Tunnelling Magnetoresistance at Low Magnetic Fields in LSMO/STO/LSMO Magnetic Tunnel Junction

Ramazan TOPKAYA1

ABSTRACT: La_{0.7}Sr_{0.3}MnO₃/SrTiO₃/La_{0.7}Sr_{0.3}MnO₃ magnetic tunnel junction was grown on (001)-oriented SrTiO₃ substrate by using pulsed laser deposition technique. La_{0.7}Sr_{0.3}MnO₃ (LSMO) was used as ferromagnetic (FM) electrodes and SrTiO₃ (STO) was used as a tunnel barrier. The effect of temperature and magnetic field on electrical transport properties has been investigated. Bottom electrode and junction resistances were observed to strongly depend on temperature. Large magnetoresistance of almost 300 % was observed at 77.3 K at low magnetic fields of a few tens of Oe. The large magnetoresistance was explained by the nearly half-metallic band structure of La_{0.7}Sr_{0.3}MnO₃. The asymmetry of TMR curves was attributed to the ferromagnetic electrodes with different thickness and the differences in the electronic states of the upper and lower STO/LSMO interfaces.

Keywords: La_{0.7}Sr_{0.3}MnO₃, magnetic tunnel junction, spintronics, tunnelling magnetoresistance

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LSMO/STO/LSMO Manyetik Tünel Ekleminin Zayıf Manyetik Alanlarda Tünelleme Manyetodirenci

ÖZET: Darbeli lazer biriktirme tekniği kullanılarak $La_{0.7}Sr_{0.3}MnO_3/SrTiO_3/La_{0.7}Sr_{0.3}MnO_3$ manyetik tünel eklemi SrTiO₃ alttaş üzerinde büyütüldü. Ferromanyetik elektrot için $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) ve tünelleme bariyeri için SrTiO₃ (STO) kullanıldı. Sıcaklığın ve manyetik alanın elektriksel özelliklere etkisi araştırıldı. Alt elektrot ve eklem dirençlerinin çok güçlü bir şekilde sıcaklığa bağlı olduğu gözlemlendi. 77.3 K sıcaklık ve zayıf manyetik alanlarda yaklaşık % 300'lük yüksek manyetodirenç gözlemlendi. Gözlemlenen yüksek manyetodirenç $La_{0.7}Sr_{0.3}MnO_3$ 'ün yarımetalik band yapısından kaynaklanır. TMR eğrilerinin asimetrik yapısı, farklı kalınlıktaki elektrotlar ile üst ve alt STO/LSMO ara yüzeylerin elektronik yapılarının farklılığına atfedildi.

Anahtar Kelimeler: La_{0.7}Sr_{0.3}MnO₃, manyetik tünel eklemi, spintronik, tünelleme manyetodirenci

Iğdır Üniversitesi, Mühendislik Fakültesi, Elektrik-Elektronik Mühendisliği, Iğdır, Türkiye Sorumlu yazar/Corresponding Author: Ramazan TOPKAYA, ramazan.topkaya@igdir.edu.tr

INTRODUCTION

In recent years magnetic tunnel junctions (MTJs) consisting of two ferromagnetic layer separated by a thin insulating tunnel barrier have been actively studied due to their potential applications in magnetoelectronics and spintronics such as magnetic random access memory (MRAM), GMR or TMR read heads and magnetic field sensors (Wolf et al., 2001; Wolf et al., 2006; Natarajarathinam et al., 2012; Boyraz et al., 2016; Sahin, 2016). The large TMR ratio arises from the ferromagnetic electrodes with high effective spin polarization values (Tsymbal et al., 2003). If two ferromagnetic layers are fully spin-polarized, i.e. they have 100 % spin polarization, TMR ratio theoretically becomes infinite.

Recently there has been a great deal of attention on manganese perovskites, La_{1-x}D_xMnO₃ (D=Ca, Sr, Ba, etc.), which exhibit a large change in resistance when subject to a high magnetic field (an order of a few Tesla). The effect is called "colossal" magnetoresistance because of very large scale of the effect (~ thousand percent). However, the high magnetic field (one Tesla or more) used to obtain the resistance changing has limited the prospects of this class of materials for potential applications in magnetoelectronics. A required magnetic field on the order of several Tesla should be reduced in order to obtain high sensitivity magnetic field sensor. On the other hand, it is known that the manganese perovskites, La_{0.7}Sr_{0.3}MnO₃ (LSMO) is a half-metallic ferromagnetic material with the spin polarization of 95 % at the Fermi level (Park et al., 1998). Therefore, a possible approach to decrease the required large magnetic field is to incorporate the LSMO into magnetic tunnel junction (Miyazaki and Tezuka, 1995; Moodera et al., 1995). Sun et al., (1996) observed a factor of 2 change in resistance in an applied magnetic field of less than 200 Oe for doped manganite perovskites by using SrTiO₂ barrier. Lu et al., (1996) obtained a large magnetoresistance ratio of 83 % in a low magnetic field of the order of few tens of Oe at T=4.2 K for epitaxial LSMO/STO/LSMO magnetic tunnel junctions.

In this study, the magnetic tunnel junction with the structure LSMO/STO/LSMO was grown on (001)-oriented STO substrate by using pulsed laser deposition technique. Due to a good lattice match between LSMO and STO, STO was used as the insulating barrier. The large magnetoresistance ratio of about 300 % was obtained at a relatively low field of almost 80 Oe at 77.3 K.

MATERIAL AND METHODS

La_{0.7}Sr_{0.3}MnO₃ (LSMO) film was grown on (001)-oriented SrTiO₂ (STO) substrate by using pulsed laser deposition with a base pressure of about 2x10 000 000⁻¹ Torr. The deposition was carried out in an UHVchamber using a KrF excimer laser (2 Hz repetition rate and 248 nm wavelength) with an energy density of 1.2 J cm^{2 -1} at a substrate temperature of 700 °C. For the LSMO and STO depositions an oxygen atmosphere has been used as 200 mTorr and 100 mTorr, respectively. After the deposition the sample was cooled down to room temperature. The thicknesses of the top and bottom LSMO electrodes are 50 and 30 nm, respectively. The thickness of the STO barrier is 6 nm. Total thickness of the MTJ structure is about 86 nm. Magnetic tunnel junction with a cross section of 6x2.5 µm² was fabricated by photolithography and Ar ion milling as detailed in Ref. (Lu et al., 1996). The electrical measurements were performed by using PPMS-Dynacool (Quantum Design Inc.), Agilent HP voltmeter and Keithley 428 current amplifier. The resistivity of the sample was measured using a standard dc four-probe technique as a function of field and temperature.

RESULTS AND DISCUSSION

The temperature dependence of the resistance of the bottom electrode (LSMO) is shown in Figure 1. The resistance strongly depends on temperature and increases almost 12 times with increasing temperature between 82-294 K. The bottom LSMO electrode shows a metallic behaviour. The temperature dependence of the bottom electrode resistance is attributed to a reduced effective ordering temperature at LSMO/insulator interface (Viret et al., 1997). The same results were observed in manganite tunnel spin valves (Viret et al., 1997), nanocrystalline $La_{0.7}Sr_{0.3}MnO_3$ synthesized on MgO nanowires (Zhang et al., 2010) and LSMO/AlO/ CoFeB magnetic tunnel junction (Rizwan et al., 2010).



Figure 1. Bottom electrode resistance versus temperature plot for LSMO(50 nm)/STO(6 nm)/LSMO(30 nm) junction

Figure 2 indicates the resistivity versus temperature for the LSMO(50 nm)/STO(6 nm)/LSMO(30 nm) junction. From the transport measurement, the transition temperature is determined to be about 109 K for the junction. With decreasing temperature, the resistance of the junction first slowly increases up to 109 K and later rapidly increases between 109-78 K, indicating a semiconductor or insulator-type behaviour. This result has also been reported previously by Gupta et al., (1995), Gong et al., (1997) and Zhang et al., (2010). In semiconductors and insulators, if the temperature increases, electrons generally get energy, go to the conduction band and become free to carry the current. In this case, the conductance increases. Therefore, the resistance decreases with rise in temperature (Wikipedia, 2017).



Figure 2. Temperature dependence of resistance of LSMO/STO/LSMO junction

The variation of the tunnelling magnetoresistance is examined by applying different bias currents on the junction. Figure 3 shows the field dependence of the tunnelling resistance magnetoresistance of the LSMO(50 nm)/STO(6 nm)/LSMO(30 nm) MTJ junction with area $6x2.5 \mu m^2$ measured at T=77.3

K for the bias current range of 100 pA-100 nA. The external magnetic field was applied along in-plane geometry. The observed maximum TMR ratios for each bias current are obtained at the relatively low magnetic fields like as ~80 Oe. It is seen from Figure 3 that with increasing the bias current (voltage), TMR ratio decreases. The increasing of the bias give rise to increase the Fermi level of emitter electrode and gives more energy to the emitted electrons, causing them to access further to empty states in the collector electrode especially in the antiparallel orientation. Therefore, the resistance decreases with increase in bias.

It was observed the asymmetric TMR peaks in terms of position and magnitude with respect to applied magnetic field for each bias current. There are the different reasons of the asymmetry in the MTJ structures. The different thickness of the two ferromagnetic electrodes (LSMO) can be responsible for the asymmetric position and magnitude of TMR peaks (Miao et. al., 2009). This asymmetry can be also caused by differences in the electronic states of the upper and lower STO/ LSMO interfaces. The same result has been reported for CoFeB/MgO/CoFeB magnetic tunnel junctions by Feng et. al (2009). The barrier height and thickness can result in the asymmetry in TMR peaks. For example, double barrier magnetic tunnel junctions exhibit high asymmetry with respect to the single barrier magnetic tunnel junctions (Feng et. al., 2009). The oxidized layer at the two interfaces or interface oxidation (Liu et. al., 2007), growth conditions, impurities, annealing regimes (Useinov and Kosel, 2011) and interface quality can give rise to the asymmetry in the TMR peaks. As the oxidized layer gets thicker, the degree of asymmetry increases in the work of Liu et. al. (2007). On the other hand, the annealing almost removes the bias asymmetry (Feng et. al., 2009).

The maximum change in magnetoresistance was observed as \sim 300 % at T= 77.3 K between the parallel and antiparallel orientations of the magnetization of the two electrodes. Table 1 compares the experimental TMR values obtained for LSMO/STO/LSMO MTJ junctions at different temperatures. The observed magnetoresistance ratio in a relatively low magnetic field of ~80 Oe is larger than the values observed for La_{0.67}Sr_{0.33}MnO₃ tunnel junctions (Lu et al., 1996), $La_{0.67}Sr_{0.33}MnO_3$ and $La_{0.67}CaO_{0.33}MnO_3$ thin films (Li et al., 1997). On the other hand, this value is smaller than that of observed in work of Bowen et al. (1800 % TMR in LSMO/SrTiO,/LSMO junctions, 2003) and Werner et. al. (2011). TMR ratio decreases with increase in temperature (Wu et al., 2015). The magnetoresistance ratios of 1800 and 1400 % were obtained at 4 K respectively by Bowen et al., (2003) and Werner et. al. (2011). In this study, the TMR ratio was obtained at 77.3 K. One of the reasons why the TMR ratio is smaller than that of observed in work of Bowen et al. (2003) and Werner et. al. (2011) is that the measurement temperature is higher than 4 K. The large magnetoresistance value is due to the half-metallic behaviour of the LSMO manganite (Lu et al., 1996; Bowen et al., 2003) and LSMO/STO interface.



Figure 3. Tunnelling magnetoresistance of LSMO/STO/LSMO MTJ junction as a function of magnetic field at different bias currents range from 100 pA to100 nA.

Structure	TMR Ratio (%)	Temperature (K)	References
La _{2/3} Sr _{1/3} MnO ₃ /SrTiO ₃ /La _{2/3} Sr _{1/3} MnO ₃	1800	4	(Bowen et. al., 2003)
$La_{_{0.67}}Sr_{_{0.33}}MnO_{_{3}}/SrTiO_{_{3}}/La_{_{0.67}}Sr_{_{0.33}}MnO_{_{3}}$	83	4.2	(Lu et. al., 1996)
$La_{0.67}Sr_{0.33}MnO_{3}/SrTiO_{3}/La_{0.67}Sr_{0.33}MnO_{3}$	15	4.2	(Li et. al., 1997)
$La_{0.65}Sr_{0.35}MnO_{3}/SrTiO_{3}/La_{0.65}Sr_{0.35}MnO_{3}$	1400	4	(Werner et. al., 2011)
$La_{0.67}Sr_{0.33}MnO_{3}/SrTiO_{3}/La_{0.67}Sr_{0.33}MnO_{3}$	100	13	(Noh et. al., 2001)
$La_{0.7}Sr_{0.3}MnO_3/SrTiO_3/La_{0.7}Sr_{0.3}MnO_3$	300	77.3	This work

Table 1. Comparison of TMR values of the magnetic tunnel junction with structure of LSMO/STO/LSMO

CONCLUSION

In conclusion, the magnetic tunnel junction with the structure LSMO/STO/LSMO was fabricated by using pulsed laser deposition. The electrical properties of the MTJ were investigated. It is shown that the LSMO electrode resistance decreases, whereas the LSMO/STO/LSMO junction resistance increases with decreasing temperature. The maximum tunnelling magnetoresistance (TMR) ratio measured at 77.3 K was found to be almost 300 % in the magnetic field of about 80 Oe, proving a half-metallic ferromagnetic property

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