Variation of Magnetization Dynamics of Co/Ni Multilayer by Capturing Magnetic Nanoparticles

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[Co (0.29 nm) / Ni (0.71 nm)]$_{10}$ multilayers were deposited on Ta (30 nm) and Pt (30 nm) buffer layers, and the variation of the magnetization dynamics by the adsorption of Fe_3O_4 nanoparticles was studied by vector network analyzer - ferromagnetic resonance (VNA-FMR) method. The surface roughness of the Co/Ni multilayer was 0.52 nm, and significantly increased after the adsorbing of the Fe_3O_4 nanoparticles which was captured by large field gradient on the demagnetized Co / Ni multilayer. The anisotropy field H_k, damping constant α, and anisotropy distribution ΔH_k of the Co / Ni on Ta buffer layer were 2.6 kOe, 0.012, and 0.32 kOe, respectively. By adsorbing of the Fe_3O_4 nanoparticles, the stray field from the nanoparticles H_{np} was applied to the Co/Ni, and $H_k + H_{np}$ was estimated to increase to 3.7 kOe. Moreover, α and ΔH_k also increased to 0.020, and 0.42 kOe by the adsorption, respectively. Similarly, $H_k + H_{np}$, α, ΔH_k of Pt buffered Co / Ni also increased from 2.4 kOe, 0.033, 0.43 kOe to 4.2 kOe, 0.055, 0.53 kOe, respectively. These results suggest the adsorption of Fe_3O_4 nanoparticles significantly modifies the magnetization dynamics of Co/Ni multilayer.

Index Terms—**Gilbert damping, Anisotropy field, Ferromagnetic resonance, Co / Ni multilayers, Fe_3O_4 nanoparticles.**

I. Introduction

Biosensor technology to detect specific biomolecules such as protein and deoxyribonucleic acid (DNA) has a potential to provide fast and accurate diagnosis of cancer and diabetes. One of the promising methods for the sensitive detection of the biomolecules is the use of magnetic nanoparticles functionalized with bio-recognition molecules, such as antibody [1]. The bio-functionalized nanoparticles are known to be biocompatible and cost-efficient for labeling specific biomolecules, and are also sensitively detected by magnetic field sensors, such as a super conducting quantum interference device (SQUID) [2] and a magneto-resistive (MR) sensor [3, 4]. The bio-functionalized magnetic nanoparticles are also attractive for drug delivery by applying a magnetic field gradient [5] and heating therapy by applying an alternating magnetic field [6]. The most promising material for the bio-functionalized magnetic nanoparticles is Fe_3O_4 nanoparticles. The Fe_3O_4 nanoparticles with a size of ~ 10 nm are ferromagnetic exhibiting a magnetization of ~ 50 emu/g, and are known to be non-toxic, non-immunogenic, and functionalized with bio-recognition molecules [1].

We consider spin wave biosensor to detect magnetic nanoparticles since the spin wave propagation and interference is considered to provide quite high sensitivity to detect the small magnetic field from magnetic nanoparticles [7]. For the spin wave magnetic field sensor, the ferromagnetic materials with low Gilbert damping are desired, and an yttrium iron garnet (YIG) film with low Gilbert damping was used in the previous study [7]. However, the YIG film exhibits a small magnetization of ~ 130 emu/cc and a small magnetic anisotropy due to its cubic crystal structure [8]. In this paper, we report the detection of Fe_3O_4 nanoparticles by the variation of magnetization dynamics of Co/Ni multilayers.

Co/Ni multilayer is known to exhibit a large magnetization and large perpendicular anisotropy [9] which is useful to create large field gradient at the surface by forming the maze domain structure at the demagnetized state [10], and the large field gradient was used to capture the Fe_3O_4 nanoparticles dispersed in liquid in this study. The magnetic properties of Co/Ni, such as magnetization and perpendicular anisotropy, are easily tuned by varying the thicknesses of Co and Ni, which is beneficial to control the domain structure of Co/Ni and produce large field gradient on the surface. Moreover, low Gilbert damping is reported in Co/Ni multilayers [11, 12], which will be effective to detect the variation of magnetization dynamics by the adsorption of Fe_3O_4 and also be beneficial for the application to the spin wave magnetic field sensor.

II. Experimental Method

Co / Ni multilayers with a stack of substrate / Ta or Pt (30) / [Ni (0.71) / Co (0.29)]$_{10}$ / Ta (2) (thickness in nm) was prepared by a DC magnetron sputtering system. The substrate was synthetic fused quartz with a thickness of 0.5 mm, and the Ar pressure during the sputtering was kept at 1.0 Pa. The magnetic properties of the Co/Ni were characterized by alternating gradient field magnetometer. Surface morphology and magnetic domain structure of the Co/Ni were measured by atomic force microscope (AFM) and magnetic force microscope (MFM), respectively. For the adsorption of

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nanoparticles, the water-based liquid containing 0.003 mol/L Fe_3O_4 nanoparticles with an average diameter of 10 nm was used. The Co/Ni multilayer was dipped into the liquid for 1 min, and then the residual liquid on the sample was naturally dried out.

The Co/Ni multilayers were micro-fabricated into 20 μm × 1500 μm by photolithography followed by Ar^+ ion etching. Some of the samples were dipped into the liquid with Fe_3O_4 after the micro-fabrication. SiN (300 nm) was sputtered on the Co/Ni with or without the nanoparticles, and GSG (ground-signal-ground) type coplanar waveguide with a signal line width of 20 μm was micro-fabricated on the Co/Ni wire. The waveguide has a stack of Cr (2) / Au (400), and the lift-off technique was used for the micro-fabrication. Vector network analyzer based ferromagnetic resonance (VNA-FMR) was used to measure the anisotropy field and damping parameter of the Co/Ni multilayers. The complex scattering parameter S_{21} was recorded varying an rf frequency under the static field up to 10 kOe along the film normal direction to estimate the resonance frequency f_{res} and linewidth Δf_{50}.

III. Results and Discussions

Figure 1 shows hysteresis loops of Co/Ni multilayers on (a) Ta and (b) Pt buffer layers. The loops were measured applying an external field along perpendicular (solid line) and parallel (dashed line) to the film plane. Both multilayers exhibited large perpendicular anisotropy, and the anisotropy field H_k was estimated to be about 3 kOe and 4 kOe for the multilayers with Ta and Pt buffer layers, respectively. The both multilayers exhibited almost the same magnetization M_s of ~670 emu/cc, and thus the effective anisotropy $K_{eff} = M_s H_k / 2$ was estimated to be 1 Merg/cc and 1.3 Merg/cc for Ta and Pt buffered Co / Ni, respectively.

FIG. 1 HERE

Figure 2 shows (a) AFM and (b) MFM images of Ta buffered Co/Ni multilayers after adsorbing of Fe_3O_4 nanoparticles. For reference, AFM and MFM images of the Co/Ni before adsorbing the nanoparticles are also shown in Figs. 2 (c) and (d), respectively. Before adsorbing nanoparticles, the multilayer was demagnetized to form the maze domain structure with a stripe width of ~ 200 nm as shown in Fig. 2 (d). The surface roughness R_a of the as-sputtered Co/Ni multilayer was estimated to be 0.52 nm from Fig. 2 (c). After adsorbing the Fe_3O_4 nanoparticles, the adsorbed nanoparticles were clearly seen as bright contrast as shown in Fig. 2 (a). The shape of the bright region in Fig. 2 (a) was well consistent with the maze domain structure seen in Fig. 2 (b). (The domain structures before and after the adsorbing were slightly different because of the different measurement position.) This means that the large field gradient produced on the surface of Co/Ni multilayer effectively captures the nanoparticles dispersed in the liquid.

FIG. 2 HERE

Figure 3 (a) shows absorption spectra of VNA-FMR for the Co / Ni multilayers on Ta buffer layer without the nanoparticles. The absorption spectra were obtained by subtracting the background spectrum measured at H_{ext} = 0 kOe from the spectra measured at various H_{ext}. Since the background spectrum at H_{ext} = 0 kOe contains the FMR absorption around 7 GHz due to the anisotropy field of Co/Ni multilayer, all the spectra shown in Fig. 3 have a positive peak around 7 GHz. Thus, we focus our attention to the negative absorption peak resulting from the ferromagnetic resonance of the Co / Ni multilayers. One can see that the resonance frequency gradually increased with increasing the external field. The absorption spectra were fitted with the Lorenzian function, which is shown as solid lines in the figure, to extract the resonance frequency f_{res} and line width Δf_{50} from the absorption spectra.

FIG. 3 HERE

The absorption spectra were also taken for the Co/Ni multilayers with magnetic nanoparticles. Figure 3 (b) shows the absorption spectra of the Ta buffered Co / Ni with Fe_3O_4 nanoparticles. Similar results to those of the Co / Ni without nanoparticles were obtained; negative peak shifted to high frequency side with increasing H_{ext}. However, by comparing Figs. 3 (a) and (b), one can see that the negative peak for the Co/Ni with Fe_3O_4 nanoparticles appeared at higher frequency than that without nanoparticles. This suggests the stray field from the nanoparticles shifted the FMR frequency of the Co/Ni as discussed later. In addition, the broad negative peaks with lower frequency than that of the resonance of the Co / Ni became obvious for $H_{ext} \geq 7$ kOe as shown in Fig. 3 (b). These peaks are considered to be due to the resonance of the Fe_3O_4 nanoparticles.

Figure 4 (a) shows the external field dependence of resonance frequency f_{res} of Ta buffered Co / Ni multilayers with and without magnetic nanoparticles. The field dependence of f_{res} was fitted with the following Kittel equation [13]

$$f_{res} = \frac{\gamma}{2\pi}\left(H_{ext} + H_k + H_{np}\right), \quad (1)$$

where $\gamma = g\mu_B / \hbar$ is the gyromagnetic ratio, and g is the g–factor, μ_B is the Bohr magneton, and $\hbar$ is the Dirac constant. Eq. (1) shows f_{res} is proportional to the effective field consisting of the external field H_{ext}, anisotropy field H_k of the Co / Ni multilayer, and the stray field H_{np} from the nanoparticles, and $H_k + H_{np}$ is estimated by the linear fit of the

H_{ext} dependence of f_{res}. From the results of the Co/Ni without nanoparticles, the anisotropy field H_k of the Co/Ni multilayer was estimated to be 2.6 kOe assuming the field from the nanoparticles H_{np} = 0. The anisotropy field H_k of the Co / Ni multilayer without nanoparticles agreed well with the hysteresis measurements as discussed in Fig. 1. The *g*–factor of the Co / Ni was 2.11 which did not depend on the adsorption of the nanoparticles. From the data of Co/Ni with nanoparticles shown in Fig. 4 (a), the field $H_k + H_{np}$ was estimated to be 3.7 kOe, suggesting H_{np} = 1.1 kOe. If the Fe_3O_4 nanoparticles are assumed to agglomerate on the Co / Ni, and the magnetic spheres with a diameter of 50 nm and a magnetization of 500 emu/cc are assumed to be arranged in a square lattice with a pitch size of 100 nm on the Co / Ni multilayer, the mean field in the Co/Ni was calculated to be 0.9 kOe, which is comparable to the H_{np} obtained from Fig. 4 (a).

Figure 4 (b) shows the dependence of linewidth Δf_{50} of the resonance curve on the resonance frequency f_{res} for the Ta buffered Co / Ni with and without magnetic nanoparticles. The damping parameter α is determined from the frequency dependence of the resonance linewidth by the following equation [14],

$$\Delta f_{50} = 2\alpha f_{res} + \frac{\gamma}{2\pi}\Delta H_k \qquad (2)$$

where ΔH_k is the anisotropy dispersion of the Co / Ni multilayer. From the linear fit of the data in Fig. 2 (b), ΔH_k and α were found to increase by adsorbing the nanoparticles. α and ΔH_k of Co / Ni was 0.012 and 0.32 kOe, respectively, before the adsorption and they increased to 0.020 and 0.42 kOe, respectively, by adsorbing the nanoparticles. The increase of ΔH_k is reasonable since the nanoparticles are captured non-uniformly as described in Fig. 2. However, the increase of α for the Co/Ni multilayer with nanoparticles suggests the energy dissipation into the nanoparticles through the magneto-static coupling between the Co / Ni multilayer and Fe_3O_4 nanoparticles.

FIG. 4 HERE

Similar results were also confirmed for Pt buffered Co / Ni multilayers as shown in Figs. 4 (c) and (d). The field $H_k + H_{np}$ increased from 2.4 kOe to 4.2 kOe by the adsorption of Fe_3O_4 nanoparticles, suggesting the stray field from the nanoparticles H_{np} = 1.8 kOe. The larger H_{np} suggests the larger amount of the adsorbed Fe_3O_4 nanoparticles, and we consider the Pt buffered Co/Ni captured more Fe_3O_4 nanoparticles than Ta buffered one. H_k of the Co / Ni multilayer on Pt was smaller than the value estimated from Fig. 1, but not so far from the result of the hysteresis measurement. The *g*-factors of the Pt buffered Co / Ni were not significantly dependent on the adsorption of nanoparticles; *g* = 2.10 for the Co / Ni without nanoparticles and *g* = 2.12 for the Co / Ni with nanoparticles. α and ΔH_k of the Co / Ni without nanoparticles were 0.033 and 0.43 kOe, respectively. The increase of α for the Pt buffered Co / Ni multilayer compared to Ta buffered Co / Ni is considered to be due to spin pumping effect in the Pt layer [15, 16]. α and ΔH_k of the Co / Ni were also increased to 0.055 and 0.53 kOe, respectively, by adsorbing the nanoparticles. The increase of the damping constant by adsorbing the nanoparticles for the Pt buffered Co/Ni was larger than that of Ta buffered Co/Ni. This may be related to the large H_{np} for Pt buffered Co/Ni. The results shown in Fig. 4 suggest the adsorption of Fe_3O_4 nanoparticles significantly modifies the magnetization dynamics of Co/Ni multilayer, and the Co/Ni is considered as a candidate material for spin wave sensors to capture and detect biomolecules.

IV. Conclusions

Co / Ni multilayers with a perpendicular magnetic anisotropy and low Gilbert damping constant were prepared and the variation of the magnetization dynamics by adsorbing Fe_3O_4 nanoparticles was studied by VNA-FMR measurements. The demagnetized Co / Ni multilayer had a maze domain structure with a stripe width of ~200 nm, and the field gradient on the Co / Ni surface by the maze domain structure was found to effectively capture the Fe_3O_4 nanoparticles dispersed in the water-based liquid. The anisotropy field H_k and damping constant α of Co / Ni on Ta buffer layer were 2.6 kOe and 0.012, respectively. The higher damping constant was observed for Co / Ni multilayer on Pt buffer layer due to the spin pumping effect. The field H_k + H_{np}, damping α, and anisotropy distribution ΔH_k were found to increase by adsorbing the Fe_3O_4 nanoparticles on the both Co / Ni multilayer on Ta and Pt buffer layers. H_{np} and the increase of ΔH_k are considered to be due to the stray field from the Fe_3O_4 nanoparticles. The increase of α suggests the energy dissipation into the nanoparticles through the magneto-static coupling between the Co / Ni multilayer and Fe_3O_4 nanoparticles. From the results obtained in the present experiments, the magnetization dynamics of Co/Ni multilayer is found to be significantly modified by the adsorption of Fe_3O_4 nanoparticles, and the Co/Ni is considered as a candidate material for spin wave sensors to capture and detect biomolecules.

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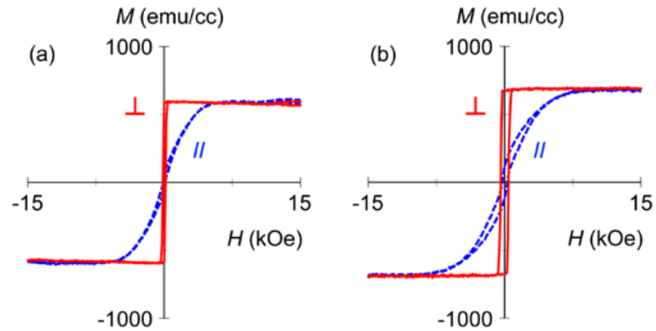

Fig. 1. Hysteresis loops of Co/Ni multilayers on (a) Ta and (b) Pt buffer layers. The loops were measured applying a external field along perpendicular (solid line) and parallel (dashed line) to the film plane.

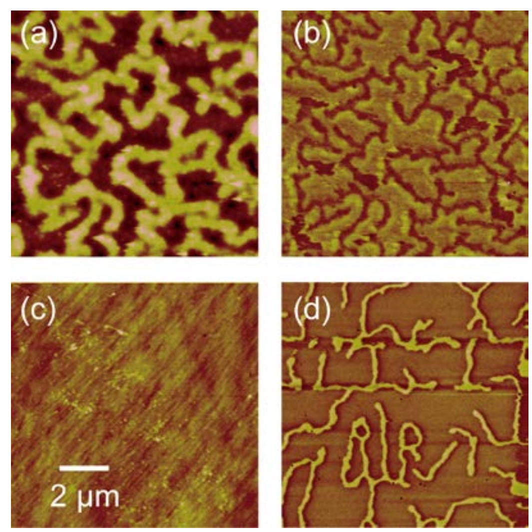

Fig. 2. (a) AFM and (b) MFM images of Ta buffered Co/Ni multilayers after adsorbing of Fe_3O_4 nanoparticles. For reference, (c) AFM and (d) MFM images of the Co/Ni before the adsorbing of nanoparticles are also shown.

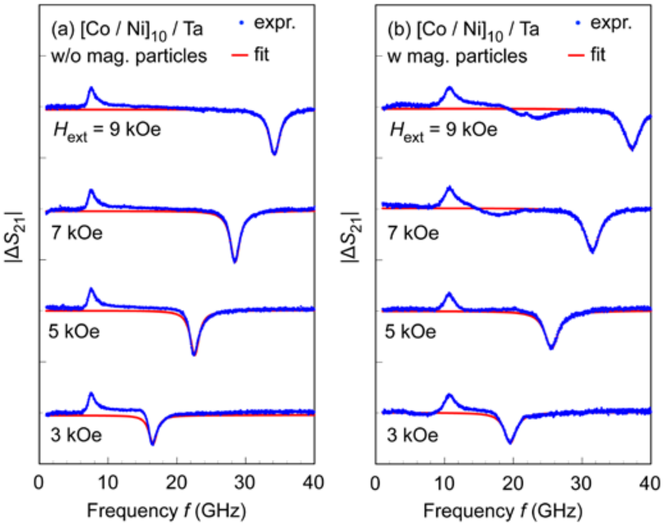

Fig. 3. Absorption spectra of VNA rf signal for the [Co (0.29) / Ni (0.71)]$_{10}$ / Ta (30) (a) without and (b) with Fe_3O_4 nanoparticles at various H_{ext}.

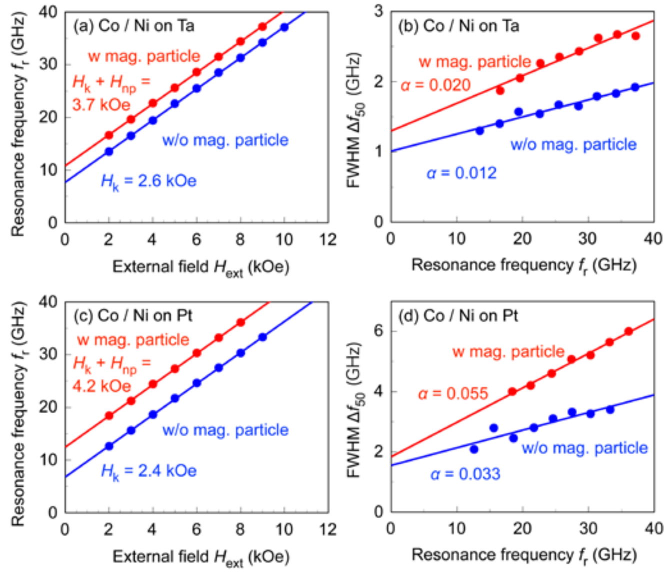

Fig. 4. External field dependence of resonance frequency f_{res} of (a) Ta buffered and (c) Pt buffered Co / Ni multilayers with and without magnetic nanoparticles, and dependence of linewidth Δf_{50} of the resonance curve on the resonance frequency f_{res} for (b) Ta buffered and (d) Pt buffered Co / Ni with and without magnetic nanoparticles.