Magnetic Anisotropy and the Magnetic Coupling in $Ni-SiO_2$ Granular Films

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In this paper we report a ferromagnetic resonance (FMR) study on a series of granular Ni_x-$(SiO_2)_{1-x}$ film with the Ni volume fraction x ranging from 0.24 to 1.0. The x-dependence of the resonance field and of the linewidth were examined with field applied parallel and perpendicular to the film surface. Our results show that the shape anisotropy of the Ni-grains increases systematically with increasing x and that the magnetic coupling between the grains is greatly enhanced at the percolation threshold.

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I. Introduction

Recently, the giant Hall effect (GHE) was observed in $Ni-SiO_2$ granular films [1]. The reported extraordinary Hall resistivity is as large as 200 $\mu\Omega$cm at 4 kG and it offers a technological promise as a sensitive device for detecting magnetic fields. In Ni_x-$(SiO_2)_{1-x}$ granular films, the Ni concentration x plays a crucial role, controlling their structural, magnetic and transport properties [2,3]. At $x \sim$ 0.5-0.6, where GHE was observed, a metallic conduction regime appears and a labyrinth structure can be observed in electron micrographs [3]. However, even in this region the conducting network in the samples is not ideally metallic and the value of GHE is greater by almost 4 orders of magnitude than that in pure nickel. Mechanisms suggested for the observed GHE include inter-granular contacts, electron interaction and localization effects [5]. An understanding of how the Ni-concentration effects the magnetic structure is important for future applications of these materials.

In this paper, we report a study of the ferromagnetic resonance (FMR) on a series of granular Ni_x-$(SiO_2)_{1-x}$ films with the Ni-concentration x varying from 0.24 to 1. The resonance fields (H_r) and their width (AH) were measured at parallel and perpendicular directions with respect to the surface of the film. We have observed a systematic increase of the separation between the parallel and the perpendicular center fields with increasing x, and an abrupt drop of AH at both directions at $x \sim$ 0.6. We relate these results to the magnetic anisotropy of the grains and to their mutual magnetic coupling.

II. Experiment

The granular Ni_x-$(SiO_2)_{1-x}$ films, with different Ni volume fraction x ranging from 0.24 to 1.0, were prepared in a Denton magnetron co-sputtering system, with Ni and SiO_2 targets mounted on two separate guns. The thickness of the films is about 1 μm. The ferromagnetic resonance (FMR) spectra were taken at room temperature with a BRUCKER EMX system operated at 9.5 GHz. Samples of about 2 x 3 mm^2 were placed on a rotating support situated in the middle of a rectangular TE_{102} resonant cavity.

III. Results and discussions

A typical FMR spectrum is shown in Fig. 1. H_r is defined as the central field of the resonance absorption peak and AH the field between the extrema of the absorption derivatives. The parallel (||) and perpendicular ($\perp$) directions are defined as zero and 90" angles between the direction of the applied field and the surface of the film. We have found that the FMR spectra in the || direction shift to lower fields and spectra in the $\perp$ direction shift to higher fields with increasing x. Fig. 2 shows the spectra for $x=$ 0.65 in the || and the $\perp$ directions respectively. The figure shows a large change of the H_r value from 1700 gauss to 5800 gauss on change of direction from || to $\perp$. A weak line also appears in the $\perp$ direction. The weak line was observed in all the samples with x > 0.5, but only in the $\perp$ direction. In Fig. 3, we plot H_r in the || direction, $H_r^{||}$ (open squares) and H_r at $\perp$ direction, $H_r^{\perp}$ (open circles) as a function of x. The plot shows that $H_r^{||}$ is nearly equal to $H_r^{\perp}$ for x = 0.24 and that the difference between $H_r^{||}$ and $H_r^{\perp}$ grows larger with increasing x. This separation of $H_r^{||}$ and $H_r^{\perp}$ implies that the shape the of the Ni-grains becomes

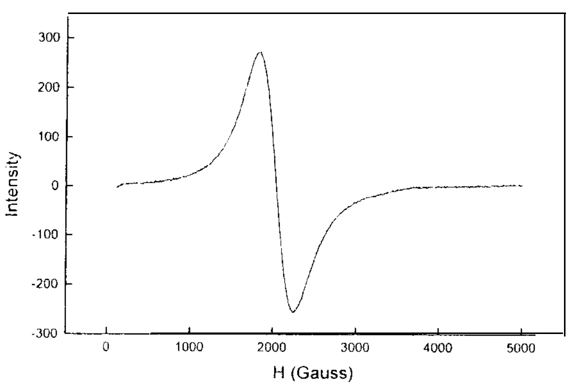

FIG. 1. A typical ferromagnetic resonance (FMR) spectrum for a Ni_x-$(SiO_2)_{1-x}$ film.

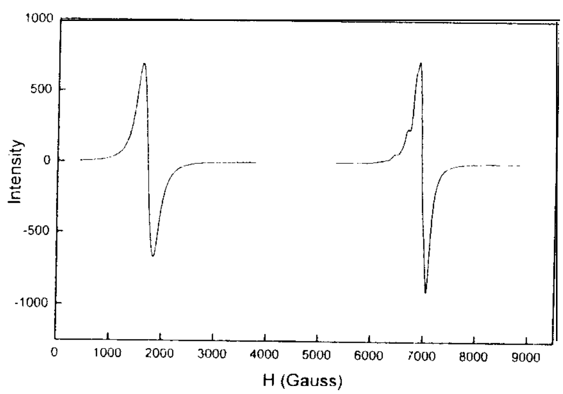

FIG. 2 Ferromagnetic resonance (FMR) spectra for $x=$ 0.65 in the parallel (left spectrum) and the perpendicular (right spectrum) direction, respectively.

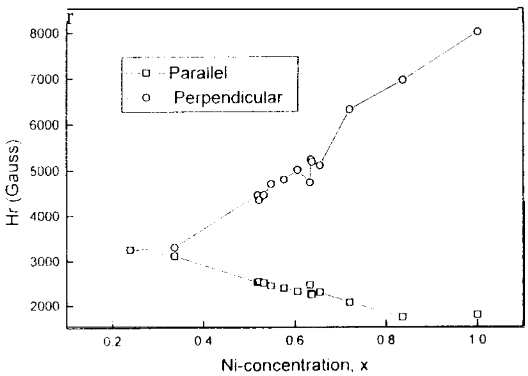

FIG. 3. Resonance field (H,) in the parallel (open squares) and the perpendicular (open circles) for x varying from 0.24 to 1.

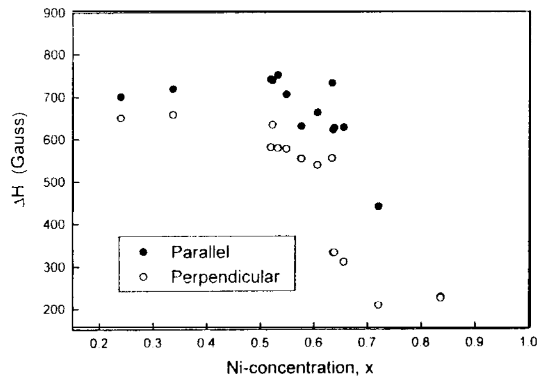

FIG. 4. Field width (AH) in the parallel (closed circles) and the perpendicular (open circles) direction for x ranging from 0.24 to 1.

more anisotropic for high x. Fig. 4 shows AH in the $\|$ direction (closed circles) and the $\perp$ direction (opened circles) vs. Ni-concentration. In both the $\|$ and the $\perp$ directions the data exhibit an abrupt drop at around x $\sim$ 0.6.

The angular dependence of FMR spectra reveals the magnetic anisotropy of the film. For a magnetic epitaxial film, or for a single crystal, the contributions of magnetic anisotropy include the diamagnetic anisotropy, the uniaxial anisotropy and the crystalline magnetic anisotropy. But for a granular film, the magnetic anisotropy is mainly determined by the shape anisotropy of the magnetic grains [5]. However, when the grain-size increases and the distance between grains decreases their exchange coupling plays a role and the increase of the separation between $H_r^{\|}$ and $H_r^{\perp}$ (Fig. 3) is an evidence of the fact that the diamagnetic dipolar field in the direction perpendicular to the film increases gradually at small x, but greatly at large x. This suggests that the growth of the Ni-grain is mainly two-dimensional along the surface of the film. Two contributions have to be considered for an interpretation of the AH $-$ x data. One is the angular distribution of the c-axis of the Ni-crystal, and the other is inhomogeneity of the internal field. The abrupt drop of AH was observed at $\|$ as well as $\perp$ direction and it should therefore not be related to the angular distribution of the crystalline axis. We thus propose that the sudden reduction of the inhomogeneity of the internal field is the cause of the observed AH drop at $x \sim$ 0.6. The concentration x = 0.6 is about the percolation threshold in this system and our suggestion thus becomes more convincing because the physical connection of Ni-grains diminishes the inter granular barrier and makes spin-interaction (between grains) stronger, so that the local field inhomogeneity is greatly depressed.

TEM study on $(NiFe)_x(SiO_2)_{1-x}$ film was published previously [6]. The sample examined was x $\sim$ 0.6 in which the giant Hall resistivity was observed. Based on their TEM data, the size of grain was ranging from 5 Å to 80 Å, and became more uniform after annealing. We believe that TEM measurements on a series of Ni_x-$(SiO_2)_{1-x}$ will be very useful for relating, directly, our FMR results to the x-dependent granular structures of the samples.

IV. Conclusion

We have measured the FMR spectra for a series of granular Ni_x-$(SiO_2)_{1-x}$ films. Our results show that the resonance field with applied field perpendicular to the film surface increases greatly with increasing Ni-concentration x. This implies enhancement of magnetic anisotropy. We have also observed a sudden drop in the width of the resonance field AH at $x \sim 0.6$. This drop of AH was related here to the physical connection between the Ni-grains, and was attributed to the enhancement of the magnetic coupling between Ni-grains at the percolation threshold.

Acknowledgments

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