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The Hall effect of Co_{0.35}Fe_{0.65}–Al₂O₃ nanogranular films

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Abstract

A series of $(Co_{0.35}Fe_{0.65})_x(Al_2O_3)_{(1-x)}$ (volume fraction) nanogranular films was fabricated using magnetron sputtering. The Hall effect of the films increases as the volume fraction of $Co_{0.35}Fe_{0.65}$ decreases near percolation. The saturated Hall resistivity of $(Co_{0.35}Fe_{0.65})_{0.43}(Al_2O_3)_{0.57}$ is about 5.5 $\mu\Omega$ cm at room temperature. This suggests that the giant Hall effect (GHE) may be due to the percolation phenomenon. The correlation of the Hall effect and tunnelling magnetoresistance (TMR) with the alloy volume fraction is discussed.

The discovery of giant magnetoresistance (GMR) in magnetic multilayers and magnetic metalnonmagnetic metal (M/NM) and tunnelling magnetoresistance (TMR) in magnetic metalnonmagnetic insulator (M/NI) compounds revived the interest in studies of magnetotransport in nanostructured inhomogeneous magnetic solids [1–5]. It is widely believed that the GMR effect originates from the spin-dependent scattering and TMR effect originates from spin-dependent tunnelling. Along with the GMR and TMR, the extraordinary Hall effect has also been studied in transition metal granular solids [6–11]. For the M/NM granular films, the extraordinary Hall resistivity was observed up to 1 $\mu\Omega$ cm, which is greater than the typical values of pure ferromagnets [6]. In M/NI granular films, the extraordinary Hall resistivity can be enhanced when the volume fraction of the magnetic metal decreases near the percolation threshold. For the Ni–SiO₂ system, the saturated Hall resistivity is even enhanced to 200 $\mu\Omega$ cm, which is almost four orders of magnitude greater than that in a pure magnetic metal sample. Pakhomov *et al* [7] named this extraordinary Hall effect the giant Hall effect (GHE), analogizing with the giant magnetoresistance effect. In this paper, we report the giant Hall effect observed in (Co_{0.35}Fe_{0.65})_x(Al₂O₃)_(1-x) nanogranular films, where x means volume fraction.

The $(Co_{0.35}Fe_{0.65})_x(Al_2O_3)_{(1-x)}$ nanogranular films with thickness of about 300 nm were deposited on glass substrates using the magnetron sputtering technique. A mosaic target was employed with bulk $Co_{0.32}Fe_{0.68}$ and Al_2O_3 in which small pieces of $Co_{0.32}Fe_{0.68}$ adhered

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compactly on the plate of Al_2O_3 around the effective sputtering region. The different volume fraction was obtained by changing the number of small pieces of $Co_{0.32}Fe_{0.68}$. It should be noted that the relative component of $Co_{0.32}Fe_{0.68}$ was changed to $Co_{0.35}Fe_{0.65}$ after deposition, determined by energy-dispersive x-ray spectroscopy using an EDAX PV-9100. The magnetism was measured at room temperature with a vibrating sample magnetometer (VSM). The Hall resistivity was measured using the conventional four-terminal method with the applied field perpendicular to the film plane. Two Hall voltages were measured either by reversing the field direction with the fixed sample or by reversing the film by 180° in an oriented field to remove the offset voltage due to the asymmetric Hall terminals. The TMR was measured in the configuration of an applied magnetic field perpendicular to the film with the current in plane using the conventional four-terminal method. All the measurements were made at room temperature.

Figure 1 shows the dependence of Hall resistivity ρ_{xy} on the applied field H for a $(Co_{0.35}Fe_{0.65})_{0.43}(Al_2O_3)_{0.57}$ nanogranular film sample at room temperature. The inset of figure 1 shows the corresponding hysteresis loop with the applied field perpendicular to the film plane. As well known, the Hall resistivity ρ_{xy} of ferromagnetic metals is the sum of two terms, one is the ordinary Hall resistivity due to the Lorentz force, which is proportional to the field, and the other is the extraordinary part originating from the scattering of the polarized conduction d electrons, which is proportional to magnetization [7]. Since the extraordinary Hall coefficient is much greater than the ordinary one, the plots of the Hall resistivity against applied field are s-shaped curves similar to the magnetization curves with a saturated field about 10 kOe. The extraordinary Hall resistivity can be characterized using its absolute value at saturation, ρ_{xys} , which can be determined by a linear extrapolation of the data at high fields to H = 0 [7]. Using this method, the saturated Hall resistivity ρ_{xys} in the (Co_{0.35}Fe_{0.65})_{0.43}(Al₂O₃)_{0.57} nanogranular film sample is about 5.5 $\mu\Omega$ cm.



Figure 1. Hall resistivity ρ_{xy} as a function of applied field *H* for a (Co_{0.35}Fe_{0.65})_{0.43}(Al₂O₃)_{0.57} nanogranular film at room temperature. The inset shows the hysteresis of the same sample with applied field perpendicular to the film plane.

Figure 2 shows the dependence of zero field resistivity ρ_{xx} measured at room temperature on Co_{0.35}Fe_{0.65} volume concentration x. It can be clearly seen that the resistivity ρ_{xx} increases drastically with decrement of x for x below 0.52, indicating that the percolation threshold in the $(Co_{0.35}Fe_{0.65})_x(Al_2O_3)_{(1-x)}$ nanogranular film locates around $x_P \sim 0.52$. Shown in figure 3 is the temperature dependence of resistivity ρ_{xx} in $(Co_{0.35}Fe_{0.65})_{0.43}(Al_2O_3)_{0.57}$ nanogranular film. The resistivity ρ_{xx} decreases with the increment of temperature. After fitting the curve, the ρ_{xx} shows a linear behaviour as log *T*, which is shown in the inset of figure 3. This behaviour is typical of sputtered granular metal close to the metal-insulator transition, due to metallic conduction via a percolation path [7].



Figure 2. The zero field resistivity ρ_{xx} measured at room temperature as a function of Co_{0.35}Fe_{0.65} volume concentration *x* in (Co_{0.35}Fe_{0.65})_{*x*}(Al₂O₃)_(1-x) nanogranular films.



Figure 3. The dependence of ρ_{xx} on temperature of the (Co_{0.35}Fe_{0.65})_{0.43}(Al₂O₃)_{0.57} nanogranular film. The inset shows the behaviour of ρ_{xx} on log *T* with the best fitting.

Figure 4(a) shows the dependence of Hall resistivity ρ_{xy} on *H* of the samples with different $Co_{0.35}Fe_{0.65}$ volume concentration. One can see that the value of ρ_{xy} at a fixed *H* increases as the metal volume fraction decreases to 0.43. At lower $Co_{0.35}Fe_{0.65}$ volume fraction, the Hall resistivity ρ_{xy} of the sample is difficult to measure. Figure 4(b) shows the dependence of ρ_{xx} on *H* for the samples with different $Co_{0.35}Fe_{0.65}$ volume concentration. The TMR effect



Figure 4. Hall resistivity ρ_{xy} (a), and resistivity ρ_{xx} (b) as functions of magnetic field for $(Co_{0.35}Fe_{0.65})_x(Al_2O_3)_{(1-x)}$ nanogranular films with various *x*, as well as both the Hall resistivity and the TMR effect (at H = 12 kOe) as a function of $Co_{0.35}Fe_{0.65}$ concentration (c).

first appears at x = 0.48, and increases with decreasing volume fraction x. So the transport properties of this system can be divided into three regions depending on x, shown as figure 4(c). It is very interesting that in region I (x < 0.43) and region III (x > 0.48) there is only the TMR effect and Hall effect respectively, whereas both the TMR effect and Hall effect coexist in region II (0.43 < x < 0.48). It is well known that the extraordinary Hall effect is due to the scattering of polarized conduction d electrons, while the TMR effect originates from the spin-dependent tunnelling between neighbouring ferromagnetic granules. For x > 0.48, there is only the Hall effect due to the formation of a granule network. For x < 0.43, the granules are separated by insulating matrix, and thus the TMR effect is dominant. For the region of 0.43 < x < 0.48, which is near to percolation, the transport in this system displays both the TMR and Hall effect based on the situation of the coexistence of the separated magnetic granules and the connected ones. In region II, as x decreases, the magnetic granules tend to disconnect and inter-granule connections become narrower, resulting in the increment of the Hall effect.

However, the mechanism of GHE near the percolation threshold in magnetic nanogranular system remains unclear. Recently Zhang *et al* [12] have shown the GHE in a $Cu_x(SiO_2)_{1-x}$ nanogranular film as *x* locates near the percolation threshold. To explain the behaviour in this system, Wan and Sheng [13] proposed a theoretical model on the mechanism of the local interface effect, in which the electron dephasing length is identified to be the intrinsic scale over which coherent electron transport has to be taken into account. According to this theory, the interference component can give rise to positive and negative Hall coefficients with equal probability for all possible random and local microstructures. Effectively, this implies an overall decrease in the charge carrier density and hence an increase in the Hall coefficient. Though the mechanisms of the Hall effect of ferromagnetic metal and nonferromagnetic metal are different, the coherent electron transport may also play an important role in the GHE in M/NI nanogranular films. Further studies are needed.

In summary, a series of $(Co_{0.35}Fe_{0.65})_x(Al_2O_3)_{(1-x)}$ nanogranular films was fabricated using magnetron sputtering. A saturated Hall resistivity of about 5.5 $\mu\Omega$ cm was observed in $(Co_{0.35}Fe_{0.65})_{0.43}(Al_2O_3)_{0.57}$ nanogranular film. The measured $\rho_{xx}-T$ curve suggests that this giant Hall effect (GHE) is due to the percolation phenomenon. The Hall effect in this system increases as the volume fraction of $Co_{0.35}Fe_{0.65}$ decreases near percolation. We believe that the signal level of the Hall effect can be improved by precisely controlling the metal volume fraction to the right percolation region or by producing some artificial structure that makes the magnetic metal particles nano-contact but avoid separating.

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