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TWO-DIMENSIONAL BEHAVIOR OF YBa₂Cu₃O_x FILMS ON SrTiO₃ SUBSTRATES

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Long-term (up to 18 months) degradation of textured YBa₂Cu₃O_x films prepared by laser ablation and magnetron sputtering on single crystal SrTiO₃, MgO and Al₂O₃ substrates were investigated. All films at each stage of exposition displayed a clearly visible quasi-2D behavior. From the measurements of the Kosterlitz-Thouless temperature it could be concluded that the oxygen content and density of charge carriers remained constant. It follows that the main consequence of degradation is the omission of some planes from the transport of supercurrent and the samples could be described in the model of layered structure with variable number of conducting planes. Using this model we find that effective thickness of an isolated CuO₂ plane in samples with greatest internal damage is $d \approx 2$ Å.

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1. Introduction

One of the more detrimental effects, which may occur in high temperature superconducting (HTSC) films, is their metastability at room temperature. As it was found by direct X-ray diffraction studies of textured YBa₂Cu₃O_x films [1], during exposition to the atmosphere they partially undergo a transformation into Cu-rich compounds Y₂Ba₄Cu₇O₁₅ and YBa₂Cu₄O₈. The 50 percent decomposition time of 123 compound was found to be approximately 4 years.

We propose to give a very simple explanation of this process. As we see it, the main effect of the appearance of 247 and 124 inclusions is the violation of continuity of some of the CuO_2 planes. From this point of view one would expect a decrease in the critical current, an increase in the sample normal state resistance (R) and a decrease in resistance slope with the temperature $\mathrm{d}R/\mathrm{d}T$. This is just what is observed experimentally. While other phenomena could produce the same effects, the main difference is in the expected quasi-two-dimensional (2D) behavior.

Quasi-two-dimensional behavior is the common feature of all HTSC, including YBa₂Cu₃O_x — the one with the smallest anisotropy. The simplest 2D effect is the Berezinskii–Kosterlitz–Thouless (BKT) transition [2–5]. When the Josephson interaction between superconducting planes of a layered structure is weak, which is the case of HTSC samples with discontinuous superconducting planes [6], surrent–voltage (I-V) characteristics of the sample can be approximated by the expression $V \propto I^n$, where the power exponent n remains equal to 1 even in the superconducting state, at temperatures lower than the mean field critical temperature $T_{\rm mf}$. Only at the temperature of Berezinskii–Kosterlitz–Thouless transition T_{KT} n undergoes a jump to n=3 and then, with the decrease in temperature, changes linearly with $\Delta T = T - T_{\rm mf}$. The slope of $n(T - T_{\rm mf})$ and the difference $\Delta T_{\rm cc}$ between $T_{\rm mf}$ and $T_{\rm KT}$ reflects the level of two-dimensionality. More isolated active planes (separated by the wrecked planes) must have a lower $n(T-T_{\rm mf})$ slope and a larger $\Delta T_{\rm cc}$.

2. Experimental results and their discussion

We have investigated a long-term (up to 18 months) degradation of textured $YBa_2Cu_3O_x$ films prepared by laser ablation and magnetron sputtering on single crystal $SrTiO_3$, MgO and Al_2O_3 substrates.

The experimental method relied on the measurement of current-voltage characteristics. The characteristics were registered in the temperature interval between 100 and 4.2 K by the standard four-probe technique. Precautions were taken in order to avoid the possible heating effects arising from a non-ideal heat removal from the samples. We monitored the difference between I-V curves registered at various rates of current sweep (each current range from 2 A to 20 μ A could be swept in a time interval between 228 s to 0.06 s), and eliminated from the analysis these branches of I-V curves which did not coincide at low rates. In order to avoid heat flow from the current leads we have elaborated good quality contact pads and maintained sufficient distance between current and potential leads. After the measurement I-V curves were plotted in double-logarithmic scale to determine the power exponent n.

We found that all the films at each stage of exposition displayed a clearly visible quasi-2D behavior with a smeared step in the temperature dependence n(T). The difference $\Delta T_{\rm cc}$ between $T_{\rm mf}$ and $T_{\rm KT}$ was smallest for films prepared by laser ablation on SrTiO₃ substrates and largest for films on MgO prepared by magnetron deposition. The Nelson-Kosterlitz jump for films on MgO and Al₂O₃ was badly smeared and rendered the investigation of the evolution of 2D behavior very difficult. For this reason the main analysis was made on YBa₂Cu₃O_x film prepared by laser ablation on SrTiO₃ substrate.

In Fig. 1 we show the $n(T/T_{\rm mf})$ dependence for one of the typical films on SrTiO₃ substrate after different times of air exposition. This sample has a rather pronounced quasi-2D behavior. Plot (a) was obtained just after the deposition. The normalization temperature $T_{\rm mf}$ in this case is 86.5 K. Let us observe that ΔT_{cc} immediately after deposition is rather small. The plots marked (b) through (d) were obtained after 6, 9 and 18 months of air exposition, respectively. $T_{\rm mf}$ for

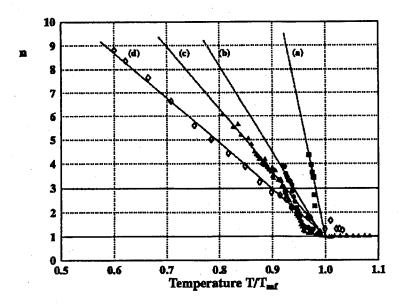


Fig. 1. Power exponent n in function of reduced temperature $T/T_{\rm mf}$.

(b) and (c) is the same as for (a). Only after 18 months of ageing we observed a noticeable decrease in this parameter to $T_{\rm mf} = 78$ K, and plot (d) is normalized to this value. Thin dashed lines in Fig. 1 represent a linear fit to the experimental data in the region n < 3.

Figure 1 shows two main features of the sample's time evolution: a substantial increase in $\Delta T_{\rm cc}$ from 1.6 K to 8 K and a decrease in the slope of the linear part of n(T) from 1.28 K⁻¹ to 0.24 K⁻¹. The increase in $\Delta T_{\rm cc}$ is caused mainly by the rapid decrease in $T_{\rm KT}$ with exposition time, while $T_{\rm mf}$ remained practically constant. From the latter observation one can conclude that the oxygen content and density of charge carriers remained the same too. On the other hand, the $T_{\rm KT}$ value is governed by the distance between the active superconducting planes. It follows that the main consequence of degradation is the omission of some planes from the transport of supercurrent. As a consequence thin films can be described in the model of the layered structure with a variable number of conducting planes [6]. $T_{\rm KT}$ and $T_{\rm mf}$ determine the temperature interval, where the I-V curves are linear, exhibiting no critical current. A larger number of wrecked planes corresponds to a larger interval of linear behavior. Such broadening of the resistive transition renders impossible many practical applications of the HTSC films.

The analysis of the quasi-2D behavior using the above-mentioned model leads to important information about the nature of superconductivity and the thickness d of superconducting layers, closely related to $\Delta T_{\rm cc}$, the difference between $T_{\rm mf}$ and $T_{\rm KT}$ [7].

The relationship between $\Delta T_{\rm cc}$ and d follows from the renormalization of effective 2D thin-film screening length Λ . As was mentioned above, $T_{\rm mf}$ remains constant, but BKT transition takes place at the temperature

$$T_{\rm KT} = \frac{q^2}{4\varepsilon_0} = \frac{\Phi_0^2}{16\pi^2 \Lambda(T_{\rm KT})\varepsilon_0},\tag{1}$$

where $\varepsilon_0 \approx 1$ is the dielectric constant, and q is the effective charge of the pancake vortices.

When the thickness of superconducting layer d is very small with respect to the in-plane coherence length ξ_0 , a good approximation for Λ is given by the slightly modified relation, frequently used for granular and dirty superconductors [8]

$$\Lambda(T) = 2\frac{\lambda^2(T)}{d} = 2\frac{\lambda^2(0)}{d} \frac{\xi_0 + d}{d} \left[\frac{\Delta(T)}{\Delta(0)} \tanh\left(\frac{\Delta(T)}{2kT}\right) \right]^{-1}, \quad d \ll \xi_0, \tag{2}$$

where λ is the London penetration depth and Δ is the superconducting energy gap. We have replaced here the normally present mean free path of charge carriers by d, as the smallest parameter of the sample.

The two-dimensional density of carriers in active superconducting planes n_{2D}

$$n_{\rm 2D} = \frac{d}{\lambda^2} \frac{mc^2}{4\pi e^2} \tag{3}$$

remains constant during the experiment, therefore the term d/λ^2 in Eq. (2) is constant too, and the changes in d affect $T_{\rm KT}$ and $\Delta T_{\rm cc}$ only through the term $(d+\xi_0)/d$. Since d is considerably smaller than ξ_0 (d is of the order of a few angstroms, while $\xi_0 \approx 16$ Å), the changes induced by this term can be rather high. The variation of the effective thickness d with the separation of the active planes is caused by varying interlayer coupling following from the charge carriers exchange [7].

Using the above outlined approximation, we find that the effective thickness of an isolated CuO_2 plane in the samples with the greatest internal damage is $d \approx 2$ Å and the corresponding ΔT_{cc} is about 70 K. It is interesting to note that in the case of two closely-spaced active layers d is increased up to 7 A and ΔT_{cc} reduced to about 50 K. The third layer in a perfect stack makes HTSC films practically 3-dimensional with ΔT_{cc} slightly exceeding 20 K. In a perfect single crystal of YBa₂Cu₃O_x with many thousands of layers ΔT_{cc} is reduced to 0.14 K [9]. In this model ΔT_{cc} increases in a stepwise manner with the number of layers [7]. This is in agreement with the observations on YBa₂Cu₃O_x films composed of a small (1,2,3...) number of unit cells [10].

Additional experiments on films prepared by laser ablation onto Al₂O₃ (sapphire) substrates and magnetron evaporation onto MgO and yttrium stabilised Zr show more broad initial resistive transitions and lower characteristic time of sample degradation.

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