

Robust spin injection via thermal magnon pumping in antiferromagnet/ferromagnet hybrid systems

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Robust spin injection and detection in antiferromagnetic thin films is a prerequisite for the exploration of antiferromagnetic spin dynamics and the development of nanoscale antiferromagnet-based spintronic applications. Previous studies have shown spin injection and detection in antiferromagnet/nonmagnetic metal bilayers; however, spin injection in these systems has been found effective at cryogenic temperatures only. Here, we experimentally demonstrate sizable interfacial spin transport in a hybrid antiferromagnet/ferromagnet system, consisting of Cr_2O_3 and permalloy, which remains robust up to the room temperature. We examine our experimental data within a spin diffusion model and find evidence for the important role of interfacial magnon pumping in the signal generation. The results bridge spin-orbitronic phenomena of ferromagnetic metals with antiferromagnetic spintronics and demonstrate an advancement toward antiferromagnetic spin-torque devices.

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I. INTRODUCTION

With their robustness against perturbation to magnetic fields and ultrafast spin dynamics, antiferromagnets (AFMs) are at the forefront of spintronics research as promising material candidates. However, efficient manipulation and readout of antiferromagnetic spin dynamics remains a major challenge [1]. Antiferromagnetic insulators [2], being of particular interest due to absent electrical shunting and low magnetic dissipation, rely on interfacial spin-charge conversion [3–7]. AFM/NM interfaces, where NM is a nonmagnetic metal with large spin-orbit coupling (e.g., Pt), have been used for accessing magnetic order [8,9] via spin Hall magnetoresistance. Furthermore, Néel vector switching has been observed in multiterminal AFM/NM devices [10–15], whereby thermal, structural, and spin-orbit phenomena have been discussed [16]. In studies of spin transport through ultra-thin AFM layers between NM and a ferromagnet, contributions of spin fluctuations and coherent evanescent spin waves have been investigated [17–20]. In spin Seebeck experiments [21–24], thermally driven spin currents across the spin-flop (SF) transition in the easy-axis AFM Cr_2O_3 have been observed [25,26]. These works have shown that the interfacial spin mixing conductance decreases substantially with increasing temperature [27] – an observation that is in agreement with temperature-dependent spin pumping mea-

surements in AFM/NM at sub-THz and GHz frequencies [25,28] and underlines the challenges of functionalizing AFM/NM interfaces for spintronic applications [1].

Ferromagnetic (FM) metals have recently gained attention due to the versatility of spin-orbitronic phenomena inherent to them [29,30]. Theoretical [29,31,32] and experimental works [33,34] on planar Hall and anomalous Hall spin injection have sparked interest for ferromagnetic metals as tunable spin injectors and detectors. Observation of magnetization-tunable spin torques in two-magnet bilayers and FM/NM multilayers [35] and recent formulation of the spin-dependent anomalous Hall effect [32,36] underline the potential of ferromagnetic metals as efficient spin-charge converters. Realization of magnetic autooscillations driven by thermal spin currents [37] as well as magnon-magnon coupling and transduction [18,38–43] further substantiate the technological relevance of the two-magnet architecture concept and raise the question whether antiferromagnet-ferromagnet hybrids have potential for the development of functional AFM-based spin-torque devices.

Here, we experimentally demonstrate thermal spin injection from a ferromagnetic metal, permalloy ($\text{Py} = \text{Ni}_{80}\text{Fe}_{20}$), into a thin film of an insulating antiferromagnet, Cr_2O_3 . We observe magnonic pumping with different spin across the SF transition that allows for sizable signals at room temperature. The results show promise for spintronics applications that take advantage of both antiferromagnetic spin dynamics and spin-orbitronic concepts inherent to ferromagnetic metals and pave the way toward AFM-FM hybrid spin-torque devices.

II. EXPERIMENT

We grow 100 nm thin Cr_2O_3 films [24] on m -plane ($10\bar{1}0$) sapphire (Al_2O_3) substrates [Fig. 1(a)] using rf-magnetron

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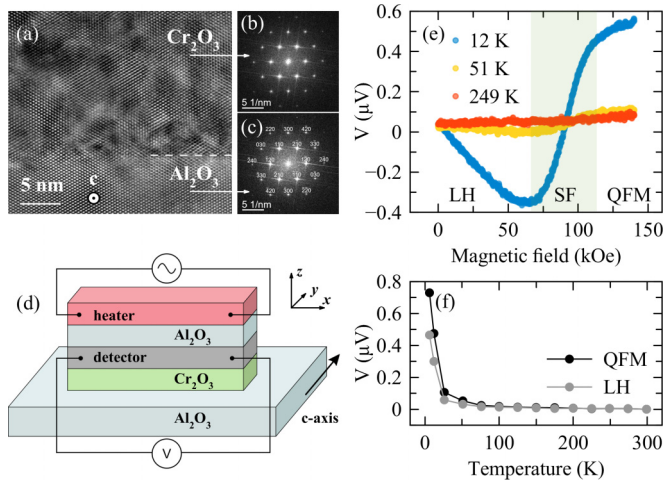


FIG. 1. (a) Cross-sectional transmission electron micrograph of the m -plane $\text{Al}_2\text{O}_3/\text{Cr}_2\text{O}_3$ interface with the c -axis [0001]-oriented out of the page and (b,c) the corresponding diffractograms of Cr_2O_3 and Al_2O_3 indicating epitaxial order. (d) Sample geometry. The magnetic field is applied parallel to the c -axis. (e) Spin Seebeck (SSE) voltage of the $\text{Cr}_2\text{O}_3/\text{Pt}$ sample shows a sign change across the SF transition. (f) SSE signals dominated by left-hand (LH) and quasiferromagnetic (QFM) magnons fall rapidly with increasing temperature.

sputter deposition. The films reveal epitaxial growth, a well-defined c -axis [0001] within the film plane [Figs. 1(b) and 1(c)], and have a typical surface roughness of 0.5 nm. The films are capped with one of three types of spin current detector layer using magnetron sputtering: (i) a nonmagnetic detector layer (4 nm)Pt, (ii) a ferromagnetic detector layer (5 nm)Py, and (iii) a combination detector layer (5 nm)Py/(2 nm)Pt. The multilayers are structured into $2.4 \times 0.25 \text{ mm}^2$ bars using optical lithography and ion milling. The axis of the bar is perpendicular to the c -axis of Cr_2O_3 . An electrically insulating (80 nm) Al_2O_3 buffer layer is deposited using atomic layer deposition. Finally, a metallic heater is deposited onto the buffer layer, with the same dimensions and lateral position as the underlying detector bar [Fig. 1(d)]. The goal of the above procedure is to obtain three sets of directly comparable devices that differ by choice of the spin current detection layer. A low-frequency electric current of 3 mA (rms) is supplied to the heater, resulting in a dissipated power of $\sim 4 \text{ mW}$ and in a periodic temperature gradient across the $\text{Cr}_2\text{O}_3/\text{detector}$ interface. The voltage generated by the detector layer is measured on the current's second harmonic using a lock-in amplifier. The measurements are performed in a cryostat with bath temperature T and carried out with the same experiment parameters for all samples.

In the $\text{Cr}_2\text{O}_3/\text{Pt}$ bilayer, the temperature gradient leads to an interfacial spin current which is converted into a voltage signal V via inverse spin Hall effect (iSHE) [44,45]. The SHE symmetry requires the spin polarization and thus the c -axis of Cr_2O_3 to lie within the film plane, as realized in our m -plane films. This spin Seebeck signal is shown in Fig. 1(e) as a function of magnetic field H applied parallel to the c -axis. The plot shows three distinct field regimes. At low fields, the spin current is dominated by the lowest-energy antiferromagnetic magnons [27,46] of Cr_2O_3 [Fig. 4(c)]. These are left-handed

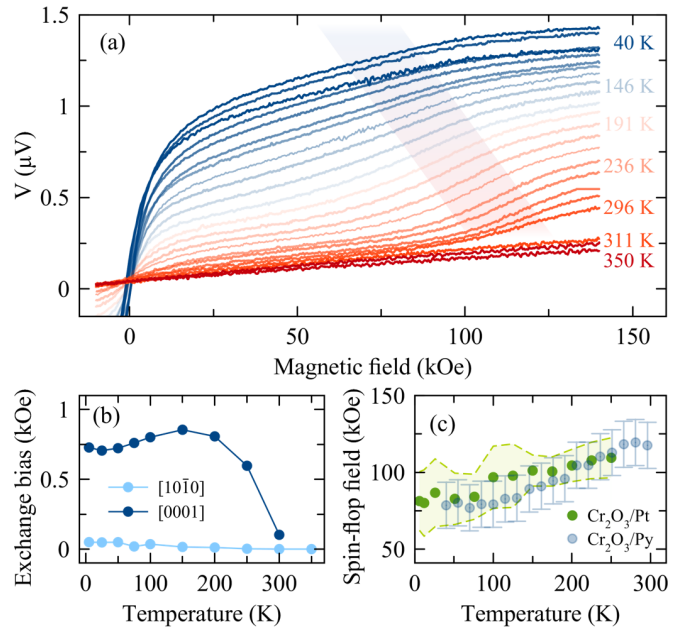


FIG. 2. (a) Thermally induced signal of the $\text{Cr}_2\text{O}_3/\text{Py}$ sample as a function of magnetic field. The Cr_2O_3 -related signal is superimposed on the sigmoidlike anomalous Nernst signal of Py magnetization reversal and disappears at the Néel temperature. The SF field region is indicated by the shading. (b) Exchange-bias field of a $\text{Cr}_2\text{O}_3/\text{Py}$ sample measured perpendicular and parallel to the c -axis. (c) The SF field regions of the $\text{Cr}_2\text{O}_3/\text{Pt}$ and $\text{Cr}_2\text{O}_3/\text{Py}$ samples overlap.

(LH) magnons with $-\hbar$ spin and generate a negative signal. With increasing magnetic field, the uniaxial two-sublattice spin system of Cr_2O_3 undergoes a SF transition, leading to a sign reversal of the spin Seebeck signal. Above the SF field, the spin current is dominated by the quasiferromagnetic (QFM) magnons which carry spin $+\hbar$. Starting from $T = 11 \text{ K}$, with increasing bath temperature, both the LH and QFM signals rapidly fall, as shown in Fig. 1(f). At room temperature the signal is negligibly small (which is consistent with previous findings [26]). The spin transport at the AFM/NM interface is robust only in the narrow range of low temperatures.

For the $\text{Cr}_2\text{O}_3/\text{Py}$ bilayer, as shown in Fig. 2(a), the thermally generated signal is dominated by the sigmoidlike shape due to the anomalous Nernst effect (ANE) of Py [47,48]. With magnetic field traversing through zero, Py magnetization undergoes a reversal, which is translated by the temperature gradient across the Py layer into a sign change of the voltage signal. In a control sample of a Py bar deposited directly on an m -plane Al_2O_3 substrate, as expected, a steplike ANE signal is observed (see the Appendix). In the $\text{Cr}_2\text{O}_3/\text{Py}$ bilayer, on the other hand, the signal approaches saturation over the entire field range. Such high-field saturation effects have been studied in ferromagnetic thin films [49–51] and discussed to originate from interfacial spin pinning. We carry out exchange-bias measurements in $\text{Cr}_2\text{O}_3/\text{Py}$ bilayer films. The observed exchange-bias field of $\lesssim 800 \text{ Oe}$ along the c -axis, shown in Fig. 2(b), corroborates the hypothesis of the interfacial pinning of Py spins [52,53].

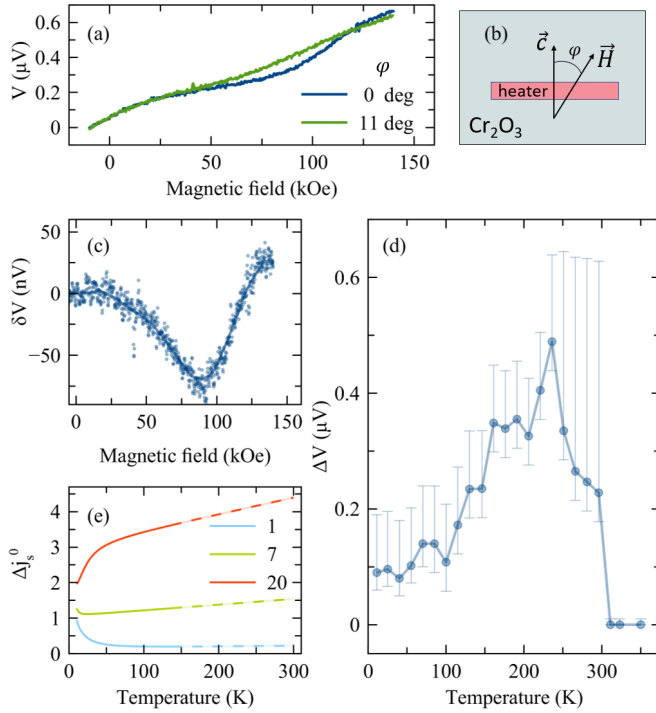


FIG. 3. (a) Thermally generated signal of $\text{Cr}_2\text{O}_3/\text{Py}$ as a function of magnetic field applied at angle ϕ with respect to the c -axis of Cr_2O_3 , as shown in the top view of the device (b). (c) Difference between the thermally generated spectra with aligned and misaligned magnetic field approximates the SSE signal, which shows a sign change across the SF transition. (d) SSE voltage jump across the SF transition in $\text{Cr}_2\text{O}_3/\text{Py}$ increases with increasing temperature, before collapsing at the Néel temperature. (e) Injected spin current jump across the SF transition (in arbitrary units) is calculated for various ratios $g_m^{\uparrow\downarrow}/g_n^{\uparrow\downarrow}$ of the spin transmission coefficients. Dashed lines represent an extension of the theory based on the magnon approximation to high temperatures.

Superimposed on the ANE signal of Py, another conspicuous signal introduces an inflection point in the 80–120 kOe field range [shaded region in Fig. 2(c)]. The inflection point field increases with increasing temperature; it follows the SF field [54], shown in Fig. 2(c), which we extract from measurements on $\text{Cr}_2\text{O}_3/\text{Pt}$ where the signal is not obscured by the ANE contributions. Moreover, the superimposed signal disappears in the temperature range of 296–311 K [Fig. 2(a)], where we find the Néel temperature $T_N = 305$ K of the Cr_2O_3 film [55]. Remarkably, the signal first increases with increasing temperature before collapsing at the Néel temperature, even though the underlying ANE signal continuously decreases with increasing temperature. These observations confirm that the signal superimposed on the ANE sigmoid originates from the Cr_2O_3 spin system and its mechanism is different from ANE. We refer to it as the spin Seebeck (SSE) signal.

The ANE background in conjunction with the interfacial pinning does not allow for an exact quantitative determination of the Cr_2O_3 -related SSE signal. We therefore employ two holistic methods for its assessment: (i) At 240 K, we intentionally misalign the magnetic field direction with respect to the c -axis of Cr_2O_3 by up to $\phi = 11^\circ$, as shown in Figs. 3(a) and

3(b). Such misalignment leads to a partial suppression of the SSE signals [56]. We then subtract the misaligned spectrum from the aligned one. This procedure cannot be used to fully subtract the underlying ANE signal, due to a residual SSE signal in the misaligned geometry and the angle-dependent nature of the the ANE signal, and provides a qualitative assessment of the SSE signal. The resultant difference curve δV [Fig. 3(c)] first decreases and then increases with increasing magnetic field, similarly to the SSE signal of $\text{Cr}_2\text{O}_3/\text{Pt}$ in Fig. 1(e). The δV difference spectrum confirms that the SSE signal in fact shows a sign change across the SF field, the latter being in agreement with the data shown in Fig. 2(c). (ii) We graphically extract the signal difference ΔV by simply taking the values of V below and above the SF from the field-dependent data of Fig. 2(a). While this procedure imposes a notable systematic error to the data (see the Appendix), it affirms the temperature dependence of the SSE signal in $\text{Cr}_2\text{O}_3/\text{Py}$ that was conspicuous from the raw data in Fig. 2(a): The signal increases with increasing temperature and collapses at the Néel temperature [Fig. 3(d)]. This behavior is in stark contrast to the thermal spin signal in $\text{Cr}_2\text{O}_3/\text{Pt}$ which vanishes above 50 K. In the $\text{Cr}_2\text{O}_3/\text{Py}$ hybrid system, the observed robust spin transport near room temperature is a prerequisite for practical AFM-based applications and calls upon a microscopic evaluation.

III. THEORETICAL MODEL

We attribute the Cr_2O_3 -related signal to the spin-dependent anomalous Hall effect (SAHE) [32] in Py, which converts the interfacial spin current into a transverse voltage, functioning as a spin detector. Under the device geometry depicted in Fig. 1(d), the spin-dependent electron transport in Py can be characterized by

$$j_x^\pm = -\sigma_\pm \frac{\partial \mu_\pm}{\partial x} + \theta_{\text{AH}}^\pm j_z^\pm + \Sigma_{\text{AN}}^\pm \frac{\partial T}{\partial z}, \quad (1)$$

$$j_z^\pm = -\sigma_\pm \frac{\partial \mu_\pm}{\partial z} - \theta_{\text{AH}}^\pm j_x^\pm, \quad (2)$$

where $\pm$ represents spin-up/down polarization along the x direction and $\mu_\pm$ is the spin-dependent chemical potential. $\sigma_\pm$, $\theta_{\text{AH}}^\pm$, $\Sigma_{\text{AN}}^\pm$ are the spin-dependent electrical conductivity, anomalous Hall, and anomalous Nernst coefficients of the Py [21]. In the linear response regime, these quantities can be parametrized as $\sigma_\pm = \sigma(1 \pm P)$, $\theta_{\text{AH}}^\pm = \theta_{\text{AH}}(1 \pm \beta)$, and $\Sigma_{\text{AN}}^\pm = \Sigma_{\text{AN}}(1 \pm \alpha)$ [29], where P , β , and α are phenomenological constants.

Defining $\mathbf{j}_c = \mathbf{j}^\uparrow + \mathbf{j}^\downarrow$ as the charge current density and $\mathbf{j}_s = \mathbf{j}^\uparrow - \mathbf{j}^\downarrow$ as the spin current density, and using $\mu_c = (\mu_\uparrow + \mu_\downarrow)/2$ as the charge chemical potential and $\mu_s = (\mu_\uparrow - \mu_\downarrow)/2$ as the spin chemical potential, we can solve the continuity conditions $\nabla \cdot \mathbf{j}_c + \partial \mu_c / \partial t = 0$ and $\nabla \cdot \mathbf{j}_s + \partial \mu_s / \partial t = -\mu_s / \tau_s$, where τ_s is the spin relaxation time. Up to linear order in $\partial T / \partial z$, P , β , and α , we obtain the spin diffusion equation and its solution as

$$\frac{\partial^2 \mu_s}{\partial z^2} = \frac{\mu_s}{\lambda^2}, \quad (3)$$

$$\mu_s = \frac{\lambda_j^0 \cosh\left(\frac{z-d}{\lambda}\right)}{2\sigma \sinh\left(\frac{d}{\lambda}\right)}, \quad (4)$$

where $\lambda \approx \sqrt{2\sigma\tau_s}$ is the spin diffusion length of the Py, and j_s^0 is the interfacial spin current density injected from the Cr_2O_3 into the Py which serves as the boundary condition for Eq. (3). We omit $\partial^2\mu_s/\partial x^2$ term because the length of the Py bar is orders of magnitude larger than its thickness, suppressing the spin diffusion effect along y direction [29].

With the open circuit boundary condition of j_c in the x direction and $\partial\mu_c/\partial x \approx -\theta_{\text{AH}}(P + \beta)\partial\mu_s/\partial z - (\Sigma_{\text{AN}}/\sigma)\partial T/\partial z$, the detected voltage along x averaged over the film thickness becomes

$$V = \frac{L\Sigma_{\text{AN}}}{\sigma d}\Delta T + \frac{L\lambda}{2\sigma d}\theta_{\text{AH}}(P + \beta)j_s^0 \tanh \frac{d}{2\lambda}, \quad (5)$$

where L and d are length and thickness of the Py bar, and ΔT is the temperature drop across the thickness of Py. While the ANE background signal is independent of Cr_2O_3 (except for interfacial spin pinning, as discussed above), the SAHE term depends on the interfacial spin injection j_s^0 and hence on the Néel vector in Cr_2O_3 .

The SF transition in Cr_2O_3 directly leads to a sign change of j_s^0 , because the reorientation of the Néel vector causes a reversal of the spin polarization of the lowest-energy magnon bands [25]. Following the formalism by Rietz *et al.* [27], we consider the spin pumping contributions by the Néel vector $\mathbf{n}$ and finite magnetization moment $\mathbf{m}$ as

$$\mathbf{j}_s^0 = g_n^{\uparrow\downarrow} \mathbf{n} \times \dot{\mathbf{n}} + g_m^{\uparrow\downarrow} \mathbf{m} \times \dot{\mathbf{m}}, \quad (6)$$

where $g_n^{\uparrow\downarrow}$ and $g_m^{\uparrow\downarrow}$ are the (real parts of) interfacial spin-mixing conductance for the Néel vector $\mathbf{n}$ and total magnetic moment $\mathbf{m}$, respectively. By integrating the magnon modes over the Brillouin zone, we can obtain the total spin current j_s^0 arising from the spin dynamics in Cr_2O_3 . Across the SF transition, j_s^0 experiences a sign change (see the Appendix), in agreement with experimental data in Fig. 3(c). Consequently, we calculate [Fig. 3(e)] the difference of spin injection across the SF transition, Δj_s^0 , which allows for comparison with the ΔV signal in Fig. 3(d).

IV. DISCUSSION

The distinct difference of the temperature dependence of thermally driven spin current in $\text{Cr}_2\text{O}_3/\text{Py}$ [Fig. 3(d)] and $\text{Cr}_2\text{O}_3/\text{Pt}$ [Fig. 1(f)] can be phenomenologically captured by the ratios of interfacial spin transmission coefficients $g_m^{\uparrow\downarrow}/g_n^{\uparrow\downarrow}$ associated with the vectors $\mathbf{m}$ and $\mathbf{n}$ [57,58]. We do not expect the theoretical curves, calculated in Fig. 3(e), to accurately reproduce the experimental data of Figs. 1(f) and 3(d), but they allow us to assess the differences between the AFM/NM and AFM/FM systems qualitatively. As shown in Fig. 3(e), $g_m^{\uparrow\downarrow}/g_n^{\uparrow\downarrow} = 20$ results in an increasing Δj_s^0 with increasing temperature, while $g_m^{\uparrow\downarrow}/g_n^{\uparrow\downarrow} = 1$ yields a decreasing Δj_s^0 . The general trend of Δj_s^0 under different $g_m^{\uparrow\downarrow}/g_n^{\uparrow\downarrow}$ clearly indicates that this quantity is larger in $\text{Cr}_2\text{O}_3/\text{Py}$ than in $\text{Cr}_2\text{O}_3/\text{Pt}$, especially at higher temperatures. A plausible mechanism underlying the enhanced value of $g_m^{\uparrow\downarrow}/g_n^{\uparrow\downarrow}$ is that magnonic excitations in Py, which are absent in Pt, play an important role. The majority electrons have a spin polarization opposite to that of the magnonic excitations in Py. Below the SF transition in Cr_2O_3 , the spin polarization of its lowest-energy magnon band matches that of the majority electrons but not

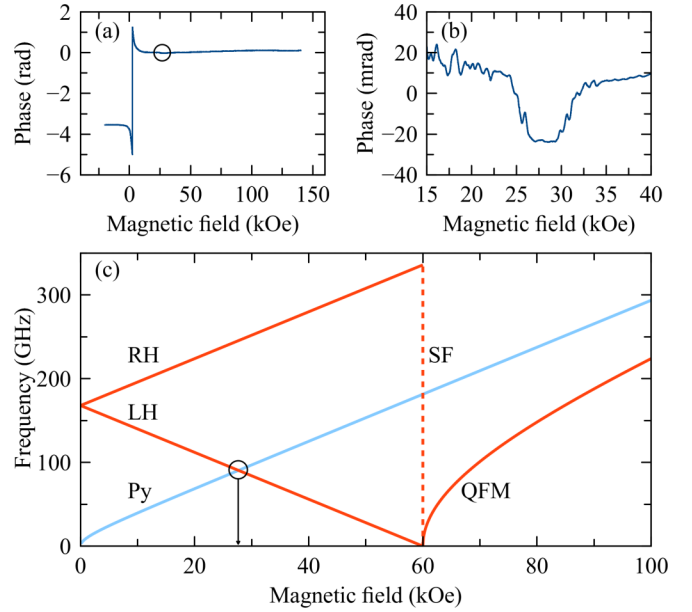


FIG. 4. (a), (b) Temporal phase of the thermal signal in $\text{Cr}_2\text{O}_3/\text{Py}$ reveals an anomaly at ~ 30 kOe. (c) Estimated frequencies of zero-wave-vector magnons in Cr_2O_3 and Py as a function of magnetic field. Left-hand (LH), right-hand (RH), spin-flop (SF), and quasiferromagnetic (QFM) modes are shown. Py and LH magnons cross at ~ 30 kOe.

the magnons in Py [25,28,59]. Therefore, the electrons in Py dominate the spin transmission processes while the magnon contribution is suppressed. Above the SF transition, on the other hand, Cr_2O_3 develops a net magnetization along the applied magnetic field. The magnons in Cr_2O_3 and in Py now have matching spin polarization, enabling the magnon-magnon spin transmission across the interface [40,41]. Once a magnon spin current in Py is generated, it drags the electron spin current, resulting in an effective enhancement of $g_m^{\uparrow\downarrow}$. This is in stark contrast to AFM/NM interfaces, where magnonic excitations in NM are absent [27].

This phenomenological explanation is further supported by an experimental observation that suggests a coupling between the magnonic subsystems of Cr_2O_3 and Py. In Figs. 4(a) and 4(b) for $\text{Cr}_2\text{O}_3/\text{Py}$, the temporal phase of the Py detector voltage $V(H)$ (i.e., the lock-in phase) in response to the periodic thermal drive of the heater is shown. The phase shows a clear π jump upon magnetization reversal of Py around zero field and remains nearly constant with increasing magnetic field. On top of this flat background, a small resonant [41,42,60,61] feature is conspicuous at the field of ~ 30 kOe. While this anomaly is only observed at high thermal drives indicating a nonlinear phenomenon, it is independently reproduced in the $\text{Cr}_2\text{O}_3/\text{Py}/\text{Pt}$ control sample [Fig. 5(b)]. Strikingly, the field at which the anomaly is observed corresponds to the crossing of the LH antiferromagnetic magnons of Cr_2O_3 and the ferromagnetic magnons of Py in the frequency-field diagram [Fig. 4(c)]. The observed anomaly points to a resonant (partial) hybridization [39] of the antiferromagnetic LH magnons of Cr_2O_3 and ferromagnetic magnons of Py, as such hybridization would modify the effective (combined via phonons and magnons) thermal

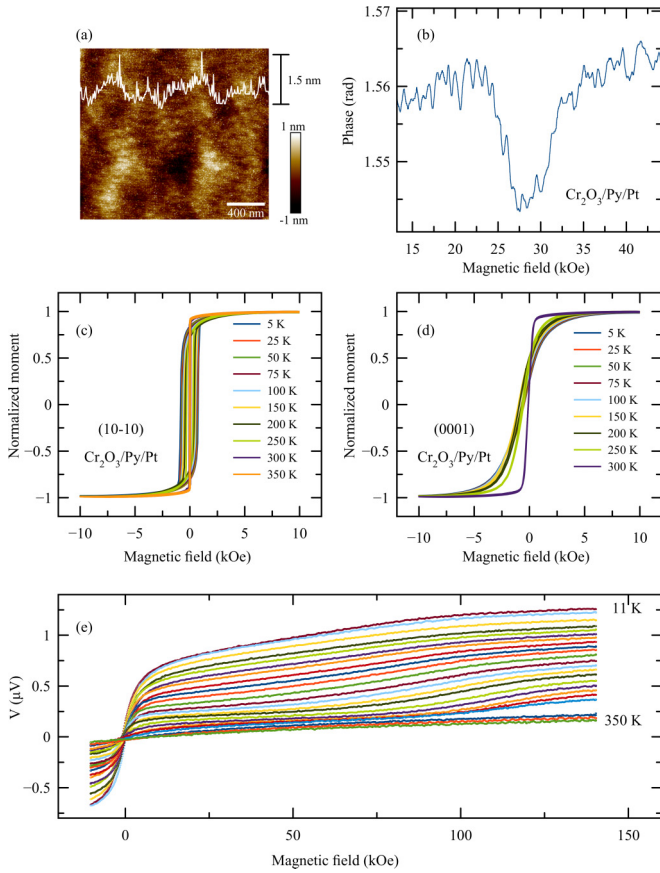


FIG. 5. (a) Atomic force micrograph of the Cr_2O_3 surface, including a line scan. (b) The phase of the thermal signal from the $\text{Cr}_2\text{O}_3/\text{Py}/\text{Pt}$ control sample shows a resonant feature near 30 kOe. (c,d) Magnetometry of $\text{Cr}_2\text{O}_3/\text{Py}/\text{Pt}$ along $[10\bar{1}0]$ and $[0001]$ directions. (e) Thermal signal from the $\text{Cr}_2\text{O}_3/\text{Py}/\text{Pt}$ control sample shows a similar behavior to that of the $\text{Cr}_2\text{O}_3/\text{Py}$ sample.

transport and thus lead to a phase shift of the the thermal wave [62] from Py into Cr_2O_3 . The frequency-field diagram [Fig. 4(c)] shows no crossing of the (zero-wave-vector) QFM magnons of Cr_2O_3 and the ferromagnetic magnons of Py, suggesting that the magnonic spin current above the SF transition is facilitated in a nonresonant manner. Recently, resonant and nonresonant magnon transduction at an interface of dissimilar ferromagnets has been observed [40,41]; our results suggest that analogous phenomena are possible at AFM/FM interfaces.

V. CONCLUSIONS

In summary, we have demonstrated thermally driven spin injection from a metallic ferromagnet into an insulating antiferromagnet. The interfacial spin current is converted into voltage signals via spin-orbit effects in Py. The spin injection in the AFM/FM system grows with increasing temperature and allows for sizable signals at room temperature. This is in stark contrast to AFM/NM systems where sizable spin transport has only been shown in the narrow range of cryogenic temperatures. Realization of robust spin transport near room temperature in thin film systems is an important step toward practical AFM-based spintronic devices,

in particular AFM-based spin-torque devices. Our theoretical model as well as experimental observation of coupling between the magnonic subsystems in the $\text{Cr}_2\text{O}_3/\text{Py}$ hybrid suggest that interfacial magnon transduction plays an important role in the spin transport. Our results motivate further studies to elucidate the microscopic mechanisms of the spin transport across AFM/FM interfaces, including magnon coupling and transduction. Our work opens avenues for spintronic and quantum information [63–65] applications based on hybrid ferromagnet/antiferromagnet systems and merges spin-orbitronic concepts of ferromagnetic metals with antiferromagnetic spintronics.

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APPENDIX

Material growth parameters. The Cr_2O_3 thin films were deposited on *m*-plane sapphire substrates by rf magnetron sputtering at 700 °C. The base pressure was 3×10^{-8} mbar and the working pressure of Ar gas was 5×10^{-3} mbar; nominal sputter rate was set to 1.72 nm/min. Prior to the deposition, the substrates were annealed at 750 °C for 10 min. Detector layers were magnetron sputtered at room temperature (dc for Py and rf for Pt) at Ar gas working pressure of 8×10^{-4} mbar. For oxidation-prone materials, a 5 nm sputtered Al_2O_3 cap layer was used.

Device fabrication. We fabricated SSE devices from the films using a combination of an optical lithography mask and negative photoresist for the detector layer. Once the $2.4 \times 0.25 \text{ mm}^2$ bar had been developed, the detector layer was ion milled down to Cr_2O_3 , leaving only the detector bar on top of the Cr_2O_3 layer. Using atomic layer deposition, an 80 nm electrically insulating layer of Al_2O_3 was deposited. Lastly, the heater layer was deposited in similar fashion to the detector except using positive photoresist and magnetron sputtering for the metallization.

Material characterization. The resistivity of the Py detector, sandwiched between Cr_2O_3 and Al_2O_3 , was measured with the two-point method (with wire bonds through the Al_2O_3 layer) and may be contaminated with contact resistance. The thus upper limit of the resistivity was obtained as $\sim 1 \mu\Omega \text{ m}$ which is 2–3 times larger than the literature values of pristine Py films [66] and in agreement with resistivity of Py grown under similar conditions on oxide substrates [67–69].

The atomic force microscopy of the Cr_2O_3 film surface is shown in Fig. 5(a).

Cross-sectional bright-field and high-resolution transmission electron microscopy (TEM) measurements were performed using an image- C_s -corrected Titan 80-300 microscope (FEI: Field Electron and Ion Company) operated at

an accelerating voltage of 300 kV. Prior to TEM analysis, the specimen was mounted in a double-tilt low-background holder and placed for 8 s into a Model 1020 Plasma Cleaner (Fischione) to remove possible contaminations. The cross-sectional TEM specimen was prepared by *in situ* lift-out using a Helios 5 CX focused ion beam (FIB) device (Thermo Fisher). To protect the sample surface, a carbon cap layer was deposited beginning with electron-beam-assisted and subsequently followed by Ga-FIB-assisted precursor decomposition. Afterwards, the TEM lamella was prepared using a 30 keV Ga FIB with adapted currents. Its transfer to a three-post copper lift-out grid (Omniprobe) was done with an EasyLift EX nanomanipulator (Thermo Fisher). To minimize sidewall damage, Ga ions with only 5 keV energy were used for final thinning of the TEM lamella to electron transparency.

The hysteresis curves of the $\text{Cr}_2\text{O}_3/\text{Py}/\text{Pt}$ show exchange bias for magnetic fields applied parallel to the c -axis [compare Figs. 5(c) and 5(d)].

Theoretical model. Equation (5) encompasses two contributions: The ANE (the second term) is a Py volume effect independent of Cr_2O_3 , which is responsible for the background signal. The SAHE component (the first term), on the other hand, depends on the interfacial spin injection j_s^0 and hence on the Néel vector in Cr_2O_3 . The SF transition in Cr_2O_3 directly leads to a sign change of j_s^0 , because the reorientation of the Néel vector causes a reversal of the spin polarization of the lowest-energy magnon bands [25], as shown in Fig. 6(a) calculated at 10 K using the formalism developed in Ref. [27]. This formalism becomes less accurate when approaching the Néel temperature, which is represented by the dashed shading in Fig. 3(e).

Thermal signal and control samples. We find the SSE signals of both $\text{Cr}_2\text{O}_3/\text{Py}$ and $\text{Cr}_2\text{O}_3/\text{Pt}$ to be anti-symmetric with respect to the polarity of the applied magnetic field, as shown in Figs. 6(c) and 6(d). The thermal signal was measured on a control sample with a Py detector layer deposited directly onto an m -plane sapphire substrate. The data in Fig. 6(b) show a steplike signal, indicative of Py magnetization reversal near zero field.

The misalignment procedure shown in Fig. 3(a) is time intensive and does not result in a quantitative determination of the SSE signal magnitudes, since the the ANE contribution itself is angle dependent and the misalignment results in partial but not full suppression of the SSE signal. We thus extract the magnitude difference of the thermal signal below and above the spin-flop transition from spectra with magnetic field fully aligned with the c -axis of Cr_2O_3 . This approach

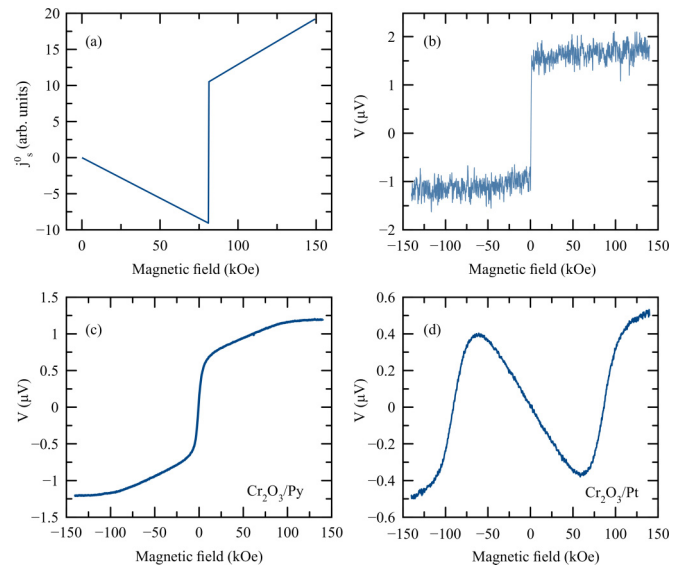


FIG. 6. (a) Calculated interfacial spin current at 10 K. (b) Thermal signal due to anomalous Nernst effect (ANE) from the $\text{Al}_2\text{O}_3/\text{Py}$ control sample at 10 K shows steplike reversal of magnetization. (c) SSE signals of the $\text{Cr}_2\text{O}_3/\text{Py}$ and (d) $\text{Cr}_2\text{O}_3/\text{Pt}$ sample are anti-symmetric with respect to the magnetic field polarity.

returns the qualitative behavior, conspicuous from the raw data in Figs. 2(a) and 5(e). The inaccuracy of this heuristic approach results in error bars due to finite width of the SF transition. The upper error bar is generally larger because the limited field range does not allow for extrapolating the behavior above the SF, especially at higher temperatures. The ANE background is not truly subtracted but, given the antagonical temperature dependences of the SSE and ANE contributions, the procedure allows us to assert that the magnitude for the maximum SSE signal in $\text{Cr}_2\text{O}_3/\text{Py}$ near room temperature is comparable to that in $\text{Cr}_2\text{O}_3/\text{Pt}$ at low temperature.

For extracting the spin-flop field of the $\text{Cr}_2\text{O}_3/\text{Py}$ sample, we smooth the raw data, calculate the derivative, and evaluate the position of the local maximum (i.e., the inflection point of the raw data) corresponding to the spin-flop field. The data are plotted in Fig. 2(c).

The thermal signal was measured on a control sample of $\text{Cr}_2\text{O}_3/\text{Py}/\text{Pt}$. The data in Fig. 5(e) show a behavior similar to that observed in the $\text{Cr}_2\text{O}_3/\text{Py}$ sample.

The phase of the thermal signal of the $\text{Cr}_2\text{O}_3/\text{Py}/\text{Pt}$ in Fig. 5(b) shows an anomaly at ~ 30 kOe similar to that of the $\text{Cr}_2\text{O}_3/\text{Py}$ sample.

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