$CoFeSiO/SiO_2$ multilayer granular films with very narrow Ferromagnetic resonant linewidth

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In order to fabricate GHz-band micro-magnetic devices, we have investigated the high-frequency permeability of $CoFeSiO/SiO_2$ multilayer granular films comprising alternate stacks of granular and insulator layers, which can enable us to control the magnetic grain size and inter-grain spacing along the film thickness direction independent of each other. A very narrow ferromagnetic resonance (FMR) linewidth was obtained under specific film conditions. A sharp FMR peak was obtained for granular layer and insulator layer thicknesses of 6 nm and 1 nm, respectively. The $CoFe/SiO_2$ volume ratio of the granular layer has a strong influence the FMR linewidth. A high $CoFe/SiO_2$ volume ratio resulted in a very narrow FMR linewidth. HR-TEM observation of a film with a high $CoFe/SiO_2$ volume ratio revealed a well-defined multilayer granular structure and a homogeneous CoFe grain size, which seem to be necessary for obtaining a very narrow FMR linewidth. The narrowest FMR linewidth observed in this study is 420 MHz, for an FMR frequency of 2.4 GHz, a permeability of 380, and a damping factor α of 0.007.

***Index Terms*—granular, ferromagnetic resonance, linewidth, permeability**

I. Introduction

According to the recent advancements in the field of GHz-band wireless networks such as cellular phones and wireless LANs, a large number of studies on high-frequency soft-magnetic thin films have been carried out in order to fabricate micro-magnetic devices [1], [2]. The permeability of soft-magnetic thin films decreases abruptly in the vicinity of the ferromagnetic resonance (FMR) frequency, which restricts the frequency range for which the films can be used. Therefore, soft-magnetic thin films used for high frequency applications must have a large anisotropy magnetic field (H_k) and high saturation magnetization (M_s) in order to obtain a high FMR frequency. In addition, their FMR linewidth should be as narrow as possible because the magnetic loss around the FMR frequency strongly depends on the FMR linewidth.

We have previously studied multilayer granular films comprising alternate granular and insulator layers. This enables us to independently control the magnetic grain size and inter-grain spacing along the film thickness direction [3], [4]. Thus, multilayer granular films not only exhibit high resistivity due to the multiple insulator layers but also superior high-frequency permeability. Furthermore, under specific conditions, these films possess a high FMR frequency with a very narrow FMR linewidth. In order to analyze the factors responsible for narrow FMR linewidth in soft-magnetic thin films, in this study, we have fabricated $CoFeSiO/SiO_2$ multilayer granular films with different granular structures and observed the microstructure of these films by high-resolution transmission electron microscopy (HR-TEM). We also report the high-frequency magnetic properties of the $CoFeSiO/SiO_2$ multilayer granular films, dependence of the FMR linewidth on the granular structure, and the microstructure of the films.

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II. Experimental Procedure

$CoFeSiO/SiO_2$ multilayer granular films were deposited on a surface-oxidized (100) silicon substrate (400 µm thick) at room temperature by co-sputtering $Co_{80}Fe_{20}$ alloy and SiO_2 targets using an inductively coupled RF sputtering system under a pressure of approximately 0.2 Pa of pure Ar gas. During deposition, a magnetic field of approximately 8 kA/m was applied to the film plane in order to introduce in-plane uniaxial magnetic anisotropy. The total thickness of the multilayer films was regulated to be approximately 300 nm.

The microstructure of the film was evaluated by HR-TEM, and the magnetization curves were measured by using a vibrating sample magnetometer (VSM). A size of VSM sample was 10 mm × 10 mm. The frequency dependence of permeability in the range from 1 MHz to 9 GHz was measured by using a high-frequency permeameter (PMM-9G1: Ryowa Electronics Co., Ltd.) with a sample size of 4 mm × 10 mm.

III. Results and Discussion

A. Magnetic properties of $CoFeSiO/SiO_2$ multilayer films

The magnetization curves and frequency dependence of the complex permeability of the $CoFeSiO/SiO_2$ film (granular layer thickness: 6 nm, SiO_2 layer thickness: 1 nm, $CoFe/SiO_2$ ratio: 5.0) are shown in Figs. 1 and 2, respectively. In-plane uniaxial magnetic anisotropy with an H_k of 3.6 kA/m is clearly confirmed, and the easy axis of magnetization corresponds to the direction of the applied field during the deposition; this implies that the uniaxial magnetic anisotropy is an induced magnetic anisotropy.

The real-part permeability is approximately 380, which remains constant up to 1 GHz, and the FMR frequency is 2.4 GHz. The value of real permeability is almost equal to the calculation result of rotation magnetization by using the measured M_s and H_k, which indicates that the origin of

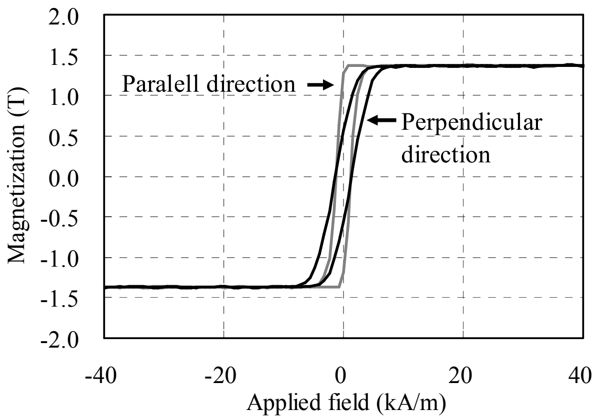

Fig. 1. Magnetization curves of $CoFeSiO/SiO_2$ multilayer granular film.

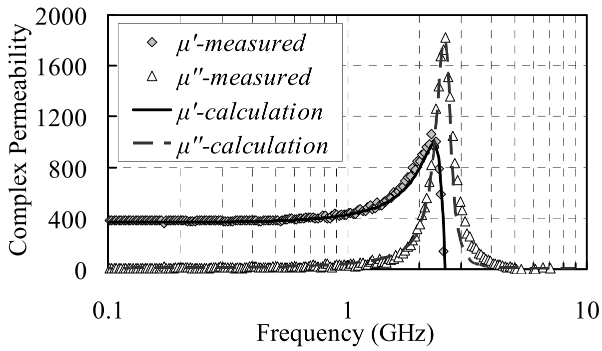

Fig. 2. Frequency dependence of complex permeability of $CoFeSiO/SiO_2$ multilayer granular film.

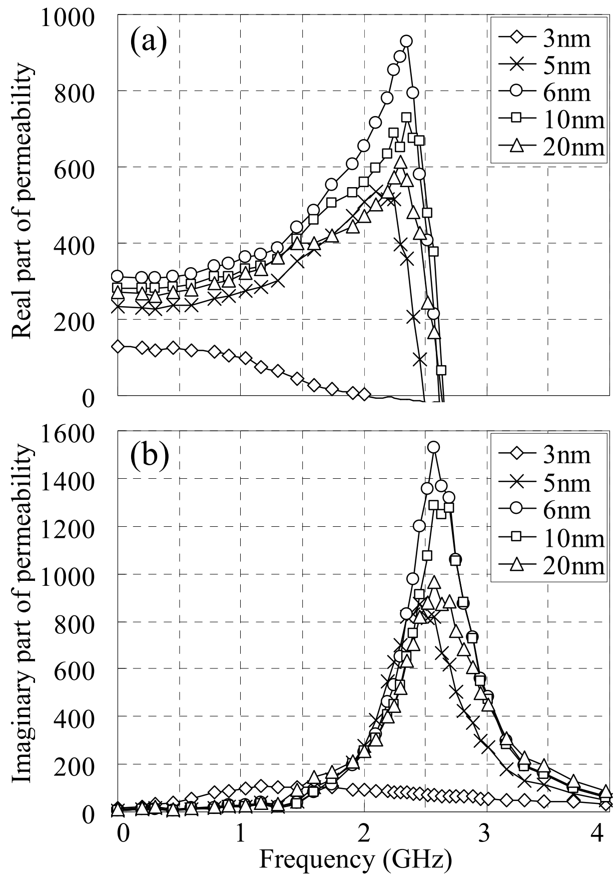

Fig. 3. Frequency dependence of permeability of $CoFeSiO/SiO_2$ films for various granular layer thickness (a) real part, (b) imaginary part.

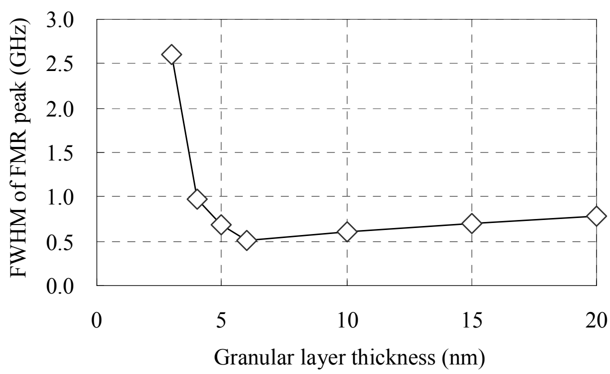

Fig. 4. FWHM of FMR peak as a function of granular layer thickness.

permeability is derived from a rotation magnetization. The imaginary-part permeability exhibits a very sharp FMR loss peak and its full width at half maximum (FWHM) is 420 MHz. The permeability calculated using the measured values of H_k, M_s and resistivity ρ is also shown in Fig. 2. The calculated permeability agrees with the measured permeability. The Gilbert damping factor α used for this calculation is 0.007; which is lower than those of commonly used soft-magnetic films [5]-[7]. The small value of α indicates a very narrow FMR linewidth, which is probably due to a relatively uniform distribution of H_k in the film. The $CoFeSiO/SiO_2$ film with a very narrow FMR linewidth and high permeability can be effectively used as the magnetic core of thin-film inductors in GHz-band applications.

B. Relation between FMR linewidth and granular structure

Figure 3 shows the frequency dependence of the complex permeability of $CoFeSiO/SiO_2$ films for various granular layer thicknesses ranging from 3 to 20 nm with a constant $CoFe/SiO_2$ volume ratio of 4.0 and a constant SiO_2 layer thickness of 1.0 nm. The film with a 6-nm-thick granular layer has the maximum real-part permeability in the low-frequency region. In the samples with a granular layer thickness greater than 6 nm, the magnetocrystalline anisotropy, induced by an increase in the CoFe grain size resulting from an increase in the granular layer thickness may influence the rotation of the magnetic moment; this influence may result in a decrease in the permeability [3]. On the other hand, a super-paramagnetic phase partially appears in the films with a granular layer thickness less than 6 nm; the appearance of this phase is a result of the small grain size of CoFe alloy.

The imaginary-part permeability exhibits a very narrow FMR linewidth for the films with a granular layer thickness greater than 5 nm. The FWHM of the FMR peak as a function of granular layer thickness is shown in Fig. 4. The minimum FWHM was observed for the film with a granular layer thickness of 6 nm. This observation corresponds to the

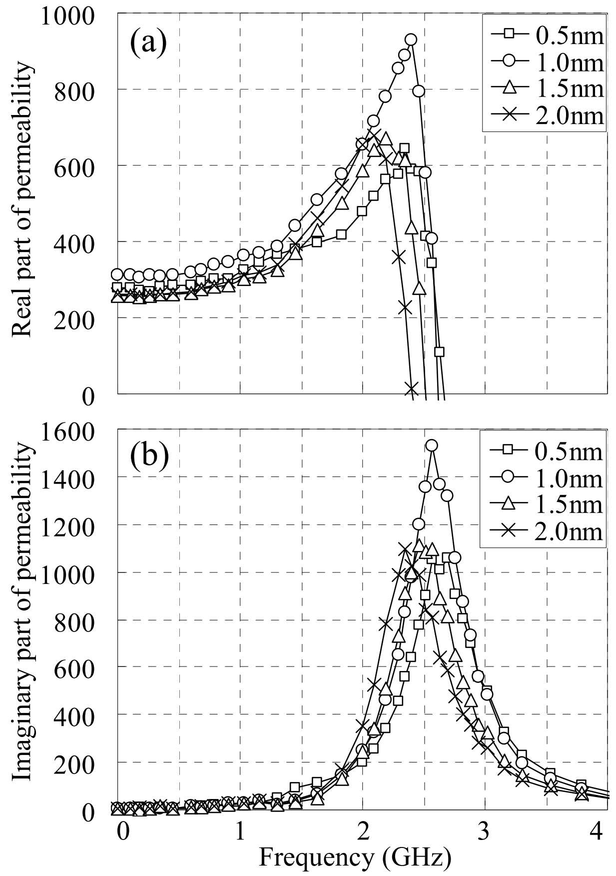

Fig. 5. Frequency dependence of permeability of CoFeSiO/SiO_2 films for various SiO_2 layer thickness (a) real part, (b) imaginary part.

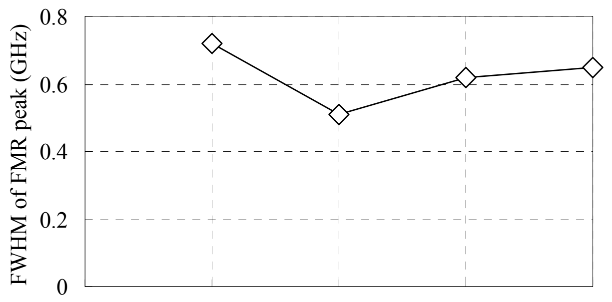

Fig. 6. FWHM of FMR peak as a function of SiO_2 layer thickness.

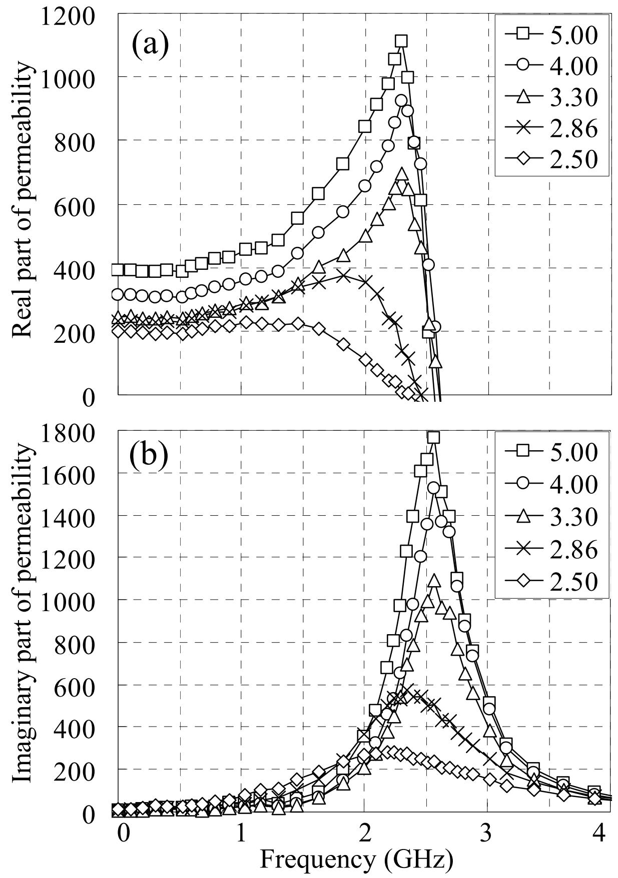

Fig. 7. Frequency dependence of permeability of CoFeSiO/SiO_2 film for various CoFe/SiO_2 volume ratio (a) real part, (b) imaginary part.

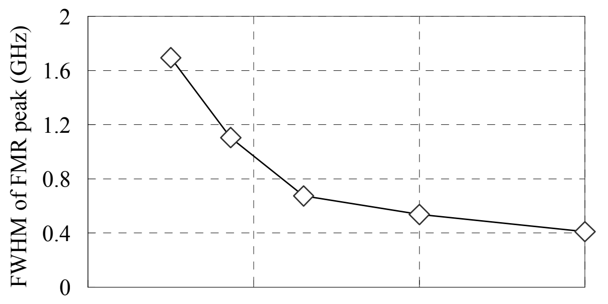

Fig. 8. FWHM of FMR peak as a function of CoFe/SiO_2 volume ratio.

appropriate grain size required for obtaining a very narrow FMR linewidth. The appearance of the super-paramagnetic phase, observed in the magnetization curves of the films with a small granular layer thickness deteriorates the homogeneity of H_k, which results in a broad FMR linewidth. Furthermore, an increase in magnetocrystalline anisotropy has an adverse effect on the exchange interaction of CoFe grains, which directly increases the dispersion of H_k.

The frequency dependence of the complex permeability and the FWHM of FMR peaks for films with SiO_2 layer thicknesses ranging from 0.5 to 2.0 nm (constant granular layer thickness: 6 nm, constant CoFe/SiO_2 volume ratio: 4.0) are shown in Figs. 5 and 6, respectively. The maximum real-part permeability at 100 MHz and the maximum imaginary-part permeability at the FMR frequency are observed for the film with a SiO_2 layer thickness of 1 nm. The FWHM of the FMR peak (Fig. 6) is also minimized for this film. A deformed multilayer structure of the film with a SiO_2 layer thinner than 1 nm and a decline of exchange interaction along the film thickness direction of the film with a SiO_2 layer thicker than 1 nm can explain the dependence of FMR linewidth on the SiO_2 thickness. Therefore, in order to form a well-defined multilayer granular structure, a SiO_2 layer with a thickness of no less than 1 nm is required.

The frequency dependence of the complex permeability and the FWHM of the FMR peaks of $CoFeSiO/SiO_2$ films with various $CoFe/SiO_2$ volume ratios of the granular layer (constant granular layer thickness: 6 nm, constant SiO_2 layer thickness: 1 nm) are shown in Figs. 7 and 8, respectively. The real-part permeability increases with an increase in the $CoFe/SiO_2$ volume ratio, i.e., increase in the M_s value of the granular layer. The imaginary-part permeability exhibits a sharper FMR peak for a higher $CoFe/SiO_2$ ratio. Accordingly, the FWHM of FMR peak decreases with an increase in the $CoFe/SiO_2$ volume ratio. In the multilayer granular structure, the $CoFe/SiO_2$ ratio of the granular layer is considered to be closely related to the thickness of SiO_2 between adjacent CoFe grains in the in-plane direction, which increases with a decrease in $CoFe/SiO_2$ ratio. Therefore, the sharp FMR peaks observed for the film with a high $CoFe/SiO_2$ ratio is probably due to a strong exchange interaction between the in-plane CoFe grains interleaved with a thin SiO_2 layer.

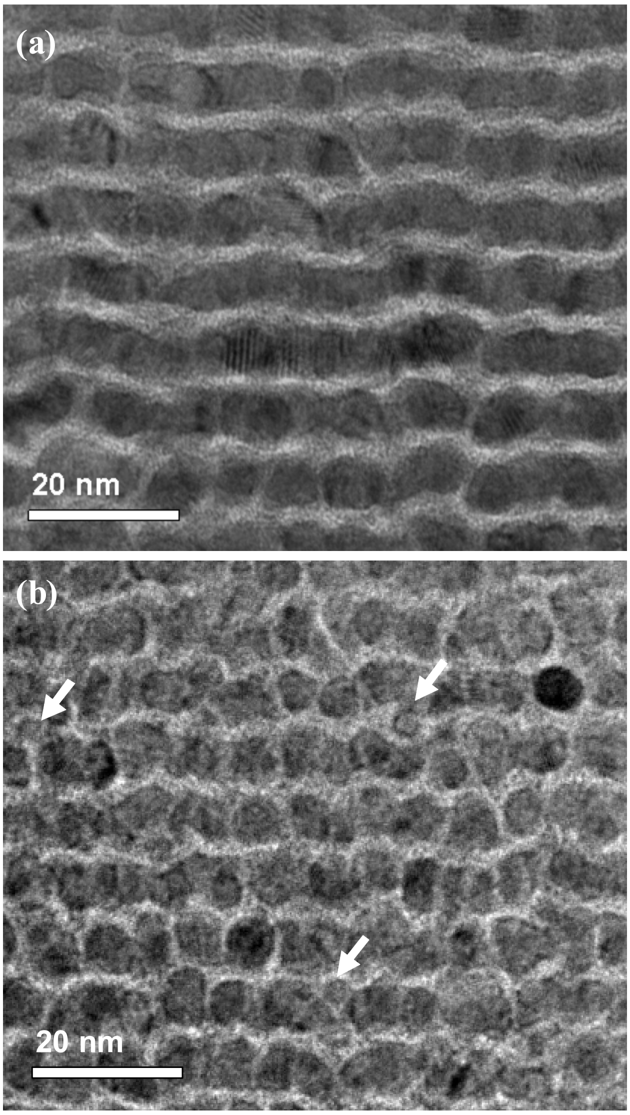

Fig. 9. Cross-sectional HR-TEM images of CoFeSiO (6 nm) /SiO_2 (1 nm) films with a $CoFe/SiO_2$ ratio of (a) 5.00, (b) 2.50.

C. HR-TEM observation of $CoFeSiO/SiO_2$ multilayer film

The cross-sectional HR-TEM images of $CoFeSiO/SiO_2$ films are shown in Fig. 9. The multilayer structure formed by alternate granular CoFeSiO and amorphous SiO_2 layers is clearly observed. The CoFe grain size is approximately 6 nm, which is almost equal to the granular layer thickness; this indicates that the SiO_2 layers suppress the grain growth of CoFe. There are CoFe grains smaller than 6 nm, shown by the arrows in Fig. 9b, therefore the uniformity in the CoFe grain size and SiO_2 layer thickness of the film with a high $CoFe/SiO_2$ volume ratio (Fig. 9a) is better than that of the film with a low $CoFe/SiO_2$ volume ratio (Fig. 9b). It is inferred that excess SiO_2 in the granular layers hampers uniform CoFe grain growth during the formation of granular layers. A well-defined multilayer granular structure and a high uniformity in the CoFe grain size seem to be necessary for obtaining a very narrow FMR linewidth as shown in Fig. 7.

IV. Conclusion

We have investigated the high frequency permeability of $CoFeSiO/SiO_2$ multilayer granular films, which exhibit a very narrow FMR linewidth. A granular layer thickness of 6 nm, a SiO_2 layer thickness of 1 nm, and a high $CoFe/SiO_2$ volume ratio are required to obtain a sharp FMR peak with an FMR linewidth of 420 MHz at an FMR frequency of 2.4 GHz. HR-TEM observation of the films with a high $CoFe/SiO_2$ ratio revealed a well-defined multilayer granular structure and homogeneous CoFe grain size, which seem to be necessary for obtaining a very narrow FMR linewidth.

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