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FeNiSiO/SiO_2 multi-layer granular magnetic films with high-resistivity

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The resistivity and high-frequency permeability of FeNiSiO/SiO_2 multilayer granular magnetic films have been investigated to fabricate a magnetic film with high resistivity suitable for high-frequency micromagnetic devices. High resistivity is obtained by improving the electrical insulation of SiO_2 associated with high-temperature deposition and concurrently by optimizing the microstructure, for example, the grain size of granular Fe-Ni metals and the thickness of SiO_2 layers, which are independently controllable by an alternate sputtering deposition of FeNiSiO granular and SiO_2. The high-frequency permeability are strongly affected by the Fe-Ni grain size as well as the Ni content, due to the influence of super-paramagnetism; the composition of the Fe-Ni grains primarily determines an anisotropy magnetic field (H_k), which is competitive with super-paramagnetism and is a key parameter that affects the high-frequency permeability. The permeability obtained in FeNiSiO/SiO_2 films with a resistivity of 1.1 Ωcm is 460 at 100 MHz.
Key words: high resistivity, permeability, granular, Fe-Ni, high frequency

1. INTRODUCTION

In the last few decades, numerous studies have been conducted on micromagnetic devices, which require superior high-frequency soft magnetic thin films with high permeability [1]-[4]. Soft magnetic films for high-frequency applications must have both a large anisotropy magnetic field (H_k) and high saturation magnetization (M_s) in order to achieve a high ferromagnetic resonance (FMR) frequency. Furthermore, the electrical resistivity of the magnetic films is also a significant factor to be considered for the application of the films in micromagnetic devices. This is because metallic magnetic films with low resistivity cause an increase in the eddy current losses in the films because of a decrease in the skin depth and also result in a parasitic capacitance between the magnetic films and metal conductive layers. Therefore, high resistivity is desirable for magnetic thin films, particularly for high-frequency applications. However, most of the high-frequency soft magnetic thin films with high permeability, which mainly include magnetic metals such as Fe, Co, and their alloys, exhibit lower resistivity than that of conventional ferrite materials [5].

Granular magnetic films composed of fine grains of ferromagnetic 3d-transition metal alloys embedded in an insulating oxide matrix of chemically active elements have been considered to be a promising candidate in high-frequency applications because of their relatively high resistivity, which is induced by the insulating oxide between the magnetic metal grains [6]-[10]. The permeability of the granular magnetic films is closely related to the suppression of the magnetocrystalline anisotropy of the individual magnetic grains and the inter-grain exchange interaction; in particular, a strong exchange coupling through a thin insulating layer is preferred for realizing a magnetic film with high permeability while maintaining high resistivity. In general, there is a tradeoff between high resistivity and high permeability; therefore, they cannot coexist even in granular-structured magnetic films.

We have already developed multilayer granular films, which consisted of alternate stacks of granular and insulator layers, and they exhibit excellent high-frequency permeability while maintaining a relatively high resistivity (approximately 1 mΩcm) [11]. Compared to a conventional self-organized granular structure prepared by phase separation phenomena in co-sputtering deposition, a multilayer granular structure enables the artificial modification of the microstructure independently in terms of grain size, thickness of the insulator layer, and the arrangement of magnetic grains, which indicates a potential for fabricating a magnetic film with high permeability and high resistivity.

In this paper, we have elucidated the relationship between the high-frequency permeability and the resistivity of FeNiSiO/SiO_2 multilayer granular films by tuning the structural parameters and the composition of the ferromagnetic granular metal.

2. EXPERIMANTAL PROCEDURE

FeNiSiO/SiO_2 multilayer granular magnetic films were deposited on a surface-oxidized (100) silicon substrate by using inductively coupled RF sputtering systems. FeNiSiO granular and SiO_2 insulator layers were fabricated by the co-sputtering of Fe, Ni, and SiO_2 targets and single sputtering of SiO_2 target, respectively. The thicknesses of the FeNiSiO granular and SiO_2 amorphous layers were controlled by the shuttering time. The operation pressure of pure Ar gas was approximately 0.12 Pa, and the background pressure was less than 1.0×10^{-6} Pa. The substrate temperature was regulated from 20°C to 200°C. The film composition ratio of Fe to Ni was controlled by the RF power of each target. During deposition, a magnetic field of approximately 100 Oe was applied to the film plane in order to introduce an in-plane uniaxial magnetic anisotropy. The total thickness of the films was regulated to be approximately 300 nm. The thicknesses of the FeNiSiO granular and SiO_2 insulator layers were controlled by the shuttering time during deposition.

The microstructure of the film was evaluated by transmission electron microscopy (TEM). The Fe and Ni contents were measured by X-ray fluorescence (XRF). The magnetization curves were measured by using a vibrating sample magnetometer (VSM). The dependence of frequency on complex permeability was evaluated using a high-frequency permeameter (PMM-9G1: Ryowa Electronics Co., Ltd.) in the frequency range from 1 MHz to 9 GHz. The DC resistivity was measured by a conventional four-point probe method.

3. RESULTS AND DISCUSSION

A cross-sectional TEM image of the FeNiSiO/SiO_2 multilayer granular film (FeNiSiO granular layer: 4 nm, SiO_2 insulator layer: 2 nm, Ni content: 31 at.%, and substrate temperature: 160ºC) is shown in Fig. 1. The circular dark contrast and a light contrast observed in this figure correspond to the crystallized Fe-Ni nano-sized grains and amorphous SiO_2 layer, which clearly shows that the sequential deposition of FeNiSiO and SiO_2 produces a multilayer granular structure appearing as layers of Fe-Ni grains isolated by continuous amorphous SiO_2 layers. This structure differs from that of conventional granular magnetic films, which show a random arrangement and inhomogeneous size of magnetic grains. The size of the Fe-Ni grain is almost equal to the thickness of the FeNiSiO granular layer, which indicates the suppression of the growth of magnetic grains and flexibility in modifying the grain size by inserting the SiO_2 insulator layer.

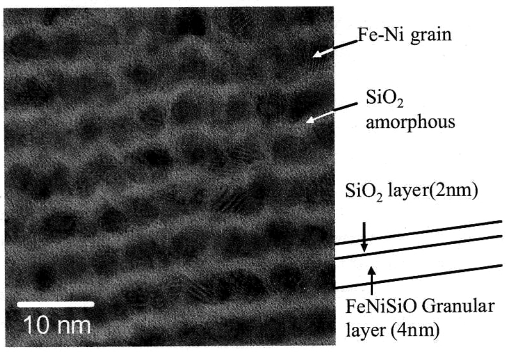

Fig. 1 Cross-sectional TEM image of FeNiSiO multilayer granular film.

Figure 2 shows magnetization curves of FeNiSiO/SiO_2 multilayer granular film (FeNiSiO granular layer: (a) 4 nm, (b) 3 nm, SiO_2 insulator layer: 2 nm, Ni content: 31 at.%, and substrate temperature: 160ºC). A distinct in-plane uniaxial magnetic anisotropy is observed in Fig. 2a. The easy axis of magnetization is in accordance with a parallel direction of the applied magnetic field during the deposition; this indicates that the uniaxial magnetic anisotropy stems from an induced magnetic anisotropy. H_k of the film was approximately 23 Oe, and M_s was 0.89 T, which shows a soft magnetism suitable for high-frequency applications. To the contrary, two magnetization curves almost accord with each other in Fig. 2b, which imply no existence of the uniaxial magnetic anisotropy. In the multi-layer granular structure, the size of the magnetic grains is almost equivalent to the thickness of the granular layers as shown in Fig. 1 [11], and the reduction in grain size causes the transition from ferromagnetism to super-paramagnetism; therefore, a disappearance of the uniaxial magnetic anisotropy as confirmed in Fig. 2b, may be attributed to the emergence of super-paramagnetic phase.

The DC resistivity and frequency dependence of the complex permeability of the FeNiSiO/SiO_2 multilayer granular films (FeNiSiO granular layer: 4 nm, SiO_2 insulator layer: 2 nm, and Ni content: approximately 30 at.%) as a function of substrate temperature are shown in Figs. 3 and 4, respectively. The DC resistivity of the films increases abruptly with an increase in the substrate temperature, and it is equal to 7.5 Ωcm at 200ºC. As shown in Fig. 4a, the real permeability at 10 MHz tends to increase with the substrate temperature, while an opposite tendency is observed at 1 GHz, which results from a large decreasing trend of the frequency dependence of real permeability of the films deposited at a higher substrate temperature. The frequency at which the real permeability starts to decrease with an increase in frequency decreases as the substrate temperature increases, and the corresponding peak in the imaginary permeability shift to a lower frequency along with a reduction in its peak maximum (Fig. 4b). Such a change in the frequency dependence associated with the

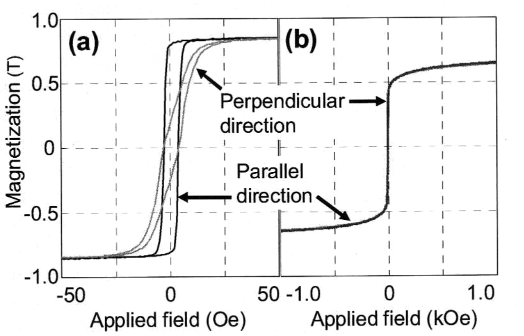

Fig.2 Magnetization curves of FeNiSiO/SiO_2 films. FeNiSiO granular layer (a) 4nm, (b) 3nm

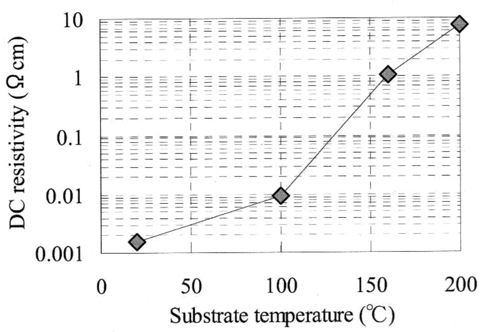

Fig. 3 Dependence of resistivity on substrate temperature.

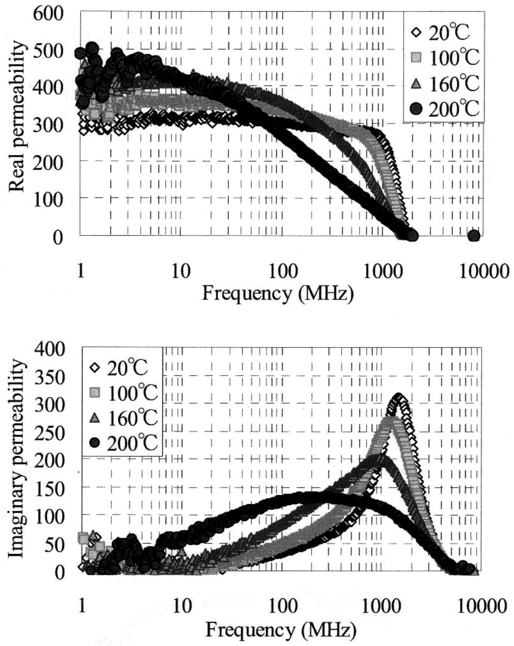

Fig. 4 Frequency dependence of permeability as a function of substrate temperature. (a) Real part and (b) imaginary part

substrate temperature can be inferred as the influence of super-paramagnetic phase induced by a reduction in a short-ranged exchange interaction between two adjacent magnetic grains [8], which is closely related to the film resistivity controlled by the inter-grain insulating amorphous layer of SiO_2. Because the grain size and the Ni content of the films are almost constant, the increase in resistivity with the substrate temperature is probably attributed to an improvement in the insulating amorphous SiO_2 layers in terms of less number of carriers and magnetic element impurities, which magnetically couple with neighboring grains. On the other hand, the most probable contribution of the oxidation of Fe-Ni grains when heated can be excluded from the results that the M_s of the films are almost constant irrespective of the substrate temperature (no show here).

The DC resistivity and frequency dependence of the complex permeability of the FeNiSiO/SiO_2 multilayer granular films (SiO_2 insulator layer: 2 nm, Ni content: approximately 30 at.%, and substrate temperature: 160ºC) as a function of FeNiSiO granular layer thickness are shown in Figs. 5 and 6, respectively. An increase in the granular layer thickness results in a sharp reduction in the resistivity as shown in Fig. 5, and it also improves the high-frequency permeability properties (Fig. 6), which suggest the effect of Fe-Ni grain size. The reduction in resistivity caused by an increase in the granular layer thickness originates from the reduction in an interface resistance by increasing the grain size of metallic Fe-Ni. The origin of the change in permeability profile is probably considered to be caused by the magnetic transition of Fe-Ni grains. For instance, the frequency dependence of the complex permeability with a granular layer thickness of 3 nm, which exhibits a decreasing trend with an increase in frequency, is

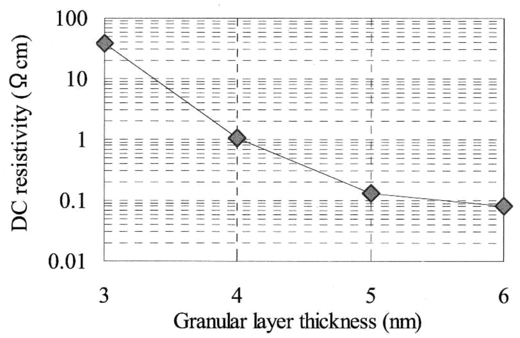

Fig. 5 Dependence of resistivity on granular layer thickness.

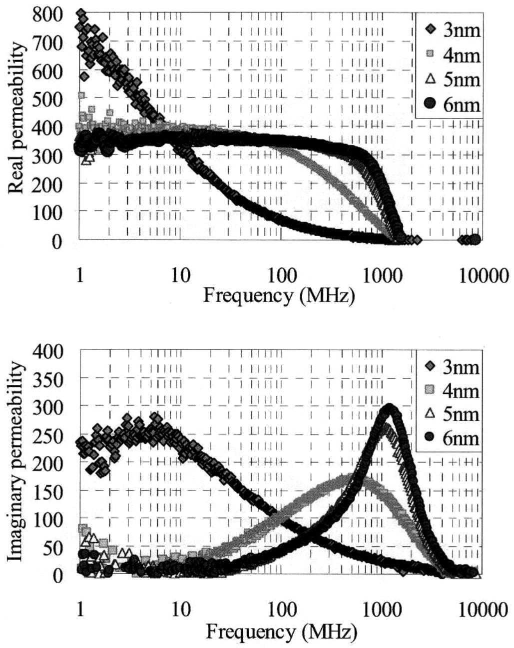

Fig. 6 Frequency dependence of permeability as a function of granular layer thickness. (a) Real part and (b) imaginary part

directly associated with the existence of super-paramagnetism as shown in Fig. 2b [8]. The super-paramagnetism appearing in small-sized grains is detrimental to high-frequency soft magnetic properties, while large-sized grains cause a reduction in the resistivity of the films. This implies that there exists an optimum grain size to realize both high-resistivity and good high-frequency magnetic properties and, in this experiment, the grain size of 4 nm is preferable and is close to the optimum size.

The DC resistivity and frequency dependence of the complex permeability of the FeNiSiO/SiO_2 multilayer granular films (FeNiSiO granular layer: 4 nm, SiO_2 insulator layer: 2 nm, and substrate temperature: 160ºC) as a function of Ni content is shown in Figs. 7 and 8, respectively. At 10 MHz, the real permeability of the film with a Ni content of 21.4 at.% is the largest; however, the maximum permeability at 100 MHz is shifted to that of the film with a Ni content of 31.1 at.%,

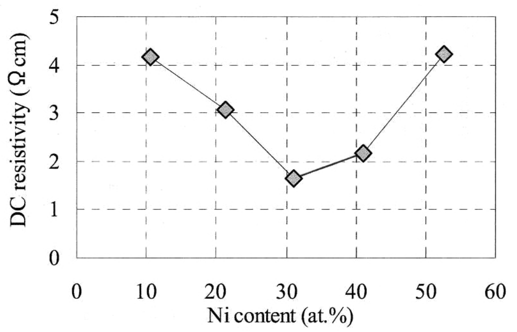

Fig. 7 Dependence of resistivity on Ni content of the films.

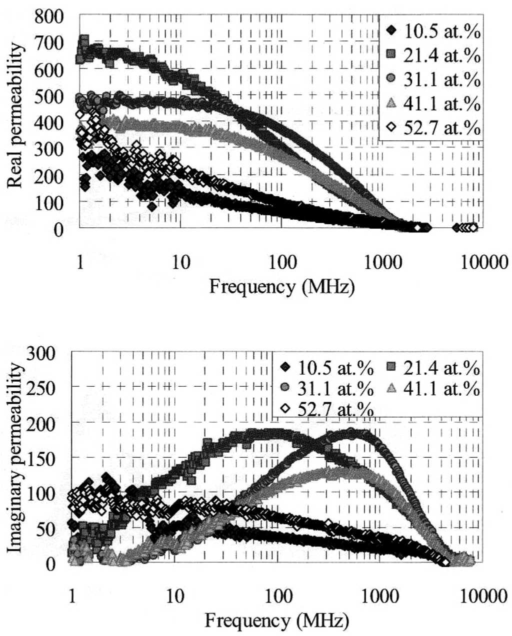

Fig. 8 Frequency dependence of permeability as a function of Ni content. (a) Real part and (b) imaginary part

because of the smaller frequency dependence of permeability at high frequency region. The peak values of the imaginary permeability are maximized in the films with a Ni content of 21.4 at.%, while the peak frequency of the imaginary permeability attains a maximum value in the film with a Ni content of 31.1 at.%, which is the largest value observed. The DC resistivity of the films is minimized at a Ni content of 31.1 at.%, which is still greater than 1 Ωcm. This value is expected to be sufficiently large to suppress the parasitic capacitance that degrades the device performance.

Figure 9 shows the dependence of M_s and H_k on the Ni content. M_s and H_k vary with the Ni composition as observed in Co-Fe alloys [9], and it amount to maximum values at an Ni content of approximately 30 at.%, which signifies the origin of variation in frequency dependence of permeability caused by the Ni content. A superior frequency dependence of permeability of the film with a Ni 31.1 at.% as shown in Fig. 8, results from the relatively large H_k of Fe-Ni grains as confirmed by Fig. 9, while the inferior results of the others is probably due to the existence of super-paramagnetic phase caused by a small H_k, which indicates the importance of H_k of magnetic grains at high-resistivity granular magnetic films.

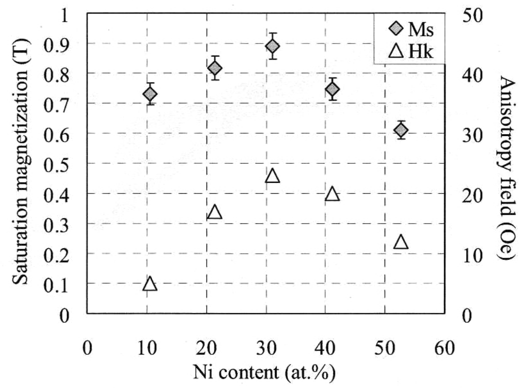

Fig. 9 Dependence of saturation magnetization and anisotropy field on Ni content.

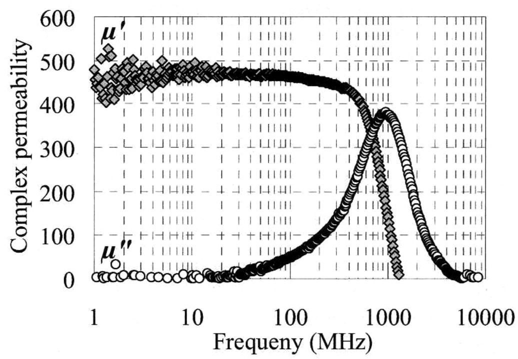

Fig. 10 Frequency dependence of the complex permeability of $FeNiSiO/SiO_2$ multilayer granular film.

The frequency dependence of the complex permeability of $FeNiSiO/SiO_2$ multilayer granular film (FeNiSiO granular layer: 4 nm, SiO_2 insulator layer: 2 nm, Ni content: 30.5 at.%, and substrate temperature: 160°C) is shown in Fig. 10. The values of real permeability are approximately 460 at 10 MHz and it remains constant up to 100 MHz, which is slightly large compared with a calculation permeability of rotation magnetization by using the measured values of H_k and M_s (Fig. 2a). The Q factor at 100 MHz is 10, and the FMR frequency is 1.3 GHz, which almost accord with estimated result by using the Landau-Lifshitz-Gilbert equation [12]. These results indicate that the permeability of $FeNiSiO/SiO_2$ films derives from the magnetic moment rotation. The DC resistivity of this film is 1.1 Ωcm, which indicates the compatibility between superior high-frequency permeability and high resistivity. $FeNiSiO/SiO_2$ films will be suitable at least for hundreds megahertz applications such as power inductor for the integrated power supply, due to its high resistivity [13].

Consequently, to realize magnetic thin films with high resistivity and excellent high-frequency magnetic properties, it is essential that the microstructure, for example, the grain size and the spacing of the grains should be regulated suitably and consistently, and magnetic grains should have a large anisotropic magnetic field in order to prevent the transition to super-paramagnetism. $FeNiSiO/SiO_2$ multilayer granular magnetic films with an optimized microstructure and composition exhibit resistivity greater than 1 Ωcm and excellent high-frequency permeability; furthermore, multilayer granular structure is effective for artificially controlling the grain size and inter-grain spacing to obtain superior high-frequency soft-magnetic properties, where the composition modification of Fe-Ni magnetic grains is essential to achieve large H_k.

4. CONCLUSION

The resistivity and high-frequency magnetic properties of $FeNiSiO/SiO_2$ multilayer granular magnetic films have been investigated to fabricate high-frequency magnetic films with high resistivity, suitable for use in high-frequency micromagnetic devices. The multilayered granular structure is modified to obtain high-resistivity magnetic films by ensuring the high electrical insulation of inter-grain SiO_2 nonmagnetic layers using a high-temperature process and by controlling the microstructure, for example, the grain size and the interlayer thickness of the grains. The high-frequency magnetic properties are strongly affected by the Fe-Ni grain size and are also varied with different Ni contents of Fe-Ni magnetic grains, suggesting a significant influence of super-paramagnetism. The reduction in the grain size of Fe-Ni particles results in the appearance of super-paramagnetism, and the composition of Fe-Ni grains determines the value of H_k, which influences the permeability, particularly in high-frequency regions. The value of real permeability obtained in the $FeNiSiO/SiO_2$ multilayer granular film with a resistivity of 1.1 Ωcm is approximately 460 at 100 MHz, and the Q factor of the film is 10 at 100 MHz. The novel $FeNiSiO/SiO_2$ granular multi-layer film will be suitable at least for hundreds megahertz applications such as power inductor for the integrated power supply [13].

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