Magnetic field dependence of voltage-current characteristics of Fe_3O_4 thin films at room temperature

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Fe_3O_4 thin films have been fabricated on glass substrate by the facing-target sputtering technique and their field-modulated voltage-current behavior was investigated. The nonlinear dependence of voltage on current density displays a switching from high-resistivity to low-resistivity states above a threshold current density. The low-resistivity state is very sensitive to the applied magnetic field, and a large negative magnetoresistance of ~−27% is observed at 300 Oe under a high current density of 100 A cm^{-2} at room temperature. Furthermore, the dependence of the magnetoresistance on the magnetic field reveals a good linear relationship. The observed results seem to favor a picture of spin-polarized intergrain tunneling through the grain boundaries. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2171791]

The half-metallic materials have received increasing attention because of their potential application in future spintronic devices. Band structure calculation for magnetite (Fe_3O_4) shows that only minority spin (spin-down) electrons are present at the Fermi level,[1,2] indicating that Fe_3O_4 is half-metallic and the conduction electrons are 100% spin polarized. Early studies by Feng *et al.* revealed a negative magnetoresistance (MR) of −7.5% at about 130 K and 2.3 T in a polycrystalline thin-film sample.[3] Here, MR is defined according to $\Delta\rho/\rho=(\rho_H-\rho_0)/\rho_0$, where ρ_0 is the zero-field resistance and ρ_H is the resistance in the applied magnetic field H. Li *et al.* found the magnitude of MR was quite small for Fe_3O_4 films grown on (100)-oriented MgO and $SrTiO_3$ substrates.[4] In particular, the MR exhibits a monotonic increase with decreasing temperature below 105 K and reaches a value of 32% at 40 kOe and 60 K.[5] Recently, a MR of −7.4% has been observed in the polycrystalline Fe_3O_4 films in an applied magnetic field of 50 kOe at RT.[6] In this letter, we report the strong modifying effect of magnetic field (H) on the voltage-current density ($V-J$) characteristics of Fe_3O_4 film. The application of a high density current above a threshold value causes resistivity jumps. This low-resistivity state is sensitive to the magnetic field and produces a large negative MR effect. Negative MR as high as −27% have been observed under a field of 300 Oe at RT.

A 100-nm-thick Fe metal film was deposited on glass substrate at a sputtering rate of 0.092 nm/s by a facing-target sputtering technique at RT.[7,8] The Fe_3O_4 thin film was then obtained by *in situ* plasma oxidation at about 350 °C in an oxygen atmosphere of 8 Pa. Following oxidation, the film was cooled to RT in a vacuum of 2×10^{-3} Pa. The structure, magnetic property, and $V-J$ behavior of the as-grown thin film were characterized by x-ray diffraction (XRD) and atomic force microscopy (AFM), superconducting quantum interference device magnetometer, and dc four-probe techniques, respectively.

Figure 1 shows the $\theta-2\theta$ scanning profile of an XRD experiment for the Fe_3O_4 film using the Cu K_α radiation. Careful XRD phase analysis determines that the only peak observed corresponds to the (531) orientation of the Fe_3O_4 phase rather than some orientations of other Fe oxides. The rocking curve obtained on this (531) peak shows a full width at half maximum (FWHM) of ~0.118°. The surface morphology was further examined by AFM (inset of Fig. 1). The root-mean-square surface roughness is ~6.8 nm averaged over an area of 1×1 μm^2. The average grain size, D ~80 nm, has been determined by the linear intercept method, i.e., from the number of the line crossings, which is consistent with the calculated result of Scherrer formula, $D=k\lambda/\mathrm{FWHM}\cdot\cos\theta$, inserting $k=0.89$, $\lambda=0.15405$ nm, FWHM=0.118°, and $\theta=32.65°$.

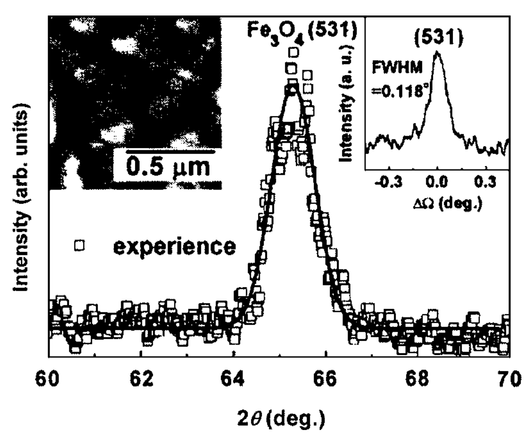

FIG. 1. Typical XRD $\theta-2\theta$ spectrum of the Fe_3O_4 thin film grown on a glass substrate. The smooth solid curve is draw for clarity. Left inset shows the AFM image of the film (image size: 1×1 μm^2). The rocking curve of the (531) peak is shown in the right inset.

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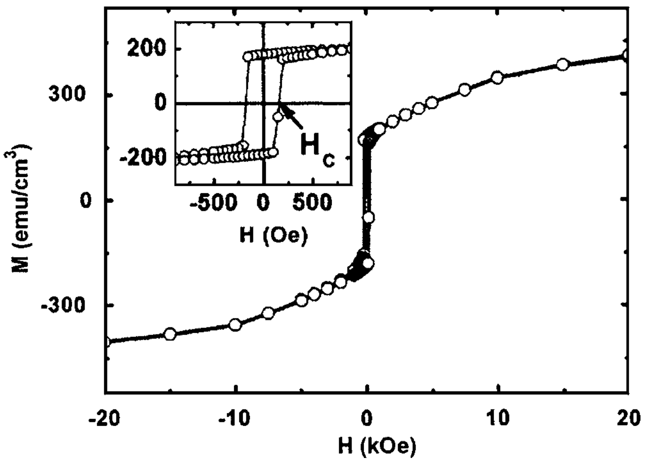

FIG. 2. Magnetic hysteresis loop of the Fe_3O_4 film after subtraction of the substrate diamagnetic contribution at RT. The magnetic field was applied in plane. The inset shows the low field zoom-in of the hysteresis curve. H_C denotes the coercive field.

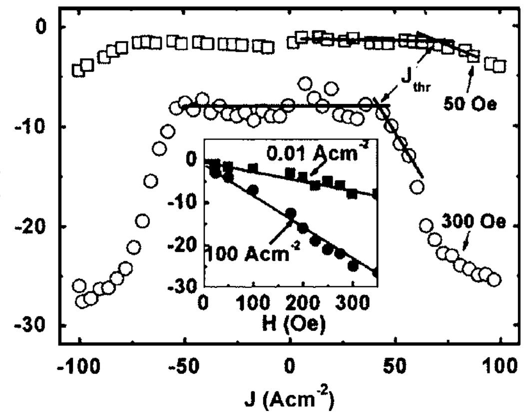

FIG. 4. MR vs bias current density under fields of 50 Oe (□) and 300 Oe (◯). The inset displays the MR as a function of applied magnetic field H for current densities of 0.01 and 100 A cm^{-2}.

Figure 2 displays the in-plane magnetic hysteresis loop measured at room temperature. The diamagnetic signal of the substrate has been already subtracted. The magnetization shows no saturation up to a magnetic field of 20 kOe, and the value of 410 emu/cm^3 at 20 kOe is less than that of 471 emu/cm^3 for the bulk Fe_3O_4.[9] As shown in the enlarged low-field hysteresis of the inset, the observed coercive field (H_C) is around 167 Oe, and the remanent magnetization reaches 50% of the magnetization at 20 kOe. The slow approach to saturation is consistent with that reported for single crystal and polycrystalline Fe_3O_4 films grown by different techniques,[5,6,10–12] suggesting the presence of antiferromagnetic exchange interactions across the atomically sharp antiphase grain boundaries (GBs) as discussed in Refs. 10 and 13.

The V-J characteristics in Fig. 3 are taken in current-in-plane geometry at RT with various magnetic fields applied parallel to the current as shown in the left inset. Every curve is highly reproducible and does not show any significant hysteretic behavior when the current is cycled. And hence, the whole curve is symmetric about the origin. When J attains a threshold value J_{thr} and corresponding voltage approaches V_{thr}, highly nonlinear conduction sets in and a low resistance state occurs above J_{thr} in the film. By fitting the non-Ohmic behaviors at $H=0$ to dynamic conductance $G=dJ/dV=G_0+kV^{\alpha}$,[14] where G_0 is the conductance in the zero-voltage limit, k is a constant, and the exponent α is a number characterizing the transport mechanism, we get α of about 1.75 at RT, indicating that the transport property of the Fe_3O_4 film is largely governed by the direct tunneling process through a potential energy barrier between the adjacent grains. According to the Simmons model for a square barrier,[15] one obtains a mean barrier height of 0.18 eV and a barrier thickness of 1.84 nm at zero field, which is close to the width ~2 nm of the GBs of Fe_3O_4 (100 nm) film, determined by Eerenstein *et al.* using the effective medium approximation.[16]

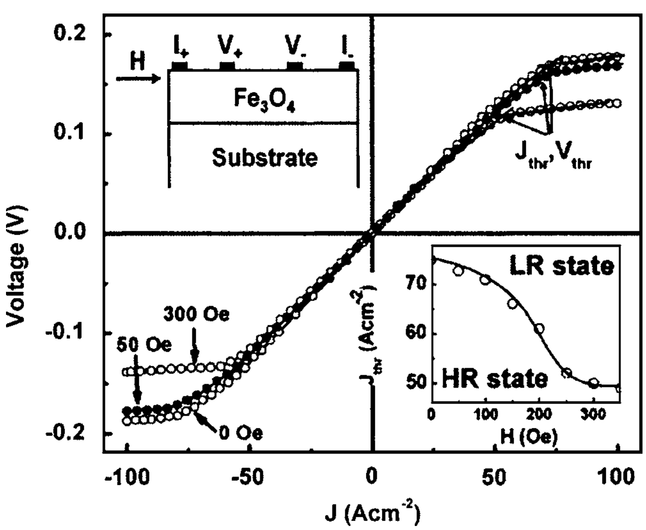

FIG. 3. V-J characteristics of the Fe_3O_4 thin film measured under different magnetic fields. The left inset is a schematic electrode setting for the measurement. J_{thr} as a function of applied magnetic field is shown in the right inset. The smooth solid lines indicate the turning points from HR to LR states.

The remarkable observation of the present work is the strong modifying effect of magnetic field (H) on the $V-J$ dependence. The application of H decreases the threshold value J_{thr}, suggesting that the ferromagnetic alignment of the grains in a magnetic field increases electron tunneling. As shown in the inset of Fig. 3, J_{thr} shows a slow drop with increasing H monotonically and exhibits a relatively fast change near the H_C followed by a saturation tendency down to 50 A cm^{-2}. It is clear that the switching from the high-resistivity (HR) to low-resistivity (LR) states takes place at smaller J_{thr} with increasing magnetic field.

It is ascertained that the influence of Joule heating is not important here. In our case, the resistivity of the Fe_3O_4 thin film is 5.6×10^{-3} Ω cm at 300 K and 0.01 A cm^{-2}, 5.0×10^{-3} Ω cm at 400 K and 0.01 A cm^{-2}, and 3.5×10^{-3} Ω cm at 300 K and 100 A cm^{-2}, respectively. One obtains the resistivity change of -2.0×10^{-5} Ω cm/A cm^{-2} and -0.6×10^{-5} Ω cm/K around RT. The former is much greater than the latter. Thus simple Joule heating cannot account for the observed effect.

Figure 4 shows the MR ratios as functions of current density. To be specific, for low J, the magnetite film shows small negative MR, −1.5% and −8% for $H=50$ and 300 Oe, while for the high J, a sharp increase in negative MR is evident at 73 and 48 A cm^{-2}, corresponding approximately to the J_{thr} for $H=50$ and 300 Oe. The ratios reach −5% and −27% for 50 and 300 Oe at 100 A cm^{-2}, respectively.

In magnetite films the presence of antiphase boundaries (APB) has been revealed as natural growth defects, and the magnetic coupling at the boundaries is antiferromagnetic (AF).[10,16] Due to the high degree of spin polarization in magnetite, the presence of these AF boundaries enhances the resistance of the films and GBs act as more highly resistive tunnel barriers. When a current is applied, the conduction electrons are spin polarized and are forced to move forward whether their spin is parallel to neighboring localized spin or not. Upon application of a magnetic field, the AF coupled spins will align themselves to some degree with the magnetic field, thus enhancing the transfer of spin-polarized electrons through APBs and causing a MR effect. The inset of Fig. 4 reviews the field dependence of MR for 0.01 and 100 A cm^{-2}. The linearity up to 350 Oe with increasing H is consistent with a hopping model discussed by Eerenstein *et al.*,[10] in which spin-polarized electrons traverse AF interfaces between the ferromagnetic grains.

In summary, we have observed the non-linear field-dependent $V-J$ behaviors in magnetite films. The switching from high-resistivity to low-resistivity states above a threshold current density is sensitive to the applied magnetic field and a large negative MR $\sim -27\%$ at 300 Oe was observed under a current density of 100 A cm^{-2} at room temperature. Such a phenomenon can be understood by considering a spin-polarized current flowing across GBs.

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