Dependence of Magnetic Damping on Temperature and Crystal Orientation in Epitaxial Fe₄N Thin Films

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In-plane and out-of-plane ferromagnetic resonance (FMR) were used to investigate the intrinsic magnetic damping constant (α) in epitaxial Fe₄N thin films deposited on MgO substrates. The dependence of α on temperature was evaluated from room temperature (RT) to 4 K. The external magnetic field (H) of FMR was applied in two directions, i.e., [100] and [110], of the Fe₄N lattice. Anisotropic α was observed from RT to 4 K. Moreover, the α for $H \parallel$ [100] exceeded the α for $H \parallel$ [110] at 180 K. Numerical calculations of α for bulk Fe₄N revealed the same behavior as that in the experiments. The temperature dependence of anisotropic α was explained by the changes in the electronic band structure depending on the directions of magnetization.

Key words: magnetic damping, ferromagnetic resonance, Fe₄N thin film

1. Introduction

Fe₄N thin films with the γ' phase have attracted attention due to the negative spin polarization of the density of states (DOS).¹⁾ Using a Fe₄N thin film as an FM electrode of the magnetic tunnel junction (MTJ), Fe₄N/MgO/CoFeB-MTJ, we reported an inverse tunnel magnetoresistive (inverse-TMR) effect and inverse current-induced magnetization switching (inverse-CIMS).²⁻⁵⁾ One of the advantages of the inverse properties is that highly efficient spin-torque oscillation can be expected in a MTJ with dual MgO barriers, for example, CoFeB (pinned layer) / MgO / CoFeB (free layer) / MgO / Fe₄N (pinned layer), because spin-torque transfers from both the pinned layers. For application of Fe₄N films in spintronics devices, the magnetic damping properties need to be understood, since the critical switching current density in CIMS strongly depends on them.

In recent years, down-sizing of magnetic elements into the several-10s of nanometer range has become indispensable for high areal densities. Nanoscale magnets exclude magnetic domain structures and the resultant anisotropic magnetic damping might be predominant. The anisotropic damping constant of epitaxial films has been reported by some groups. For example, R. Yilgin *et al.* reported that maximum and minimum “intrinsic” damping constants (α) were observed at room temperature (R.T.) in the hard-axis and the easy-axis of epitaxial Co₂MnSi films, respectively.⁶⁾ Other reports have revealed anisotropic α at R.T. in hcp and bcc-Co epitaxial films.⁷⁾ On the other hand, anisotropy has not been reported in Fe and fcc-Co epitaxial films.⁸⁻¹⁰⁾ These reports have led us to consider that not all films have anisotropic α , and it is not clear what determines the occurrence of anisotropic α .

For the microscopic origins of magnetic damping, several researchers have proposed the spin-orbit interaction, which is the coupling between spin and lattice, and the spin-flip, due to impurities.¹¹⁻¹⁴⁾ Such intrinsic origins become predominant by excluding the thermal fluctuation of spins. Kambersky and Gilmore *et al.* introduced the electron lifetime (τ), which can be converted to temperature, and explained the experimental data regarding the temperature (down to 4 K) dependence of α conducted by Bhagat *et al.*¹⁵⁻¹⁷⁾; nevertheless, they did not address the issues of the mechanisms of anisotropic α .

We investigated α of “poly”-crystalline Fe₄N films in a previous study.¹⁸⁾ In contrast, epitaxial Fe₄N films, considered an important spintronics material for the future, excited our curiosity based on how its magnetocrystalline anisotropy acts upon anisotropic α . The present work was intended to evaluate the temperature dependence of α by means of in-plane and out-of-plane ferromagnetic resonance (FMR), as well as numerical calculations, to promote our understanding of the mechanism of anisotropic α .

2. Experimental and analysis procedure

100-nm-thick epitaxial Fe₄N films were deposited on MgO substrates using an ultra-high-vacuum magnetron sputtering system. Prior to the deposition, a dry-etching process on the MgO substrate was performed with an RF power density of 0.4 W/cm². To promote chemical ordering of γ' -Fe₄N films, *in situ* IR thermal treatment was performed at 300 °C for 15 min immediately after deposition of the Fe₄N film. After cooling the film to R.T. in 30 min, a 1-nm-thick Ta cap layer was deposited on the Fe₄N film surface. The

Fig. 1 (a) Definition of polar angle (θ) and azimuthal angle (ϕ) of external field (H) and magnetization (M). (b) Schematic layout of coplanar waveguide (CPW) and vector network analyzer (VNA).

optimum conditions to achieve a single-phase Fe_4N film were determined by X-ray diffraction (XRD) structural analysis, and measurements of the saturation magnetization (M_s) and negative anisotropic magnetoresistance properties.¹⁹⁾ Using a torque magnetometer, the in-plane magnetocrystalline anisotropy constant (K_1) was determined using the saturated torque amplitude. The resonance field (H_r) and line-width (ΔH) under the FMR state were measured using the electron cyclotron resonance (ESR) machine with X-band (9.89 GHz) microwaves. Rectangular samples with dimensions of $2 \times 2 \text{ mm}^2$ were placed on a quartz rod and introduced into the microwave cavity (TE_{011}). In the in-plane case, H was applied along $[100]$ and $[110]$ of the epitaxial Fe_4N film. In the out-of-plane case, H was rotated in the (010) and (110) planes of the film to evaluate α from ΔH . The sample temperature (T) was changed from R.T. to 4 K. Another FMR experiment was performed as a reference measurement by scanning the applied microwave frequency using a vector network analyzer (VNA) and a coplanar waveguide (CPW). The setups are shown in Fig. 1(b).

The total free energy density of magnetization in this study is expressed as

$$E = -M_s H [\sin \theta_H \sin \theta_M \cos(\phi_H - \phi_M) + \cos \theta_H \cos \theta_M] + (2\pi M_s^2 - K_\perp) \cos^2 \theta_M + \left(\frac{K_1}{4} \right) [\sin^4 \theta_M \sin^2 \phi_M + \sin^2 2\theta_M] \quad (1)$$

where M_s , H , $\theta_{H(M)}$, $\phi_{H(M)}$, and K_1 represent the saturation magnetization, external field, polar angle of the external field (magnetization) with respect to the $[001]$ direction, azimuthal angle of the external field (magnetization) with respect to the $[100]$ direction, and the magnetocrystalline anisotropy constant, respectively. Based on Eq. (1), the actual equation in the FMR condition is given by

Fig. 2 (a) In-plane XRD profile of 100-nm-thick epitaxial Fe_4N film measured with X-ray incidence angle of 0.4° with respect to film surface. Upper and lower profiles were observed with scattering vector (Q) points to $[100]$ and $[110]$ of Fe_4N , respectively. (b) ϕ scan profile with $2\theta_\chi = 47.8^\circ$, which corresponds to the $\text{Fe}_4\text{N}(200)$ diffraction angle. (c) In-plane magnetization hysteresis loops (M - H loops) at RT with external field applied to $[100]$ (solid line) and $[110]$ (dashed line).

$$\frac{\partial E}{\partial \theta_M} = 0 \quad \text{and} \quad (2)$$

$$\left(\frac{2\pi f_r}{\gamma} \right)^2 = \left[\frac{\partial^2 E}{\partial \phi_M^2} \frac{\partial^2 E}{\partial \theta_M^2} - \left(\frac{\partial^2 E}{\partial \phi_M \partial \theta_M} \right)^2 \right] = [H_r \cos(\theta_H - \theta_M) + A] \times [H_r \cos(\theta_H - \theta_M) + B]. \quad (3)$$

Here, A and B are functions of $\theta_{H(M)}$ and $\phi_{H(M)}$. To extract α from the effective magnetic damping constant (α_{eff}), which includes the contribution attributable to inhomogeneous magnetic properties, the θ_H dependence of ΔH was evaluated. We assumed $\Delta H = \Delta H_\alpha + \Delta H_{\Delta 4\pi M_{\text{eff}}} + \Delta H_{\Delta \theta} + \Delta H_{K_1}$, where ΔH_α , $\Delta H_{\Delta 4\pi M_{\text{eff}}}$, $\Delta H_{\Delta \theta}$, and ΔH_{K_1} represent the line-width due to intrinsic magnetic damping, the distribution of the magnitude of demagnetization, the distribution of the H direction, and distribution of K_1 , respectively. The respective formulas are expressed as

$$\Delta H_\alpha = \frac{1}{\sqrt{3}} [\alpha (2H_r \cos(\theta_H - \theta_M) + A + B)] \left| \frac{d(2\pi f_r / \gamma)}{dH_r} \right|^{-1}, \quad (4)$$

$$\Delta H_{\Delta 4\pi M_{\text{eff}}} = \frac{1}{\sqrt{3}} \left| \frac{dH_r}{d(4\pi M_{\text{eff}})} \right| \Delta 4\pi M_{\text{eff}}, \quad (5)$$

$$\Delta H_{\Delta \theta} = \frac{1}{\sqrt{3}} \left| \frac{dH_r}{d\theta_H} \right| \Delta \theta_H, \quad (6)$$

$$\Delta H_{K_1} = \frac{1}{\sqrt{3}} \left| \frac{\partial H_r}{\partial (2\pi f_r / \gamma)} \frac{\partial (2\pi f_r / \gamma)}{\partial K_1} \right| \Delta K_1. \quad (7)$$

Fig. 3 (a) Torque curve of 100-nm-thick epitaxial Fe₄N film with diameter of 8 mm. (b) Magnetocrystalline anisotropy constant (K_1) as a function of the reciprocal of the applied field ($1/H$). The open circles, triangles, and squares correspond to experimentally obtained K_1 values for thicknesses of 50, 100, and 200 nm, respectively. The solid lines plot the linear fitting results using the least-squares method.

In this study, by fitting the experimental ΔH as a function of θ_H using Eqs. (4)-(7), α was evaluated.

3. Experimental results

3.1 Crystallographic textures and magnetic properties of epitaxial Fe₄N thin film

Figure 2(a) shows the XRD profile with grazing incidence, the ϕ -scan. Both profiles for the X-ray scattering vectors (Q) set to [100] and [110] of Fe₄N are shown on the same panel. Diffraction from the Fe₄N (100) and (110) superlattice appeared at 23.6° and 33.5°, respectively. Judging from the estimation using the Bragg's formula, small diffractions that appeared at 44.3° for $Q \parallel [100]$ and at 64.6° for $Q \parallel [110]$ might correspond to the bcc-Fe (110) and (200), respectively. Such the speculation implies the existence of ultra-thin Fe film, an initial growth layer, with α phase at the interface between MgO substrate and Fe₄N film due to its large lattice misfit to be 10.8 %. The ϕ scan profile is shown in Fig. 2(b). Diffraction peaks every 90° appeared with almost the same intensity. The results revealed that a Fe₄N film was successfully grown on the MgO(100) single crystal substrate. Figure 2(c) shows the in-plane magnetization hysteresis loops (M - H loops) measured at R.T. with $H \parallel [100]$ and $[110]$ of the Fe₄N crystal. In the specimen, magnetization saturates at $H = 400$ Oe and 700 Oe for [100] and [110], respectively. M_s of the epitaxial thin film was measured to be 1390 emu/cm³, which agreed with that of the bulk Fe₄N (1400 ~ 1500 emu/cm³) (e.g. Ref. 20) as well as the film (1470 emu/cm³) fabricated using molecular beam epitaxy.²¹⁾ However, the M_r / M_s ratio becomes less than 1 for [100]. That might be explained by an ultra-thin bcc-Fe initial growth layer, of which hard-axis ([110]) is parallel to

Fig. 4 (a) In-plane FMR spectra obtained by varying the azimuthal angle of the external field (ϕ_H) with respect to the [100] crystallographic axis of the epitaxial Fe₄N film. (b) Resonance field (H_r) as a function of ϕ_H . The open circles and solid line represent experimental data and calculated results using Eq. (3) with $\theta_M = \theta_H = 90$ deg. (c) Experimentally obtained line-width (ΔH) as a function of ϕ_H .

the [100] of Fe₄N.

Figure 3(a) shows the torque-curve of the 100-nm-thick Fe₄N film. H was rotated in the (001) plane. A four-fold symmetric curve was observed with negligibly small uniaxial anisotropy. Judging from the curve, $\langle 100 \rangle$ and $\langle 110 \rangle$ correspond to the magnetization easy-axis and hard-axis, respectively, which was also confirmed by the M - H loops shown in Fig. 2(c).

Figure 3(b) shows the reciprocal of H ($1/H$) dependence of the K_1 values evaluated using the saturated torque amplitude for 50, 100, and 200-nm-thick epitaxial Fe₄N films. The intrinsic K_1 was determined to be the value for $1/H = 0$, that is $H \rightarrow \infty$, with extrapolation of the least-squares fitting line so that domain walls could be extracted. K_1 for 50 nm thickness was half the values of K_1 for 100 nm and 200 nm thicknesses, which were similar. These results imply that the Fe₄N with highly chemical ordering dominates the film with increasing thickness, and that biaxial anisotropy might be saturated at a thickness around 100 nm. The following FMR experiment was conducted for the 100-nm-thick Fe₄N film.

3.2 External field dependence of in-plane FMR at R.T.

Figure 4(a) shows the in-plane FMR spectra obtained by varying the azimuthal angle of the external field (ϕ_H) with respect to the [100] for the epitaxial Fe₄N film. The spectrum indicates the magnitude of microwave power absorption by magnetization precession, and a dispersion-type spectrum is observed by H modulation. From 0 to 180 deg., the spectra systematically shifted

Fig. 5 Typical FMR spectra ($|S_{11}|$) measured at R.T. using coplanar waveguide (CPW) and vector network analyzer (VNA) for (a) $H \parallel [100]$ and (b) $H \parallel [110]$. The spectra were fitted with dispersion and Lorentz functions. (c) Resonance frequency (f_r) as a function of H . The open circles and triangles correspond to experimental f_r with $H \parallel [100]$ and $[110]$. The lines are calculated f_r values obtained by varying azimuthal angle (ϕ_H) with respect to $[100]$. (d) Frequency linewidth (Δf) as a function of H . The solid lines are calculated Δf values using the equation: $2\pi\Delta f = 2\alpha_{\text{eff}}\gamma H_{\text{eff}}$.

depending on ϕ_H .

Figure 4(b) shows the ϕ_H dependence of H_r , which is defined as the intersection field at which the horizontal baseline of the background crosses the spectrum itself. The experimental data show that four-fold symmetric behavior such that $H_r = 910$ Oe (310 Oe) was observed at $\phi_H = \langle 110 \rangle$ ($\langle 100 \rangle$). We confirmed that a large (small) H_r was observed for the magnetic hard (easy) direction. This must imply the presence of biaxial magnetocrystalline anisotropy in the (001) plane. The FMR resonance condition in the presence of magnetocrystalline anisotropy was considered by Kittel for the case in which H was applied parallel to the film plane.²²⁾ Using Eqs. (1), (2), and (3) with $\theta_M = \theta_H = 90$ deg. and $\phi_H = \phi_M$, the modified FMR relationship is given by

$$\left(\frac{2\pi f_r}{\gamma}\right)^2 = \left[H_r + \frac{K_1}{M_s}(2 - \sin^2 2\phi_M) + 4\pi M_s\right] \left[H_r + \frac{2K_1}{M_s} \cos 4\phi_M\right], \quad (8)$$

where f_r and γ represent the resonance frequency of 9.89 GHz and the gyromagnetic constant in CGS units, $\gamma = g\mu_B/2m_e c = 17.588 \times g$ MHz/Oe, respectively. Here, we assumed that the perpendicular anisotropy was omitted ($4\pi M_{\text{eff}} = 4\pi M_s - 2K_1/M_s \approx 4\pi M_s$) because our specimen had sufficiently large M_s compared to K_1 . With $M_s = 1390$ emu/cm³, $K_1 = 2.3 \times 10^5$ erg/cm³ and $g = 2.09$, the calculation showed good agreement with the maximum and minimum H_r of the experimental data. M_s and K_1

had similar values as observed in the M - H loop and the torque-curve as mentioned above. The slight difference in intermediate H_r might be due to a slightly different azimuthal angle between ϕ_H and ϕ_M .

Figure 4(c) shows the ϕ_H dependence of ΔH . ΔH is defined as the field difference at positive and negative FMR peaks. Likely to the H_r behavior, the experimental data show four-fold symmetric properties; $\Delta H = 240$ Oe (175 Oe) was observed at $\phi_H = \langle 110 \rangle$ ($\langle 100 \rangle$). Converting ΔH to α_{eff} using the Gilbert damping equation: $\Delta H = (2/\sqrt{3})\alpha(2\pi f)/g$, α_{eff} was estimated to be 0.062 and 0.045 for $\langle 110 \rangle$ and $\langle 100 \rangle$, respectively.

3.3 Frequency dependence of in-plane FMR via CPW and VNA at R.T.

To confirm anisotropic α_{eff} in the epitaxial Fe₄N film observed using ESR, α_{eff} in the film plane was also investigated by means of CPW and VNA by scanning the microwave frequency. Figures 5(a) and 5(b) show the reflection intensity spectra ($|S_{11}|$) as a function of the applied frequency (f) with $H \parallel [100]$ and $H \parallel [110]$, respectively. Since $|S_{11}|$ included the reflection component attributable to the CPW itself as background ($|S_{11}(\text{b.g.})|$), after applying sufficiently large H in the film plane so that spectrum attributable to FMR component disappears, $|S_{11}(\text{b.g.})|$ was subtracted from $|S_{11}|$. The imaginary part of S_{11} , corresponding to the phase-lag from the input power, dominated the spectra in our specimen. For this reason, in order to determine the resonance frequency (f_r) and the frequency line-width (Δf), we performed fitting using the following Lorentz and its derivative functions, namely, the dispersion function with proportional constants (P, Q).

$$|S_{11}| = P \frac{\Delta f}{(f - f_r)^2 + \Delta f^2} + Q \frac{\Delta f(f - f_r)}{[(f - f_r)^2 + \Delta f^2]^2}. \quad (9)$$

Both of the experimental spectra were reproduced by Eq. (9). Though the dip signals appeared at $f = 20$ GHz for $H \parallel [100]$ and $f = 17$ GHz for $H \parallel [110]$, this might be attributed to the CPW and not the FMR component, since we confirmed that f_r of the dips were independent of H in the experiments.

Figure 5(c) shows a summary of f_r as a function of H , the directions of which were $H \parallel [100]$ and $[110]$. In the case of $H \parallel [100]$, f_r monotonically increased from $f_r = 7.5$ GHz to 20 GHz with increasing H . On the other hand, in the case of $H \parallel [110]$, after the parabolic dependence appeared up to $H = 750$ Oe, a monotonic increase appeared. The relationship between f_r and H in the specimen is given by Eq. (8). The solid lines show the calculation results for $\phi_H = 0$ ($H \parallel [100]$), 15, 30, and 45 deg. ($H \parallel [110]$). The same parameters including $M_s = 1390$ emu/cm³, $K_1 = 2.3 \times 10^5$ erg/cm³ and $g = 2.09$ are substituted. However, Eq. (8) is valid only when H is greater than $H_{K1} = (2K_1/M_s)\cos 4\phi_M$, in which case the

Fig. 6 (a) Resonance field (H_r) and (b) linewidth (ΔH) of the out-of-plane FMR as a function of the polar external field angle (θ_H) with respect to [001]. Each line represents the calculated results with the parameters indicated in the panels. The open circles and triangles correspond to experimental data for $\phi_H = 0$ and 45 deg., respectively, as schematically shown in (c).

magnetization direction can maintain $\phi_M = \phi_H$. Modifying Eq. (8), the FMR condition formula below H_{K1} is given by

$$\left(\frac{2\pi f_r}{\gamma} \right)^2 = \left[-H_r + \frac{K_1}{M_s} (2 - \sin^2 2\phi_M) + 4\pi M_s \right] \left[-H_r + \frac{2K_1}{M_s} \cos 4\phi_M \right], \quad (10)$$

and this result is shown by the dashed lines in Fig. 5(c). The same value of f_r can be obtained for $H = 0$ Oe, which means the FMR is achieved by H_{K1} pointing to [100], easy-axis. The calculation revealed that the experimental f_r for $H \parallel [100]$ was fitted well with $\phi_H = 0$ for all H . In the case of $H \parallel [110]$, only f_r above $H = 900$ Oe could be reproduced by the calculation. Below 900 Oe, it is possible to interpret this as ϕ_M gradually changing from 45 to 0 deg. because of H_{K1} with decreasing H , and finally reaching complete 0 deg. ([100] easy-axis) at $H = 0$, which means that FMR condition is achieved by only H_{K1} at $H = 0$. Note that $H_r = 900$ Oe (250 Oe) was observed at $f = 9.89$ GHz, which was consistent with the H_r obtained by ESR.

Figure 5(d) shows Δf as a function of H , the direction of which is $H \parallel [100]$ and $[110]$. Δf is defined as the FWHM of the Lorentz function expressed by Eq. (9) after fitting the spectra. We can confirm that a large Δf for [110] was observed for all values of H . Here, α_{eff} was evaluated using the simple FMR equation: $2\pi\Delta f = 2\alpha_{eff}\gamma H_{eff}$, where H_{eff} includes H , $4\pi M_s$, and H_{K1} . By fitting the experimental data where a constant Δf was

observed in the high H region, α_{eff} for [100] and [110] were determined to be 0.033 and 0.046, respectively, with $M_s = 1390$ emu/cm³, $K_1 = 2.3 \times 10^5$ erg/cm³, and $g = 2.09$. Consistent with the data obtained by ESR, α_{eff} of [100] is obviously larger than that of [110] in our specimen at R.T.

3.4 Anisotropic intrinsic damping constant and its temperature dependence

In order to extract α from α_{eff} , experiments on out-of-plane FMR were conducted. Figure 6(a) shows the θ_H dependence of H_r at R.T. when H rotates in the (010) plane ($\phi_H = 0$ deg.) and the (110) plane ($\phi_H = 45$ deg.), respectively. Because of the H limitation of our ESR machine, data below $\theta_H = 5$ deg are not shown. Both H_r increased with decreasing θ_H due to the demagnetization field generated perpendicular to the film plane. H_r was enhanced for $\phi_H = 45$ deg. compared to H_r for $\phi_H = 0$ deg. for all θ_H . By substituting $\phi_H = 0$ and 45 deg. in the modified Eq. (3), the calculation was conducted with $M_s = 1300$ emu/cm³, $K_1 = 2.2 \times 10^5$ erg/cm³ and $g = 2.09$. The parameters were consistent with values obtained in the in-plane FMR. The calculations reveal that the difference in H_r must be due to the presence of K_1 .

Figure 6(b) shows the θ_H dependence of ΔH at R.T. for $\phi_H = 0$ and 45 deg. The calculation of ΔH was also conducted assuming the extrinsic contributions mentioned in Eqs. (4), (5), (6), and (7). Except for the less than $\phi_H = 20$ deg. in the $\phi_H = 45$ deg. case (7 deg. in the $\phi_H = 0$ deg. case), we found that the calculations agreed with experimental data. In prior research on out-of-plane FMR in Ni-Fe, Co-Fe-B, Co₂MnSi, and even poly-crystalline Fe₄N, the whole ΔH was successfully fitted by calculation.^{6,18,23-26} Even though the mechanism is not yet fully identified, judging from the fact that the unmatched fitting result appears stronger for $\phi_H = 45$ deg., domain structures in small θ_H are considered as a possible mechanism because of K_1 in the epitaxial Fe₄N film. Such an additional resonance has been presumed to be domain structure excitation below magnetic saturation in crystals with strong magnetic anisotropy.^{27,28} For example, Nagamiya examined the additional resonance observed in Fe₃O₄ where the magnetic anisotropy has tetragonal symmetry.²⁹ Therefore, we performed the fitting for the large θ_H region, where a single domain structure could be expected.

Figure 7 shows the temperature dependence of α determined by data fitting of the out-of-plane FMR experiments mentioned above for $\phi_H = 0$ and 45 deg. The error bars attached to each data point show the value of 2σ assuming that our experiments and analysis events follow the normal distribution. We can confirm that the distributions of magnetic-properties-related extrinsic damping components such as $\Delta 4\pi M_{eff}$, $\Delta \theta_H$,

Fig. 7 Temperature dependence of the intrinsic magnetic damping constant (α) for $\phi_H = 0$ ($H // [100]$) and 45 deg. ($H // [110]$). Error bars (2σ) have been included. Solid and dashed lines are visual guides.

and ΔK_1 were successfully excluded, therefore α reflects the damping phenomena for our material itself. Note that anisotropic α was clearly observed at R.T., that is, α for $\phi_H = 45$ deg. was larger than that for $\phi_H = 0$ deg., and both α were enhanced with decreasing T . Surprisingly, α for $\phi_H = 0$ deg. exceeded that for $\phi_H = 45$ deg. at $T \approx 180$ K. The phenomenon has not yet been reported. To understand the principles such as $T \approx 180$ K as well as the magnitude relation between α for [100] and [110], the first principle numerical calculation assuming spin-orbit interaction of Fe_4N is indispensable.

4. Discussion

Anisotropic α , in other words, different α depending on the magnetization direction with respect to the lattice, was clearly observed in the epitaxial Fe_4N film. To discuss the mechanism for the occurrence of anisotropic α , we examined the numerical calculation of α for the Fe_4N crystalline model (bulk) based on the torque correlation model developed by Kambersky and Gilmore et al.^{16,17} Figure. 8 shows the calculation result conducted for both cases in which the magnetization direction points along [100] and [110] using the linear-muffin-tin orbital (LMTO) method under the local spin density functional approximation (LSDA).³⁰ The actual form of α in our calculation is

$$\alpha \propto \sum_k \sum_{m,n} \left(|\Gamma_{mk,nk}|^2 \frac{\hbar/2\tau}{(E_F - E_{mk})^2 + (\hbar/2\tau)^2} \frac{\hbar/2\tau}{(E_F - E_{nk})^2 + (\hbar/2\tau)^2} \right) \quad (11)$$

where $\Gamma_{mk,nk}$, E_F , and $E_{mk(nk)}$ represent the matrix element of the torque due to spin-orbit interaction, the Fermi energy, and the electron energy of the m (n) band at k -space, respectively. Since τ represents the electron lifetime attributable to the electron-lattice interaction, $1/\tau$ of the lateral axis indicates temperature. We can find two facts from the calculation results: first, both α decrease exponentially with increasing temperature;

Fig. 8 Numerical calculation of intrinsic damping constant (α) as a function of the inverse of electron lifetime ($1/\tau$) of the epitaxial Fe_4N . The solid and dashed lines correspond to the results when the external field (H) was applied along [100] and [110].

second, α for $H // [110]$ is larger than α for $H // [100]$ above $1/\tau \approx 0.005 \times 10^{14} \text{ s}^{-1}$, although its magnitude relation is reversed below $1/\tau$. The interpretation for the first result is that the monotonic decrease with $1/\tau$ indicates that the events of excitation and relaxation of electrons take place within the same band ($m = n$), corresponding to the “intra-band” transition. The second result can be explained by considering that Eq. (11) is constituted of two Lorentzian spectra with energy broadening of $1/\tau$. Therefore, if the states connected by the matrix elements $\Gamma_{mk,mk}$ are sharply distributed around the E_F , a small value of $1/\tau$ is superior for α , and then we infer that this may be the case for $H // [100]$ situation below $1/\tau \approx 0.005 \times 10^{14} \text{ s}^{-1}$. Contrarily, in the case of $H // [110]$, it is considered that the energy distribution of the state crucial for $\Gamma_{mk,mk}$ is wide compared to the $H // [100]$ case. In the presence of spin-orbit interaction, the electron band structure changes depending on the magnetization direction and then the effects of energy broadening $1/\tau$ on the excitation spectra may be different. Based on our calculations, the results lead us to conclude that the experimental anisotropic α and its T dependence reflect the difference in electron band structures of Fe_4N depending on H and T .

However, T at which α for $H // [100]$ exceeds that for $H // [110]$ (crossover T) comes out to be extremely low in the calculation. In the electrical conduction model, resistivity (ρ) is generally given by

$$\rho = \frac{m_e}{ne^2} \frac{1}{\tau}, \quad (12)$$

where m_e , n , and e represent the electron mass, the free-electron density, and the elementary charge, respectively. Using the formula, $1/\tau \approx 0.005 \times 10^{14} \text{ s}^{-1}$ is converted to $\rho = 0.018 \mu\Omega\text{cm}$ with $m_e = 9.1 \times 10^{-31} \text{ kg}$, $n = 1 \times 10^{29} \text{ m}^{-3}$, and $e = 1.6 \times 10^{-19} \text{ C}$. According to the experimental data for the T dependence of ρ in the epitaxial Fe_4N film,³¹ $\rho = 0.018 \mu\Omega\text{cm}$ cannot be

explained within the finite T regime since even the residual ρ is 100 times as high as the converted $\rho = 0.018 \mu\Omega\text{cm}$. Though further investigation is warranted, in our speculation, the difference in temperature might be attributed to the actual τ of the Fe_4N film possibly being extremely short due to scattering by impurities and/or dislocations in the thin films.

5. Summary

Studies on in-plane FMR and out-of-plane FMR were carried out for 100-nm-thick epitaxial Fe_4N films. Large (small) H_r and ΔH were observed for $H \parallel [110]$: hard-axis ($H \parallel [100]$: easy-axis). Calculation of H_r using a modified Kittel's function agreed with the experimental H_r with reasonable parameters: $M_s = 1390 \text{ emu/cm}^3$, $K_1 = 2.3 \times 10^5 \text{ erg/cm}^3$ and $g = 2.09$. As a comparison, in-plane FMR using CPW and VNA was performed at R.T., and it was confirmed that the above three parameters were also valid to explain the experimental data. Based on the in-plane FMR study at R.T., the presence of anisotropic α was reliably observed in the epitaxial Fe_4N film. Using the out-of-plane FMR, temperature variation of the anisotropic α was investigated. We found that α for $\phi_H = 0 \text{ deg.}$ was significantly enhanced with decreasing T , and exceeded that for $\phi_H = 45 \text{ deg.}$ at $T = 180 \text{ K}$. According to our numerical calculation of α as a function of $1/\tau$, the observed anisotropic α was at least qualitatively explained by the change in the electronic band structure depending on the magnetization direction.

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