Transport Properties of La$_{0.7}$Sr$_{0.3}$MnO$_3$/MgB$_2$ Random Nanocomposites

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Abstract—We have investigated electrical transport properties of random binary networks composed of superconducting MgB$_2$ micro-particles and half-metallic ferromagnet La$_{0.7}$Sr$_{0.3}$MnO$_3$ nano-particles. Unusually large value of the threshold for a percolation transition to a superconducting state of the nanocomposite has been detected for MgB$_2$. We explain the observed behavior by significant geometric disparity between particles of the constituents and unconventional (triplet) superconducting state induced in the nanocomposite due to the proximity effect. The effect of high-frequency irradiation on a superconducting transition of the nanocomposite was investigated as well. The influence of a high-frequency signal on the critical current magnitude and superconducting transition of a nanocomposite is revealed. A nonthermal effect of an external electromagnetic irradiation on the nanocomposite characteristics has been found.

Index Terms—nanocomposite, percolation transition, proximity effect, Josephson junctions

I. INTRODUCTION

The first realization of a Josephson junction utility as tunable microwave sources and detectors can be traced back to the earliest works by Josephson. It was also very early realized that a single Josephson junction emits too little energy to be useful as a microwave source. This and other shortcomings can be eliminated by employing arrays of Josephson junctions. Such arrays can be obtained by creating structures representing a composition of a superconductor (S) separated by normal inclusions. In this case, it is possible to obtain a set of superconductor-normal metal-superconductor (S-N-S) contacts. Sensitivity of current-voltage (I-V) characteristics of the S-N-S contacts to high-frequency electromagnetic fields is due to a superconducting proximity effect. Namely, the proximity induced phase-coherent state is established between superconducting sides of the contact and thus the Cooper pairs flow through the normal interlayer. When a magnetic or electromagnetic field is applied to a contact, the phase coherence between the contact’s sides is broken and a contact transfers into a resistive state. In this case, the I-V characteristic of a contact demonstrates the absence of a critical current and a strong nonlinearity at low voltages, $V < V_c = I_c R_n$, on the contact (here $I_c$ is the critical current of the contact, $R_n$ is the contact resistance in a normal state). With increasing of an electromagnetic radiation, the nonlinearity of I-V characteristics of the contact in the $V_c$ region is rapidly suppressed and the contact transfers into a normal state. In practice, when conventional Ss are used, $V_c$ does not exceed several dozen microvolts. This circumstance strongly limits the dynamic range of Josephson junction functionality and thus its applicability in devices. To expand the dynamic range and significantly enlarge magnitude of $V_c$, it is proposed to use a three-dimensional structure of weakly coupled contacts - the so-called 3D Josephson medium. The creation of such a medium can be realized artificially by means of a direct contact of various materials with superconducting material. As a result, new compounds with unique characteristics not realized for ‘parent’ components can be obtained [1-7]. In particular, interest to superconducting proximity effect in S-N structures is initiated by the possibilities of new applications of these structures as novel functional elements of nano-electronics [1-3].

One of such S-N 3D Josephson medium nontrivial implementation can be a composite of a half-metallic ferromagnet (hmF) with an S metal. Indeed, an existence of proximity effect in hybrid hmF and high-$T_c$ S film’s structures has been demonstrated in Refs. [8-11]. Long-range proximity effect and induced superconducting order in such hmF as manganese was detected, in particular, by Andreev reflection in experiments on the manganite-S point contacts [12-15] (manganite = La$_{0.5}$Ca$_{0.5}$MnO$_3$, La$_{0.5}$Sr$_{0.5}$MnO$_3$, S = Pb, Nb, MgB$_2$). In addition to the spectroscopic manifestation of the proximity effect in contacts of an S with manganites, a superconducting current has been detected in numerous studies of hybrid S-manganite-S film’s structures [8-11,16]. Overall, the superconducting proximity effect was observed in half-metallic manganites being in contact with Ss of different symmetries: as $s$-wave symmetry Pb, Nb, MgB$_2$ [12-14], as well as $d$-wave symmetry cuprates [8-11,16,17], of the order parameter. Yet, the question, what mechanism is responsible for the transfer of Cooper pairs through the hmF remains still open and is the subject of intensive discussions [1,3,4,5].

II. EXPERIMENT

We have studied transport properties of superconductor (MgB$_2$) – hmF (manganite La$_{0.7}$Sr$_{0.3}$MnO$_3$, LSMO) composites with different bulk contents of the components. The specific feature of the composites was that MgB$_2$ is a powder of a micrometer size (5-10 μm), while LSMO is a nanometer powder with an average particle size about 20-30 nm. The homogeneous composition of the samples was obtained by

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mixing the components in alcohol, followed by drying and additional mechanical mixing. From the resulting mixture, plates with the dimension $S = 0.2\times10^{-3}\,\text{mm}^2$ were prepared at pressures of 40-60 kbar. The current and potential measuring contacts were prepared by pressing the plates with fine-dispersed silver in the contact’s area. Resistance of the contact pads was $R = 10\,\Omega$ and was practically independent on the composite composition. As a proof of a homogeneous distribution of the components over a volume of the sample we attract attention to the fact that the resistance of various plates of the same composition is equal, and a linear dependence of the resistance on the length of the plate region, which was obtained with the help of six potential contacts. The samples were not additionally sintered to avoid mutual diffusion and chemical reactions of the components. Thus, the nanocomposites under consideration are random mixtures of two components.

As is known, the electric transport characteristics of composites depend not only on the properties of its constituent elements, but also on the structure of the percolation cluster that appears in the composite [18]. Special case is when the components greatly differ on their conductivity [3]. If an infinite (percolate) conducting cluster over a highly conductive components greatly differ on their conductivity [3] . If an additional mechanical mixing. From the resulting mixture, plates with the dimension $S = 0.2\times10^{-3}\,\text{mm}^2$ were prepared at pressures of 40-60 kbar. The current and potential measuring contacts were prepared by pressing the plates with fine-dispersed silver in the contact’s area. Resistance of the contact pads was $R = 10\,\Omega$ and was practically independent on the composite composition. As a proof of a homogeneous distribution of the components over a volume of the sample we attract attention to the fact that the resistance of various plates of the same composition is equal, and a linear dependence of the resistance on the length of the plate region, which was obtained with the help of six potential contacts. The samples were not additionally sintered to avoid mutual diffusion and chemical reactions of the components. Thus, the nanocomposites under consideration are random mixtures of two components.

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In the LSMO/MgB$_2$ composite, the high-resistive phase is LSMO and the low-resistive phase is MgB$_2$. By varying the concentration ($p$) of LSMO, it enables to define a threshold ($p_c$) above which the transport characteristics of the composite are determined by the current flow through chains – LSMO – MgB$_2$ – LSMO – MgB$_2$ –. For the samples with $p > p_c$, the observation of a superconducting current is the evidentiary fact of the superconducting proximity effect in manganite LSMO.

III. EXPERIMENTAL RESULTS

Fig. 1 shows the dependence of the LSMO/MgB$_2$ nanocomposite resistivity on the volume content of LSMO at temperature $T = 300\,\text{K}$. A rapid change in the sample’s resistivity occurs in the region of 20% volume concentration of LSMO. The temperature dependence of the resistance for pure MgB$_2$ and a composite with 10% and 26% LSMO volume content is shown in the inset in Fig. 1. It can be seen from the figure that already 10% of LSMO substantially broadens the resistive superconducting $R(T)$ transition of the composite as compared to the pure MgB$_2$ sample. The resistance of the sample with 26% LSMO in MgB$_2$ remains finite throughout the temperature range under study, although the onset of the $R(T)$ decrease coincides with the $T_c$ of pure MgB$_2$. Note that the resistance-versus-temperature dependences were taken from transport current values that do not affect the $R(T)$ transition behavior.

Based on the traditional models [18] of a percolation transition in the sense of geometric contact, small additions of LSMO can not affect the formation of a MgB$_2$ infinite cluster since in the case of equal-size components its formation is achieved already at 20% of magnesium diboride. However, in our case, the percolation path per MgB$_2$ is disturbed due to a significant difference in the particle sizes of MgB$_2$ and LSMO (5-10 µm and 20-30 nm, respectively). Indeed, due to the mixing and pressuring, the manganite nanoparticles envelop the MgB$_2$ microcrystals. As a result, a direct contact of the MgB$_2$ granules is eliminated and the resistance of the composite is determined by the – LSMO – MgB$_2$ – LSMO – MgB$_2$ – chains, which leads to a significant broadening of the $R(T)$ curve.

The results of the sample density measurements after pressing support this assumption. The density of MgB$_2$ plates obtained at pressures of 40 - 60 kbar was (72 ± 3)% of the single crystal MgB$_2$. The density of the compacted LSMO nano-powder at the same pressures was only (68 ± 3)% of the LSMO; the density of the MgB$_2$ - LSMO composite (26% of LSMO) was (96 ± 3)% of the calculated density. Such high density of the composite points that the LSMO nano-powder under high uniaxial pressures "spreads" throughout the sample volume, filling the pores around large MgB$_2$ granules.

Fig. 2 shows the I-V characteristics of the LSMO/MgB$_2$ samples (10% and 26% of LSMO). The critical current, $I_c$, is
observed at 10% of LSMO, but for 26% of LSMO the I-V dependence does not exhibit the presence of a critical current. Yet, it has a shape typical for a medium of weakly coupled contacts with an excess current, i.e., the current in the superconducting state exceeds the current in the normal state. These results most naturally can be interpreted in terms of a 3D Josephson medium - a bulk of a large number of Josephson S-N-S contacts.

An effective way to show the nature of the current flow in the composite is the response of its transport characteristics on a hydrostatic pressure. It is well known that for any structure of a grid of weakly connected S-N-S contacts their superconducting characteristics will be improved under the pressure effect. It follows from the exponential dependence of a current through a weak connection from its thickness $d$, $I \approx I_c \exp(-d/\xi_n)$, $\xi_n$ is the coherence length of the N layer.

We carried out experiments on high hydrostatic pressures effect on the temperature dependence of the superconducting transition and the critical current of the nanocomposites. Samples of LSMO/MgB$_2$ nanocomposites with a volume concentration of 26% LSMO were placed in a high-pressure chamber of a piston-cylinder type. The pressure transfer medium was a kerosene-oil mixture. In addition to the composite sample, a platinum wire temperature sensor and a pressure sensor were placed in the chamber.

![Image](image_url)

Figure 3. Effect of hydrostatic pressure on the temperature dependence of the resistivity of a nanocomposite (26% LSMO). Inset: the effect of pressure on the temperature dependence of the MgB$_2$ resistance.

In Fig. 3, the resistivity-versus-temperature dependences are shown for the sample LSMO/MgB$_2$ (26% LSMO) at zero (atmosphere) and P = 12 kbar pressures. Under the influence of pressure, the beginning of the transition shifted toward low temperatures, and the smearing region, starting from the middle of the R(T) transition, shifted to high temperatures. Such pressure effect on the temperature dependence of the composite resistance is determined by two factors – a negative value of $dT_c/dP$ for MgB$_2$ and a response of the intergranular regions to the pressure. The inset in Fig. 3 shows the influence of pressure on the dependence of the superconducting R(T) transition for pure MgB$_2$. Under the effect of pressure, the critical transition temperature drops with a rate of $dT_c(P)/dP \approx 0.1$ K/kbar. Note that the $dT_c/dP$ value for MgB$_2$ obtained for our compacted samples is in the complete agreement with the data given in Ref. [19] for pure magnesium diboride. The decreased smearing of the R(T) curve for the composite appears due to the decrease in the resistance of the LSMO grains in the MgB$_2$-LSMO-MgB$_2$ contacts, which is reflected in the decrease of the composite total resistance under pressure. The same result was also displayed in the I-V characteristics of MgB$_2$ and nanocomposite samples. In Fig. 4, dependence of the nanocomposite I-V characteristics on the applied pressure are shown. Under pressure, the critical current of the composite increased, i.e., the resistance of the sample decreased. The inset in Fig. 4 shows the dependence of the I-V characteristic for pure MgB$_2$. It can be seen that in this case the behavior is opposite: the critical current decreases under pressure. Thus, the experiments on the pressure effect on the LSMO/MgB$_2$ nanocomposite transport characteristics prove the formation of a structure of weakly coupled S-N-S type contacts in the composite.

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Figure 4. The effect of a hydrostatic pressures on the current-voltage characteristic of a nanocomposite (26% LSMO). Inset: the effect of pressure on the current-voltage characteristic of MgB$_2$.

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Figure 5. Effect of a high-frequency irradiation on the resistive superconducting transition of the LSMO/MgB$_2$ composite (26 % LSMO).

As is known, in the ferromagnetic phase, manganites are the so-called half-metals, in which carriers with only one direction of spins present at the Fermi level. For such
compounds, one cannot expect the realization of the classical s-wave proximity effect because there is no possibility to form a Cooper pair with zero total spin. However, if the surface layer of a manganite is spin-active, it provides possibility to transform Cooper pairs with the s-wave symmetry of the order parameter into Cooper pairs with the p-wave symmetry [1,20-22]. Then through the S-hmF-S contacts a spin-polarized current of p-wave Cooper pairs can flow. It is of interest to study dynamic characteristics of such contacts. In particular, as is known, in conventional S-N-S contacts the formation of so-called Josephson vortices - closed superconducting ring currents – takes place [23]. Unlike Abrikosov vortices, Josephson vortices do not have a normal core. Therefore, the frequency range of the electromagnetic signal that causes a response in typical characteristics of the Josephson medium is very wide and is limited only by the superconducting energy gap.

We have investigated the effect of a high-frequency irradiation, \( f = 50 \text{ MHz} \), on the composite I-V characteristics. The irradiation was supplied from the main output of the generator G4-102 with a voltage \( V_{\text{max}} = 0.5 \text{ V} \). The signal has been fed to the sample through a capacitance \( C = 0.1 \mu \text{F} \) along a thin measuring wire.

Fig. 5 illustrates the effect of a high-frequency signal of different amplitudes on the resistive superconducting transition of the LSMO/MgB\(_2\) nanocomposite (27% LSMO). As the amplitude of the applied signal increases, the superconducting transition curve becomes smeared, but the beginning of the transition does not shift. For the pure MgB\(_2\) sample the effect of the high-frequency signal on the critical current value and the superconducting transition has not been detected. It indicates a nonthermal nature of the electromagnetic signal effect on the R(T) characteristic of the nanocomposites under study.

The irradiation effect on the I-V characteristic of a nanocomposite sample is shown in Fig. 6. With increasing of external electromagnetic irradiation, the critical current of the sample decreases and the I-V dependence demonstrates a resistive state of the sample but with an excess current.

**REFERENCES**