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Barrier height and tunneling aspects in (110) CrO$_2$ with its natural barrier

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Abstract: We have investigated (110) CrO$_2$/natural barrier/Co magnetic tunnel junctions for their barrier and magneto-transport properties. A negative tunnel magnetoresistance (TMR) of over 5% was observed in micro-fabricated devices at 4.2 K, which is comparable to TMR values obtained with (100) CrO$_2$. Both transport and cross-sectional transmission electron microscopy analysis reveal a natural barrier thickness 3.5 ± 0.5 nm. However, we obtain a low effective barrier height of 0.4 eV from transport measurements. The inelastic electron tunneling spectroscopy showed significant bias dependence with peak positions showing vibrational modes, which deviate from stoichiometric Cr$_2$O$_3$. We conclude that the transport characteristics are controlled by defects within the natural barrier, consistent with recent theoretical reports.

I. INTRODUCTION

Because of the experimentally observed high spin-polarization in CrO$_2$ thin films, magnetic tunnel junctions with CrO$_2$ as an electrode have been the focus of intense research for both its fundamental and technological implications. So far, tunnel magnetoresistance (TMR) value consistent with its high spin-polarization has not been reported in a device with CrO$_2$ as an electrode. Observed TMR in CrO$_2$/natural barrier/Co junctions at 4.2 K has been ~8% with Leo et al. showing that the sign of TMR changes from negative to positive with the insertion of a thin Mg layer probably by preventing the growth of the natural barrier. The thin, natural, stable and insulating oxide of Cr on top of CrO$_2$ thin films that is used as the tunneling barrier in these junctions has been attributed to be the most stable oxide of Cr, i.e., Cr$_2$O$_3$ or an oxygen-deficient variant (CrO$_x$, x < 1.5). In this work we probe the transport characteristics through the natural barrier, grown on (110)-oriented CrO$_2$ junction. I-V characteristics estimate the barrier height in the CrO$_2$/natural barrier/Co junction and provide information about the barrier itself and valuable physical insight into the transport mechanism. Also, all the previously reported CrO$_2$ devices are on epitaxial (100) CrO$_2$ films grown on isostructural TiO$_2$ substrates. As has been recently found by our group, epitaxial (100) CrO$_2$ films are subject to substrate-induced strain up to significantly high film thickness (~200 nm) while (110) CrO$_2$ films grow strain-free when grown on respective TiO$_2$ substrates with (100) CrO$_2$ films following layer-by-layer growth while (110) CrO$_2$ films showing island-like growth modes. The room temperature magnetization value of (110) CrO$_2$ is also substantially enhanced compared to (100) CrO$_2$ due to strain-free growth. Therefore, if exploited properly, it is expected to translate to better CrO$_2$ device performance. With this motivation, we investigated devices with (110) CrO$_2$ as the high spin-polarization electrode.

II. EXPERIMENT

For our experiment, we deposited the CrO$_2$ layer on (110) TiO$_2$ substrate using the selective growth method described previously. The (110) CrO$_2$ film thickness in case of all the results reported here is 50 nm. To get the robust natural insulating barrier, the CrO$_2$ film is kept under ambient conditions for about two weeks. The film is then patterned using standard photolithography technique to define the TMR devices for our experiment. Co (~60 nm) and Au (~30 nm) were sputtered in a vacuum chamber with base pressure less than $5 \times 10^{-7}$ Torr to grow the free magnetic layer and the capping layer, respectively, in TMR devices. We observed that the devices are shorted and did not show any TMR if they are fabricated immediately after selectively growing the (110) CrO$_2$ film. This indicates that the natural barrier evolves with time. The cross-sectional images were obtained in a FEI Tecnai F20 transmission electron microscope (TEM). Fabricated devices were tested for their TMR properties in a home-made measurement set-up at liquid nitrogen temperature (~78 K) since the devices did not show any room temperature TMR—as reported previously. Distinct magnetic switching is important to observe TMR effect, which in our case is 100 Oe and 400 Oe for the Co and CrO$_2$ layers, respectively, at 78 K. No significant effect of shape anisotropy was observed on the switching and TMR behavior and the reported result here is for a typical junction of size $3 \times 6 \mu m^2$. Temperature-dependent TMR data and I-V properties were obtained in a Quantum Design physical property measurement system (PPMS). Inelastic electron tunneling spectroscopy (IETS) on the device(s) was collected with our home-made measurement set-up and attaining the low temperature of 2 K for this measurement in the PPMS. Details of our IETS measurement are described later with the results.

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All reported I-V and IETS spectra here are averaged from several repeated scans between ±100 mV.

III. RESULTS AND DISCUSSION

We first present our TEM analysis which provides information about the barrier thickness and film microstructure in an unambiguous manner. Figure 1(a) shows the bright-field TEM images of CrO$_2$/natural barrier/Co tunnel junction deposited on (110) TiO$_2$ substrates taken along the (110) zone-axis. As shown in this figure, the insulating barrier (the bright contrast between Co and CrO$_2$) was found to have relatively sharp interfaces with a thickness of about 3.5 ± 0.5 nm (agrees well with the Brinkman, Dynes, and Rowell (BDR) fit for barrier thickness, as discussed later). The variation in barrier thickness (0.5–1.0 nm) is consistent with inherent roughness of (110) CrO$_2$ films.\(^5\) The high resolution TEM image in Fig. 1(b) shows the homo-epitaxial growth of (110) CrO$_2$ with lattice fringes corresponding to the orientation relationship (110) CrO$_2$||(110) TiO$_2$ and also polycrystalline growth of the Co layer on top of the natural barrier. The TiO$_2$/CrO$_2$ interface shows the presence of misfit dislocations. Note that the CrO$_2$/natural barrier interface shows little contrast owing to the strain-free growth of natural barrier. As seen in the figure, the natural barrier exhibits a crystalline structure close to CrO$_{2-x}$ as reported previously.\(^4\) However, the natural barrier/Co interface exhibits a dark contrast owing to stacking fault-like defects, which could also be a major source of spin-scattering.

As shown in Fig. 2, the junction resistance of the device increases from 300 K to 2 K when measured in PPMS with four point probe method and dc current of 1.0 µA. Junction resistance is close to 2 kΩ from 300 K to ~200 K, and it increases almost 3 times (~5.5 kΩ) at 2 K. This indicates that tunneling is the most likely mode of transport in this device. An increase in junction resistance can be attributed to a few factors. Both metallic electrodes’ (CrO$_2$ and Co) resistance decreases significantly with decreasing temperature (not shown). Also, thermal fluctuations are reduced with decreasing temperature. Increasing magnetic moment with decreasing temperature\(^6\) also contributes to the increase in the resistance of the junction via spin-dependent tunneling.\(^8\) TMR observed in this device is −5.3% at 4 K (Fig. 3), which is similar to negative TMR observed in devices with (100) CrO$_2$.\(^1,4\) Also, no noticeable TMR was observed at temperatures above 200 K (data not shown), which is also similar to (100) CrO$_2$ device performances.

Assuming spin polarization of CrO$_2$ to be 98% and that of Co to be −42%, then according to the Julliere model,\(^9\) TMR value for this junction is expected to be about −57% at low temperature. The observed TMR is much lower than the expected value. This indicates that there is significant spin-flip process(es) taking place at the interface(s) with the barrier. As CrO$_{2-x}$—the presumed tunneling barrier in our devices is known to be weakly antiferromagnetic,\(^10\) significant spin-flip scattering at the barrier interfaces is likely to reduce the TMR effect. Also, (110) CrO$_2$ follows island-like growth mode on isostuctural TiO$_2$ substrates and roughness values of about 1.0 nm for a 50 nm film\(^5\) might possibly make the barrier less effective to observe a significant TMR effect, e.g., by promoting ferromagnetic (FM) coupling of the electrodes or intermixing. The sign of TMR in a CrO$_2$-based device is also very...
interface sensitive. However, in our devices, observed negative TMR is attributed to the negative spin polarization of top Co layer while the CrO$_2$ layer maintains a positive spin polarization due to conduction of only majority electrons. We next focus on the tunnel barrier properties.

Like in other tunnel junctions, the barrier in CrO$_2$/natural barrier/Co junction can be studied by observing its I-V characteristics (as shown in the schematic energy diagram Fig. 4). Barrier height ($\Phi$) is the potential barrier provided by the insulator of thickness $d$ between the ferromagnetic electrodes. Asymmetry in the barrier arises because of the different work functions involved with CrO$_2$ and Co. Based on the I-V characteristics of our tunneling devices, we used two models to estimate the barrier height in the junctions. The Brinkman, Dynes, and Rowell (BDR) model assumes a trapezoidal barrier as shown in Fig. 4. According to this model, conductance of the junction is expressed as a polynomial function of bias voltage up to 2nd order,

$$\frac{dI}{dV} = A + BV + CV^2$$

and, thus giving current in the device against bias voltage:

$$I = AV + \frac{BV^2}{2} + \frac{CV^3}{3},$$

where $A$, $B$, and $C$ are constants evaluated by fitting experimental data with, $I(V = 0) = 0$. On getting the constants $A$, $B$, and $C$ from fitting barrier thickness (in angstroms), barrier height (in electron volts) and barrier asymmetry (in electron volts) can be calculated from the BDR model according to the following expressions:

$$d^2 = -5.368 \sqrt{\frac{C}{A}} \ln \left( 1.747 \times 10^{-10} \sqrt{\frac{C}{A}} \right),$$

$$\Phi = 0.03281 \left( \frac{A}{C} \right) d^2,$$

$$\Delta\Phi = -23.42 \left( \frac{B}{A} \right) \frac{\phi^{3/2}}{d}.$$  

Both current and conductance in the device are fitted with the BRD model as shown in Figs. 5(a) and 5(b), respectively. We extract a barrier thickness of 4.0 nm, using both current and conductance fitting, which is in good agreement with direct barrier thickness estimation in TEM, i.e., 3.5 ± 0.5 nm. Also, this barrier thickness is close to three times the lattice parameter $c$ in Cr$_2$O$_3$ ($a = 0.496$ nm, $c = 1.36$ nm for its hexagonal corundum structure, assuming, of course, that structural lattice parameter of the barrier is close to Cr$_2$O$_3$.

Further, we also fitted the current density curves using the Simmons model, as comparison between the barrier heights is still relevant as it is not a priori clear, beyond the reasons mentioned above, which model best describes transport properties. The model proposed by Simmons assumes a rectangular shaped ideal barrier between two metallic electrodes. According to his model, current density across the junction for intermediate voltages can be approximated by

$$J = \beta (V + \gamma V^3),$$

FIG. 4. (Color online) Schematic energy diagram of IETS phenomenon between two metal electrodes sandwiching an insulating barrier. The increase in current due to inelastic tunneling of electrons at a particular voltage and corresponding changes in its first and second derivative are schematically shown.

FIG. 5. (Color online) Fitting of current (a) and conductance (b) against bias voltage with BDR model. (c) Current density against bias voltage in the device with Simmons fit.
The chemical nature of the natural barrier is probed by the IETS technique. Compared to the total current through the junction, the increase in current due to IETS (shown schematically in Fig. 4) is very small—within 1% of the total current and therefore may not be more significant than the electrical noise level at room temperature. The full width at half maxima (FWHM) of the IETS peak has a temperature dependence of 5.4k_BT (Ref. 20) and therefore to obtain a significant resolution, spectra were taken at 2 K, which corresponds to a thermal broadening of 0.93 mV. Because of asymmetry in barrier energy, inelastic tunneling electrons also see different barrier height depending on the bias polarity and accordingly their peak intensities can depend on the bias voltage polarity of the device.21 The IETS signal is obtained, considering a small ac modulation (V_m cos ωt) with the dc bias (V_b) across the junction as the 2nd order term in the following expansion:

$$I(V_b + V_m \cos \omega t) = I(V_b) + \frac{dI}{dV_b} |_{V_b} V_m \cos \omega t + \frac{d^2I}{dV_b^2} |_{V_b} V_m^2 \cos^2 \omega t + \cdots. \quad (9)$$

For a bias voltage of 150 mV, modulation voltage was 1–2 mV with modulation frequency ~1 kHz to get rid of any low frequency noise. Calculated minimum energy resolution due to ac modulation is 1.22 V_m (Ref. 20) and therefore for an ac modulation of 2.0 mV, IETS spectra can be confirmed to be correct within ±2.5 mV. Modulation broadening of the IETS spectra is therefore on the same order as thermal broadening at 2 K, which is about 1.0 mV.

Figure 6 shows the IETS spectra obtained on one of our devices at 2 K for positive and negative biases. Observed intensities in the spectra were smaller for positive bias between CrO_2 and Co than negative bias and correspondingly peak intensities are also smaller for positive bias.

![Figure 6](image-url)
Energies of the vibrational modes in bulk Cr$_2$O$_3$ obtained with infrared spectroscopy are shown in Fig. 6 by the vertical lines. From left to right, the positions of these lines are 303, 419, 444, 532, and 613 cm$^{-1}$, respectively. Considering the peak positions to be correct within ±2.5 mV and comparing with the available data, these peak positions can be assigned to off-stoichiometric Cr$_2$O$_3$ excitations. Also, because of the strong bias and temperature dependence of our IETS spectra, it was not corrected for the elastic tunneling background. As discussed in our transport analysis, we conclude in general that precise characterization of the natural barrier, apart from the electrode material, in CrO$_2$ devices can play a decisive role in determining the magneto-transport properties of tunnel junctions.

IV. SUMMARY

In summary, micro-fabricated (110) CrO$_2$/natural barrier/Co tunnel junctions were investigated for their TMR, I-V characteristics, and inelastic tunneling spectroscopy (IETS). The TMR response and its temperature dependence are similar to previously reported results with (100) CrO$_2$ as an electrode. The barrier height obtained from the I-V characteristics of the junction is approximately 0.40 eV, which is lower than anticipated values. However, the estimated barrier thickness from I-V characteristics matches reasonably well with the thickness of the natural oxide layer on CrO$_2$ film observed in TEM. The IETS spectra had significant bias dependence and the IETS peak positions showed vibrational modes to deviate from stoichiometric Cr$_2$O$_3$. Based on the overall transport characteristics, we conclude that tunneling properties to be determined by defects within the barrier material and not by the high spin-polarized CrO$_2$ or its crystallographic orientation.

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