Journal of Superconductivity: Incorporating Novel Magnetism, Vol. 15, No. 1, February 2002 (© 2002)

Spin-Injection Quasiparticle Nonequilibrium in Cuprate/Manganite Heterostructures

John Y. T. Wei[1]

Received 1 November 2001

Quasiparticle nonequilibrium due to spin-polarized current injection in perovskite superconductor/ferromagnet (S/F) thin-film heterostructures has been studied with the technique of cryogenic scanning tunneling spectroscopy. The spin-injection heterostructures consisted of epitaxial bilayers of the high-T_c superconductor $YBa_2Cu_3O_{7-\delta}$ and the half-metallic ferromagnet $La_{0.7}Ca_{0.3}MnO_3$, with a spin-polarized quasiparticle current injected into the cuprate layer from the manganite layer. The tunneling conductance measured at 4.2 K on the cuprate layer showed a distinctly nonequilibrium quasiparticle spectrum as a result of the spin injection. Quantitative analysis of the tunneling spectral evolution versus the injection current yielded an estimate of the quasiparticle spin-diffusion depth and the spin-relaxation time in the superconducting cuprate.

KEY WORDS: spin injection; high T_c; cuprate; manganite; heteroepitaxial; thin films; STM; tunneling; quasiparticle; spin relaxation.

High-T_c superconductivity can be suppressed by the injection of a spin-polarized current [1]. This phenomenon has been observed in epitaxial cuprate/manganite thin-film heterostructures, and is believed to be due to magnetic pair breaking in the superconducting cuprate by spin-polarized quasiparticles injected from the ferromagnetic manganite [2]. Magnetic pair breaking is an important subject in the field of superconductivity [3]. Quasiparticle (QP) injection effects in superconducting thin-films have also been extensively studied, particularly in relations to nonequilibrium superconductivity [4]. The combination of these mechanisms holds great potential in device applications, involving the rapid tuning of superconductivity by a small current [5]. These device possibilities include superconducting spin switches, spin transistors, and spin-dependent logic elements [6]. The realization of these high-speed and low-noise device concepts may be most promising in high-T_c superconductors [7], by virtue of their large gap anisotropy, low carrier density, and strong electron correlations, factors which are conducive to quasiparticle injection, nonequilibrium, and pair breaking [8].

In order to understand this spin-injection phenomenon for device applications, we have carried out a tunneling study of cuprate/manganite heteroepitaxial films. These thin-film samples, consisting of superconducting $YBa_2Cu_3O_{7-\delta}$ (YBCO) and ferromagnetic $La_{0.7}Ca_{0.3}MnO_3$ (LCMO) layers, each 100 nm thick and separated by a 2 nm monolayer of yttria-stabilized zirconia (YSZ), were grown by pulsed laser-ablated deposition on $SrTiO_3$ substrates [9]. Magnetic and electrical transport measurements indicated a critical temperature $T_c \approx 87$ K in the cuprate, and a Curie temperature $T_m \approx 260$ K in the manganite layer. Scanning tunneling spectroscopy (STS) was then used to measure the QP density-of-states spectrum in the cuprate at 4.2 K, well below both T_c and T_m [10].

The scenario of QP spin injection into a cuprate superconductor is portrayed in Fig. 1. On the left side, itinerant electrons in the ferromagnetic (F) manganite layer are represented by the filled circles, with the parallel-aligned arrows indicating their near-100% spin polarization at the Fermi level (horizontal line). The right side shows the QP spectrum for the superconducting (S) cuprate with a predominant d-wave

[1]Department of Physics, University of Toronto, 60 St. George Street, Toronto, Ontario, Canada M5S1A7.

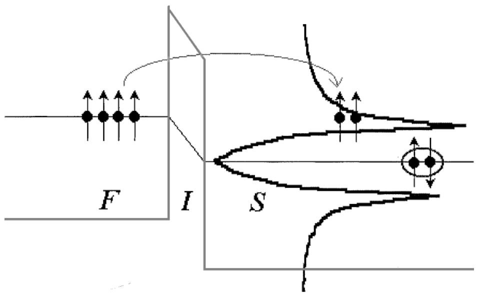

Fig. 1. Schematic of spin-polarized quasiparticle (QP) injection from a half-metallic ferromagnes (F) into a d-wave superconductor (S) via an insulating (I) barrier. The itinerant electrons in F are represented by the filled circles, with their spin-polarization at the Fermi level indicated by parallel arrows. In order for the injected QPs in S to recombine into singlet d-wave pairs (antiparallel arrows within the oval), they must first randomize their spin orientation, thus making spin relaxation the limiting factor for QP equilibrium. These "spin-bottlenecked" QPs could then persist into the bulk of S to cause pair-breaking effects, which are directly observable by tunneling spectroscopy.

pairing symmetry [11], where the filled circles represent injected QPs and the open oval represents a d-wave Cooper pair. The insulating (I) YSZ monolayer is represented by the trapezoidal injection barrier, indicating a bias voltage applied between the F and S layers. Note that in order for QPs injected from F to S to recombine into the spin-singlet pairs, the QPs must first randomize their spins, making spin relaxation the limiting factor for QP equilibrium. This spin relaxation is expected to be considerably slower than charge relaxation [12], effectively "spin bottlenecking" the QP recombination process. Therefore, one would expect this spin-polarized nonequilibrium QP distribution to persist into the bulk of the S layer and cause pair breaking via two mechanisms: (1) raising the effective, nonequilibrium QP temperature, thus weakening the pairing interaction and (2) breaking the time-reversal symmetry of the d-wave pairs, through itinerant exchange interactions similar to the well-known spin-flip scattering involving local moments [13]. Either mechanism could introduce spectral smearing and reduce the effective pair potential for QP tunneling.

Results of the STS measurements are summarized in Fig. 2. The left inset shows the experimental setup, with the YBCO thin film under QP spin injection from the LCMO sublayer and probed by STS with a piezo-driven Pt tip [10]. The main plot displays the normalized tunneling conductance versus bias voltage (dI_t/dV_b vs. V_b) data under various

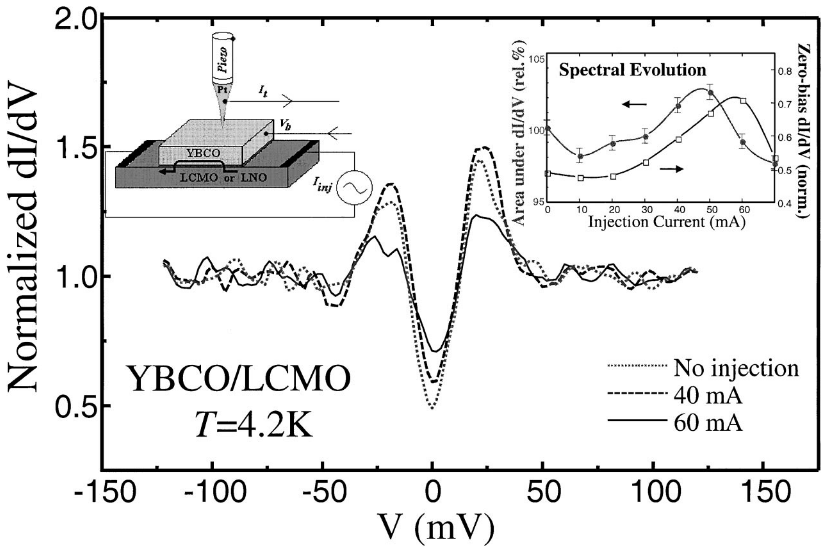

Fig. 2. Summary of the scanning tunneling spectroscopy (STS) measurements at 4.2 K. The left inset shows the experimental setup, with the YBCO thin film under QP spin injection from the LCMO sublayer and probed by STS with a piezo-driven Pt tip. The main plot displays the normalized tunneling conductance versus bias voltage (dI/dV vs. V) data under various injection currents (I_{inj}). These spectra can be identified with the d-wave QP density-of-states. The right inset shows how the tunneling spectra evolve with the injection, indicating (1) nonconservation of the spectral area under dI/dV, and (2) nonmonotonic variation of the zero-bias dI/dV. This spectral evolution is clearly nonthermal in character, and attributable to itinerant magnetic pair breaking by the spin-polarized QPs.

injection currents (I_{inj}). These spectra can be identified as the QP spectrum [11] associated with the predominant d-wave pairing symmetry [14], and analyzed with well-established theoretical formalism [15]. Details of how the QP spectrum evolves with the injection are displayed in the right inset, showing (1) nonconservation of the spectral area under dI/dV and (2) nonmonotonic variation of the zero-bias dI/dV. The spectral evolution is clearly nonthermal in character, and attributable to itinerant magnetic pair breaking by the injected spin-polarized QPs. In specific, the creation of QP bound states by the pair-breaking perturbation [16], in competition with QP relaxation from nonequilibrium, which involves both excess spins and charges [17], could qualitatively explain both the spectral nonconservation and nonmonotonicity. Alternatively, spin-charge separation in the fundamental QP excitations could also account for this nontrival spectral evolution [18,19]. For comparison, a controlled sample with the paramagnetic perovskite $LaNiO_3$ (LNO) as the sublayer was also made and measured. In contrast to the YBCO/LCMO sample, the YBCO/LNO sample showed no disturbance in the QP spectrum under the injection current, consistent with it being spin neutral and thus carrying no itinerant moment needed for magnetic pair breaking [20].

Quantitative information about the QP spin relaxation could also be determined from the tunneling data. First, assuming that the injected QPs must spin relax before they could recombine into singlet pairs, we could model the QP recombination by the rate equation $\partial_t n = D\partial_x^2 n - n/\tau_s$, where n is the excess QP density, τ_s is the QP spin-relaxation lifetime, and D is the spin-diffusion constant. Then, by taking the steady-state solution, where λ_s is the spin-diffusion depth, x is the distance from the cuprate/manganite interface, and n_0 is the excess QP concentration at $x = 0$, we could determine both λ_s and τ_s. From the n/n_0 data taken at $x = 100$ nm, we estimate that $\lambda_s \approx 20$ nm along the c-axis for YBCO. And, using the approximation $D \approx v_f l$, with the c-axis Fermi velocity $v_f \approx 10^5$ cm/s and the mean-free path $l \approx 1$ nm, we estimate that $\tau_s \approx 40$ ns. These characteristic length and time scales for QP spin relaxation are sizable as compared with those reported for QP charge relaxation in the cuprates [5], and therefore have positive implications for using spin-injection mechanisms in high-T_c devices.

In summary, our tunneling spectroscopy measurements on cuprate/manganite spin-injection devices clearly revealed a nonequilibrium QP distribution with a spin-enhanced relaxation time. These results provide strong evidence for dynamic pair breaking by the spin-polarized QP injection. Further study is needed to determine whether the itinerant spins break the time-reversal symmetry of the Cooper pairs, as one would expect in the magnetic pair breaking scenario which introduces finite pair-lifetime via exchange scattering: $\Delta \rightarrow \Delta + i\Gamma$, where Δ is the order parameter and Γ represents the exchange energy. In the context of d-wave pairing, this would imply the possibility of electronically tuning the pairing symmetry, e.g. $d \rightarrow d + is$, providing a novel design concept for high-T_c spin-injection devices.

ACKNOWLEDGMENTS

This work was done in collaboration with N. C. Yeh at the California Institute of Technology and R. P. Vasquez at the Jet Propulsion Laboratory in the United States. The author gratefully acknowledges funding support from NSERC, CFI, OIT, CIAR, and the Connaught Fund in Canada.

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