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Observation of the intrinsic Gilbert damping constant in Co/Ni multilayers independent of the stack number with perpendicular anisotropy

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We investigate the intrinsic Gilbert damping constant in perpendicular magnetic anisotropy Co/Ni multilayer system by means of an all-optical method. We find that the intrinsic Gilbert damping constant does not depend on the stack number and the perpendicular magnetic anisotropy when the magnetic field is high enough. In contrast, the extrinsic Gilbert damping is strongly correlated with the inhomogeneous anisotropy distribution in the low-field regime, as observed in magneto-optical images. Intriguingly, the intrinsic Gilbert damping is consistently reduced with decreasing length scale in the measurements, providing a concrete means to determine the intrinsic Gilbert damping.

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Perpendicular magnetic anisotropy (PMA) architectures are desired to improve memory density and to overcome the thermal stability problems in spin-transfer torque magnetic random access memory. The PMA architecture typically has a smaller switching current density $J_s$ because it is free from demagnetization fields. However, PMA materials typically have larger Gilbert damping constants than typical in-plane ferromagnetic materials, leading to larger values of $J_s$ because $J_s$ is proportional to the Gilbert damping constant $\alpha$. Therefore, the determination of the Gilbert damping constant of PMA materials is important. Furthermore, it is widely accepted that $\alpha$ is closely related to the spin-orbit interaction, which is also a main source of PMA. Thus, the relationship between the Gilbert damping and PMA has been widely investigated. Recently, this connection has been supported by many research groups for various PMA systems, such as Pt/Co/Pt (Ref. 4) and [Co/Pd]$_N$. In contrast, other groups have reported that there are no correlations between the Gilbert damping and PMA in Co$_{60}$Fe$_{10}$/Ni, Co$_{90}$Fe$_{10}$/Pd, and Ta-buffered Co/Ni (Ref. 9) multilayer systems.

In another aspect, the determination of the intrinsic $\alpha$ in a multilayer system is controversial. Generally, it is believed that $\alpha$ value increases with an increase in the stack number $N$ due to the plausible increase of the roughness and defects with an increase in the number of interfaces. However, some recent reports have shown that $\alpha$ is independent of $N$ (Ref. 9) or inversely correlated to $N$. Hence, the understanding of $\alpha$ mechanism remains incomplete, especially in multilayer system with different numbers of stacks.

Furthermore, the precise determination of $\alpha$ is an elaborate procedure that depends on the samples and the measurement techniques. Usually in real samples, spin waves are additionally generated from the non-magnetic or magnetic inhomogeneity such as the roughness, defects, the unsaturated magnetization state, or the anisotropy distribution. Therefore, the measured $\alpha$ includes not only the intrinsic $\alpha$ but also extrinsic $\alpha$ from the additional spin wave contribution, such as the apparent dephasing of various excited spin-wave precession frequencies and the additional direct and indirect relaxation via the interaction among the spin waves. The apparent dephasing is easily excluded by applying a strong enough external magnetic field because the strong magnetic field aligns the spins in one direction and reduces the incoherent precessional motion coming from the inhomogeneous anisotropy and demagnetization field distribution. Interestingly, the extent of the incoherent precessional motion can also be reduced with decreasing length scale in the measurement.

In this letter, we studied $\alpha$ in [CoNi]$_N$ multilayer system, which have a low switching current and strong thermal stability, while systematically controlling PMA with changing $N$ from 6 to 27. In this sample system, we measured $\alpha$ as a function of both $N$ and PMA by using an all-optical method. To identify the apparent effect, we investigated $\alpha$ by varying the external magnetic field $H$. Intriguingly, it was also found that the all-optical measurement technique with decreasing beam size can reduce the long-range inhomogeneity effect remarkably.

The Co/Ni multilayer films were prepared on a Si/SiO$_2$ (001) substrate by DC magnetron sputtering with a base pressure of $5 \times 10^{-9}$ Torr. The stack structure of the sample series is Si/SiO$_2$/Al$_2$O$_3$/(100 Å)/Ti(50 Å)/Co(3 Å)/[Ni(7 Å)/Co(3 Å)]$_N$/TaN(50 Å). We changed $N$ as follows: increase by three consecutively from 6 to 27. We used a polar magneto-optical Kerr effect (p-MOKE), a vibration sample magnetometer (VSM), and a superconducting quantum interference device (SQUID) to measure the magnetic properties of the samples. Figure 1(a) shows the hysteresis loops measured by p-MOKE at room temperature. By utilizing the in-plane VSM and SQUID, the effective PMA energy density $K_{eff}$ is estimated from $K_{eff} = M_sH_s/2$, where $M_s$ is the saturation magnetization and $H_s$ is the saturation magnetic field in the...
The $K_{\text{eff}}$ value is plotted as a function of $N$ in Fig. 1(b). In general, the $K_{\text{eff}}$ increases as the $N$ increases. Because magnetic inhomogeneity is an important issue in the PMA samples, we also investigated the magnetic inhomogeneity using a magneto-optical microscope magnetometer (MOMM).

Field-swept domain reversal patterns are visualized from the initial saturated state with $N = 6$, 15, and 24 in Fig. 1(c). This figure shows the domain reversal pattern in color codes with the field sweeping rates of 0, 5, and 15 Oe/s near the coercive field. As $N$ increases, the field-swept domain reversal pattern becomes more nucleation-dominant which indicates increased anisotropy-distribution. Also, the sweeping field range increases concomitantly, which implies growth of local coercivity distribution or $\Delta H_c$ by the accumulation of the number of stacks.

Measurements of $x$ were carried out by using time-resolved MOKE. A Kerr-lens mode-locked Ti:Sapphire oscillator generates laser pulses with a center wavelength of $\sim$800 nm. The laser pulse has a pulse width of 30 fs and a repetition rate of 82 MHz. We doubled the frequency of the probe beam used here with a BBO crystal; thus, the laser beam is split into a pump beam and a probe beam. The probe beam intensity is much weaker than that of the pump beam (1:200). The pump and probe laser pulses are fed into a $50 \times (0.5$ numerical aperture) polarization-conserving objective lens, as shown in Fig. 2(a). The probe beam is incident along the normal axis of the sample. The spot diameters of the pump and probe beams are $\sim 2 \mu m$ and $\sim 1 \mu m$, respectively. The pump fluence $F$ is in the range of 4–20 mJ/cm$^2$. To check thermal effect from the pump beam, we measured dynamic Kerr loops with $N = 6$ at $F = 8$ mJ/cm$^2$ (not shown here). We found out that the transient heating and cooling is negligible unless the experimental error really matters. In addition, compared the loop between with and without the pump beam, we estimated that the sample was heated up to $\sim 360$ K. This result is in good agreement with the previous report. To reduce the noise, we used balanced detection and lock-in amplifier filtering via mechanical chopping on the pump beam at 1300 Hz. The external magnetic field was applied with the angle $\theta_H$ of 60° from the normal direction of the film plane.

Figure 2(b) shows the time-resolved MOKE data with $N = 6$–27 at $H = 5.0$ kOe, $\theta_H = 60^\circ$, and $F = 8$ mJ/cm$^2$. In the Kerr signals, the step-like decrease at 2 ps reflects the demagnetization, and the damped harmonic oscillation after a few ps reflects the precessional motion. The Kerr signals are fitted to a damped-harmonic function expressed as
$\theta = \theta_0 + A e^{-t/\tau_0} + B e^{-t/\tau_0} \sin(2\pi f t + \phi)$,$^19$ where the first term \(\theta_0\) represents the offset background, the second term is the remagnetization with relaxation time \(\tau_0\), and the final term is the precessional motion with the resonance frequency \(f\), the relaxation time \(\tau\), and the initial phase \(\phi\). The solid curves in Fig. 2(b) are the fitted lines.

Figure 2(c) shows \(f\) as a function of \(H\) with \(N = 6, 15\), and 24. Here, \(\theta_T = 60^\circ\) and \(F = 12\) mJ/cm$^2$. In this figure, the value of \(f\) increases as \(H\) increases. In order to understand this tendency, we fitted the data with the Kittel equation, as shown by the solid curves in Fig. 2(c). In the samples with a large \(N\), the polar hysteresis loops in Fig. 1(a) do not show a square sharp shape. This suggests that the second-order anisotropy \(K_2\) exists, and the \(K_2\) value increases as the total Co thickness increases.$^20$ Therefore, we consider the following Kittel equation with \(K_2\): $\theta(f) = \gamma / 2\pi \sqrt{H_1 + H_2}$, where $H_1 = H \cos(\theta_T - \theta) + H_{\text{eff}}^1 \cos^2 \theta - H_{\text{eff}}^2 \cos^4 \theta$ and $H_2 = H \cos(\theta_T - \theta) + H_{\text{eff}}^1 \cos^2 \theta + H_{\text{eff}}^2 \sin^2 \theta - \cos^4 \theta$. Here \(\gamma\) is the gyromagnetic ratio defined as \(\gamma = \gamma g \mu_B / h\). The equilibrium angle \(\theta\) is calculated using the energy equilibrium relationship of \(\sin^2 \theta = 2H/H_{\text{eff}}^1 \sin(\theta_T - \theta) + (H_{\text{eff}}^1 / H_{\text{eff}}^2) \sin^2 \theta \), where $H_{\text{eff}}^1 = 2K_{\text{eff}} / M_s + 4K_{\text{eff}} / M_s - 4xM_s$ and $H_{\text{eff}}^2 = 4K_{\text{eff}} / M_s$.$^19$ We used the $H_{\text{eff}}^1$ value from the experimental data measured by the in-plane VSM and SQUID. By considering $H_{\text{eff}}^1$, we obtained fairly well-fitted matching curves, as shown in Fig. 2(c). As expected, the fitted $H_{\text{eff}}^1$ values increase notably as \(N\) increases. We obtained $g$ values that were close to the \(g\) value of bulk Co. The final best fit values of $\gamma$ and $H_{\text{eff}}^1$ are summarized in Table I.

With these fitting parameters, we first estimated the possible single effective damping constant of $x_{\text{eff}}$ by fitting the experimental data of $\tau^{-1}$ as a function of \(H\) when $\tau^{-1} = \gamma x_{\text{eff}} / 2 (H_1 + H_2)$. The estimated single damping constants are $0.024 \pm 0.001$, $0.026 \pm 0.002$, and $0.031 \pm 0.005$ for $N = 6, 15$, and 24, respectively. The value of $x_{\text{eff}}$ increases slightly with increasing \(N\), and the corresponding error also increases. For $N = 6$ shown with the black square symbols in Fig. 2(d), the experimental values of $\tau^{-1}$ are well fitted to the $\tau^{-1}$ curve using a single instance of $x_{\text{eff}}$. However, as $N$ becomes large, the fitted curves deviate more from the experimental data in the small-field regime. Therefore, the error ranges are very large, and the $x_{\text{eff}}$ values are far from intrinsic value. Silva et al.$^6$ showed that $x$ is well-described by the weighted average of the individual contributions of $x$ from the various atomic species in Co$_{66}$Fe$_{34}$Ni multilayers and alloys. Concretely, by using bulk values, Silva et al. calculated $\frac{c}{x_{\text{ave}}}$ and reported that $\frac{c}{x_{\text{ave}}}$ is in excellent agreement with the experimental data. We calculate $\frac{c}{x_{\text{ave}}}$ in our Co/Ni multilayers by using the same method and the values of bulk $c$, Co, and Ni.$^5$ The estimated $\frac{c}{x_{\text{ave}}}$ value is $\sim 0.22$, which is well matched with the $x_{\text{ave}}$ value of $N = 6$.

Figure 2(e) shows the experimental effective damping parameter $x_{\text{eff}}$ as a function of $H$. The value of $x_{\text{eff}}$ is obtained from the simple approximate equation of $x_{\text{eff}} = (2\pi f \tau)^{-1}$ using the experimental data of $f$ and $\tau$ in Fig. 3.$^{22,23}$ Here, $x_{\text{eff}}$ accounts not only for the intrinsic $x$ but also for the extrinsic damping such as spin pumping and additional spin wave contribution. Therefore, the intrinsic $x$ is smaller than the $x_{\text{eff}}$ values.$^{11,19}$ The value of $x_{\text{eff}}$ decreases as the external field increases in Fig. 2(e). Because the effective damping changes as the applied $H$ is varied, the change in $x_{\text{eff}}$ originates from the additional spin-wave damping or the apparent effect of the incoherent spin waves, which easily arise in samples with an inhomogeneous anisotropy distribution in a low $H$.$^{11,24}$ This explains why the variation of $x_{\text{eff}}$ for $N = 24$ is larger than that for $N = 6$ because the inhomogeneity of $K_{\text{eff}}$ for $N = 24$ is larger than that for $N = 6$, as shown in Fig. 1(c). Clearly, the $x_{\text{eff}}$ values converge to the nearly identical minimum values under a high external magnetic field irrespective of the stack number $N = 6, 15$, and 24. This value is similar to $\sim 0.22$ obtained by $x_{\text{ave}}$.

As described in the supplementary material,$^{25}$ we checked the effect of magnetization tilted angle on $x_{\text{eff}}$ for example, by short wavelength spin waves. However, in our samples, the influence of short wavelength spin waves on damping is less operative when $H$ is strong enough, which is well matched with the previous report.$^{22}$

To analyze this in more detail with respect to $N$ as well as $K_{\text{eff}}$, we measure $x_{\text{eff}}$ under weak and strong $H$ for various $N$. Figure 3(a) shows the $x_{\text{eff}}$ dependences on $N$ at 3.2 kOe and 5.0 kOe when $\theta_T = 60^\circ$. When $H$ is not strong enough (3.2 kOe), $x_{\text{eff}}$ increases as $N$ increases noticeably after $N = 9$. However, $x_{\text{eff}}$ shows nearly a constant value with increasing $N$ at sufficiently high external magnetic fields.

**TABLE I.** Fitted and extracted values obtained in Co/Ni multilayer films when $N = 6, 15$, and 24. The $H_{\text{eff}}^1$ values are determined by VSM and SQUID measurements while the other values are obtained by the fitting procedure.

<table>
<thead>
<tr>
<th>$N$</th>
<th>$H_{\text{eff}}^1$ (Oe)</th>
<th>$H_{\text{eff}}^2$ (Oe)</th>
<th>$\gamma$ (Mrad/s Oe)</th>
<th>$g = \gamma h / \mu_B$</th>
</tr>
</thead>
<tbody>
<tr>
<td>6</td>
<td>1080</td>
<td>0 ± 570</td>
<td>20.9 ± 0.7</td>
<td>2.38 ± 0.08</td>
</tr>
<tr>
<td>15</td>
<td>4200</td>
<td>2350 ± 100</td>
<td>19.0 ± 0.2</td>
<td>2.16 ± 0.02</td>
</tr>
<tr>
<td>24</td>
<td>5790</td>
<td>4220 ± 40</td>
<td>18.8 ± 1.3</td>
<td>2.14 ± 0.14</td>
</tr>
</tbody>
</table>

FIG. 3. (a) A dependence of $x_{\text{eff}}$ on the stack number $N$ and (b) the effective PMA energy density $K_{\text{eff}}$. Red and black solid circles represent the data of the low external magnet field $H = 3.2$ kOe and the high external magnet field $H = 5.0$ kOe, respectively.
Therefore, our result shows that the intrinsic $\zeta$ is indeed independent of $N$ from 6 to 27 within the experimental error. This calls attention to the other possible relationships between the PMA and the Gilbert damping.

To elucidate the relationship between the PMA and $\zeta_{\text{eff}}$, we plot $\zeta_{\text{eff}}$ as a function of $K_{\text{eff}}$, as shown in Fig. 3(b). The value of $\zeta_{\text{eff}}$ increases rapidly when the anisotropy field level exceeds the external magnetic field. However, the value of $\zeta_{\text{eff}}$ is independent of $K_{\text{eff}}$ under a high external magnetic field. This means that the intrinsic $\zeta$ is independent of $K_{\text{eff}}$ in our Ti-buffered Co/Ni multilayer system.

To verify the inhomogeneity effect at low field in another way, we first carried out experiments by varying the magnification of the objective lenses at $10 \times$, $20 \times$, $50 \times$, and $100 \times$ with numerical apertures of 0.25, 0.4, 0.5, and 0.95, respectively. Figure 4(a) shows the value of $\zeta_{\text{eff}}$ as a function of the probe beam spot area. We used a pump power of 20 mW, in the front of the objective lens and a weak external magnetic field of $H = 3.0$ kOe at $\theta_{\text{ill}} = 60^\circ$. Compared to the result of $H = 3.0$ kOe, similar but weak trend (not shown here) is obtained in a strong external magnetic field of $H = 5.0$ kOe. Surprisingly, we noted an increasing relation between $\zeta_{\text{eff}}$ and the probe beam area. Since varying the magnification changes not only the probe area but also $F$ in our experimental configuration, we also checked $\zeta_{\text{eff}}$ while varying the pump fluence with fixed magnification of $50 \times$ lens. As shown in Fig. 4(b), $F$ has no clear relationship to $\zeta_{\text{eff}}$. In addition, $\zeta_{\text{eff}}$ with $N = 24$ was significantly decreased compared to that of $N = 6$. This means that the probe beam area influences the determination of $\zeta_{\text{eff}}$ in addition to the strength of the external field. This phenomenon can be consistently explained by the reduced contribution from the small length-scale excitation and detection of the apparent dephasing or the spin-wave damping, which comes from the inhomogeneous $K_{\text{eff}}$ distribution, as witnessed in the domain evolution pattern in Fig. 1(c). This implies that the smaller beam spot pump-probe technique further mitigates the apparent dephasing or the additional spin-wave damping.

It has been reported that $\zeta$ has a proportional relationship with PMA in Co/Pd and Co/Pt systems. In these systems, spin pumping causes by the Pt or Pd layers which have strong spin orbit coupling and $d$-$d$ hybridization occurs at the Co atomic layer in contact with the Pd or Pt layer. Contrary to the interfacial spin pumping effect, the $\zeta$ in our experiment is not inversely proportional to the ferromagnetic layer thickness, as shown in Fig. 3(a). Also, the spin-orbit coupling constant of Ti is more than 10 times weaker than that of Pd or Pt. Therefore, we believed that the spin-pumping effect is negligible in our system. The $3d$-$3d$ hybridization is much weaker than $3d$-$4d(5d)$ hybridization. In addition, PMA is caused by not only magnetocrystalline anisotropy but also shape anisotropy which arises from the dipole-dipole interaction in Co based multilayer system. Therefore, there is no noticeable correlation between the PMA and the intrinsic $\zeta$ in our samples. Also, we could not find any noticeable relationship between the intrinsic $\zeta$ and the sample properties such as $N$ and magnetic inhomogeneity. This indicates that the intrinsic $\zeta$ is well determined by the average damping of the atoms and layers, where the spins are coupled by strong exchange interactions and precessing in unison. Therefore, the highly local characteristic mainly contributes to the intrinsic $\zeta$ in this system.

In conclusion, we systematically studied the Gilbert damping constant $\zeta_{\text{eff}}$ as a function of the probe beam spot area with various degrees of magnification by an objective lens at $10 \times$, $20 \times$, $50 \times$, and $100 \times$ when $H = 3$ kOe at $\theta_{\text{ill}} = 60^\circ$. (b) The value of $\zeta_{\text{eff}}$ as a function of the pump fluence $F$. The black and red dashed lines are visual guide lines whose values are obtained from Fig. 2(e) with $H = 5.5$ kOe.

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25See supplementary material at http://dx.doi.org/10.1063/1.4795013 for the Gilbert damping constant dependences on the magnetization tilt angle.