Efficient terahertz emission from R-plane α-Fe$_2$O$_3$/Pt

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We report on the generation of ultrafast spin currents through femtosecond laser excitation of epitaxial bilayer thin films comprising α-Fe$_2$O$_3$ (an insulating antiferromagnet) and Pt (a heavy metal). The epitaxial thin films of α-Fe$_2$O$_3$ were simultaneously grown in three distinct orientations, namely, R, A, and C planes, employing pulsed laser deposition. By using the R plane α-Fe$_2$O$_3$, we demonstrate a substantial enhancement of the THz signal, nearly one order of magnitude greater compared to the A and C planes α-Fe$_2$O$_3$/Pt. We perform a detailed investigation into the sample azimuthal and laser polarization dependence of the THz emission. Our investigations establish that the sample azimuthal dependence of THz emission in the α-Fe$_2$O$_3$/Pt system is linked to the spin domain distribution as well as the crystalline symmetry of the orientation of α-Fe$_2$O$_3$. These results contribute significantly to understanding the ultrafast dynamics of spin currents in antiferromagnets.

I. INTRODUCTION

Antiferromagnets (AFMs) have recently attracted significant attention for the development of the next generation of spintronic devices [1–4]. In an AFM, the two coupled sublattices with spontaneous magnetic moments in opposite directions create the net zero magnetic moment. This inherent property provides notable advantages, such as the lack of stray fields and remarkable stability to external magnetic fields. These qualities make AFMs promising for high-density spintronic memory applications [5]. Another remarkable advantage of AFMs is their extremely high intrinsic frequency (THz) of antiferromagnetic dynamics, offering the potential to elevate the processing speed of memory elements from gigahertz to terahertz regimes [1,6].

Another emerging phenomenon of antiferromagnetic materials in spintronics involves the generation of terahertz (THz) emission by using laser-induced ultrafast spin current. Such a process of spintronic-based THz emission was first discovered in ferromagnet/heavy metal heterostructures [7–9]. In such structures, an ultrafast spin current generated in the ferromagnet is converted into charge current through the inverse spin Hall effect in the heavy metal layer. The resulting ultrafast transient charge current emits electromagnetic radiation in the THz frequency range [10–15]. Recently, this phenomenon was observed in antiferromagnetic/heavy metal heterostructures [16–19]. While it was first observed in NiO/Pt [16], it has also been recently demonstrated in α-Fe$_2$O$_3$/Pt [18].

Similar to NiO, α-Fe$_2$O$_3$ or hematite is also an insulating antiferromagnet at room temperature. α-Fe$_2$O$_3$ is antiferromagnetic between the Morin transition temperature of $\approx$260 K [20,21] and Néel temperature of 950 K. Furthermore, α-Fe$_2$O$_3$ possesses a low Gilbert damping constant and a large spin diffusion length [22–25]. Recently, the spin pumping from the acoustic resonant mode in α-Fe$_2$O$_3$ was also reported [26]. These reports indicated that α-Fe$_2$O$_3$ is a highly promising material for various spintronics-based devices [21,27–30].

The recent study of THz emission on α-Fe$_2$O$_3$/Pt was performed on (0001)-oriented α-Fe$_2$O$_3$ films grown on C plane Al$_2$O$_3$ substrate. [18] To date, no studies of THz emission have been performed on other orientations of α-Fe$_2$O$_3$ films. Previous studies have indicated that the R plane of α-Fe$_2$O$_3$ is highly promising for efficient generation of spin-orbit torque [31] as well as for long-range spin transport [23]. Consequently, investigating THz emission in other low-index directions, specifically the A and R plane α-Fe$_2$O$_3$, is particularly interesting.

In this work, we report on zero field THz emission from α-Fe$_2$O$_3$/Pt thin films for three distinct orientations of α-Fe$_2$O$_3$, namely, the C plane, A plane, and R plane. First, we demonstrate the growth of high-quality epitaxial thin films of the C plane, A plane, and R plane α-Fe$_2$O$_3$ on the sapphire substrate. Then, we systematically studied THz emission from α-Fe$_2$O$_3$/Pt having different orientations as a function of sample azimuth and laser polarization. We find a substantial enhancement of the THz signal in the R-plane α-Fe$_2$O$_3$, nearly one order of magnitude greater compared to the C plane and A plane α-Fe$_2$O$_3$/Pt. We show that the symmetry of the sample azimuthal dependence of THz emission in our α-Fe$_2$O$_3$/Pt system is related to the spin domain distribution, as well as the crystalline symmetry.

α-Fe$_2$O$_3$ has a rhombohedral corundum crystal structure with Fe^{+3} ions on the basal plane with AFM coupling between adjacent planes. The atomic arrangement in the basal plane is hexagonal due to which several studies have shown epitaxial

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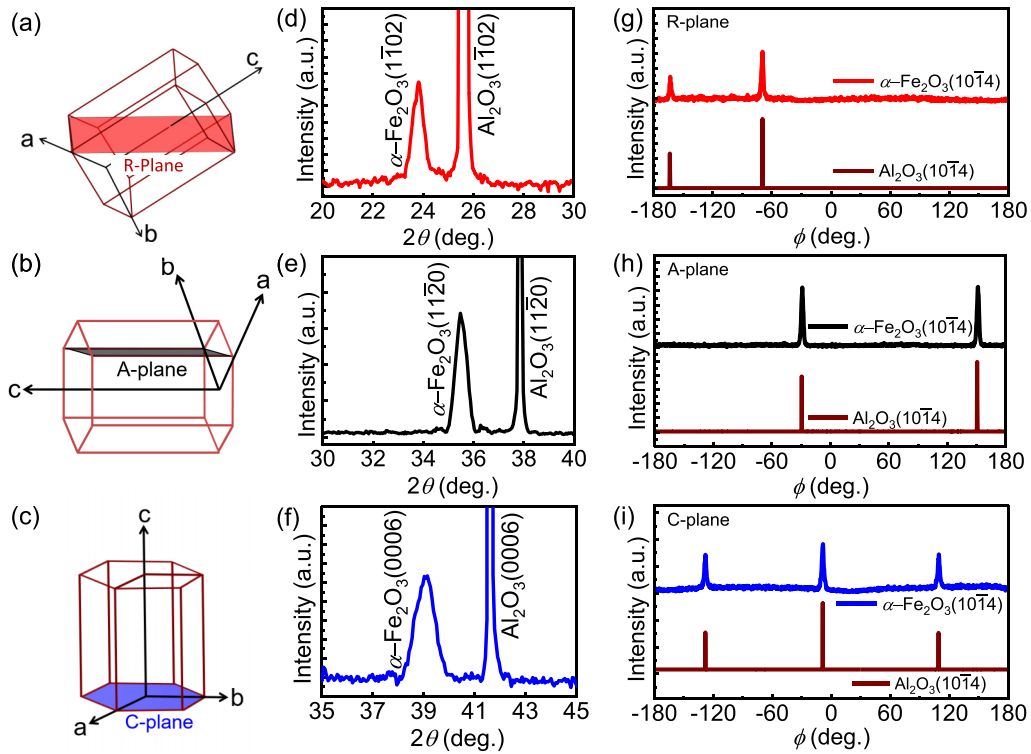‡muduli@physics.iitd.ac.in

FIG. 1. Unit cell of α-Fe$_2$O$_3$ showing (a) R plane, (b) A plane, and (c) C plane. The XRD θ-2θ scan of (d) R plane, (e) A plane, and (f) C plane α-Fe$_2$O$_3$ thin films. The ϕ scan data of (10$\bar{1}$4) reflection of both α-Fe$_2$O$_3$ and Al$_2$O$_3$ for (g) R pane, (h) A plane, and (i) C plane. For clarity, different intensity scales are utilized for Al$_2$O$_3$ and α-Fe$_2$O$_3$ in figures (g)–(i).

growth of α-Fe$_2$O$_3$ on hexagonal Al$_2$O$_3$ [32,33] despite the lattice mismatch of 5.8% between Al$_2$O$_3$ and α-Fe$_2$O$_3$. We employ the pulsed laser deposition method to *simultaneously* grow epitaxial thin films of α-Fe$_2$O$_3$ on (0001)-oriented (C plane), (11$\bar{2}$0)-oriented (A plane), and (1$\bar{1}$02)-oriented (R plane) Al$_2$O$_3$ substrates, respectively. The three orientations of the α-Fe$_2$O$_3$ are shown schematically in Figs. 1(a) to 1(c).

II. EXPERIMENTAL DETAILS

KrF excimer pulsed laser source with a wavelength of 248 nm, laser energy density of 1.1 J/cm^2, and a repetition rate of 10 Hz is employed for the deposition of α-Fe$_2$O$_3$ thin films. The substrate-to-target distance was set at 5.6 cm throughout the deposition and the Fe$_2$O$_3$ target was rotated continuously at 5 rpm for uniform growth. We find the growth rate of α-Fe$_2$O$_3$ thin films to be 0.028 Å per laser shot. The base pressure of the chamber was better than 2×10^{-6} Torr. During the deposition, oxygen partial pressure of 3.75×10^{-3} Torr was maintained. The best quality of films was obtained for the substrate temperature of 550 °C. After the growth of α-Fe$_2$O$_3$, the sample was moved to a different ultrahigh vacuum (AJA Orion 8) sputtering system to grow the Pt layer. The base pressure of the sputtering chamber was better than 4×10^{-8} Torr. The α-Fe$_2$O$_3$ film was exposed to air during the sample transfer from the pulsed laser deposition system to the sputtering system. Therefore, before the Pt layer deposition, the α-Fe$_2$O$_3$ film surface was cleaned by removing 3 nm-α-Fe$_2$O$_3$ (at etch rate of 0.125 Å/s) using RF substrate bias.

Subsequently, a 3-nm-thick Pt layer was deposited at room temperature with a working pressure of 2×10^{-3} Torr. The growth rate of the Pt layer was 0.4 Å/s.

High-resolution x-ray diffraction (HRXRD) was employed to characterize the crystallographic properties of α-Fe$_2$O$_3$ thin films. The epitaxial relationship of α-Fe$_2$O$_3$ was determined through ϕ-scan analysis. The film thicknesses were determined using x-ray reflectivity (XRR). These measurements were performed using a PANalytical x-ray diffractometer, utilizing Cu-K_α radiation with a wavelength of 1.54 Å. The Raman measurement was performed using a Renishaw inVia confocal microscope with a 532-nm-laser wavelength and 2400 lines per mm grating at 10-mW power. We conducted THz emission measurements on α-Fe$_2$O$_3$/Pt samples using a time-domain THz spectroscopy experimental setup [34]. The samples were excited by a femtosecond laser at normal incidence. Optical excitation was achieved using the output from a Ti:sapphire regenerative amplifier, providing approximately 35 femtosecond pulses at a 1-kHz repetition rate and centered around 800 nm. The laser output was split into two parts using a beam splitter, with a stronger portion serving as the optical pump for excitation and a weaker portion as the probe for detecting the THz pulses. A pair of parabolic mirrors directed the emitted THz radiation to a 1-mm-thick [110]-oriented ZnTe nonlinear crystal, which was utilized for detecting the THz pulses. All the measurements are performed without any magnetic field. THz-pulse detection employed the electro-optic sampling technique, where the probe beam was collinearly overlapped with THz pulses on the ZnTe crystal. A computer-controlled linear translational stage controlled the

delay between them. The THz pulse-induced birefringence in the probe was detected using a balanced photodetector, lock-in amplifier, quarter-wave plate, and Wollaston prism. The differential signal obtained was directly proportional to the THz electric field. All measurements were conducted under ambient conditions of temperature and humidity.

III. RESULTS AND DISCUSSION

Figures 1(d) to 1(f) illustrates the HRXRD θ-2θ scan for 20-nm-thick R, A, and C planes α-Fe$_2$O$_3$ thin films. On the R-plane Al$_2$O$_3$ substrate, Bragg peaks corresponding to the reflection from (1$\bar{1}$02)-planes of Fe$_2$O$_3$ were observed at 23.75°, as depicted in Fig. 1(d). When grown on the A-plane Al$_2$O$_3$ substrate, the Fe$_2$O$_3$(11$\bar{2}$0) reflection was positioned at 35.27°, as shown in Fig. 1(e). On the C-plane Al$_2$O$_3$ substrate, Bragg peaks corresponding to the reflection from (0006) planes of Fe$_2$O$_3$ was observed at 38.93°, as illustrated in Fig. 1(f). The long-range HRXRD θ-2θ scan is shown in the Appendix information (Appendix A). The absence of other peaks indicates the epitaxial growth of the Fe$_2$O$_3$ film on C-, A-, and R-plane Al$_2$O$_3$ substrates.

To further confirm the epitaxial relationship, we performed ϕ scan measurements. We used (10$\bar{1}$4) reflection of Al$_2$O$_3$ ($2\theta = 35.148°$) and α-Fe$_2$O$_3$ ($2\theta = 33.15°$) for all three cases R, A, and C planes α-Fe$_2$O$_3$. The sample surface was tilted by an angle, χ for the (10$\bar{1}$4) reflection, which was 47°, 57.56°, and 38.2° for the case of R, A, and C planes, respectively. In the case of R-plane α-Fe$_2$O$_3$ thin films, the ϕ scan exhibits two peaks with a 90° interval were observed for both the substrate Al$_2$O$_3$ and α-Fe$_2$O$_3$ as depicted in Fig. 1(g). In the case of A-plane α-Fe$_2$O$_3$ thin film, the ϕ scan exhibits two peaks with a 180° interval for both the substrate Al$_2$O$_3$ and the thin film α-Fe$_2$O$_3$ as depicted in Fig. 1(h). Finally in the case of C-plane α-Fe$_2$O$_3$ thin film, the ϕ scan exhibits three peaks with a 120° interval for both the substrate Al$_2$O$_3$ and the thin film α-Fe$_2$O$_3$ as depicted in Fig. 1(i). These results indicate that the in-plane directions of α-Fe$_2$O$_3$ and Al$_2$O$_3$ are parallel to each other without any twinning or rotation, which is similar to previous studies [32,35].

Raman measurements were employed to confirm further the crystalline quality and phase of the grown α-Fe$_2$O$_3$ thin films. Figure 2 presents the Raman measurement for 20-nm-thick R, A, and C planes α-Fe$_2$O$_3$ films. Seven phonon modes, including $5E_g$ and $2A_g$, were observed, corresponding to 247, 294, 379, 416, and 573 cm^{-1} for $5E_g$ modes and 226 and 495 cm^{-1} for $2A_g$ modes, respectively. These findings confirm the corundum structure of the grown α-Fe$_2$O$_3$ thin film [36–38]. The presence of an infrared (IR) active mode around 665 cm^{-1} in all the three orientations of α-Fe$_2$O$_3$ indicates the presence of disorder in the film. This peak is normally observed in thin films of α-Fe$_2$O$_3$. [39] The mode observed around 740 cm^{-1} arises from the Al$_2$O$_3$ substrate [40]. A weak and broad vibrational mode at around 834 cm^{-1} is assigned to a magnon excitation [39], confirming the anti-ferromagnetic nature of α-Fe$_2$O$_3$ films. The signature of weak two-magnon mode at around 1550 cm^{-1} is also observed. Finally, a broad peak around 1320 cm^{-1} is assigned to the $2E_u$(LO) mode, which originates from two-phonon scattering and is known to be resonantly enhanced [39]. We also observe

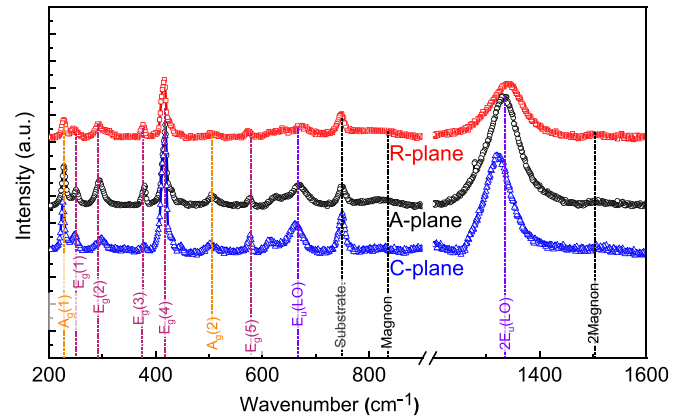FIG. 2. Raman spectra obtained using 532-nm-laser excitation of α-Fe$_2$O$_3$/Pt heterostructure grown in R, A, and C planes.

a small shift of the $2E_u$(LO) mode in the C-plane sample towards the lower wave number, which is likely due to the stretching of the Fe-O bond, as previously reported in Fe$_2$O$_3$ by using combined Raman and x-ray absorption near-edge structure measurements [36]. The bond stretching in the C plane may be due to a slightly different state of strain compared to other orientations.

The surface roughness and morphology were determined by atomic force microscopy (AFM), which are shown in Appendix A. The surface roughness of the α-Fe$_2$O$_3$ films with R, A, and C planes are comparable (<0.4 nm), which is expected as the films are grown under identical conditions. The magnetization measurements performed on R, A, and C planes α-Fe$_2$O$_3$ films were found to be dominated by the diamagnetic contribution of the substrate with no noticeable difference in magnetization for the three orientations as shown in Appendix B. The behavior is consistent with the antiferromagnetic nature of the α-Fe$_2$O$_3$ films.

Figure 3(b) shows the example of the detected THz emission signal performed on the R, A, and C planes $\alpha - Fe_2O_3$/Pt heterostructure. The coordinate system (xyz) is defined in the laboratory frame as shown in Fig. 3(a). The laser light is incident along the y-axis. For the linear polarization of the laser, the polarization direction, θ_P, is defined w.r.t the x-axis. The sample azimuth, θ_S is also defined w.r.t the x-axis. The THz emission spectra in Fig. 3 are recorded at θ_S, which shows maximum THz amplitude for $\theta_P = 0°$. The amplitude of the THz emission signal is found to depend strongly on the orientation of α-Fe$_2$O$_3$ thin films. The THz signal in the R-plane α-Fe$_2$O$_3$/Pt system exhibits an order of magnitude higher amplitude compared to the C- and A-plane α-Fe$_2$O$_3$/Pt. A comparison of the amplitude with the reference Fe/Pt is shown in the Appendix C. The fast Fourier transformation (FFT) of these signals is shown in the inset of Fig. 3, which shows that the bandwidth of generated THz emission is similar (∼2 THz) for all three cases. Note that our measurement setup bandwidth is about 3 THz. Measurements done on the bare α-Fe$_2$O$_3$ do not show any THz signals, as shown in the Appendix D. Hence, the THz signal originates primarily from the bilayer α-Fe$_2$O$_3$/Pt via the spin-to-charge conversion (SCC), which is similar to the recent report by Qiu et al. [18].
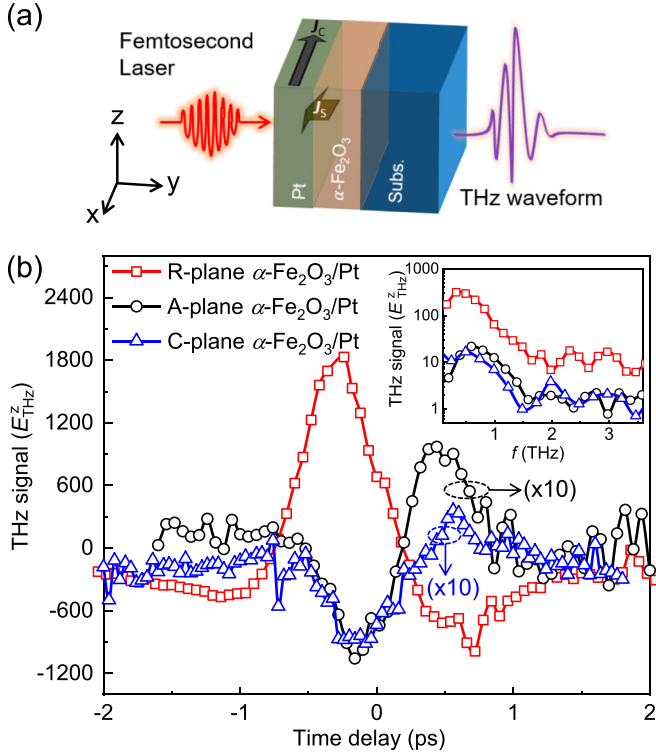
FIG. 3. (a) Schematic of the transmission-type THz spectroscopy setup. The xyz coordinate system is defined in the laboratory's frame. The sample is placed in the x-z plane, while the laser is incident along the y-direction. (b) THz signal, E^z_{THz} from R, A, and C planes α-Fe$_2$O$_3$/Pt heterostructure grown on Al$_2$O$_3$ substrates. The inset displays the corresponding Fourier spectra of the THz signal.

To understand the origin of THz generation in α-Fe$_2$O$_3$/Pt system, we performed a detailed study of THz emission as a function of sample azimuth (first column) and laser polarization (second column) as shown in Fig. 4. The sample azimuthal-dependent THz emission measurements are taken for $\theta_P = 0°$. In the R-plane α-Fe$_2$O$_3$/Pt, the sample azimuthal dependence of THz amplitude displays one-fold symmetry [Fig. 4(a)], while the corresponding XRD ϕ-scan data [Fig. 1(g)] exhibits a two-fold symmetry. Consequently, factors other than crystal symmetry influence the sample azimuthal dependence of the THz amplitude for the R-plane α-Fe$_2$O$_3$/Pt. However, in the A-plane α-Fe$_2$O$_3$/Pt, the azimuthal dependence of THz amplitude exhibits two-fold symmetry [Fig. 4(b)], which is consistent with the XRD ϕ-scan data [Fig. 1(h)], which also exhibits a two-fold symmetry. Finally, for the C-plane α-Fe$_2$O$_3$/Pt, the sample azimuthal dependence of THz amplitude displays one-fold symmetry [Fig. 4(c)], while the corresponding XRD ϕ-scan data [Fig. 1(i)] exhibits a three-fold symmetry. Hence, factors other than crystal symmetry are also found to influence the sample azimuthal dependence of the THz amplitude for the C-plane α-Fe$_2$O$_3$/Pt. This result appears to contradict the results of Qiu *et al.* [18], which was performed only for the C-plane α-Fe$_2$O$_3$/Pt. The sample azimuthal dependence of THz amplitude in the work by Qiu *et al.* exhibited three-fold symmetry, in agreement with the crystal symmetry of C-plane α-Fe$_2$O$_3$.

The second column of Fig. 4 shows the behavior of THz amplitude with laser polarization angle, θ_P. For the R-plane α-Fe$_2$O$_3$ the measurement is performed at $\theta_S = 0°$ [Fig. 4(d)]. However, for A- and C-planes α-Fe$_2$O$_3$/Pt, these measurements are performed for $\theta_S = 180°$, and $270°$, respectively [Figs. 4(e) and 4(f)]. These values of θ_S were chosen to maximize the signal to noise as the THz amplitude was found to be very weak for A- and C-planes α-Fe$_2$O$_3$/Pt thin films. The THz signal can be effectively fitted with $P_0 + P_1 \cos 2(\theta_P)$ terms for all three orientations. In the case of R- and A-planes α-Fe$_2$O$_3$/Pt the polarization modulation (P_1/P_0) of THz amplitude is about 10%, whereas it is about 30% for the C-plane α-Fe$_2$O$_3$/Pt. The tunability of THz amplitude in the α-Fe$_2$O$_3$/Pt heterostructure with pump polarization differs from spintronic THz emitter based on ferromagnets and heavy metal [7–9], but is similar to that of AFM/HM systems such as NiO/Pt [16,17,41] and α-Fe$_2$O$_3$/Pt [18].

Now, we will discuss the mechanism of orientation-dependent THz emission in α-Fe$_2$O$_3$/Pt system. In the case of AFM/HM system, the ultrafast spin current can be produced through both incoherent and coherent mechanisms. The incoherent mechanism includes the pyrospintronic effect (PSE) [7,42,43] and ultrafast spin Seebeck effect (SSE) [8,42,44], while the coherent mechanism includes impulsive stimulated Raman scattering (ISRS). In the incoherent processes, PSE and SSE, the ultrafast spin current is produced by heating-induced spin-voltage and a temperature gradient across the AFM/HM interface, respectively. These methods require a nonzero net magnetization within the magnetic layer. In our case, all the measurements are performed at zero magnetic field. We also did not observe any significant difference in the magnetization of R-, A-, and C-planes α-Fe$_2$O$_3$/Pt, which can explain the observed difference in THz peak amplitude for the three orientations. Hence, we neglect the incoherent driving forces as a possible mechanism to explain the observed behavior.

On the other hand, the coherent ISRS mechanism does not require a nonzero net magnetization and has been applied to explain the THz emission from both NiO/Pt [16] and α-Fe$_2$O$_3$/Pt [18] systems. The ISRS process induces an impulsive magnetization $\Delta M(t)$ in the α-Fe$_2$O$_3$ layer, which produces an ultrafast antiferromagnetic spin pumping to the adjacent Pt layer. Consequently, the injected ultrafast spin current in the Pt layer is converted to THz pulses via the inverse spin Hall effect. A signature of the ISRS process [45,46] is the dependence of THz amplitude on the laser polarization, which we observe in Fig. 4. The THz emission in our geometry due to the ISRS origin can be phenomenologically described as [16]

$$E^z_{THz} \propto A_0 + A_1 \cos(\theta_S - \delta) + A_2 \cos(2\theta_S - \delta), \quad (1)$$

where coefficients A_0, A_1, and A_2 are functions of nonlinear susceptibility tensor, $\chi^{(2)MEE}_{i'j'k'}$ and a ratio η_n, which represents the contribution from the nth domain. The nonlinear susceptibility tensor $\chi^{(2)MEE}_{i'j'k'}$ describes the relationship between AC magnetization and the laser electric field, which is parametrically generated by simulated Raman-type nonlinear process [16,47–49]. Here, $i'j'k'$ refers to the spin coordinate system of the α-Fe$_2$O$_3$ lattice. We assume that our samples consist of

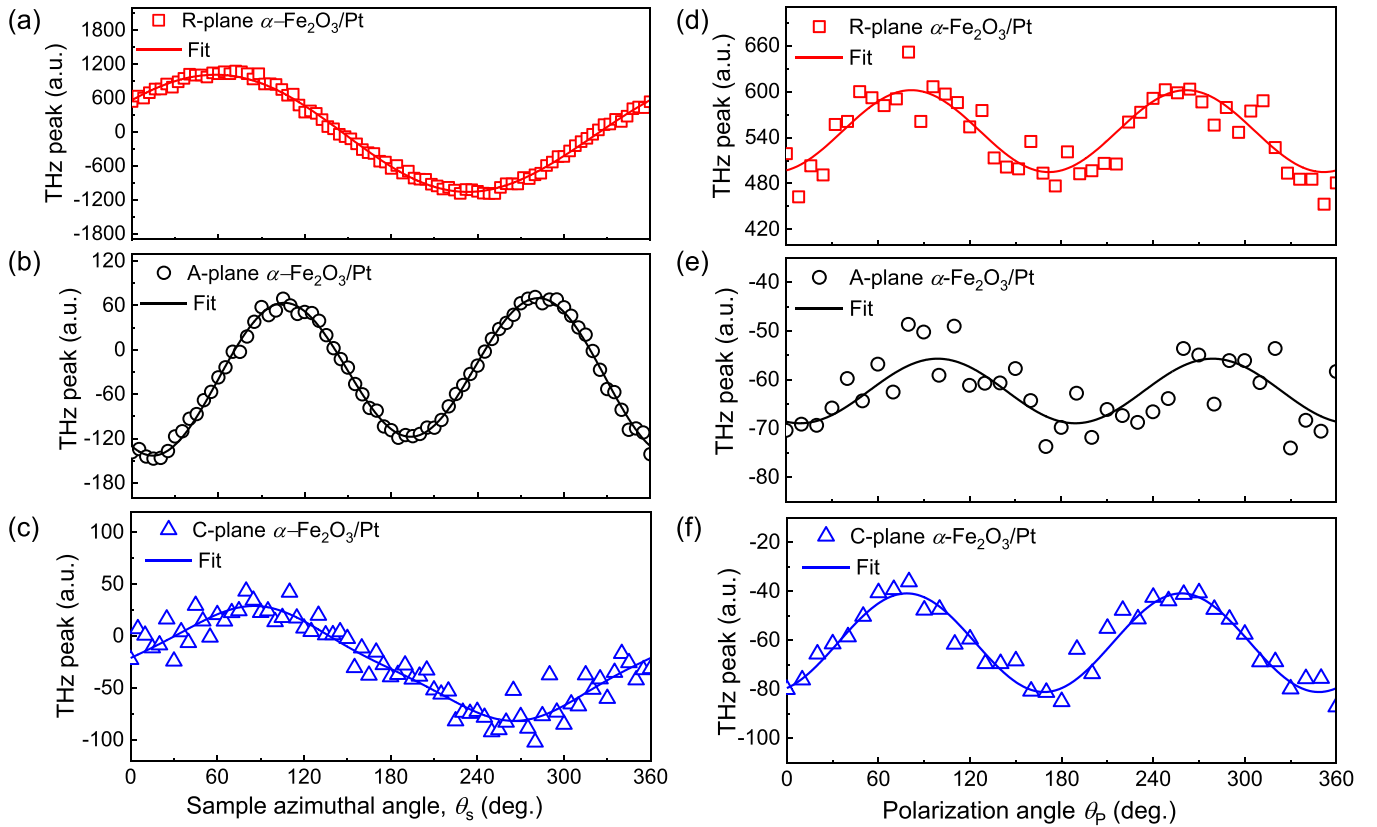FIG. 4. Sample azimuthal angle (θ_S) dependent THz emission in α-Fe$_2$O$_3$ heterostructures in three different orientations: (a) R-plane α-Fe$_2$O$_3$/Pt, where $\theta_S = 0°$ corresponds to the [1$\bar{1}$00] axis of the R plane of the Al$_2$O$_3$ substrate; (b) A plane α-Fe$_2$O$_3$/Pt, where $\theta_S = 0°$ corresponds to the [0001] axis of the A plane of the Al$_2$O$_3$ substrate; (c) C plane α-Fe$_2$O$_3$/Pt, where $\theta_S = 0°$ corresponds to the [1$\bar{1}$00] axis of the C plane of the Al$_2$O$_3$ substrate. Polarization angle (θ_P) dependent THz emission in thin film of (d) R-plane α-Fe$_2$O$_3$/Pt at constant $\theta_S = 0°$, (e) A-plane α-Fe$_2$O$_3$/Pt at constant $\theta_S = 180°$, and (f) C-plane α-Fe$_2$O$_3$/Pt at constant $\theta_S = 270°$.

randomly distributed domains. The domain sizes of α-Fe$_2$O$_3$ are typically of the order of 1 μm, [50] which is significantly smaller than the size of the laser spot, which is about 1.5 mm. Finally, the angle δ accommodates offset arising from the experimental setup. We fit the sample azimuthal dependence with the above equation as shown by the solid lines in Fig. 4. The term A_1 (one-fold) are found to be the most dominant term for R and C planes, while both the terms A_1 (one-fold) and A_2 (two-fold) are found essential for the fitting of sample azimuthal dependence of THz peak for A-plane α-Fe$_2$O$_3$/Pt. Furthermore, the value of A_1 is found to be 1035 ± 4 for the R plane, which is about 20 times higher than that of the C-plane ($A_1 = -52 \pm 2$).

To explain this behavior, we invoke the crystalline and magnetic symmetry. According to Eq. (1), the amplitude of THz emission depends on the summation of spin currents generated in each magnetic domain via the ISRS mechanism. Hence, the distribution of spin domains can significantly influence both the amplitude and the sample azimuthal dependence of THz emission. In α-Fe$_2$O$_3$, the easy axes are along all $\langle 1100 \rangle$ axes, which changes the three-fold crystalline symmetry of the C plane and R plane, while it does not affect the two-fold symmetry of A-plane α-Fe$_2$O$_3$. [18] This explains why the sample azimuthal dependence of THz peak for A-plane α-Fe$_2$O$_3$/Pt still exhibits two-fold symmetry consistent with the XRD ϕ-scan data [Fig. 1(h)]. The spin do-

main distribution in the C-plane and R-plane α-Fe$_2$O$_3$ changes the symmetry of the sample azimuthal dependence of THz emission. Qiu *et al.* [18] showed a three-fold symmetry of the sample azimuthal dependence of THz emission in the C plane, while we observed a one-fold symmetry. We attribute this difference to different spin domain distributions in our samples.

Finally, we discuss the possible origin of the large THz emission observed in R-plane α-Fe$_2$O$_3$/Pt. We believe the mechanism is related to a more efficient generation of ultrafast spin current by laser-induced transient magnetization. The ISRS process involves impulsive magnetization $\Delta M(t)$ in the α-Fe$_2$O$_3$ layer, giving spin pumping on a subpicosecond time scale. Recent studies have shown that spin-orbit torque is more efficient in switching the Néel vector in R-plane α-Fe$_2$O$_3$/Pt for which the easy plane is tilted [31]. The R-plane α-Fe$_2$O$_3$ has a tilt angle of about 58° with the easy plane (C plane). Hence, the laser-induced effective fields can have components in the easy plane containing two sublattice magnetic moments m_A and m_B. This can rotate the two sublattices more constructively, leading to the highly efficient spin current generation in R-plane α-Fe$_2$O$_3$. Antiferromagnetic spin pumping experiments as a function of orientation of α-Fe$_2$O$_3$ such as those used by Vaidya *et al.* [51] and Li *et al.* [52], as well as spin-torque-driven antiferromagnetic resonance experiments reported recently by Zhou *et al.* [53] can help to confirm our hypothesis.
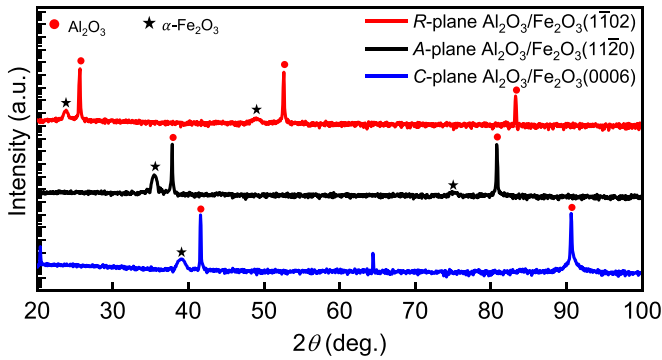
FIG. 5. Long range θ-2θ x-ray diffraction scan of R, A, and C plane α-Fe$_2$O$_3$ thin films grown on R-, A-, and C-plane Al$_2$O$_3$ substrates, respectively. The black star and red dot are used to indicate the Bragg peaks corresponds to α-Fe$_2$O$_3$ film and Al$_2$O$_3$ substrate, respectively.

We note that laser-induced terahertz emission in antiferromagnetic/nonmagnetic structures is an emerging phenomenon. To fully understand the mechanism of the efficient THz emission from the R-plane α-Fe$_2$O$_3$/Pt system, more comprehensive theoretical studies, particularly as a function of the orientation of α-Fe$_2$O$_3$, are necessary. In this regard, theoretical studies similar to the recent work on off-resonant THz spin pumping by Sun *et al.* [54] could be beneficial.

IV. CONCLUSION

In conclusion, we demonstrated that THz amplitude and polarity can be tuned by adjusting the sample azimuthal orientation and the orientation of α-Fe$_2$O$_3$ thin films without needing an external magnetic field. We show an order of magnitude improvement in THz emission from epitaxial thin films of R-plane α-Fe$_2$O$_3$/Pt as compared to the identical films grown with C- and A-plane orientations. Furthermore, we show that the orientation of α-Fe$_2$O$_3$ films also affects the symmetry of sample azimuthal dependence of THz peak amplitude, which is determined by both crystalline symmetry and magnetic symmetry of the orientations. Our work shows that the R-plane α-Fe$_2$O$_3$ is highly promising for THz spintronics. The results obtained from this study are highly promising and have tremendous potential for the advancement of spintronics-based THz devices as well as the field of antiferromagnetic spintronics.

APPENDIX A: STRUCTURAL CHARACTERIZATION OF α-Fe$_2$O$_3$/Pt HETEROSTRUCTURE

1. X-ray diffraction

Figure 5 illustrates the long-range XRD θ-2θ scan for the 20-nm-thick R-, A-, and C-planes α-Fe$_2$O$_3$ thin films. On the R-plane Al$_2$O$_3$ substrate, Bragg peaks corresponding to the reflection from (1$\bar{1}$02) and (2$\bar{2}$04) planes of α-Fe$_2$O$_3$ were observed at 23.75° and 48.94°, respectively. When grown on the A-plane Al$_2$O$_3$ substrate, the Fe$_2$O$_3$(11$\bar{2}$0) reflection and (22$\bar{4}$0) reflection were observed at 35.27° and 75.03°, respectively. On the C-plane Al$_2$O$_3$ substrate, Bragg peaks corresponding to the reflection from (0006) and (0012) planes of Fe$_2$O$_3$ were observed at 38.93° and 83.87°, respectively. The absence of peaks from other orientations indicates the
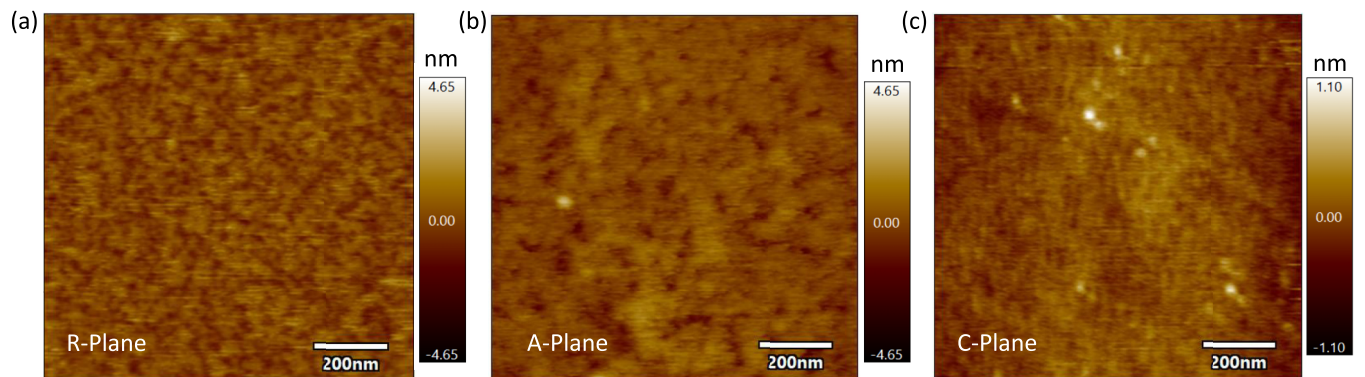

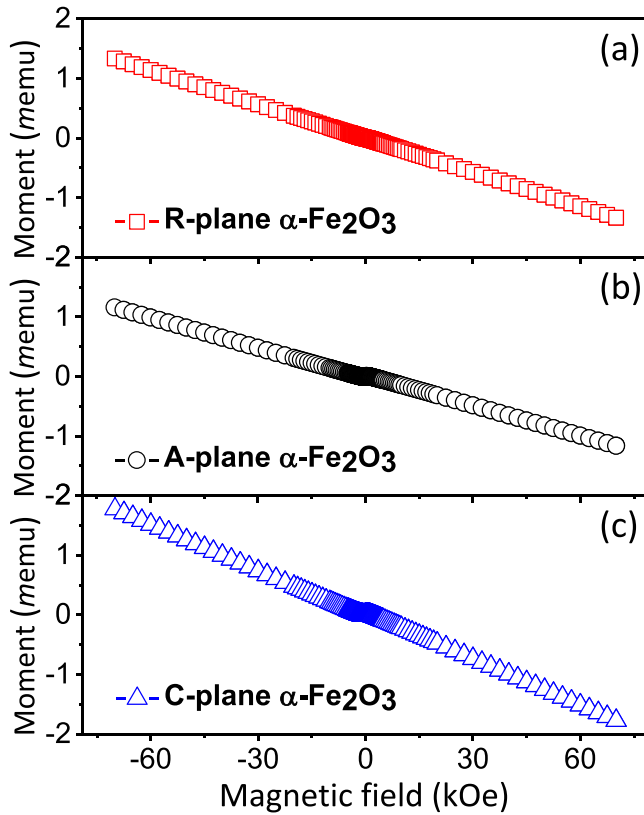FIG. 6. Atomic force microscopy images of (a) R-, (b) A-, and (c) C-plane-oriented α-Fe$_2$O$_3$ thin films.

FIG. 7. Magnetization measurement in thin film of α-Fe$_2$O$_3$ grown in (a) R-, (b) A-, and (c) C-plane orientation using quantum design PPMS system.

epitaxial growth of the Fe$_2$O$_3$ film on R-, A-, and C-plane Al$_2$O$_3$ substrates.

2. Atomic force microscopy

Figure 6 represents the topography of α-Fe$_2$O$_3$ thin films with a scan area of $1\,\mu m \times 1\,\mu m$. All the measurements were performed at room temperature in tapping mode using Asylum research, MFP-3D system. The value of root-mean-square (rms) roughness of R-, A-, and C-plane α-Fe$_2$O$_3$ films were found to be 0.36 nm, 0.34 nm, and 0.11 nm, respectively. The value of rms roughness for all the films is less than 0.4 nm, indicating smoother films surface, which is consistent with the previous reports on α-Fe$_2$O$_3$ [18]. Overall, there is no major difference in surface roughness for the three different orientations of α-Fe$_2$O$_3$ thin films.

APPENDIX B: MAGNETIC CHARACTERIZATION OF α-Fe$_2$O$_3$ THIN FILM

The in-plane magnetization curves were obtained at room temperature using the magnetic property measurement system (MPMS) with a superconducting quantum interference device (SQUID). Figure 7, the magnetic moment was plotted as a function of external magnetic field (by sweeping the magnetic field from 70 kOe to -70 kOe) for the (a) R-, (b) A-, and (c) C-plane α-Fe$_2$O$_3$ thin films. For all three cases, we observed a dominating diamagnetic contribution arising from their respective R-, A-, and C-planes Al$_2$O$_3$ substrate. However, a
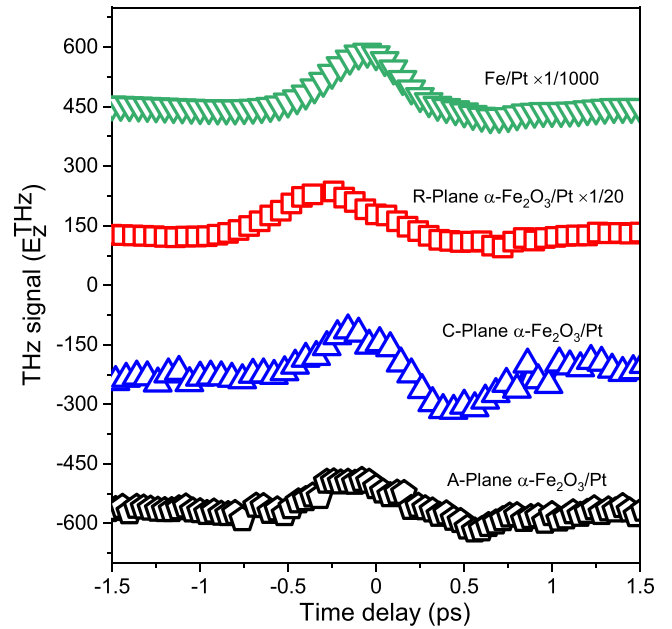
weak "ferromagnetic-like" signal were observed for the all films in the low field region, which might be due to the flux-pinning of the superconducting solenoid. We also measure the magnetization curve for pristine Al$_2$O$_3$ substrate without films and observe a small ferromagnetic signal, indicating the "ferromagnetic-like" signal was not originated from the film.

APPENDIX C: COMPARISON OF THZ SIGNAL AMPLITUDE OF α-Fe$_2$O$_3$/Pt WITH REF Fe/Pt

We compared the THz amplitude of the R-, A-, and C-plane oriented α-Fe$_2$O$_3$/Pt heterostructures with a Fe/Pt sample, which is our reference sample. The results are presented

FIG. 8. Comparison of THz signal amplitude of R-, A-, and C-plane oriented α-Fe$_2$O$_3$/Pt heterostructure with the reference Fe/Pt sample. The THz amplitudes are shifted vertically for clarity.
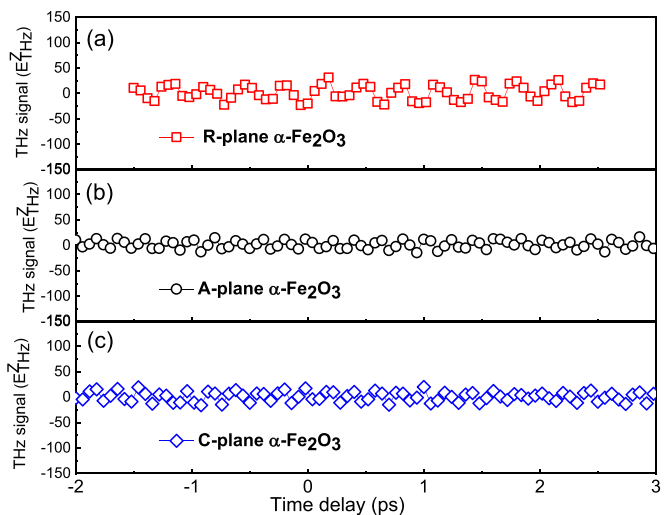

FIG. 9. THz measurement of only α-Fe$_2$O$_3$ thin film grown in (a) R-, (b) A-, and (c) C-plane orientation.

in Fig. 8. We used identical setup conditions for all measurements. The THz amplitude of our C-plane α-Fe$_2$O$_3$/Pt sample is found to be three orders less compared to the reference Fe/Pt sample. The same order of THz amplitude was reported by Qiu *et al.* for C-plane α-Fe$_2$O$_3$/Pt [18]. The R-plane-oriented sample, on the other hand, shows one order of magnitude greater amplitude than A and C plane oriented α-Fe$_2$O$_3$/Pt. Our comparison confirms that the R-plane α-Fe$_2$O$_3$/Pt sample is more efficient for THz emission, even when compared with Qiu *et al.* [18].

APPENDIX D: THZ TIME-DOMAIN SPECTROSCOPY OF PURE α-Fe$_2$O$_3$ THIN FILMS

To verify the contribution of the substrate and the α-Fe$_2$O$_3$ thin films to the terahertz emission, we conducted THz measurements on R-, A-, and C-planes α-Fe$_2$O$_3$ thin films, as shown in Fig 9. We found no signal from the α-Fe$_2$O$_3$ thin films alone. This confirms that no significant terahertz signal is present from the ultrafast dipole radiation. The terahertz signal is known to be weaker in pure antiferromagnetic materials, as reported in previous studies [16,55].

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