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METALS AND SUPERCONDUCTORS

Andreev Reflection in Natural Grain Boundaries of Polycrystalline High-T_c Superconductor $La_{1.85}Sr_{0.15}CuO_4$

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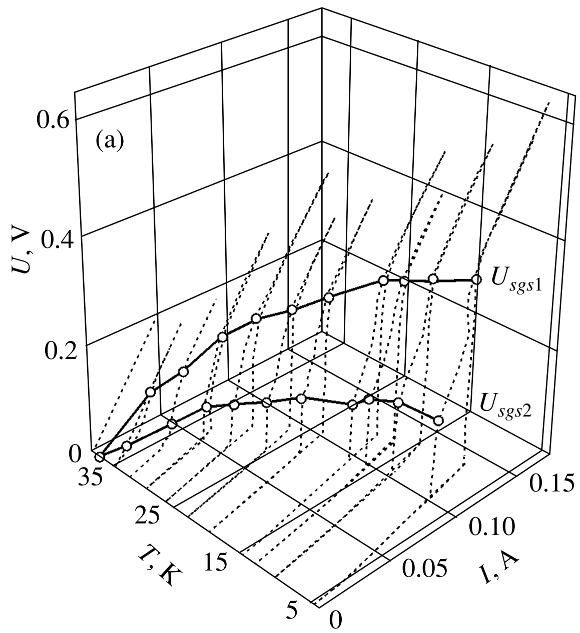
Abstract—The temperature evolution of the current–voltage characteristic (CVC) of a "break junction" with metal-type conductivity on the polycrystalline $La_{1.85}Sr_{0.15}CuO_4$ high-temperature superconductor is investigated. The CVC exhibits gap peculiarities and hysteresis, which is observed in the region of negative differential resistance. The experimental results are described well in terms of the Kümmel–Gunsenheimer–Nicolsky theory for an S–N–S junction (S is a superconductor, N is a normal metal) this theory takes into account multiple Andreev reflection of quasiparticles. It is shown that the shape of the CVC and the existence and the shape of hysteresis are determined by the ratio of "long" and "short" grain boundaries in the polycrystal under investigation. *© 2003 MAIK "Nauka/Interperiodica".*

1. INTRODUCTION

The investigation of the current–voltage characteristic (CVC) of a Josephson junction makes it possible to obtain information on the physical properties of superconductors. The peculiarities of a CVC contain information about the energy gap [1, 2] and may depend on the symmetry of the superconductor order parameter [3]. Since the discovery of high-temperature superconductivity (HTSC), different Josephson structures [4] and polycrystalline high-T_c materials [5–7] in which Josephson medium is realized [8] have been actively studied. Technically, it is very difficult to prepare a single Josephson junction with high-quality superconducting "banks" because of the high chemical activity of high-T_c compounds, and on polycrystalline samples inevitable heating makes it difficult to measure the temperature evolution of a CVC in a wide range of currents, including the range where the CVC becomes linear. Many experimental investigations of the transport properties of polycrystalline superconductors with different compositions have been carried out with the use of break junctions [9–12]. Break-junction technology allows one to decrease the self-heating of a sample significantly. Break junctions prepared on bulk samples require small measuring currents, like films, but they are free from a number of drawbacks inherent to the latter (lower critical temperature, smaller energy gap). While a microcrack develops, the cross section of the sample decreases until only a narrow conducting channel is left and a tunneling junction is formed in the limit. In the first case, the current density flowing through the crystallites in the break region significantly exceeds the current density in the sample volume. Thus, the break region determines the critical current in the whole sample. This fact allows one to use relatively small measuring currents to obtain CVC sections reflecting the gap peculiarities of the superconductor. In the present work, break junction CVCs of a exhibiting a hysteretic behavior are measured on $La_{1.85}Sr_{0.15}CuO_4$ at different temperatures. The first measurements on a polycrystalline sample of this system [13], which represents a network of weak links, were carried out soon after the discovery of HTSC. In the experiment in [13], the CVC of a sample had a number of peculiarities, which probably resulted from the presence of foreign phases and self-heating of the sample. This makes comparison with the theoretical characteristics of weak links junctions extremely difficult. From the presence of excess voltage on the CVC in [13], it follows that the boundaries between superconducting granules in ceramics were probably insulating and, thus, a chaotic network of Josephson junctions was formed in the material. The synthesis technology for high-T_c superconductors of lanthanum and yttrium systems has been significantly improved since the pioneering work performed in [13] and make it possible to provide natural boundaries of metal character between high-T_c superconductor crystallites.

2. EXPERIMENTAL

$La_{1.85}Sr_{0.15}CuO_4$ is prepared using the solid-state reaction technique. Samples with a typical size of $2 \times 2 \times 10$ mm^3 were sawed out from synthesized tablets. The samples were glued onto a sapphire substrate. The central part of a sample was ground down to a cross section $S \sim 0.2 \times 1$ mm^2. A further decrease in S is



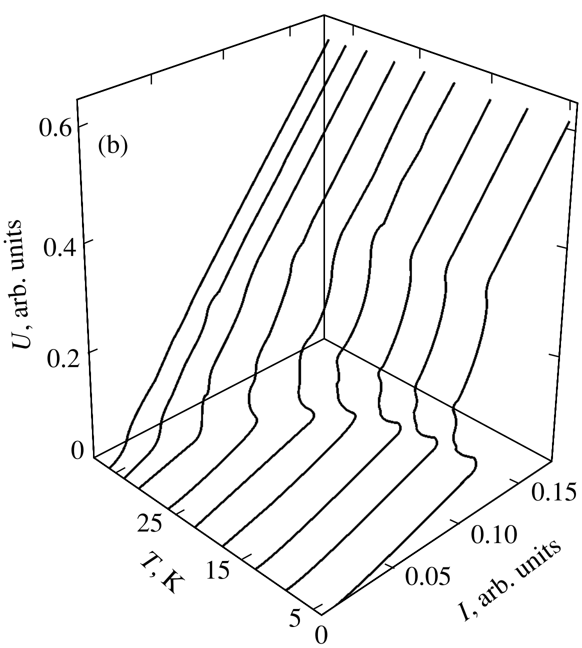


Fig. 1. Temperature evolution of the CVC of a break junction: (a) experiment and (b) theory.

extremely difficult to control because of the inevitable mechanical stresses at current- and potential-lead terminals. To obtain a break junction, a sample with the cross-sectional area *S* mentioned above, together with the substrate, was bent by means of screws on pressed current-lead terminals, which caused a microcrack to appear in the part of the sample between the potential-lead terminals. In this case, either a tunneling junction (resistance $R > 100\ \Omega$) or a junction with metal-type conductivity ($R < 10\ \Omega$) appeared. For the measurements carried out in this work, the samples with the lowest resistance were chosen. During the measurements, samples were held in a heat-exchange helium atmosphere. CVC measurements were carried out under isothermal conditions by slowly scanning a bias current.

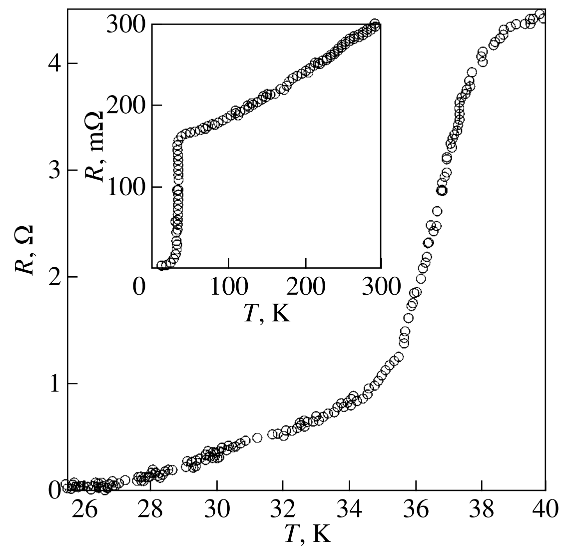


Fig. 2. Temperature dependence of the break-junction resistance. Inset: temperature dependence of the resistance of a bulk sample.

3. RESULTS AND DISCUSSION

The temperature evolution of the CVC of a break junction on $La_{1.85}Sr_{0.15}CuO_4$ is shown in Fig. 1a. All CVCs are characterized by the presence of a critical current and a region with a small differential resistance; at low temperatures, this region is followed by a hysteretic jumplike increase in voltage *U*. In the region of high values of current *I* and *U*, the *U*(*I*) dependence is close to linear; its extrapolation to the value $U = 0$ gives the value of excess current I_{ex}, the existence of which confirms the metallic character of the conductivity of the junction under investigation [14]. The hysteretic peculiarity of a CVC obtained in the current-scanning mode is often observed on S–N–S junctions [5, 7, 14]. Such a peculiarity was shown in [15] to appear if there is a region of negative differential resistance (NDR) on a CVC; this region can be observed only in the bias voltage regime on an S–N–S junction.

Figure 2 presents the temperature dependence of the resistance *R*(*T*) of the break junction. The inset to Fig. 2 shows the *R*(*T*) dependence measured up to 300 K before the break formation. The linear character of the *R*(*T*) dependence above T_c confirms the metal type of conductivity of the sample. A comparison of the resistance *R* of the sample just above, superconducting the transition temperature before (0.15 Ω) and after the microcrack appearance (4 Ω) indicates that the contact area decreased by approximately 27 times. After the break junction was created, the temperature at which the resistance disappears became 2.5 K lower than that in a bulk sample. It is known that thermal fluctuations in weakly coupled superconductors (thermally activated phase slip [16]) decrease the transition temperature from a resistive state into the state with zero resistance. The dispersion of the parameters of individual weak-link junctions leads to dispersion of the temperatures at which the resistances of separate weak-link junctions disappear. In a bulk sample, current flows through the "best" weak links and the influence of the dispersion of these parameters is insignificant. In a break junction, the majority of percolation paths are broken up; therefore, "poorer" weak links (with smaller critical currents and lower temperatures at which the resistance disappears) begin to influence the transport characteristics, in particular, to decrease the temperature at which the resistance of the whole sample disappears. Thus, when a microcrack is formed, the current flows not through a three-dimensional network but rather through a network of a smaller dimensionality.

Consider a chain of weak links connected in series which have different thickness of normal-metal region between the superconductive banks. The current–voltage relation for this chain is

$$U(I, T) = \sum_i V_i U_i(I, T, S_i, d_i), \tag{1}$$

where $U_i(I, T, S_i, d_i)$ is the CVC of an individual S–N–S junction whose N layer has a thickness d_i and cross section S_i and V_i is a weighting coefficient showing the degree of influence of this junction on the resulting CVC of the chain ($\sum_i V_i = 1$). In the model under investigation, the dispersion of cross sections is ignored and $S_i = S$ (the dispersion of cross sections, as well as the presence of parallel-connected junctions, smears the of the CVC).

There are several theories which could be applied to calculate $U_i(I, T, d_i)$ of a single S–N–S junction. The RSJ model and its modifications [17–19], in our opinion, cannot adequately describe the physical processes operating in S–N–S junctions. The current flowing through an S–N–S junction and the CVC peculiarities accompanying it are determined by the Andreev reflection [20]. Currently, a number of theories [1, 2, 21–23] are used for the description of CVCs of weak-link junctions. Kümmel–Gunsenheimer–Nicolsky theory (KGN) [2], unlike the other theories, describes the appearance of an NDR region in the CVC of an S–N–S junction observations; the other theories do not take into account the contribution from bound states in the S–N–S junction to the current [24]. The KGN theory deals with weak-link in which the Fermi velocities in the superconductor and the normal metal are equal. We assume that high-T_c ceramics meet this requirement and, thus, the KGN theory can be used to calculate $U_i(I, T, d_i)$ in Eq. (1). The KGN theory is also convenient because in this theory the ratio d_i/l (where l is the mean free path of electrons in the N metal) is used as the weak-link parameter determining the CVC shape. The current flowing through a weak link in the KGN theory [2] is given by

$$I = \frac{1}{d}\frac{e}{m}\sum_k \sum_{n=1}^{\infty} P_N(E_k) \tag{2}$$
$$\times \{[f(E_k)k_e - (1 - f(E_k))k_h]e^{-nd/l}(|A_n^-|^2 - |A_n^+|^2)\},$$

where $f_0(E_k)$ is the Fermi function describing the energy distribution of quasiparticles, P_N is the probability of the presence of quasiparticles in the N region, e is the electronic charge, m is the electronic mass, n is the number of Andreev reflections undergone by a quasiparticle before it escapes from the quantum well (the normal metal between the superconductors), $A_n^-(E)$ and $A_n^+(E)$ are the probabilities of the nth Andreev reflection of quasiparticles with directions of hole propagation parallel or antiparallel to the electric field, and k_e and k_h depend on the energy and direction of motion of electrons and holes and are determined in [2].

The experimental CVC obtained can be qualitatively described using only one term in sum (1). However, the inclusion of longer junctions (with larger values of d) significantly improves the agreement between the experimental data and the theoretic dependence. With two terms in sum (1), the calculated curve well describes the experimental CVC (Fig. 1). In this case, the best-fit curve corresponds to the values $d_1/l = 0.2$, $V_1 = 0.93$, $d_2/l = 0.6$, and $V_2 = 0.07$. By using our results and the data from review [25], we obtained $l \sim 10$ Å for $La_{1.85}Sr_{0.15}CuO_4$; therefore, $d_1 = 2$ and $d_2 = 6$ Å.

This model allowed us to describe the unusual shape of experimental CVCs. The arch-shaped peculiarity of an experimental CVC corresponds to the last arch-shaped peculiarity on the calculated curve. This peculiarity is due to multiple Andreev reflection in the S–N–S junction. According to the theories mentioned above, multiple Andreev reflection of quasiparticles leads to the appearance of a subharmonic gap structure on the CVC of the S–N–S with minima at $U = 2\Delta(T)/en$, where Δ is the energy gap of the superconductor. The last arch-shaped peculiarity corresponds to $n = 1$ and 2.

In [3], the authors come to the conclusion that, in the case of d-wave symmetry of electron pairs in the superconductor, the peculiarities of the CVC of a weak-link junction that correspond to subharmonics of the energy gap are heavily suppressed. The arch-shaped peculiarities distinctly visible on our CVCs probably confirm that the symmetry of the superconducting order parameter is different from the d-wave symmetry.

The literature data on the symmetry and temperature dependence of the energy gap in a high-T_c superconductor are contradictory (see, e.g., reviews [26–30]). Special points $U_{sgs1}(T)$ and $U_{sgs2}(T)$, marking the arch-shaped peculiarity, are shown on the experimental CVCs in Fig. 1. The relations $U_{sgs1}(T) = 2\Delta(T)/e$ and $U_{sgs2}(T) = \Delta(T)/e$ are not strictly satisfied because the current flows through several weak-link junctions. However, the proportionality to $\Delta(T)$ should remain for these special points. The observed $U_{sgs1}(T)$ and $U_{sgs2}(T)$ dependencies are slightly different from the temperature dependence of the energy gap in the BCS theory.

In polycrystalline high-T_c superconductors, crystallites are also distributed in orientation [8] and, due to a strong anisotropy of these crystallites, there is a dispersion of energy gap values on a current-flow path. One can simply, but not sufficiently correctly, take this dispersion into account by substituting different energy gap values into the KGN equation for different terms in Eq. (1). This operation improves the agreement of the theoretical curves with the experimental CVCs only insignificantly, but the number of fitting parameters increases in this case. It should be noted that the thickness distribution function of grain boundaries and the energy-gap distribution function of crystallites on a current-flow path may be related because of the peculiarities of ceramic synthesis. Our further investigations will be devoted to this issue.

4. CONCLUSIONS

Thus, we have successfully described both the CVC shape with a hysteretic peculiarity and its temperature evolution by means of the KGN theory [2], which takes into account multiple Andreev reflection. This allows us to conclude that, for natural grain boundaries of a metallic type in the polycrystalline high-T_c superconductor $La_{1.85}Sr_{0.15}CuO_4$, Andreev reflection determines the characteristic features of the current-voltage curve.

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