Order parameter oscillations in Fe/Ag/Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ tunnel junctions

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We have performed temperature dependent tunneling conductance spectroscopy on Fe/Ag/Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (BSCCO) planar junctions. The multilayered Fe counterelectrode was designed to probe the proximity region of the ab-plane of BSCCO. The spectra manifested a coherent oscillatory behavior with magnitude and sign dependent on the energy, decaying with increasing distance from the junction barrier, in conjunction with the theoretical predictions involving d-wave superconductors coupled with ferromagnets. The conductance oscillates in antiphase at $E = 0$ and $E = \pm \Delta$. Spectral features characteristic to a broken time-reversal pairing symmetry are detected and they do not depend on the geometrical characteristics of the ferromagnetic film.

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In recent years numerous theoretical and experimental studies have been devoted to the spin polarized transport in ferromagnet(FM)-superconductor(SC) structures. This interest is equally motivated by the rapidly developing applications, e.g. in quantum two-level systems (qubits) based on phase shifts [1], as well as by the very interesting physics involving unconventional superconducting states. The exchange energy $\hbar$ in the ferromagnet leads to a spatially inhomogeneous state with the order parameter presenting phase changes by $\pi$ in the proximity region at the interface with the SC. Such a modulated state is called Larkin-Ovchinnikov-Fulde-Ferrell (LOFF) from the names of the authors who first predicted it [2]. In the FM, the Cooper paired electrons have different energies and Fermi momenta, so they dephase on a scale of several ferromagnetic coherence lengths. Consequently, this region presents a diversity of intensely explored phenomena like spatially dumped oscillations of the density of states (DOS) and gapless superconductivity [3]-[6], oscillations of the density of states (DOS) and gapless superconductivity. The conductance oscillates in antiphase at $E = 0$ and $E = \pm \Delta$. Spectral features characteristic to a broken time-reversal pairing symmetry are detected and they do not depend on the geometrical characteristics of the ferromagnetic film.

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An extensive attention was dedicated to the LOFF state in the cuprates, while intrinsically interesting, may also offer information about what makes the high-Tc superconductivity different from the conventional one. A multitude of theoretical models were developed to calculate the specific transport properties, spin and charge relaxation in tunneling experiments involving d-wave HTSC with FM counterelectrodes. The most popular ones are those based on the Blonder-Tinkham-Klapwijk (BTK) approach [13]. However, these models consider just the transmission and reflection processes at the interface with the FM film modeled as a semi-infinite domain. In this paper we will use a model [5] based on a ballistic quasiclassical formalism with the DOS averaged over all classical trajectories of multiple reflections in the FM domain of finite thickness $\delta$.

Hoping to contribute to the experimental information about this topic, we report in this paper the transport properties in a multilayered-Fe/Ag/Bi$_2$Sr$_2$CaCu$_2$O$_{8-\delta}$ (BSCCO) ab-plane oriented planar tunneling junction. The BSCCO ab-plane is probed successively from 5 overlapping Fe thin films with the same thickness, through an intermediate thin layer of Ag designed to minimize the barrier strength and avoid the appearance of a spin-glass phase at the junction interface [15]. The spectra are collected on the thicker and thicker counterelectrodes and studied as function of the temperature and thickness of the FM domain. The planar junctions were built on freshly cleaved monocrystal samples of slightly underdoped BSCCO grown by the self-flux method. Two silver paste leads were attached to the crystal and its lower side was molded in epoxy resin. Before the epoxy dried, the upper side was fixed between two quartz glass slabs with coplanar surfaces. The epoxy penetrated between the sides of the slabs so that the crystal was caught between two smooth glass surfaces attached by
thin epoxy walls. While the surface of the glass was protected, we performed a clean mechanical polishing along the ab-plane of the crystal, laterally embedded in epoxy. We monitored the process with a profilometer, until the exposed edge of the crystal was at the same level as the glass surfaces. The counterelectrode sandwich was then evaporated using a metallic shadow mask shaped as in Fig. 1, forming thin film strips 0.1 mm in width. The deposition was controlled using a calibrated quartz thickness-monitor. Above a 30Å silver film, five 30Å iron films were deposited at a rate of 0.1 Å/s for uniformity. The residual resistivity ratio of the Fe film $R_{RRR} = R(300K)/R(40K) \approx 8.3$ indicates a good deposition quality. The tunneling barrier is formed naturally on the exposed edge of BSCCO. It had a resistance $R_B(300K) \approx 450\Omega$, for the typical junction presented here, showing a high transparency. We evaluated the elastic mean free path $l$ of conduction electrons in one of our Fe films using an expression derived from Pippard relations: $v_F l = (\pi k_B/e)^2 (\sigma/\gamma)$. With the iron film conductance $\sigma \approx 4 \times 10^3$ S/m$^2$, electronic specific heat $\gamma = 5 \times 10^{-3}$ J/K$^2$, and Fermi velocity $v_F = 1.98 \times 10^6$ m/s, we obtained $l \approx 100\AA$. Since the quasiclassical description of conductance assumes $l < d$, we see that such a model is applicable to our first three Fe layers. We performed conventional 4-point measurements, with a constant current driving bias.

The tunneling spectra at the interface with the BSCCO crystal were obtained successively for the six layers, starting with the Ag counterelectrode, then probing an increasing thickness in the FM region from 0 to 150Å, by 30Å increment for each electrode. Each layer was measured at different temperatures, from 150 to 4.2 K. Fig. 2 represents the temperature dependence for the first three layers, each curve being normalized by the spectrum at 150K. All layers manifested a sudden change in the background conductance at $T_c$. This phenomenon can be attributed to the competition between a screening spin-accumulation close to the interface, responsible for a drop in the barrier conductance, and the onset of Andreev reflections, with an adverse effect. In an Andreev reflection, the electron incident on the barrier with energy lower than the superconductive gap is reflected as a hole, while a Cooper pair is transmitted into the SC. In the case of d-wave superconductors the Andreev reflections are phase sensitive. This leads to the formation of bound states at the junction interface with amplitudes dependent on the angle of incidence. In a quasiclassical picture, the particles move along trajectories reflected between the boundaries of the counterelectrode, the density of states per trajectory being correlated to the length of the trajectory or number of reflections which in turn depends on the energy and exchange potential. The tunneling conductance spectrum is a measure of the energy resolved total density of states and is regarded as a sum of local bound states averaged over all trajectories distributed by length. Therefore, the thickness of the counterelectrode influences the amplitude of the bound states and thus the spectral features, like the ZBCP. For our Ag terminal, since the amplitude of the Andreev states is low (Fig. 2 and 3), and it is in direct contact with the next iron layer, the background conductance at $T_c$ decreases with decreasing temperature, due to the predominance.
of a spin accumulation at the junction barrier. The temperature dependence of the Ag/BSCCO tunneling spectrum shows no ZBCP (Fig. 2a). This may indicate a predominant injection normal on the [100] plane combined with a rarefied distribution of long trajectories due to the very low thickness of the film, leading to the suppression of low energy states. The energy resolved DOS shows signs of a minigap (Fig. 3) at zero energy due to the very low thickness of the film, leading to the suppression of low energy states. The energy resolved DOS was averaged over a Gaussian distribution of thicknesses around each \( d \). The best fits imposed a value \( h = 20 \) meV for the first iron layer and \( h = 170 \) meV for the second one (consistent with the accepted value \( 16 \)). The fits for all other Fe layers were obtained by keeping constant the exchange energy \( h = 170 \) meV, the gap \( \Delta = 44 \) meV, and the barrier transparency \( Z = 0.51 \). The only adjusted variable was the thickness scaled by the coherence length, \( d/\pi\xi \). The procedure resulted into five values for the layer thickness remarkably close to the actual ones. c) \( T = 60 \) K. The ZBCP for \( d = 150 \) Å is not split at this higher temperature approaching \( T_c \), due to the suppression of the BTRS and the bigger effect from the spin-splitting field on the superconductivity since \( \Delta \) decreases while the \( h \) remains constant.

Next we probed the FM region of the junction. With each Fe layer, 30 Å are added to the total distance accessible to the pairs injected into the counterelectrode. The pairs survive along a distance of several coherence lengths \( \xi \approx 38 \) Å as the SC order parameter decays oscillatory around zero. We performed theoretical fits for the DOS spectra at the intermediate temperature 40 K. The model we employed calculates the DOS taking into account the thickness \( d \) of the FM film when the interface with the SC is highly transparent and \( d \) is smaller than the electron mean free path \( l \). With \( l \) calculated above, the model is applicable mostly for the first few layers, which are anyway the most suggestive. Since we expect our boundaries to be rough, leading to an admixture of trajectories and a smearing of the spectral features, the DOS was averaged over a Gaussian distribution of thicknesses around each \( d \). The best fits imposed a value \( h = 20 \) meV for the first iron layer and \( h = 170 \) meV for the second one (consistent with the accepted value \( 16 \)). The fits for all other Fe layers were obtained by keeping constant the exchange energy \( h = 170 \) meV, the gap \( \Delta = 44 \) meV, and the barrier transparency \( Z = 0.51 \). The only adjusted variable was the thickness scaled by the coherence length, \( d/\pi\xi \). The procedure resulted into five values for the layer thickness remarkably close to the actual values (see Fig. 2b), confirming the validity of the model and also the regularity of the thin films. We also take advantage from the BTK type models to underline the effect of the d-wave (possibly mixed with s-wave) pairing symmetry of BSCCO.

The temperature dependent spectra measured on the first FM layer \( (d = 30 \) Å \( ) \) also has a pseudogap. The \( T_c \) is slightly smaller and the background conductance after the transition increases, indicating the growing influence of the Andreev reflections. The most remarkable features (Fig. 3), are the dips replacing the \( \pm\Delta \) coherence peaks. Due to the direct contact with the Ag layer, the spectrum rather shapes up the effect of a distribution of spin-polarized quasiparticles in the Ag film, so that the theoretical fit on this curve imposed an exchange energy \( h = 20 \) meV, lower than that for the other Fe layers. For \( 0 < h < \Delta \), the \( \pm\Delta \) peaks move to subgap energies, being replaced by minima in the DOS. The resulted subgap peaks are expected to merge into a ZBEP with increasing \( d \), and to exhibit local oscillatory variations. Indeed, the next FM layers show the oscillations, but the peaks do not merge with increasing \( d \). This may be due to properties of the junction unaffected by the thickness of the FM environment, like the presence of a mixed su-
perpendicuor phase with broken time-reversal symmetry (BTRS) at the junction interface (e.g., (d+is)-wave)\[11\]. This splits the ZBCP, forming a gaplike feature with the size given by the magnitude of the sub-component s, and influenced by temperature. In this case, the s-wave gap remains unchanged with increasing d, while the subgap DOS oscillates up and down as the decaying order parameter alternatively takes negative and positive values respectively.

As seen in Fig. 2, the next Fe layer (d = 60Å) measures a flatter pseudogap at high temperatures. The conductance enhancement at the superconductive transition is larger, consistent with the high amplitude of the Andreev peaks. The electronlike and holelike quasiparticle branches of the spectrum are asymmetric at lower temperatures. The split ZBCP is high and it becomes broader and less asymmetric with increasing temperature (Fig. 3a,c). This asymmetry may be attributed to the presence of a spin dependent contribution to the ZBCP split at lower temperatures. If a spin imbalance is present in the barrier, the ZBCP corresponding to up (down) spin currents are shifted at higher (lower) energies. The polarization in the FM layer decreases the spin-up peak and increases the spin-down peak. The ratio of the peaks can be used to evaluate the spin polarization P in iron. In our case, \( P_F \approx 43\%\), which is consistent with the values reported in literature [10]. The broader ZBCP at temperatures closer to \( T_c \) may be the effect of a thermal smearing and the suppression of superconductivity due to the diminishing \( \Delta \).

The next three layers (d = 90-150Å) complete an oscillatory cycle. The curves are too noisy to discern the more delicate features but the general behavior is sufficiently clear: the superconductivity is highly suppressed and the LOFF states develop \( d \)-dependent coherent oscillations, with smaller and smaller amplitude. The sign of these oscillations depend on the energy. In Fig. 4 the reduced DOS, \( G(E)/G_0 - 1 \), oscillates in antiphase at energy \( E = 0 \) with respect to the \( E = \pm \Delta \) values. At lower temperatures the zero energy conductance is lower than the theoretically predicted curve (Fig. 4a), due to the significant presence of the BTRS induced ZBCP split. The BTRS states disappear at a temperature lower than BSCCO \( T_c \), so that the 150Å spectrum presents a 60K ZBCP without the split. Consequently, the points taken at temperatures nearer to \( T_c \) approach better the theoretical expectation.

In summary, we constructed planar Fe/Ag/BSCCO junctions, with the Fe region imparted in layers of equal thickness. Temperature dependent tunneling spectra were collected on each layer, probing a reproducible spatial dependent instance of the LOFF state. The spectral features are affected by the pair-breaking exchange field in the FM region, the thickness of this area and the unconventional pairing symmetry in the BSCCO. As the Fe thickness is increased, the conductance spectra show decaying oscillations with energy dependent amplitude and sign. The ZBCP is split regardless the thickness, but dependent on the temperature, due to the presence of a subcomponent added to the dominant d-wave component at the junction interface to form a BTRS state. The spectrum is asymmetric at low temperature due to the spin-polarization.

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