AIP Applied Physics Letters

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Citation: Applied Physics Letters **76**, 3968 (2000); doi: 10.1063/1.126837
View online: http://dx.doi.org/10.1063/1.126837
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/76/26?ver=pdfcov
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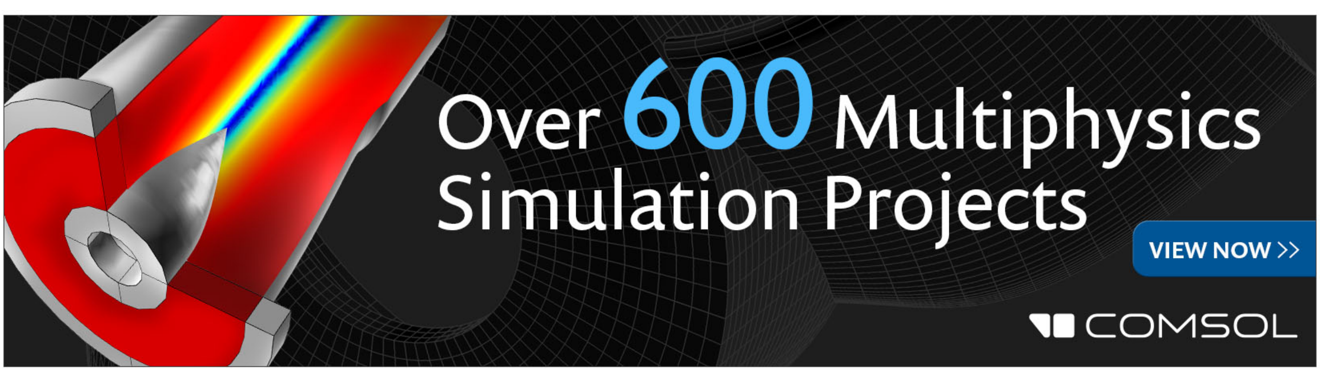

Direct measurement of the dependence of granular giant magnetoresistance on the relative orientation of magnetic granules

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(Received 22 March 2000; accepted for publication 28 April 2000)

Experiments have been designed to vary the relative angle between the magnetic moments of different Co granules in $Cu_{80}Co_{20}$ granular system. The moments of granules are mostly aligned in the same direction by field cooling to low temperature in a high magnetic field. A small field applied at an angle relative to the cooling field rotates the moments of a portion of the granules that have small particle size and coercivity. It is found that the giant magnetoresistance (GMR) varies linearly with $\cos\phi$, where ϕ is the relative angle between the magnetic axes of granules. This behavior disappears if the sample is cooled in zero fields, or if the rotating field is too large or small, or if the measuring temperature is higher than the blocking temperature. Our results show that the GMR in granular structures has the same angular dependence as the layered films and confirm the existing theories and recent microscopic models of granular GMR suggesting a crucial role of the relative orientations of the magnetic granules in determining the spin dependent scattering. © *2000 American Institute of Physics.* [S0003-6951(00)01326-7]

Granular magnetic systems, where magnetic metallic particles are embedded in nonmagnetic metallic host, show a remarkable negative giant magnetoresistance (GMR). Since Berkowitz *et al.*[1] and Xiao *et al.*[2] reported the GMR found in Cu–Co granular systems in 1992, this effect has attracted a lot of interest. It is known[2–6] that GMR depends on granular size and distribution and is suppressed if the electron mean-free path is reduced. Generally the magnetic granules are treated as single domains and the main mechanism of GMR is usually analyzed in terms of the spin dependent scattering at the granule surfaces or within the granules. Several theoretical studies have been done in granular systems.[7–12] However, more detailed theoretical works are still needed to understand the spin dependent scattering in these particle-matrix systems. For example, in granular systems, the space distribution of magnetic field, the electron spin diffusion process and the spin transitions are much more complex than the multilayered systems.

Basically, GMR can be descried as the result of the alignment of magnetic granules by the external magnetic field, which reduces the spin dependent scattering of the system. A general observation is $\mathrm{GMR}\propto(M/M_S)^2=\langle\cos\theta\rangle^2$, where M is the global magnetization, M_S is the saturation magnetization, θ is the angle between the magnetic moment of a granule and the external field, and $\langle\cos\theta\rangle$ is the average value of $\cos\theta$.[2,3] This angular dependence has been explained by Zhang *et al.*[7,11] and Asano *et al.*[8] based on the spin dependent scattering at the granule-matrix interfaces.

The $\langle\cos\theta\rangle^2$ dependence also suggests[2] that the GMR is related to the *relative orientations* of the magnetic granules. This is a generally accepted concept and is experimentally demonstrated in trilayer or multilayered magnetic films, where the GMR varies linearly with the cosine of the relative angle between the moments of the magnetic layers.[13–20] It can be easily shown that, in a system of random distribution, $\langle\cos\theta_{ij}\rangle=\langle\cos\theta\rangle^2$, where ϕ_{ij} is the relative angle between magnetic axes of uncorrelated magnetic granules. Pogorelov *et al.*[12] have introduced a microscopic theory of granular GMR and have shown that the spin dependent conductance in granular systems can be sensitive to the short-range magnetic order in addition to the long-range order. It is of interest to measure directly the resistance as a function of the relative orientation of the magnetic granules and find out the relationship between the two. As mentioned earlier, in spin-valve type structures, the linear dependence of MR on the cosine of the angle between the magnetic moments of the two ferromagnetic layers has been well documented with both experimental results[13–17] and theoretical models.[18–20] In those experiments, the angle between the magnetization of the two ferromagnetic layers is changed by applying a suitable magnetic field that rotates one layer but not the other. However, the same method does not work and controlling the relative angle between magnetic moments is difficult in the granular systems.

In this work, we have designed an experiment in which one can statistically adjust the relative angle ϕ between the magnetic moments of different granules in $Cu_{80}Co_{20}$. It is found that GMR varies linearly with $\cos\phi$. The experiments were conducted as follows. First, the granular film was field cooled (FC) down to $T=5$ K, in a high field $\boldsymbol{H}_1=5$ T. This way, the magnetic moments of granules were pointed in their easy axes close to the field direction $\boldsymbol{H}_1$ and on average were aligned in the same direction as $\boldsymbol{H}_1$. Second, a small field $\boldsymbol{H}_2$ was applied at an angle ϕ relative to $\boldsymbol{H}_1$ after $\boldsymbol{H}_1$ was removed. Since there is a size distribution of the granules with a corresponding distribution in the coercivity, granules having high coercivity may stay with the initial field $\boldsymbol{H}_1$ and those having low coercivity rotate with $\boldsymbol{H}_2$. Thus, one can

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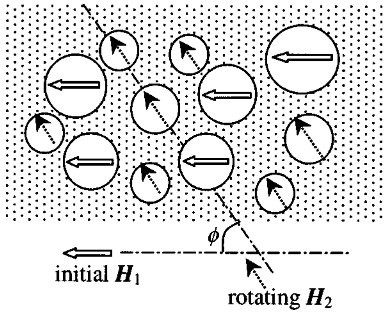

FIG. 1. Illustration of changing the relative orientations of magnetic granules.

rotate the magnetic moments of a fraction of the granules by choosing an appropriate value of $\boldsymbol{H}_2$. Figure 1 gives a schematic view of the process. One may roughly divide the granules into two groups: those rotating with $\boldsymbol{H}_2$ and those staying in the original direction of $\boldsymbol{H}_1$. The angle between the moments of two groups of granules is exactly the angle between $\boldsymbol{H}_1$ and $\boldsymbol{H}_2$. Here we assume the interparticle interaction is weak and ignore the effects of those granules that may not follow exactly one of the two directions.

The samples used in our experiments are typical GMR granular Cu–Co films. $Cu_{80}Co_{20}$ films were deposited by vacuum magnetron sputtering from a composition target. The film thickness was 80 nm and the GMR value was ~10% at $T=5$ K. Samples were cut into 1 mm×4 mm rectangles. Four-terminal direct current resistivity measurement was performed using a Quantum Design physical properties measurement system. Magnetic susceptibility and magnetization measurements were made with a Quantum Design superconducting quantum interference device. Figure 2(a) shows the susceptibility χ versus temperature T curves for both zero filed cooled (ZFC) and FC runs. It indicates a blocking temperature T_B of about 40 K and implies the existence of size distribution of the Co granules with small and large particles. The inset shows the MR ratio of the $Cu_{80}Co_{20}$ granular film at 5 K. It does not saturate even in a high field of $H=6$ T, which suggests the Co granules have a wide size distribution. Figure 2(b) shows the magnetization curve at 300 K, above the blocking temperature. The shape of the curve is consistent with the Langevin function associated with a distribution of granule size. The inset gives the low field hysteresis loop at 5 K, and it indicates coercivity H_C of about 200 Oe.

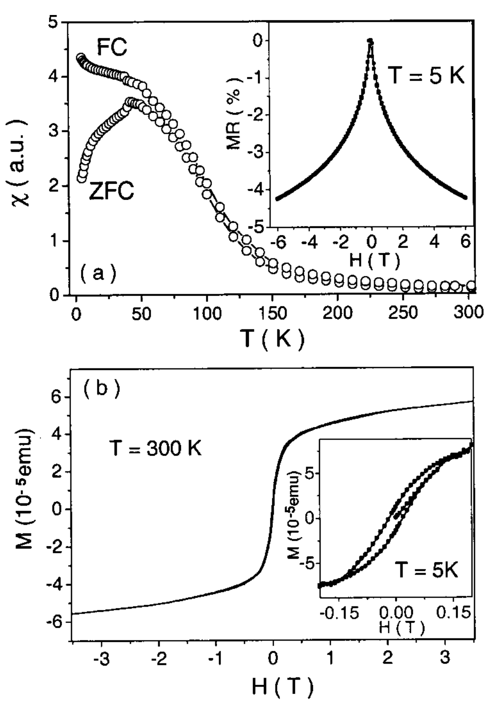

FIG. 2. (a) Magnetic susceptibility χ as a function of temperature. The measuring field equals to 500 Oe. Inset shows the MR at 5 K. (b) Magnetization curve at 300 K; inset shows the low field hysteresis loop at 5 K.

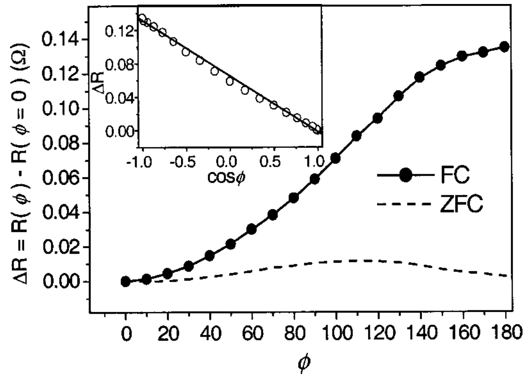

FIG. 3. Resistance as a function of ϕ and cos ϕ (inset) at $T=5$ K, sample is FC in $\boldsymbol{H}_1=5$ T. Rotating field $\boldsymbol{H}_2=600$ Oe. Dashed line shows the same measurement after ZFC and the cos ϕ dependence disappears.

In Fig. 3, the solid line and symbol shows the MR dependence on angle ϕ. The experimental data fit well to the cos ϕ function in the entire range between 0° and 180°, and the inset shows the resistance is linearly dependent on cos ϕ. The resistance of the sample R can be expressed as

$$R \approx 31.201 + 0.065(1-\cos\phi)(\Omega). \quad (1)$$

The same measurement has been repeated for a ZFC sample, and it is found that the cos ϕ dependence of the GMR disappears, as shown in Fig. 3 (dashed line). Since the moments of the ZFC sample are blocked in random directions, a cos ϕ dependence is not expected. Rather the MR should be independent of ϕ, which is consistent with our experiment. This result lends support to that the cos ϕ dependence of GMR for FC is due to the change in the angle between the moments of those granules rotating with $\boldsymbol{H}_2$ and those fixed in $\boldsymbol{H}_1$. Our result confirms that the GMR depends on the relative orientations of the magnetic granules.

The field strength of $\boldsymbol{H}_2$ has been varied and the earlier experiment repeated. It is found that when $\boldsymbol{H}_2$ is either too low or too high the cos ϕ dependence of the GMR is weak, and the optimal field of $\boldsymbol{H}_2$ is around $H_{2C} \approx 650$ Oe at which the GMR is most sensitive to the change of ϕ. This is not difficult to understand: when $\boldsymbol{H}_2$ is too small (<100 Oe), only a small portion of the granules with very small particle sizes can be rotated and most of the granules stay in the initial orientation of $\boldsymbol{H}_1$. The MR change is small due to the small number of those involved in the scattering associated with the angle change. On the other hand, when $\boldsymbol{H}_2$ is too large (>1000 Oe), most of the granules will align with the rotating $\boldsymbol{H}_2$ and it also reduces the contribution from the scattering due to the relative orientations of the granules. Figure 4 shows the $\Delta R = R(\phi) - R(\phi=0)$ as a function of ϕ

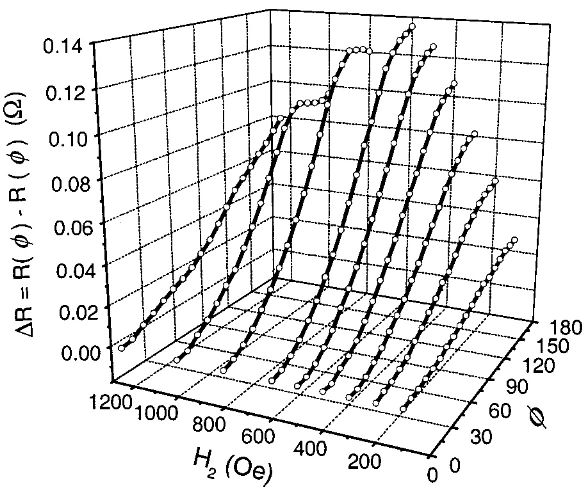

FIG. 4. The cos ϕ dependence of GMR at $T=5$ K in different rotating field $\mathbf{H}_2$.

measured at different rotated field $\boldsymbol{H}_2$. One can see that the cos ϕ dependence of GMR weakens when $\boldsymbol{H}_2$ is greater than H_{2C}. This result is significant in that it indicates the angle ϕ one measures in our experiment is that between the moments of two groups of the magnetic granules and GMR varies linearly with the cosine of that angle. In order to observe a significant cos ϕ dependence of the GMR, one needs to chose a rotating field $\boldsymbol{H}_2$ of reasonable strength and try to balance the number of granules of the two groups: those staying in the initial field direction $\boldsymbol{H}_1$ and those rotating with $\boldsymbol{H}_2$.

It has been found in our experiment that the cos ϕ dependence of the GMR becomes weaker when the temperature is higher and disappears when temperature is higher than the blocking temperature. Figure 5 shows the GMR as a function of ϕ at different temperatures with $\boldsymbol{H}_2=600$ Oe. This result is expected considering that the coercivity of the system in the blocked state decreases with increasing temperature and becomes zero at the blocking temperature. Raising the temperature is equivalent to increasing the rotating field $\boldsymbol{H}_2$ in that it is easier to rotate the majority of the magnetic granules.

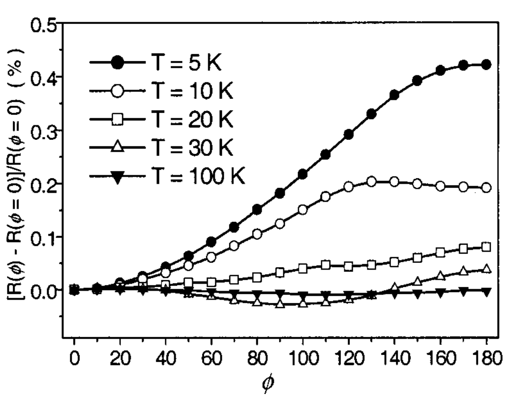

FIG. 5. The cos ϕ dependence of GMR at different temperatures, rotating field $\boldsymbol{H}_2=600$ Oe.

In summary, using a uniquely designed experiment, we have demonstrated a linear dependence of granular GMR on the cosine of the relative orientation between the magnetic axes of granules in the particle-matrix systems. This is similar to the linear dependence found in layered films, where the relative angle between the magnetic layers is easily controlled. The result confirms the existing theories and recent microscopic models of granular GMR and provides direct experimental proof that the relative orientations of the magnetic moments of the granules is a crucial factor in determining the spin dependent scattering in granular GMR materials.

The authors would like to thank Dr. L. Malkinski and Dr. J. Q. Wang for helpful discussions. This work was supported by DOD/DARPA Grant No. MDA972-97-1-0003 through AMRI/UNO.

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