*Transactions of the Materials Research Society of Japan* 29[4] 1437-1440 (2004)

# Preparations and Evaluations of Magnetoelectric Oxides / Superconductor Multilayers

Nobuyuki Iwata, Koji Matsuo and Hiroshi Yamamoto

College of Science and Technology, Nihon University, Japan

Fax : +81-47-467-9683

We proposed the introduction of magnetoelectric (ME) materials as a gate insulator to the Josephson field effect transistor (JFET). Magnetic field is induced with electric filed applied to the ME materials. We named it Josephson ME field effect transistor (JMEET). Critical current $I_C$ and/or superconducting transition temperature $T_C$ should be modulated considering the Fraunhofer pattern. We grew representative Cr2O3 thin films on ITO substrate. Calculated magnetic field was 1.26 gauss at Josephson junction area, which was sufficient to modulate the $I_C$. Multilayer of Cr2O3 / YBa2Cu3OX was grown and evaluated on MgO substrate.

Key words : magnetoelectric, JFET, JMEET, Cr2O3

## 1. INTRODUCTION

Since the discovery of high temperature superconducting oxides, strongly correlated oxides have been investigated energetically. In such oxides, spin, charge, orbit and light are strongly coupled. So the oxides have been paid attention as various functional materials, of which diverse phenomena are varied by doping, pressure, chemical composition and magnetic field[1]. For example, in the LaMnO3 series, magnetic and electric transitions occur upon doping. Those transitions caused by double exchange interaction are the result due to the coupling between magnetic and electric phenomena. Noticeable point is that the electric behavior is a parameter to control the function of the materials. In other words, if the strongly correlated oxides are insulator without doping, such the oxides are not functional any more.

Meanwhile, we actively paid attention to control the magnetic properties by electric insulator. This attempt led to the novel function except for that in strongly correlated oxides, which was a function of field effect. Magnetoelectric (ME) effect was worthy of remark[2].

Magnetoelectric effect is characterized by the appearance of an induced magnetization with electric field applied, and the appearance of an induced electric polarization with magnetic field applied. Antiferromagnetic insulator, Cr2O3, is a representative among ME materials. As shown in Fig.1, when the electric field was applied, the position of $Cr^{3+}$ ions was shifted relative to the lattice of negative oxygen ions. This produces a change in the orbital contribution to the magnetic moment of the $Cr^{3+}$ ions. Between + site and – site, the super exchange interaction change, strengthening the magnetic ordering for + site, while loosening it for – site[2,3]. Totally magnetic field appears.

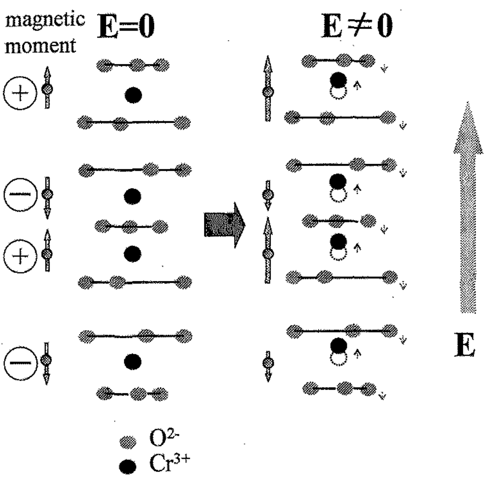


Fig.1 : Electric field effect of Cr2O3

By the way various kinds of superconducting three-terminal devices have been developed, however, the change of $I_C$ or $T_C$ are not so large[4]. We proposed new type Josephson field effect transistor (JFET) as adopting the ME materials to a gate insulator. We named it Josephson ME field effect transistor (JMEET). In JMEET device large $I_C$ modulation was expected as the induced magnetic field, controlled by electric field, penetrated through the Josephson junction as shown in Fig.2. Josephson junction was generated by itself due to the large lattice mismatch between YBa2Cu3OX (YBCO) and MgO substrate. In this study, we prepared and evaluated Cr2O3 thin films and Cr2O3 on YBCO thin films.

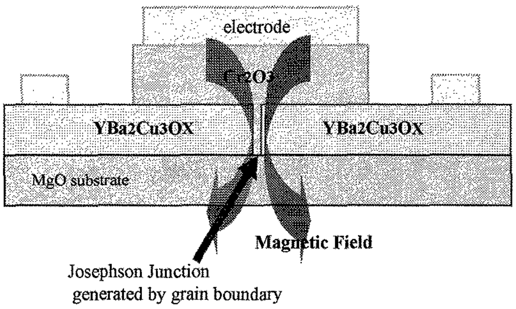


Fig.2 : Schematic image of Josephson magnetoelectric field effect transistor (JMEET)

## 2. EXPERIMENTAL PROCEDURES

Thin films of Cr2O3 were grown on ITO substrates for 2 - 4 h by off-axis RF magnetron sputtering method without heating as shown Fig.3. Target was sintered Cr2O3 with the size of 80 mm ϕ in diameter. The sputtering gas pressure and RF power were 2.0 Pa (100% Ar) and 100W.

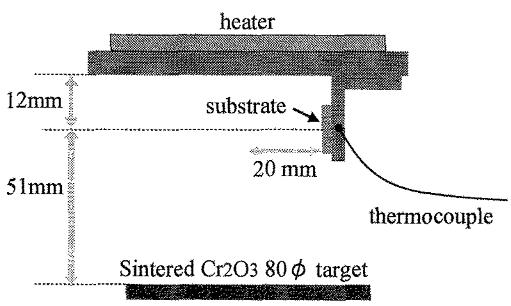


Fig.3 : Growth chamber of RF magnetoron sputtering for Cr2O3 thin films

As shown in Fig.4 the YBCO thin films were grown on MgO substrates by off-axis DC-RF magnetron sputtering method. MgO substrates were annealed in air at 1000 °C for 4 h in advance. Annealed substrates were hold at the sample holder inserted with Pt foil between substrates and sample holder. Back of the sample holder was heated by infrared radiation and the temperature at the side of the holder was detected by thermocouple while growing, which was 800 °C. Sintered YBCO 52 mm ϕ target was sputtered with RF power of 50W and DC current of 0.3A. Sputtering atmosphere was 26 Pa with the flow rate of 45 ccm in Ar and O2 (Ar : O2 = 1: 1 ). Growth was done for 0.5 and 2 h.

The surface morphology was obtained by atomic force microscopy (AFM, SII : SPI3800). Resistance of the YBCO was measured as a function of temperature, ranging from room temperature to liquid nitrogen temperature by four probe method. Gate voltage was applied at the electrode of top of the Cr2O3 thin films as shown in Fig.2.

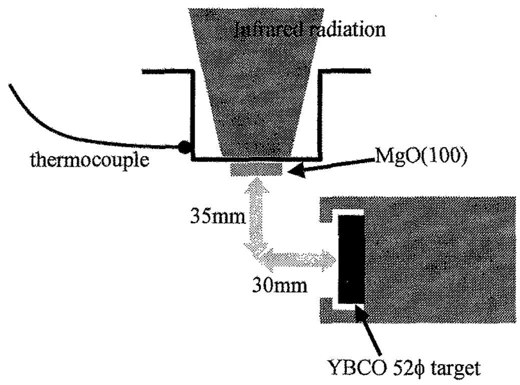


Fig.4 : Growth chamber of DC-RF magnetoron sputtering for YBa2Cu3OX thin films

## 3. RESULTS and DISCUSSION

In Fig.5, the I-V characteristic normal to the film plane of Cr2O3 thin film on ITO substrate is shown. Film thickness and electrode diameter was 22 nm and 0.45 mm ϕ. At 0.28 MV / cm, leakage current was $5.2\times10^{-3}$ A / cm$^2$. Below 0.3 MV /cm, grown Cr2O3 worked as gate insulator.

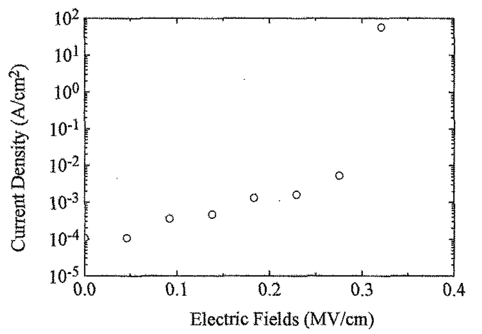


Fig.5 : I-V characteristic of Cr2O3 thin film with 22 nm thickness. The size of measurement electrode was 0.45 mmϕ .

We calculated the induced magnetic field at 0.3 MV /cm. Assuming that the linear magnetoelectric susceptibility of Cr2O3 was $10^{-6}$ , induced magnetization *Pm* was $10^{-3}$ emu by the equation (1), which was correspondence with magnetic filed *B* of $1.26 \times 10^{-2}$ gauss. Assuming that the ratio of Josephson junction and superconducting area was 1 versus 100, and Meissner effect made the induced magnetic field concentrate at the junction. Magnetic field *B* at the junction was 1.26 gauss. In the case of 10μm$^2$ JMEET, magnetic flux was almost correspondence with fluxoid Φ0. Considering the

Fraunhofer pattern, junction *I*C should be modulated to zero by the induced magnetic field.

$$Pm = \alpha E \qquad (1)$$

Figure 6 shows the 5μm² AFM images and line profiles of YBCO thin films with thickness of about (a) 50 nm and (b) 200 nm on annealed MgO substrates. Superconducting transition temperatures *T*C were 77K and 92K, respectively. Many grains were clearly seen and separated by deep grain boundaries. In Fig.6(a), some of the boundaries, the depth was almost correspondences with the film thickness. In Fig.6(b), the deepest boundary was about 60 nm.

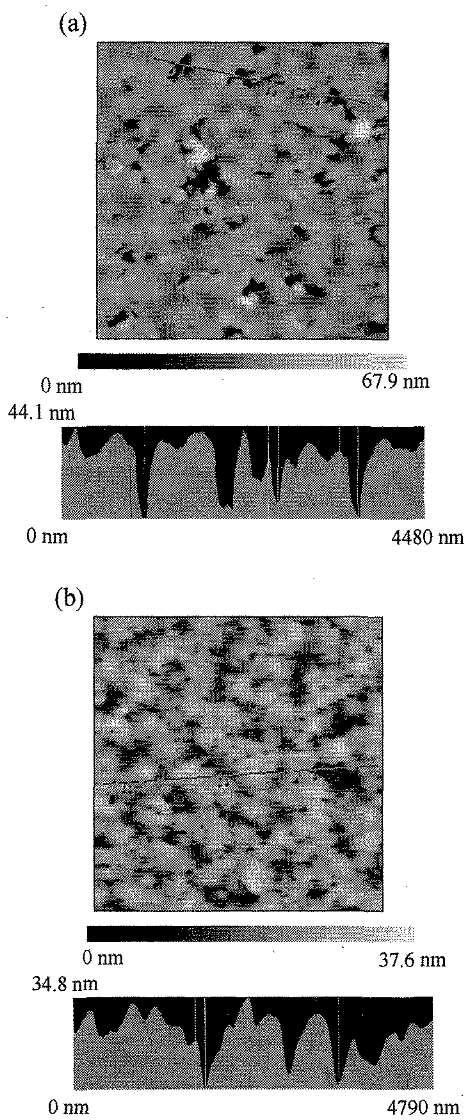


Fig.6 : 5μm² AFM images and line profiles of (a) 50 nm thick YBCO and (b) 200 nm thick YBCO films.

After deposition of Cr2O3 and annealing at 400 °C for 6h in O2 flow to compensate the oxygen vacancies of YBCO film, we measured the temperature dependence of resistivity of 50 nm thick YBCO film as shown in Fig.7. With decreasing temperature, resistivity was increasing like semiconductor. Superconducting YBCO thin film became semiconductor after deposition and annealing.

In Fig.8 temperature dependence of resistivity of 200 nm thick YBCO film after Cr2O3 deposition and annealing. The YBCO thin film seemed to show superconducting transition at 87.8 K. But large residual resistivity of $9.4\times10^{-4}$ Ωcm remained.

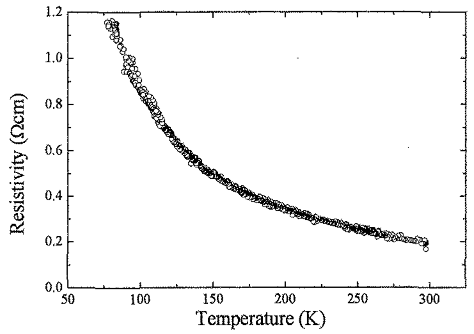


Fig.7 : Temperature dependence of resistivity of 50 nm thick YBCO film after Cr2O3 deposition and annealing.

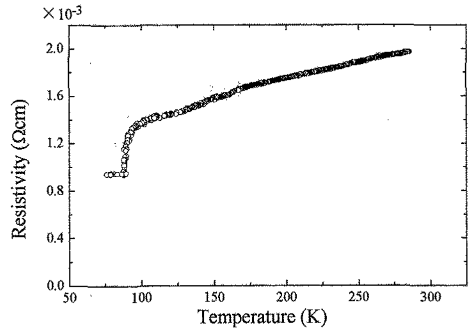


Fig.8 : Temperature dependence of resistivity of 200 nm thick YBCO film after Cr2O3 deposition and annealing.

Figure 9 shows the temperature dependence of resistivity of 200 nm thick YBCO film without annealing. Comparing the annealed YBCO thin film after Cr2O3 deposition in Fig.8, residual resistance was relatively small but remained. At 77K in the films of Fig.8 and 9, gate voltage was applied up to 0.3 MV /cm, remarkable change was not observed.

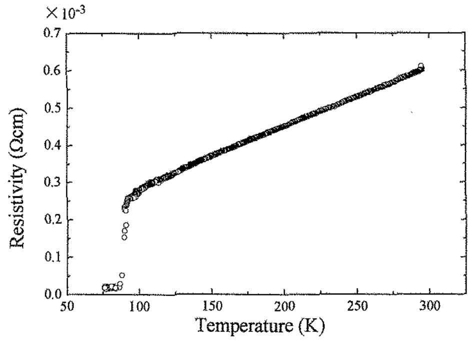


Fig.9 : Temperature dependence of resistivity of 200 nm thick YBCO film after Cr2O3 deposition without annealing.

After annealing to compensate the oxygen vacancies of YBCO films, the *TC* was slightly recovered but residual resistivity was increased. These results indicated that the superconducting YBCO thin film, on which no Cr2O3 was deposited with masking, was oxidized as we thought. But we expected that the annealing induced a diffusion of Cr into YBCO layer, on which Cr2O3 was deposited, and then lead to normal metal. Film with 50 nm thick did not show even metallic behavior any more. On such the Cr diffused YBCO films although the magnetic moment was induced, the modulation of *IC* and/or *TC* was not observed. In the future investigation we have to insert the suitable blocking layer for Cr diffusion between Cr2O3 and YBCO.

## 4. SUMMARY

We proposed new type Josephson field effect transistor, JMEET, in which the magnetoelectric materials were adopted as a gate insulator. We expected the large modulation of *IC* and *TC* by induced magnetic field with electric field applied. Representative Cr2O3 thin films were grown and calculated magnetic field was 1.26 gauss at the Josephson junction area. In the multilayered structure of Cr2O3 / YBCO, we expected the Cr diffusion into the YBCO layer.

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(Received October 13, 2003; Accepted July 1, 2004)