Role of crystalline and damping anisotropy to the angular dependences of spin rectification effect in single crystal CoFe film

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Abstract
The angular dependence of the microwave-driven spin rectification (SR) effect in single crystalline Co₀.₅Fe₀.₅ alloy film is systematically investigated. Due to the strong current-orientation dependent anisotropic magnetoresistance (AMR), the SR effects in CoFe film strongly deviate from the ordinary sin²ϕ_M relation with ϕ_M defined as the magnetization angle away from the current. A giant Gilbert damping anisotropy in the CoFe film with a maximum–minimum ratio of 520% is observed, which can impose a strong anisotropy onto magnetic susceptibility. The observed unusual angular dependence can be well explained by the theory including current-orientation dependent AMR and anisotropic magnetic susceptibility. Our work also suggests that the strong current-orientation dependent AMR in single crystalline CoFe film could exist up to the gigahertz frequency range.

1. Introduction
The electrical detection of magnetization dynamics through the spin rectification (SR) effect attracted great interest in dynamics spintronics community in the last two decades [1–16]. In the micro- or nano-structured magnetic structures, the magnetization precession can be driven by the microwave field or spin torque induced by the microwave current, thus a dc voltage can be generated through rectification between the microwave current and the oscillating electrical resistance due to anisotropic magnetoresistance (AMR). Because of the capability of detecting microwave spin excitations with dc measurements, the SR effect has been widely applied to study ferromagnetic resonance (FMR) [2–4], spin wave resonance [5, 6], nonlinear magnetization dynamics [7, 8], and domain wall dynamics [9–11]. The SR effect can also be applied in novel microwave applications, such as spintronic microwave sensing [12, 13], imaging [14, 15] and wireless energy harvesting [16].

In the devices consisting of ferromagnetic metal/heavy metal bilayer, the SR signals can also arise from the spin-pumping-driven and/or Oersted-field-driven magnetization dynamics [1, 17–22], and it was assumed that the contributions from the spin pumping effect and the Oersted field effect can be separated by the angular dependent measurements of the microwave-induced dc voltage. A comprehensive understanding on the angular dependence of SR is essential to study the spin pumping effect and quantify the spin Hall angle [23–25]. The angular dependence of SR is determined by AMR and the microwave field. AMR is usually assumed as ΔR ∝ cos²ϕ_M with ϕ_M as the angle between the magnetization and current. However, such cos²ϕ_M dependence of AMR may not be appropriate for crystalline films due to the additional influence from the crystalline symmetry [26]. In single crystalline samples, AMR can exhibit additional four-fold symmetry [27–29], asymmetric behavior [30, 31], and strong dependence on the
Figure 1. (a) Schematic of device for measuring the current-orientation dependent AMR. The ADMR curves from a 20 nm CoFe film with the current along (b) $\theta = 45^\circ$, (c) $\theta = 0^\circ$ and (d) $\theta = 15^\circ$ away from the CoFe[110] direction.

current orientation [29, 31–34]. Such unusual AMR symmetries can be used to engineer the SR signals for new features in detecting magnetization dynamics.

In this paper, we have investigated the angular dependence of the SR effect in the single crystalline Co$_{0.5}$Fe$_{0.5}$ (001) alloy film. The CoFe alloy is an important material for industry due to its large magnetization [35] and very low damping with large crystalline anisotropy [34, 36, 37]. We find that the angular dependence of the SR voltage in CoFe film strongly differs from those reported in the polycrystalline films, and can be well explained by the SR theory under consideration of the unusual angular dependent magnetoresistance (ADMR) in the CoFe film. The strong anisotropy ratio of the damping constant up to 520% is determined, which can induce the strong anisotropy of the resonant susceptibility driven by the microwave field.

2. Experiment

Single-crystal CoFe films were grown on MgO(001) substrates at room temperature by molecular beam epitaxy in an ultrahigh vacuum chamber with a base pressure of $2 \times 10^{-10}$ Torr [28, 29, 31]. The MgO(001) single-crystal substrate was first annealed at 650 °C for half an hour. Then a 10-nm-thick MgO seed layer was grown on the substrate at 500 °C. The high-quality surface was confirmed by in-situ reflective high energy electron diffraction. The CoFe alloy films were deposited via co-evaporation using Fe and Co sources at room temperature. The composition ratio was determined by the growth rate measured by a calibrated quartz thickness monitor.

We first prepared a 20 nm CoFe film for the current-orientation dependent ADMR measurement, which is covered by a 6 nm MgO layer for protection. The CoFe film was patterned into many standard Hall bars with different crystalline orientations by standard photolithography and Ar$^+$ ion milling, as shown in figure 1(a). All the Hall bars have the identical width of 100 $\mu$m and length of 300 $\mu$m.

The SR measurements were conducted on a Pt(3 nm)/MgO(6 nm)/CoFe (10 nm) trilayer at room temperature, as shown in figure 2(a). To conduct the dynamic measurements in the devices with different crystalline directions, the sample was fabricated into a dozen of 100 $\mu$m $\times$ 10 $\mu$m bars. All the bars orient along a series of crystalline orientations with the increment of 15°. For electrical measurements, Au(150 nm)/Cr(30 nm) contacts were prepared by magnetron sputtering. As shown in figure 2(a), the microwave current $I_{ac}$ flowing through the top Pt layer can generate the in-plane Oersted field $h_y$ in the CoFe layer, thus exiting the magnetization dynamics in CoFe film. A dc voltage $V_{dc}$ can be detected due to the rectification of $I_{ac}$ with the magnetoresistance variation caused by the magnetization precession [1, 2]. The 6 nm MgO insulating layer should be thick enough to suppress the spin pumping effect between CoFe and Pt [38, 39]. Thus, the magnetization dynamics should be dominantly induced by the microwave field $h_y$. 
3. Results and discussion

We first performed the ADMR measurement on the single crystalline CoFe film with different current orientations at room temperature. Figure 1(a) shows the typical device geometry, which contains many Hall bars with the current along different crystalline directions ($\theta$). The ADMR measurement was conducted by rotating the in-plane field direction ($\phi_H$) with respect to the bar. The applied rotating field is 2500 Oe, which is much larger than the magnetic anisotropy field, i.e. less than 300 Oe for the samples. Figures 1(b)–(d) show the typical ADMR curves as a function of $\phi_H$ from the devices with different current directions. The ADMR for $I|[010]$ ($\theta = 45^\circ$) in figure 1(b) shows the ordinary two-fold dependence, similar to that in the polycrystal films. The ADMR for $I|[110]$ ($\theta = 0^\circ$) in figure 1(c) contains a two-fold term and a four-fold term. Moreover, the ADMR in the Hall bar with $\theta = 15^\circ$ in figure 1(d) shows the asymmetric two-fold dependence with the maximum deviating away from 0$^\circ$ and 180$^\circ$. The AMR ratio $[=(R_{\max} - R_{\min})/R_{\min}]$ in CoFe film is 2.1$\%$ for $I|[010]$, and only 0.2$\%$ for $I|[110]$, which indicates the strong current-orientation dependence. The ADMR curves in figure 1 significantly deviate from the traditional AMR in the polycrystalline system, but can be well explained by the phenomenological model [26, 27, 29–33].

Figures 2(b) and (c) show the typical resonant voltage spectra from the device with the current along [110], and the magnetic fields are along the easy axis ([110], figure 2(b)) and hard axis ([100], figure 2(c)), respectively. All the SR spectra can be well fitted with a combination of symmetric and antisymmetric Lorentzian functions. The measured SR voltage in figures 2(b) and (c) is dominated by the antisymmetric signal $V_a$, since the SR effect in our devices is mainly excited by $h_y$. Through fitting, we can obtain the resonance field $H_r$ and the linewidth $\Delta H$. 

Figure 2. (a) Schematic of the sample and circuit for the SR measurement on the Pt(3 nm)/MgO(6 nm)/CoFe(10 nm) sample. (b) and (c) The FMR spectra with the ac current applied along the [110] direction with the magnetic field along (b) easy axis ([110]) and (c) hard axis ([100]). The frequency-dependent (d) $H_r$ and (e) $\Delta H$ from the device with the [110] orientation. The solid lines in (b) and (c) are the fitting by the combination of symmetric and antisymmetric Lorentzian functions. The lines in (d) are the fitting by equation (1), and the lines in (e) represent the linear fitting.
αH

Note that the electronic detection of anisotropic damping was performed on the Pt/CoFe bilayer in reference [34]. The frequency of applied microwave is 13 GHz. (c)–(e) The \( \varphi_M \)-dependence of antisymmetric voltage \( V \), the four-fold anisotropy field with easy axis along \([110] \) and \( H_a \) is the uniaxial anisotropy field with easy axis along \([010] \). By fitting the data with the dispersion relation, we determined the saturation magnetization \( 4\pi M_s = 24.5 \text{ kOe} \), the Landé g factor \( g = 2.13 \), the uniaxial anisotropy field \( H_a = 140 \text{ Oe} \), and the four-fold anisotropy field \( \mu_0 H_a = 270 \text{ Oe} \). The spectrum in figures 2(b) and (c) show the significant different linewidths. Figure 2(e) summarizes the fitted \( \Delta H \) as a function of \( f \) which shows significantly different slope for \( H || [110] \) and \( H || [100] \). The Gilbert damping \( \alpha \) can be extracted by fitting the data with the formula \( \Delta H = \Delta H_{inh} + 2\pi \alpha / \gamma \). \( \Delta H_{inh} \) is the inhomogeneous broadening due to the disorders in lattice structures, and the similar values around \( \approx 11 \text{ Oe} \) can be obtained for both field orientations. The fitted damping constants are \( \alpha_{[100]} = 0.0151 \pm 0.0002 \) for \( H || [100] \) and \( \alpha_{[110]} = 0.0029 \pm 0.0001 \) for \( H || [110] \), so our study provides a further experimental evidence of the giant anisotropic damping by the SR technique in single crystalline CoFe film. The measured anisotropy ratio of the damping constant is 520%, which echoes with our previously reported large value of 440% in reference [34]. Note that the electronic detection of anisotropic damping was performed on the Pt/CoFe bilayer in reference [34], and the experimental results on the Pt/MgO/CoFe trilayer may better reflect the intrinsic damping property of the CoFe layer.

Next, we studied the angular dependence of SR voltage in the devices with different current orientations. Although the SR voltage with the field along \( \varphi_M = 45^\circ \) direction from a Pt/CoFe bilayer has been reported in reference [34], the role of crystalline and damping anisotropy to the angular dependences of SR effect was still missing. For the in-plane excitation field \( h_y \), the \( \varphi_M \)-dependence of SR voltage can be expressed as

\[
V_a = \frac{I_{ac}}{2M_s} A_{as} h_y \cos \varphi_M \frac{dR}{d\varphi_M}.
\]  

Here, \( I_{ac} \) is the microwave current, \( A_{as} \) is the amplitude of antisymmetric term in magnetic susceptibility, which depends on the magnetic anisotropy and magnetic damping of the sample, \( \cos \varphi_M \) describes the projection of transverse magnetization motion onto the current direction as to modulate the magnetoresistance, and \( dR/d\varphi_M \) is the partial derivative of ADMR. By considering the magnetic anisotropies, the \( R(\varphi_M) \) relation can be calculated from the experimental \( R(\varphi_M) \) curve [31, 33], thus the \( dR/d\varphi_M \) curve can be experimentally determined. In the polycrystal systems, the ADMR is expressed as

Figure 3. The typical dc voltage spectra measured with different magnetic-field directions for the devices with (a) \( \theta = 45^\circ \) \(([010]) \) and (b) \( \theta = 0^\circ \) \(([110]) \). The frequency of applied microwave is 13 GHz. (c)–(e) The \( \varphi_M \)-dependence of antisymmetric voltage \( V \), from the devices with (c) \( \theta = 45^\circ \) \(([010]) \), (d) \( \theta = 0^\circ \) \(([110]) \) and (e) \( \theta = 15^\circ \). The lines in (a) and (b) are fittings using a combination of symmetric and antisymmetric Lorentzian functions, and the lines in (c)–(e) are the calculated \( R_{SR} \).
$R(\varphi_M) = R_0 + \Delta R \cos 2\varphi_M$, so the angular dependence of $V_s$ follows the $\sin^2 \varphi_M \cos \varphi_M$ function. However, figure 1 shows that the ADMR curves in single crystalline systems could obviously deviate from the $\cos 2\varphi_M$ function, thus the SR signal could have the angular dependence different from the simple $\sin^2 \varphi_M \cos \varphi_M$ relation.

Figure 3(a) shows the representative voltage spectra as a function of $\varphi_H$ from the [010]-oriented device ($\theta = 45^\circ$). Since the ADMR with $I||[010]$ in figure 1(b) follows the traditional $\cos 2\varphi_M$ relation, the SR should follow the ordinary $\sin 2\varphi_M \cos \varphi_M$ relation, so the measured SR signal in figure 3(a) is negligible for $\varphi_H$ at $0^\circ$ and $90^\circ$, and reaches the maximum at $\varphi_H \approx 45^\circ$. The angular dependence of $V_s$ in figure 3(c) from this device is close to the ordinary $\sin 2\varphi_M \cos \varphi_M$ relation. In the [110]-oriented device ($\theta = 0^\circ$), figure 1(c) shows that the AMR is a combination of the two-fold and four-fold terms, so $dR/d\varphi_M$ has a sign reversal for $\varphi_H$ between $0^\circ$ and $90^\circ$, resulting in the sign reversal of the SR signal at $\varphi_H \approx 50^\circ$ in figure 3(b).

Figure 3(d) summarizes the angular dependence of $V_s$ from this device, which obviously deviates from the $\sin 2\varphi_M \cos \varphi_M$ relation. Note that $V_s$ in figure 3(d) is one order smaller than that in figure 3(c). Figure 3(e) shows the angular dependence of SR voltage for the device with $\theta = 15^\circ$, which does not contain the conventional symmetric behavior at $\varphi_H = 180^\circ$ due to the non-symmetric behavior of ADMR shown in figure 1(d). So, our results demonstrate that the SR effect in the single crystalline system can have very different angular dependence than that in the polycrystalline system.

In order to quantitatively compare the angular dependence of SR signal with AMR, we measured the ADMR in each device shown in figure 2(a). The measured ADMR with different current orientation is very similar to the data in figure 1, but the AMR ratio is slightly smaller due to the contact resistance from the two-terminal measurements and the shunting effect from the thin Pt layer. The AMR ratio in figure 4(a) strongly depends on the current orientation, with a 12 times difference for the current along the $\langle 110 \rangle$ and $\langle 100 \rangle$ directions.

With the measured ADMR curve for each device, we can numerically calculate the angular dependence of $V_s$ using equation (2). Usually, it is difficult to experimentally determine $I_{ac}$ and $h_y$, which can be normalized by calculating the effective SR resistance $R_{SR} = 2V_sM_s/(I_{ac}h_y) = A_{xx}\cos \varphi_M dR/d\varphi_M$. It should be noted that both $A_{xx}$ and $dR/d\varphi_M$ could strongly depend on $\varphi_M$. $A_{xx}$ can be determined by [40, 44, 45]

$$A_{xx} = \frac{\gamma H'}{2\pi \alpha (H' + H'')}.$$  \hspace{1cm} (3)

Here, $H'$ and $H''$ are defined in equation (1). Anisotropic $A_{xx}$ could be induced by magnetic anisotropy and the anisotropy of $\alpha$. As shown in figure 2, $\alpha$ with the magnetization along $\langle 100 \rangle$ and $\langle 110 \rangle$ shows more than 500% difference. In this study, the detailed relation between $\alpha$ and the magnetization angle $\varphi_M$ relative to the [110] crystal direction was not determined. However, the $\alpha(\varphi_M)$ relation of CoFe film has been measured in reference [34], which is replotted as the circles in figure 4(c). So, in order to understand how the anisotropic $\alpha$ influences $A_{xx}$, we used the $\alpha(\varphi_M)$ relation represented by the black line in figure 4(c),

\begin{figure*}[ht]
\centering
\includegraphics[width=\textwidth]{figure4}
\caption{(a) The current-orientation dependent AMR ratio from the 10 nm CoFe sample. (b) The measured $V_{ac}^{meas}$ and the calculated $R_{SR}^{cal}$ from the devices with different orientation angle $\theta$. (c) The anisotropic damping constant $\alpha$ as a function of magnetization angle $\varphi_M$, with respect to the [110] direction for calculating $R_{SR}$. The circles are the experimental anisotropic $\alpha$ of CoFe film replot from reference [34]. (d) The calculated $\delta M$-dependent magnetic susceptibility term $A_{xx}$ due to the anisotropic damping constant $\alpha$.}
\end{figure*}

\begin{align}
A_{xx} &= \frac{\gamma H'}{2\pi \alpha (H' + H'')}.
\end{align}
which also contains a four-fold symmetry from the CoFe(001) plane. Figure 4(d) shows the calculated $A_{xx}$ as a function of $\phi_M$, which contains a strong anisotropy ratio of $\sim$5. The minimum locates at 45° and 135° due to the maxim $a$ for the magnetization along the (100) directions. We also calculated the $A_{xx}(\phi_M)$ relation with a constant $\alpha$, and found that $A_{xx}$ only varies $\sim$0.11%. So, the strong anisotropy of $A_{xx}(\phi_M)$ in figure 4(d) is mainly attributed to the giant anisotropic $\alpha$ in the CoFe film.

From the experimental ADMR curves, we can determine the angular dependence of $d\theta/d\varphi_M$, then the $\varphi_M$-dependent $R_{SR}$ can be further calculated. Figures 3(c)–(e) show that the calculated $R_{SR}$ well reproduces the angular dependence of $V_{\alpha}$, which proves that the SR effect can be correctly described by equation (2). Therefore, in the single crystalline system, it may not be proper to only use the $\sin 2\varphi_M \cos \varphi_M$ relation to separate the contribution from the ordinary SR effect and the spin pumping effect. On the other hand, we notice that both $V_{a}$ and $R_{SR}$ in figures 3(c)–(e) are zero at $\varphi_M = 90^\circ$ and 270°, so the spin pumping signal can still be separated from the SR effect if aligning the magnetization perpendicular to the microwave current direction [1, 24, 25].

We also quantify the maxim values from the measured $V_{\alpha}(\varphi_M)$ curves and the calculated $R_{SR}(\varphi_M)$ curves, and both $V_{\alpha,max}$ and $R_{SR,max}$ in figure 4(b) strongly depend on the device orientation with an in-plane four-fold symmetry, in good agreement with the orientation-dependent AMR ratio in figure 4(a). The amplitude of the measured SR signal could have 13 times difference for the devices along the (110) and (100) directions, so our dynamic measurements confirm the giant current-orientation effect of the AMR up to the gigahertz frequency range.

4. Conclusions

In conclusion, we have demonstrated very large current-orientation dependent SR signal anisotropy, up to 13 times, due to a combination of crystalline AMR anisotropy up to 12 times and giant Gilbert damping anisotropy up to 520% in single crystalline CoFe film, with both anisotropies absent in ordinary polycrystalline magnetic films. The SR theory can well explain the unusual angular dependence of SR signals by considering both the unconventional ADMR and the anisotropic susceptibility in single crystalline CoFe film. Our measurements show that introducing crystalline symmetry into the magnetic system can significantly modify the microwave detection efficiency at different orientations using SR signals, which provides a new approach to engineer the microwave characteristics in microwave spintronics.

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