Crystallographic anisotropy of the intrinsic Gilbert damping for single-crystalline Fe film

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We experimentally investigated the crystallographic anisotropy of the intrinsic Gilbert damping constant in single crystalline Fe(001) film deposited on MgO(001) substrate. The frequency-domain ferromagnetic resonance (FMR) spectrum was measured at given external magnetic fields via a vector network analyzer. From the field-domain FMR linewidth obtained as a function of resonant frequency, the intrinsic Gilbert damping constants along the crystallographic axes in the (001) plane were successfully obtained by evaluating the extrinsic influence of two magnon scattering and inhomogeneity. A four-fold crystalline anisotropy of the intrinsic Gilbert damping constant, similar to the magnetic crystallographic anisotropy in the (001) plane, was clearly observed. Furthermore, we found that the intrinsic Gilbert damping constant along the easy axis was smaller than that along the hard axis.

Key words: Gilbert damping, single crystal, thin film, ferromagnetic resonance

1 Introduction

Single-crystalline ferromagnetic thin films will be predominantly used in future spintronics devices. Controlling the Gilbert damping constant α in ferromagnetic thin films is crucial for future spintronics applications. In ferromagnetic materials, the strength of the spin-orbit interaction is responsible for both the static and dynamic properties of magnetization, for example, a crystalline magnetic anisotropy and magnetization damping.1–19) Thus, similar to the crystalline magnetic anisotropy, one can expect that the Gilbert damping constant governing the magnetization damping will also show a crystallographic anisotropy. However, it is still unclear whether the Gilbert damping constant has crystallographic anisotropy. Various theoretical approaches have been proposed to describe the anisotropy of the Gilbert damping term in the Landau–Lifshitz–Gilbert equation, which describes the precessional motion of magnetization.1,4–11)

There are two types of anisotropy of α, a rotational anisotropy and an orientational anisotropy. The former is due to the change in damping for different directions of the temporal change in magnetization &dalpha;/dt, and the latter is attributed to the damping dependence on the momentary orientation of \( \mathbf{M}(t) \). Consequently, the Gilbert damping term can be defined by a damping matrix.7–9)

In the case of uniform mode, the orientational anisotropy mainly determines the Gilbert damping term. Seib et al.9) and Gilmore et al.7) numerically found that the Gilbert damping constant for bcc-Fe was smallest along (100), larger along (110), and at its largest along (111). Recently, Meckenstock et al.17) and Isogami et al.18) experimentally found that the Gilbert damping constant along the magnetic easy axis was smaller than that along the hard axis for single-crystalline Fe and Fe3N films.

To develop a better understanding of its anisotropic behavior, it is important to interpolate the Gilbert damping constant for many crystallographic orientations between the easy and hard axes. Furthermore, it is important to assess the common experimental methods used to measure the intrinsic Gilbert damping constant of single-crystalline ferromagnetic thin films. Generally, the ferromagnetic resonance (FMR) linewidth is measured to determine the Gilbert damping constant.12–19) The linewidth is, however, not simply in proportion to the intrinsic Gilbert damping constant because extrinsic contributions such as two magnon scattering (TMS) and inhomogeneous line-broadening (ILB) also increase the FMR linewidth.11,16) To evaluate the intrinsic Gilbert damping constant precisely, those extrinsic contributions must be excluded. The out-of-plane angular dependence of the linewidth is typically measured to extract the intrinsic Gilbert damping constant from the FMR spectrum data.12–19)

However, in the case of single-crystalline films, the out-of-plane angular dependence of the damping constant should be considered. Namely, we need an alternative method to evaluate the extrinsic contributions at a given crystallographic axis. Note that the frequency dependence of the field-domain linewidth \( \Delta H \) owing to the intrinsic Gilbert damping totally differs from the frequency dependence of both TMS and ILB. Thus, vector network analyzer (VNA)-FMR spectroscopy, which can measure the frequency dependence of \( \Delta H \) without changing the direction of magnetization, is an ideal method to examine the crystallographic anisotropy of the intrinsic Gilbert damping constant.

In this paper, we experimentally demonstrated the anisotropy of the intrinsic Gilbert damping constant for single-crystalline Fe(001) film via VNA-FMR spectroscopy. A clear four-fold symmetry of the intrinsic Gilbert damping was observed in the Fe(001) plane. Similar to the result of previous numerical studies,7) we found that the intrinsic Gilbert damping constant along the easy axis was smaller than that along the hard axis.
Fig. 1 (a) RHEED image of the Fe(001) film. The incident electron beam was parallel to the MgO[100] direction. The spots of (200), (211), and (21T) derived from Fe(001) plane were observed. (b) Epitaxial orientation in Fe(001)/MgO(001). The gray and white circles represent Fe and MgO, respectively.

2 Experimental

Single-crystalline Fe(001) film was deposited on a MgO(001) substrate using a molecular beam epitaxy in ultrahigh vacuum (UHV-MBE). Before the deposition, we cleaned the MgO substrate according to the following procedure. The MgO substrate was washed for 10 minutes each with 2-butanone, acetone, and 2-propanol using an ultrasonic cleaner. After the cleaning, the MgO substrate was set in the MBE chamber and flushed under 5.0 × 10⁻⁷ Pa at 400 °C for 2 h. Then, the substrate was cooled to 100 °C, and the Fe was deposited on the MgO substrate at 0.01 nm/s. The thickness of the Fe film was 50 nm. After the deposition, the Fe film was annealed at 400 °C for 2 h. Later, in-situ reflection high energy electron diffraction (RHEED) and X-ray diffraction (XRD) were observed (see Figs. 1 and 2).

In order to observe the FMR spectrum of the Fe(001) film via a VNA, the film was patterned into wire-shaped structures using electron beam lithography and an Ar⁺ milling technique. The wires were 240-μm long and 10-μm wide. In total, 12 wires with different crystalline orientations were fabricated. The angle between Fe[100] and the longitudinal axis of the wire, δ(100), was fabricated between 0° and 165° in 15° increments. After fabricating the Fe wires, a 150-nm-thick insulating SiO₂ layer was sputtered onto the film plane. Finally, an electrically shorted coplanar waveguide (CPW) comprising Ti (5 nm) / Au (80 nm) was fabricated onto each of the Fe wires. The 10-μm-wide center signal line of CPW was overlapped onto the Fe wire. An optical micrograph of a representative sample and the schematic cross-section are shown in Figs. 3(a) and 3(b), respectively.

The sample was placed on an in-plane biaxial electromagnet that generates x- and y-axes magnetic fields. The CPW was electrically connected to the VNA using a ground-signal-ground-type microwave probe, which produces an alternating magnetic field.

Fig. 2 XRD profiles of 50-nm-thick Fe thin film deposited on a MgO(001) substrate. (a) θ–2θ scan profile. The Fe(001) plane is parallel to the MgO(001) plane. (b) φ scan profile of the Fe(110) plane. Four-fold crystalline symmetry is observed. ω–2θ scan profile of (c) Fe(200) and (d) MgO(220). The linewidths measured by the Cu-Kα₁ source are 0.208° ± 0.003° for Fe(110) and 0.044° ± 0.001° degree for MgO(200). (e) In-plane XRD profile of Fe(110). The linewidth Δφ = 0.576° ± 0.002°.
$h_{ac}$ along the y-axis. The microwave absorption due to FMR in the Fe wire can be detected as the change in the reflection coefficient $S_{11}$ of the CPW. In order to evaluate the background noise of the measurement system, we measured the frequency dependence of $S_{11}$ at an external magnetic field of 3140 Oe along the y-axis. The microwave absorption due to FMR in this field orientation was sufficiently small because $h_{ac}$ was applied parallel to the magnetization along the y-axis. After measuring $S_{11}^{ref}$, an external magnetic field $H_{ex}$ was applied parallel to the x-axis between 2380 and -2380 Oe, at which $S_{11}$ was measured. At each amplitude of $H_{ex}$, the difference between $S_{11}$ and $S_{11}^{ref}$, $\Delta S_{11}$ ($= S_{11} - S_{11}^{ref}$), corresponding to the microwave absorption due to FMR, was measured as a function of microwave frequency.

3 Results and Discussion

Figure 1 shows the RHEED image of the annealed Fe film. The incident e-beam was parallel to the direction of MgO[001]. The image indicates (200), (211) and (21T) spots as reciprocal lattices of the Fe(001) plane. On an MgO(001) plane, an Fe film grows, with Fe[110] parallel to MgO[100].

Out-of-plane and in-plane XRD results are shown in Figs. 2(b) and (c). The X-ray source used was Cu-K$_x$ (wavelength $\lambda = 0.154$ nm). As shown in the $\theta$–2$\theta$ measurement result, Fig. 2(a), the peaks observed at 2$\theta$ = 43.3° and 65.3° are derived from MgO(002) and Fe(002), respectively. From Bragg’s law, the lattice constants of MgO, $a_{MgO}$, and Fe, $a_{Fe}$, are estimated to be 4.18 and 2.86 Å, respectively. Therefore, the lattice mismatch at the interface is $\frac{a_{Fe} - a_{MgO}}{a_{MgO}} \times 100 = -3.30\%$ along the MgO[110] direction. Figure 2(b) shows the result of the $\phi$ scan measured along the Fe(110) plane. Four-fold symmetry was clearly observed here. Figures 2(c) and (d) show the results of the 2$\theta$–$\omega$ scan for Fe(200) and MgO(220). By the Scherrer equation, the crystallite size $\tau_{Fe}$ was estimated as

$$\tau_{Fe} = \frac{K_{Sch} \cdot \lambda_{X-ray}}{\beta_{Sch} \cdot \cos \theta_{Fe}}.$$  (1)

where $K_{Sch}$ represents the shape factor (generally $\approx 0.9$) and $\lambda_{X-ray}$ is the wavelength of the X-ray source. $\beta_{Sch}$ and $\theta_{Fe}$ are the full-width at half-maximum (FWHM) and the Bragg angle, respectively. In this experiment, $\lambda_{X-ray} = 0.154$ nm, $\beta_{Sch} = 0.208 \pm 0.003^0$ and $\theta_{Fe} = 64.9^0$, respectively. From those values, $\tau_{Fe} = 46.30 \pm 0.75$ nm was calculated. As shown in Fig. 2(e), the FWHM of the $\phi$–$2\theta_{Fe}$ scan, $\Delta \phi$, is estimated from the Gaussian fitting as 0.576° $\pm$ 0.002°.

Figure 4 shows the normalized in-plane magnetization curve measured using a superconducting quantum interface device with the magnetic field applied along Fe[100] and [110]. The saturation magnetization $M_s$ was 1446 emu/cm$^3$ and the saturation field was approximately 600 Oe at room temperature. As shown in Fig. 4, the hysteresis curve of Fe[110] has the knee at the magnetic field of 600 Oe, which corresponds to the anisotropy field in the (001) plane.

Figure 5 shows the FMR spectrum for the Fe wire with $\delta_{[100]} = 0^0$ measured at $H_{ex} = 620$ Oe. A dip attributed to the microwave
Absorption caused by FMR can be clearly seen at 15.1 GHz. For the VNA-FMR measurement, the real part of $\Delta S_{11}$ is given by

$$\Re [\Delta S_{11}] = (C'\chi' + C''\chi'')f,$$

(2)

with the complex susceptibility $\chi = \chi' - i\chi''$ given by

$$\chi' = \frac{M_f f_1 f_2^2 - (1 - \alpha_{total}^2) f_2^2}{f_2^2 - (1 + \alpha_{total}^2) f_2^2 + 4\alpha_{total}^2 f_2^2},$$

(3)

and

$$\chi'' = \frac{\alpha_{total} M_f f_1 f_2^2 + (1 + \alpha_{total}^2) f_2^2}{f_2^2 - (1 + \alpha_{total}^2) f_2^2 + 4\alpha_{total}^2 f_2^2}.$$

(4)

Here, $\alpha_{total}$ is the effective damping constant consisting of the intrinsic Gilbert damping constant and the extrinsic contributions from TMS and ILB. $f_1$ is the resonant frequency and $C'$, and $C''$ are coefficients that depend on the CPW geometry.\(^\text{23}\) Note that the imaginary part in Eq. (2) is used to separate out the influence of a small error in calibrating the $S$-parameter and/or a capacitive component of CPW. Experimental results were fitted by Eq. (2), and $\alpha_{total}$, $f_1$, and $C'$ and $C''$ were evaluated as fitting parameters. As indicated by the dashed line in Fig. 5, the experimental data were well fitted by Eq. (2). The frequency-domain FMR linewidth was given by $\Delta f = 2\alpha_{total} f_1$. The external field dependence of the FMR frequency along Fe[100] and Fe[110] are shown in Fig. 6.

The value of $f_1$ satisfies following equation proposed by Shu\(^\text{24}\),

$$\left(\frac{f_1}{\gamma_0}\right)^2 = \frac{1}{M_f^2 \sin^2 \theta_M} \left\| \frac{\partial^2 E}{\partial \theta \partial \phi} - \left(\frac{\partial^2 E}{\partial \phi \partial \phi}\right) \right\|_{\theta_M, \phi_M, \theta_M, \phi_M},$$

(5)

where $\gamma_0$ is the gyromagnetic ratio. The free energy $E$ is given by the summation of the Zeeman energy $E_{Zeeman}$, demagnetizing energy $E_d$, and magnetic crystalline anisotropy energy $E_a$ such that

$$E = E_{Zeeman} + E_d + E_a,$$

(6)

where

$$E_{Zeeman} = - M \cdot H_{ex}.$$

(7)

$$E_d = \frac{1}{2} \left( - \tilde{N}_d M \right) \cdot M,$$

(8)

and

$$E_a = K_1 (\alpha_1^2 \alpha_2^2 + \alpha_2^2 \alpha_3^2 + \alpha_3^2 \alpha_1^2).$$

(9)

Here, $\tilde{N}_d$ is the demagnetizing tensor, $K_1$ is the first-order crystalline magnetic anisotropy constant, and $\alpha_i$ ($i = 1, 2, 3$) are the direction cosines of the magnetization with respect to the [100], [010], and [001] axes, respectively. The angle of the external field (magnetization) with respect to the direction perpendicular to plane is given by $\theta_M(\theta_M)$, and $\phi_M(\theta_M)$ represents the angle between the direction of the external field (magnetization) and longitudinal axis of the wire (see Fig. 3(c)). In this paper, we assumed $H_{ex}$ was applied in the film plane, i.e., $\theta_M = \frac{\pi}{2}$. After

The external magnetic field dependence of the FMR frequency for Fe[100] and Fe[110]. Both the experimental results are fitted by Eq. (5).

The calculation, the elevation angle $\theta_M \approx \frac{\pi}{4}$ was obtained. By fitting $f_i$ versus $H_{ex}$ data for the [100] direction with Eq. (5), we evaluated $K_1 = 4.48 \times 10^6$ erg/cm$^3$ and $\gamma_0 = 2.93 \times 10^6$ Hz/Oe.

In order to evaluate the saturation field of the magnetization along (110) from the calculated value of $K_1$, we calculated the stable azimuthal angle of the magnetization as a function of $H_{ex}$. Figure 7 shows the calculated azimuthal angle of the equilibrium direction of magnetization as a function of magnetic field applied along both [100] and [110]. As the strength of $H_{ex}$ was smaller than 0.6 kOe, the magnetization deviates from the longitudinal axis because of the crystalline anisotropy of the Fe(001) film. Therefore, the bending in the $H_{ex}$ dependence of $f_i$ for Fe[110] is attributed to the inability of the magnetization to saturate along Fe[110] until the magnetic field becomes larger than 0.6 kOe. This is consistent with the magnetic field needed to saturate the magnetization along [110] (see Fig. 4). As a consequence, we can neglect the linewidth broadening due to a field dragging effect\(^\text{19,20}\) for fields greater than 0.6 kOe.

Figures 8(a) and 8(b) respectively show the polar plots of $f_i$ and $\Delta f$ with respect to $\delta_{100}$ measured at 2380 Oe. Clear fourfold symmetry in $f_i$ is attributed to the four-fold symmetry of the
crystalline magnetic anisotropy of Fe(001). Similar symmetry was also observed in $\Delta f$. However, this is not direct evidence for the existence of crystalllographic anisotropy of the intrinsic Gilbert damping constant $\alpha_{int}$ in Fe(001) because there remains extrinsic contributions to the linewidth. To explore the crystallographic anisotropy of the Gilbert damping constant, we excluded the extrinsic contributions as follows. According to the theoretical model proposed by Krivosik\(^1\), the effective FMR linewidth is represented by

$$\Delta H_{\text{eff}} = \frac{2}{\gamma_0} \alpha_{int} f_i + \Delta H_{\text{TMS}} + \Delta H_{\text{ILB}}. \quad (10)$$

The first term of the right-hand side of Eq. (10) is the linewidth due to the intrinsic damping with $\alpha_{int}$, while the second and third terms of Eq. (10) are the line-broadening contributions from TMS and ILB, respectively. $\Delta H_{\text{TMS}}$ is given by\(^1\)

$$\Delta H_{\text{TMS}} = \frac{\partial f_i}{\partial H_{\text{ex}}} \xi^2 \left(\frac{\partial H_{\text{ex}}}{\partial f_i}\right)^2 \int_{|k| \leq \frac{\xi}{2}} \delta(f_k - f_i) dk. \quad (11)$$

Here, $\delta H_{\text{ex}}$ represents the distribution of the in-plane crystalline anisotropy field, $f_i$ is the magnon frequency with wave number $k$, and $\xi$ is the magnon mean free path. The magnon frequency $f_k$ is written as

$$f_k = \gamma_0 \sqrt{H_{k,x}H_{k,y}}. \quad (12)$$

with

$$H_{k,x} = H_{ex} + \frac{A}{M_s} k^2 + \left(N_y - N_z\right) M_s \left(1 - \frac{e^{-kd}}{kd}\right) \sin^2 \psi_k. \quad (13)$$

and

$$H_{k,y} = H_{ex} + \frac{A}{M_s} k^2 + \left(N_z - N_x\right) M_s \frac{1 - e^{-kd}}{kd}. \quad (14)$$

Here, $N_x$, $N_y$, and $N_z$ are the components of the demagnetizing tensor in the $x$, $y$, and $z$ axes, $A$ is the exchange stiffness constant, $d$ is the thickness of the sample and $\psi_k$ represents the angle between the direction of the magnon’s propagation and magnetization direction. The $\delta$-function in Eq. (11) should be integrated when TMS occurs, i.e., $f_i = f_k$. The magnon mean free path $\xi$ is considered to be less than or about the same as the crystallite size because magnetic properties differ between neighboring crystallites. Thus, the magnon wavenumber $|k|$ should be smaller than the inverse of the crystallite size, $|k| \geq \frac{2\pi}{\xi}$. Additionally, in a single-crystalline ferromagnet, $\xi$ is considered to be approximately equal to the crystallite size $\tau_{Fe}$ ($\xi \approx \tau_{Fe}$). Meanwhile, we can estimate $\delta H_{ex}$ as $\delta H_{ex} \approx \frac{D_{Fe} \xi}{\tau_{Fe} \xi} = 0.93 \text{ Oe}$.

Figure 9 shows $\Delta H_{\text{TMS}}$ as a function of $f_i$ at $\xi$ in the range 40–50 nm.

![Fig. 9](image9.png)

**Fig. 9** The external magnetic field dependence of the field-domain FMR linewidth for Fe[100] ($\delta_{(100)} = 0^\circ$) and Fe[110] ($\delta_{(110)} = 90^\circ$). The dashed lines represent the linear fit for Eq. (15).

![Fig. 10](image10.png)

**Fig. 10** Calculated $\Delta H_{\text{TMS}}$ as a function of $f_i$ at $\xi$ in the range 40–50 nm.

In the case of [110], $\Delta H_{\text{eff}}$ shows a marked increase with decreasing $f_i$ in the range below 13.5 GHz. The line-broadening observed at $f_i < 13.5$ GHz is mainly caused by an increase in the change of $f_i$ with $H_{ex}$, i.e., $\frac{\partial f_i}{\partial H_{ex}}$ in [110]. In order to evaluate the TMS contribution, we calculated the frequency dependence of $\Delta H_{\text{TMS}}$ for different magnon mean free path $\xi$ of 40, 45, and 50 nm (Fig. 10). The values of $\Delta H_{\text{TMS}}$ are much smaller than that of $\Delta H_{\text{eff}}$, and thus the line-broadening...
As per Eq. (15), $\Delta H_{\text{eff}}$ is independent of the FMR frequency $f_i$. From the difference in these frequency dependencies, we can evaluate those contributions independently. $\Delta H_{\text{eff}}$ was fitted by the linear function of $f_i$ as dashed lines in Fig. 9, and $\sigma_{\text{int}}$ was estimated from the gradient of the dashed line. Figure 11 shows the polar plot of $\sigma_{\text{int}}$ with respect to $\alpha_{[100]}$. As shown in Fig. 11, $\sigma_{\text{int}}$ shows four-fold symmetry in Fe(001). Small inconsistencies between the crystallographic axes and directions showing the local minima of $\sigma_{\text{int}}$ may be due to imperfections in the microfabrication of the Fe wire. Figure 11 also suggests that $\sigma_{\text{int}}$ along [100] is smaller than that along [110]. This tendency is consistent with the numerical study conducted by Gilmore et al. Namely, the anisotropy in the spin–orbit interaction responsible for the crystalline magnetic anisotropy also leads to the anisotropy of $\sigma_{\text{int}}$ with similar symmetry. Our experimental result implies that the anisotropy of $\sigma_{\text{int}}$ has a great effect on the performance of spintronics devices comprising single-crystalline ferromagnetic thin films.

4 Summary

We have demonstrated the crystalline anisotropy of the intrinsic Gilbert damping for single-crystalline Fe(001) film. FMR spectra with given crystalline axes were measured via VNA-FMR spectroscopy. The extrinsic contributions to the FMR linewidth from two-magnon scattering and inhomogeneous line-broadening were separated from the intrinsic damping effect by way of a theoretical approach. A four-fold symmetry in $\alpha_{\text{int}}$ was clearly observed, and the value of $\alpha_{\text{int}}$ along the easy axis was smaller than that along the hard axis. This interesting characteristic should be considered when epitaxial ferromagnets are applied to spintronics devices of the future.

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