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# Dynamically Enhanced Supercurrents in Granular Superconducting Y-Ba-Cu-O Thin Films

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*Thin superconducting films of Y-Ba-Cu-O with* $T_{c0} \cong 89$ *K have been prepared by the aerosol deposition method. The voltage current characteristic of microbridges (typically* $10 \times 10\ \mu m^2$*) made from the finely granular films, which behave like a network of boundary Josephson junctions, is highly nonlinear and shows besides thermal effects evidence for dynamic enhancement of the supercurrent. Our experimental data are in qualitative agreement with a theory developed for conventional superconductors by Aslamazov and Larkin and also by Tinkham. A value for the electron phonon scattering time has been derived:* $\tau_E \cong 15$ *ps.*

## 1. INTRODUCTION

It is a widely accepted idea, that grain boundaries in granular ceramic superconducting thin films can be considered as Josephson junctions (boundary JJs or BJJs).[1] The geometrical dimension of a JJ is related to the coherence length, which is extremely short in this kind of superconductor, 11–16 Å in the a–b plane, and 1.5–3 Å in the *c*-direction.[2] These small distances are comparable to boundary layers of grain some few atoms thick.[3] With the present state of the art it is very difficult to prepare in a controlled way by nanolithography or the formation of extremely thin sandwich structures a JJ of ceramic material.[4] With the present progress in material processing, however, one is able to prepare thin films with a few BJJs or, as will be described in the following, a network of hundreds of BJJs.

There is a large amount of literature available on experiments with JJs from conventional and ceramic superconductors, as a wide spectrum of applications can be expected or have been realized. From the theoretical point of view, these experiments help to understand the nature of superconductivity and especially of dynamic, nonequilibrium effects. One of these effects is the enhancement of superconducting properties, like the critical



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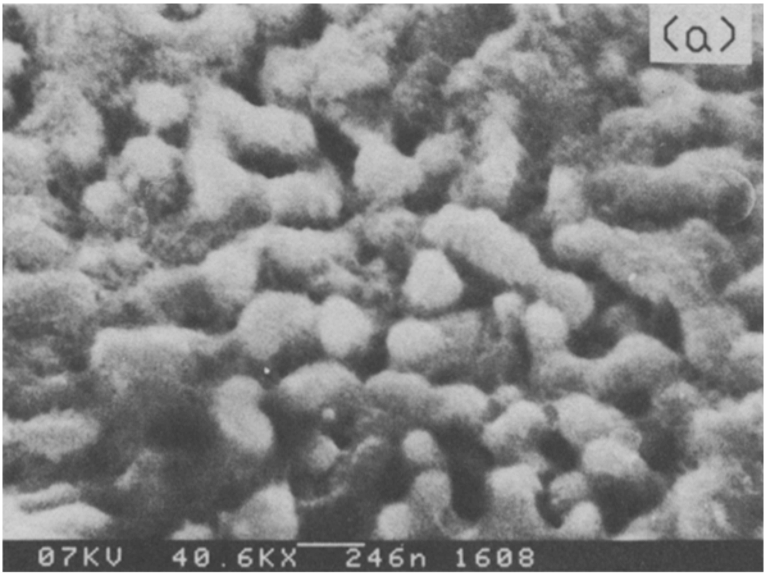
current $I_c$, which has been reported in weak links of conventional superconductors[5–7] and has been explained by a model of Aslamazov and Larkin (AL)[8] and in a more intuitive way by Tinkham[9] using a similar approach.

In the following we present the various pieces of evidence, that we are able to prepare a network of BJJs in a good quality superconducting $YBa_2Cu_3O_{7-x}$ film, which shows an enhancement of the supercurrent and a voltage step in the voltage-current characteristic (VCC). In sec. 3 we compare the AL[8] model with our experimental results. Finally some conclusions are given.

## 2. EXPERIMENTAL RESULTS

Our superconducting thin films have been prepared by the non-vacuum chemical aerosol deposition technology using the solution of metal organic $\beta$-diketonates of Y, Ba and Cu as precursors.[10–12] In the present study we used $SrTiO_3$ (100) as a substrate; the resulting films exhibit, after an anneal heat treatment at 850°C, a $T_{c0}$ and transition width of 89 and 2 K respectively. The film thickness is typically 250 nm and $I_c$ in the order of $10^4$ A/cm$^2$ at 77 K. X-ray diffractometry indicates the orthorhombic perovskite structure with the $c$-axis perpendicular to the substrate. The film is highly granular with a grain size of approximately 150 nm, as can be observed by SEM pictures, see Fig. 1a. For critical current measurements microbridges were fabricated from the superconducting film by standard photo lithography and ion-beam milling. As an example a SEM picture of a $10 \times 10\ \mu m^2$ microbridge with the regular granular structure is given in Fig. 1b.

The first type of measurements were VCCs taken at different temperatures. For these we used a continuous flow $^4$He cryostat (Oxford Instruments CF 1204) operating between 4 and 300 K. The sample was placed on a copper block together with a calibrated RhFe thermometer and a heater inside the sample space, and was cooled by exchange gas ($^4$He at 1 bar). The electrical measurements were done by a four-probe technique using a current source, operating at dc or at low frequency ac ($\leq 100$ Hz). Figure 2 shows as an example a VCC (sample 597-1, $T_{c0} = 86$ K) at a temperature, measured by the RhFe thermometer, of 77 K; the microbridge, however, can be at higher temperature at higher currents, as will be shown below. Three types of critical current can be observed: $I_{c0}$, up to this current no voltage is generated across the bridge, $I_{c1}$, the extrapolation of the linear high current state to zero voltage, and $I_s^*$ and the corresponding $V_s^*$, the coordinates of a sudden voltage step, which exhibits an increasing hysteresis at low temperatures. The curve from $I_{c0}$ up to $I_s^*$ is often referred to as a "foot," the curve above the voltage step can be compared to an RSJ (resistive shunted junction) model calculation,[13] with a shifted asymptote. In this



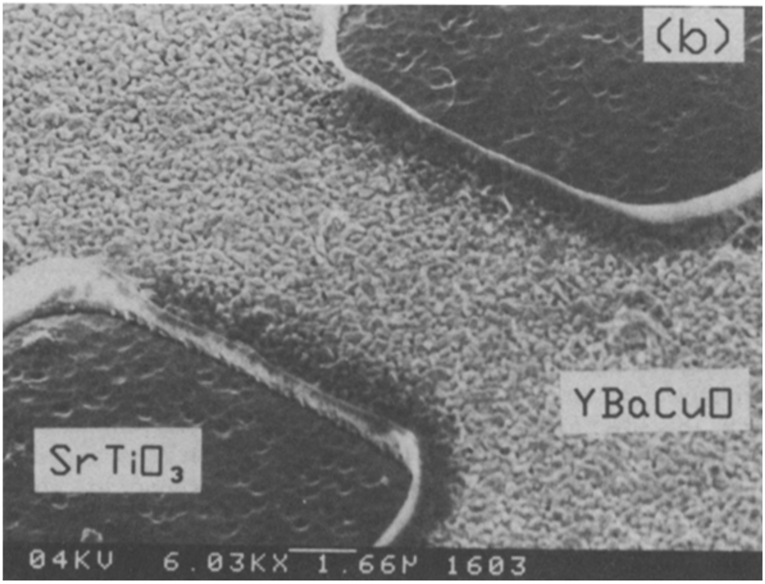




Fig. 1. SEM picture of our highly granular YBaCuO thin film made by aerosol deposition on a $SrTiO_3$ substrate (after heat treatment). (a) detail with large magnification (bar indicates 246 nm). (b) view of a $10 \times 10\ \mu m^2$ microbridge made by ion-beam milling from the same film as in (a) (bar indicates 1.66 $\mu$m).

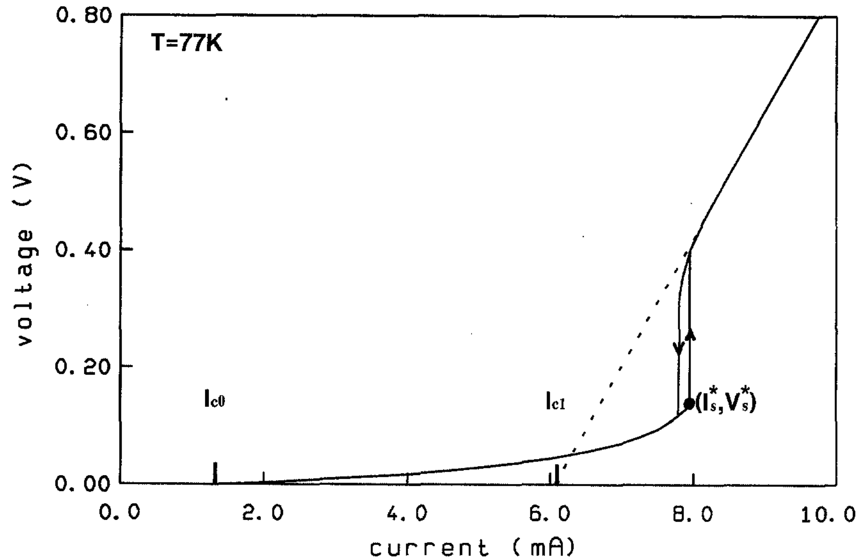



Fig. 2. A VCC of a 10×10 $\mu m^2$ bridge (sample No. 597-1) at 77 K. Arrows identify the two branches of the voltage step region.

region the current grows slowly with the voltage, since its change is caused only by the increase of the normal component.

Figure 3 shows the evaluation of the VCC nearby $T_c$ as a function of temperature. Besides the qualitative features, development of a "foot" and a voltage step, the most striking aspect is the resistance of the normal bridge at $T_c$ (the slope of line $K$) being approximately 400 Ω, whereas the differential resistance in the foot, even at large currents (more than three times $I_{c0}$) is less than 25 Ω. Qualitatively the same results as in Fig. 3 have been obtained with at least five different samples and three different dimensions of the bridge (2×4, 10×10, and 10×100 $\mu m^2$).

Looking at the development of the voltage step below $T_c$ one is tempted to ascribe this effect to purely thermal Joule heating. The power dissipated in the microbridge, e.g. at 73 K (curve $A$ in Fig. 3) is 1.2 and 15 mW at the lower and higher ends of the voltage step, which corresponds to a considerable power density of 1.2 and 15 kW/$cm^2$ respectively. A simple calculation[14] of the temperature change of a 10×10 $\mu m^2$ area heated by 1 mW power in thermal connection with a thermal semi-infinite reservoir with a thermal conductivity of 0.2 W/K cm (as is the case of $SrTiO_3$ [Ref. 15]), predicts a rise of approximately 2 K. The curves up to ($I_s^*$, $V_s^*$) are therefore taken at essentially constant temperature, whereas probably the lines above the voltage step are taken around or above $T_c$ (measured at zero current). The

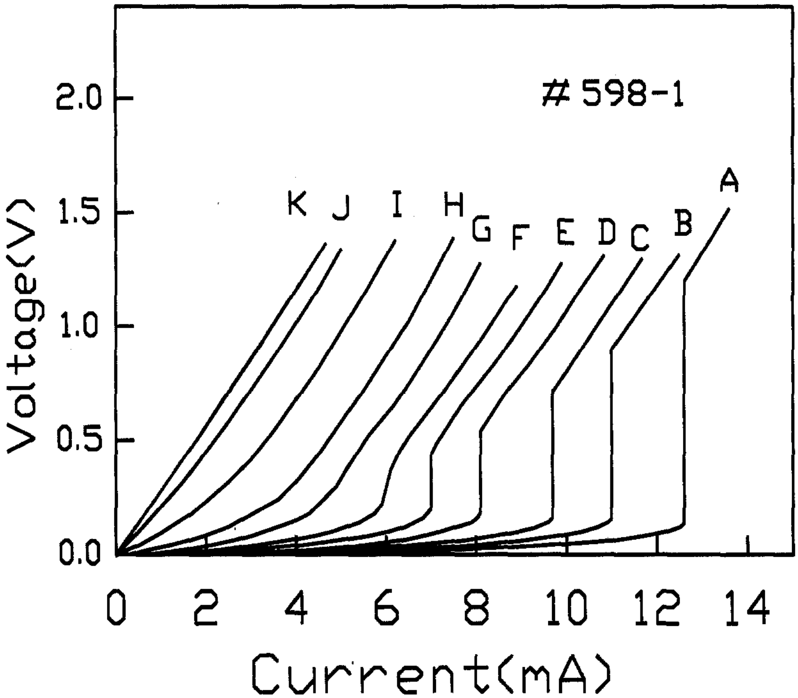



Fig. 3. A set of VCCs of a $10 \times 10\ \mu m^2$ bridge (sample 598-1) at different temperatures: A: 73 K, B: 75 K, C: 77 K, D: 79 K, E: 81 K, F: 83 K, G: 85 K, H: 87 K, I: 89 K, J: 91 K, and K: 93 K; at the voltage step region only the branch with increasing current is given.

constant slope above the voltage jump support this conclusion, there is no sign of a power induced transition, whereas at e.g. the highest point of line *D* at 79 K the power dissipated (approximately 15 mW) should give a temperature rise definitively above $T_c$. If the occurrence of the voltage step is due to Joule heating, the power $I_s^* \times V_s^*$ needed to induce the jump, should be expected to increase with decreasing temperature. However, it remains approximately constant.

In order to obtain a better insight in the thermal effects, we repeated the measurement at 77 K, but now with the sample in direct contact with liquid $N_2$, no change in the VCC could be detected. The set of VCCs of Fig. 3 were repeated at low ac frequencies (up to 20 Hz) with positive and negative polarity; the curves were completely symmetric and not frequency dependent. Measurements of the light response of the junction[12] revealed a strongly enhanced response, when the bias current was set in the voltage step region. This response showed only a weak frequency dependence (measured up to 4 KHz), significantly less than when the bias current was set at other values.

The main argument, however, for dealing with a non-purely-thermal effect in the VCC is taken directly from the curves in Fig. 3. The bridge can be evidently considered as a kind of thermometer, where both, the position in the $V-I$ plane, i.e. the resistance, and the slope, the differential resistance, should be considered. At 93 K, curve $K$, the sample is in the normal state. The VCC is a straight line, and the resistance is identical to the differential resistance. As the $R-T$ curve is monotonously ascending one could expect a position of a VCC-point with temperature above $T_c$ always lying on or above line $K$ or that $T_c$ has to be current dependent. For all lines starting below $T_c$, even at highest currents, there is a nonnormal component present in the total current, as the differential resistance at no point equals the ohmic value.

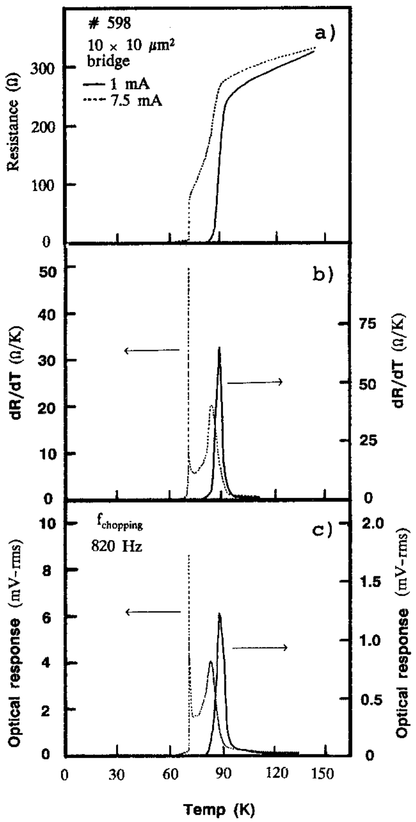



Fig. 4. The resistance $R$ (a), its thermal derivative $dR/dT$ (b), and the optical response (c) of bridge #598 as a function of temperature at a bias current of 1 and 7.5 mA.

The voltage step in the VCC is clearly not a transition to the normal state. The resistance ($R = V/I$) above the step, is, for curve $A$ in Fig. 3, e.g., at least a factor of two lower than the normal state resistance above $T_c$. Fig. 4 shows in more detail the temperature dependence of the resistance (Fig. 4a) and the optical response (Fig. 4c) for two constant currents, 1 and 7.5 mA, as a function of temperature. As already discussed before, the temperature is measured at the sample-holder. The actual temperature of the microbridge is approximately 2 K/mW higher due to the electrical power dissipation. If one takes in account the recent data of Nahum *et al.* about the thermal boundary resistance of YBaCuO films,[16] even 3 K/mW would be expected. In Fig. 4a clearly the voltage step in the curve measured with 7.5 mA can be seen, but also the still rising resistance up to the normal state value. This curve is in agreement with optical response measurements, Fig. 4c, which show a sharp peak at the voltage step. This peak has been followed up to 4 kHz with only a slight change in light response.[12]

Frenkel *et al.*[17] measured VCCs of microbridges patterned from epitaxial Y-Ba-Cu-O thin films made by pulsed laser deposition. In their paper they distinguish, similarly to our results, a foot and an abrupt voltage transition at high bias currents. Above the voltage transition, their films are in the normal state, in contrast to results on our films. Their conclusion about the purely thermal nature of the voltage step transition based on SEM voltage image analysis is therefore not totally valid for our highly granular type of films.

In conclusion: In our experiments we find intriguing VCCs, which exhibit unexpected behavior: a foot in the VCC curve, a voltage step, which eventually rises the temperature of the bridge above $T_c$ (determined at zero current), and a high voltage part, which clearly shows nonnormal behaviour.

## 3. COMPARISON WITH THEORY

Evidently we are not the first to observe the kind of VCCs described above. For classical superconductors Octavio *et al.*,[5,6] Daalmans *et al.*,[18] and Guthmann *et al.*[7] present experimental data for microbridges. For high-$T_c$ superconductors the low voltage part of VCCs with a "foot" and a voltage step have been reported by Frenkel *et al.*[17,19] Theoretical work on the VCC of weak links made from conventional superconductors have been done by Aslamazov and Larkin,[8] Aslamazov and Volkov,[20] and Tinkham,[9] who applied the AL theory representing a more physically transparent explanation of the observed effect (see also Kadin and Goldman[21]]).

The Tinkham theory[9] can be summarized as follows: For a very short bridge the quasiparticles inside the bridge are in equilibrium with the particle distribution in the banks, provided their energies are at least as great as the energy gap $\Delta_0$ of the banks. If the current through the bridge is increased above the critical current $I_{c0}$, a voltage $V$ is generated across the bridge, which due to the ac Josephson effect generates an rf field of frequency

$$\omega = 2eV/\hbar = d\Phi/dt \tag{1}$$

with $\Phi$ the phase difference between the order parameters of the two banks. The rf field cyclically decreases the gap according to the solution of the static Ginzburg-Landau equation for a short bridge of length $L$, oriented in the $x$-direction ($x=0, L$ on the banks):

$$|\Delta(x,\Phi)|^2 = \Delta_0^2\left[1 - \frac{4x}{L}\left(1-\frac{x}{L}\right)\sin^2\frac{\Phi}{2}\right] \tag{2}$$

As the quasiparticles with energy below $\Delta_0$ are isolated from the banks and trapped in the bridge, the effective temperature $T^*$ of the bridge can vary according to the cyclically decreasing gap. Through inelastic scattering the quasiparticles can leave the bridge region. This process is governed by $\tau_E$, the inelastic scattering time of quasiparticles at the Fermi surface, the electron phonon scattering time. Tinkham then considers two extreme cases (see Fig. 7, p. 255 in Ref. 9): (i) $\omega\tau_E \ll 1$ and (ii) $\omega\tau_E \gg 1$. In the first case the departure from equilibrium is small. Tinkham shows that $\delta T^* < 0$, giving an enhancement of $I$ on the forward half cycle of the Josephson current cycle; but $\delta T^* > 0$, giving a reduced reverse current in the second half cycle. This results in an increase of the forward current in both half cycles, the increase being proportional to $\omega\tau_E \propto V$. Thus at low voltage $V \ll \hbar/2e\tau_E$ the average supercurrent acquires a dc component; it is allowed to speak of a dynamically enhanced supercurrent. In case (ii), however, the cyclic gap reduction results in a cyclic reducing of $T^*$ in both the forward and backward directions, resulting in an ac enhancement of the supercurrent, but without a dc component. AL[8] give an expression for the maximum voltage $V_s^*$ of the foot:

$$V_s^* \cong \hbar T/e\tau_E\Delta_0(T) \tag{3}$$

The theoretical model of AL[8] and Tinkham[9] at least qualitatively explains our results, but a difficult point remains: the high voltages—in the order of volts—measured with our bridges. The explanation is, that we are dealing with a network of hundreds of BJJs (approximately 50 BJJs/$\mu$m$^2$, see Fig. 1), where the voltages of the individual bridges add up. A similar observation, giant Shapiro steps in 2D Josephson arrays, has been reported by Benz *et al.*[22]

In Fig. 5 we plot $V_s^*$ as a function of $T/T_c$ for two $10\times 10$ $\mu$m$^2$ bridges. For a comparison with theory, we assume that $V_s^*$ is a sum of $n$ identical

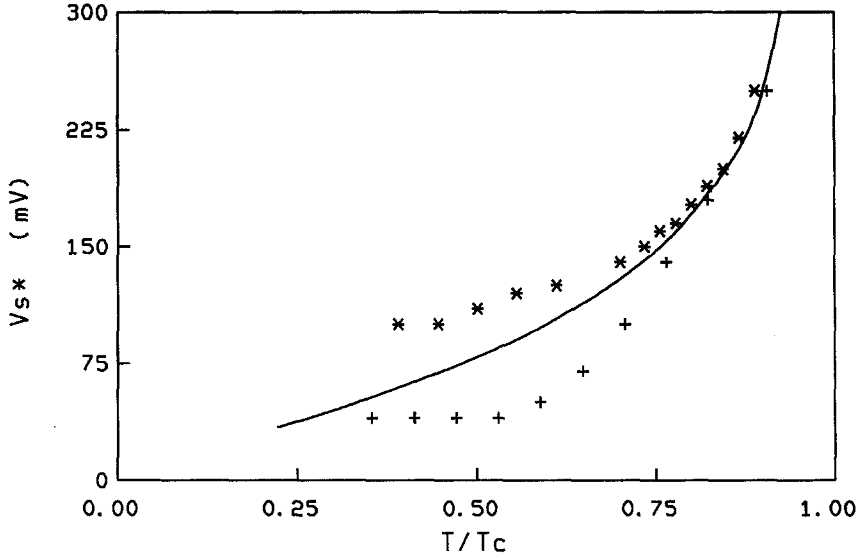



Fig. 5. The voltage $V_s^*$ as a function of $T/T_c$ for a $10\times10\ \mu m^2$ bridge. Experiment: "+" bridge No. 597-1; "*" bridge No. 598-1. Theory: solid line, fit according to Eq. (3).

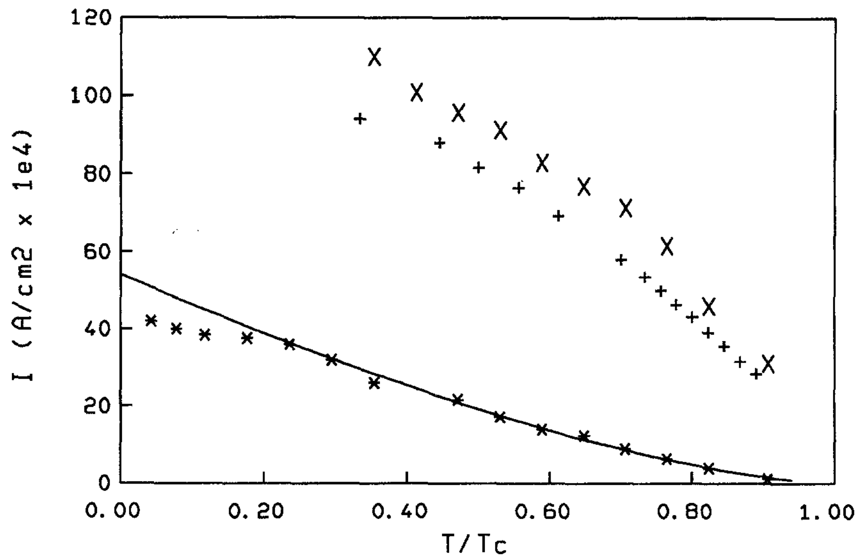



Fig. 6. Critical current as a function of $T/T_c$ for a $10\times10\ \mu m^2$ bridge. Experiment: "*" $I_{c0}$ of bridge No. 598-2, "+" $I_s^*$ of bridge No. 598-1, "×" $I_s^*$ of bridge No. 597-1. Theory: solid line, fit assuming $I_{c0}\propto(1-T/T_c)^{3/2}$.

individual junctions, in our case $n \cong 60$, as can be seen from SEM pictures (see Fig. 1). The solid line in Fig. 5 is a calculation with the aid of relation (3), where for $\Delta_0(T)$ we use the BCS gap energy. With only one fit parameter $\tau_E/n = 2.5 \times 10^{-13}$ s, there is a qualitative agreement between theory and experiment. The resulting value for $\tau_E \cong 1.5 \times 10^{-11}$ s is in agreement with the value of Gershenzon *et al.*,[23] obtained for YBaCuO at 1.6 K: $\tau_E \cong 10^{-10}$ s. In Fig. 6 we show $I_s^*$ and the critical current as a function of temperature. The solid line is a fit according to $I_c \propto (1 - T/T_c)^{1.5}$; this relation, an indication of JJ-like behaviour, is valid above 0.2 $T_c$.

## 4. CONCLUSION

We have given evidence that we are able to prepare networks of BJJs which show characteristic features in the VCCs: a foot and an abrupt voltage step transition to a still nonnormal state. Therefore an explanation based only on heating effects within the bridge is rejected. The theoretical model of Tinkham[9] for conventional superconductors based on the work of Azlamazov and Larkin[8] can be used to explain our experimental data for ceramic superconductors. In analogy with Octavio *et al.*[5] we interpret our VCCs as an evidence of dynamic enhancement of the supercurrent. From our data and the theory it is possible to derive a value of the electron phonon scattering time $\tau_E \sim 15$ ps (at $T \cong T_c$).

A possible explanation for the VCCs could be as follows: at low current below $T_c$ there is a complete superconducting state. At higher currents, a foot appears, but the superconducting component still increases due to the current induced cooling of the BJJs. The cooling is getting less effective at high voltages with consequently higher Josephson frequency. A voltage step appears, which ends around the zero current $T_c$. Increasing the current one still remains in a nonnormal state.

As already has been shown by Frenkel *et al.*,[19] the transition from the superconducting to the current induced resistive state (the foot in our description) is possible within 250 ps. From our value for $\tau_E$, we estimate a lower limit for the transition time to the voltage step in the order of $10^{-11}$ s. Application in the field of ultra-fast electronics of a device using the highly nonlinear VCC can be expected.

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