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We achieved the electrical control of a magnetic state by using an external magnetic field via electric field-induced resistance (EIR) change phenomena. We prepared a La$_{0.7}$Sr$_{0.3}$MnO$_3$ (LSMO) film which showed EIR changes. Before the EIR treatment, the sample barely showed negative magnetoresistance (MR). After the EIR treatment, the value of negative MR became six times larger than that before the EIR treatment. The magnetization curves also changed from paramagnetism to superparamagnetism. These behaviors revealed that the EIR treatment changed not only the resistance state but also the magnetic state in LMSO film. © 2013 AIP Publishing LLC
[http://dx.doi.org/10.1063/1.4802483]

Electrically controlled magnetism has long been a topic of interest in electronics. Several types of electrically controlled magnetism have been investigated, such as multiferroics,[1,2] tunnel magnetoresistance,[3] and current-driven magnetic domain wall motion.[4] Herein, we propose another type of electrically controlled magnetism. We focused on perovskite oxide material because it has many attractive features such as a half-metal band structure, colossal magnetoresistance, and Mott insulators.[5,6] In 2008, we reported that (Pr,Ca)MnO$_3$ (PCMO), which is one of the perovskite oxides, showed electric field-induced resistance (EIR) changes and negative differential resistance (NDR) behaviors.[7] We proposed that an oxidation/reduction process between an electrode and a base material could play an important part in the EIR phenomenon. However, the magnetism of perovskite oxides is explained by the double exchange interaction via spin-polarized conduction electrons. In this mechanism, spin-polarized conduction electrons are itinerant between two manganese via the oxygen.

Here, we propose a way to electrically control magnetism. If we can change a certain area of the oxidization state—which is excess/deficient oxygen—using the EIR phenomenon, the magnetism or magnetic order must be changed. In the present study, we prepared a thin La$_{0.7}$Sr$_{0.3}$MnO$_3$ (LSMO) film because with such a film the EIR effect works more effectively. We investigated the EIR properties of samples, and we also measured the magnetoresistance (MR) and magnetic properties of the sample before and after the EIR treatment.

A stoichiometric La$_{0.7}$Sr$_{0.3}$MnO$_3$ (LSMO) target was prepared by a solid-state reaction. The ratios of La and Sr used were 0.7 and 0.3, respectively. An Nb-doped SrTiO$_3$ substrate was used as a substrate and bottom electrode. The base pressure before introducing the sputtering gas was 3.0×10^{-4} Pa, and the gas pressure during deposition was 8.0×10^{-1} Pa. The gas used was a mixture of Ar and O$_2$, and the ratios were 9:1. The growth temperature was also fixed at $400\,^{\circ}$C. The thicknesses of the LSMO films were 45 nm. Square-shaped Pt with a side length of 300 μm was used as a top gate electrode and a source-drain electrode. The thickness of the electrode was

50 nm. We performed the structural analysis of the films using an X-ray diffractometer (XRD; Rigaku Rad-B, Rigaku, Tokyo) and Cu Kα radiation. All diffraction angles were precisely corrected using Au polycrystal patterns. We also checked an epitaxy using Reflection High Energy Electron Diffraction (RHEED; VACUUM PRODUCTS ARH-100). The electric properties of the thin films were measured at room temperature. The current-voltage (I-V) property was measured using a picoammeter (model 6487, Keithley, Cleveland, OH) and measured using a coaxial prober system (model K-157MP, Kyowariken, Tokyo). A sweep voltage of 50 mV/s was used for the measurement. The magnetic properties of the sample were measured using a superconducting quantum interface device (SQUID, Quantum Design Japan, MPMS-5, Tokyo). The surface morphology was measured using an atomic force microscope (AFM, TOYO Corporation, nano-R, Tokyo).

Figure 1 shows the XRD and RHEED patterns of the LSMO sample, an AFM image of the sample, and the surface profile indicated by an arrow in the AFM image. The XRD pattern revealed that the film has an n00 diffraction peak from LSMO. RHEED patterns revealed that the sample is epitaxially grown on the substrate. These patterns showed relatively spotty patterns. It might be due to the low crystallinity of the sample.[8] The AFM image and the surface profile also revealed that the sample surfaces are smooth. The root-mean square roughness of the sample is approx. 1 nm, which was smooth enough to uniformly apply the voltage on the metal electrode.

Figure 2(a) shows the initial I-V curve of the sample. The I-V curve showed both NDR behavior and EIR behaviors. From our previous work, we revealed that this initial hysteresis was induced by migrating oxygen barely oxidizing the top electrode. The current maximum indicates the saturation of oxygen migration caused by an electric field, and the corresponding voltage indicates the ease of oxygen migration. Figure 2(b) shows the resistance change repeatability of the sample. The pulse voltages of 10 V, 1 ms were used for resistance switching. It is clear that the resistance was changed by the application of each electric field at least eight times.
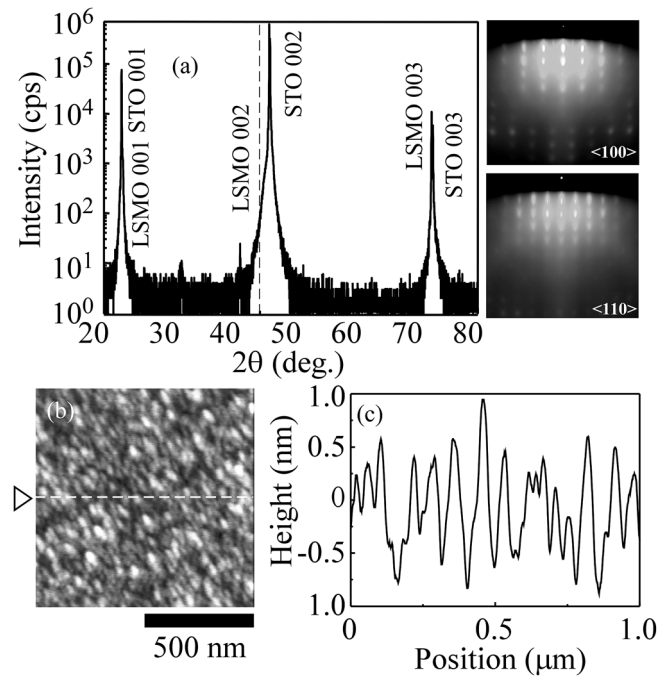
FIG. 1. (a) The XRD pattern of the LMSO sample, (b)an AFM image, and (c) the surface profile (indicated by the arrow in the AFM image).

Since we were able to prepare the sample with EIR properties, we investigated magnetic property changes before and after switching treatment. Figure 3(a) is a schematic illustration of the sample for MR measurement. First, we measured the MR between the electrodes of (1) and (3),
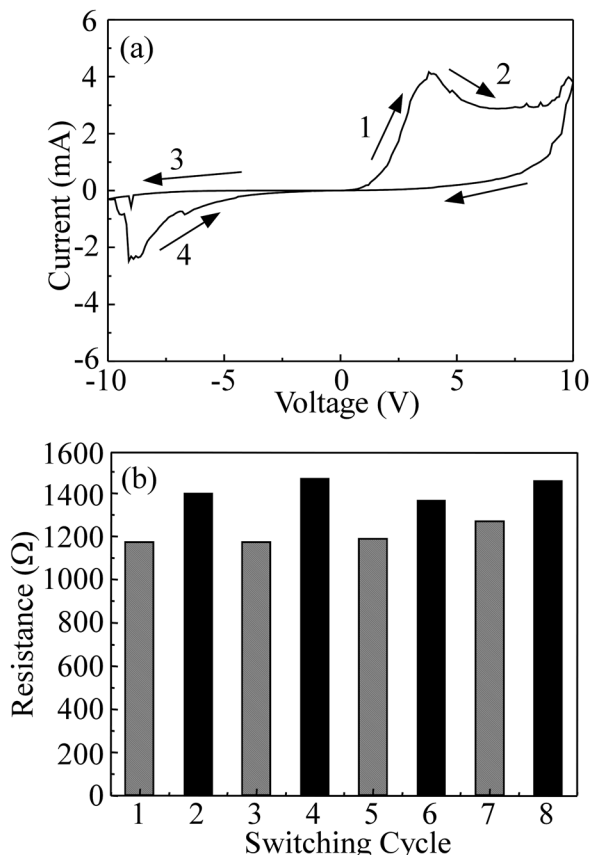

FIG. 2. Initial I-V curves of the LMSO sample (a). Arrows 1 and 2 indicate NDR behavior, and 3 and 4 indicate EIR behavior. The resistance change repeatability of the sample (b). The switching pulse voltage was 10 V and 1 ms.
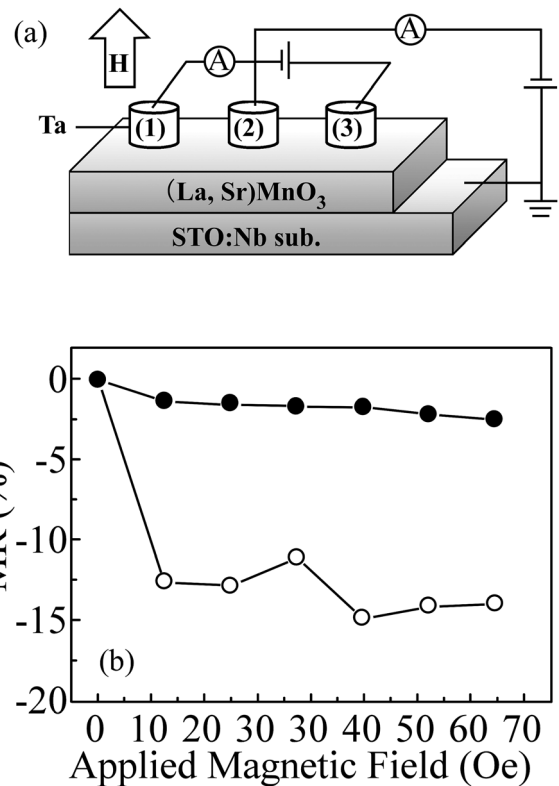

FIG. 3. (a) A schematic illustration of the sample for MR measurement. The ⟨electrode (2)⟩ − ⟨Nb-doped $SrTiO_3$ substrate⟩ was used for writing a resistance state. The ⟨electrode (1)⟩ − ⟨electrode(3)⟩ was used for MR measurement. The magnetic field was applied in the direction perpendicular to the current direction (⟨electrode (1)⟩ − ⟨electrode(3)⟩). (b) The MR before (closed circles)/after (open circles) EIR treatment.

which is indicated by a black line in Figure 3(a). Then we measured the EIR properties of electrode (2) (indicated by a gray line in Fig. 3(a)). Next, we measured the MR between electrodes (1) and (3) again. The results are shown in Figure 3(b). Both processes showed negative MR within the measurement magnetic field. After the EIR treatment, the MR value of the sample became six times larger than that of the initial sample. This result indicates that the sample after the EIR treatment had a positive magnetic order, and each magnetic moment became strongly coupled.

To investigate the magnetic ordering state of the sample, we measured the magnetization curve and temperature dependence of the magnetization of the sample before and after the EIR treatment. Figure 4(a) is a schematic illustration of the sample for magnetization measurement. We prepared a sample with 36 electrodes (Fig. 4(a)). First, we measured the magnetization curve and temperature dependence of the magnetization of the sample. We then wrote an initial memory state into each electrode by sweeping voltages (Fig. 2(a)) and then a certain memory state at the voltage of 4 V. The magnetic properties were measured in the same manner. Figure 4(b) shows the results of magnetization curves before and after the EIR treatment.

The magnetization curves of each measurement were positive, indicating that this sample has a positive magnetic order which is paramagnetism or ferromagnetism. In the case of the sample before EIR treatment, paramagnetism was observed. In contrast, the sample after EIR treatment showed high magnetic susceptibility at a low magnetic field, and it became small above 0.1 T. Although the sample showed a
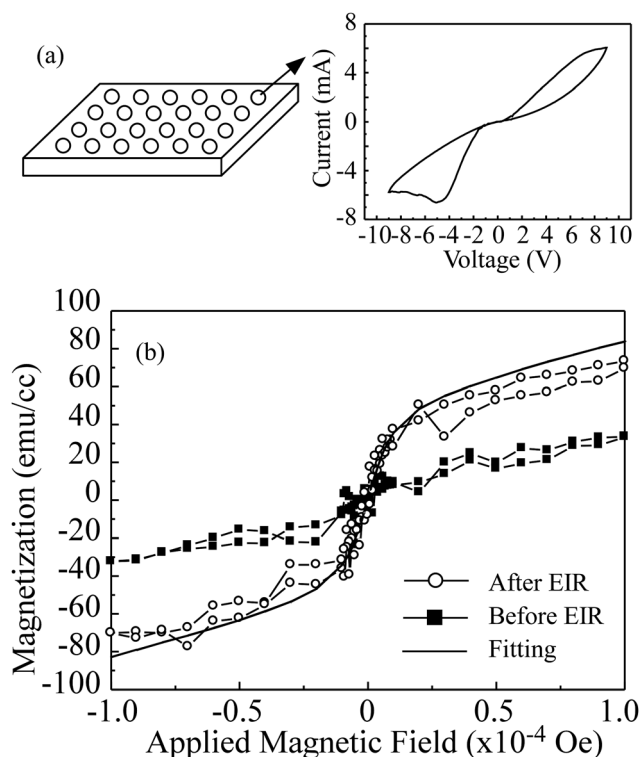
FIG. 4. A schematic illustration and an example of EIR behavior of the sample used for magnetization measurement (a). The sample had 36 electrodes. Each electrode shows EIR behavior. (b) Magnetization curves of the sample before/after EIR treatment. The solid line in the curves after EIR treatment is a fitting line of the Langevin Function.

typical ferromagnetic magnetization curve, it did not have any hysteresis window. This might be due to the too-small ferromagnetic regions, i.e., superparamagnetism.[9] To estimate the small ferromagnetic region, we fitted the magnetization curve using the Langevin Function, which is defined by the following equation:

$$L(x) = \coth(x) - 1/x, \quad x = g\mu_B JH/K_B T,$$

where g is the g-factor, μ_B is the Bohr magneton, $J \rightarrow$ infinity, K_B is the Boltzmann constant, T is the temperature, and H represents the applied magnetic field. The magnetic moment against an applied magnetic field can be calculated by the equation

$$M = Ng\mu_B L(x),$$

where N is the number of atoms per unit volume. The result of fitting is indicated by the solid line in Figure 4(b). According to the fitting result, we calculated that the area of the ferromagnetic region in the sample is approx. 10 nm. We also measured the temperature dependence of magnetization at 0.05 T (M-T) before/after the EIR; the results are shown in Figure 5. The M-T curve before EIR treatment exhibits a spin-glass like magnetization peak at around 50 K. The M-T curve after EIR treatment showed an almost constant magnetization value above 50 K. These results are supported by the hypothesis mentioned above, i.e., the sample before and after EIR treatment is paramagnetism and superparamagnetism at room temperature, respectively. These data also explain the
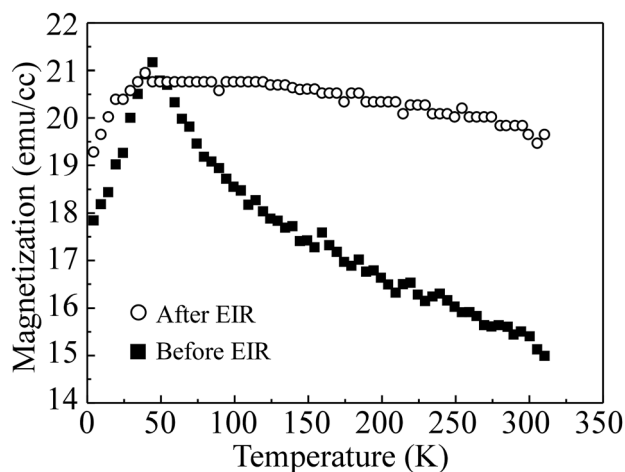

FIG. 5. The temperature dependence of the magnetization curve measured at 0.05 T. Black squares: the sample before EIR treatment. Open circles: the sample after EIR treatment.

MR behavior. The sample beneath the electrode was changed from paramagnetism to superparamagnetism by the EIR treatment. In this case, magnetic moments are suddenly ordered by the application of a magnetic field. Then, the electron scattering that originated from the magnetic moments decreases. Therefore, the sample showed a negative MR.

In this work, we demonstrated the magnetic change using EIR treatment. The sample showed resistance switching behavior due to the application of an electric field. The resistance switching ratio was about 14%. This switching behavior was reproducible at least eight times. The sample before EIR treatment barely showed negative MR, whereas the sample after EIR treatment showed negative MR approx. six times larger than that of the sample before EIR treatment. The magnetization measurement revealed that the sample was changed from paramagnetism to superparamagnetism by the EIR treatment. This magnetic state change is responsible for the MR change.

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