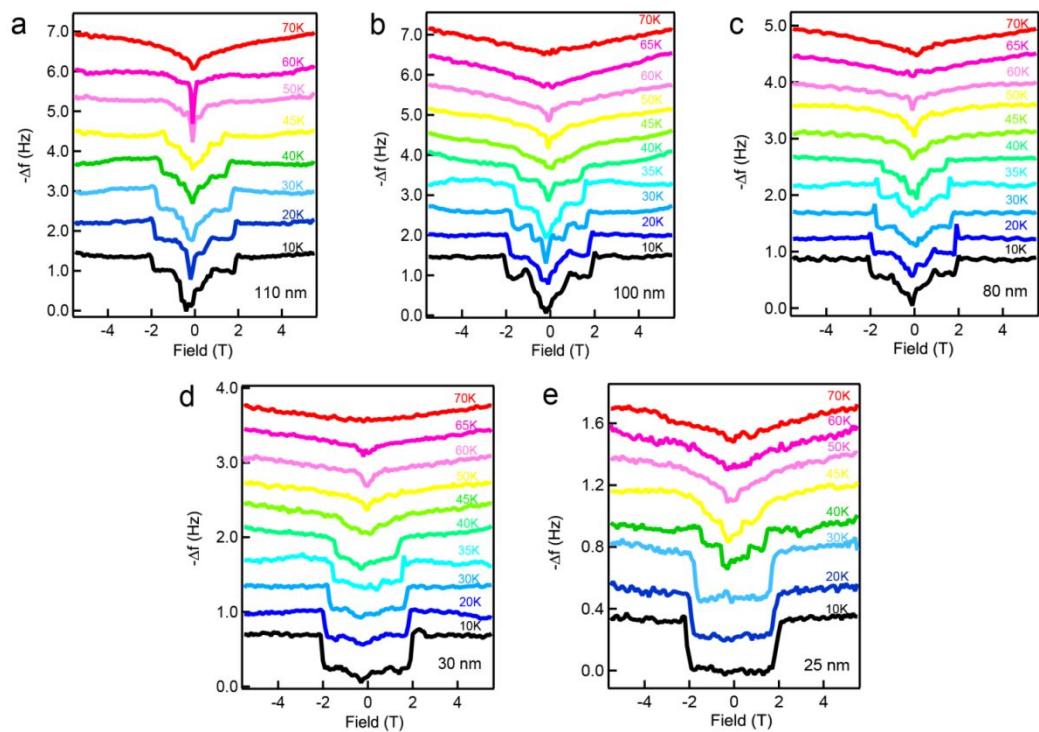


Figure S8. Temperature dependence of MFM vs B curves in samples of various thicknesses. (a) 110 nm, (b) 100 nm, (c) 80 nm, (d) 30 nm, (e) 25 nm.