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A large low-field tunneling magnetoresistance of $CrO_2/(CrO_2/Cr_2O_3)$ powder compact with two coercivities

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Two channels of the conductance *G* exist in cold-pressed powder compacts of $CrO_2/(CrO_2/Cr_2O_3)$, where two types of granules have different coercivities. One of the channels is the spin-dependent intergranular tunneling conductance, and the other one is the spin-independent higher-order inelastic hopping conductance. The conductance is mainly related with the spin-dependent tunneling channel at low temperature. A large low field tunneling magnetoresistance (TMR) of 30.4% is achieved in 950 Oe field at 2 K. The reason is the relative orientation of the magnetization that tends to be antiparallel between the two coercivities. Field dependence of the tunneling magnetoresistance shows two better-separated peaks than that of CrO_2 powder compact. The results suggest that the introduction of CrO_2/Cr_2O_3 improves low field sensitivity, TMR, and switching characteristics. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1868080]

I. INTRODUCTION

The magnetotransport properties of chromium dioxide (CrO_2) have attracted much attention recently because it was predicted theoretically and practically to be a half-metallic ferromagnet.[1–4] It is suggested that half-metallic ferromagnets are ideal materials for the electrodes in spin-dependence tunneling devices. High values of tunneling magnetoresistance (TMR) have been reported in several experiments on CrO_2 polycrystalline films and powder compacts. Manoharan[5] and Coey[6] studied the cold-pressed powder samples and found that their TMR can reach as high as 30%–50% in magnetic fields from 1 to 5 T in 1998. The magnetic field is so strong that it severely limited their practical utilities. As proposed by Coey,[6] aligning the particles, which makes the magnetization of neighbors lay either parallel or antiparallel, might achieve improved magnetoresistance ratios. Dai and Tang[7] studied the field-aligned CrO_2 powder and found that TMR can reach as high as 41% in about 1000 Oe field at 5 K in 2001. However, in that experiment, the high resistance state is achieved only at the coercive field, which is too narrow a region for its application in switches and other devices. In addition, even at the coercive field, there are always those junctions where the two neighboring particles are aligned parallel to each other, thus reducing the TMR. Dai and Tang proposed that high TMR is possible if one can appropriately control and maximize the antiparallel alignment of the neighboring particles at the coercive field. Unfortunately, no experiments give evidence about it at low field up to now. Figure 1 (Ref. 8) shows magnetoresistance of $CoFe/Al_2O_3/Co$ junction plotted function of *H*. The two ferromagnetic layers with different coercive fields are generally used in order to realize experimentally both parallel and antiparallel magnetization alignment in a magnetic tunneling junction.[8,9]

In this work, based on the consideration of parallel and antiparallel magnetization alignment in a magnetic tunneling junction, $CrO_2/(CrO_2/Cr_2O_3)$ powder compacts were studied. CrO_2 powder with lower coercive field was introduced into the pure commercial CrO_2 powder for magnetic recording supplied by Dupont Corp. A large low-field TMR, enhanced field sensitivity and switching characteristics of $CrO_2/(CrO_2/Cr_2O_3)$ powder compacts are reported.

II. EXPERIMENT

CrO_2/Cr_2O_3 powder is CrO_2 particles enwrapped by Cr_2O_3. It was prepared by partially reducing the CrO_2 in air at 500 °C for 15 min. The ferromagnetic powder was characterized by powder x-ray diffraction (XRD) and transmis-

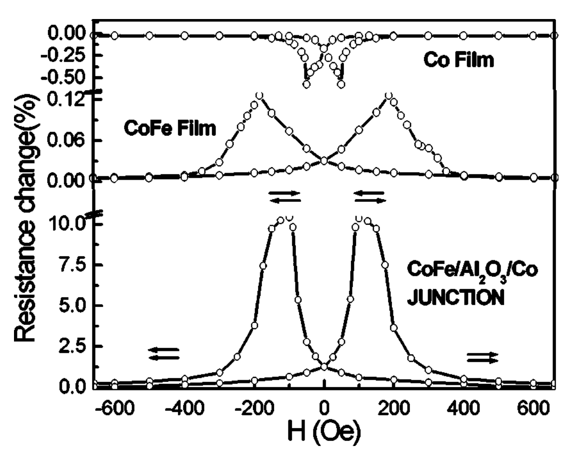

FIG. 1. Magnetoresistance of $CoFe/Al_2O_3/Co$ junction plotted function of H in the film plane, at 295 K. Also shown is the variation in CoFe and Co film magnetoresistance (see Ref. 8).

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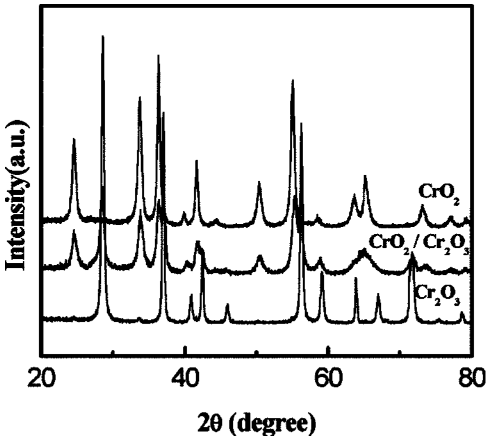

FIG. 2. X-ray diffraction patterns of CrO_2, CrO_2/Cr_2O_3, Cr_2O_3.

sion electron microscopy (TEM). The size and shape of CrO_2/Cr_2O_3 are the near same of that of CrO_2, with rod shape with an average length of 400 nm and a diameter of 50 nm. The magnetization curves were measured by a vibrating sample magnetometer 7300 (VSM). Different coercivities were found for CrO_2 and CrO_2/Cr_2O_3 at 1000 Oe for the former and at 600 Oe for the latter at 2 K. Two samples were prepared for the measurement. Sample 1 was the original commercial CrO_2 powder for magnetic recording supplied by DuPont Corp. Sample 2 was a composite where a weight fraction of 50% of the CrO_2 powder was mixed with CrO_2/Cr_2O_3 powder. The cold-pressed compacts were made under a pressure of 1.5×10^8 N/m^2. Transport measurements were performed using a Physical Properties Measurement System (Quantum Design Co. Ltd.).

III. RESULTS AND DISCUSSION

Figure 2 shows the XRD patterns of CrO_2, CrO_2/Cr_2O_3, and Cr_2O_3. As seen from Fig. 2, more Cr_2O_3 appears after reducing for 15 min. Field dependence of the magnetization is given in Fig. 3. In Fig. 3, it is obvious that the sample of CrO_2/Cr_2O_3 shows nearly saturated magnetization at 4000 Oe. The phase percentage can be determined from the saturated magnetization of samples and they are listed in Table I.

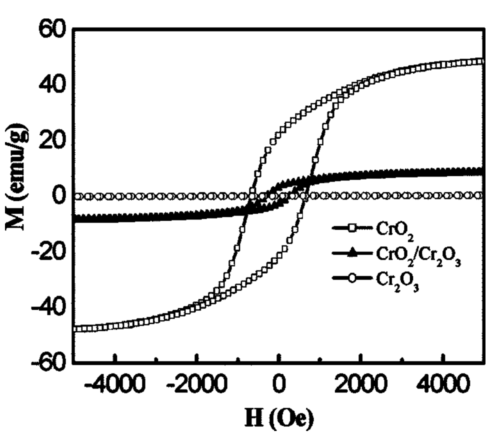

FIG. 3. Magnetization curves of CrO_2, CrO_2/Cr_2O_3, Cr_2O_3 at room temperature.

TABLE I. Annealing effect of CrO_2 powder compacts.

Powder compact	Annealing temperature (°C)	Annealing Time (min)	Content of Cr_2O_3 (wt %)
1	No annealing	0	<5
2	500	15	≈ 70
3	550	120	>98

The conduction mechanism of the CrO_2 powder compact has been analyzed by Coey *et al.*[6] and it is associated with the spin-dependent intergranular tunneling across the grain boundaries. The relationship between tunneling conductance and temperature is given as follows:[10–12]

$$G_{SDT} = G_0(1 + P^2m^2)\exp[-(\Delta/T)^{1/2}], \quad (1)$$

$$\mathrm{LnG} \propto T^{-1/2}, \quad (2)$$

where P is the spin polarization, $m=M/M_S$ is the relative magnetization, G_0 is a constant and Δ is proportional to the Coulomb charging energy and barrier thickness. Figure 4 shows the logarithmic conduction *LnG* as a function of $T^{-1/2}$ for sample 1 and sample 2. It is observable from Fig. 4 that the relationship between *LnG* and $T^{-1/2}$ is linear below 47 K, which is typical of spin-dependent intergranular tunneling. Δ can be determined from the slope of the linear part of *LnG vs* $T^{-1/2}$. It is $\Delta \approx 4.4$ K, and 11.1 K for sample 1 and 2, respectively. The resistivity of sample 2 increases significantly due to the introduction of Cr_2O_3, a good insulator, which causes the dilution effect and the increase in barrier thickness.

It is also found from Fig. 4 that *LnG* starts to deviate from the linear when $T>47$ K. This phenomenon suggests G_{SDT} is not the major conduction mechanism and other mechanism may become dominant at high temperature. As proposed by Glazman[13] and confirmed by Xu,[14] with increasing temperature, an inelastic hopping conductance with high-order spin-independence (G_{SI}) becomes dominant. This hopping channel follows a power law. So the total conductance is given as follows:

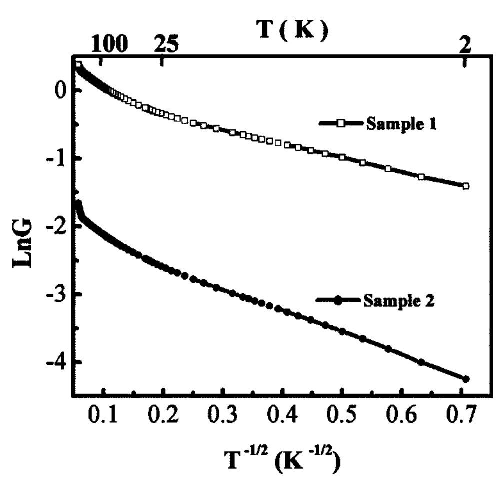

FIG. 4. LnG as a function of $T^{-1/2}$ for samples 1 and 2.

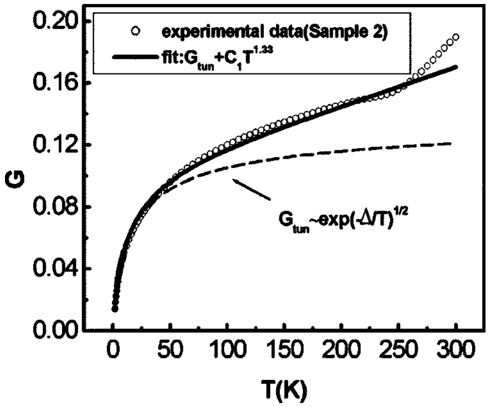

FIG. 5. Illustrations of the theoretical fitting of G as a function of T for sample 2.

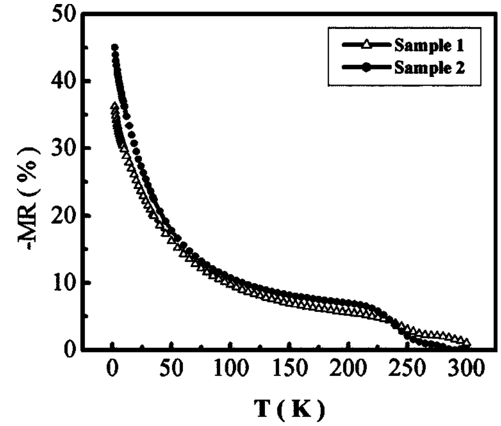

FIG. 6. Temperature dependence of TMR for samples 1 and 2.

$$G = G_{SDT} + G_{SI} = C_1 \exp[-(\Delta/T)^{1/2}] + C_2 T^{\gamma}, \quad (3)$$

where C_1 and C_2 are constants, $\gamma = N - [2/(N+1)]$. N is the number of steps in a multistep tunneling process. Equation (3) fits the experimental data surprisingly well when temperature is below 250 K. Therefore, the conductance of sample 2 can be fitted generally by

$$G = G_{SDT} + G_{SI} = C_1 \exp[-(\Delta/T)^{1/2}] + C_2 T^{1.33}. \quad (4)$$

The first and the second terms represent the spin-dependent channel and the spin-independent channel, respectively. The third-order ($\gamma=2.5$) hopping becomes nonnegligible when temperature is above 250 K. In other words, the conductance is determined by the spin-dependent tunneling and the second-order ($N=2$) hopping when $T < 250$ K. Figure 5 shows the theoretical fits for the temperature dependence of the conductance in details. Increasing temperature favors the higher-order hopping in agreement with the theory as reported by Glazman and Matveev.

In addition to the spin-dependent direct tunneling, the spin-independent high order inelastic hopping is also proposed to account for the TMR-T dependence.[15] In fact, TMR is given by:

$$\mathrm{TMR} = (R_H - R_0)/R_0, \quad (5)$$

where $R_H = R_{SDT} + R_{SI}$, $R_0 = R_{SDT,0} + R_{SI}$, therefore, TMR can be expressed as

$$\mathrm{TMR} = (R_{SDT,H} - R_{SDT,0})/(R_{SDT,0} + R_{SI}) \quad (6)$$

with increasing temperature, R_{SI} rapidly increases and result to decrease rapidly in TMR (Fig. 5). In addition, the decrease of TMR is associated with the spin-flip effect at high temperature and suppressed spin polarization of CrO_2.[16,17]

Figure 6 shows the temperature dependence of the TMR for sample 1 and sample 2. It is also evident that TMR of sample 2 is larger than that of sample 1. The higher TMR of sample 2 is associated with the improved barrier layer due to the precipitation of Cr_2O_3 at the grain boundaries. As proposed by Stauffer,[18] the dilute composites show much greater resistivity and magnetoresistance ratios. Several groups have studied CrO_2/Cr_2O_3 powder compacts, and obtained greater resistivity and magnetoresistance than CrO_2. Those results are listed in Table II. The TMR as a function of applied field for sample 2 at 2 K is shown in Fig. 7. The TMR is defined as $(R-R_H)/R_H$, where R_H is the resistance at applied field. The TMR of sample 2 is 30.4% at $T=2$ K and shows two better-separated peaks near $H \approx \pm 950$ Oe than CrO_2 [Fig. 7(a)]. The origin of the higher low-field magnetoresistance and well-separated peaks for sample 2 is discussed below.

A tunneling path is open when the relative magnetizations of the two neighboring particles are parallel at high field, and a tunneling path is closed when the relative magnetizations of the two neighboring particles are antiparallel, usually at coercive field. Extremely high TMR is possible if one can appropriately control the magnetization alignment of the neighboring particles. Dai and Tang studied the field-aligned CrO_2 powder compact, where magnetization of neighboring CrO_2 lie ideally parallel at high field, and found that TMR can reach as high as 41% at about 1000 Oe field at 5 K.[7] However, in their experiment, even at the coercive field, there always are those junctions where the two neighboring particles are open. So they proposed that high TMR is also possible if one can appropriately control and maximize the antiparallel orientation of the neighboring particles at the coercive field. In our work, sample 2 was prepared by the similar ways realizing magnetization alignment in magnetic tunneling junction and can be approximately described as a network of many magnetic tunneling junctions, which

TABLE II. Magnetoresistance of CrO_2 powder samples.

Composition	Preparation	MR	Field (Oe)	Source (Ref.)
CrO_2	Cold pressed	42%(∼30%)	50 000(10000)	2
CrO_2	Cold pressed	30%(∼23%)	50 000(5000)	3
25%CrO_2, 75% Cr_2O_3	Cold pressed	∼50%	50 000	3
$CrO_2/(CrO_2/Cr_2O_3)$	Cold pressed	∼30.4%	950	This work

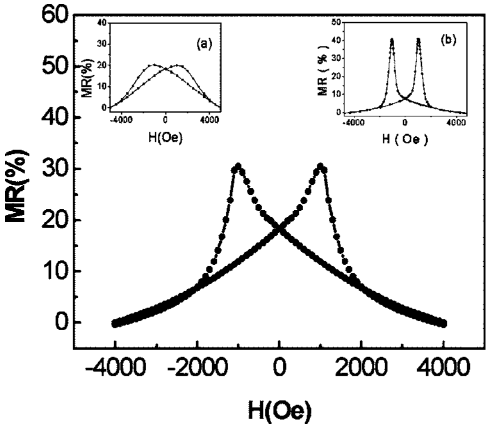

FIG. 7. TMR of sample 2 at 2 K. Inset (a) and inset (b) show the TMR of the cold-pressed CrO_2 powder and field-aligned CrO_2 powder at 5 K, respectively.

formed by CrO_2, (CrO_2/Cr_2O_3) particles and the boundary between them. Between the two coercivities the magnetizations of (CrO_2/Cr_2O_3) particles with smaller H_c have already aligned into the field direction but the magnetizations of CrO_2 particles with larger H_c are still pointing in the opposite direction. Therefore, one gets a region where the resistance is high between the two coercivities. This can be seen in Figure 7. A sudden slope change near 600 Oe indicates the realignment of the CrO_2/Cr_2O_3 particles and the peak at ~1000 Oe indicates the realignment of CrO_2. More junctions are in the open state between the two coercivities in sample 2 than sample 1 at coercivity. Thus sample 2 has a larger low field magnetoresistance and two well-separated broad peaks. However, the TMR of the field-aligned CrO_2 powder compact is still higher than that of sample 2 [Figure 7(b)], which consists of random powder. For the field-aligned samples, as the field decreases to zero from the saturation, nearly all moments remain in the original direction, which results in the low resistance state and the higher TMR. This circumstance is different from that of the random powder where a large part of the moments is already misaligned as soon as the field reduced to near zero, which leads to a large increase in resistance and a decrease in TMR. In fact, the antiparallel alignment of the neighboring particles could not be maximized but it can be improved at the coercive field in this work. It is the reason that clearly broad peaks of TMR could not be seen in Fig. 7. It is also the cause that two coercivities could not be observed on magnetization loop of sample 2.

In order to achieve a larger TMR, higher field sensitivity and a broader region with high TMR, maximization the antiparallel alignment of the neighboring particles at the coercive field is a promising approach. Aligning of half-metallic particle with a different coercive field is in progress.

IV. CONCLUSIONS

The magnetic and transport properties of $CrO_2/(CrO_2/Cr_2O_3)$ powder compact have been studied. Better control of the magnetization alignment of the neighboring particles is realized by using CrO_2 particles with different coercive field. Between the two coercivitys, one can get a region where the resistance is large. Sample 2 shows a larger low-field magnetoresistance and two better-separated broad peaks than sample 1. A negative TMR of 30.4% has been obtained at 2 K in $H=950$ Oe. The spin-dependent intergranular tunneling and inelastic hopping are the major conduction mechanisms of the $CrO_2/(CrO_2/Cr_2O_3)$ powder compact. It suggests that the improved switching characteristics of $CrO_2/(CrO_2/Cr_2O_3)$, as well as the broader field region of high resistance state, may have useful applications.

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