Anomalous superconductivity of $Pb/La_{0.7}Sr_{0.3}MnO_3$ point contacts

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Recent experimental results indicate the possible realization of an even frequency *p*-wave triplet superconducting state in proximity affected $La_{0.65}Ca_{0.35}MnO_3-X$ (X=Pb, MgB_2) point contacts (PCs). Motivated by this we present a study of the current-voltage characteristics and of the dynamic conductance of PCs between Pb and another member of a ferromagnetic manganite family $La_{0.7}Sr_{0.3}MnO_3$ (LSMO). For proximity affected contacts we have observed a spectacular drop of the contact's resistance with the onset of the Pb superconductivity and, for small voltage, an excess current and doubling of the normal-state conductance. We also detected the subharmonic gap resonances and found that proximity induced superconducting state of LSMO corresponds to that with the energy of the quasiparticle gap much larger than that of Pb. The results most reasonably can be explained by assuming a conversion of spin singlet pairs into triplet pairs at the Pb/LSMO interface and intrinsic superconductivity of LSMO with the actual gap independent on Pb. The mechanism of a conversion has been discussed. Systematic character and repeatability of a number of principal experimental facts suggest that some general physical phenomena have been documented in transport properties of proximity affected singlet superconductor/half-metallic manganite heterostructures.

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I. INTRODUCTION

At energies below the superconducting gap, a charge transport through a normal nonmagnetic (N) metal being in contact with a superconductor (S) is possible only due to a specific process. That is the so-called Andreev reflection (AR) (Ref. 1): a two-particle process in which, in the N metal, an incident electron above the Fermi energy E_F and an electron below E_F with an opposite spins are coupled together and transferred across the interface in the S side forming a Cooper pair in the condensate. Simultaneously, an evanescent hole with opposite momentum and spin appears in the N metal. The charge doubling at the interface enhances the subgap conductance[2] and this phenomenon has indeed been observed in the case of a perfectly transparent interface. The picture is significantly modified when spin comes into play. If the N metal is a ferromagnet, there is an imbalance between spin-up and spin-down populations, which suppresses the AR and reduces the subgap conductance below the normal-state value.

An alternative but equivalent way of thinking about AR processes is through a superconducting proximity effect (PE).[3] The electron and the hole involved in the AR are coherently coupled. The phase-coherent electron-hole conversion results in a nonzero pair amplitude in the N metal. Theory predicts that in the extreme case of a completely spin polarized metal being in contact with a singlet *s*-wave pairing superconductor (sS) the PE (and the AR) is absent.[4] Therefore one might expect that the influence of the superconducting PE on transport properties of an sS/half-metallic ferromagnet (hmF) heterostructures should be negligibly small. However, an unconventional mutual influence of superconductivity and ferromagnetism in hybrid sS/hmF structures was observed recently by a few groups[5,6] (one can find references on earlier publications in reviews[3]). Simultaneously, several models were put forward suggesting unconventionally long-range proximity effects.[7–9] Despite the existing experimental evidences supporting unconventional PE in sS/hmF structures and efforts of theorists understanding of this phenomenon still remains unclear and needs further exploration.

There is a general consensus now that the AR, being the central mechanism for the superconducting PE at S/ferromagnet (F) interface, strongly depends on the degree of magnetic homogeneity. Theories[7–9] predict the appearance of the long-range PE if there is a spatial variation of the magnetization in the F. Such inhomogeneities, hence the effect, may in principle be artificially generated in ferromagnets. But existing technology cannot create them in a controlled way at the S/F interface with nanoscale precision so that the most realistic scenario is to use ferromagnet with intrinsic magnetic inhomogeneity. This was, for example, realized by Sosnin *et al.*[6] The authors, using a rare-earth metal with helical magnetic structure as the F, obtained strong experimental evidence for unconventional pairing.

Recently, we addressed the problem of interplay between a spinless Cooper pair current and a spin polarized quasiparticle current in sS/hmF structures.[10,11] Measurements of the subgap current-voltage characteristics and of the dynamic conductance have been performed for $La_{0.65}Ca_{0.35}MnO_3$-X (X=Pb, MgB_2) point contacts (PCs). $La_{0.65}Ca_{0.35}MnO_3$ (LCMO) is a typical mixed-valence manganite and theoretically predicted to be a half-metallic ferromagnet. In our opinion, specific origin of magnetic and transport properties of these colossal magnetoresistive materials, the so-called a double-exchange interaction, makes them promising for exploration of long-range PE. Indeed, within the double-exchange interaction model[12] the itinerant charge carriers (electrons or holes) provide *both* the magnetic interaction between nearest Mn^{3+}-Mn^{4+} ions and the electrical conductivity. Due to the short mean free path (that is typically the distance of about a lattice parameter), the charge carrier probes the magnetization on a very short length scale. As a result, for these systems a strong interplay between local magnetic order and electric properties exists. It is also

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well established now that the existence at the surface of broken exchange bonds, local structure disorder, defects, etc., generates randomness in the double-exchange interactions. Thus, in manganites a surface is intrinsically magnetic inhomogeneous and materials can be used for detection of an unconventional PE. Our motivation was that if the conversion from singlet to triplet pairing exists in sS/hmF heterostructures, the sS/LCMO PCs have to reveal features (such as excess current and doubling of a normal-state conductance) that are typical for sS/N contacts. Indeed, for some of the contacts the measured AR spectrum demonstrates very distinct characteristics, which we interpret as the manifestation of a triplet *p*-wave even frequency pairing.[10,11] The most unexpected features were observation of large induced energy gap of LCMO (much larger than that of Pb or MgB_2) and the so-called subharmonic gap structure (SGS) of the dynamic conductance. By processing the data we get to the conclusion that this combination of observation is a manifestation of an *intrinsic superconductivity* of LCMO; i.e., at low temperature, the LCMO is thermodynamically very close to a triplet *p*-wave superconducting state, which can be fixed by an external superconducting tip.[11]

Note that the compound $La_{0.65}Ca_{0.35}MnO_3$ is a representative of the perovskites manganite family $La_{1-x}A_xMnO_3$ (where A is alkaline-earth doping ions; A=Ca, Sr, Ba, ...), the so-called colossal magnetoresistive manganites with common electronic and magnetic properties.[12] If the phenomenon of a conversion from singlet to triplet pairing is indeed realized in sS/LCMO structures and, at low temperature, the LCMO is thermodynamically very close to a triplet superconducting state, it is reasonable to assume that the peculiarities, which have been observed for sS/LCMO PCs, have to be detected for other members of the manganites family, too. These have not yet been demonstrated in experiment until now. It was our motivation to investigate the current-voltage characteristics (*I-V*) and the dynamic conductance $dI/dV=G(V)$ of superconducting microconstrictions between Pb tips and plates of another representative member of a ferromagnetic manganite family $La_{0.7}Sr_{0.3}MnO_3$ (LSMO). For some of Pb/LSMO PCs very specific properties, which cannot be explained by any simple theory of PE, have been observed. Namely, with decreasing temperature, a spectacular drop of the contact's resistance has been detected with an onset of the Pb superconductivity. For small voltages, an excess current and doubling of the normal-state conductance have been found; the subharmonic gap resonances are also observed. It was documented that the character of $G(V)$ vs voltage dependence corresponds to that for the induced superconducting energy gap of LSMO much larger than that of Pb. We explain the observations as due to the long-range penetration of an unusual triplet component of the order parameter that is generated at the sS/hmF interface and is maintained by the intrinsic superconducting fluctuations of the hmF; i.e., as for LCMO, we argue that, at low temperature, the LSMO is thermodynamically very close to a superconducting state with *triplet p*-wave even frequency pairing.

This report may be considered as the next step in our efforts[10,11] to understand the phenomenon of mutual influence of singlet *s*-wave superconducting pairing and non-Fermi half-metallic ferromagnetic state of doped manganites.

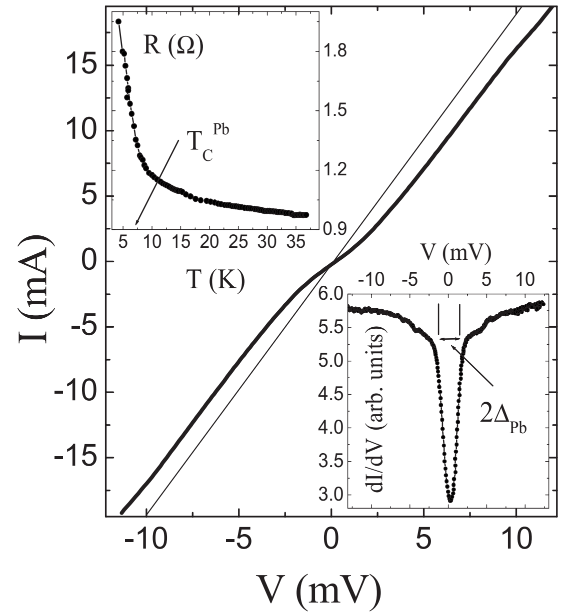


FIG. 1. The current-voltage (*I-V*) dependence of the Pb/LSMO contact without visible superconducting proximity effect; T =4.2 K. Top inset: the temperature dependence of the contact's resistance $R(T)$; arrow indicates $T_C^{Pb}=7.2$ K. Bottom inset: the contact's Andreev reflection spectra; $T=4.2$ K; arrows indicate superconducting gap of Pb.

II. EXPERIMENTAL DETAILS

The samples were prepared and explored using methods as described in Refs. 10, 11, and 13. Briefly, textured LSMO plates were grown using standard ceramic technique. In particular, ceramic powder plates sized $0.1\times1\times10$ mm^3 were pressed ($\sim$20 Kbar) and then subjected to annealing for 8 h at 1250 °C. This leads to an increase in the average size values of crystallites up to values of about 5–10 μm. Metallic contacts between LSMO plate and superconducting wire were formed by pressing slide-squash up a needle-shaped superconductor by a micrometric screw against the polished LSMO surface. The resistivity and the *I-V* characteristics were measured by the standard four-probe method with the low-frequency ac technique. The transition resistance of the current and potential contacts was $R \sim 10^{-8}$ Ω cm^2. The junction's resistance was much larger ($\sim 1\div30$ Ω) so that the rescaling effects can be neglected. The analysis showed that we deal with the so-called Sharvin contacts[14] and the contact's transport regime corresponds to the intermediate region, i.e., is neither ballistic nor diffusive one. (One can find the details in Refs. 11 and 13.)

III. RESULTS

For further references, we start with the results from a sample that reveals conventional properties of sS/hmF PC. In Fig. 1, the representative characteristics of the Pb/LSMO

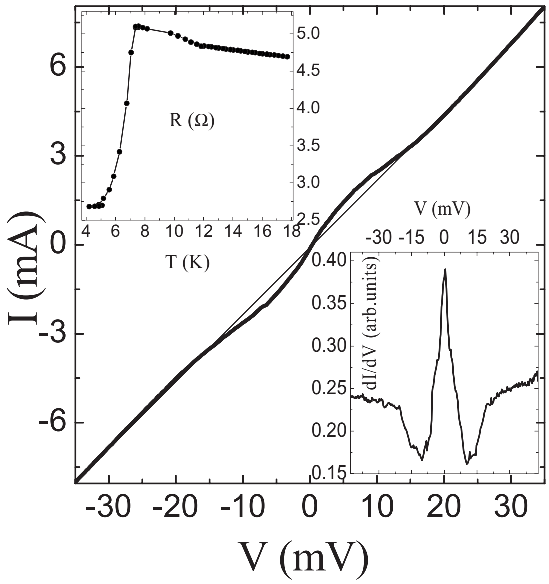


FIG. 2. The current-voltage (I-V) dependence of the proximity affected Pb/LSMO contact (PS 1); T=4.2 K. Top inset: the temperature dependence of the contact's resistance $R(T)$. Bottom inset: the contact's Andreev reflection spectra at T=4.2 K.

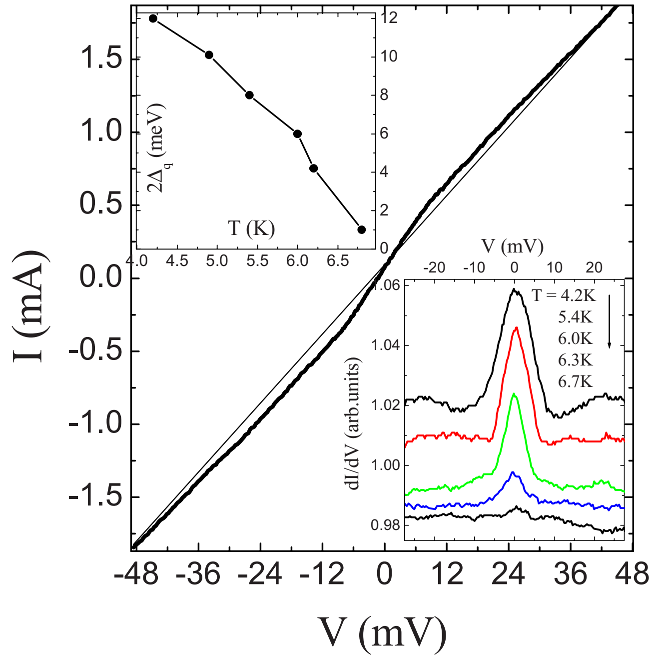


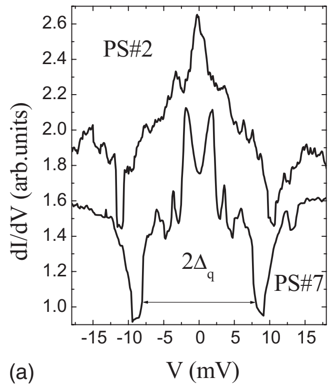
FIG. 3. (Color online) The current-voltage (I-V) dependence of the proximity affected Pb/LSMO contact, PS 11, at T=4.2 K. Top inset: the evolution of the proximity induced quasiparticle gap $2\Delta_q$ with temperature. Bottom inset: the temperature dependence of the contact's PS 11 Andreev reflection spectra. The curves are shifted for clarity.

contact without PE are exposed: the I-V dependence at T=4.2 K is shown in the main panel, the top inset exhibits the temperature dependence of the contact's resistance $R(T)$, and the bottom inset illustrates the contact's dynamic conductance at T=4.2 K. As one can see, in contrast to the AR at an S/N interface characterized by an excess current, here an excess voltage V_{exc} is observed. The almost constant V_{exc} value is detected for $|V| \leq 20$ mV. This proves the suggestion that heating effects could be neglected. The sharp increase in the $R(T)$ at T=7.2 K corresponds to the superconducting transition of Pb wire and shows that its superconducting properties are not affected by the underlying LSMO film. Any visible features of a superconducting PE have not been detected, and we will refer to these contacts as the contacts "without PE." Indeed, for singlet pairing, the superconducting coherence length $\xi_F \sim (D_F/E_{ex})^{1/2}$ for a hmF, like a manganite, is extremely short $\sim 5 \div 7$ Å (here D_F is the diffusion coefficient and E_{ex} is the exchange energy; see, for example, Ref. 3). Contribution of this small region to the contact's resistance is less than 1%. The suppression of the AR in sS/hmF PCs was observed experimentally by several groups (see, e.g., Ref. 15) and qualitatively was explained by de Jong and Beenakker's theory.[4]

Figures 2–4 focus on the key results of the report obtained on the PE contacts. The label data of the PE contact is a quite visible drop of the contact's resistivity just after a superconducting transition of Pb. We observed pronounced picture of the PE on few junctions. Figure 2 shows the representative characteristics of the Pb/LSMO PE contact (contact PS 1). The I-V dependence is shown in the main panel, the top inset exhibits the temperature dependence of the contact's resistance $R(T)$, and the bottom inset illustrates the contact's AR spectra. As one can see, in contrast to the case in Fig. 1, at $T<7.2$ K a sharp *drop* of the contact's resistivity is observed. Reduction in the resistance for PS 1 PC is about 50%, a theoretical limit that one can expect for a perfect S/N junction. At low voltage, an excess current I_{exc} is also unambiguously detected.

In Fig. 3 we documented the properties of one more Pb/LSMO PC (contact PS 11). In comparison with the PS 1 (see Fig. 2), the normal-state resistivity of this contact is about six times larger; however, it demonstrates all specific features of the proximity affected contact, namely: excess current, anomalous AR spectra, proximity induced gap, that is much larger than that for Pb (see discussion below). In bottom inset, the evolution of the AR spectra of the PS 11 with temperature is shown. These data directly prove that the anomalous behavior of the junction is due to the superconducting state of Pb. In top inset, the temperature dependence of proximity induced quasiparticle gap is shown.

In Figs. 4(a) and 4(b) the data for the differential conductance $G(V)$ of three other Pb/LSMO proximity affected PCs is presented. Similar to a conventional AR at an S/N interface and opposite to the results in Fig. 1, a doubling of the normal-state conductivity has been observed. It is well established now that the AR acts as a quasiparticle current channel for an applied voltage $eV<\Delta_q$, where Δ_q is the single-particle gap at the interface. From the experimental results in Figs. 2, 3, and 4(a), we extract that the proximity induced single-particle gap at the Pb/LSMO interface is $\Delta_q \approx 6 \div 8$ meV. For an exclusive case shown in Fig. 4(b) we observe the proximity gap as large as $\Delta_q \approx 18$ meV; that is a magnitude of Δ_q earlier had been detected for Pb/LCMO and MgB_2/LCMO PCs.[10,11] Note that the detected Δ_q is much



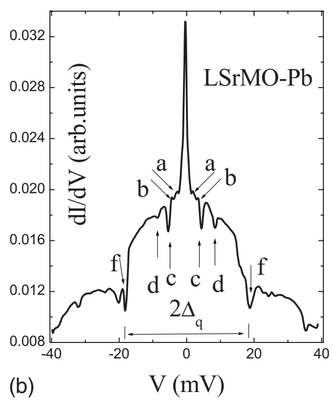


FIG. 4. The conductance $G(V)$ vs voltage dependences for proximity affected Pb/LSMO point contacts at 4.2 K: (a) PS 2 and PS 7. The conductance PS 7 is an example of a complex AR spectra has been observed for some Pb/LSMO contacts; (b) PS 4; see text for a classification of the resonances.

larger than that of Pb; the latter is as large as $\Delta_{Pb}(T=0) = 1.41$ meV.

In Fig. 4, for voltage eV $< \Delta_q$ a subharmonic gap structure due to multiple AR is definitely visible and can be classified as described below. The concept of multiple AR was first introduced by Klapwijk *et al.*,[16] for *SNS* junctions. Then it was shown[17,18] that this mechanism is the reason for subgap quasiparticle current in junctions between superconductors of any type, and specifically in superconducting PCs. It is worthy to note here that multiple AR can be observed only if *both electrodes* are in a superconducting state. So, the observation of the SGS is a strong argument in favor of the fact that the LSMO is in a superconducting state with *actual gap independent on Pb*.

Let us summarize the main results we have detected for proximity affected PCs (see Figs. 2–4). First, we observe the principal fact such as spectacular drop of the contact's resistance with the onset of the Pb superconductivity. Second, the subharmonic gap resonances due to multiple AR are directly visible. Third, in proximity affected PCs, the magnitude of a proximity induced gap of the LSMO is much larger than that of the Pb and may be as large as $\Delta_q \approx 18$ meV. These strongly suggest that both electrodes are in a superconducting state with independent gaps. All these anomalies are observed only in the superconducting state of the Pb tip ($T < T_C^{Pb}$).

IV. DISCUSSIONS

At present there are a few models of the subgap transport in sS/hmF heterostructures. At this stage of investigations, we found that the findings for proximity affected PC are most likely consistent with the theory of Ref. 9. This model predicts the appearance at sS-*F* interface of superconducting *p*-wave triplet even frequency correlations with an unusually long penetration length in the ferromagnetic metal. The appearance of triplet superconductivity (tS) requires the interplay of two separate interface processes: a spin mixing and a spin destroying scattering. The spin mixing effect generates at the sS side of the sS/*F* boundary the triplet correlation with "zero-spin" component. Similar to the wave function of the singlet pair, this component penetrates into the *F* on a short distance. (In the case of a fully spin polarized ferromagnet the conversion singlet pairs into triplet takes place entirely within the singlet superconductor.[19]) Spin destroying scattering induces in the sS both "nonzero-spin" triplet components of the pair amplitudes. These equal spin triplet correlation penetrates on an unusually long length $\xi_T \sim (D_F/2\pi T)^{1/2}$ into the hmF (typically temperature T is much smaller than the exchange energy E_{ex}).

The main condition for spin triplet channels pairing to be induced at sS/*F* interface is the so-called "spin active" interface, i.e., the ability of the sS/*F* interface to convert a singlet pair into triplet one. For manganites, several theoretical models and numerous experimental data suggest that nanoscale nonhomogeneity is an intrinsic feature of these compounds (see, e.g., Ref. 12). Another characteristic important for our discussion is that, due to Hund's interaction (for Mn^{3+} the Hund's energy ~ 1 eV), spin disorder serves as strong spin scattering center for charge carriers. By assuming that the surface of manganites is spin active one, for proximity affected contacts, we suggest that the conditions for the unconventional PE are fulfilled, i.e., depending on the local magnetic nonhomogeneity at the sS/LSMO boundary, the LSMO surface causes coherent equal spin *p*-wave even frequency pairing correlations, which spread over large distance into the manganite's bulk.

However, the induction of pairing correlations in the normal region is not enough for the realization of multiple AR. The observation of the subharmonic structure requires *the existence of actual gaps in both superconducting electrodes*.[17,18] Thus, experimental finding of the subharmonic structure proves that the proximity induced superconducting state of LSMO possesses *intrinsic superconducting gap* independent on Pb, which is much larger then Δ_{Pb}.

Following the physics described, in Fig. 5 a spatial structure of the current through the proximity affected Pb/LSMO contact is shown. The figure explains a mutual conversion of the currents along the contact. In fact, due to a long-range PE, in the case of the proximity affected sS/LSMO contact we deal with a charge transport through an sS-tS-hmF heterostructure. Namely, there is a region at sS-hmF interface where a conversion from spin singlet pairs into spin triplet pairs takes place. The equal spin triplet supercurrent flows through the hmF, while the singlet part is completely blocked.[19] The sum of the singlet and triplet currents is constant, obeying the continuity equation. At the boundary of

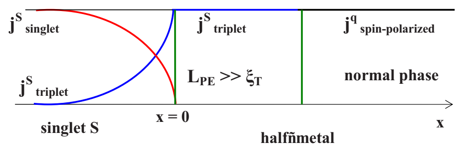


FIG. 5. (Color online) Spatial structure of the current through the proximity affected Pb/LSMO contact. The x axis is directed perpendicular to the sS/hmF interface that is at $x=0$; the hmF is placed in the region $x>0$, while the sS is located at $x<0$. Proximity affected region L_{PE} is much longer than the superconducting coherence length $\xi_T \sim (D_F/2\pi T)^{1/2}$ and $L_{PE} \gg \xi_T$.

superconducting and normal phases of the LSMO a spin polarized supercurrent is continued as a quasiparticle current due to the usual AR mechanism. Indeed, at both side of the tS-hmF interface the charge current is spin polarized and there is no restriction (because of spin) on the AR. As a result, an excess current and a doubling of the normal-state conductance have to be observed.

Let us now consider in detail the region where a conversion between spin singlet and spin triplet pairs takes place (see Fig. 6). Figure 6(a) illustrates the so-called "semiconductor picture" of the proximity affected PC. Figure 6(b) explains the mechanism of conversion between spin singlet and spin triplet pairs due to multiple ARs. At the sS-tS interface we deal, in fact, with a weak link (or constriction) between two different superconductors. The "weak link" here is a region where both singlet and triplet pairing interactions are suppressed, or, in other words, the region where both the singlet and triplet Cooper pairs are destroyed [see Fig. 6(a)]. As was shown by Octavio *et al.*[17] in the semiclassical picture, for a given voltage $V<\Delta/e$ across the weal link a quasiparticle accelerated from Fermi surface suffers $n\sim\Delta/\mathrm{eV}$ ARs until it reaches the top of the pair potential well. For short constriction between two different singlet superconductors, sS_1-sS_2, it was obtained[18] that this multiple AR manifests itself in dc current as current steps at voltages Δ_1/n, Δ_2/n, or $(\Delta_1+\Delta_2)/m$, with $n=1,2,3\ldots$ and $m=1,3,5\ldots$ Generalizing this picture, we describe the charge current through the sS-tS$\Rightarrow$sS-weal link-tS interface as follows.

In the particular case shown in Fig. 6(b), an incoming electron (hole) of a given spin subband and under the energy gap Δ_{sS} cannot enter in the triplet superconducting electrode. It is spin flipped and then is Andreev reflected as a hole (electron) back to the sS, simultaneously adding a *triplet* Cooper pair to the condensate in the tS. This hole (electron) is spin flipped and then is reflected by Andreev mechanism as an electron (hole) back to the tS, simultaneously adding a *singlet* Cooper pair to the condensate in the sS. For a given voltage across the sS-tS interface a quasiparticle undergoes $n\sim\Delta_{sS}/\mathrm{eV}$, Δ_{tS}/eV, or $m\sim(\Delta_{sS}+\Delta_{tS})/\mathrm{eV}$ reflections (depending on electrodes and energy, it starts) until it reaches the top of the pair potential well. As was already indicated, a crucial condition for conversion processes under consideration is the spin-flip scattering at the interface. Thus, the physical mechanism of mutual conversion between the spinless Cooper pair and the spin polarized Cooper pair is a quasiparticle flip-scattering by following Andreev reflection.

To proceed further with SGS classification, we should take into account a degradation of manganite's surface properties.

It is well established now that the origin of manganite's surface degradation is due to the existence of broken bounds at the surface and the translational symmetry breaking of the lattice that generate randomness in the double-exchange interactions. Although it is difficult to directly assert that disorder is confined in a well delimited surface layer, the boundary with structural disorder is estimated to be of order of a few nanometers.[20–24] These reasons are general for all manganites; however, it was found that the size of the surface layer for the LSMO is larger than for the LCMO compound.[20,21] Also, for the LSMO the surface terminating atomic layer is assigned to the MnO_2 layer,[22] while for the LCMO the surface exhibits a large enhancement in the Ca concentration compared to the bulk material.[22,23] The origin of this distinction in surface segregation is not understood at the moment, but we can conclude that most probably the carrier concentration and the magnetic state of the LCMO and the LSMO surfaces differ. That is why, in our opinion, the induced quasiparticle gap typically detected for the Pb/LSMO PCs, $\Delta_q\approx 8$ meV [see Fig. 4(a)], differs from that for the Pb/LCMO PCs $\Delta_q\approx 18$ meV.[10,11] Only for an exclusive case, shown in Fig. 4(b), we have a chance to detect for the LSMO the induced quasiparticle gap as large as for the LCMO compound, $\Delta_q\approx 18$ meV. Taking this remark into

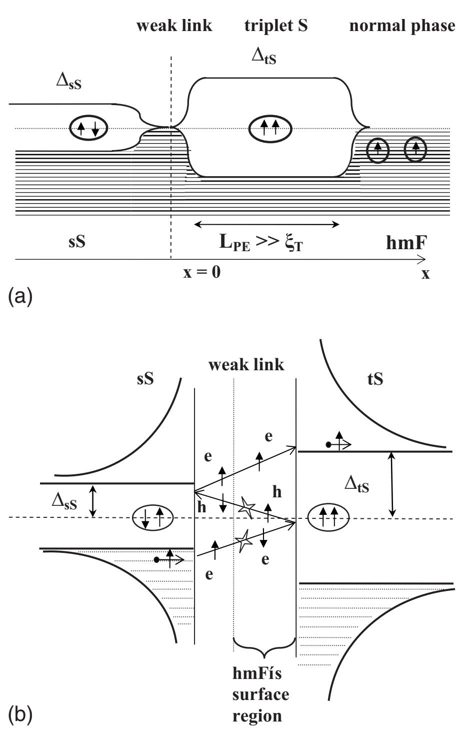


FIG. 6. (a) Semiconductor picture of proximity affected Pb/LSMO contact. Weak link here is a region where both singlet and triplet pairing interactions are suppressed. (b) Trajectory of a quasiparticle that is accelerated out of the condensate by the electric field suffering multiple Andreev reflections. In the case of singlet and triplet superconducting electrodes every Andreev reflection is foregone with a spin-flip scattering. Spin-flipping processes are illustrated by stars.

account, we can classify the SGS voltage marked in Fig. 4(b) by the labels as follows: $a \to \Delta_{Pb}$, $b \to (\Delta_{Pb}+\Delta_{LSMO})/5$, $c \to \Delta_{LSMO}/3$, $d \to \Delta_{LSMO}/2$, and $f \to \Delta_{LSMO}$, where $\Delta_{Pb} \approx 1.4$ meV and $\Delta_q = \Delta_{LSMO} \approx 18$ meV.

Let us now make some comment on such an important question as the origin of the quasiparticle gap Δ_q the magnitude of which cannot be explained in terms of conventional theory of proximity effect.[3] At this stage of investigation, we can only speculate about different mechanisms due to which the proximity induced single-particle gap at the sS-manganite interface is much larger than the superconducting gap of Pb or MgB_2. To overcome this problem, we assumed in Ref. 11, as a likely hypothesis, that the manganites are thermodynamically very close to a triplet *p*-wave superconducting state, i.e., at low temperature, the local triplet superconducting fluctuations are essentially sustained in a manganite and a singlet superconductor only fixes the phase coherency of a superconducting state. The role of an external stimulus is important here because the half-metallic ferromagnetic state of manganites corresponds to a metal in a "dirty limit;" i.e., in a metallic phase the coherency of *p*-wave symmetry superconducting state with an intrinsic gap is destroyed.[25] A few possible mechanisms due to which the triplet superconducting fluctuations may be quite strong have been listed in Ref. 11. In brief, the situation with manganites may be similar to superconducting properties of strontium ruthenate Sr_2RuO_4. Convincing experimental data have been obtained in favor of the spin-triplet *p*-wave superconductivity of clean Sr_2RuO_4. However, the superconductivity in impure phase of Sr_2RuO_4 has not been found (see Ref. 26 and references therein). The electron-electron interaction of *p*-wave symmetry may also be quite strong due to an exchange of magnons.[27] An interesting suggestion is also a "latent superconductivity in doped manganites" considered in Ref. 28.

In the end of the section, let us briefly discuss other possible explanations and/or mechanisms, which could be related to our observations.

Recently, it was predicted that in sS/F structures, the so-called *odd frequency* pairing could take place.[8,29] In this case, the Cooper pair wave function is symmetric under exchange of spatial- and spin-coordinates but antisymmetric under exchange of time-coordinates. The study of such pairing in sS/*F* junction was addressed by a number of authors over the last years.[19,30] However, if the superconducting correlations are odd in frequency, a pairing interaction has to be additionally a *frequency dependent* (due to strong retardation effect) in order to have a nonzero intrinsic gap in the LSMO. We think this scenario is more exotic than we suggested.

Nonlocal or crossed AR in which an electron from one magnetic domain is Andreev reflected as a hole into oppositely polarized domain while a pair is transmitted into a superconductor,[31] is, in principle, possible. However, it seems improbable that in all the proximity affected PCs the portion of domains with opposite magnetization is exactly equal.[11]

A conversion of spinless Cooper pair into spin polarized Cooper pair and vice versa is also possible due to absorption (respectively, emission) of a magnon.[32] If this mechanism is governing, the junction's current-voltage characteristic has to be, at low temperatures, asymmetric with respect to the base voltage. The $I(V)$ characteristics of all PCs we have explored are symmetric, and thus the magnon assisted mechanism does not control the charge transport in our PCs.

A large resistance switching effect has been recently discovered in contacts of colossal magnetoresistive materials with nonmagnetic metals[33–37] and is actively discussed in connection with resistive switching memory (the so-called "memresistor," see Ref. 38 and references therein). Resistivity switching is observed after the bias voltage overcame a threshold value (typically $\sim \pm 3 \div 5$ V), and is reversible and nonvolatile. The effect is shown to occur over an extended region, about 100 nm around the contact interface into manganite. A few possible models of the resistance switching are under debate. Some research groups proposed an electrochemical migration of oxygen ions at the interface as the origin of resistance instability.[37] Another probable model is the insulator-metal transition in which the electronic charge injection acts like a doping to induce a Mott-like transition.[34,35] Some authors argued that the switching effect can be regarded as a charging effect at the Schottky-like interface.[36] Anomalous of the current-voltage characteristics, we have observed in proximity affected sS/hmF contacts have no hysteretic properties and no thresholds; they appear only when nonmagnetic electrode is a superconducting state. Therefore, we consider that the mechanisms of the resistance instability cannot directly relate to our findings.

V. SUMMARY

To study mutual influence of competing ordered states, we have prepared point contacts of a low-temperature superconductor, Pb, and a half-metallic ferromagnet, $La_{0.7}Sr_{0.3}MnO_3$, and investigated their transport properties. The subgap charge current in these contacts is due to the Andreev reflection. Several evidence of existence of unconventional (triplet) superconducting pairing and long-range PE are detected. The obtained experimental data can be understood within a model incorporating the concepts (i) of magnetic disorder-induced superconducting triplet correlations at sS/hmF interface with a long-range decay length and (ii) a competing state in which pairing correlations with energy gap Δ_q exist without long-range superconducting phase coherence. The physical mechanism due to which a conversion between singlet Cooper pairs and spin polarized triplet Cooper pair is realized is the Andreev reflection with foregoing spin flipping. The model proposed leads to a natural explanation of four of the five findings of the experiment: (i) a spectacular decrease in the resistance just after the superconducting transition of a tip (Pb), (ii) an excess current, (iii) doubling of the conductance, and (iv) a subharmonic gap structure. The origin of anomalously large proximity induced gap Δ_q remains, however, unclear to date.

Systematic character and repeatability of a number of principal experimental facts suggest that some general physical phenomena have been observed in transport properties of proximity affected singlet superconductor-half-metallic manganite contacts. Whether the origin of these features can be traced to a thermodynamic state of manganites with local triplet superconducting fluctuations, is a matter of further investigations. The results obtained are of great relevance for spin dependent electronics devices, which are based on exploration of nanometric superconducting and half-metallic magnetic materials.

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