Subgap Structures in Intrinsic Josephson Junctions of $Tl_2Ba_2Ca_2Cu_3O_{10+\delta}$ and $Bi_2Sr_2CaCu_2O_{8+\delta}$

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We report on pronounced structures in the current-voltage characteristics of intrinsic Josephson junctions in the high-T_c superconductors $Tl_2Ba_2Ca_2Cu_3O_{10+\delta}$ and $Bi_2Sr_2CaCu_2O_{8+\delta}$. The structures appear well below the superconducting gap, independent of sample geometry, magnetic field, and temperature up to $0.5T_c$. We discuss possible explanations and show that the structures could be explained by a single peak in the quasiparticle current, possibly arising from a subgap structure in the quasiparticle density of states. For superconducting superlattices a subgap peak has been predicted and could explain our results. [S0031-9007(96)00449-8]

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The high-T_c superconductors form natural superconducting multilayers where the superconducting order parameter is periodically modulated along the c axis. At least in the highly anisotropic compounds—like $Bi_2Sr_2CaCu_2O_{8+\delta}$ (BSCCO) or $Tl_2Ba_2Ca_2Cu_3O_{10+\delta}$ (TBCCO)—the modulation is strong enough that adjacent superconducting copper oxide double or triple layers are only weakly coupled by the Josephson effect. This behavior can be directly observed in c axis transport measurements in sufficiently small single crystals [1–4]. Every pair of adjacent copper oxide double or triple planes, together with the intervening nonsuperconducting layers, forms an intrinsic Josephson junction. The periodic order parameter variation should also manifest itself in the quasiparticle density of states. From solutions of the Bogoliubov–de Gennes equations for a periodically modulated superconducting order parameter global [5–7] and local [7] densities of states have been predicted which are quite different from that of a homogeneous BCS superconductor. A special feature of them is a subgap peak. Physically, this subgap peak originates from de Gennes–Saint James bound states in the normal region between two superconducting banks [8] which broaden to a band structure in superconducting superlattices. The above-mentioned calculations are based on SNS superlattices or a modification thereof [7] for which properties similar to a SINIS superlattice are to be expected. Here, S, N, or I stands for alternating superconducting, normal, and insulating layers. A SINIS model seems resonable for BSCCO and TBCCO, since band structure calculations yield insulating behavior for SrO layers [9] and tunneling experiments showed that the BiO and TlO layers can be metallic at least for higher doping levels [10,11]. It seems improbable that the peculiar structures of the quasiparticle density of states in a superlattice can be observed by tunneling experiments with artificial tunnel structures [6]. They effectively only probe the outermost few Å of the sample and might suffer from surface roughness or surface degradation. Intrinsic Josephson junctions, on the other hand, are perfectly epitaxial layer by layer. They are insensitive to surface degradation and can provide local information about quasiparticle conductivity between pairs of CuO_2 bilayers or trilayers located in the bulk of the sample. Therefore, natural intrinsic junctions might be a unique tool to study the intrinsic properties of high-T_c superconductors. A close examination of their current-voltage (I-V) characteristics might be a clue to the electronic density of states and thus provide an improved understanding of high-T_c superconductors. In this Letter we will show that the I-V characteristics of intrinsic Josephson junctions in TBCCO and BSCCO exhibit a subgap structure that occurs on each quasiparticle branch. Its striking regularity indicates that the structure is a general property of all intrinsic Josephson junctions in the sample, and not only a parasitic effect of some of them. The fact that we observed these subgap structures in two different materials might indicate a common property of strongly anisotropic layered superconductors.

Measurements have been performed on step stacks fabricated on thin TBCCO films and on mesas patterned on BSCCO single crystals. The step stacks are made on TBCCO thin films deposited on $LaAlO_3$ substrates with ion etched step edges. Film thicknesses were betweeen 250 and 450 nm. Bridges of widths between 3 and 24 μm were patterned across the step thus defining a stack of typically 100–200 intrinsic junctions at the step [12]. The

BSCCO crystals were grown in an oxygen atmosphere by floating zone technique [13]. X-ray diffraction confirms that the crystals are single phase Bi-2212, and energy dispersive x-ray analysis indicates a cation stoichiometry of approximately Bi:Sr:Ca:Cu = 2.2:1.8:1.1:2 [14]. Using standard photolithography and Ar-ion etching, mesas of 20×20 and 40×40 μm^2 across and of varying heights were fabricated on the *a*-*b* face of the crystal. For transport measurements a four-point technique was used for the TBCCO step stacks. BSCCO samples were measured in a two-terminal configuration with low resistance gold contacts sputtered onto the freshly cleaved *a*-*b* faces of the crystals. The two-terminal configuration results in contact resistance in the *I*-*V* data, which has been subtracted for data evaluation. To reduce the influence of external noise, low-pass filters were used in the current and voltage leads. Some of the measurements were performed in a shielded room.

The *I*-*V* characteristic of every individual junction in an intrinsic Josephson junction stack exhibits a superconducting and a resistive branch, similar to the *I*-*V* characteristics of standard Josephson tunnel junctions. All junctions in the stack can be switched into the resistive state individually, such that the total *I*-*V* characteristic consists of many branches differing by the number of junctions in the resistive state [1]. As an example, Figs. 1(a) and 2(a) show *I*-*V* characteristics of a BSCCO mesa and a TBCCO step stack at $T = 4.2$ K. After exceeding the critical current of an individual junction in the stack, the *I*-*V* characteristic exhibits a characteristic voltage jump V_c. At low temperatures, $V_c \approx 21$ mV in BSCCO mesas and $V_c \approx 27$ mV in TBCCO step stacks, which is about 50% of $2\Delta/e$ [15,16]. The latter would be obtained for a standard SIS tunnel junction. Figures 1(b) and 2(b) show the subgap structures for a BSCCO mesa and for a TBCCO step stack, respectively. We find two hysteretic regions on each branch. On the first branch we observe two voltage jumps with increasing bias current, at voltages denoted V_1^1 and V_1^2 in Fig. 1(c). In BSCCO $V_1^1 = 6.15$ mV and $V_1^2 = 8.05$ mV, in TBCCO $V_1^1 = 7.5$ mV and $V_1^2 = 9.6$ mV. On the *n*th branch, corresponding to *n* junctions being resistive, there are *n* hystereses in each region. This strongly indicates that every individual junction exhibits a quasiparticle branch consisting of three subbranches denoted "*a*," "*b*," and "*c*" in Fig. 1(c). On the *n*th branch this results in *n* possible subbranches in each region. Here, with increasing bias current the voltage jumps first at V_n^1 in the lower region and at V_n^2 in the upper region. For example, at voltages below $V_2^1 \approx 2V_1^1$ on the second branch in Fig. 1(c) both resistive junctions are on their subbranch *a*. After the first voltage jump one junction has switched to branch *b*, and after the second jump both junctions are on their subbranch *b*. At $V_2^2 \approx 2V_1^2$ first one and finally both junctions switch to *c*. Thus, if all junctions in the stack exhibit identical features, V_n^1 and V_n^2 (when all *n* resistive junctions are in states *a* and *b*, respectively) divided by the branch number *n* should essentially be independent of *n*. The result, shown in the inset of Fig. 3, for a BSCCO mesa clearly confirms this interpretation.

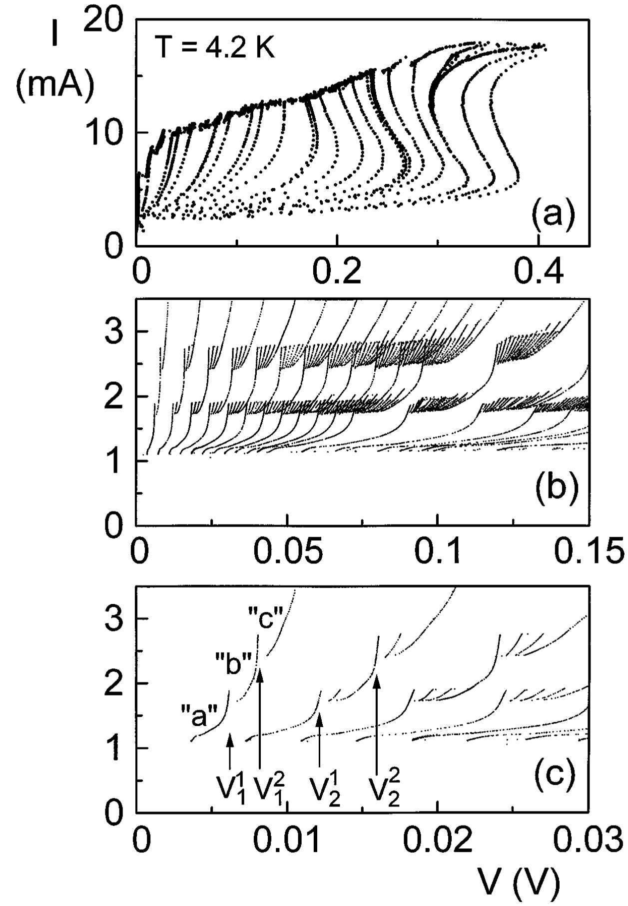

FIG. 1. Overall *I*-*V* characteristic of BSCCO sample D1 exhibiting multiple branching in the resistive state (not all branches are traced out for clarity). (b) Same *I*-*V* characteristic on expanded scales showing subgap structures at bias currents of about 1.9 and 2.7 mA. (c) Enhanced resolution of first three branches of (b) with definition of subbranches "*a*," "*b*," and "*c*" and voltages V_n^1 and V_n^2.

We found the subgap features in a variety of samples, measured in different experimental setups. V_n^1 and V_n^2 were independent of the geometrical size of the samples. In magnetic fields of up to 2 T we found that the structures broadened but V_n^1 and V_n^2 remained unchanged. We can therefore exclude the possibility that the observed features arise from geometric self-resonances or vortex motion [17], since such resonances should depend on geometry and magnetic field. Also, V_n^1 and V_n^2 are temperature independent below $0.5T_c$ (Fig. 3). A temperature increase only results in a decreased hysteresis, the intensity, i.e., the amount of excess current above the (extrapolated) quasiparticle curve, remains essentially constant. Above about $0.5T_c$ the characteristic voltage V_c drops below V_1^1 and V_1^2, making further evaluation impossible (Fig. 3). We also investigated the subgap

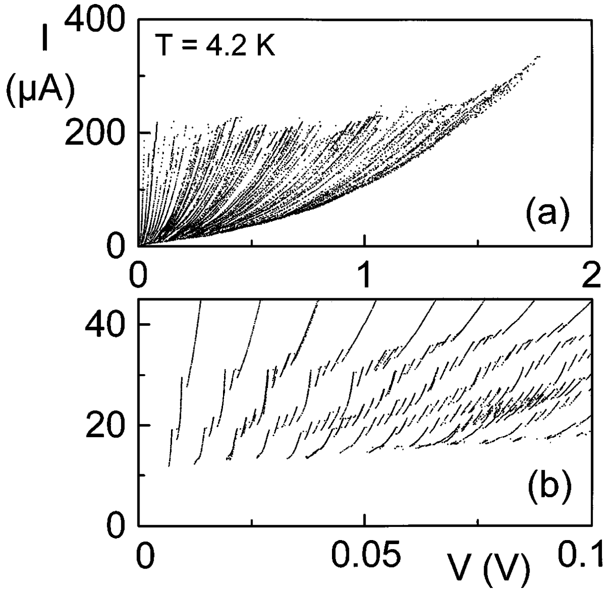

FIG. 2. (a) Overall *I-V* characteristic of TBCCO sample V40a34 exhibiting multiple branching in the resistive state. (b) Same *I-V* characteristic on expanded scales showing subgap structures at bias currents of about 20 and 30 μA.

structures for BSCCO crystals under different annealing conditions. For example, the as-grown sample D6, with a T_c of 88 K and a critical current density of 660 A/cm^2, exhibited weak, nonhysteretic structures. After oxygen doping by annealing in air the critical current density is raised to 1630 A/cm^2, and the critical temperature lowered to 80 K. The subgap structures are now more pronounced. Within the measurement error the structures still occur at the same voltages (cf. Table I).

The observed structures may not directly map voltage dependent features of the quasiparticle current, due to the presence of ac Josephson currents at nonzero voltages. We therefore calculated the *I-V* characteristic by including the ac Josephson effect. Since the total *I-V* characteristic of the whole stack of intrinsic Josephson junctions can essentially be obtained from combining *I-V* characteristics of the individual junctions, it seems sufficient to calculate the *I-V* characteristic of a single junction. A phenomenological approach is the resistively shunted junction model, which, in its simplest form, assumes a quasiparticle current $I_q = V/R$ [18], together with the Josephson equations for the supercurrent. In order to include voltage dependent features we parametrize I_q by

$$I_q(V) = I_p \exp\left[-\left(\frac{V - V_p}{V_\sigma}\right)^2\right] + \frac{V/R}{1 + \exp[(V_g - V)/V_b]}, \qquad (1)$$

where V is the voltage, and $I_p, R, V_p, V_g, V_\sigma, V_b$ are fitting parameters. The first term is to model a (subgap) peak in I_q, located at a voltage V_p. The second term accounts for the global gap structure. We write the total current I through one on the junctions of the stack as the sum of the Josephson current, the quasiparticle current, and the displacement current, $I = I_J + I_q + I_d$. With the use of the Josephson relations, $I_J = I_c \sin(\gamma)$, $\hbar d\gamma/dt \equiv \hbar\dot{\gamma} = 2eV$, and $I_d = CdV/dt$ we get

$$I = I_c \sin\gamma + I_q\left(\frac{\hbar\dot{\gamma}}{2e}\right) + \frac{\hbar C}{2e}\ddot{\gamma}. \qquad (2)$$

We finally obtain the *I-V* characteristic by calculating the time average of $V(t)$. The result is shown in Fig. 4, together with the functional form of I_q. Surprisingly, one peak in I_q has turned out to be sufficient to produce the observed double structure in the *I-V* characteristic. The actual peak position is located at subbranch *b*, at about 7.4 mV for BSCCO and 9.0 mV for TBCCO. A peak in I_q seems to be the simplest explanation for the observed subgap structures. However, other mechanisms might be able

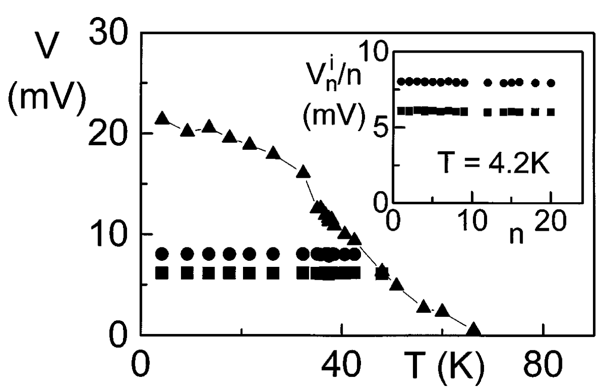

FIG. 3. Voltages V_1^1 (squares) and V_1^2 (circles), as defined in Fig. 1(c), of annealed BSCCO sample D6, together with characteristic voltage V_c (triangles), vs temperature; inset shows V_n^1/n (squares) and V_n^2/n (circles) of BSCCO sample D7 vs branch number n at 4.2 K.

TABLE I. Critical temperature and voltages V_n^1 and V_n^2 of various samples.

Sample	Material	T_c	V_n^1 (mV)	V_n^2 (mV)
V40a34	TBCCO	109	7.43 ± 0.05	9.60 ± 0.05
V75a12	TBCCO	108	7.65 ± 0.05	9.65 ± 0.05
V75b24	TBCCO	115	7.57 ± 0.1	9.60 ± 0.1
V86c7	TBCCO	115	7.44 ± 0.05	9.64 ± 0.05
V91c1	TBCCO	114	7.54 ± 0.05	9.71 ± 0.05
V91c8	TBCCO	114	7.36 ± 0.1	9.57 ± 0.1
D6a	BSCCO	91	6.02 ± 0.05	7.90 ± 0.05
D9b	BSCCO	87	6.05 ± 0.05	7.97 ± 0.05
D6	BSCCO	88	5.95 ± 0.5	7.97 ± 0.5
D6[a]	BSCCO	80	6.15 ± 0.05	8.05 ± 0.05
D1[a]	BSCCO	84	6.16 ± 0.05	8.06 ± 0.05
D7[a]	BSCCO	87	6.09 ± 0.05	8.05 ± 0.05

[a]Annealed in air for 10 h at 350 °C.

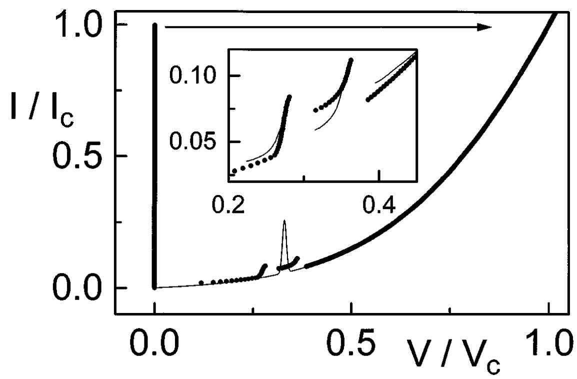

FIG. 4. Functional form of quasiparticle current as used for simulations (solid line) and computed *I-V* characteristic (markers). Inset compares simulated *I-V* characteristic (markers) with measured *I-V* characteristic of TBCCO sample V40a34 at 4.2 K (solid line). Abscissa in units V/V_c, ordinate in units I/I_c.

to account for the structures. First, we can rule out resonances of the ac Josephson current involving sample geometry, as discussed above. Second, ac Josephson currents could couple to excitations like phonons. This mechanism would very likely require two excitations at frequencies $2eV_1^1/\hbar$ and $2eV_1^2/\hbar$. Although it would be an exciting observation to see that ac Josephson currents are able to couple to phonons at several THz, we note that, in contrast to our observations, a phonon based mechanism should show a strong temperature dependence of the intensity of the structures. Third, the observed features might be associated with subharmonic gap structures at voltages $2\Delta/n$ arising from either multiparticle tunneling [19], Josephson self-coupling [20], or multiple Andreev reflection [21]. Even with the assumption of an extremely low value of Δ of, say, 12 meV we would infer $n = 3$ or larger. It seems unlikely that such high orders appear as the most pronounced features in the subgap regime. Finally, the effect could be associated with a superconducting gap in the BiO layers. However, the critical temperature of the BiO layer would have to be well above 50 K, since no temperature dependence was observed below this temperature. Also we should have seen an appreciable voltage shift with annealing. Returning to our first interpretation we have seen that one peak in the quasiparticle current, possibly arising from a subgap peak in the quasiparticle density of states, might account for the observed structures. This peak would have to be at 3.7 meV in BSCCO and at 4.5 meV in TBCCO, corresponding to about 0.2Δ. One might argue that the peak should scale with Δ and thus with temperature. However, we note that we were able to evaluate V_n^1 and V_n^2 only up to $0.5T_c$, where the gap is known to vary only marginally [15,16]. Finally, a structure at 0.2Δ might be too low in energy to be compatible with features arising from the layered nature of the high-T_c superconductors. This question remains open and has to be clarified by theory.

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