Tunneling spectroscopy study of spin-polarized quasiparticle injection effects in cuprate/manganite heterostructures

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Scanning tunneling spectroscopy was performed at 4.2 K on epitaxial thin-film heterostructures comprising $YBa_2Cu_3O_{7-\delta}$ 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. The quasiparticle tunneling characteristics observed were consistent with *d*-wave pairing symmetry, with a gap-maximum $\Delta_0 \approx 22$ meV, up to at least 35 mA (7×10^3 A/cm^2) injection. Spectral smearing observed at higher injections could be fitted to elevated effective quasiparticle temperatures, even though negligible sample heating was detected by *in situ* thermometry. The overall spectral evolution with the injection current also appears to be nonthermal in character, showing a nonmonotonic change in both the zero-bias tunneling conductance and the area under the conductance spectrum. We discuss general implications of these results for the scenario of dynamic pair breaking by a nonequilibrium distribution of spin-polarized quasiparticles.
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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 nonequilibrium distribution of spin-polarized quasiparticles from the manganite.[5] Vas'ko *et al.*,[1] Chrissey *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 for spin-polarized injection and the absence of this attenuation for unpolarized injection. This latter study also indicated an I_C enhancement trend ($dI_C/dI_M>0$) at low injection levels and lower temperatures for samples with thin insulating barriers.

II. EXPERIMENT

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 ~mA signals, this experimental approach is inherently *microscopic* as a local (~nm) and nonperturbative (~nA) probe of the superconducting order parameter. The measurements were made at 4.2 K to minimize Joule-heating effects related to the resistiveness of the LCMO underlayer.[4] Figure 1 is a schematic of the STM setup, showing a platinum tip on a piezotube 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 *in situ* by calibrating the actual resistivity ρ of the LCMO layer against the $\rho(T)$ profile measured with very low-current and thus negligible Joule heating.[4]

The samples measured were $YBa_2Cu_3O_{7-\delta}$/yttria-stabilized-zirconia/$La_{0.7}Ca_{0.3}MnO_3$, (YBCO/YSZ/LCMO) epitaxial heterostructures, 100 nm/2 nm/100 nm in thickness and 6 mm×6 mm (YBCO=6 mm×2 mm) in size. The heterostructures were grown by pulsed laser deposition on (100) $LaAlO_3$ substrates, with the stepped geometry (see Fig. 1)

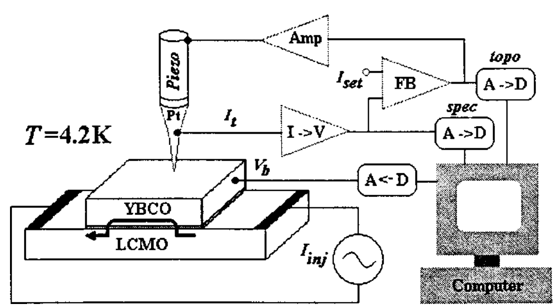

FIG. 1. Schematic of the STM setup, showing a Pt tip on a piezotube driven by a feedback circuit interfaced with a computer which monitors the tunneling current I_t and controls the bias voltage V_b on the sample. A spin-polarized quasiparticle current can be injected into the superconducting YBCO layer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

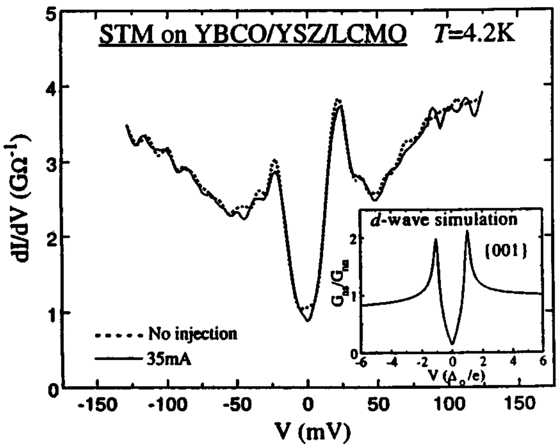

FIG. 2. STM tunneling data taken on an YBCO/YSZ/LCMO heterostructure at 4.2 K, showing a distinct gap structure. The dashed curve is for 0 mA injection and the solid curve is for 35 mA (7×10^3 A/cm^2) injection. The inset shows the spectral simulation from Ref. 8 for *c*-axis quasiparticle tunneling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

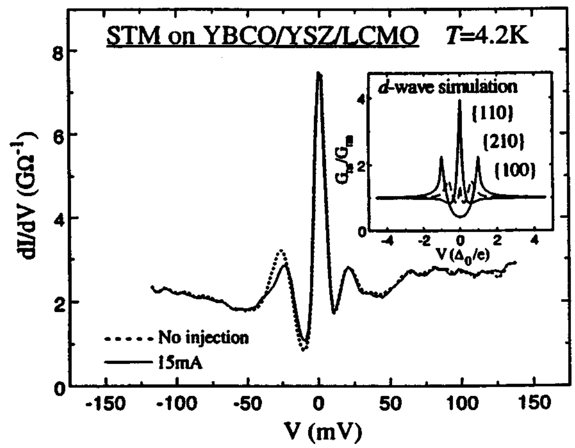

FIG. 3. STM tunneling data taken on an YBCO/YSZ/LCMO heterostructure at 4.2 K, showing a zero-bias conductance peak structure. The dashed curve is for 0 mA injection and the solid curve is for 15 mA (3×10^3 A/cm^2) injection. The inset shows the spectral simulations from Ref. 11 for various *ab*-plane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

defined by shadow masking.[4] Chemical compositions of the samples were 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 ($T_M\approx260$ K)[6] and the YBCO layer to have a sharp (~1 K) superconducting transition at $T_C\approx87$ 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 $J_C\approx5\times10^4$ A/cm^2.[4] This value is substantially smaller than in YBCO films without the ferromagnetic underlayer, and can be understood in terms of self-injection effects associated with a thin insulating barrier.[4]

III. RESULTS

Two types of tunneling conductance dI/dV spectra were observed on the YBCO layer at 4.2 K. Figure 2 plots the predominant type, showing a distinct gap structure with finite zero-bias conductance, asymmetric peaks and a linear spectral background. The dashed curve is for 0 mA injection and the solid curve is for 35 mA injection. The gap-structure can be attributed to *c*-axis quasiparticle tunneling[7,8] on a superconductor with a *d*-wave order parameter $\Delta_k=\Delta_0(\cos k_x-\cos k_y)/2$,[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 $\Delta_0\approx22$ meV, up to at least 7×10^3 A/cm^2 injection. Note in the 35 mA data the co-occurrence of 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 on thin-barrier samples for low-level injection.[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 0 mA injection and the solid curve is for 15 mA injection, 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 junction orientations. 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 $d+is$ or $d+id'$ scenario.[13,14] Such ZBCP splitting was not seen in our *ab*-plane tunneling data up to 35 mA injection, suggesting the *d*-wave pairing symmetry to be invariant under perturbation by a spin-polarized current density of up to 7×10^3 A/cm^2.

Figure 4 shows the spectral dependence of the *c*-axis

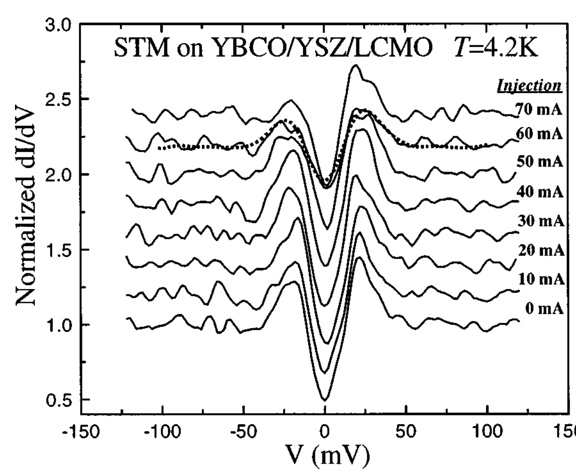

FIG. 4. Spectral dependence of the *c*-axis tunneling data on the injection current, plotted in normalized units after dividing out the linear background and offset vertically for clarity. The dashed curve is a thermally smeared fit of the 0 mA data to the 60 mA data, indicating an elevated *effective* quasiparticle temperature of $T_{\mathrm{eff}}\approx60$ K, even though negligible *thermal* heating was actually detected for this injection level.

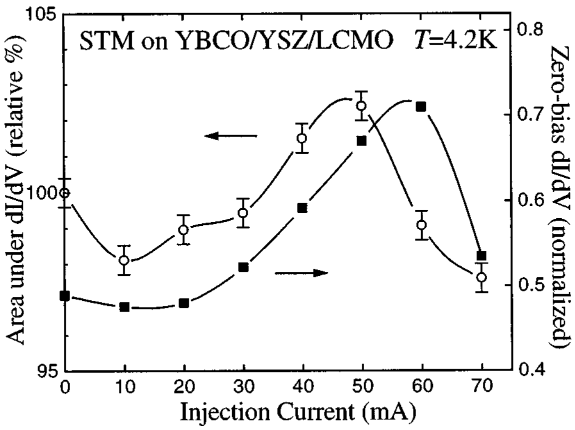

FIG. 5. Quantitative illustration of how the spectra shown in Fig. 4 evolve with the injection current. The area under the dI/dV conductance, plotted as open circles in units of % relative to the 0 mA data, is clearly *not conserved*. The zero-bias conductance values, plotted as solid squares in normalized units, show a similarly *non-monotonic* evolution.

tunneling data on the injection current, plotted in normalized units after dividing out the linear backgrounds and offset vertically for clarity. Note that, while the overall spectral features are preserved, the gap edges show marked broadening with higher injection, particularly above ~30 mA. This spectral broadening does not appear to be thermal in origin, since relatively little heating was detected by our *in situ* thermometry for each injection level. As an example, the dashed curve gives a thermally smeared fit of the 0 mA data to the 60 mA data, using the temperature in the Fermi–Dirac function as a parameter. This fit indicates an elevated *effective* quasiparticle temperature of $T_{\mathrm{eff}} \approx 60$ K for 60 mA (1.2×10^4 A/cm^2) injection, even though negligible *thermal* heating (<5 K) was actually detected for this injection level at 4.2 K.

Figure 5 illustrates more quantitatively how the tunneling spectra shown in Fig. 4 evolve with the injection current. First, the area under the dI/dV conductance spectrum, plotted as open circles in units of % relative to the 0 mA data, is clearly *not conserved*. Second, the zero-bias conductance values, plotted as solid squares in the normalized units, show a similarly *nonmonotonic* dependence on the injection current. Both spectral evolutions appear nonthermal in character, since Joule heating would be expected to monotonically increase the mid-gap density of states, which is proportional to the zero-bias dI/dV, and to simply redistribute the quasiparticle spectrum without changing the total number of states, which can be quantified as the spectral area under dI/dV.

IV. DISCUSSION

A possible physical origin of the effective quasiparticle temperature is the presence of a nonequilibrium excess quasiparticle distribution with a long relaxation time in the YBCO layer. This is a plausible scenario, since the quasiparticles injected from the manganite are *spin polarized* and cannot readily recombine into *spin-singlet* Copper pairs in the cuprate.[15] Detailed verification of this latter scenario would require a more quantitative spectral analysis. Experimentally, samples of different geometries and thicknesses could be measured by tunneling spectroscopy to determine the actual spin-diffusion depth[16] in the superconducting cuprate. Theoretically, the BCS gap-equation would need to be self-consistently solved,[17] incorporating a nonequilibrium quasiparticle distribution with a *d*-wave pairing potential which is also directly perturbed by exchange interaction with the spin-polarized quasiparticles.[18] An *s*-wave formulation of such a theory has indicated the possibility for spatial texturing due to topological phase-segregation of the nonequilibrium quasiparticles.[19] One feature of this model is the formation of quasiparticle bags associated with the spin-polarized injection, giving rise to subgap states which add to the total density-of-states spectrum. Another aspect of the model is the critical concentration of spin-polarized quasiparticles needed to form the topological bags, suggesting a threshold-like spectral evolution. Both tendencies would offer consistent explanations for the highly nontrivial spectral behavior seen in our tunneling data.

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