Giant Hall Effect of $Fe_{45.51}(Al_2O_3)_{54.49}$ Nano-granular Film *

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A series of $Fe_{45.51}(Al_2O_3)_{54.49}$ nano-granular films were prepared using ion-beam sputtering technique. A saturated hall resistivity of about 12.5 $\mu\Omega$·cm at room temperature was observed. The transmission electron microscopy image showed that very small Fe particles of smaller than 1 nm are embedded in Al_2O_3 matrix, and connected into network. The measured $\rho - T$ curve indicated that this giant Hall effect may originate from the percolation phenomenon. With different annealing temperature (T_A) up to 300° C, the saturated Hall resistivity decreased only a little. The good thermal stability of $Fe_{45.51}(Al_2O_3)_{54.49}$ nano-granular Films showed potential application for magnetic sensor.

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The most common magnetic field sensors are based on the Hall effect in semiconductors.[1] In normal metals, the Hall coefficient is lower than semiconductors because of the high density of carriers. In ferromagnetic metals, there is an additional extraordinary Hall effect, which is proportional to magnetization of sample.[2] This extraordinary term is typically a factor of 10 or more stronger than the ordinary one. The discovery of giant magnetoresistance (GMR) in magnetic multilayers[3,4] and granular magnetic-nonmagnetic metal alloys[5,6] revived the interest in the studies of magnetotransport in nanostructured inhomogeneous magnetic solids. It was found recently that in magnetic particles embedded in nonmagnetic insulator matrix, the extraordinary Hall resistivity was enhanced to several orders larger than that of the pure magnetic metals when the magnetic metal particles are just above the percolation threshold.[7-10] This effect was called giant Hall effect (GHE) by Pakhomov *et al.*,[7] similar to the GMR effect. Based on the giant Hall effect and the weak temperature dependency of Hall resistivity, it was proposed that percolation ferromagnetic granular metal-insulator films can be an alternative candidate material structure for high sensitive Hall sensors. In this paper, we report the study on the Hall resistivity and the microstructure of $Fe_{45.51}(Al_2O_3)_{54.49}$ nano-granular films, the influence of annealing temperature on saturated Hall resistivity.

The $Fe_{45.51}(Al_2O_3)_{54.49}$ nano-granular films about 150 nm thick were deposited on glass substrates using ion-beam sputtering technique. A mosaic target was employed. The target consisted of pure Al_2O_3 and Fe, and small pieces of Al_2O_3 adhered compactly on the plate of Fe to get the right volume fraction. The exact Fe volume fraction of the deposited films was determined by energy-dispersive x-ray spectroscopy using an EDAX PV-9100. The magnetization M was measured at room temperature with a vibrating sample magnetometer (VSM). Hall resistivity was measured by using conventional four-terminal method with the applied magnetic field perpendicular to the film plane. Two Hall voltages were measured either by rotating the film by 180° in a fixed field or by reversing the field direction to remove the offset voltage due to the asymmetric Hall terminals. The $Fe_{45.51}(Al_2O_3)_{54.49}$ nano-granular film used for TEM observation was deposited on carbon films supported by a fine copper mesh.

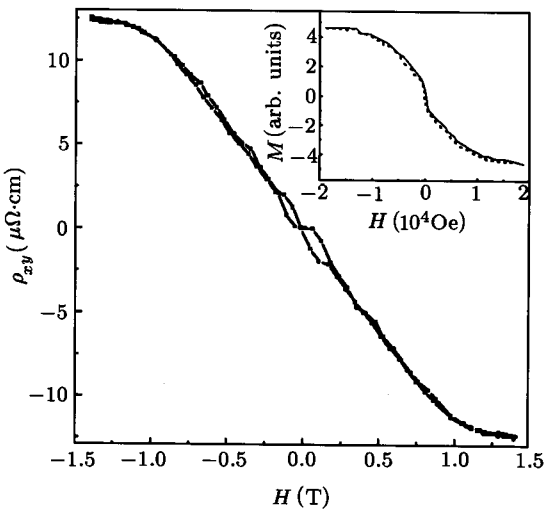


Fig. 1. Hall resistivity ρ_{xy} as a function of applied magnetic field H for an as-deposited $Fe_{45.51}(Al_2O_3)_{54.49}$ nano-granular film sample at room temperature. The inset shows the magnetization hysteresis loop of the same sample with magnetic field applied perpendicularly to the film plane.

Figure 1 shows the Hall resistivity ρ_{xy} as a function of applied magnetic field H for an as-deposited $Fe_{45.51}(Al_2O_3)_{54.49}$ nano-granular film sample at room temperature. The inset shows the magnetization hysteresis loop of the same sample with applied magnetic field perpendicular to the film plane. Note that

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the curve of the Hall resistivity against field H is similar to the magnetization curve. The Hall resistivities were negative in the positive field. The extraordinary Hall resistivity can be characterized by its absolute value at saturation, ρ_{xys}. Figure 1 shows the saturated Hall resistivity ρ_{xys} of the as-deposited $Fe_{45.51}(Al_2O_3)_{54.49}$ nano-granular film at room temperature which was about 12.5 $\mu\Omega$·cm. The saturated field was high, about 1.3 T, which was consistent with the magnetization curve. The signal level based on these percolating films was smaller than that of the best semiconductor sensors. It was believed that by employing a better soft magnetic material (larger saturation magnetization and smaller coercivity) to replace Fe in the two components composite, signal level could be further improved.

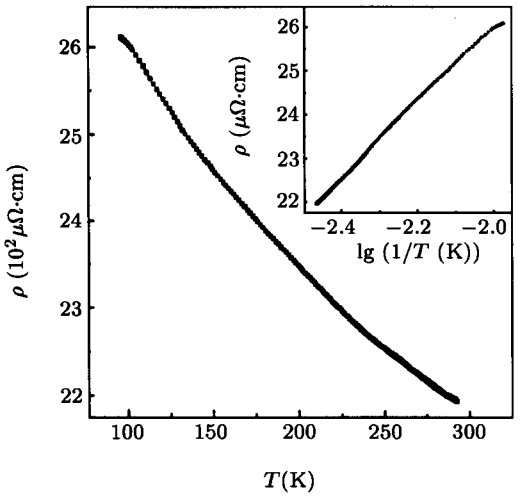


Fig. 2. The $\rho - T$ curve of the as-deposited $Fe_{45.51}(Al_2O_3)_{54.49}$ nano-granular film, the inset was the ρ vs logarithm of temperature.

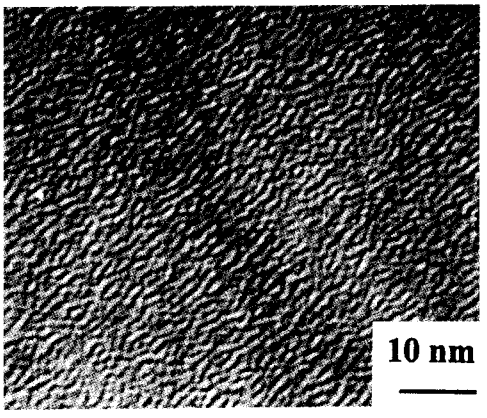


Fig. 3. TEM image of as-deposited $Fe_{45.51}(Al_2O_3)_{54.49}$ nano-granular film.

Figure 2 shows the $\rho - T$ curve of the as-deposited sample. The resistivity ρ decreased with the increase of ρ temperature. The inset shows the curve of versus $\log(1/T)$. The ρ vs T dependence was close to logarithmic. This dependence is typical of granular sputtered metals close to the metal-insulator transition. Pakhomov *et al.*[7] attributed it to metallic conduction via percolation paths, with enhanced electron–electron interaction effects. Figure 3 shows the transmission electron microscopy (TEM) image of the as-deposited sample. The Fe particles embedded in Al_2O_3 matrix were smaller than 1 nm, and almost connected into network. The TEM image further demonstrated that the volume fraction of Fe in these granular films was near percolation threshold.

Compared with the studied of the Hall effects of the magnetic metals embedded in SiO_2 matrix, the Hall effects of these nano-granular films all increased drastically when the volume fraction of the magnetic metal decreased to the percolation threshold.[8,10] The large enhancement of Hall effects in these films can not be explained via a simple percolation theory, which predicts that the largest enhancement is less than 20.[11] The mechanism why the magnetic metal-nonmagnetic insulator granular film near percolation exhibited giant Hall effect was still under study. We would like to attribute this anomalous giant Hall effect to the enhanced electron–electron interaction[8] and the decrease of electron density n when the volume fraction of magnetic metals was near percolation threshold.

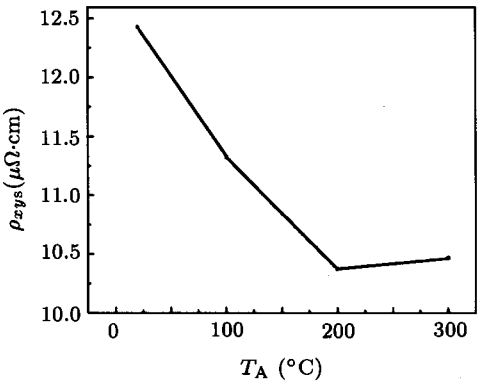


Fig. 4. Saturated Hall resistivity measured at room temperature for $Fe_{45.51}(Al_2O_3)_{54.49}$ nano-granular films annealed at different temperatures up to 300°C.

The giant Hall resistivity in the percolating granular magnetic system suggests a number of interesting applications as Hall sensors. The main advantages of the GHE system are that the Hall element has a linear response with the external field without requiring a bias field, which is necessary in sensors based on the magnetoresistance or the spin valve effect, and that it consists of only one single layer sensing element prepared via a simple sputtering deposition procedure. These two features place the GHE

device into a unique category in various magnetic sensors. From a Hall sensor application point of view, the sensor should have a good thermal stability. Figure 4 shows the saturated Hall resistivity measured at room temperature for $Fe_{45.51}(Al_2O_3)_{54.49}$ nanogranular films annealed at different temperature up to 300°C. Even when the annealing temperature was as high as 300°C, the saturated extraordinary Hall resistivity ρ_{xys} did not decrease much. The thermal stability of $Fe_{45.51}(Al_2O_3)_{54.49}$ nano-granular films was good for temperature as high as 300°C, and thus may be considered as a magnetic field sensor for operating temperatures below 300°C.

In summary, a series of $Fe_{45.51}(Al_2O_3)_{54.49}$ nanogranular films with different annealing temperature were fabricated by using ion-beam sputtering technique, and a saturated Hall resistivity of about 12.5 $\mu\Omega$·cm at room temperature for the as-deposited sample was observed. The $\rho - T$ curve and the TEM image of the as-deposited sample demonstrated that the volume fraction of Fe in granular films was near percolation threshold and indicated that the anomalous giant Hall effect may originate from the percolation. With increasing annealing temperature up to 300°C, the saturated Hall resistivity decreased only a little. The good thermal stability below 300°C made the $Fe_{45.51}(Al_2O_3)_{54.49}$ nano-granular film a plausible candidate as the Hall sensor in the areas requiring relatively high temperatures.

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