CrO₂/Ag/YBCO Interface Study with a Flip-Chip Configuration

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Transport across a CrC₂/Ag/YBCO interface was studied using a flip-chip configuration. The results were interpreted in the Andreev reflection scenario. It is shown that the surface spin-polarization of CrO_2 film, even after exposing to air, remained close to 100% to the T_c of $YBa_2Cu_3O_{7-x}$, a temperature limited by this method.

KEY WORDS: CrO_2; half metallic material; tunneling junction; spin-polarization; Andreev bound states; Flip-Chip configuration; Andreev reflection scenario; YBCO

CrO_2 has been predicted to be a half-metal theoretically with an exchange energy of 1 eV [1,2]. It has a Rutile structure and the lattice constants are $a = b = 4.419$ Å, $c = 2.912$ Å. The Curie temperature of CrO_2 is about 391 K [3]. With the breakthrough in the metalor-organic chemical vapor (MOCVD) deposition of thin films on TiO_2 or Al_2O_3 substrates [4,5,7,8], CrO_2 has became a popular candidate for high-temperature spintronic applications. For device application, the spin-polarization at or near the surface is critical. Experiments, such as spin-resolved photo-emission [9], vacuum tunneling [10], and tunneling through different barrier [11–14], have demonstrated close to 100% surface spin-polarization below liquid helium temperature, one of the highest among all the tested materials. Measurements of surface spin-polarization at higher temperature, however, have been absent. Some half metals, for example, $La_{0.7}Sr_{0.3}MnO_3$ (LSMO), a colossal magnetoresistance (CMR) material, has

shown rather rapid degradation of surface spin-polarization [14–17] with increasing temperature. This lowered spin-polarization at higher temperature is evidently undesirable for high temperature applications [18]. Therefore, to measure the surface spin-polarization of CrO_2 is a prerequisite for determining the feasibility of high-temperature spintronic device based on CrO_2.

In our previous study of the temperature dependence of surface spin-polarization of LSMO, the LSMO/$YBa_2Cu_3O_{7-x}$ (YBCO) interface was employed. YBCO is known to be a d-wave superconductor [19,20]. Its superconducting order-parameter changes sign under a 90° rotation. As a direct result of this d-wave symmetry, when tunneling into (110) plane, the normally reflected electron current and Andreev-reflected hole current have a 180° phase difference. These currents undergo constructive interference and create zero energy surface bound states, Andreev-bound states. As a manifestation of the Andreev-bound states, the surface conductivity peaks around zero bias; the zero-bias conductance peak (ZBCP) was observed in tunneling spectrum of YBCO with various metals [21]. In reality, because of the surface facets, such as those exposed by the screw dislocation typical to YBCO thin film, ZBCP can be observed in the tunneling characteristic along other directions as well [22]. Since the ZBCP is a result of d-wave symmetry and Andreev reflection, it has been predicted that the ZBCP will be suppressed

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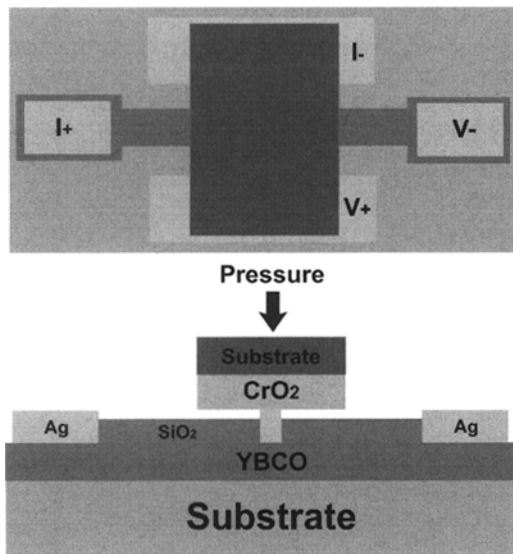
Fig. 1. A schematic of the CrO_2/Ag/YBCO flip-chip testing configuration.The junction resistance was adjust to the minimum by applying pressure before the measurement.

at a ferromagnet/d-wave superconductor interface [23–26], just as the suppression of Andreev reflection at such an interface [27]. The measurement of the ZBCP thus can reveal the surface spin polarization of the ferromagnet material. This method is a modification of the experiment in the literature, including a Nb tip and a ferromagnent interface [11]. While their experiment was limited by the critical temperature T_c of the low-temperature superconductor, our method could measure much higher temperatures due to the high T_c of YBCO, which is about 90 K.

In this letter we apply this method to a CrO_2/YBCO interface to obtain the surface spin-polarization of CrO_2 films at high temperature. Since the CrO_2 and YBCO films cannot be epitaxially grown together, an intrinsic junction geometry such as in our previous study [17] cannot be realized. Instead, a flip-chip scheme is adopted, as shown in the Fig. 1.

A 3000 Å Ag film was thermally evaporated on the surface of a 2000 Å YBCO film. This heterostructure was then patterned and ion-milled in Ar ambient to the YBCO layer, forming a 10 μm × 10 μm Ag island. A 1000 Å SiO_2 was then deposited by Ar plasma sputtering for insulation. The chip was glued on a chip carrier. We then placed a 4000 Å CrO_2, face down, on top of the Ag/YBCO junction and mechanical force was applied to adjust the contact resistance. The contact was formed at the spot of CrO_2 and Ag contact. Two large patches of the original Ag/YBCO

film were left on the substrate during the processing. Contacts to the CrO_2 were made through these two patches. All connections were made by wire bonding. This testing structure was a true CPP (current perpendicular to the plane) configuration. A special dip probe was designed so that mechanical pressure can be applied from outside to the junction even when the probe is in a cryogenic system. Special measures were also taken to prevent sample breakage. Typical force applied to the entire chip was in the order of 10 N. Four probe I–V measurement was taken on the CrO_2/Ag/YBCO junction and the conductance was retrieved by taking the derivative digitally.

The flip-chip configuration is often sensitive to vibration [29]. Relatively soft and malleable Ag can insulate the vibration and reduce the noise. Also it avoids the direct contact between the two films, therefore protects them from being damaged. After pressing, the Ag was usually thinner than 1500 Å. The spin diffusion length in noble metals is in the order of μm [30], thus adding an Ag layer does not reduce the spin polarization of the current reaching the YBCO surface.

The CrO_2 films we used were supplied by Dr A. Gupta's group at IBM's T. J. Watson Research Center. They were fabricated by the two-chamber MOCVD technique. The substrates were (100) orientated TiO_2 single crystal, the film thickness was 4000 Å. The X-ray diffraction spectrum of the film is shown in Fig. 2a. A 0.2° full width at half maximum (FWHM) of the rocking curve around the CrO_2 (200) peak is shown in the inset of Fig. 2a. The X-ray spectrum indicates the (100) orientation and high quality crystallinity of the film. Magnetic hysteresis loops along the magnetic hard axis (010) and the magnetic easy axis (001) were also measured at room temperature by vibrating sample magnetometery (VSM). As shown in the magnetization curve in Fig. 2b, the sample is ferromagnetic at room temperature with a coercivity of 25 Oe along the c-axis, the easy axis of magnetization. The resistivity of the film is about 300 $\mu\Omega$ at room temperature and shows a metallic behavior from room temperature down to 4.2 K. The YBCO film was grown using pulsed laser deposition (PLD). A (001) $SrTiO_3$ substrate was held at 800°C in 150 mTorr O_2 pressure. The laser energy density was kept at ~1.7 J/cm² and a 2000 Å thick film was grown and then cooled down in 400 Torr oxygen. The surface grown in these condition will have enough surface roughness to expose large amount of ab-plane facets [17]. The YBCO film after all the process has a T_c of 90 K.
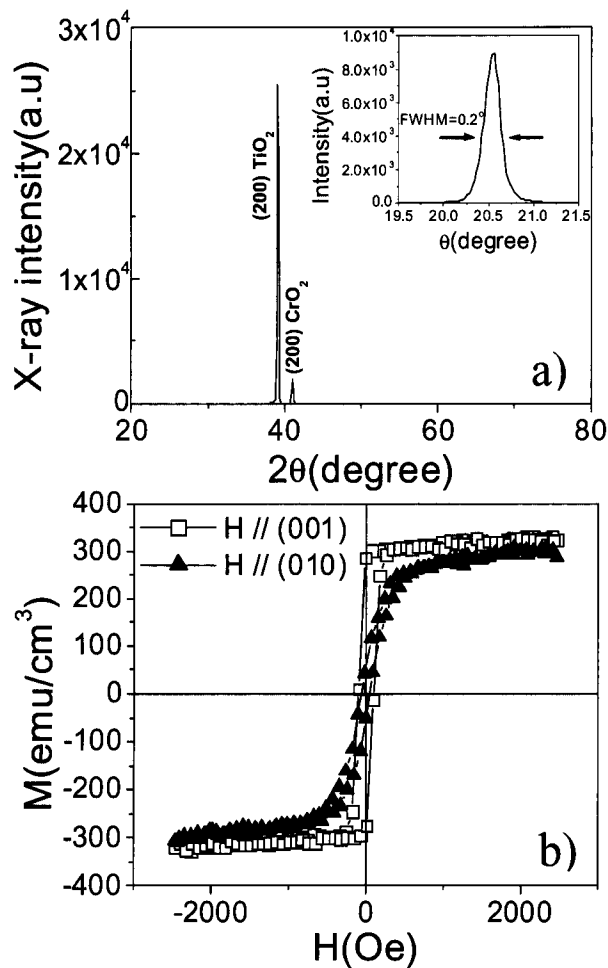
Fig. 2. (a) An X-ray diffraction scan of the CrO₂ film. The inset shows a rocking curve around the CrO₂ (200) peak. (b) The room temperature magnetization data of the CrO₂ film obtained by VSM along the magnetic hard axis (010), and the magnetic easy axis (001).

Fig. 3. (a) The curves of a Ag/YBCO junction at different temperatures. (b) The G–V curves of a CrO₂/Ag/YBCO junction. No ZBCP is present at all the temperatures, indicating that the high spin polarization at the surface of the CrO₂ does not decay rapidly with temperature.

To indicate the validity of the flip-chip method, a 4000 Å Ag film was thermally evaporated on a STO substrate, and then pressed onto the Ag/YBCO junction. By adjusting the pressure, a junction resistance of 0.5 Ω was achieved. Figure 3a shows a set of temperature dependent G–V curves. These features are presumed to arise from the Ag/YBCO interface. ZBCP formations can be seen for temperatures below 35 K. The shape of the G–V curves and the width of the onset voltage of ZBCP are very similar to the previous studies where *in-situ* junctions were used [17,21]. All curves converge at the YBCO superconducting gap value. No shoulder structure was observed. This shoulderless feature is a manifestation of the low-tunneling barrier strength in this junction [31]. The presence of ZBCPs in the Ag/Ag/YBCO
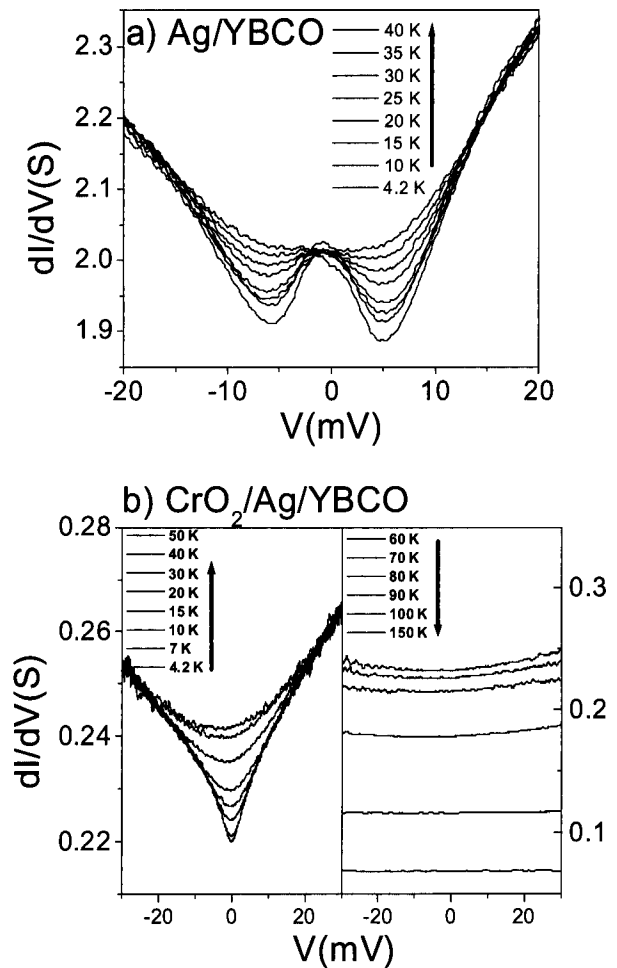
structure proved that the flip-chip configuration is capable of recognizing the fine structures in the transport study.

Then the Ag film was replaced with a 4000 Å orientated CrO₂ film, and the junction resistance was about 5 Ω. It corresponds to an areal resistivity of about 1×10^{-5} Ω-cm². This value is similar to the LSMO/YBCO junction we measured in the previous study [17]. The resultant G–V curves are shown in Fig. 3b. No ZBCP structure was observed in the tested temperatures. For temperatures below 60 K, the G–V curves converge at the YBCO superconducting gap value. This value is slightly larger than those in Fig. 3a. This deviation is due to the inclusion of the contact resistance between Ag and CrO₂. Since it is basically a metal–metal contact we assume

it to be constant over the small voltage range. Compared with the gap value we observed in Fig. 3a, a value of ~ 0.75 Ω was retrieved, corresponding to an areal resistivity of about 1.5×10^{-6} Ω cm^2. The lack of a shoulder structure again indicates a low Z junction. For temperature above 60 K, the YBCO surface starts to lose its superconductivity. An up-shift of the entire curve can be seen. For temperature beyond the bulk T_c, the bound states do not exist anymore, and junction behaves as a metal–metal contact.

As stated earlier, we assume that the electrons from CrO$_2$ maintain much of their spin polarization when they reached the Ag/YBCO interface. In the Andreev bound state scenario, the absence of the ZBCP is an evidence of near 100% spin polarization at the CrO$_2$ surface, just as in the LSMO/YBCO interface. However, unlike in the LSMO/YBCO interface, the ZBCP was not resurgent at higher temperature. The similar theoretical effort as in the previous study on LSMO/YBCO interface [17] did not yield meaningful results. One reason is the surface density of state for CrO$_2$ is not readily available, even in an empirical form. Without this information, a quantitative extraction of the exact spin polarization of CrO$_2$ surface is not possible.

In conclusion, we have studied a CrO$_2$/Ag/YBCO junction with a flip-chip configuration. Analyzed in the Andreev bound scenario, the spin polarization at the surface of CrO$_2$ has been shown to be near 100%. The high spin polarization was maintained up to at least the YBCO critical temperature, 90 K. No decay of this surface spin polarization at high temperature was observed. Indicating little surface insulating layer even though our films were exposed to air for several weeks. In the future work, more vigorous analysis effort will be spent to extract quantitative results.

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