

Evidence of Topological Hall Effect in Pt/Antiferromagnetic-Insulator

Bilayers

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Abstract

Topological Hall effect has been a primary indicator of non-trivial spin textures in magnetic materials. We observe the evidence of topological Hall effect in Pt/Cr₂O₃ bilayers grown on Al₂O₃(0001) and Al₂O₃(11 $\bar{2}$ 0), where Cr₂O₃ epitaxial film is an antiferromagnetic insulator. The Pt/Cr₂O₃ bilayers exhibit topological Hall resistivity for Cr₂O₃ thicknesses below 6 nm near and above room temperature, which is above the Néel temperature of Cr₂O₃, revealing the key role of thermal fluctuations in the formation of spin textures. The similarity of topological Hall signals in (0001)- and (11 $\bar{2}$ 0)-oriented Cr₂O₃ films indicates that the emergence of spin textures is insensitive to crystalline orientation.

Topological spin textures, such as skyrmions, have attracted intense interest in recent years.¹⁻⁸ The competition between exchange interaction, magnetic anisotropy, and Dzyaloshinskii-Moriya interaction (DMI) generates chiral spin textures in bulk non-centrosymmetric materials⁹ and magnetic multilayers.¹⁰ Electric transport measurement via the topological Hall (TH) effect is a powerful tool for detecting spin textures. In conducting skyrmion materials, the TH effect arises when electrons flowing in the spin textures experience an emergent electromagnetic field through interaction with the textured spins¹ and are scattered off of the textures, producing a uniquely identifiable TH voltage.¹¹

Magnetic skyrmions have been studied almost exclusively in bulk ferromagnets (FM) or FM multilayers. Antiferromagnets (AF)¹²⁻¹⁴ have also been predicted to harbor topological spin textures such as AF skyrmions.¹⁵⁻¹⁷ To date, except some non-collinear AFs with intrinsic chirality,¹⁸ only a few experiments have included AFs in their heterostructures to show spin chirality,^{19,20} where the AFs solely play an auxiliary role, such as providing an exchange bias¹⁹ or an AF heavy metal (HM) to generate a large interfacial DMI²⁰ while spin chirality emerges in the neighboring layer. Recently, thermally-driven spin chirality fluctuations have been predicted in materials with spin frustrations^{21, 22} and verified in 2D Ising FMs²³ even above their Curie temperatures (T_C). Here, we report the detection of topological spin textures in Pt/Cr₂O₃ bilayers using TH effect near and above the Néel temperature (T_N) of AF insulator (AFI) Cr₂O₃ films.

Epitaxial Cr₂O₃ thin films are grown on Al₂O₃(0001) and Al₂O₃(11 $\bar{2}$ 0) substrates using off-axis sputtering at a substrate temperature of 500°C.²⁴⁻²⁷ Figure 1(a) shows $2\theta/\omega$ X-ray diffraction (XRD) scans for Cr₂O₃(30 nm) films on Al₂O₃(0001) and Al₂O₃(11 $\bar{2}$ 0). Figure 1(b) shows an XRD rocking curve of the Cr₂O₃ (0006) peak for the (0001)-oriented film [the (11 $\bar{2}$ 0)-oriented film gives a similar rocking curve]. The clear Laue oscillations and narrow full-width-at-half-maximum

(FWHM) of the rocking curves indicate the high crystal quality of the Cr_2O_3 films in both orientations. Figure 1(c) shows an X-ray reflectometry (XRR) scan of the Cr_2O_3 film on $\text{Al}_2\text{O}_3(0001)$, where the fitting gives a roughness of 0.1 nm. The smooth surface is confirmed by atomic force microscopy (AFM) as shown in Fig. 1(d) with a roughness of 0.1 nm. The scanning transmission electron microscopy image for a 30 nm Cr_2O_3 film on $\text{Al}_2\text{O}_3(0001)$ in Fig. 1(e) reveals the epitaxy and single crystal ordering of Cr_2O_3 .

Given that Cr_2O_3 is insulating, the predicted topological spin textures in the AF, if exist, cannot be directly detected electrically. We deposit a 2 nm Pt layer on Cr_2O_3 films of various thicknesses (t) at room temperature using off-axis sputtering, where the Pt layer provides a channel for Hall measurement as well as interfacial DMI needed for the formation of spin textures. The $\text{Pt}(2 \text{ nm})/\text{Cr}_2\text{O}_3(t)$ bilayers are patterned into Hall bars with a current channel width of 100 μm .

Figure 2(a) shows the Hall resistivity for a $\text{Pt}(2 \text{ nm})/\text{Cr}_2\text{O}_3(5 \text{ nm})$ bilayer on $\text{Al}_2\text{O}_3(0001)$ at a temperature (T) of 345 K with an out-of-plane field (H) and a DC current of 100 μA . In general, there are three terms in the Hall resistivity, $\rho_{xy} = \rho_{\text{OH}} + \rho_{\text{AH}} + \rho_{\text{TH}}$, including the ordinary Hall (ρ_{OH}), anomalous Hall (ρ_{AH}), and topological Hall (ρ_{TH}) resistivities.^{7,28} ρ_{OH} has a linear field dependence. At 345 K, ρ_{AH} in the $\text{Pt}/\text{Cr}_2\text{O}_3$ bilayer is due to the spin-Hall anomalous Hall effect (SH-AHE).^{29,30} Since 345 K is higher than the $T_N = 307 \text{ K}$ ³¹ of bulk Cr_2O_3 and $\sim 280 \text{ K}$ for 20 nm $\text{Cr}_2\text{O}_3(0001)$,³² we use the Langevin function to fit the SH-AHE data as shown in Fig. 2(a), which agrees well with the experimental data at $|H| < 0.2 \text{ T}$ and $|H| > 1.4 \text{ T}$. However, there is a pronounced upturn at $0.2 \text{ T} < |H| < 1.4 \text{ T}$ that cannot be explained by the SH-AHE. We think it is very likely that the topological Hall effect (THE) leads to this upturn feature, which can be extracted by subtracting the OHE and SH-AHE. The obtained ρ_{TH} vs. H plot is presented in Fig. 2(b), which shows a TH region within $|H| < 1.4 \text{ T}$ with a peak value of 0.45 $\text{n}\Omega \text{ cm}$ at $\pm 0.4 \text{ T}$ and

sign switching at $H = 0$.

Following the same process, we measure ρ_{xy} of the Pt(2 nm)/Cr₂O₃(5 nm) bilayer on Al₂O₃(0001) from $T = 250$ to 385 K and fit the data using the Langevin function, as shown in Fig. 2(c). Figure 2(d) shows the corresponding ρ_{TH} vs. H plots and the inset gives the temperature dependence of ρ_{TH} , which indicate that ρ_{TH} disappears below 300 K and above 365 K. The observed THE in our Pt/Cr₂O₃ bilayer indicates spin chirality in the Cr₂O₃ film, which is attributed to the DMI at the Pt/Cr₂O₃ interface (For more details about the DMI at Pt/Cr₂O₃ interface, see *Supplementary Materials*). The spin chirality at $T > T_C$ has been observed in FM SrRuO₃ and V-doped Sb₂Te₃,²³ which was explained as spin chirality fluctuations in quasi-two-dimensional FMs with weak DMI. Unlike the FMs where the THE appears within a small temperature window near and above T_C , the THE in the AF Cr₂O₃ films is robust even at 60 K above its bulk T_N .

We next conduct Hall measurement for various Cr₂O₃ thicknesses at 345 K, as shown in Figs. 3(a) and 3(b). The TH peaks persist at $t = 3$ to 5 nm and disappear at 6 nm. Besides, the ρ_{TH} peak position moves from 0.8 to 0.4 T as t increases from 3 to 5 nm.²³ Figure 3(c) shows the thickness dependence of ρ_{TH} , which drops suddenly to zero from 5 to 6 nm, indicating that the DMI is not large enough to stabilize spin textures in thicker Cr₂O₃ films.²³ Figure 3(e) summarizes the temperature-thickness phase diagram of ρ_{TH} for the Pt/Cr₂O₃ bilayers on Al₂O₃(0001).

To better understand the observed THE, we use Monte-Carlo (MC) simulations to model the spin textures. Cr₂O₃ has a corundum structure and a rhombohedral unit cell. According to neutron measurements and first-principle calculations,^{33, 34} the first and second-nearest neighbor exchange coupling constants, J_1 and J_2 , are dominant. Thus, the Hamiltonian can be written as,

$$H = -\sum_{\langle i,j \rangle} J_1 (\mathbf{S}_i \cdot \mathbf{S}_j) - \sum_{\langle k,l \rangle} J_2 (\mathbf{S}_k \cdot \mathbf{S}_l) + \sum_{\langle i,j \rangle} \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j) - B_z \sum_i S_i^z - K \sum_i (S_i^z)^2, \quad (1)$$

where $\mathbf{S}$ is the unit spin vector, the third term is the DMI, the fourth term is the Zeeman energy,

and the last term is the uniaxial anisotropy along $\text{Cr}_2\text{O}_3(0001)$. We choose $J_1 = -1, J_2 = -0.74$ based on first-principle calculations, and $K = 1$. D ranges from 0.1 to 0.5 to simulate DMI of different Cr_2O_3 thicknesses, which forbids spin chirality at low temperatures. We use six layers with 31×31 spins in each layer to rebuild the exchange coupling in the Cr_2O_3 rhombohedral unit cell along (0001) with the periodic boundary condition. The DMI is only exerted on the first layer and the spin chirality is indicated by the topological charge Q of the top layer²²

$$Q = \frac{1}{4\pi} \int d^2 \mathbf{r} \mathbf{S} \cdot (\partial_x \mathbf{S} \times \partial_y \mathbf{S}), \quad (2)$$

which can be calculated by the summation of the solid angle determined by three neighboring unit spin vectors in the first layer of spin lattice.³⁵ Figure 3(d) shows the field dependence of Q at $D = 0.5$ and $T = 1.05 T_N$, where T_N is determined by the temperature dependence of susceptibility χ through MC simulation.³⁶ The antisymmetric curve of Q in Fig. 3(d) for Cr_2O_3 is similar to the reported simulations for FMs.²³ Since $\rho_{\text{TH}} \propto Q$, our simulations confirm the presence of spin chirality at $T > T_N$. Figure 3(f) summarizes the phase diagram of Q for $\text{Cr}_2\text{O}_3(0001)$ with varying T and $1/D$ ($D \propto 1/t$ for interfacial DMI³⁷). The simulation captures the key features of the experimental data that spin chirality of $\text{Cr}_2\text{O}_3(0001)$ appears near and above T_N .

Cr_2O_3 is a uniaxial AF with spins aligned along the easy axis $[0001]$. To explore how the crystallographic and spin orientations affect the formation of spin textures, we perform the same measurements of ρ_{xy} for Pt/ Cr_2O_3 bilayers grown on $\text{Al}_2\text{O}_3(11\bar{2}0)$ with an in-plane easy axis. Figure 4(a) shows the $\rho_{xy} - \rho_{\text{OH}}$ vs. H data for a Pt(2 nm)/ Cr_2O_3 (5 nm) bilayer on $\text{Al}_2\text{O}_3(11\bar{2}0)$ from $T = 250$ to 385 K with are Langevin fitting for ρ_{AH} . The TH peaks are observed from 300 to 385 K, as compared to 325-365 K for Pt/ $\text{Cr}_2\text{O}_3(0001)$. In addition, the TH peak appears at smaller fields, 0.2 T at 345 K as compared to 0.4 T for Pt/ $\text{Cr}_2\text{O}_3(0001)$. Figure 4(b) shows the field dependence of ρ_{TH} obtained by subtracting the fitting curve of ρ_{AH} from $\rho_{xy} - \rho_{\text{OH}}$. The magnitude

of ρ_{TH} reaches the maximum at 325 K, as seen in the inset of Fig. 4(b). Surprisingly, there is small non-zero TH signal even at 385 K. The differences in the TH signals between the bilayers grown on (0001)- and (11 $\bar{2}$ 0)-orientated Al_2O_3 may be attributed to the different magnitude of uniaxial anisotropy, and T_{N} due to strain,^{32, 38} the direction of uniaxial anisotropy or spin configurations at the surface layer of Cr_2O_3 . The detailed mechanism needs further exploration. However, we can conclude that the $\text{Pt}/\text{Cr}_2\text{O}_3(11\bar{2}0)$ bilayers share similar topological Hall features with the $\text{Pt}/\text{Cr}_2\text{O}_3(0001)$ bilayers.

Figure 5(a) shows the thickness dependence of $\rho_{xy} - \rho_{\text{OH}}$ for $\text{Pt}/\text{Cr}_2\text{O}_3(t)$ bilayers on $\text{Al}_2\text{O}_3(11\bar{2}0)$ at 345 K, from which ρ_{TH} is extracted as shown in Fig. 5(b). The TH signal persists from 3 to 6 nm and vanishes at $t = 10$ nm. Meanwhile, the TH peak position is lowered from 0.6 to 0.2 T as t increases from 3 to 6 nm, which is similar to the $\text{Pt}/\text{Cr}_2\text{O}_3(0001)$ bilayers. The magnitude of ρ_{TH} has the maximum at 3 nm and then gradually decreases as Cr_2O_3 becomes thicker [Fig. 5(c)]. Figure 5(d) shows the MC simulation of topological charge Q in a varying out-of-plane field. For $\text{Cr}_2\text{O}_3(11\bar{2}0)$, we use four layers to rebuild the AF exchange coupling to the second-nearest neighbor. The last term in Eq. (1) is rewritten as $-K \sum_i (S_i^x)^2$ because of the in-plane easy axis. The similarity of Q between $\text{Cr}_2\text{O}_3(11\bar{2}0)$ and $\text{Cr}_2\text{O}_3(0001)$ is consistent with our experimental results. Compared with the $\text{Cr}_2\text{O}_3(0001)$ in Fig. 3(e), $\text{Cr}_2\text{O}_3(11\bar{2}0)$ has a broader $T-t$ range for THE as shown in Fig. 5(e). Figure 5(f) shows the simulation, which indicates a larger phase space of Q , possibly due to the different crystal orientation and anisotropy.

The similar phase diagrams of spin textures in the Cr_2O_3 films with different crystal and spin orientations indicate the minor role played by the uniaxial anisotropy. We believe the spin textures in our $\text{Pt}/\text{Cr}_2\text{O}_3$ bilayers have similar fundamental origin as that in FM SrRuO_3 and V-doped Sb_2Te_3 .²³ The broken inversion symmetry at the $\text{Pt}/\text{Cr}_2\text{O}_3$ interface provides the DMI. Due

to the relatively small DMI at Pt/Cr₂O₃ interface compared to the exchange interaction in Cr₂O₃,³⁹ spin textures can only emerge near or above T_N with the assistance of thermal fluctuations to overcome the strong AF exchange interaction. In conducting FM or AF layers that exhibit THE, conduction electrons are deflected by the emergent electromagnetic field due to the spin chirality. Proximity effect^{40, 41} in Pt/Cr₂O₃ bilayers is another possibility to explain the mapping of spin textures from Cr₂O₃ to Pt; while to date, the existence of proximity in Pt/Cr₂O₃ is debatable⁴¹⁻⁴³ and it is unlikely to preserve the moment in Pt at $T > T_N$.⁴⁴ We propose a new mechanism, spin Hall topological Hall effect (SH-THE),^{45, 46} which is similar to the SH-AHE in Pt/FMI and AFI bilayers. The SH-THE arises from the following process: 1) as a current flows through the Pt layer, it generates spin accumulation via the spin Hall effect at the Pt/Cr₂O₃ interface; 2) the spin accumulation exerts a spin torque on the AF moments in Cr₂O₃, which form spin textures at appropriate thickness and temperature range; 3) the spins in Pt at the interface experience a reaction torque from the winding AF spin textures in Cr₂O₃ and acquire a Berry phase, resulting in a topological Hall effect. This process should apply to both FMI and AFI based bilayers. Similar SH-THE has also been observed in Pt/Tm₃Fe₅O₁₂ bilayers.⁴⁶

In summary, we observe strong evidence of topological Hall effect in Pt/Cr₂O₃ bilayers at temperatures near or above the T_N , revealing the emergence of spin textures in collinear AF insulator films. This first evidence of topological Hall effect in HM/AFI bilayers significantly expands our materials base to include the large family of AF insulators for the exploration of AF-based skyrmion technology.

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Figure Captions:

Figure 1. (a) $2\theta/\omega$ XRD scans of a 30 nm Cr_2O_3 epitaxial film grown on $\text{Al}_2\text{O}_3(0001)$ and $\text{Al}_2\text{O}_3(11\bar{2}0)$ substrates. (b) XRD rocking curve of the Cr_2O_3 (0006) peak for the film on $\text{Al}_2\text{O}_3(0001)$. (c) XRR scan of a 30 nm Cr_2O_3 film on $\text{Al}_2\text{O}_3(0001)$, where the fitting (black curve) gives a Cr_2O_3 thickness of 30 nm and a roughness of 0.1 nm. (d) AFM image of a 30 nm Cr_2O_3 film on $\text{Al}_2\text{O}_3(0001)$ with a surface roughness of 0.1 nm. (e) STEM image of a 30 nm Cr_2O_3 film on $\text{Al}_2\text{O}_3(0001)$.

Figure 2. Topological Hall (TH) measurements of a $\text{Pt}(2\text{ nm})/\text{Cr}_2\text{O}_3(5\text{ nm})$ bilayer on $\text{Al}_2\text{O}_3(0001)$. (a) Hall resistivity ρ_{xy} of $\text{Pt}(2\text{ nm})/\text{Cr}_2\text{O}_3(5\text{ nm})/\text{Al}_2\text{O}_3(0001)$ at 345 K, where the black curve is the fitting for ρ_{AH} with Langevin function and linear background due to ρ_{OH} . (b) TH resistivity after subtraction of the fitting curve for ρ_{AH} and ρ_{OH} in (a). (c) $\rho_{xy} - \rho_{\text{OH}}$ at various temperatures from 250 to 385 K, where the black curves are the fitting for ρ_{AH} with Langevin function. (d) ρ_{TH} after subtraction of the fitting curve for ρ_{AH} in (c) at each temperature. Inset: ρ_{TH} as a function of temperature.

Figure 3. (a) Hall resistivity $\rho_{xy} - \rho_{\text{OH}}$ of $\text{Pt}(2\text{ nm})/\text{Cr}_2\text{O}_3(t)$ bilayers on $\text{Al}_2\text{O}_3(0001)$ at 345 K with different thickness (t) of Cr_2O_3 , where the black curves are the fitting for ρ_{AH} with Langevin function. (b) ρ_{TH} of the bilayers after subtraction of the fitting curve in (a). (c) ρ_{TH} as a function of Cr_2O_3 thickness measured at 345 K. (d) Monte-Carlo simulations of field dependence of topological charges Q at $T = 1.05T_{\text{N}}$ ($K = |J_1| = 1$ and $D = 0.5|J_1| = 0.5$ with out-of-plane easy axis anisotropy). (e) Contour plot of ρ_{TH} of $\text{Pt}(2\text{ nm})/\text{Cr}_2\text{O}_3(t)$ bilayers on $\text{Al}_2\text{O}_3(0001)$ as a function of temperature and Cr_2O_3 thickness. (f) Contour plot of topological charges Q as a function of temperature T/T_{N} and $1/D$ based on Monte-Carlo simulation. The red circles in (e) and (f) are the data points.

Figure 4. (a) Hall resistivity $\rho_{xy} - \rho_{OH}$ of a Pt(2 nm)/Cr₂O₃(5 nm) bilayer on Al₂O₃(11 $\bar{2}$ 0) at various temperatures from 250 to 345 K, where the black curves are the fitting for ρ_{AH} with Langevin function. (b) TH resistivity ρ_{TH} after subtraction of the fitting curve for ρ_{AH} in (a) at each temperature. Inset: ρ_{TH} as a function of temperature.

Figure 5. (a) Hall resistivity $\rho_{xy} - \rho_{OH}$ of Pt(2 nm)/Cr₂O₃(t) bilayers on Al₂O₃(11 $\bar{2}$ 0) at 345 K with different Cr₂O₃ thicknesses where the black curves are the fitting for ρ_{AH} with Langevin function. (b) ρ_{TH} of the bilayers after subtraction of the fitting curve in (a). (c) ρ_{TH} as a function of Cr₂O₃ thickness measured at 345 K. (d) Monte-Carlo simulations of field dependence of topological charges Q at $T = 1.05T_N$ ($K = |J_1| = 1$ and $D = 0.5 |J_1| = 0.5$ with easy in-plane anisotropy). (e) Contour plot of ρ_{TH} of Pt(2 nm)/Cr₂O₃(t) bilayers on Al₂O₃(11 $\bar{2}$ 0) as a function of temperature and Cr₂O₃ thickness. (f) Contour plot of topological charges Q as a function of temperature T/T_N and $1/D$ based on Monte-Carlo simulation. The red circles in (e) and (f) are the data points.

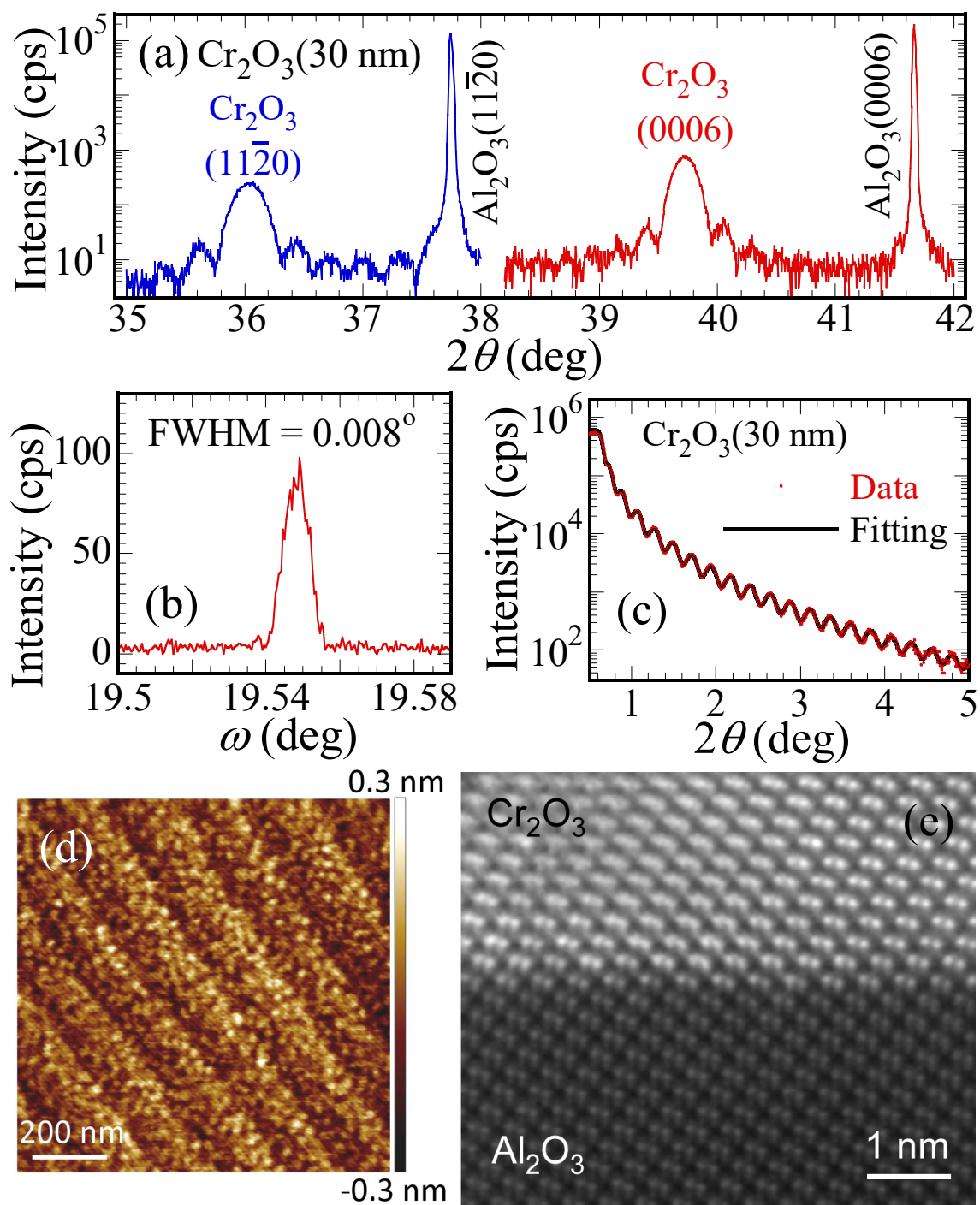


Figure 1.

Pt(2 nm)/Cr₂O₃(5 nm)/Al₂O₃(0001)

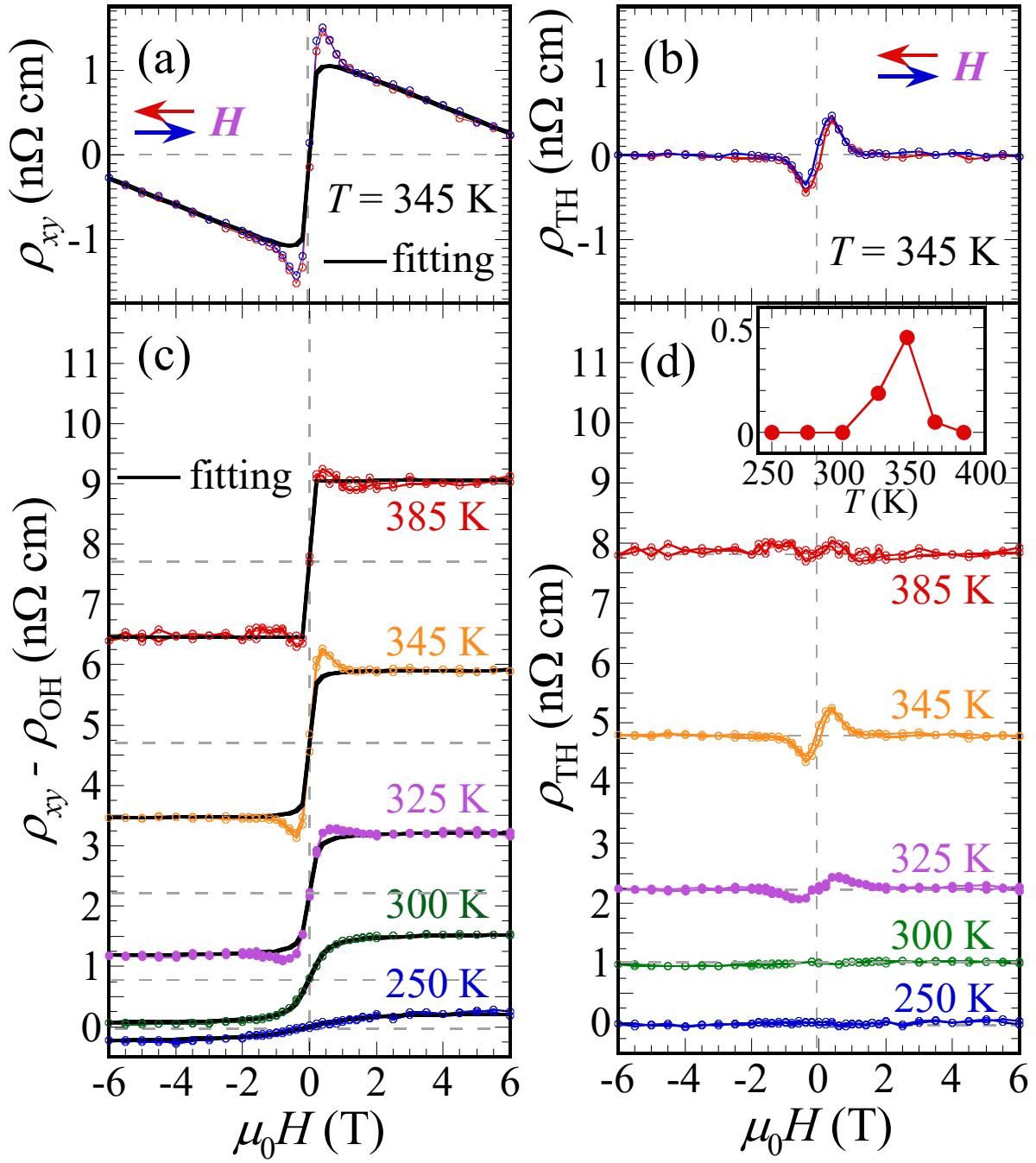


Figure 2.

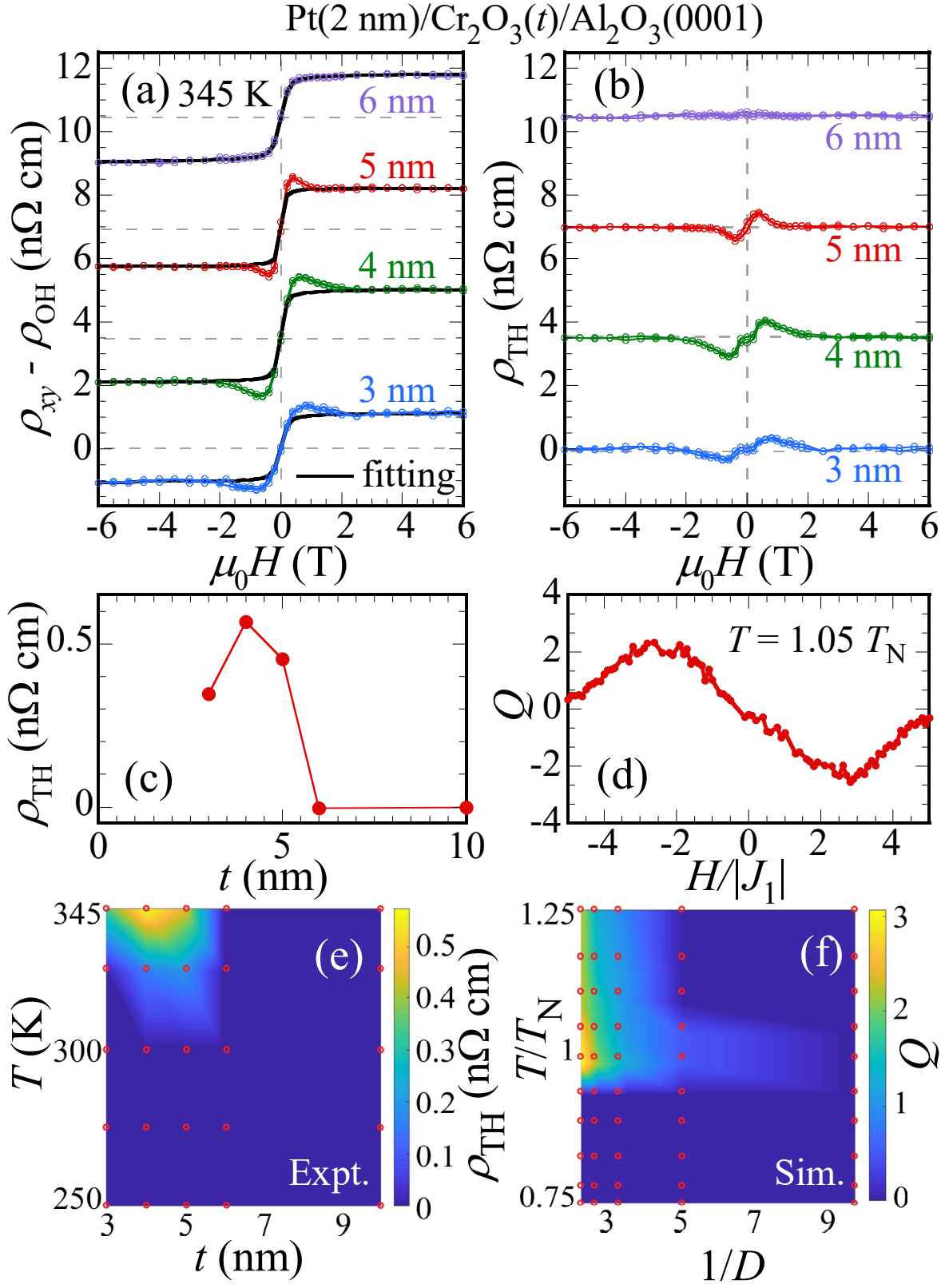


Figure 3.

Pt(2 nm)/Cr₂O₃(5 nm)/Al₂O₃(11 $\bar{1}$ 20)

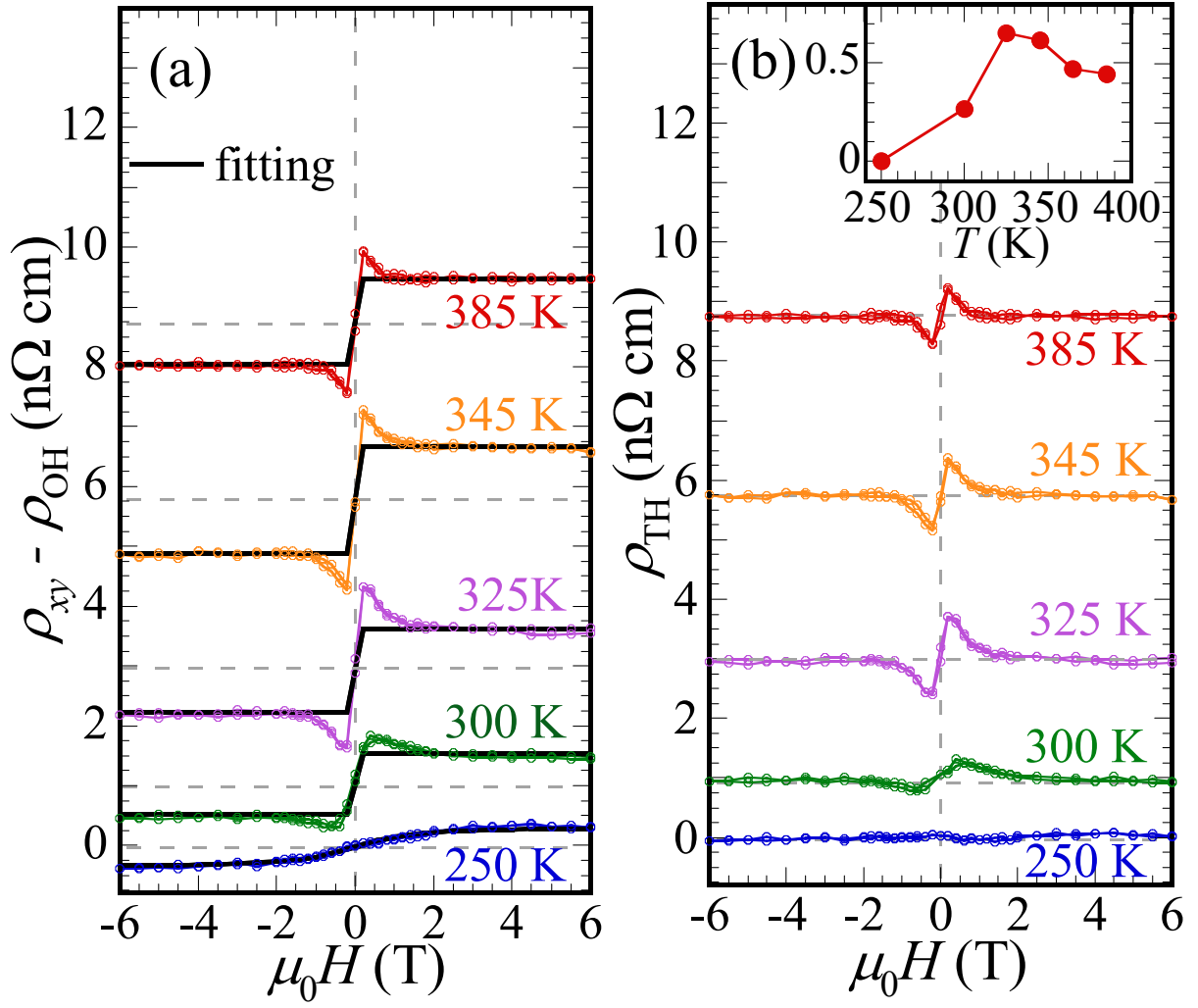


Figure 4.

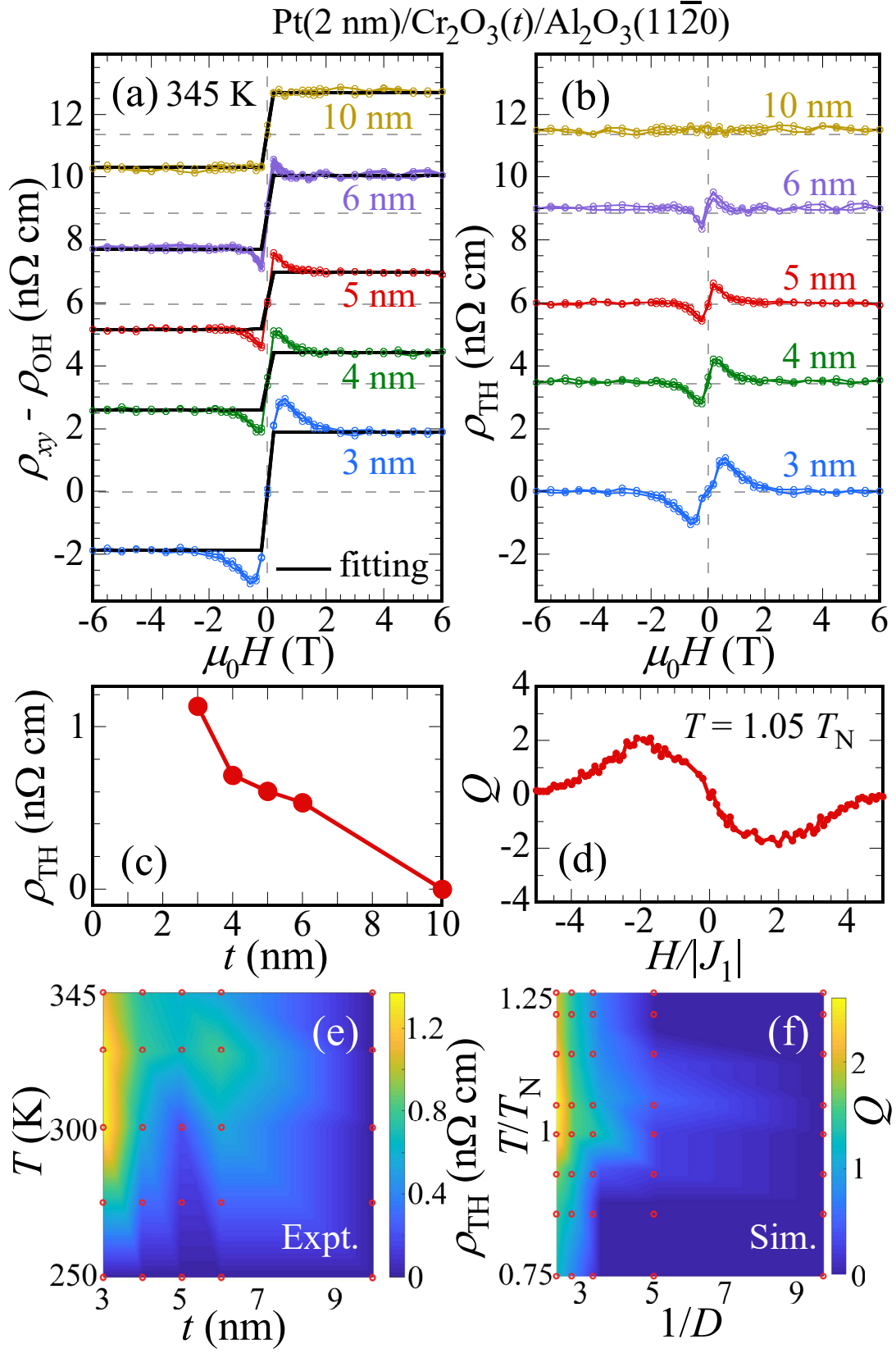


Figure 5.