

Spin Pumping of an Easy-Plane Antiferromagnet Enhanced by Dzyaloshinskii–Moriya Interaction

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Recently, antiferromagnets have received revived interest due to their significant potential for developing next-generation ultrafast magnetic storage. Here, we report dc spin pumping by the acoustic resonant mode in a canted easy-plane antiferromagnet $\alpha\text{-Fe}_2\text{O}_3$ enabled by the Dzyaloshinskii–Moriya interaction. Systematic angle and frequency-dependent measurements demonstrate that the observed spin-pumping signals arise from resonance-induced spin injection and inverse spin Hall effect in $\alpha\text{-Fe}_2\text{O}_3$ —metal heterostructures, mimicking the behavior of spin pumping in conventional ferromagnet–nonmagnet systems. The pure spin current nature is further corroborated by reversal of the polarity of spin-pumping signals when the spin detector is switched from platinum to tungsten which has an opposite sign of the spin Hall angle. Our results reveal the intriguing physics underlying the low-frequency spin dynamics and transport in canted easy-plane antiferromagnet-based heterostructures.

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Over the past decade, magnetic resonance-driven spin pumping has been demonstrated to be a powerful experimental technique for detecting pure spin currents in a range of magnet–nonmagnet heterostructures. Originally demonstrated in ferromagnetic metals and alloys [1–10], this measurement platform has been quickly extended to magnetic insulators with reduced energy dissipation [11–15], enabling the development of functional magnonic devices [16–18]. More recently, with the progress of high-frequency microwave technologies and emerging interest in antiferromagnetic (AFM) spintronics, subterahertz spin pumping has been demonstrated in antiferromagnets, such as Cr_2O_3 [19] and MnF_2 [20]. In contrast to (ferro) ferrimagnets, antiferromagnets promise to serve as a transformative platform enabling high-density memory, ultrafast data processing speeds, and robust strategies against malignant perturbations to develop next-generation spin-based information technologies [21–26]. Many of these advantages result from the ultrafast spin dynamics and vanishing net magnetic moment of the Néel order, which opens new opportunities for outperforming their ferromagnetic counterparts. A broad family of AFM materials have been intensively explored at the frontier of spintronic research [27–29].

Despite the growing availability of antiferromagnets with exceptional functionalities, AFM spin pumping remains a formidable challenge at the current state of the art. The major difficulty derives from the exchange-amplified magnon gaps in typical uniaxial antiferromagnets [19,20,30], which usually require (sub)terahertz frequency

microwave capabilities that are not available in most commercially available electron paramagnetic resonance instruments. On the other hand, easy-plane antiferromagnets, which have smaller magnon gaps and accessible resonant frequencies, are believed to be not applicable for spin pumping because the pertaining spin dynamics is linearly polarized—a fundamental distinction from the uniaxial antiferromagnets. In this Letter, we report robust gigahertz spin pumping from the canted magnetization in easy-plane antiferromagnet $\alpha\text{-Fe}_2\text{O}_3$ [31–34]. Theoretical analysis reveals the critical role of the Dzyaloshinskii–Moriya (DM) interaction which enhances the spin-pumping efficiency by orders of magnitude through modifying the ellipticity of the precessional motion of the canted magnetic moment. Our results shed light on the intriguing physics underlying the low-frequency spin dynamics and transport in canted easy-plane antiferromagnet-based heterostructures [35–41].

We first briefly review the pertinent magnetic properties of $\alpha\text{-Fe}_2\text{O}_3$. $\alpha\text{-Fe}_2\text{O}_3$ is an insulating antiferromagnet with a corundum crystal structure, as shown in Fig. 1(a). Above the Morin transition temperature, the Fe^{3+} magnetic moments stay in the (0001) plane and stack antiferromagnetically along the c axis. The magnetic moment of the two AFM spin sublattices is slightly canted by the DM interaction, leading to a weak ferromagnetic moment at equilibrium [31–34]. The DM interaction also drastically changes the spin dynamics of the acoustic (low-frequency) resonant mode, as illustrated in Fig. 1(b). Suppose that an external magnetic field H is applied along the y axis in the

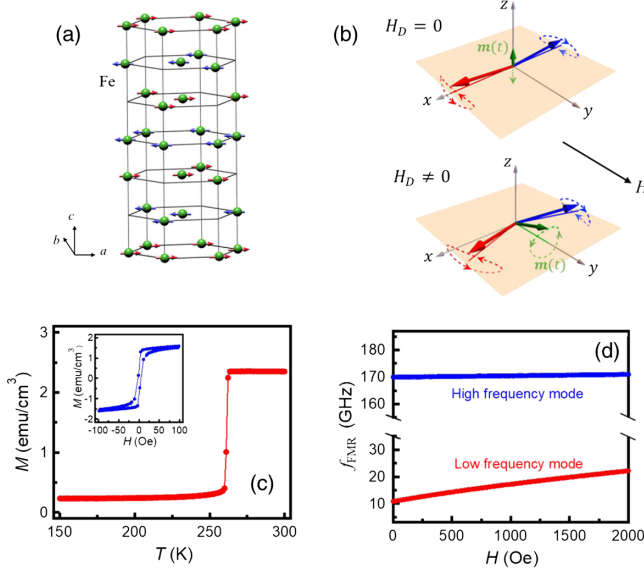


FIG. 1. (a) Schematic of the crystal structure of $\alpha\text{-Fe}_2\text{O}_3$ (oxygen atoms not shown). The symbols and arrows indicate the Fe atoms and the spins associated with them. (b) Schematic illustration of the acoustic resonant mode in an easy-plane antiferromagnet with and without the DM field H_D . The external magnetic field H is applied along the y axis. The solid green arrow which we labeled as $\mathbf{m}(t)$ is the instantaneous magnetic moment while the dashed green curve illustrates the trajectory of $\mathbf{m}(t)$. (c) Temperature dependence of the magnetization of the $\alpha\text{-Fe}_2\text{O}_3$ crystal with an external magnetic field of 1000 Oe applied in the (0001) plane. The inset shows field-dependent magnetization of $\alpha\text{-Fe}_2\text{O}_3$ when $T = 295$ K. (d) The expected resonant frequency f_{FMR} of the low- and high-frequency modes as a function of external magnetic field. H_D , $2H_E H_A$ and $2H_E H_{A'}$ are set to be 22 kOe, 15 kOe², and 3200 kOe² in the calculations [33,34].

magnetic easy plane of $\alpha\text{-Fe}_2\text{O}_3$. In the absence of the DM interaction, the two sublattice magnetic moments $\mathbf{m}_1$ and $\mathbf{m}_2$ precess elliptically in a right-handed chirality around their individual equilibrium directions, where the major (minor) axes of the trajectories are along the y (z) axis and angles have been exaggerated for visual clarity. Consequently, unless H becomes extremely strong to induce an appreciable canting of $\mathbf{m}_1$ and $\mathbf{m}_2$ along the y axis, the total magnetic moment $\mathbf{m} = \mathbf{m}_1 + \mathbf{m}_2$ will exhibit an almost linear oscillation along the z direction, as illustrated in the top panel of Fig. 1(b). Correspondingly, the oscillation of $\mathbf{m}(t)$ can only give rise to an ac spin pumping while the dc component is negligible. Moreover, an x -polarized microwave magnetic field is not able to drive the resonance of $\mathbf{m}(t)$ because of the polarization mismatch. This is a universal property of the conventional easy-plane antiferromagnets, such as NiO and MnO. When the DM field H_D is involved, however, $\mathbf{m}_1$ and $\mathbf{m}_2$ will be canted toward the y axis at equilibrium and precess elliptically around the canted directions, resulting in an elliptically circulating $\mathbf{m}(t)$ around the y axis, as illustrated

in the bottom panel of Fig. 1(b). Because the elliptical trajectory of the canted magnetic moment now has a finite projection along the x axis direction, the microwave magnetic field polarized along the Néel vector can effectively drive the acoustic resonant mode of $\alpha\text{-Fe}_2\text{O}_3$, hence a dc spin pumping. As H_D increases, the precession of $\mathbf{m}(t)$ becomes more circular, leading to a significantly enhanced dc spin-pumping efficiency in comparison with the conventional easy-plane antiferromagnets.

Our spin-pumping measurements use 0.5-mm-thick commercially available $\alpha\text{-Fe}_2\text{O}_3$ crystals (see Supplemental Material [42] Sec. 1). Figure 1(c) shows a temperature dependence of the magnetization of an $\alpha\text{-Fe}_2\text{O}_3$ crystal with an external magnetic field H of 1000 Oe applied in the (0001) plane. The measured magnetization curve exhibits a sharp, steplike variation around the Morin transition temperature $T_M \sim 263$ K. When $T < T_M$, $\alpha\text{-Fe}_2\text{O}_3$ becomes a uniaxial antiferromagnet with a negligible ferromagnetic moment. When $T > T_M$, a weak canted magnetic moment of ~ 2 emu/cm³ is observed [32–34], which is also confirmed by the field-dependent magnetization curve shown in the inset. At room temperature, two magnetic resonance modes coexist in $\alpha\text{-Fe}_2\text{O}_3$. When an external magnetic field H is applied in the (0001) plane, the low-frequency (acoustic) mode described above has an eigenfrequency [33,34]

$$\left(\frac{2\pi f_{\text{FMR}}}{\gamma}\right)^2 = H(H + H_D) + 2H_E H_A, \quad (1)$$

where γ is the gyromagnetic ratio, f_{FMR} is the resonant frequency, H_E is the exchange field, H_A is the effective field associated with the basal plane anisotropy, and H_D is the DM field. The high-frequency mode (also known as optical mode) corresponding to an out-of-plane oscillation of the Néel vector has a resonant frequency [33,34]

$$\left(\frac{2\pi f_{\text{FMR}}}{\gamma}\right)^2 = H_D(H + H_D) + 2H_E H_{A'}, \quad (2)$$

where $H_{A'}$ is the anisotropy field along the [0001] axis (the hard axis). Figure 1(d) plots the expected resonant frequency f_{FMR} of both modes as a function of H . In the magnetic field range of interest, f_{FMR} of the low-frequency one lies in the low-gigahertz frequency regime [33], which is accessible by the commercial microwave units. Our magnetic resonance and spin-pumping measurements presented below will focus on the branch of the low-frequency mode of $\alpha\text{-Fe}_2\text{O}_3$.

To detect the resonance-driven spin currents, we use magnetron sputtering to deposit platinum (Pt) or tungsten (W) films on top of the $\alpha\text{-Fe}_2\text{O}_3$ crystals (see Supplemental Material [42] Sec. 1). The spin-pumping measurements are performed using a microwave transmission line on the $\alpha\text{-Fe}_2\text{O}_3$ /metal samples at room temperature (see Supplemental Material [42] Sec. 2). Figure 2(a) shows the schematic of the sample structure. A static magnetic field H is applied in the magnetic easy-plane (x - y plane) and the

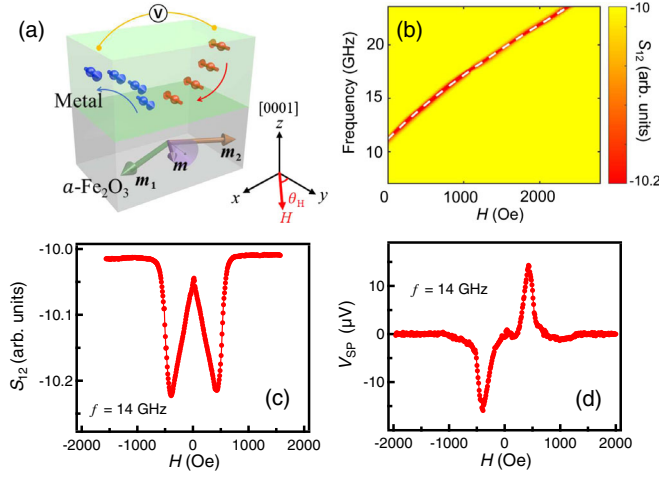


FIG. 2. (a) Schematic of dynamic spin injection and inverse spin Hall effect in an $\alpha\text{-Fe}_2\text{O}_3$ /metal heterostructure. $\mathbf{m}_1$, $\mathbf{m}_2$, and $\mathbf{m}$ represent the magnetic moment of the two spin sublattices and the canted ferromagnetic moment of $\alpha\text{-Fe}_2\text{O}_3$, respectively. (b) Color-coded spin wave transmission spectrum S_{12} measured as a function of external magnetic field H and microwave frequency f . The white dashed line represents the fitting curve from which H_D and $H_E H_A$ are extracted. (c) S_{12} vs H spectrum of $\alpha\text{-Fe}_2\text{O}_3$ measured at $f = 14$ GHz and an input microwave power $P_{rf} = 1$ mW. (d) V_{SP} vs H spectrum measured at $f = 14$ GHz, $\theta_H = 0^\circ$, and $P_{rf} = 1000$ mW.

spin-pumping signal V_{SP} is measured across the long axis (x axis) of the sample. θ_H is defined as the relative angle between the in-plane magnetic field H and the y axis. At resonance condition, an oscillating Oersted field (linearly polarized along the x axis) produced by microwave currents flowing a proximal transmission line compensates the Gilbert damping of $\alpha\text{-Fe}_2\text{O}_3$, then the canted magnetization can freely oscillate around the equilibrium position, transferring spin angular momenta to the conduction electrons of the neighboring metal layers through interfacial exchange coupling. The injected spin current J_s is converted to a charge current: $J_c \propto \theta_{SH} J_s \sigma \cos \theta_H$ by the inverse spin Hall effect (ISHE) [1–4], where θ_{SH} is the spin Hall angle of the metal layer and σ is the spin polarization parallel to the external magnetic field.

Figure 2(b) shows the normalized spin wave transmission spectrum measured as a function of magnetic field H and microwave frequency f . At resonance condition, a significant amount of microwave power is absorbed by the $\alpha\text{-Fe}_2\text{O}_3$ crystal, leading to a reduced microwave transmission efficiency characterized by S_{12} . The dispersion curve of the observed spin wave mode shown in Fig. 2(b) agrees well with the theoretical prediction of the acoustic mode of $\alpha\text{-Fe}_2\text{O}_3$, by which H_D and $2H_E H_A$ are fitted to be $(2.07 \pm 0.041) \times 10^4$ Oe and $(1.59 \pm 0.029) \times 10^7$ Oe², quantitatively agreeing with the values reported in previous studies [33]. Figures 2(c) and 2(d) show the field-dependent magnetic resonance and spin-pumping spectra of an

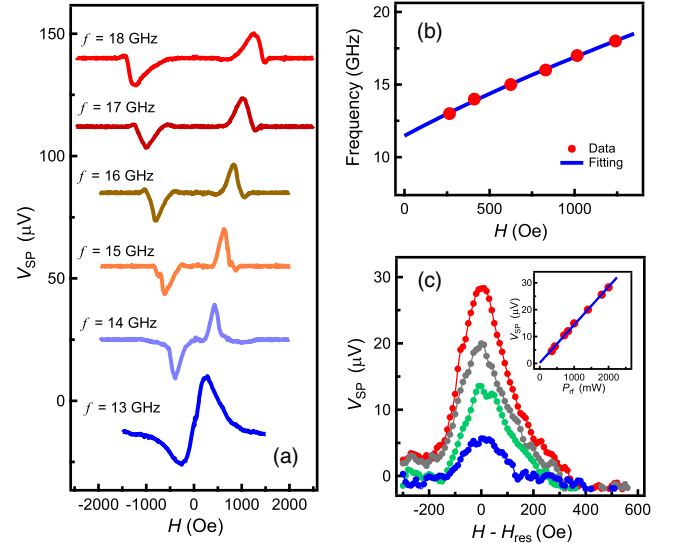


FIG. 3. (a) Spin-pumping spectra of the $\alpha\text{-Fe}_2\text{O}_3$ /Pt sample measured at different frequencies with $\theta_H = 0^\circ$. The curves are offset for clarity. (b) Peak position of the measured spin-pumping signals as a function of the microwave frequency. A theoretical fitting (blue curve) using Eq. (1) agrees well with the experimental results (red points). (c) V_{SP} vs $(H - H_{res})$ spectra of the $\alpha\text{-Fe}_2\text{O}_3$ /Pt sample measured with input microwave power of 440, 1000, 1400, and 2000 mW (blue, green, black, and red curves, respectively) at $f = 14$ GHz and $\theta_H = 0^\circ$. Inset: microwave power dependence of the measured spin-pumping signals. The blue line is a linear fitting of the experimental data.

$\alpha\text{-Fe}_2\text{O}_3$ /Pt (5 nm) sample measured at $f = 14$ GHz and $\theta_H = 0^\circ$. For spin-pumping measurements, the input microwave power P_{rf} of 1000 mW is modulated at a frequency of 161 Hz. To obtain a better signal-to-noise ratio, a preamplifier is used to amplify the electrical voltage signals with a gain of 500 (spin-pumping data shown below were taken after amplification unless stated otherwise). It is clear that the electrical signals are observed at the magnetic fields satisfying resonance conditions. In addition, the measured spin-pumping signals change sign when the external magnetic field is reversed from $\theta_H = 0^\circ$ to 180° , which is consistent with the theoretical model of ISHE. Figure 3(a) shows a series of spin-pumping spectra of the $\alpha\text{-Fe}_2\text{O}_3$ /Pt sample measured from 13 to 18 GHz with $\theta_H = 0^\circ$. At all frequencies, the observed voltage signals show the expected sign reversal when the polarity of the external magnetic field changes. The variation of the absolute magnitude of the spin-pumping signals results from the frequency-dependent microwave transmission efficiency. Figure 3(b) plots the magnetic field H with the maximum spin-pumping signals as a function of the microwave frequency, which essentially follows the acoustic resonance as described by Eq. (1). Figure 3(c) shows the spin-pumping spectra of the $\alpha\text{-Fe}_2\text{O}_3$ /Pt sample measured at microwave powers of 440, 1000, 1400, and 2000 mW with a frequency of 14 GHz. All the spectra have been

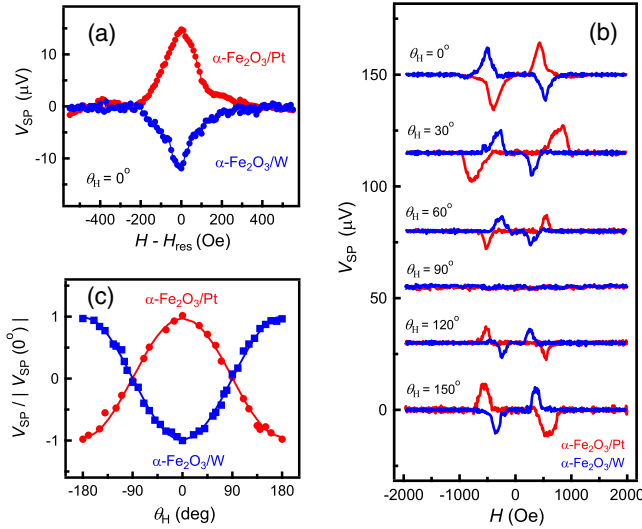


FIG. 4. (a) V_{SP} vs $(H - H_{res})$ spectra of the $\alpha\text{-Fe}_2\text{O}_3/\text{Pt}$ sample (red curve) and the $\alpha\text{-Fe}_2\text{O}_3/\text{W}$ sample (blue curve) measured at $f = 14$ GHz and $\theta_H = 0^\circ$. (b) Spin-pumping spectra of the $\alpha\text{-Fe}_2\text{O}_3/\text{Pt}$ (red curves) and $\alpha\text{-Fe}_2\text{O}_3/\text{W}$ (blue curves) measured at a different in-plane field angle θ_H . The curves are offset for clarity. (c) Angular dependence of $V_{SP}/|V_{SP}(0^\circ)|$ for $\alpha\text{-Fe}_2\text{O}_3/\text{Pt}$ and $\alpha\text{-Fe}_2\text{O}_3/\text{W}$. The red and blue curves show $\cos\theta_H$ and $-\cos\theta_H$, respectively.

offset by the magnetic resonance field H_{res} for clarity. The inset shows the linear microwave power dependence of V_{SP} measured at $\theta_H = 0^\circ$.

To further confirm that the measured electrical voltages indeed result from ISHE, we employ tungsten (W) which has a negative spin Hall angle as an alternative spin detector [46]. Figure 4(a) shows the V_{SP} vs H spectra for two $\alpha\text{-Fe}_2\text{O}_3/\text{Pt}$ and $\alpha\text{-Fe}_2\text{O}_3/\text{W}$ samples measured at $P_{rf} = 1000$ mW and $\theta_H = 0^\circ$. Notably, a robust voltage signal of $11.9 \mu V$ is observed in the $\alpha\text{-Fe}_2\text{O}_3/\text{W}$ sample, showing the opposite sign in comparison with the $\alpha\text{-Fe}_2\text{O}_3/\text{Pt}$ sample. When H is rotated in the magnetic easy plane of $\alpha\text{-Fe}_2\text{O}_3$, the canted magnetic moment remains essentially parallel to H at all angles since the resonance field H_{res} exceeds the basal plane anisotropy field. Figure 4(b) shows a series of V_{SP} vs H spectra for varying in-plane field angle θ_H . When H is rotated from 0° to 90° , V_{SP} observed in both samples gradually vanishes and approaches zero. Figure 4(c) summarizes the angular dependence of V_{SP} for both $\alpha\text{-Fe}_2\text{O}_3/\text{Pt}$ and $\alpha\text{-Fe}_2\text{O}_3/\text{W}$ samples normalized by the maximum magnitude of spin-pumping signals measured at $\theta_H = 0^\circ$ (see Supplemental Material [42] Sec. 2). The clear cosinusoidal shape corroborates that the observed electrical voltages indeed arise from ISHE.

Next, we develop a theoretical model to quantitatively analyze the observed spin pumping in $\alpha\text{-Fe}_2\text{O}_3/\text{metal}$ bilayers. According to the device geometry depicted in Fig. 2(a), a dc spin current density I_s polarized along the y axis can be generated by an x -polarized microwave magnetic field h_{rf} as follows (see Supplemental Material [42] Sec. 3):

$$I_s = \hbar g_r \left\langle \mathbf{m} \times \frac{d\mathbf{m}}{dt} \right\rangle = \hbar g_r f \text{Im}[\chi_{xx}^* \chi_{zx}] h_{rf}^2, \quad (3)$$

where $\hbar$ is the Planck constant, f is the driving microwave frequency, χ_{xx} and χ_{zx} are two components of the dynamical susceptibility such that $m_i(t) = \text{Re}[\chi_{ij} h_{rf,j} e^{i2\pi f t}]$, and g_r is the interfacial spin-mixing conductance [47,48]. In uniaxial antiferromagnets with circular polarization, the factor $\text{Im}[\chi_{xx}^* \chi_{zx}]$ is proportional to $[H_A/(H - H_{res})^2 H_E]$ on resonance [24,49]. For easy-plane antiferromagnets with DM interaction, however, this factor becomes complicated due to the elliptical precession, so we resort to numerical calculations. Owing to the ISHE and the spin diffusion effect in the heavy metal, I_s obtained from Eq. (3) converts into a spin-pumping signal V_{SP} [50]:

$$V_{SP} = \xi \tilde{g}_r \frac{L \theta_{SH} \rho e}{2\pi} \frac{\lambda}{d_N} \tanh\left(\frac{d_N}{2\lambda}\right) h_{rf}^2, \quad (4)$$

where e is the electron charge, L , θ_{SH} , λ , ρ , and d_N are the length, the spin Hall angle, the spin diffusion length, the resistivity, and the thickness of the heavy-metal layer, respectively. The factor $\tilde{g}_r = \{g_r/[1 + (2\lambda e^2 \rho g_r/h) \coth(d_N/\lambda)]\}$ is the effective spin-mixing conductance incorporating the spin backflow effect in the heavy metal [51]. Here, we have defined $\xi = 2\pi f \text{Im}[\chi_{xx}^* \chi_{zx}]$ as the spin-pumping susceptibility to characterize the conversion efficiency of microwave power into dc spin current density. ξ sensitively depends on the polarization status of $\mathbf{m}(t)$, which is determined by the DM field H_D , the driving frequency f , and the external magnetic field H . We calculate ξ for $f = 14$ GHz and other parameters extracted from experiments [52], where the Gilbert damping $\alpha = 4.6 \times 10^{-4}$ is obtained by comparing the measured linewidth with the numerical linewidth (see Supplemental Material [42] Sec 4). Normalized to γ/H_E , Fig. 5 plots ξ as a function of H for different strengths of H_D with $H_D^{\text{Max}} = 2.07 \times 10^4$ Oe being the actual value we measured. Notably, the DM interaction significantly enlarges the peak value ξ_p along with a reduction of the resonance field H_{res} . As plotted in the insets, when H_D increases from zero to its actual value H_D^{Max} , the resonance field is reduced by 7 times while the peak value ξ_p increases by almost 350 times. This drastic enhancement confirms the essential role of DM interaction underlying the observed dc spin-pumping signals. Using $\theta_{SH} = 0.1(-0.14)$ [14], $t_N = 5$ nm, $\lambda = 7.3(2.1)$ nm [14], and $\rho = 3.3 \times 10^{-7}(2.5 \times 10^{-6}) \Omega \text{m}$ for the Pt (W) layer, and other relevant material parameters: $L = 4$ mm, $d_N = 5$ nm, and $h_{rf} = 0.2$ Oe at a frequency of 14 GHz (see Supplemental Material [42] Sec. 5), the effective spin-mixing conductance $\tilde{g}_r$ is estimated to be $7.1 \times 10^{20} \text{ m}^{-2}$ and $7.4 \times 10^{19} \text{ m}^{-2}$ for the $\alpha\text{-Fe}_2\text{O}_3/\text{Pt}$ and $\alpha\text{-Fe}_2\text{O}_3/\text{W}$ interfaces, respectively. These values are 2 orders of magnitude larger than those reported in (anti)ferromagnet-heavy-metal systems [1,5,7,13,14,20,53]. A recent study indicated that when the

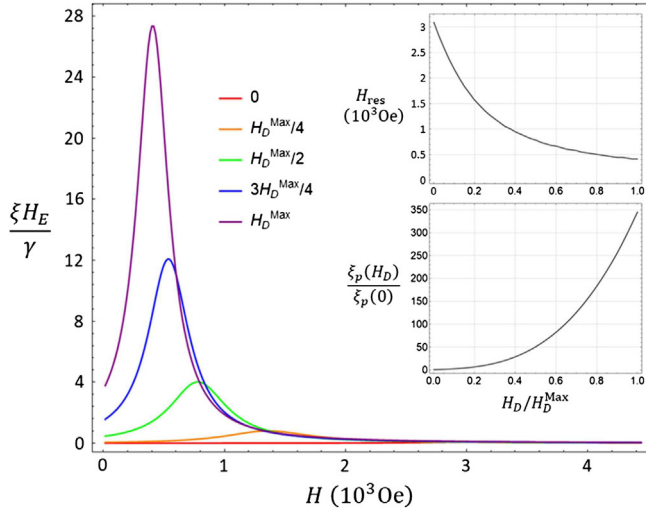


FIG. 5. Calculated spin-pumping susceptibility ξ (scaled by γ/H_E) as a function of the external magnetic field H for different DM fields, where $H_D^{\text{Max}} = 2.07 \times 10^4$ Oe is the actual value we measured. Insets: resonant field H_{res} and resonant peak ξ_p (normalized to its value at $H_D = 0$) as functions of H_D/H_D^{Max} . Parameters for the calculation are chosen from experiments [52] and the Gilbert damping coefficient $\alpha = 4.6 \times 10^{-4}$ is obtained by comparing the measured linewidth with the numerical values (see Supplemental Material [42] Sec. 4).

Néel vector contribution is suppressed, the spin-mixing conductance characterizing the spin pumping from $\mathbf{m}(t)$ could indeed be much larger than that for the Néel vector [54], which is consistent with our results. However, it remains an open question as to why the spin-mixing conductance becomes significantly enhanced when the contribution of Néel vector precession vanishes. A possible reconciliation is that if we use the unit vector $\hat{\mathbf{m}}$, rather than $\mathbf{m}$ itself, to define spin pumping in Eq. (3), then the value of $\tilde{g}_r$ will shrink by a factor of $(H_D/H_E)^2$, yielding $5.5 \times 10^{17} \text{ m}^{-2}$ and $5.8 \times 10^{16} \text{ m}^{-2}$ for the $\alpha\text{-Fe}_2\text{O}_3/\text{Pt}$ and $\alpha\text{-Fe}_2\text{O}_3/\text{W}$ interfaces, respectively.

In conclusion, we have demonstrated robust gigahertz-frequency spin pumping of a canted easy-plane antiferromagnet $\alpha\text{-Fe}_2\text{O}_3$. By modifying the ellipticity of the resonant motion of the canted magnetic moment, the DM interaction could significantly enhance the spin-pumping efficiency by orders of magnitude in spite of the detrimental role of hard-axis anisotropy, leading to robust inverse spin Hall signals measured in adjacent heavy-metal layers. Our results suggest the exciting possibility of harnessing hard-axis antiferromagnets in developing next-generation functional magnetic devices.

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Note added.—Recently, a related paper by Boventer *et al.*, appeared [55]. They have also observed dc spin-pumping signals in $\alpha\text{-Fe}_2\text{O}_3/\text{metal}$ bilayers.

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