Compositional Dependence of *g*-Factor and Damping Constant of GdFeCo Amorphous Alloy Films

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Time-domain magnetization dynamics of sputtered GdFeCo (30 nm) amorphous alloy films was measured by pump-probe method using high-power ultra-short pulse fiber laser. The effective g-factor $g_{\rm eff}$ and effective damping constant $\alpha_{\rm eff}$ of the GdFeCo films were estimated by using a numerical calculation of Landau–Lifshitz–Gilbert equation. The precessional frequency took a maximum near the magnetization compensation composition $C_{\rm M}$ of the GdFeCo, while the estimated $g_{\rm eff}$ and $\alpha_{\rm eff}$ increased around the angular momentum compensation composition $C_{\rm A}$. The compositional dependences of $g_{\rm eff}$ and $\alpha_{\rm eff}$ were roughly described by a mean-field model. The $g_{\rm eff}$ and $\alpha_{\rm eff}$ were also estimated from the ferromagnetic resonance (FMR) spectra, and the data from the FMR spectra agreed well with those from the pump-probe measurement except for the composition near $C_{\rm M}$. The FMR method was unable to excite the magnetization near $C_{\rm M}$ because of the small net magnetization.

***Index Terms*—Amorphous magnetic films, magnetic resonance, optical fiber lasers, rare earth alloys, time domain analysis.**

I. Introduction

UNDERSTANDING of the magnetization dynamics in magnetic thin films is one of the recent critical issues to develop ultra-high-speed magnetic sensors and memories. In general, magnetic loss increases at very high frequencies because of the occurrence of a magnetic resonance, and the loss limits the maximum driving frequency of the magnetic materials. In magnetic thin films, the resonance frequency will be raised to several GHz by the increase of the magnetization, i.e., the increase of the demagnetizing field. However, at present, high magnetic moment CoFe based alloy layers have been already used in many magnetic devices such as spin valve heads and magnetic random access memories, and drastic increase of the magnetization from CoFe alloys is not thought to be possible.

Ferrimagnetic rare-earth (RE) transition metal (TM) alloy systems are known to be possible to compensate their angular momentums even at the RE-TM composition having a small saturation magnetization due to the difference of the *g*-factors between RE and TM sublattice magnetic moments. The compensation of the angular momentum is reported to lead to the divergence of the effective *g*-factor as well as the effective Gilbert damping constant [1], [2], which will contribute to the fast magnetization reversal. Some works have already reported to investigate the divergence of the effective *g*-factor and Gilbert damping constant in RE-TM alloy system. Soohoo *et al.* have reported compositional dependence of the effective *g*-factor of GdFe alloys by using X-band ferromagnetic resonance (FMR), and pointed out the increase of $g_{\rm eff}$ near the magnetization compensation point [3]. Recently, spin dynamics of GdFeCo [4], [5] and GdCo [6] alloys were measured by pump-probe setup using an amplified Ti:sapphire laser, and the temperature dependences of the precessional frequency and effective damping factor have been reported. They observed the increase of Gilbert damping constant near the angular momentum compensation point [5], [6]. They also pointed out the increase of the effective *g*-factor due to the angular momentum compensation, but the temperature dependence of the anisotropy field was not taken into account in their estimations.

Development of the ultra-short pulse laser enables us to investigate time-domain spin dynamics of various magnetic thin films. Many reports utilize a Ti:sapphire ultra-short pulse laser to observe the precessional motion of the magnetization after the optical excitation with ~ 100 fs laser pulse [4]–[7]. Recently, ultra-short pulse fiber laser with fiber chirp pulse amplification system has attracting significant interest since it has characteristics of high output pulse energy of ~ 1 μJ–10 μJ, sub-watt-level average power, excellent stability [8], [9]. These characteristics are quite important for the pump-probe experiment to study time-domain spin dynamics of magnetic thin films, and the ultra-short pulse fiber laser is considered to be a compact and maintenance free practical light source for such researches. In this paper, we first describe the application of the ultra-short pulse fiber laser to excite the magnetization precession, and report the magnetization dynamics of GdFeCo with various compositions measured by pump-probe method using the fiber laser. The compositional dependence of *g*-factor and damping constant of GdFeCo films have been evaluated and discussed together with the data taken by ferromagnetic resonance (FMR).

II. Experimental Method

SiN (140 nm)/$\mathrm{Gd}_C(\mathrm{Fe}_{90}\mathrm{Co}_{10})_{100-C}$ (30 nm)/$\mathrm{Al}_{67}\mathrm{Cu}_{33}$ (100 nm)/SiN (10 nm)/oxidized Si substrate was prepared by using a magnetron sputtering system. The Gd content C of the GdFeCo layers was controlled by varying the number of Gd chips on the $\mathrm{Fe}_{90}\mathrm{Co}_{10}$ target. The SiN layer was sputtered to enhance Kerr rotation at $\lambda = 1560$ nm (wavelength of the fiber laser) as well as to protect the GdFeCo layer from oxidization. The 100 nm $\mathrm{Al}_{67}\mathrm{Cu}_{33}$ underlayer enhances the reflectivity of the sample. Hysteresis loops were measured by

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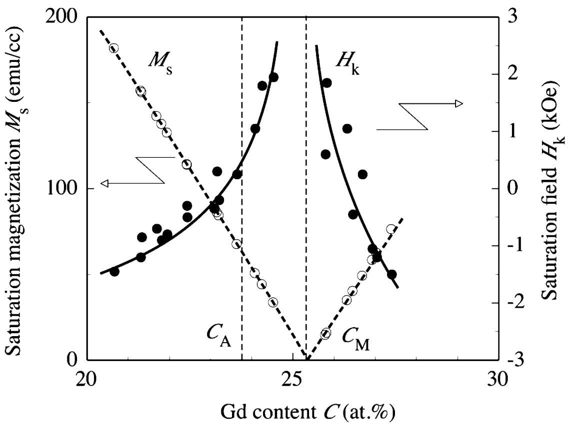

Fig. 1. Compositional dependence of magnetization and anisotropy field estimated from M–H loops of GdFeCo films. The Gd content was estimated from the magnetization of GdFeCo at room temperature by using a mean field approximation. The parameters of the mean field model are in the text.

an alternating gradient field magnetometer (AGM), and Kerr loops were checked by a polarized angle modulation method. Time-domain magnetization dynamics of GdFeCo was measured by pump-probe method using high-power fiber laser with a wavelength of 1560 nm, a pulse width of 1 ps, a maximum energy of 2 μJ/pulse, and a repetition frequency of 200 kHz. The pump and probe beams were focused onto GdFeCo films with a diameter of 60 μmϕ and 15 μmϕ, respectively. The probe beam was incident normal to the film surface and the polarization of the reflected probe beam was analyzed to monitor the perpendicular component of the magnetization, M_z, after the illumination of the pump beam. Typical fluences of the pump and probe beams are 2–4 mJ/cm^2 and 0.3 mJ/cm^2, respectively. During the measurements, an external field of 2 kOe was applied 45 deg from the film normal direction. Ferromagnetic resonance (FMR) spectra at room temperature were obtained by an X-band (9.2 GHz) spectrometer applying an external field parallel or perpendicular to the film plane. The maximum external field for FMR measurements was 13 kOe.

III. Results and Discussions

Fig. 1 shows compositional dependence of the magnetization M_s and anisotropy field H_k of the sputtered GdFeCo films. The Gd content can be estimated from the magnetization of GdFeCo at room temperature by using a mean field approximation [10]. For the estimation of Gd content, it was assumed that the exchange constants are $J_{TM-TM} = 6.3 \times 10^{-15}$ erg, $J_{Gd-TM} = -2.1 \times 10^{-15}$ erg, $J_{Gd-Gd} = 0.2 \times 10^{-15}$ erg, and angular momenta are $S_{TM} = 0.87$, $S_{Gd} = 3.5$, and g-factors are $g_{TM} = 2.16$, $g_{Gd} = 2.0$. Using these values, the angular momentum compensation composition C_A and magnetization compensation composition C_M at room temperature are estimated to be 23.8 at% and 25.2 at%, respectively, and these are indicated in Fig. 1. The calculated C_M agrees roughly with the experimental value of GdFe [10]. The anisotropy field H_k was estimated from the M–H loop, and positive anisotropy field means the easy axis perpendicular to the film plane. From Fig. 1 the H_k became positive in Gd content of 23.5–26.5 at%, and diverges at C_M.

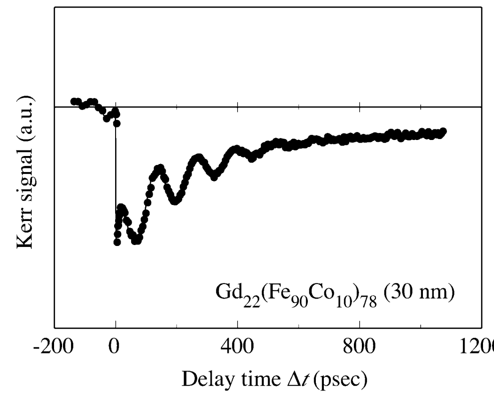

Fig. 2. Dynamic response of perpendicular component of magnetization M_z of the $Gd_{22}(Fe_{90}Co_{10})_{78}$ film after the excitation of the pump beam of 3.6 mJ/cm^2 produced by high-power ultra-short pulse fiber laser.

Fig. 2 shows a typical result of the measured magnetization dynamics of the GdFeCo film. The fluence of the pump beam was estimated to be 3.6 mJ/cm^2. Just as in the reported papers [4], [5], the M_z component shows a sudden drop at delay time of $\Delta t \sim 0$, due to the laser induced demagnetization. The drop of the M_z at $\Delta t \sim 0$ depends on the pump fluence and corresponds to about 20% of the saturation value M_s in this case. Major differences between the Ti:sapphire laser and the present fiber laser will be pulse width and wavelength. Even though the fiber laser has 10 times longer pulse width than Ti:sapphire laser, pump fluence necessary to start the precession of magnetization was almost the same as that of Ti:sapphire laser, which means the laser induced demagnetization is an adiabatic process. The wavelength of the present fiber laser of $\lambda = 1560$ nm is also different from that of Ti:sapphire laser of $\lambda \sim 800$ nm, but this difference may not be critical for the metallic samples.

After the laser induced demagnetization, a clear oscillation during the recovery of M_z was confirmed as shown in Fig. 2. The recovery process of GdFeCo can be described by the following Landau–Lifshitz–Gilbert (LLG) equation based on the mean field model:

$$\frac{d\mathbf{M}}{\mathrm{d}t} = -\gamma_{\mathrm{eff}}(\mathbf{M} \times \mathbf{H}) + \frac{\alpha_{\mathrm{eff}}}{M}\left(\mathbf{M} \times \frac{d\mathbf{M}}{\mathrm{d}t}\right) \quad (1)$$

where γ_{eff} and α_{eff} are effective gyromagnetic ratio and effective Gilbert damping constant, respectively [1]. Numerical calculation of LLG equation [11] yields $\gamma_{\mathrm{eff}} = g_{\mathrm{eff}}\,\mu_B/\hbar$ and α_{eff} for each GdFeCo film, which are estimated to be $g_{\mathrm{eff}} = 3.1$ and $\alpha_{\mathrm{eff}} = 0.28$ from Fig. 2.

The g_{eff} and α_{eff} can be evaluated from conventional FMR measurements. Fig. 3 shows compositional dependence of the resonance field H_{res} and line width ΔH_{pp} estimated from differentiated FMR spectra of GdFeCo films. The resonance field and line width were measured applying a magnetic field parallel and perpendicular to the film plane. For the samples having negative H_k (See Fig. 1), the resonance fields from in-plane FMR spectra were smaller than those from out-of-plane. Unfortunately, the clear absorption spectra were not obtained near C_M due to the small net magnetization of GdFeCo, since the resonance of magnetization is excited by an alternating magnetic field of the microwave. The line width ΔH_{pp} increased with the

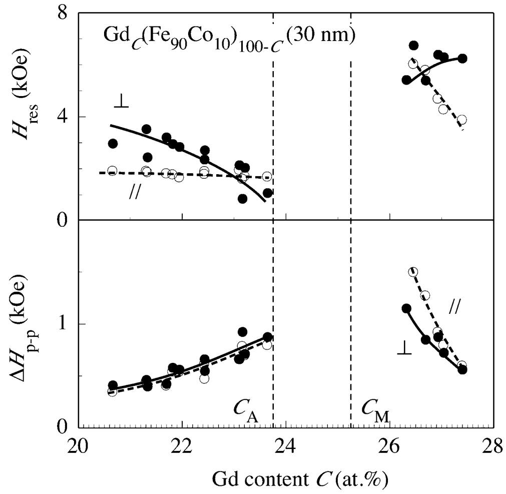

Fig. 3. Compositional dependence of the resonance field $H_{\rm res}$ and line width $\Delta H_{\rm pp}$ estimated from differentiated FMR spectra of GdFeCo films. The resonance field and line width were measured applying a magnetic field along and normal to the film plane, which are shown as open and closed circles, respectively.

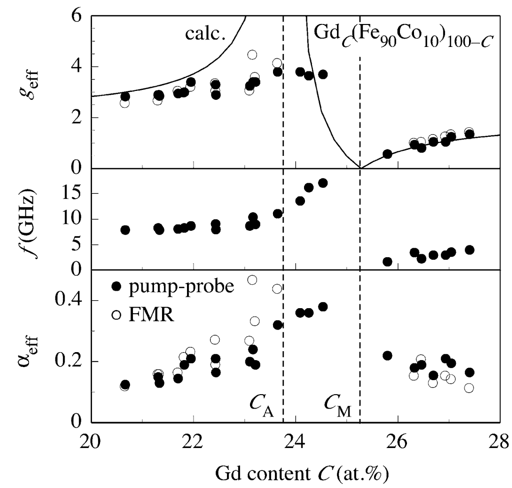

Fig. 4. Compositional dependences of effective g-factor $g_{\rm eff}$, precession frequency f, and effective Gilbert damping constant $\alpha_{\rm eff}$, of GdFeCo films. The closed and open circles represent the data from pump-probe and FMR methods, respectively. The solid line indicates the $g_{\rm eff}$ calculated by the mean-field model [1].

composition approaching to $C_{\rm M}$. This will be due to the increase of the effective damping constant $\alpha_{\rm eff}$ and/or effective field $H_{\rm eff}$ acting on the magnetization during the precession, since the relationship between the $\alpha_{\rm eff}$ and $H_{\rm eff}$ is given by

$$\alpha_{\rm eff} = \sqrt{3}\frac{\Delta H_{\rm PP}}{H_{\rm eff}}. \qquad (2)$$

The effective field $H_{\rm eff}$ can be estimated using Kittel's formula [12], and $\sqrt{3}$ is conversion factor of the line width between differentiated spectra and absorption spectra assuming Lorenzian form of the absorption. The effective field will increase near $C_{\rm M}$ due to the increase of $H_{\rm k}$. The line width of FMR spectrum is known to depend on the angle of the applied field due to the dispersions of direction of easy axis of the anisotropy and of the magnitude of the effective anisotropy [13]. However, the difference in the line width between out-of-plane and in-plane directions was quite small in our GdFeCo as shown in Fig. 3 except for near $C_{\rm M}$, which will indicate the negligible contribution of such dispersions to the line width. One of the reasons of the small contribution of the dispersions to the line width is that the resonance frequency is not significantly dependent on the angle of the applied field, since the effective perpendicular anisotropy of the GdFeCo is small, typically $\sim 10^5$ erg/cc. Thus, in this study, we used (2) to reduce parameters for the estimation of $\alpha_{\rm eff}$.

Fig. 4 shows the compositional dependences of precessional frequency f, effective g-factor $g_{\rm eff}$, and effective Gilbert damping constant $\alpha_{\rm eff}$ of the GdFeCo films obtained from the pump-probe data (closed circles) and FMR spectra (open circles). The solid line is the calculated $g_{\rm eff}$ by the mean field model [1] assuming g-factors of $g_{\rm TM} = 2.16$ and $g_{\rm Gd} = 2.0$. For the evaluation of $g_{\rm eff}$ and $\alpha_{\rm eff}$ from FMR spectra, we used $H_{\rm res}$ and $\Delta H_{\rm pp}$ measured applying a magnetic field along the easy axis. The data from the FMR spectra agree well with those from the pump-probe measurement except for near angular momentum compensation composition $C_{\rm A}$. The difference between FMR and pump-probe results at around $C_{\rm A}$ is considered to be due to the difference in the excitation method. In the pump-probe measurement, the pump illumination necessary to start the precession will disturb the spin alignment in the sample, which will be discussed later. As previously mentioned the FMR method cannot excite the magnetization near $C_{\rm M}$ because of the small net magnetization. However the pump-probe measurement utilize the laser pulse to excite the spin system, so that it was possible to observe the oscillation of the magnetization and possible to estimate $g_{\rm eff}$ and $\alpha_{\rm eff}$.

The precessional frequency f evaluated from the pump-probe measurement took a maximum near the magnetization compensation composition $C_{\rm M}$. This tendency is consistent with the previous reports [5], [6]. On the other hand, $g_{\rm eff}$ increases around the angular momentum compensation composition $C_{\rm A}$. The different trend between the precessional frequency f and effective g-factor $g_{\rm eff}$ is due to the increase of the anisotropy field $H_{\rm k}$ near $C_{\rm M}$ as shown in Fig. 1. The increase of $H_{\rm k}$ results in the increase of effective field $H_{\rm eff}$ acting on the magnetization, which will increase the precessional frequency. In this study, the increase of the anisotropy field $H_{\rm k}$ near $C_{\rm M}$ (Fig. 1) was taken into account in the fitting of the precessional motion. Thus it can be concluded that the $g_{\rm eff}$ did not increase as much as the precessional frequency f, and the increase of f near $C_{\rm M}$ is mainly due to the increase of the anisotropy field $H_{\rm k}$ near $C_{\rm M}$. The $\alpha_{\rm eff}$ also shows an increase near $C_{\rm A}$, which is consistent with the previous report [5], [6].

The compositional dependences of $g_{\rm eff}$ and $\alpha_{\rm eff}$ are roughly described by a mean-field model [1], [2]. The mean-field model shows divergences of $g_{\rm eff}$ and $\alpha_{\rm eff}$ at $C_{\rm A}$ [2], which is qualitatively consistent with the experiment. However, the $g_{\rm eff}$ near $C_{\rm A}$ estimated from pump-probe is much smaller than the calculated value. The mean field model assumes a perfect antiferromagnetic alignment of Gd and FeCo moments, but the pump illumination to start the precession may disturb such alignment. For the pump-probe measurement near $C_{\rm A}$, the pump fluence was set as small as possible to start the precession, typically ~ 2 mJ/cm^2, but the spin alignment may be slightly disturbed by the illumination, which will result in the decrease of the estimated $g_{\rm eff}$. Moreover, slight increase of the sample temperature during the precession of the magnetization may influence the estimation.

IV. Conclusion

Ferrimagnetic amorphous GdFeCo films with various compositions were prepared by magnetron sputtering method, and their magnetization dynamics; effective g-factor $g_{\rm eff}$ and effective Gilbert damping constant $\alpha_{\rm eff}$, were investigated both by pump-probe measurement and by ferromagnetic resonance. The high-power ultra-short pulse fiber laser was used for the pump-probe measurement and the fiber laser was found to excite the magnetization precession of the GdFeCo films. The $g_{\rm eff}$ and $\alpha_{\rm eff}$ estimated from the pump-probe and FMR methods agree well with each other, and they showed increases near angular momentum compensation composition $C_{\rm A}$. The FMR method was hard to excite the magnetization near the magnetization compenation composition $C_{\rm M}$ because of the small net magnetization, while it was possible to estimate $g_{\rm eff}$ and $\alpha_{\rm eff}$ near $C_{\rm M}$ for the pump-probe method. The increases of $g_{\rm eff}$ and $\alpha_{\rm eff}$ near $C_{\rm A}$ were qualitatively described by the mean field model assuming perfect antiferromagnetic alignment of Gd and FeCo moments. However, there was a quantitative difference between the $g_{\rm eff}$ estimated from pump-probe method and calculation near $C_{\rm A}$. The difference between the pump-probe experiment and the mean field model may be due to the disturbance of the spin alignment after the illumination of the pump beam.

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