Josephson Effect in Hybrid Oxide Heterostructures with an Antiferromagnetic Layer

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(Received 24 November 2006; revised manuscript received 30 April 2007; published 6 July 2007)

Josephson coupling between an s- and d-wave superconductor through a 50 nm thick Ca1−xSrxCuO2 antiferromagnetic layer was observed for the hybrid Nb/Au/Ca1−xSrxCuO2/YBa2Cu3O7−δ heterostructures and investigated as a function of temperature, magnetic field, and applied millimeter-wave electromagnetic radiation. The magnetic field dependence of the supercurrent Ic(H) exhibits anomalously rapid oscillations, which is the first experimental evidence of the theoretically predicted giant magneto-oscillations in Josephson junctions with antiferromagnetic interlayers.

DOI: 10.1103/PhysRevLett.99.017004

The coexistence of superconducting and magnetic ordering in solids is of great interest for fundamental studies and electronic applications. In bulk compounds the exchange mechanism of ferromagnetic (FM) order tends to align spins of superconducting (S) Cooper pairs in the same direction preventing singlet superconducting pairing [1,2]. At the interfaces between superconducting and magnetic materials, however, superconducting and magnetic order parameters may interact due to the proximity effect resulting in interplay between superconducting and magnetic ordering and novel physical phenomena. For instance, superconductivity in ferromagnetic/superconducting/ferromagnetic (FM/S/FM) spin-valve junctions can be controlled by spin orientations in the FM electrodes [3]. Another effect is the damped oscillatory behavior of the superconducting order parameter induced in the ferromagnetic layer across S/FM interfaces that may lead to a π phase shift in the supercurrent of S/FM/S Josephson junctions [4].

We would expect interesting mesoscopic physical effects also at interfaces between superconductors and antiferromagnets (AFM). Monotonous suppression of the superconducting order parameter at the S/AFM interface with band-type AFM and novel low-energy Andreev bound states originating from spin dependent quasiparticle reflections at S/AFM interfaces have been theoretically predicted [5,6]. The observation of the Josephson effect in Nb/Cu/FeMn/Nb polycrystalline thin film heterostructures has been demonstrated in [7], where the γ-Fe50Mn50 alloy was used as metallic AFM layer. Although strong suppression of superconductivity has been observed in AFM FeMn layers similar to the ferromagnetic case, the supercurrent modulation with an applied magnetic field Ic(H) shows rather conventional Fraunhofer pattern [7]. If, instead, the polycrystalline metallic AFM is substituted by an array of ferromagnetic layers with alternating directions of magnetization, then according to the theoretical calculations by Gor’kov and Kresin the Ic(H) dependence should exhibit rapid oscillations originating from a gradual weak canting of the magnetic moments in the presence of an applied magnetic field [8].

Investigations of S/AFM interfaces composed of oxide high-temperature superconductors (HTS) and antiferromagnetic materials are relevant in order to understand the pairing mechanism of high-temperature superconductivity. No superconducting pairing has been found experimentally in La1.85Sr0.15CuO4/La2CuO4/La1.85Sr0.15CuO4 S/AFM/S heterostructures with a one-unit-cell thick antiferromagnetic insulator La2CuO4, thus, counteracting the mixing of antiferromagnetism and high-temperature superconductivity [9]. If the La2CuO4 barrier is doped across the insulator-metal transition and becomes HTS itself (La2CuO4+δ), the so-called “giant proximity effect,” namely, a pronounced Josephson supercurrent, has been observed in the S/N′/S heterostructures above the superconducting transition temperature of the La2CuO4+δ N′ barrier with a thickness of up to 20 nm, which is well beyond the short coherence length of the cuprates [10]. In [10] the “giant proximity effect” is explained by resonant tunneling through the pair states in the barrier. The alternative explanation has been suggested in [11], where the experimentally obtained high value of the supercurrent is explained by the presence of microshorts through the interlayer.

In spite of recently renewed interest in superconducting structures with magnetically active materials, there is still a lack of experimental results on Josephson junctions with antiferromagnetic weak links, in particular, comprised of oxide HTS and AFM materials. For instance, no experimental observations of the theoretically predicted rapid oscillations of Ic(H) have been demonstrated so far [8].

Here we report on the experimental studies of dc and rf current transport in hybrid thin film S-N-AFM-D antiferromagnetic junctions fabricated in the form of Nb/Au/Ca1−xSrxCuO2/YBa2Cu3O7−δ mesa heterostructures with areas from 10×10 up to 50×50 μm2 [see inset (a) of Fig. 1] [12,13]. Here Nb is a conventional s-wave superconductor (S), YBa2Cu3O7−δ (YBCO) is an oxide d-wave superconductor (D), Au is a normal metal.

The Josephson effect in hybrid oxide heterostructures with an antiferromagnetic layer can be understood within a simple model [14]. The existence of the Josephson effect was predicted for a N/S/N junction with a non-ideal phase-matching condition. The applied magnetic field Hc provides the condition of phase matching in the Josephson junction. The Josephson current Ic is proportional to the parameter l0/λ_0, where l0 is the phase-breaking length and λ_0 is the London penetration depth. The parameter l0/λ_0 is proportional to cos(θ/2), where θ is an angle between the phase of the order parameter of the superconducting junction and the macroscopic magnetic field.

The Josephson current Ic in hybrid oxide heterostructures with an antiferromagnetic layer can be expressed as [15]

Ic = (2e/ℏ) δ cos(θ/2)

where δ is the thickness of the antiferromagnetic layer and cos(θ/2) is the cosine of the angle between the phase of the order parameter of the superconducting junction and the macroscopic magnetic field.

The Josephson effect in hybrid oxide heterostructures with an antiferromagnetic layer can be observed in a magnetic field Hc that provides the condition of phase matching in the Josephson junction. The Josephson current Ic is proportional to the parameter l0/λ_0, where l0 is the phase-breaking length and λ_0 is the London penetration depth. The parameter l0/λ_0 is proportional to cos(θ/2), where θ is an angle between the phase of the order parameter of the superconducting junction and the macroscopic magnetic field.

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ping conductivity and a Neél temperature magnetic resonance measurements of the CSCO thin films AFM layer suggested in [8]. Electric and electronic paramagnetic resonance measurements presented in this Letter have been carried out. The Ic(T) dependence of the Nb superconducting gap (ΔNb) derived from the I-V curve of the Nb/Au/YBCO junction (solid line in Fig. 1) [17]. However, the Ic(T) curve does not follow a dependence typical for S/N/S junction and the values of Vc = IcRN (RN is resistance of the junction in the normal state) calculated from the I-V curves of different samples do not show any pronounced dependence on d5 [18]. Thus, we may conclude that the superconducting coherence length in the CSCO AFM layer ξAFM, which determines the penetration depth of the superconducting ordering in the AFM layer, is not small with respect to the thickness of the CSCO layer d5. A qualitative estimation of ξAFM in the case of ballistic electron transport (quasiparticle mean free path l > d5) can be performed by modeling of the AFM as a stack of -FM-FM'-thin ferromagnetic layers FM and FM' with alternating direction of exchange field ξAFM ~ min{l, hνF/(2πkT)} [8], where νF is the quasiparticle Fermi velocity, h and k are the Planck and Boltzmann constants, respectively. Thus, we obtain ξAFM ~ 7 nm at T = 4.2 K for one of the measured AFM junctions. Note that the value of ξAFM depends on the properties of the CSCO AFM layer (oxygen content, Fermi velocity, mean free path, etc.) and might vary in different experimental samples.

Our experimental data show that Ic and RN of the AFM junctions vary considerably between the samples since the transparency value of the Au/CSCO interface is not technologically reproducible (see Table I). However Ic and RN are the correlated parameters and the values of Vc in the different AFM junctions are consistent and systematically 2–3 times higher than those of the Nb/Au/YBCO junctions. Thus, one can conclude that the reason for the observed enhancement of the supercurrent in the AFM junctions is related to the presence of the AFM CSCO interlayer. A possible reason for this enhancement is the complex superconducting order parameter in YBCO thin films with the dominant d-wave and minor s-wave components [19]. The latter arises even in the bulk, in particular, because of orthorhombic crystal structure of YBCO [17,19]. However, change of symmetry of the superconducting order parameter may occur across the AFM/D interface in the AFM junctions. For instance, the d-wave

(N), and Ca1-xSrxCaCu2O2 (CSCO) is a quasi-two-dimensional Heisenberg AFM, where the quasiparticle spin direction alternates in the neighbor (111) ferromagnetic planes [see inset (b) of Fig. 1] [14]. Taking into account weak magnetic coupling between the neighbor magnetic planes we can consider current transport along (111) CSCO being equivalent to the case of an A-type AFM layer suggested in [8]. Electric and electronic paramagnetic resonance measurements of the CSCO thin films in the temperature range of 4.2–300 K demonstrate hopping conductivity and a Neél temperature TNeel = 90–120 K, respectively [15]. Thus, the CSCO films are in a G-type antiferromagnetic Mott-insulating state within the temperature range of 4.2–40 K, where the electrical measurements presented in this Letter have been carried out.

In order to avoid microshorts in the AFM junctions, several precautions were taken: (i) the deposited CSCO films (d5 = 20 or 50 nm) are thicker than the rms surface roughness of the YBCO layer; (ii) the Nb/Au bilayer is used as superconducting counterelectrode in our junctions. Direct Nb deposition on top of the CSCO/YBCO heterostructure results in the formation of a Nb/CSCO interface with very high resistance (~1 Ω cm²) due to Nb oxidation. Thus, if the Au layer was locally damaged because of the finite surface roughness of the CSCO/YBCO heterostructure, then a high resistivity niobium oxide is formed to heal any shunting conductivity at that point.

A 4-point measurement technique was used for electrical characterization of our AFM junctions with two contacts to the YBCO electrode and two contacts to the Nb counterelectrode [see inset (a) of Fig. 1]. The difference in electronic parameters of Au and CSCO and the surface characteristics of the CSCO/YBCO bilayer determine the potential barrier Ib at the Au/CSCO interface and the junction resistance [16]. Thus, our AFM junction can be considered as an S/N/Ib/AFM/D structure, where the superconducting order parameter is induced in the normal Au and the AFM CSCO layers by the proximity effect to Nb and YBCO electrodes, respectively [17].

Figure 1 shows temperature dependences of the supercurrent Ic(T) obtained using dc transport measurements of the AFM junction. The Ic(T) dependence of a Nb/Au/YBCO junction (d5 = 0) is presented for comparison. We observe a good qualitative agreement of all normalized Ic(T) dependences with the temperature dependence of the Nb superconducting gap (ΔNb) derived from the I-V curve of the Nb/Au/YBCO junction (solid line in Fig. 1) [17]. However, the Ic(T) curve does not follow a dependence typical for S/N/S junction and the values of Vc = IcRN (RN is resistance of the junction in the normal state) calculated from the I-V curves of different samples do not show any pronounced dependence on d5 [18]. Thus, we may conclude that the superconducting coherence length in the CSCO AFM layer ξAFM, which determines the penetration depth of the superconducting ordering in the AFM layer, is not small with respect to the thickness of the CSCO layer d5. A qualitative estimation of ξAFM in the case of ballistic electron transport (quasiparticle mean free path l > d5) can be performed by modeling of the AFM as a stack of -FM-FM'-thin ferromagnetic layers FM and FM' with alternating direction of exchange field ξAFM ~ min{l, hνF/(2πkT)} [8], where νF is the quasiparticle Fermi velocity, h and k are the Planck and Boltzmann constants, respectively. Thus, we obtain ξAFM ~ 7 nm at T = 4.2 K for one of the measured AFM junctions. Note that the value of ξAFM depends on the properties of the CSCO AFM layer (oxygen content, Fermi velocity, mean free path, etc.) and might vary in different experimental samples.

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symmetrical component of the superconducting pairing of YBCO may be transformed across the CSCO/YBCO interface into a superposition of the spin-triplet $p_x$-wave and the extended s-wave components, thus, enhancing the values of $V_c$ [20].

In S/FM/S Josephson junctions the “giant Josephson effect” caused by an anomalously large penetration of the superconducting order parameter into the ferromagnetic layer with spiral-type magnetization may be explained by a triplet superconducting pairing generated at the S/FM interface between the s-wave superconductor and the ferromagnetic film [2,21]. We emphasize that in our case, unlike [21], the value of the Josephson supercurrent may be qualitatively explained only by a singlet s-wave component of the superconducting pairing induced at the CSCO/YBCO interface. Moreover, low-energy Andreev bound states at the CSCO/YBCO interface may act as an additional channel for the supercurrent in our AFM junctions [6].

Integer and half-integer Shapiro steps at voltages $V_n = n(hf_e/2e)$ and $V_{n-1/2} = (n - 1/2)hf_e/2e$ ($n = 1, 2, 3, \ldots$) are observed in the I-V curves of the investigated AFM junctions irradiated by an external millimeter-wave electromagnetic source at frequencies $f_e = 36$–120 GHz (see Fig. 2 for $f_e = 56$ GHz). The oscillating dependences of the critical current, $I_c(I_e)$, and the amplitudes of the Shapiro steps, $I_n(I_e)$, versus the millimeter-wave electromagnetic current amplitude $I_e$ were derived from the I-V curves of the AFM junctions (Fig. 3). Values of $I_e$ were calculated as the square root of power of the external radiation. The experimentally obtained $I_n(I_e)$ dependences follow the resistively shunted junction (RSJ) model of Josephson junctions, where electrical current of the junction is considered as superposition of superconducting and quasiparticle parts [22].

In the high frequency limit of the RSJ model $f_e > f_c = (2e/h)I_cR_N$, the $I_n(I_e)$ dependence is proportional to zero order of the modified Bessel function $J_0(2\alpha)$ and, similarly, $I_n(I_e) \sim J_n(2\alpha)$, where $\alpha = eI_cR_N/hf_e$. Good approximation of the experimentally obtained $I_n(I_e)$ dependence by our simulations within the RSJ model is a supplementary indication of the absence of microshorts in the AFM junctions and Josephson origin of the supercurrent. The absence of excess currents in the I-V curves of the AFM junctions is further evidence against the formation of microshorts [17]. The half-integer Shapiro steps (Fig. 2) may originate from a second harmonic component $I_{c2} \sin2\varphi$ in superconducting current-phase relation of the AFM junctions $I(\varphi) = I_c1 \sin\varphi + I_{c2} \sin2\varphi$ as well as from their finite capacitance $C$ [Stewart-McCumber parameter $\beta_c = (2e/h)I_cR_N^2C$] [23]. The modified RSJ model, which takes into account nonzero values of $C$ and $I_{c2}$, fit well the experimental $I_n(I_c)$ and $I_1(I_c)$ curves in Fig. 3 [24]. In particular, the $I_{c2}$ value used to fit the experimentally obtained $I_n(I_c)$, $I_1(I_c)$, and $I_{1/2}(I_c)$ curves (Figs. 2 and 3) is as high as $q = I_{c2}/I_{c1} = 0.2$ [24].

Figure 4 shows $I_c(H)$ dependences of an AFM junction and a Nb/Au/YBCO junction of the same area $50 \times 50 \mu$m$^2$. The $I_c$ values in Fig. 4 were obtained from the dc measurements of the I-V curves. In order to perform high sensitivity magnetic field measurements in the microTesla range we used a magnetic field coil with a dc source and amorphous multilayer $\mu$-metal shielding. The magnetic field is applied parallel to the thin film surfaces, i.e., in plane of the Josephson junction. The widths $H_0$ of the central $I_c$ peaks of the $I_c(H)$ curves are 3.2 and 88 $\mu$T for the AFM junction and the Nb/Au/YBCO junction, respectively. Another specific feature of the AFM junction is the additional rapid modulation of the $I_c(H)$ pattern with a period of $H_1 = 0.7 \mu$T, which is about 5 times smaller.

### Table I: Parameters of the AFM junctions at $T = 4.2$ K and $H = 0$. $d_{sc}$ is the thickness of the Ca$_{1-x}$Sr$_x$CuO$_2$ interlayer, $x$ characterizes the Sr doping level in Ca$_{1-x}$Sr$_x$CuO$_2$. Parameters $A$, $I_c$, $R_N$, and $V_c = I_cR_N$ are the area, supercurrent, normal resistance, and characteristic voltage of the junctions, respectively.

<table>
<thead>
<tr>
<th>Sample no.</th>
<th>$d_{sc}$ (nm)</th>
<th>$x$</th>
<th>$A$ ($\mu m^2$)</th>
<th>$I_c$ (µA)</th>
<th>$R_N$ (Ω)</th>
<th>$V_c$ (µV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>20</td>
<td>0.5</td>
<td>$10 \times 10$</td>
<td>400</td>
<td>0.8</td>
<td>320</td>
</tr>
<tr>
<td>2</td>
<td>50</td>
<td>0.15</td>
<td>$20 \times 20$</td>
<td>555</td>
<td>0.4</td>
<td>222</td>
</tr>
<tr>
<td>3</td>
<td>50</td>
<td>0.15</td>
<td>$10 \times 10$</td>
<td>48</td>
<td>3.5</td>
<td>170</td>
</tr>
<tr>
<td>4</td>
<td>50</td>
<td>0.5</td>
<td>$50 \times 50$</td>
<td>70</td>
<td>2.9</td>
<td>203</td>
</tr>
<tr>
<td>5</td>
<td>0</td>
<td>0</td>
<td>$50 \times 50$</td>
<td>270</td>
<td>0.2</td>
<td>54</td>
</tr>
<tr>
<td>6</td>
<td>0</td>
<td>0</td>
<td>$20 \times 20$</td>
<td>18</td>
<td>3.6</td>
<td>65</td>
</tr>
</tbody>
</table>
where $J$ is the calculated current from the millimeter-wave electromagnetic current $I_R$ of the AFM junction no. 4 (filled circles) and the $I_c$ of the AFM junction no. 3 at $V = I_c R_N$ = 0.0017 $\mu$V were demonstrated in the Nb/Au/CSCO/YBCO junctions with up to 50 nm thick CSCO AFM layers. The ac Josephson effect is manifested in multiple Shapiro steps, which are well fitted by the RSJ Josephson junction model. The experimentally observed rapid oscillations of $I_c(H)$ dependence qualitatively agree with the theory [8].

The authors are grateful to V. V. Demidov, F. Lombardi, V. A. Luzanov, I. M. Kotelyanski, and U. Poppe for helpful discussions. This work was supported by the Russian Academy of Sciences, the ESF AQDJJ and THIOX, Swedish KVA, SI and SSF “OXIDE,” and EU NANOXIDETTC.

FIG. 3. Dependences of the supercurrent $I_c(I_0)$, circles] and the first integer Shapiro step $I_1(I_0)$, triangles] versus the applied magnetic field. The data are derived from the $I$-$V$ curves of the AFM junction no. 3 at $f_e = 56$ GHz and $T = 4.2$ K. The solid and dashed lines correspond to the $I_c(I_0)$ and $I_1(I_0)$ curves numerically calculated from the modified RSJ model with $q = I_2/I_1 = 0.2$ [24]. The dotted line shows the calculated $I_1(I_0)$ dependence for $q = 0$, i.e., $I_1 \propto J_1(2\alpha)$, where $J_1$ is a first order Bessel function and $\alpha = e I_c R_N/h f_e$.

than the $H_0$. The shape of the rapid oscillations strongly depends on the direction of the applied magnetic field. For instance, in-plane rotation of the applied magnetic field $H$ by 90° makes the $I_c(H)$ pattern asymmetric and doubles $H_0$ and $H_1$. Thus, the observed rapid oscillations of $I_c(H)$ qualitatively correspond to the theoretically predicted ones in the case of S/AFM/S Josephson junctions with layered AFM with alternating magnetic moments [8].

In conclusion, we have experimentally demonstrated dc and ac Josephson effects in Nb/Au/CSCO/YBCO thin film junctions with antiferromagnetic CSCO layers. Values of the Josephson characteristic voltage $V_c = I_c R_N$ = 100–200 $\mu$V were demonstrated in the Nb/Au/CSCO/YBCO junctions with up to 50 nm thick CSCO AFM layer. The ac Josephson effect is manifested in multiple Shapiro steps, which are well fitted by the RSJ Josephson junction model. The experimentally observed rapid oscillations of $I_c(H)$ dependence qualitatively agree with the theory [8].

FIG. 4. Magnetic field dependences of the supercurrent $I_c(H)$ of the AFM junction no. 4 (filled circles) and the Nb/Au/YBCO junction no. 5 (open circles) at 4.2 K. The $I_c$ is normalized by $I_{c_{\text{max}}} = I_c(4.2$ K). The inset shows a large-scale $I_c(H)$ graph of the Nb/Au/YBCO junction [25].

[13] We have observed no deviation of dc and rf properties of the AFM junctions deposited on differently oriented NdGaO$_3$ substrates. Thus, we do not include the thin film orientation issue in the following discussion of the obtained experimental data.
[25] The smaller $I_c$ = 165 $\mu$A for the junction 5 presented in Fig. 4 was measured after multiple cooling cycles 2 years later than $I_c$ = 270 $\mu$A given in Table I.