Magnetic and transport properties of spin-filtering tunnel junctions with magnetic insulator La$_2$NiMnO$_6$

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Abstract. Magnetic tunnel junctions (MTJs) La$_{0.7}$Sr$_{0.3}$MnO$_3$ (LSMO)/LaTiO$_3$/La$_2$NiMnO$_6$ (LNMO)/Cr/Au were fabricated on SrTiO$_3$ substrates to examine spin-filtering tunneling effect of the magnetic insulator LNMO by the measurement of tunnel magnetoresistance (TMR) through the ferromagnetic LSMO and LNMO layers. The crystal structures and magnetic properties of LNMO films were improved by growing them in high background oxygen pressure. The MTJ where the LNMO layer was prepared in oxygen pressure of 400 mTorr was found to show magnetoresistance ratio of about 10% at 50 K.

1. Introduction
Spin-polarized electron injection into nonmagnetic metals and semiconductors is of interest because it enables us to create new spintronics devices such as spin transistors [1]. Spin injection into a semiconductor from a ferromagnetic metal, however, is difficult due to the conductivity mismatch between metals and semiconductors [2]. The use of spin-filtering effect through a ferromagnetic insulator [3-6] is one of candidates to solve this problem. Moodera et al. have reported the spin-filtering effect in junctions with EuS barriers [3]. Gajek et al. have shown the spin-filtering effect of a multiferroic material La$_{0.1}$Bi$_{0.9}$MnO$_3$ (LBMO) from the tunnel magnetoresistance (TMR) effect through ferromagnetic LBMO and La$_2$/Sr$_{1/3}$MnO$_3$ layers at 60 K [6]. However, it is still difficult to realize this effect around room temperature, because ferromagnetic insulators generally have complex crystal structures and relatively low Curie temperatures. One of the ferromagnetic insulators which has high Curie temperature (280 K) is La$_2$NiMnO$_6$ (LNMO) [8]. Hashisaka et al. prepared tunneling structures composed of ferromagnetic insulator LNMO, ferromagnetic metal La$_{0.7}$Sr$_{0.3}$MnO$_3$ (LSMO), and nonmagnetic LaTiO$_3$ spacer (LTO) between them, and demonstrated that the magnetoresistance of -0.16% was observed at 150 K [9]. The observed TMR effect was very small, so that further works are required to realize larger spin-filtering effect through LNMO. In this paper, we report on the magnetoresistance of LSMO/LTO/LNMO/Cr/Au tunnel junctions. Positive TMR of about 10% was observed at 50 K for this kind of tunnel junctions.

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2. Experiments

LNMO(100 nm) films and LSMO(150 nm)/LTO(0.8 nm)/LNMO(3.2 nm)/Cr(2 nm)/Au(5 nm) magnetic tunnel junctions (MTJs) were prepared on SrTiO$_3$ (STO) substrates using a pulsed laser deposition (PLD) at 600 °C for the oxide layers and an electron beam (EB) deposition method at room temperature for the Cr and Au layers. The ferromagnetic metal LSMO was used to detect the spin-filtering effect by the TMR through the LSMO and LNMO layers, and the nonmagnetic insulator LTO was inserted to reduce the magnetic interaction between the LSMO and LNMO layers. Stoichiometric polycrystalline oxide targets prepared by a conventional ceramic fabrication technique were ablated by KrF excimer laser ($\lambda = 248$ nm). The LNMO films were deposited in various oxygen pressures to search for the optimal condition of the film growth. For the tunnel junctions, the LSMO and LTO layers were grown in background oxygen pressure of 300 mTorr, and the LNMO layers in oxygen pressure of 300 and 400 mTorr. After the deposition of LSMO/LTO/LNMO layers, Cr/Au layers were deposited on them. The crystallographic qualities and the lattice constants of the LNMO films were investigated using X-ray diffraction (XRD) (Cu K$_\alpha$ radiation). Hysteresis loops and temperature dependence of magnetization were investigated for the LNMO films using a superconducting quantum interference device (SQUID) magnetometer. The MTJs were fabricated using photolithography and dry etching by ion-milling. The MTJs were designed as ellipse-shaped pillars of several $\mu$m$^2$.

3. Results and Discussion

The XRD patterns of LNMO films grown in oxygen pressure of 10, 100, 200 and 300 mTorr are shown in figure 1(a). The LNMO(008) peaks were observed for the films grown in the oxygen pressure of 10, 200 and 300 mTorr. For the films deposited in the oxygen pressure of 100 mTorr, the LNMO(008) peak was not observed, because the LNMO(008) and STO(004) peaks overlapped. Figure 1(b) shows the lattice constant of LNMO films as a function of background oxygen pressure. This result indicates that the substrate-induced lattice distortion occurred in the LNMO films grown in low oxygen pressure. LNMO films with small lattice distortion were obtained by grown in the pressure of more than 200 mTorr, which agrees with the result by Hashisaka et al. [9]. Figure 2(a) shows the magnetic hysteresis loops for the LNMO films measured at 5 K in the field range -5000 Oe $\leq$ H $\leq$ 5000 Oe. It is clear that the saturation magnetization increased with increasing oxygen pressure during deposition, and the highest saturation magnetization was obtained for the film grown in 300 mTorr. The saturation magnetization for the sample with 300 mTorr at 5 K is about 5 $\mu$B per unit cell. This value is consistent with the bulk magnetization of LNMO [7]. Figure 2(b) shows the temperature dependence of magnetization in 10 kOe for the LNMO films from 5 to 300 K. For the films deposited in the oxygen pressure of 300 mTorr, the residual magnetization disappeared at about 170 K due to the ferromagnetic-paramagnetic transition. The Curie temperature of this film is lower than that of bulk LNMO (280 K). The Curie temperature increased with increasing the background oxygen pressure during the deposition. This result implies that the magnetic properties including Curie temperature can be improved by depositing the LNMO layers in oxygen pressure of over 300 mTorr.

On the basis of the above results for the LNMO films, we prepared the MTJs with the LNMO layers deposited in oxygen pressure of 300 and 400 mTorr. The magnetoresistance curve at 10 K for the MTJ with the LNMO layer deposited in oxygen pressure of 300 mTorr is shown in figure 3(a). The resistance increased monotonically with sweeping the field from -3000 Oe and a sharp drop occurred at a positive field. This behavior can be attributed to the TMR effect which reflects the parallel and antiparallel magnetic alignment between the LNMO and LSMO films. The magnetoresistance ratio of this MTJ was about 9% at 10 K but not detectable at 50 K. Figure 3(b) shows the magnetoresistance curve at 50 K for the MTJ with the LNMO layer deposited in oxygen pressure of 400 mTorr. The magnetoresistance ratios of this MTJ were
about 7% at 5K and 10% at 50 K. The fully antiparallel alignment may not be achieved because of the remaining ferromagnetic coupling between the LNMO and LSMO layers. We note that the positive sign of TMR ratio was obtained in this study. This result is inconsistent with the previous result by Hashisaka et al., where inverse TMR ratio of -0.12% was observed [9]. This contradiction may be connected with the difference of the surface condition or crystal defects in the junctions, which affect the magneto-transport properties. Further research is needed to get a deeper insight into the spin-filtering effect of LNMO barriers.

4. Conclusion

The LNMO films and LSMO/LTO/LNMO/Cr/Au MTJs were fabricated using a PLD and an EB deposition method. XRD profiles showed that the LNMO films with small lattice distortion were obtained in oxygen pressure of over 200 mTorr during the PLD process.
Figure 3. The magnetoresistance of the spin-filtering tunnel junctions, with the magnetic field sweeping from -3000 to 3000 Oe. (a) The magnetoresistance curve at 10 K for the sample with the LNMO layer deposited in oxygen pressure of 300 mTorr. (b) The magnetoresistance curve at 50 K for the sample with the LNMO layer deposited in 400 mTorr.

measurements demonstrate that the saturation magnetization of LNMO films grown in oxygen pressure of 300 mTorr corresponded to that of bulk LNMO and that the Curie temperature of LSMO films increased with increasing the background oxygen pressure during the deposition. The magnetoresistance ratio of about 10% at 50 K was observed in the magnetoresistance measurement for the tunnel junction when the LNMO layer was deposited at 400 mTorr.

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