Tunneling Spectroscopy Study of Spin-Polarized Quasiparticle Injection Effects in Cuprate/Manganite Heterostructures

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Scanning tunneling spectroscopy was performed at 4.2K on epitaxial thin-film heterostructures comprising YBa$_2$Cu$_3$O$_{7-\delta}$ and La$_{0.7}$Ca$_{0.3}$MnO$_3$, to study the microscopic effects of spin-polarized quasiparticle injection from the half-metallic ferromagnetic manganite on the high-$T_c$ cuprate superconductor. Quasiparticle tunneling characteristics consistent with $d$-wave pairing symmetry were observed, with a gap-maximum $\Delta_0=22$meV and showing little spectral variation up to 35mA ($7\times10^3$ A/cm$^2$) injection. Spectral smearing was observed at higher injection and could be fitted to an elevated effective quasiparticle temperature ($T_{\text{eff}}=60$K for 60mA injection), eventhough negligible sample heating (<3K at 4.2K) was detected by in-situ thermometry. We discuss general implications of these results for the scenario of dynamic pair-breaking by a non-equilibrium distribution of spin-polarized quasiparticles.
I. INTRODUCTION

Recent experiments have indicated that high-temperature superconductivity in a thin film can be suppressed by the injection of current from a lattice-matched half-metallic ferromagnet [1-4]. The phenomenon has been reported by transport measurements on epitaxial cuprate/insulator/manganite heterostructures, and is believed to be due to dynamic pair-breaking in the cuprate by a non-equilibrium distribution of spin-polarized quasiparticles from the manganite [5]. Vas'ko et al. [1], Chrisey et al. [2] and Dong et al. [3] have shown the superconducting critical-current \( I_C \) in the cuprate to be attenuated by a current \( I_M \) injected from the ferromagnetic manganite, with a much larger \( I_C/I_M \) ratio than in quasiparticle injection devices without spin-polarization. More recently, Yeh et al. [4] have employed a pulsed-current technique to minimize the spurious effects of Joule heating, verifying the \( I_C \)-attenuation near \( T_C \) but also indicating an \( I_C \)-enhancement trend \( (dI_C/dI_M > 0) \) with low injection levels at lower temperatures.

II. EXPERIMENTAL

To examine the physics underlying these effects, we have performed tunneling spectroscopy on similar spin-injection heterostructures, using a low-temperature scanning tunneling microscope (STM). Unlike the macroscopic transport measurements which involve \( \sim \text{mA} \) signals, this experimental approach is inherently microscopic as a local \( \sim \text{nm} \) and non-perturbative \( \sim \text{nA} \) probe of the superconducting order parameter. The measurements were made at 4.2K to maximize the cooling power and minimize the
resistiveness of the LCMO underlayer. Figure 1 is a schematic of the STM setup, showing a Platinum-tip on a piezo-tube driven by a feedback circuit interfaced with a computer which monitors the tunneling current $I_t$ and controls the bias voltage $V_b$ on the sample. A spin-polarized quasiparticle current can be injected into the superconducting YBCO layer by passing a current $I_M$ through the ferromagnetic LCMO layer, as indicated by the arrow. For a given injection current, the sample temperature $T$ can be monitored \textit{in-situ} by calibrating the actual resistivity $\rho$ of the LCMO layer against the $\rho(T)$ profile measured with very low-current and thus negligible Joule heating [4].

The samples measured were YBa$_2$Cu$_3$O$_{7.8}$/yttria-stabilized-zirconia/La$_{0.7}$Ca$_{0.3}$MnO$_3$, (YBCO/YSZ/LCMO) epitaxial heterostructures, 100nm/2nm/100nm in thickness and 6mm x 6mm (YBCO=6mm x 2mm) in size. The heterostructures were grown by pulsed laser deposition on (100) LaAlO$_3$ substrates, with the stepped geometry (see Fig.1) defined by shadow-masking [4]. Chemical composition of the samples was verified by X-ray photoelectron spectroscopy. Surface morphology as seen by optical microscopy indicated predominantly c-axis oriented epitaxy, with some ab-plane outgrowth attributable to the slight YBCO/YSZ lattice-mismatch. Electrical resistivity measurements showed the LCMO layer to have a colossal magnetoresistivity peak near its Curie temperature ($T_M=260\text{K}$) [6] and the YBCO layer to have a sharp ($\sim1\text{K}$) superconducting transition at $T_C=87\text{K}$, values which are comparable to those in bulk materials. Critical current density of the YBCO layer was determined by a pulsed-current technique to be $J_C=5\times10^4\text{ A/cm}^2$, substantially smaller than in YBCO films without the ferromagnetic underlayer [4].
III. RESULTS

Two types of tunneling conductance spectra were observed on the YBCO layer at 4.2K. Figure 2 plots the predominant type, showing a distinct gap-structure with finite zero-bias conductance, asymmetric peaks and linear background. The dashed curve is for 0mA injection and the solid curve is for 35mA injection, showing little spectral variation. The gap-structure can be attributed to c-axis quasiparticle tunneling [7,8] on a superconductor with a d-wave order parameter $\Delta_z = \Delta_0 (\cos k_x - \cos k_y) / 2$ [9], as exemplified by the simulation from Ref. 8 shown in the inset. This suggests the YBCO layer to have a d-wave pairing symmetry with gap-maximum $\Delta_0 = 22$meV up to at least $7 \times 10^3$ A/cm$^2$ injection. Note in the 35mA data, however, an apparent discrepancy between the slight gap-edge smearing and gap-bottom sharpening, the former suggesting pair-suppression but the latter suggesting pair-enhancement. This latter feature is consistent with the observation of slight $I_C$-enhancement by pulsed-current measurements for low-level injection at lower temperatures [4].

Figure 3 shows the other type of tunneling spectra observed, with a pronounced zero-bias conductance peak (ZBCP) flanked by dip and peak structures. The dashed curve is for 0mA injection and the solid curve is for 15mA injection, also showing little spectral variation. The ZBCP feature can be attributed to tunneling in the ab-plane of a d-wave superconductor [7], whose phase sign change about its gap-nodes ($k_x = \pm k_y$) allows the formation of zero-energy Andreev-bound surface states [10,11], as demonstrated in the inset by the spectral simulations from Ref. 11 for various ab-plane junctions. It is worth noting that a split ZBCP would be expected if the time-reversal symmetry of the d-
wave were broken \[12\], such as in a complex-symmetry $d+i\pi$ or $d+id'$ scenario \[13,14\]. Such ZBCP-splitting was not seen in our $ab$-plane tunneling data up to 35mA injection, suggesting the $d$-wave pairing symmetry to be invariant under perturbation by a spin-polarized current up to $7 \times 10^3 \text{ A/cm}^2$.

Figure 4 shows the spectral dependence of the $c$-axis tunneling data on the injection current, plotted in arbitrary units and offset vertically for clarity. Above 40mA ($8 \times 10^3 \text{ A/cm}^2$) injection, the gap-bottom begins to rise and the gap-edges to broaden while the overall spectral features are preserved. The dashed curve is a thermally smeared fit of the 0mA data to the 60mA data, using the temperature in the Fermi-Dirac function as a parameter. This fit provides an estimate of the effective quasiparticle temperature $T_{\text{eff}}=60\text{K}$ for 60mA ($1.2 \times 10^4 \text{ A/cm}^2$) injection, which is $\sim 25\%$ of the maximum critical current.

**IV. DISCUSSION**

Possible physical origins of the effective quasiparticle temperature are: 1) Joule heating of the YBCO layer by the YSZ and LCMO layers; 2) a non-equilibrium excess quasiparticle distribution with a long relaxation time in the YBCO layer. The first possibility could be discounted by our in-situ thermometry, which indicated negligible ($<3\text{K}$) sample heating for 60mA injection at 4.2K. The second possibility seems more plausible, since the quasiparticles injected from the manganite are spin-polarized and cannot readily recombine into spin-singlet Cooper-pairs in the cuprate \[15\]. Detailed verification of this latter scenario would require more quantitative spectral analysis.
Experimentally, samples of different dimensions and insulator materials could be used to further minimize Joule-heating effects in tunneling spectroscopy and to determine the actual spin-relaxation depth in the cuprate [16]. Theoretically, the BCS gap-equation would need to be self-consistently solved, incorporating a non-equilibrium quasiparticle distribution with a $d$-wave pairing potential which is also directly perturbed by the spin-polarized quasiparticles [17-19].

V. CONCLUSION

STM spectroscopy was performed on YBCO/YSZ/LCMO spin-injection heterostructures at 4.2K. Tunneling spectra consistent with $d$-wave pairing symmetry were observed in the YBCO layer. The $d$-wave appears to be robust against a spin-polarized current perturbation up to $7 \times 10^3$ A/cm$^2$, or $\sim 15\%$ of the maximum critical current. Spectral smearing was observed at higher injection, consistent with pair-suppression due to an elevated quasiparticle temperature. This effective temperature does not appear to be thermal in origin, but more likely to reflect a non-equilibrium distribution of spin-polarized quasiparticles with a long relaxation time and acting as dynamic pair-breakers.

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7
FIGURE CAPTIONS

Fig. 1 Schematic of the STM setup, showing a Pt-tip on a piezo-tube driven by a feedback circuit interfaced with a computer which monitors the tunneling current $I_t$ and controls the bias voltage $V_b$ on the sample. A spin-polarized quasiparticle current can be injected into the superconducting YBCO layer via the YSZ barrier (in grey) by passing a current $I_{inj}$ through the ferromagnetic LCMO layer, as indicated by the arrow.

Fig. 2 STM tunneling data taken on an YBCO/YSZ/LCMO heterostructure at 4.2K, showing a distinct gap-structure. The dashed curve is for 0mA injection and the solid curve is for 35mA ($7 \times 10^3$ A/cm$^2$) injection. The inset shows the spectral simulation from Ref. 8 for c-axis quasiparticle tunneling on a d-wave superconductor.

Fig. 3 STM tunneling data taken on YBCO/YSZ/LCMO heterostructure at 4.2K, showing a zero-bias conductance peak structure. The dashed curve is for 0mA injection and the solid curve is for 15mA ($3 \times 10^3$ A/cm$^2$) injection. The inset shows the spectral simulations from Ref. 11 for various ab-plane tunneling on a d-wave superconductor.

Fig. 4 Spectral dependence of the c-axis tunneling data on the injection current, plotted in arbitrary units and offset vertically for clarity. The dashed curve is a thermally smeared fit of the 0mA data to the 60mA ($1.2 \times 10^4$ A/cm$^2$) data, using an effective quasiparticle temperature $T_{eff}=60$K in the Fermi-Dirac function as parameter.
Fig. 1
Wei et al.
Fig. 2
Wei et al.
Fig. 3
Wei et al.
Fig. 4
Wei et al.

STM on YBCO/YSZ/LCMO  $T=4.2\text{K}$