Third harmonic characterization of antiferromagnetic heterostructures

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Electrical switching of antiferromagnets is an exciting recent development in spintronics, which promises active antiferromagnetic devices with high speed and low energy cost. In this emerging field, there is an active debate about the mechanisms of current-driven switching of antiferromagnets. For heavy-metal/ferromagnet systems, harmonic characterization is a powerful tool to quantify current-induced spin-orbit torques and spin Seebeck effect and elucidate current-induced switching. However, harmonic measurement of spin-orbit torques has never been verified in antiferromagnetic heterostructures. Here, we report harmonic measurements in Pt/α-Fe2O3 bilayers, which are explained by our modeling of higher-order harmonic voltages. As compared with ferromagnetic heterostructures where all current-induced effects appear in the second harmonic signals, the damping-like torque and thermally-induced magnetoelastic effect contributions in Pt/α-Fe2O3 emerge in the third harmonic voltage. Our results provide a new path to probe the current-induced magnetization dynamics in antiferromagnets, promoting the application of antiferromagnetic spintronic devices.

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**Antiferromagnetic (AFM) spintronics is an emerging research field with great potential for ultrafast, energy-efficient future technology**¹⁷. In the past several years, current-induced switching of AFM Néel order has been demonstrated in several antiferromagnetic materials, including metallic AFM CuMnAs and Mn₂Au as well as heavy-metal (HM)/AFM-insulator bilayers such as Pt/NiO and Pt/α-Fe₂O₃.⁸⁻¹⁵ These recent developments generate intense interests in active AFM devices. However, there is ongoing debate on the mechanism of the Néel order switching, which could be induced by spin-orbit torque (SOT) or the magnetoelastic effect as well as artifact signals from heavy metals and the relation to AFM grain morphology⁸⁻¹⁹.

Lock-in detection technique has been widely used to investigate current-induced spin torque contributions in HM/ferromagnetic (FM) systems by measuring the first and second harmonic voltages¹⁸⁻²⁰. For AFMs, the second harmonic measurement has been used for identifying 180° Néel vector reversals in CuMnAs.¹³ However, it requires that the AFM has both broken time and space inversion symmetry. Whether harmonic measurement can be used in characterizing the current induced effect in other AFMs is still an open question.²²⁻²⁸

In this article, we report harmonic measurements in HM/AFM bilayer Pt/α-Fe₂O₃. As compared to the HM/FM bilayers where spin torques only contribute to the second harmonic signals, our results shown that for HM/AFMs, the damping-like SOT, as well as the magnetoelastic effect, appear in the third harmonic response. Our theoretical modeling, together with the temperature-dependent harmonic measurements, indicate that the magnetoelastic effect could have an important contribution to current-induced AFM switching.

**Results**

α-Fe₂O₃ is an easy plane AFM at room temperature with the Néel order in ab-plane (0001). Due to the Dzyaloshinskii–Moriya interaction (DMI), there is a small in-plane canting of Néel order, which exhibits a very weak moment.⁷⁷ We grow epitaxial α-Fe₂O₃ films on Al₂O₃ (0001) substrate by off-axis sputtering.⁸⁻³⁰ X-ray diffraction scan (see Supplemental Materials) of a 30 nm α-Fe₂O₃ film shows Laue oscillations, demonstrating high crystal quality of the α-Fe₂O₃ film. Subsequently, we grow a 5 nm Pt layer on α-Fe₂O₃ by off-axis sputtering at room temperature. We pattern the Pt/α-Fe₂O₃ bilayers into a 5 μm wide Hall cross using photolithography and Ar ion etching, as schematically shown in Fig. 1a. For the harmonic measurement, we apply a 4 mA ac current I at 17 Hz and measure the first (1ω), second (2ω), and third (3ω) harmonic voltages using a lock-in amplifier.

**First harmonic Hall signals.** We first show the angular dependence of first harmonic voltage for a Pt/α-Fe₂O₃(30 nm) bilayer at a temperature (T) of 300 K in the presence of an in-plane magnetic field (H) from 0.3 to 14 T. Figure 1b schematically illustrates the two spin sublattices m₁ and m₂ of the in-plane magnetic field applied at an angle φ₁ relative to the x axis. We also define the unit vector of Néel order n = m₁ - m₂ and net magnetization m = m₁ + m₂, as shown in Fig. 1c. The orientations of these relevant vectors, m₁, m₂, n, and H are represented by their polar angle θ and azimuthal angle φ. Figure 1d shows the ϕ₁-dependence of first harmonic voltage V₁H, which is the same as the transverse spin Hall magnetoresistance (TSMR) in DC measurements (see Eq. S1 in Supplemental Materials for more details). Based on the theory of spin Hall magnetoresistance (SMR), when the current is applied along the x direction, the generated spin current with spin polarization σ is along the y direction. Depending on the relative angle between σ and n, the transverse voltage V₁H ∝ nₓnᵧ⁶⁰. For our α-Fe₂O₃ films, we showed previously⁸⁻²⁹ that the spin-flop transition occurs at the critical field of <1 T, where the Néel order is perpendicular to the magnetic field, nH. Then,

\[ V₁H = -V_{TSMR} \sin 2\phi₁. \]

Such TSMR has been demonstrated in many Pt/AFM bilayer systems³¹⁻³³. Fitting the angular-dependent V₁H curves in Fig. 1d with Eq. (1), we extract TSMR for each value of the magnetic field, which is plotted in Fig. 1e. The magnitude of V_{TSMR} saturates near μ₀H = 1 T, which is consistent with our previous results³¹, indicating single domain AFM state at μ₀H > 1 T. One notes that there is a small decrease of V_{TSMR} at high field. This is due to the tilting of the AFM spins at high field, which lowers the value of Néel vector n₃⁴.

**Second harmonic Hall signals.** In addition to the first harmonic signals, we simultaneously measure the second and third harmonic voltages. For the second harmonic voltage V₂H, our modeling (see Supplemental Materials for details) shows that it consists of two components, the field-like (FL) SOT and the spin Seebeck effect (SSE), which can be written as,

\[ V₂H = V₂^{FL} + V₂^{SSE} = V_{TSMR} \frac{H}{H_{eff}} \cos(2\phi₁)\cos(\phi₁) + V_{SSE}\cos(\phi₁). \]

where \(H_{eff}\) is the effective field of field-like torque and \(V_{SSE}\) is the SSE voltage. Figure 2a shows the in-plane angular dependent V₂H curves at different magnetic fields from 1 to 14 T. Each curve in Fig. 2a is fitted by Eq. (2), such as those shown in Fig. 2b for μ₀H = 5 T.
where $V_{3\omega}^{DL}$, $V_{3\omega}^{ME}$, and $V_{3\omega}^{AR}$ are the damping-like torque, magnetoelastic (ME) effect, and change of the resistivity ($\Delta R$) term, respectively. $H_{ex}$, $H_{DM}$, $H_{K}$, $H_{DL}$, and $H_{ME}$ are the exchange field, DMI effective field, easy-plane anisotropy field, damping-like torque effective field, and ME-induced effective easy-axis anisotropic field along $x$, respectively. $V_{3\omega}^{AR}$ mainly originates from the change of Pt resistivity due to the applied current. In previous reports of electrical switching of AFMs, thermally-induced Pt resistivity change has led to saw-tooth shaped artifact in switching signals^{8,10,16,37}. And there could be a very minor contribution to $V_{3\omega}^{AR}$ due to the heating induced softening of magnetization given the very high Néel temperature of $\alpha$-Fe$_2$O$_3$^{20}. Equation (3) reveals why damping-like torque and ME only appear in the third harmonic voltage as $H_{DL}^{\perp}$ and $H_{ME} \propto I^3$, whereas in FM, linear dependence on $H_{DL}$ appears in the second harmonic voltage.

Figure 3a shows the in-plane angular dependence of $V_{3\omega}$ at different magnetic fields, which is fitted by Eq. (3). Figure 3b, c shows the fitting of $V_{3\omega}$ for 0.3 and 10 T, respectively, with separate $\sin^2\phi_4$ and $\sin^4\phi_4$ components. At 0.3 T, the $V_{3\omega}^{DL}$ and $V_{3\omega}^{ME}$ contribution with a $\sin^4\phi_4$ dependence is comparable to the $V_{3\omega}^{AR}$ term with a $\sin^2\phi_4$ dependence. However, at 10 T, $V_{3\omega}^{AR}$ dominates the third harmonic voltage. Figure 3d shows $V_{3\omega}^{AR}$ as a function of the magnetic field and Fig. 3e shows $V_{3\omega}^{AR}$ normalized by $V_{TSMR}$, which is essentially field independent, indicating its nonmagnetic origin. Since $V_{3\omega}^{DL}$ and $V_{3\omega}^{ME}$ have the same angular dependence, Fig. 3f combines them as $V_{3\omega}^{DL+ME}$, which shows a quick decay as the field increases.

To better understand the contribution from $V_{3\omega}^{DL}$ and $V_{3\omega}^{ME}$, we make the same harmonic measurement at lower temperatures. For bulk $\alpha$-Fe$_2$O$_3$, when the temperature is lower than the Morin transition temperature $T_M \approx 260$ K, it experiences a spin reorientation transition, where the $\alpha$-Fe$_2$O$_3$ becomes an easy-axis AFM^{38}. However, for (0001)-orientated $\alpha$-Fe$_2$O$_3$ thin films, $T_M$ is much lower or even does not exist due to epitaxial strain^{39,40} as confirmed by the similar angular dependence in the DC and harmonic measurements. Thus, in our measured temperature range (100–300 K) the $\alpha$-Fe$_2$O$_3$ is still an easy-plane AFM. Figure 4a shows the normalized $V_{3\omega}^{DL+ME}$ by $V_{TSMR}$ at $T = 300$, 200, and 100 K, which is fitted by Eq. (3). We find that $V_{3\omega}^{DL+ME}$ decreases at lower temperatures and basically vanishes at 100 K. The effective anisotropic field of the magnetoelastic effect $H_{ME}$ is induced by thermoelastic stress $\Delta \sigma$^{41}. We use the finite-element simulation (see Supplementary Materials for more details) to estimate $\Delta \sigma$ in our Hall cross at the corresponding temperatures. Then we obtain $H_{ME} = \frac{2\lambda_1 \Delta \sigma}{M_s}$, where $\lambda_1 = 1.4 \times 10^{-6}$ is the magnetostrictive coefficient of $\alpha$-Fe$_2$O$_3$ and $M_s = 759$ emu/cm$^3$ is the sublattice magnetization^{43}.

Figure 4b shows the simulated $H_{ME}$ together with the fitted $H_{ME} - H_{ME}^{\text{eff}}$, where $H_{ME}^{\text{eff}} = \frac{H_{ME}^{2}}{H_{DL}^{2} + H_{ME}^{2} - H_{DL}^{2}}$, from Eq. (3) at different temperatures using $H_{ex} = 9 \times 10^6$ Oe and $H_{DM} = 1.78 \times 10^6$ Oe^{44,45}. From Fig. 4b, we can estimate the magnitude of $H_{ME}$ in our experiment is ~0.1 Oe at 300 K. The damping-like torque effective field, however, is challenging to quantify here since it has a quadratic dependence. In Fig. 4b, the simulated $H_{ME}$ is slightly larger than the values extracted from the experimental data and the difference is larger at higher temperatures. This could be due to the parameter choice or the contribution of $H_{DL}^{\text{eff}}$. If we believe the larger $H_{ME}$ is due to $H_{DL}^{\text{eff}}$ and assume the easy-plane anisotropic field $H_{K} \approx 100$ Oe^{46}, we can evaluate that $H_{DL}$ has the order of 1 Oe. One notes that this is an order of magnitude smaller than $H_{DL}$ which may be related to the insulating nature of $\alpha$-Fe$_2$O$_3$. It is known that FL(DL)-SOT is determined by the imaginary (real) part of spin mixing...
Further research in HM/AFM-insulator is needed to better understand the SOTs in AFM heterostructures.

**Discussion**

As harmonic measurements have been used in many FM materials, we show that they also serve as a powerful tool in investigating current-induced effects in HM/AFM systems. Usually, AFMs have very large magnetic anisotropies and remain in multiple-domain states even under a strong magnetic field. The multiple-domain state of AFMs hinders the quantitative analysis of current-induced magnetization change. Further research in HM/AFM-insulator is needed to better understand the SOTs in AFM heterostructures.

**Methods**

**Sample preparation.** Epitaxial α-Fe$_2$O$_3$ films are grown on Al$_2$O$_3$(0001) substrates using radio-frequency off-axis sputtering in a 12.5 mTorr sputtering gas of Ar + 5% O$_2$ at a substrate temperature of 500 °C. Pt/α-Fe$_2$O$_3$ bilayers, as well as Pt single
layers on Al₂O₃, are patterned into the Hall cross structure using photolithography and Argon ion milling for electrical measurements.

Harmonic measurement: The in-plane angular dependence measurements are performed using a Quantum Design 14 T Physical Property Measurement System (PPMS). An ac current J with an amplitude of 4 mA and frequency 17 Hz is applied by a Keithley 6221 current source while the harmonic voltage is measured by Stanford SR865A lock-in amplifier.

Data availability
All data generated in this study are presented in the paper and the Supplementary Information.

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Author contributions
Y.C., R.D.R., and J.J.M. fabricated the samples. Y.C. performed the harmonic measurements, analyzed the data, built the theoretical model, and drafted the manuscript. E.C. and N.N.S. contributed to the second harmonic experiment. F.Y. and A.D.K. supervised the project. All authors discussed the results and commented on the manuscript.

Competing interests
The authors declare no competing interests.
