Film heterostructure with soft ferromagnetics to enhance low-field magnetoresistance

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A heterostructure composed of two NiFe films and a perovskite $La_{0.7}Ca_{0.3}MnO_3$ film is prepared using pulsed-laser deposition and magnetron sputtering. An enhanced effective low-field magnetoresistance (LFMR) is observed in this heterostructure because the NiFe thin films concentrate magnetic induction lines onto the small-area $La_{0.7}Ca_{0.3}MnO_3$ film. The dimension of this heterostructure can be reduced to several micrometers by lithography technique, and a great potential for enhancing the effective LFMR for $La_{0.7}Ca_{0.3}MnO_3$ at room temperature is predicted. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1522130]

Colossal magnetoresistance (CMR), which refers to a huge decrease in resistance in response to a magnetic field **H**, was observed in manganese oxides with perovskite structure.[1] Although the CMR effect is attractive, the materials must be submitted to a large field at low temperature in order to gain the CMR. Currently, there are at least three challenges before CMR materials can be used for industrial applications. First, they must be used at room temperature and up to 100 K above room temperature. Second, a value of MR ($=\Delta\rho/\rho(0)$, where ρ is the resistivity) as high as 20% is required under $\mathbf{H}=100$ Oe. Finally, the magnetotransport properties must remain roughly unchanged in patterned films of a dimension as small as ~ 100 nm.[2] Although the two-phase perovskite oxides and bilayered manganite $La_{2-2x}Sr_{1-2x}Mn_2O_7$ also show high low-field magnetoresistance (LFMR) in the low temperature range, the effect shows a rapid decay with increasing temperature T, unfavorable for device applications.[3,4]

By utilizing the high magnetic susceptibility of soft ferromagnetics to concentrate magnetic induction lines onto a small area where a magnetoresistive material is developed, a macroscale structure was fabricated to enhance the LFMR and TMR response.[5,6] In this structure, a thin, flat single crystal of $La_{2/3}Ca_{1/3}MnO_3$ (0.1 mm thick) was coordinated in the gap of two long columns of polycrystalline $(Mn,Zn)Fe_2O_4$ (each piece $1.473\times1.473\times24.2$ mm^3). The MR of the $La_{2/3}Ca_{1/3}MnO_3$ crystal by itself is 0.85% under $\mathbf{H}=400$ Oe at room temperature, while that of the same sample in this structure is 20%. However, such a structure is too bulky (typically 5 mm) for integrated applications. In this letter, we fabricate a heterostructure entirely made up of films and composed of soft ferromagnetic materials, and show some of its advantages.

As shown in Fig. 1(a), the heterostructure we propose is made up of two trapeziform soft magnetic films and one foursquare perovskite manganite film, in order to concentrate the magnetic induction lines onto the centered small area. The lengths of the long and short sides, and the height and the thickness of the trapeziform film are denoted by w, g, $(L-g)/2$, and t, respectively. As shown in Fig. 1(b), there is a gap between the two soft magnetic films (GTSM), whose side length and thickness are g and t, respectively.

In the heterostructure proposed here, the metallic NiFe alloy (Ni=80%, Fe=20%) is used to replace $(Mn,Zn)Fe_2O_4$, because NiFe is an excellent soft magnet (relative magnetic susceptibility $\mu_r\sim10^4$) and conductor (conductivity $\sigma\sim10^3\ \Omega^{-1}\,\mathrm{m}^{-1}$, resistivity $\sim10\ \mu\Omega$ cm) over $70\sim350$ K. Therefore, the NiFe in thin films can be used as electrodes while they concentrate magnetic induction lines, and this function is very important for reducing heterostructure to several micrometers. In fact, the thin film NiFe retains the high susceptibility even though the grain size is down to submicrometer. However, $(Mn,Zn)Fe_2O_4$ in thin film form shows quite low μ_r since the grain size is small. In addition, the time for domain relaxation in the NiFe is about 10^{-9} s, also favorable for high-frequency applications.[7,8] Because perovskite manganites show much higher resistivity than NiFe alloy and the area of GTSM is small, the resistivity of the heterostructure, as shown in Fig. 1, is ascribed to the perovskite manganite, which is polycrystalline $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) thin film confined in the GTSM area.[9–11]

If we first deposit two trapeziform NiFe films on the substrate and then deposit the LSMO film into area GTSM, as shown in Fig. 1(b), the two layers (LSMO/NiFe) keep good contact. However, the LSMO thin film must be deposited in the oxygen ambient at high temperature (600–800 °C), the pre-deposited NiFe layer will be oxidized. In our experiment, we shall first deposit LSMO film and pattern it so that only the GTSM area is left. Subsequently, two trapeziform NiFe films are deposited and a small LSMO/NiFe overlap is permitted.

The LSMO thin film (about 300 nm thick) was deposited on Si substrate using pulsed-laser deposition (PLD).[12] De-

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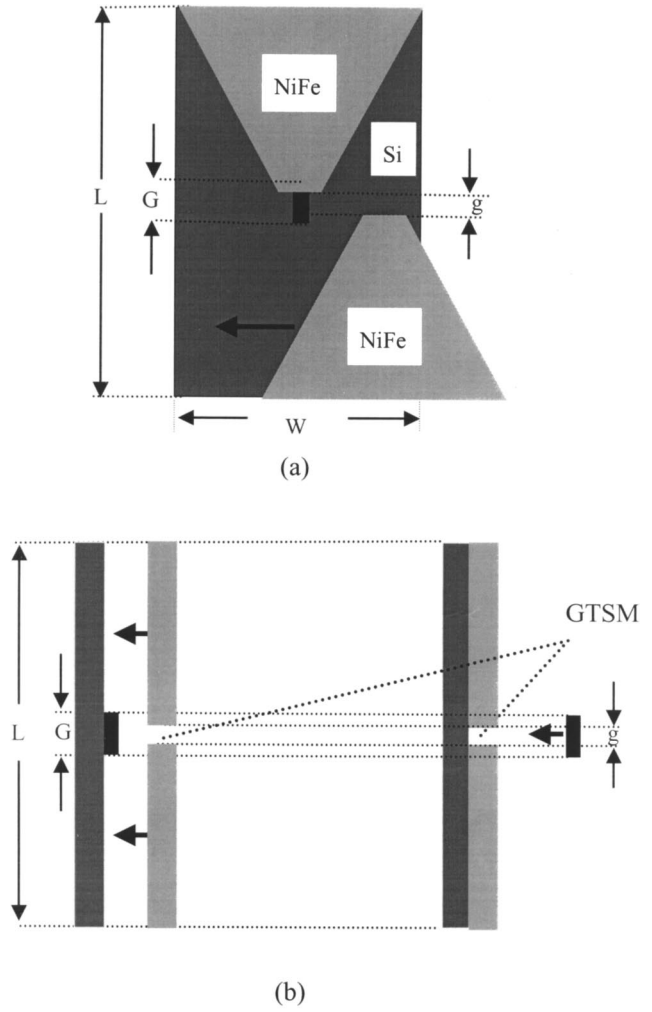

FIG. 1. (a) Surface schematic drawing of the heterostructure. (b) Left: Transverse schematic drawing of the heterostructure in order to prevent oxidation of the NiFe film. Right: Transverse schematic drawing of the heterostructure in order to deposit LSMO completely into the GTSM.

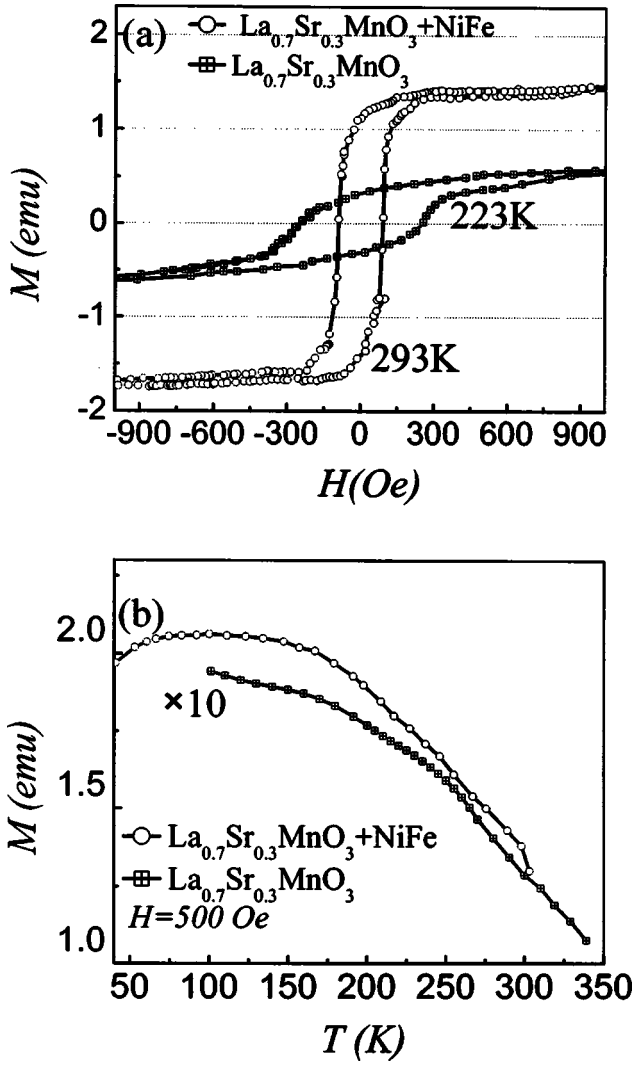

FIG. 2. (a) **M**–**H** hysteresis loops of NiFe at 293 K and LSMO film at 223 K. (b) Magnetization **M** of NiFe and LSMO films as a function of temperature T under 500 Oe.

tails of the PLD procedure were reported earlier.[13] Permalloy (Ni=80% and Fe=20%) film (about 150 nm thick) were deposited on Si substrate, using single-target rf magnetron sputtering under a base pressure of 2×10^{-5} Pa at room temperature. The length of the rectangle for total LSMO film (G) is 2 mm, and the lengths of the rectangles for one-layer LSMO film (g) and two-layer NiFe/LSMO film $[(G-g)/2]$ are 1 mm and 0.5 mm, respectively. The applied electric current flows through the NiFe films and LSMO film. The triangle-wave ac magnetic field, 3 kOe in magnitude and 0.01 Hz in frequency, was applied to the heterostructure. The resistivity as a function of temperature over 77~300 K was measured. The Lakesshore vibrating sampling magnetometer (VSM) was utilized to probe the magnetic property. Magnetization (**M**) of the sample was measured under a magnetic field of 500 Oe from 80 K to 320 K.

The measured **M**–**H** hysteresis loops of NiFe and LSMO films are shown in Fig. 2(a). A coercive magnetic field $\mathbf{H}_c\sim80$ Oe for NiFe and $\mathbf{H}_c\sim240$ Oe for LSMO can be determined. The magnetization **M** of NiFe is much larger than that of LSMO, especially at low **H**. However, the magnetization of NiFe saturates at a much lower field ($\sim\mathbf{H}_c$), while the saturated magnetic field $\mathbf{H}_s$ for LSMO is much higher than its own $\mathbf{H}_c$. Therefore, in the fabricated heterostructure, the magnetic fluxes can be effectively concentrated onto the sandwiched area GTSM by the neighboring NiFe thin films. As shown in Fig. 2(b), **M** of the NiFe film is ten times larger than that of the LSMO film under 500 Oe over 70~300 K.

Figure 3(a) shows a schematic drawing of the magnetic induction lines in the heterostructure, which is parallel or perpendicular to the electric current I ($\mathbf{H}\|I$, $\mathbf{H}\perp I$). For $\mathbf{H}\|I$, the measured LFMR is enhanced due to the concentrated magnetic fluxes within the area GTSM. This opposite effect is observed in the case of $\mathbf{H}\perp I$, since the NiFe film partitions the magnetic induction lines across the GTSM. On the basis of the spin-polarizd transport across grain boundaries, the maximal LFMR under a saturated field takes the form: $\mathbf{M}R\propto\mathbf{M}_0^2$, where $\mathbf{M}_0$ is the normalized magnetization.[14] The coercivity $\mathbf{H}_c$ is equal to $\mathbf{H}_{\text{peak}}$, corresponding to MR=0 for normal CMR materials. As shown in Fig. 2(a), $\mathbf{H}_c\sim90$ Oe for NiFe and $\mathbf{H}_c\sim250$ Oe for LSMO. However, $\mathbf{H}_{\text{peak}}\sim85$ Oe is observed for LSMO in our heterostructure, which is consistent with $\mathbf{H}_c$ of the NiFe film. That means that the real magnetic field in the area GTSM is about ~250 Oe, although the applied field is only 85 Oe, demonstrating that

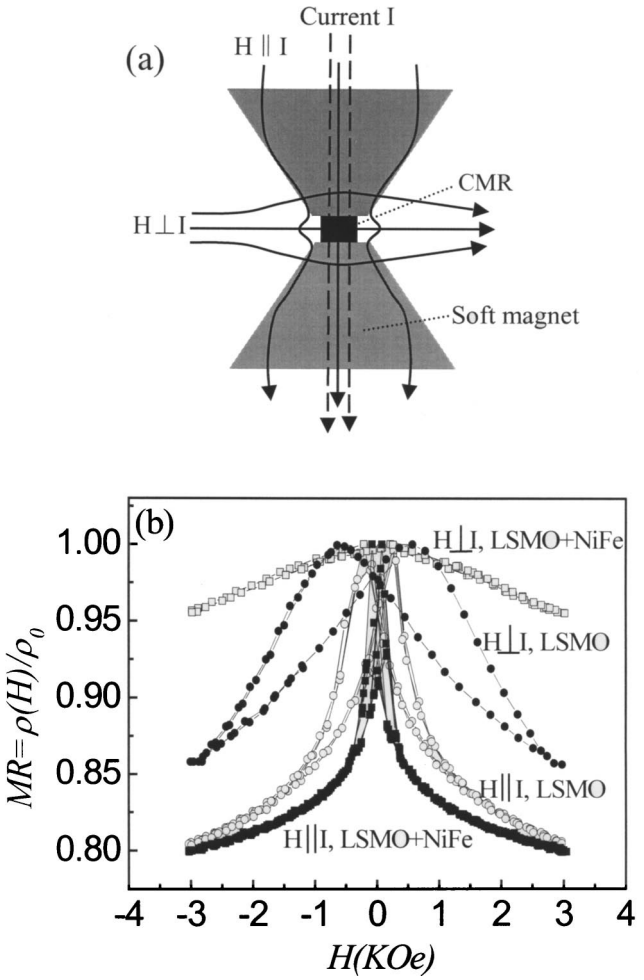

FIG. 3. (a) A schematic drawing of the magnetic induction lines in the heterostructure, which is parallel or perpendicular to the electric current I ($\mathbf{H}\|I$, $\mathbf{H}\perp I$). (b) Normalized resistivity of LSMO with or without heterostructures (LSMO+NiFe or LSMO) as a function of applied magnetic field ($\mathbf{H}\|I$, $\mathbf{H}\perp I$) at 77 K.

the present structure works well and efficiently. The consequent feature is the rapid decreasing of the sample resistivity under a low magnetic field, which benefits especially to the enhancement of LFMR. For the LSMO thin film sample, the LFMR in mode $\mathbf{H}\perp I$ is about 70% of that in model $\mathbf{H}\|I$ under the same field. However, in our heterostructure, the LFMR of LSMO thin film in mode $\mathbf{H}\perp I$ is only 0.5% under 300 Oe but reaches 13.5% in model $\mathbf{H}\|I$ under the same field.

Although the enhancement of the LFMR in our heterostructured sample is significant, it is still far lower than the theoretically predicted value. A qualitative analysis allows us to argue that the LFMR in our heterostructure depends on four factors: width of the GTSM (g), thickness of the soft magnetic films (t), quality of the deposited layers and the geometric pattern. If g decreases with other conditions being the same, the magnetic resistance [$\propto (g/\mu_r\mu_0)$, where μ_r is relative magnetic susceptibility of LSMO] in the GTSM will decrease too, which will correspondingly enhance the capability to fortify the magnetic field in the GTSM. The thicker the NiFe film, the stronger its capability to fortify magnetic fields in the GTSM. In the case of $g \sim t$, the LFMR enhancement reaches the maximal. Therefore, the perovskite film should be completely deposited into the GTSM, where the strongest magnetic field is available. If a soft magnet with a very low electrical resistivity but high resistance against oxidation at elevated temperature is available, we can deposit the soft magnetic film first, followed by the deposition of the perovskite manganite, as shown in Fig. 1(b). Some of nanocrystalline alloy, such as $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$, can keep good soft magnetism and conductance even after air annealing. Thus, they can also be used in the heterostructure. Of course, the coercivity $\mathbf{H}_c$ of NiFe films may be far smaller than 85 Oe. For example, the measured coercivity $\mathbf{H}_c$ along the easy axis of NiFe film is as small as 16.3 Oe if an in-plane magnetic field is applied to induce a uniaxial anisotropy.[8] When the size of the GTSM is as small as 1.0 μm, for example, using photolithography, and the thickness of the NiFe films is as thick as 1.0 μm, the LFMR under a field of 300 Oe can be as high as 30% at room temperature.

In conclusion, we have fabricated a heterostructure composed of soft magnet NiFe thin films and perovskite manganite LSMO layer, and have predicted its application potentials as magnetoresistance devices. Our experiment has demonstrated that the magnetic field $\mathbf{H}$ in the GTSM is significantly enhanced under an applied field $\mathbf{H}$ ($\|I$) and suppressed under $\mathbf{H}$ ($\perp I$). The LFMR will increase remarkably if the heterostructure can be fabricated as small as possible.

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