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Citation: *Journal of Applied Physics* **99**, 08J108 (2006); doi: 10.1063/1.2165588

View online: <http://dx.doi.org/10.1063/1.2165588>

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# Large room-temperature spin-dependent tunneling magnetoresistance in a $\text{Fe}_3\text{O}_4$ -polymer composite system

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(Presented on 3 November 2005; published online 24 April 2006)

Powders of polymer and  $\alpha\text{-Fe}_2\text{O}_3$  nanoparticles were pressed into pellets. The samples were annealed at 250 °C in pure hydrogen flow. X-ray diffraction spectra indicate that there is a complete phase transformation from  $\alpha\text{-Fe}_2\text{O}_3$  to  $\text{Fe}_3\text{O}_4$ . Giant negative magnetoresistance (MR) was observed at room temperature and the MR ratio is over 14% in an applied field of 5.5 T. The maximum MR ratio is higher than 22% at 130 K. The drastic enhancement of the MR ratio is attributed to the fact that the polymer is an excellent barrier material and, more importantly, prevents the oxidation of the surface of  $\text{Fe}_3\text{O}_4$ . Our results clearly suggest that there is a high degree of spin polarization at room temperature for half-metallic  $\text{Fe}_3\text{O}_4$ . © 2006 American Institute of Physics.

[DOI: [10.1063/1.2165588](https://doi.org/10.1063/1.2165588)]

## I. INTRODUCTION

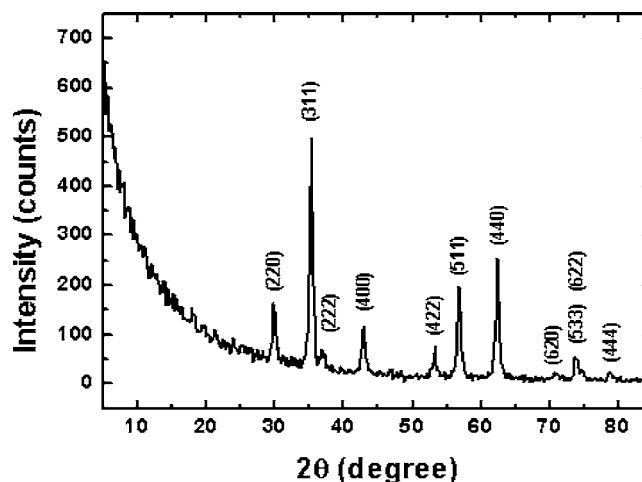
There has been much interest in the half-metallic magnetite  $\text{Fe}_3\text{O}_4$  as its highly spin polarized nature (supposedly  $\sim 100\%$ ) and high Curie temperature ( $T_C \sim 840$  K) are desirable for spin electronic devices.<sup>1,2</sup> Principally, high spin polarization should result in significant tunneling magnetoresistance (TMR), because TMR arises from spin-dependent transport between ferromagnetic (FM) particles of granular solids or FM electrodes of tunnel junctions, and is proportional to the spin polarization of the tunneling electrons.<sup>3–5</sup> Research activities have been focusing on the magnetoresistance (MR) in the magnetite of different forms including single crystals, epitaxial and polycrystalline films, powders, and tunnel junctions involving  $\text{Fe}_3\text{O}_4$  as electrodes.<sup>6–10</sup> Unfortunately, the large MR expected has not been observed in  $\text{Fe}_3\text{O}_4$ , especially at room temperature. In fact, it is well known that, in polycrystalline specimens and powder compacts of magnetic oxides, the surfaces and interfaces have rather different magnetic properties than the bulk.<sup>11–13</sup> This is a natural consequence of the off-stoichiometry, strain, defects, and bonding effects located in the area close to the surface and interface. The surface and interface thus have dramatically reduced spin polarization. In this paper we report, by using polymer polystyrene (PS) as a coating layer, that the TMR effect has been greatly enhanced to 14% in an applied field of 5.5 T at room temperature.

## II. RESULTS AND DISCUSSION

Powders of polymer PS and  $\alpha\text{-Fe}_2\text{O}_3$  nanoparticles (1:2 in weight ratio) were mixed together by first dissolving the polymer in dimethylbenzene, then adding  $\alpha\text{-Fe}_2\text{O}_3$  particles

and stirring, and finally evaporating the solvent. The samples were annealed at 200 °C in pure hydrogen flow, and then pressed into pellets under a pressure of  $5 \times 10^8$  N/m<sup>2</sup>. The samples were annealed again at 200 and 250 °C in pure hydrogen flow.

X-ray diffraction spectra were collected with a Philips X'Pert diffractometer using Cu  $K\alpha$  radiation. Figure 1 shows the diffraction pattern of our sample. It indicates that there is a complete phase transformation from  $\alpha\text{-Fe}_2\text{O}_3$  to  $\text{Fe}_3\text{O}_4$  after annealing in hydrogen. The magnetization  $M$  of the reduced sample shows the unique character of the Verwey transition (a sudden drop in  $M$  at the transition), which shows that the sample has transformed from  $\alpha\text{-Fe}_2\text{O}_3$  to  $\text{Fe}_3\text{O}_4$ . Scanning electron microscopy (SEM) was done with a LEO 1530 VP field-emission SEM. Figure 2 is a SEM image of our sample. It shows that  $\text{Fe}_3\text{O}_4$  particles are spherical and their size is between 10 and 30 nm. They are dispersed in the

FIG. 1. X-ray diffraction pattern of polystyrene-coated  $\text{Fe}_3\text{O}_4$ .<sup>a)</sup>Electronic mail: [jtang@uno.edu](mailto:jtang@uno.edu)

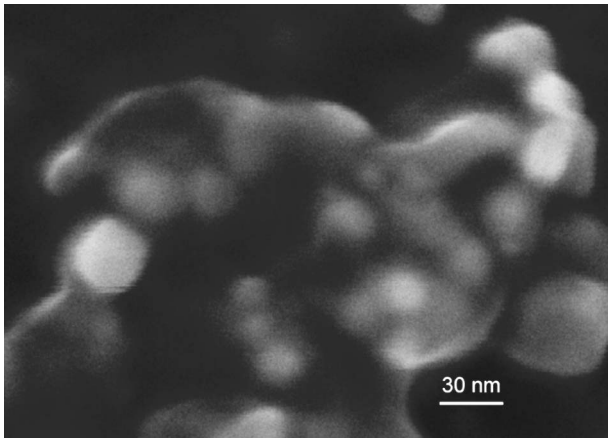


FIG. 2. Field emission SEM image of polystyrene-coated Fe<sub>3</sub>O<sub>4</sub>.

PS matrix. Some particles are close but not connected to each other. This implies that tunneling of electrons between Fe<sub>3</sub>O<sub>4</sub> particles is possible.

The resistance (*R*) of sample measured with a standard four-point method is shown in Fig. 3(a). The change in the resistance at Verwey transition is not as sharp as those observed in some single crystal samples. In Fig. 3(b), *R* is plotted in a logarithmic scale as a function of  $T^{-1/2}$ . The good linear behavior suggests intergranular tunneling is the mechanism of transport.

Magnetoresistance of the sample was measured in a Quantum Design physical property measurement system up

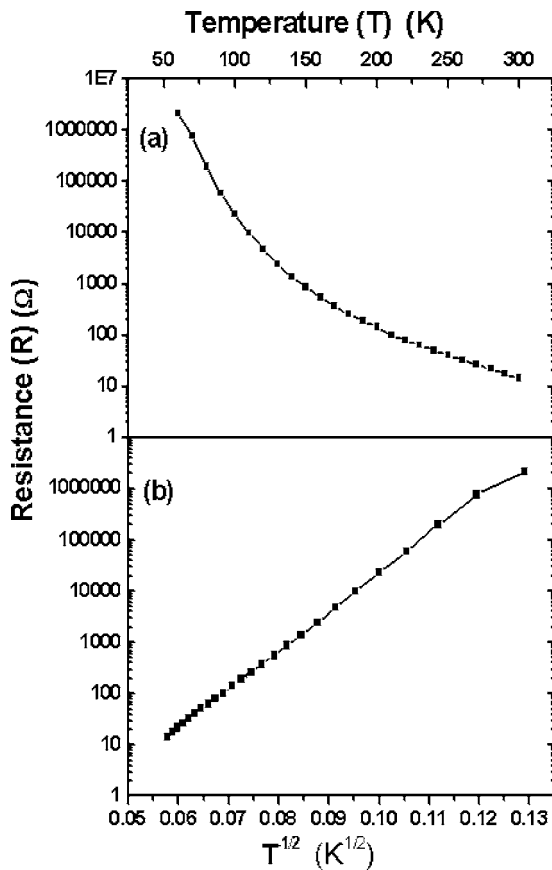


FIG. 3. Resistance as a function of temperature: (a) *R*–*T* plot, (b) *R*– $T^{-1/2}$  plot. *R* is plotted in a logarithmic scale.

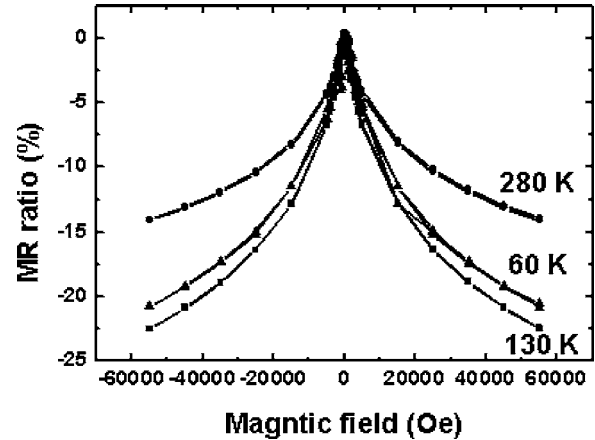


FIG. 4. Magnetoresistance as a function of the magnetic field at 60, 130, and 280 K, respectively.

to a magnetic field of 5.5 T. The magnetic field was applied perpendicular to the current. Figure 4 shows the  $MR = (R_H - R_0)/R_0$  of the sample at 60, 130, and 280 K. Giant negative MR was observed at room temperature and the MR ratio is over 14% in an applied field of 5.5 T. The MR ratio is higher than 22% at 130 K; however, it slightly decreases to 20% at 60 K. These MR values are much higher than reported data in pressed Fe<sub>3</sub>O<sub>4</sub> powders and polycrystalline films,<sup>9,14–16</sup> which have MR ratios typically near 4%–5% or lower at room temperature. It is well known that the surface state of Fe<sub>3</sub>O<sub>4</sub> is different from the bulk state. X-ray photoelectron spectroscopy (XPS) study indicates that the top two layers of the surface are rich in Fe (III) compared to the bulk in molecular beam epitaxy- (MBE) grown Fe<sub>3</sub>O<sub>4</sub> film.<sup>17</sup> However, in our sample, PS is coated on the surface of Fe<sub>3</sub>O<sub>4</sub>. PS is a very good insulator and contains no oxygen. Coating with PS will help prevent the oxidation of the surface of Fe<sub>3</sub>O<sub>4</sub>, which is believed to alter the half-metallic state at the surface. The drastic enhancement of the MR ratio clearly suggests that there is high degree of spin polarization at room temperature for half-metallic Fe<sub>3</sub>O<sub>4</sub>.

Figure 5 shows the temperature dependence of MR ratio in an applied field of 5.5 T. In Fe<sub>3</sub>O<sub>4</sub> there is a Verwey transition that is characterized by an increase in resistivity by about two orders of magnitude at the transition temperature

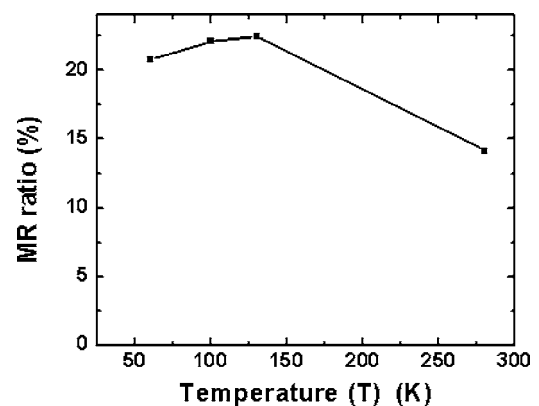


FIG. 5. Temperature dependence of the MR ratio in an applied field of 5.5 T.

$T_v \sim 120$  K. In our sample, MR ratios before and after the Verwey point do not have significant change. This implies that the Verwey transition does not change the spin polarization of  $\text{Fe}_3\text{O}_4$  significantly. The slight drop of MR ratio below the Verwey point may probably be attributed to the rapid increase of resistivity of  $\text{Fe}_3\text{O}_4$ , which will diminish the spin-dependent tunneling current.

## ACKNOWLEDGMENT

This work was supported by Sharp Laboratories of America and Louisiana Board of Regents Support Fund Grant No. LEQSF(2004-07)-RD-B-12.

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