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Progressive evolution from giant magnetoresistance to anisotropic magnetoresistance in CoNi–Al_2O_3 granular films

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A series of CoNi–Al_2O_3 granular films were prepared with a magnetron controlled sputtering system. Their magnetic-transport properties were measured by the conventional four-probe method. Results indicate that the longitudinal GMR effect reaches a maximum of –2.1% at about 35 vol. % of CoNi. Meanwhile, the GMR effect is dominant when the volume fraction is less than about 50 vol. %, whereas AMR predominates in films when the volume fraction is larger than about 50 vol. %. This suggests that there exists a progressive evolution from GMR to AMR through the percolation region in CoNi–Al_2O_3 granular films.

Key words: *CoNi–Al_2O_3 granular film; giant magnetoresistance; anisotropic magnetoresistance*

1. Introduction

Extensive studies of spin-dependent transport properties were triggered recently by the discovery of the so-called giant magnetoresistance (GMR) effect. This novel effect which was first discovered in the magnetic Fe/Cr multilayer [1] and later in heterogeneous granular films with ferromagnetic granules embedded in a non-magnetic metallic matrix [2, 3] shows that the resistance of films could exhibit large changes – of the same order of magnitude as the resistance itself – upon applying a magnetic field and has potential technical applications in the information industry. Although the exact mechanism of GMR remains elusive, it is attributed to spin-dependent scattering being dominant at the interface between ferromagnetic granules and a matrix, as well as within the ferromagnetic granules [4]. Another direction in the research of spin-dependent transport phenomena in heterogeneous granular films is granular systems where small ferromagnetic granules are embedded in an immis-

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cible insulating matrix. Giant magnetoresistance and the relative magnetic properties of a mixture depend on the volume fraction of ferromagnetic granules. At low-volume fractions, ferromagnetic granules are isolated from each other and electric transport proceeds by intergranular tunnelling, thus in these systems the GMR effect is also called tunnelling magnetoresistance (TMR). When the volume fraction of ferromagnetic granules increases above a certain threshold, individual granules will form a network structure [5] with a continuous metallic conductance path. In the latter, the films will exhibit anisotropic magnetoresistance (AMR), an effect first discovered in ferromagnetic metals by Willam Thomson in 1857 [6] and used in magnetic field detectors for digital recording and magnetic bubbles.

Being different from the mechanism of GMR or TMR, the theoretical basis of AMR takes into account spin-orbit interaction and the d band splitting commonly found in bulk ferromagnetic materials [7]. Therefore, the GMR (or TMR) and AMR effects should have a certain relationship in granular films. Until now, however, little work has been done to study the relationship between GMR and AMR. Since CoNi –Ag alloys can exhibit both AMR and GMR effects [8] and Co–Al_2O_3 granular films can show a typical GMR effect [9], a new CoNi–Al_2O_3 granular system can be expected to be a good candidate for understanding the relationship between GMR and AMR. In this paper, we reported the transport properties of a series of CoNi–Al_2O_3 granular films. We found that they exhibit mainly GMR effects below the ferromagnetic percolation threshold and AMR effect above the ferromagnetic percolation threshold, and at the passage of the percolation threshold there is a progressive evolution of TMR to AMR.

2. Experimental procedure

The CoNi–Al_2O_3 granular films were sputtered onto a glass substrate at room temperature (300 K) with an spc350 multi-target magnetron controlled sputtering system. A CoNi target (the weight ratio of Co and Ni 1:1) and Al_2O_3 target (99.9% purity) were separately installed on two independently controlled R.F. cathodes, and were alternatively employed to sputter the films. The thickness of films for MR measurement was about 300 nm as measured by α-step meter. The volume fraction of CoNi in the CoNi–Al_2O_3 films was controlled by changing the sputtering power of the CoNi target and was determined by an energy dispersive spectrum (EDS) obtained with a JEM-2010 transmission electron microscope (TEM). The base pressure was about 3×10^{-3} Pa and the sputtering gas (Ar) pressure was 1.7×10^{-1} Pa. The MR effect was measured by the conventional four-probe method for applied fields up to 1.25 T.

3. Results and discussion

The variation of resistivity of CoNi–Al_2O_3 granular films as a function of volume fraction of granules was measured without any applied field at room temperature, as shown in Figure 1. The resistivity can change by over four orders of magnitude when the volume fraction of ferromagnetic granules changes from about 17 vol. % to 65 vol. %. With the increasing volume fraction of granules, the resistivity decreases slowly at first, and changes sharply at about 50 vol. % when the percolation threshold of granular films is crossed [10], indicating that conduction proceeds by metallic conducting over a ferromagnetic network above 50 vol. % and by tunnelling conduction below 50 vol. %.

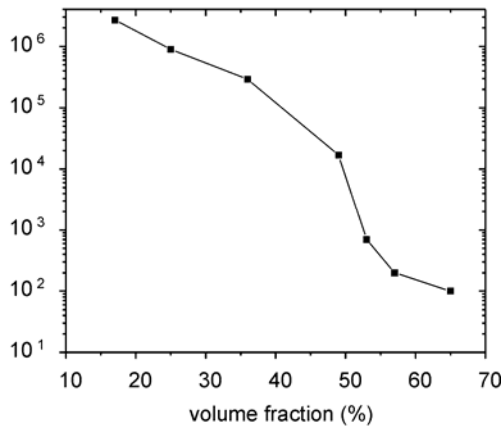

Fig. 1. Measured room resistivity vs. volume fraction of granules in CoNi–Al_2O_3 films

As mentioned above, it is clear that the conduction mechanism of CoNi–Al_2O_3 granular films varies with the volume fraction of granules, leading to the conclusion that the GMR or AMR effect dominates in granular films at different volume fractions. Figure 2 shows the dependence of longitudinal GMR and transverse GMR on the applied field for various volume fractions of CoNi granules. When the volume fraction of granules is less than about 49%, the longitudinal MR is negative (Fig. 2a–c), and for volume fractions above 49% it is positive (Fig 2d, e) except for a little part of negative MR at higher applied fields (Fig. 2d). On the other hand, transverse GMR is always negative in the above films.

Figure 3 shows the dependence of longitudinal GMR on the volume fraction of granules. Here, longitudinal GMR effects are calculated according to

$$\mathrm{GMR} = \frac{\Delta\rho}{\rho} = \frac{\rho(H) - \rho(0)}{\rho(0)}$$

It is clear that longitudinal GMR first increases up to about –2.1% at the volume fraction of about 35% CoNi, and then decreases with the increasing volume fraction. Being different from longitudinal GMR, however, the AMR effect increases monotonically with increasing volume fraction, as seen in Figure 4. In this case

$$\mathrm{AMR} = \frac{\rho_{\parallel}(H) - \rho_{\perp}(H)}{\rho_{\parallel}(0)/3 + 2\rho_{\perp}(0)/3}$$

where $\rho(H)$ and $\rho(0)$ stand for the resistivities in the applied field and initial zero field, respectively, and $\rho_{\parallel}$ and $\rho_{\perp}$ represent resistivities with the current parallel and perpendicular to the applied field in the plane of the samples [7].

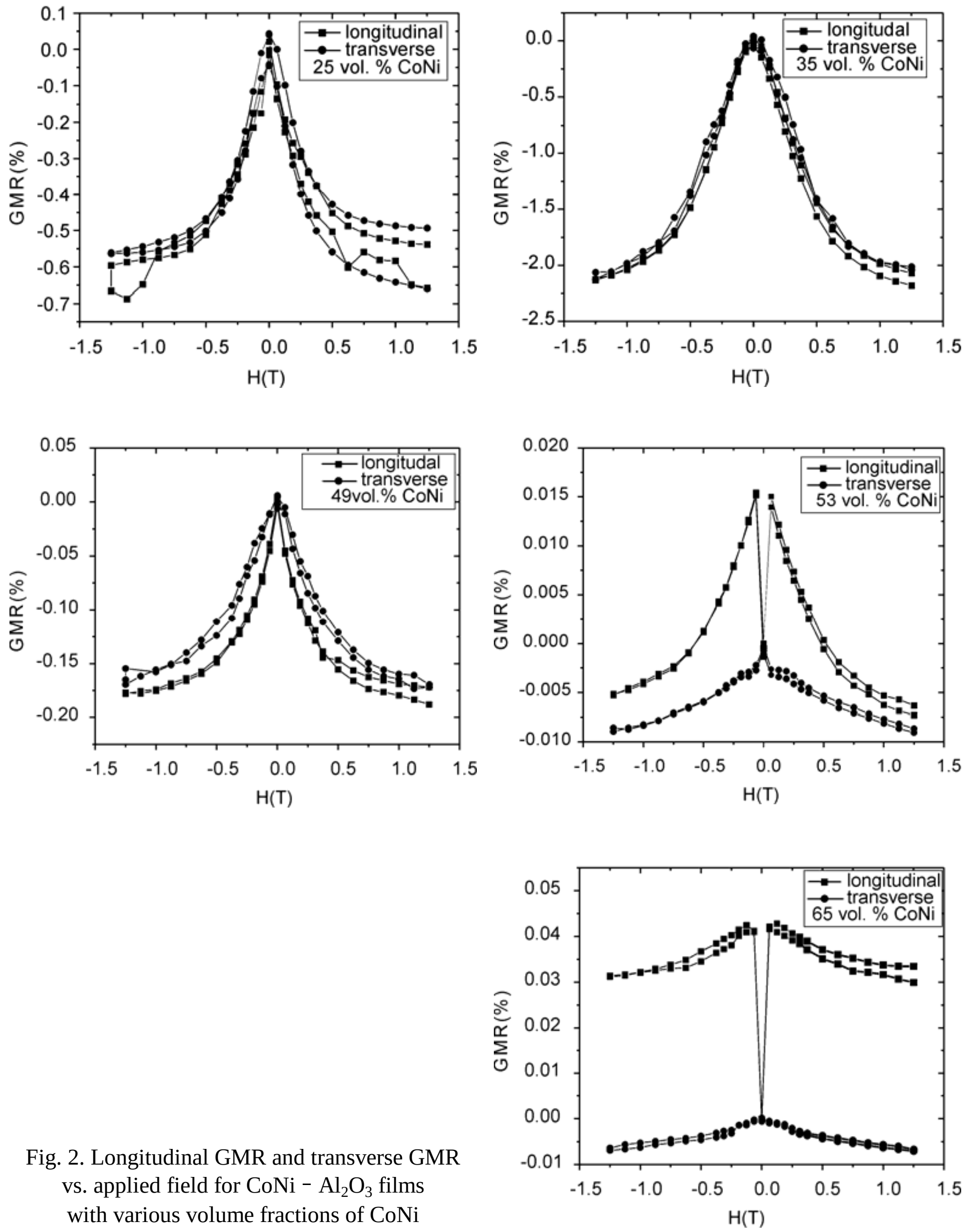

Fig. 2. Longitudinal GMR and transverse GMR vs. applied field for CoNi – Al_2O_3 films with various volume fractions of CoNi

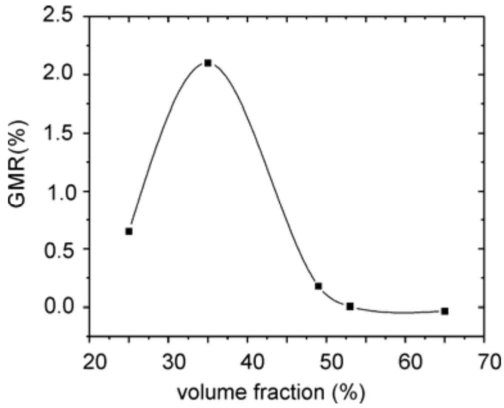

Fig. 3. Dependence of longitudinal GMR on the volume fraction of CoNi granules in CoNi–Al_2O_3 films

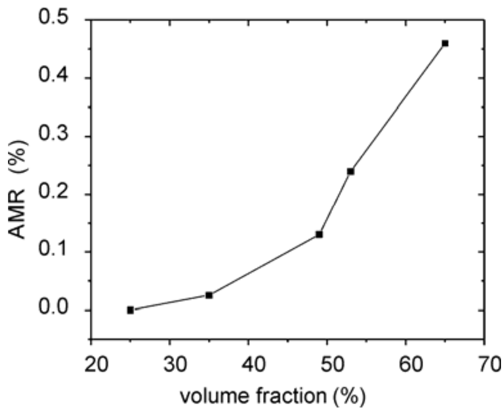

Fig. 4. Dependence of AMR on the volume fraction of CoNi granules in CoNi–Al_2O_3 films

All values of GMR and AMR were measured at H = 12.5 kOe (1×10^6 A/m). Comparing Figure 3 with Figure 4, it is apparent that in samples with lower volume fractions of granules, longitudinal GMR is negative while the AMR effect is negligible, indicating that GMR dominates in magnetic granular films. For the volume fractions of granules larger than 35 vol. %, the longitudinal GMR decreases, while AMR increases gradually and reaches 0.46% in our experiments at the volume fraction of 65 vol. %, indicating that the AMR effect predominates in films with higher volume fractions. The AMR effect will tend to the value of CoNi bulk alloy with increasing volume fraction, which can be expressed as follows. It is obvious that when the volume fraction of granules is larger than the percolation threshold, all CoNi particles coalesce and gradually form a connecting network with metallic conductance, and as a consequence, GMR vanishes. In the case of a connecting network, however, spin-orbit coupling can be enhanced as in the CoNi ferromagnetic bulk, and thus AMR dominates in granular films. When the volume fraction of granules gradually falls below the percolation threshold, electric transport will start to be governed by the tunnel current between CoNi particles separated by the Al_2O_3 matrix [11], since the connecting network is broken and large multidomain CoNi granules are separated into small single-domain or superparamagnetic granules. It follows that when these magnetic particles are magnetized under an applied field, the GMR effect will appear due to the spin-dependent tunnelling effect, and increase with decreasing granule volume fraction until a peak value is reached. After this, as the volume fraction decreases further, CoNi particles are far apart, i.e. the thickness of Al_2O_3 tunnelling barriers become larger than the spin diffusion length, and the spin-flip tunnelling process may occur, giving rise to a drop in GMR [12]. On the other hand, spit-orbit coupling gradually weakens due to granules being separated when their volume fraction is less than the percolation threshold, leading to a weak AMR effect.

4. Conclusions

In summary, in CoNi–Al_2O_3 films, the longitudinal GMR effect reaches a maximum of –2.1% at about 35 vol. % CoNi and the GMR effect is dominant when the volume fraction is less than about 50 vol. %, whereas AMR predominate in films when the volume fraction is larger than about 50 vol. %. This suggests that a progressive evolution from GMR to AMR occurs in the percolation region of CoNi–Al_2O_3 granular films.

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