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Anomalous Transport Properties of a Paramagnetic $NiTiO_3$ + HTSC Two-Phase System Representing a Random Josephson Junction Network

M. I. Petrov*, D. A. Balaev, K. A. Shaĭkhutdinov, and S. I. Popkov

Kirenskiĭ Institute of Physics, Siberian Division, Russian Academy of Sciences, Krasnoyarsk, 660036 Russia

**e-mail: smp@iph.krasnoyarsk.su*

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Composites representing a network of random Josephson junctions and characterized by the compositions 92.5 at. % $Y_{3/4}Lu_{1/4}Ba_2Cu_3O_7$ + 7.5 at. % $NiTiO_3$ and 92.5 at. % $Y_{3/4}Lu_{1/4}Ba_2Cu_3O_7$ + 7.5 at.% $MgTiO_3$ are synthesized, and their magnetoresistance properties are studied. The temperature dependence of the resistance $R(T)$ measured for the composite that contains the paramagnetic $NiTiO_3$ compound exhibits a characteristic feature below the superconducting transition temperature T_c of the high-T_c superconductor, namely, a region where R is independent of the current j and weakly depends on the magnetic field H. Below a certain temperature T_m, a strong dependence of R on j and H is observed, which is peculiar to a network of Josephson junctions. The dependences $R(T, j, H)$ obtained for the "reference" samples with the nonmagnetic $MgTiO_3$ compound exhibit no such features. The anomalous behavior of the HTSC + $NiTiO_3$ composite is explained by the effect produced by the magnetic moments of Ni atoms in the insulating barriers on the transport current. © *2002 MAIK "Nauka/Interperiodica".*

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Josephson structures with different types of magnetic ordering in the barriers (superconductor–ferromagnet–superconductor or superconductor–paramagnet–superconductor) have been intensively studied on the basis of conventional superconductors for years, both theoretically and experimentally [1–13]. These structures attract the interest of researchers, because they exhibit such effects as the nonmonotone temperature dependence of the critical current [6, 7, 10–12], the presence of π junctions [6, 7, 10–12], the reduction of the superconducting properties, and the characteristic behavior of magnetoresistance [13]. The strong chemical activity of high-T_c superconductors (HTSCs) hinders the fabrication of single Josephson junctions of the aforementioned kinds on their basis. However, the characteristic features of the resistive state of such junctions can be observed on two-phase composites whose one ingredient is an HTSC and the other is a compound with magnetic ordering. Technologically, the fabrication of such composites presents no difficulties. The necessary condition for their synthesis is the absence of strong chemical interaction between the initial ingredients of the composite. Earlier, it was shown [14–18] that, below the transition temperature T_c of the superconducting grains, the transport properties of composite HTSCs prepared by the fast sintering technique represent a Josephson junction network.

We studied composite samples with the volume content of the HTSC 92.5 vol % and the content of $NiTiO_3$ (or $MgTiO_3$) 7.5 vol %. The samples were prepared as follows. First, the initial components were synthesized. The $Y_{3/4}Lu_{1/4}Ba_2Cu_3O_7$ superconductor was synthesized by the conventional ceramic technology. The $NiTiO_3$ and $MgTiO_3$ compounds were obtained from NiO, MgO, and TiO_2 (high purity grade) also by the ceramic technology, by heating at 1250°C within 100 h with five intermediate grindings. Then, the initial components of the future composite were taken in the necessary proportion and grinded and mixed in an agate mortar, after which they were pressed into pellets. The pellets were placed for 2 min in an furnace heated to 910°C and then placed in another furnace heated to 350°C, in which they were annealed within 3 h to saturate them with oxygen. After this, the pellets were cooled together with the furnace. For the samples used in our study, we introduce he following notation: S+7.5(Ni) for composites with $NiTiO_3$, and S+7.5(Mg) for composites with $MgTiO_3$. Since the $MgTiO_3$ compound is nonmagnetic and isostructural with $NiTiO_3$, the S+7.5(Mg) composites were used as reference to reveal the effect of the magnetic moments of nickel.

Magnetic measurements performed for the synthesized $NiTiO_3$ showed that this material is an antiferromagnet with the Néel point ~ 22 K and, above 22 K, it is a paramagnet. The experimental value of the effective moment per atom was found to be ~4 μ_B. These data agree well with the results obtained in [19].

The X-ray structure studies of S+7.5(Ni) and S+7.5(Mg) composite samples revealed the presence of only two phases (within the accuracy of the X-ray analysis): the 1-2-3 structure and the ilmenite structure. No foreign reflections were observed. The relative intensities of the reflections correspond to the volume contents of the composite components. The measurements of the temperature dependences of magnetization *M*(*T*) for the S+7.5(Ni) and S+7.5(Mg) composite samples showed that both composites have a single superconducting phase with the transition temperature T_c = 93.5 K, which coincides with T_c of the initial polycrystalline $Y_{3/4}Lu_{1/4}Ba_2Cu_3O_7$ HTSC, which was annealed in the same way as the composites.

Figure 1 presents the dependences *R*(*T*) measured for the S+7.5(Mg) composite by the four-probe method at different strengths of the external magnetic field (the magnetic field is perpendicular to the current) (Fig. 1a) and at different values of the transport current *j* (Fig. 1b). The behavior of these dependences is typical of the composites with an insulator, such as HTSC + CuO [15] and HTSC + Pb_2ScTaO_6 [18], with a short sintering time. The onset of the superconducting transition is observed at T_c = 93.5 K for all transport currents in small magnetic fields (<1 kOe), which coincides with T_c of the initial HTSC. At this temperature, the superconducting transition of the HTSC grains is observed in the form of a sharp decrease in the resistance. The magnitude of the resistivity drop is independent of both the external field and the current and is determined by the volume content of the superconducting component in the composite. The inset in Fig. 1b shows (on an enlarged scale) the dependences *R*(*T*, *j*) for the composite. One can see that all these dependences are functions of the transport current (the *I*–*V* characteristics are nonlinear) immediately below the superconducting transition temperature. The dependences *R*(*T*, *H*) (Fig. 1a) behave in a similar way. The broadening of the superconducting transition in magnetic fields 1–60 kOe is related to the penetration of the magnetic field into the superconducting crystallites. As the temperature decreases further, the resistance smoothly decreases, which corresponds to the transition of the network of Josephson weak links. The temperature at which the resistance becomes zero decreases with increasing current and magnetic field. These dependences, *R*(*T*, *j*) and *R*(*T*, *H*), can be described in terms of the thermally activated phase slippage mechanism [20], as was done for HTSC + CuO composites in [15].

Figure 2 shows the temperature dependences of the resistance *R*(*T*) of the S+7.5(Ni) composite for different strengths of the external magnetic field (0–60 kOe) (Fig. 2a) and different transport currents *j* (Fig. 2b). The onset of the superconducting transition looks similar to that observed for the S+7.5(Mg) samples. However, as the temperature decreases, the dependences *R*(*T*) exhibit a plateau (see the inset in Fig. 2b) within which the dependences *R*(*T*, *j*) are not functions of the current (the *I*–*V* characteristics are linear) and *R* weakly

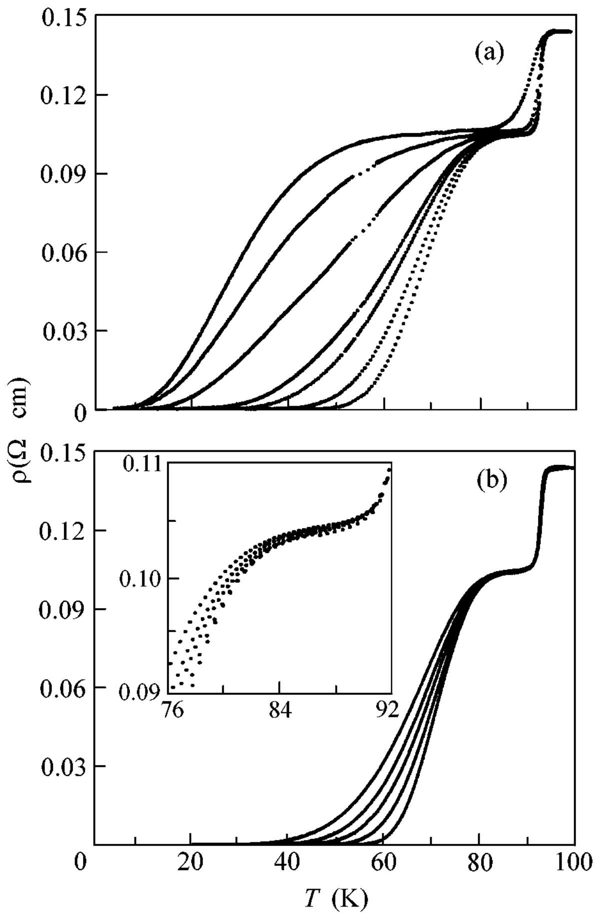

Fig. 1. Temperature dependences of the resistivity of an S+7.5(Mg) sample (a) for different strengths of the external magnetic field *H* at *j* = 27 mA/cm^2 and (b) for different values of the current density *j* at *H* = 0: (a) *H* = 20 Oe, 38 Oe, 80 Oe, 116 Oe, 1 kOe, 10 kOe, and 60 kOe (from right to left); (b) *j* = 27, 135, 270, 407, and 520 mA/cm^2 (from right to left).

depends on the magnetic field. Only starting from a certain temperature T_m do the dependences *R*(*T*) become functions strongly dependent on both the magnetic field (Fig. 2a) and the current (Fig. 2b), as in the case of the S+7.5(Mg) composites. In high magnetic fields (<10 kOe), the point T_m becomes spread, but no displacement of this temperature is observed. The latter is confirmed by Fig. 3, which presents the temperature dependence of the derivative *dR*/*dT*. As the transport current and the magnetic field increase, the temperature corresponding to the zero resistance value decreases. We also note that the dependences *R*(*T*, *H*) and *R*(*T*, *j*) behave in different ways, which is presumably related to the effect of the distribution function of the parameters of the Josephson junctions formed in the composites on the behavior of *R*(*T*). The dependences *R*(*T*, *j*) for the S+7.5(Ni) composite (Fig. 2b) cannot be described in the framework of the approach proposed in [20] and used for the HTSC + CuO composites in [15].

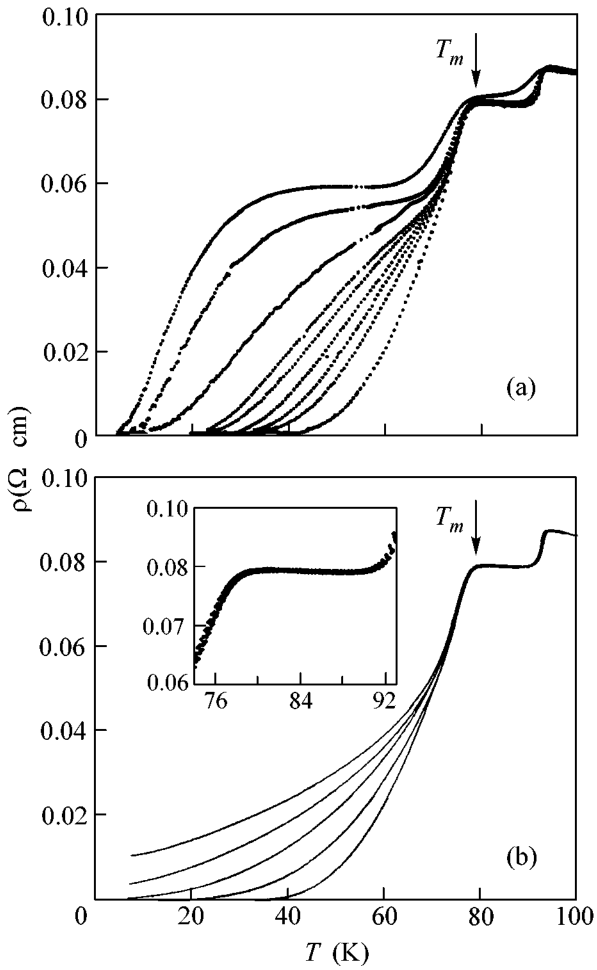

Fig. 2. Temperature dependences of the resistivity of an S+7.5(Ni) sample (a) for different strengths of the external magnetic field H at $j = 20$ mA/cm^2 and (b) for different values of the current density j at $H = 0$: (a) $H = 0$, 20 Oe, 38 Oe, 80 Oe, 116 Oe, 200 Oe, 1 kOe, 10 kOe, and 60 kOe (from right to left); (b) $j = 20$, 100, 200, 300, and 400 mA/cm^2 (from right to left).

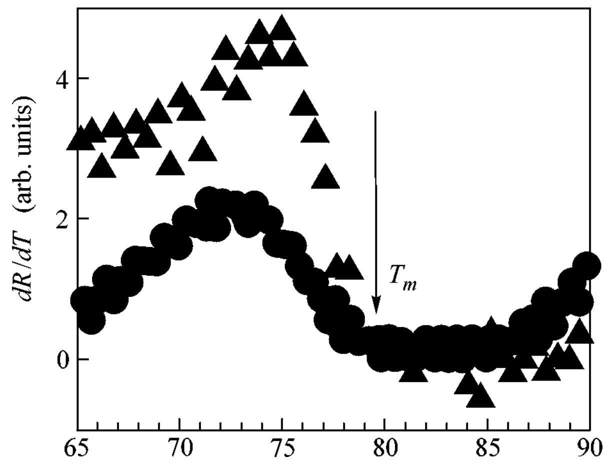

Fig. 3. Fragments of the temperature dependences of the derivative dR/dT for the dependences obtained at $H = 0$ (triangles) and 60 kOe (circles) for an S+7.5(Ni) sample.

The appearance of the aforementioned feature for the S+7.5(Ni) composites, i.e., the appearance of the temperature T_m, cannot be explained by any kind of chemical mechanism (this statement is confirmed by the S+7.5(Mg) sample used as reference). Taking into account that both types of samples were prepared by identical procedures, one can conclude that the feature in question is associated with the use of the $NiTiO_3$ paramagnet as the second component of the composite. It is the presence of magnetic moments of nickel atoms that gives rise to the anomalous behavior of the dependences $R(T)$.

This behavior can be explained by the mechanism similar to that proposed in [21, 22]. The cited publications report on the resistance measurements along the c axis of $YBa_2Cu_3O_7$ [21] and $Bi_2Sr_2CaCu_2O_8$ [22] single crystals placed in a strong magnetic field up to 18 T coplanar with the a–b plane of the crystal. A broadening of the resistive transition was observed, and the curves $\rho(T)$ showed two characteristic regions: one immediately below T_c with ρ independent of j and the other below a certain point T_m with ρ not being a function of the current. The authors of these publications [21, 22] believe that the point T_m is the melting temperature of the Abrikosov vortex lattice. Evidently, in the aforementioned geometry ($H \parallel a, b$), the magnetic field penetrates into the nonsuperconducting layers of the quasi-two-dimensional single crystal to a greater extent than into the superconducting layers with the generation of Abrikosov vortices. When the current flows along the c axis, the charge carriers tunnel from the superconducting layers through the nonsuperconducting ones, in which the spins of the carriers interact in the Zeeman way with the external field. In addition, Abrikosov vortices can move, and, at temperatures above T_m, they move without pinning, which leads to a specific form of the temperature dependence of magnetoresistance.

Our HTSC + $NiTiO_3$ composite can be considered as a macroscopic analog of the experiment described above. In fact, the charge carriers tunnel between the superconducting grains through the paramagnet in which the spins of the carriers become involved in the exchange interaction (an approximate analog of the Zeeman interaction) with the magnetic moments of nickel. The probability of such an interaction is close to unity, because $a_0 < \xi_0$, where a_0 is the lattice constant of $NiTiO_3$, i.e., the distance between the magnetic moments, and ξ_0 is the coherence length. The paramagnetic material $NiTiO_3$ induces a fluctuating effective magnetic field (evidently, its time average strength is equal to zero), which penetrates into the superconducting grains to the depth λ (~1000 Å) [23]. This field causes the formation of Abrikosov vortices in the nearsurface layer of the HTSC adjacent to $NiTiO_3$. In the interval between the melting temperature of the Abrikosov vortex lattice T_m and the superconducting transition temperature T_c, the resistance of the sample does not depend on the transport current, as in the case of [21, 22]. Below this interval, the Abrikosov vortices are pinned inside the HTSC grains. In this case, the *I–V* characteristic depends on the transport current density and a strong dependence of the resistance on the magnetic field takes place (Fig. 2).

We note that the dependences we obtained resemble the results reported in a recent publication [13] where the transport properties of single Josephson junctions with conventional superconductors, Nb/Al/Gd/Al/Nb (superconductor–ferromagnet–superconductor), were studied and similar dependences $R(T)$ were obtained. Below the superconducting transition temperature, these dependences also displayed two characteristic regions. Immediately below T_c, the dependences $R(T)$ in [13] did not depend on the transport current, and, only starting from a certain temperature (analogous to T_m), the curves $R(T)$ were found to depend on j. However, the effect of the ferromagnetism of Gd on the tunneling of charge carriers through this kind of barrier was unfortunately not analyzed in [13].

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REFERENCES

1. I. O. Kulik, Zh. Éksp. Teor. Fiz. **49**, 1211 (1965) [Sov. Phys. JETP **22**, 841 (1966)].
2. H. Shiba and T. Soda, Prog. Theor. Phys. **41**, 25 (1969).
3. L. N. Bulaevskiĭ, V. V. Kuziĭ, and A. A. Sobyanin, Pis’ma Zh. Éksp. Teor. Fiz. **25**, 314 (1977) [JETP Lett. **25**, 290 (1977)].
4. J. L. Paterson, J. Low Temp. Phys. **35**, 371 (1979).
5. J. Niemeyr and G. von Minnigerode, Z. Phys. B **36**, 57 (1979).
6. L. N. Bulaevskiĭ, A. I. Buzdin, and S. V. Panjukov, Solid State Commun. **44**, 539 (1982).
7. A. I. Buzdin, B. Bujicic, and M. Yu. Kupriyanov, Zh. Éksp. Teor. Fiz. **101**, 231 (1992) [Sov. Phys. JETP **74**, 124 (1992)].
8. S. V. Kuplevakhskiĭ and I. I. Fal’ko, Fiz. Tverd. Tela (St. Petersburg) **34**, 183 (1992) [Sov. Phys. Solid State **34**, 96 (1992)].
9. A. S. Borukhovich, Usp. Fiz. Nauk **169**, 737 (1999).
10. M. Fogelström, Phys. Rev. B **62**, 11812 (2000).
11. Y. Tanaka and S. Kashiwaya, J. Phys. Soc. Jpn. **69**, 1152 (2000).
12. V. V. Ryazanov, V. A. Oboznov, A. Yu. Rusanov, *et al.*, Phys. Rev. Lett. **86**, 2427 (2001).
13. O. Bourgeois, P. Gandit, A. Sulpice, *et al.*, Phys. Rev. B **63**, 064517 (2001).
14. M. I. Petrov, D. A. Balaev, S. V. Ospishchev, *et al.*, Phys. Lett. A **237**, 85 (1997).
15. M. I. Petrov, D. A. Balaev, K. A. Shaĭkhutdinov, and K. S. Aleksandrov, Fiz. Tverd. Tela (St. Petersburg) **41**, 969 (1999) [Phys. Solid State **41**, 881 (1999)].
16. M. I. Petrov, D. A. Balaev, K. A. Shaĭkhutdinov, and S. G. Ovchinnikov, Fiz. Tverd. Tela (St. Petersburg) **40**, 1599 (1998) [Phys. Solid State **40**, 1451 (1998)].
17. M. I. Petrov, D. A. Balaev, S. V. Ospishchev, and K. S. Aleksandrov, Fiz. Tverd. Tela (St. Petersburg) **42**, 791 (2000) [Phys. Solid State **42**, 810 (2000)].
18. D. Berling, B. Loegel, A. Mehdaoui, *et al.*, Supercond. Sci. Technol. **11**, 1292 (1998).
19. G. S. Heller, J. J. Stickler, S. Kern, and A. Wold, J. Appl. Phys. **34**, 1033 (1963).
20. V. Ambegaokar and B. I. Halperin, Phys. Rev. Lett. **22**, 1364 (1969).
21. M. Charalambous, J. Chaussy, and P. Lejay, Phys. Rev. B **45**, 5091 (1992).
22. M. S. Fuhrer, K. Ino, K. Oka, *et al.*, Physica C (Amsterdam) **282–287**, 2041 (1997).
23. A. P. Malozemoff, in *Physical Properties of High Temperature Superconductors*, Ed. by D. M. Ginsberg (World Scientific, Singapore, 1989; Mir, Moscow, 1990), Vol. I.

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