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# Strong temperature dependence of the $c$-axis gap parameter of $Bi_2Sr_2CaCu_2O_{8+\delta}$ intrinsic Josephson junctions

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Stacked series arrays of intrinsic Josephson tunnel junctions have been fabricated on the surfaces of $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystals using photolithography and Ar-ion milling together with *in situ* monitoring of the resulting current-voltage ($I$-$V$) characteristics. The number of junctions in the stack (along the $c$ axis) is varied from 1–5 to ~200 in a controlled way. Both Josephson coupling and multiple branches with well developed gap features are seen in the $I$-$V$ dependences of the arrays at $T \lesssim 90$ K. The gap parameter $\Delta_c$ is 10–13 meV at 4.2 K, which is approximately half the value reported in the literature. The temperature dependence of $\Delta_c$ deviates strongly from the BCS one. Proximity induced superconductivity of the Bi-O layers may possibly explain both the reduced gap parameter and its temperature dependence.

The electronic and superconducting properties of layered $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) high-$T_c$ materials have proved to be a rich source of new physics because of its large anisotropy. One of the most fundamental superconducting characteristics of the new material is the superconducting gap parameter $\Delta$. Of the traditional methods used to determine $\Delta$, one of the most direct is electron tunneling spectroscopy, which can also determine gap anisotropy in single-crystal specimens. A complication in performing and interpreting tunneling measurements is that they are surface measurements, and often these surfaces are poorly characterized.

The layered, extremely anisotropic Bi2212 high-$T_c$ material exhibits intrinsic Josephson (IJ) effects.[1] The Josephson tunneling in this compound is believed to occur between superconducting double Cu-O layers of thickness 3 Å separated by intermediate Sr-O and Bi-O layers of thickness $d=12$ Å.[1] Thus, tunnel junctions are natural parts of the crystal structure and therefore offer a possibility of measuring inherent superconducting characteristics of the interplane tunneling. A difficulty with such a model[1] is that the superconducting coherence length in the $c$-axis direction $\xi_c$ ($\approx 1$ Å) (Ref. 2) is much less than the distance $d$ between double Cu-O planes in the Bi2212 compound. This makes it difficult to adopt the scenario of direct Josephson tunneling across such a long distance. From our measurements we have reason to believe that the picture is more complicated.

In this paper we report on the reduced value of the $c$-axis gap parameter $\Delta_c$ of IJ tunnel junctions and its strong temperature dependence. $\Delta_c$ at low temperatures is approximately half the expected value and $\Delta_c(T)$ deviates widely from the BCS temperature dependence. These observations conform with the assumption of proximity induced superconductivity of the intermediate Bi-O layers. The Josephson tunneling then takes place between adjacent Bi-O layers with small order parameters while there is a strong proximity coupling between Bi-O and Cu-O layers.

To observe the IJ effects in the single crystals of Bi2212 one has to work with very small tunnel areas in order to reach the superconducting critical current $I_c$ without too much heating in the contacts.[1] Bi2212 single crystals are usually several micrometers thick. This means that there are several thousand junctions in series and it becomes difficult to extract the tunneling properties of an individual junction.

Small stacks of different areas and heights can be fabricated on the surface of a bulk single crystal by photolithography.[3–5] The advantages of this method are that the number of junctions in series may be reduced to only a few and several contacts may be fabricated on top of the stacks, enabling, e. g., four probe measurements.[3,4]

The fabrication process for making stacks of approximately 1–5 to 200 junctions (10×20, 10×50, 20×30, and 20×70 $\mu m^2$ large by 0.01–0.3 $\mu$m thick) on the surface of a Bi2212 single crystal[6] uses standard photolithography and Ar-ion milling.[4] By varying the intensity of the Ar-ion beam, etching time and monitoring the resulting current-voltage ($I$-$V$) characteristics *in situ* at $\approx 80$ K (using a cryocooled sample holder in a vacuum chamber) we succeeded in making only a few junctions in stacks. The minimal number of junctions, made in a reproducible and controlled way, was 4–5. At several occasions, we observed only one junction in the $I$-$V$ characteristic in the whole range of currents (0–15

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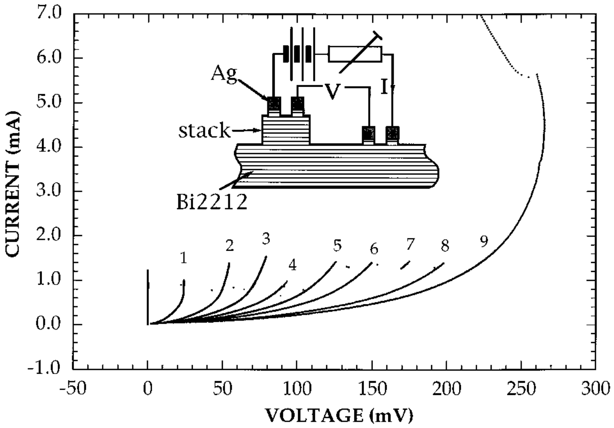

FIG. 1. $I$-$V$ dependence for the sample having nine junctions in the stack. The temperature is 20 K. The numbers near the curves denote the quasiparticle branches of successive junctions going normal as the current increases. The inset shows a schematic layout of the four probe measurement. Due to the fabrication process there are usually 2–3 extra layers underneath each contact, which separate the surface of the stack under study from the normal metal and have no influence on the four probe measurements.

mA). Several contacts to the top of the stack and to the etched-down lower region of the single crystal are made by photolithography and ion milling or chemical etching of a $\sim$2000-Å-thick Ag film. The contact resistance between the Ag thin film and the surface of the single crystal is typically a few ohms at room temperature. Hard baked photoresist is used as isolation. No subsequent heat treatment is needed to clearly see the superconductor-insulator-superconductor Josephson-like behavior of these series arrays.

Figure 1 shows the $I$-$V$ dependence of an array of nine junctions at 20 K. The number of junctions is determined from the number of quasiparticle branches seen in the $I$-$V$ dependence. Each of the branches was traced, one by one, in multiple runs with a slow bias current sweep in a similar way as described in Ref. 1. The $I$-$V$ curves were highly hysteretic, typical for Josephson tunnel junctions with large capacitances. The ratio $I_c/I_r$ between the critical current and the drop-back current $I_r$ (i.e., the smallest return current in the voltage state) was about 50–100 for different samples at low temperature.

One can see in Fig. 1 that the first branch has a well-developed gap structure, the $I$-$V$ dependence is almost vertical at the characteristic (gap) voltage $V_g\approx$ 25 mV. Another sample with a stack of 17 junctions ($I$-$V$ dependence is not shown) has $V_g\approx 18.5$ mV and the sample with only one junction has $V_g\approx$ 27 mV at 4.2 K. Stacks with larger number of junctions always have less distinct current rise at $V=V_g$, but still $V_g$ ranges between 18 and 20–22 mV.[7] It is also seen in Fig. 1 that the subgap current is relatively large and $I_c$ is small as compared with the current rise at the sum-gap voltage.

For the case of tunneling between two identical superconductors, a sharp increase of the current should take place at $V_g=2\Delta(T)/e$, where $\Delta(T)$ is the temperature-dependent superconducting gap parameter and $e$ is the electron charge.[8] The value of $\Delta$ obtained in the majority of tunneling experiments on Bi2212 is approximately 25 meV at low temperature.[9] This value was recently reported for high-quality break tunnel junctions made from bulk Bi2212 single crystals.[10] Careful scanning tunneling microscope (STM) experiments also show $\Delta$ close to 25 meV.[11] In the data presented here, $V_g\approx 20$–26 mV corresponds to $\Delta_c\approx 10$–13 meV for the interplane tunneling in the $c$ direction.

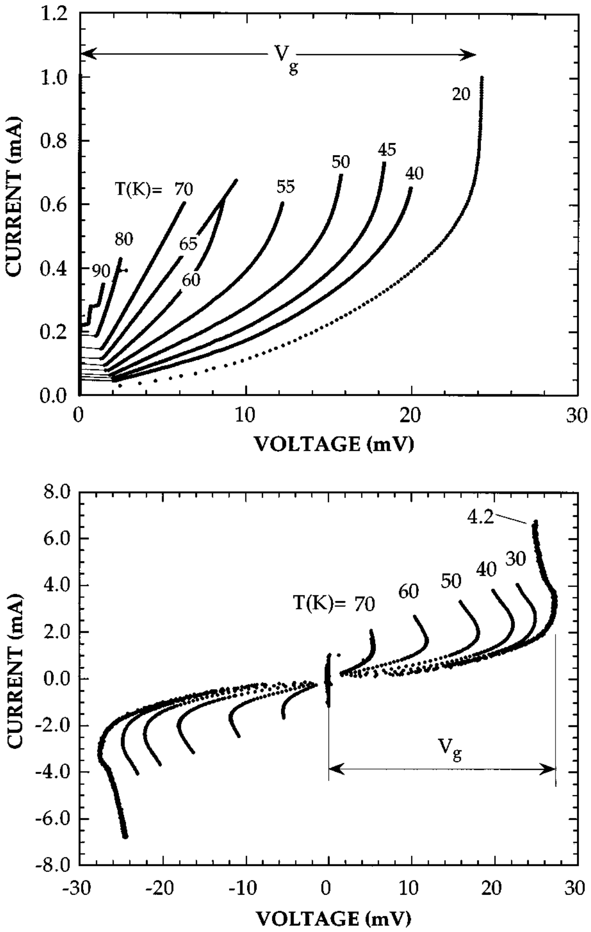

FIG. 2. (Top) $I$-$V$ characteristics for junction No. 1 having the lowest $I_c$ of the same stack as in Fig. 1 at different temperatures. There are several branches shown for $T=90$ K. Note the change in the shape of the $I$-$V$ dependence at $T\approx 60$–65 K. (Bottom) Temperature-dependent $I$-$V$ characteristics for the single-junction sample.

The fact that the gap feature is well developed in our tunneling curves allows us to accurately trace the temperature dependence of the gap voltage. Figure 2 (top) shows the $I$-$V$ dependence of the junction with lowest critical current (see Fig. 1, No. 1) and Fig. 2 (bottom) shows the $I$-$V$ dependence of a single-junction sample at various temperatures. It is clearly seen that the gap voltage at least doubles in the temperature region from 60 down to 20 K. Furthermore, the curved character of the $I$-$V$ dependence changes to nearly linear beyond 60 K for the samples with 9 and 17 junctions in the stack. This makes it impossible to judge what the gap voltage is between $T=60$ K and $T\leq T_c$.

Figure 3 shows the temperature dependences of the gap voltage, normalized to its value at 4.2 K, $V_g(T)/V_g(4.2)$, for several samples, having 1, 3, 9, and 17 junctions in the stacks. $V_g(T)/V_g(4.2)$ for all samples are similar. The gap

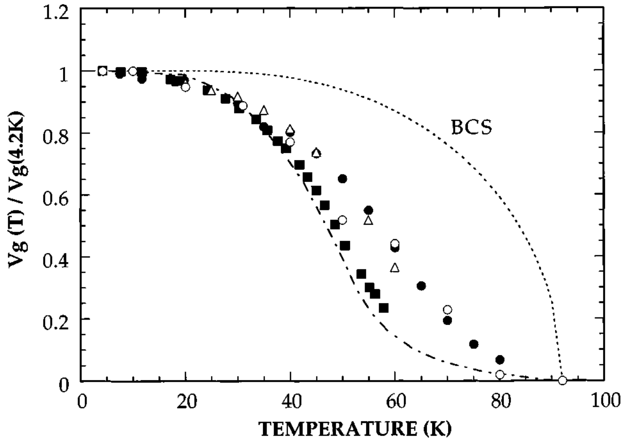

FIG. 3. Normalized gap voltage, $V_g(T)/V_g(4.2\text{ K})$, vs temperature for the stacks with 1 (filled circles), 3 (open circles), 9 (triangles), and 17 (squares) junctions. BCS temperature dependence (dotted line) and the temperature dependence of the normalized gap parameter $\Delta_{-1}(T)/\Delta_{-1}(0)$ from Fig. 1(b) of Ref. 14 (dashed/dotted line) are shown for comparison.

voltage decreases much more rapidly with increasing temperature than expected for a BCS gap.[12] The single-junction sample allows us to trace the temperature dependence of the gap voltage very close to $T_c$ and we conclude that the superconducting energy gap is finite everywhere below $T_c$. The energy gap does not go to zero at $T\approx 65$–$70$ K as might be concluded from an extrapolation of the $V_g$ dependence for samples with 9 and 17 junctions.

A scenario of coherent Josephson tunneling in between Cu-O double planes seems unlikely when comparing the tunneling distance $d\approx 12$ Å with the coherence length $\xi_c\sim 1$ Å. The role of the Bi-O layers in the $c$-axis superconducting properties has not previously been assessed and we believe that they give an important contribution in our observations.

In several models the tunneling in the $c$ direction is approached by a consideration of the high-$T_c$ superconductors as natural superlattices which consist of layers with different electronic and superconducting properties.[13,14] Such a model, as applied to the Bi2212 superconductor, assumes that the pairing coupling in the Cu-O layers is strong ($S$ layers) while in the Bi-O layers it may be weaker or completely absent ($S'$ or $N$ layers). The Sr-O layers are assumed to be insulating. The behavior of such a system with nonequivalent layers strongly depends on the ratio $t/kT_c$, where $t$ is the hopping integral between layers and $k$ is the Boltzmann constant. Furthemore, the Bi-O(3) bond, which joins adjacent sheets in the $Bi_2O_2$ bilayer is almost as long as 3 Å.[15] This relatively long and weak Bi-O(3) bond parallel to the $c$ axis results in the very weak interlayer bonding and micalike mechanical behavior of the Bi2212 superconductor which easily cleaves between the Bi-O layers.[17] Thus, it is not unreasonable to assume that the dominant tunneling barrier in the structure will be the one between Bi-O layers.

In such a way one can describe the layered Bi2212 compound as clusters of strongly superconducting $CuO_2$ bilayers covered from both sides by weakly superconducting single Bi-O layers: $S'SS'$-$S'SS'$-$\cdots$. The intracluster hopping integral $t$ can be relatively large ($t\gg kT_c$) while intercluster one, $t'$ (between adjacent Bi-O layers), can be small ($t'<kT_c$). According to this picture the Josephson tunneling takes place between the outermost layers (Bi-O) of clusters while in between the layers of cluster there is a strong proximity coupling. Early angle-resolved photoemission studies (ARPES)[16] and some STM experiments[18] demonstrated indeed, that the surface Bi-O layers can be metallic or even superconducting.

This model allows us to explain both the reduced value of the gap voltage $V_g$ and its strong temperature dependence. $V_g$ in this case is equal to $2\Delta_{S'}/e$, where $\Delta_{S'}$ is the small proximity induced order parameter of the Bi-O layers. The temperature dependence of $\Delta_{S'}$ can be quite unusual.[14,19] From Fig. 1(b) of Ref. 14 and $\Delta_{S'}\approx\Delta_S/2$, which corresponds to our case of having $V_g$ two times smaller than expected value, we can extract the hopping integral $t$ between Bi-O and Cu-O layers of about $0.35k_BT_c\approx 3$ meV.[14] Clearly, slightly different values of $V_g$ at low temperature for different samples may be attributed to a small variation of the oxygen content $\delta$ which is known to change the anisotropy (interlayer coupling) in Bi2212.

Although this model provides reasonable explanations for the experimental observations, one should examine other obvious possibilities, like a nonequilibrium current injection of quasiparticles or gap anisotropy.

(i) If the thickness of the superconducting electrodes of the stacked Josephson tunnel junctions is less than the quasiparticle diffusion length, the quasiparticles generated in one junction can easily penetrate into adjacent junctions and deteriorate their superconducting properties. The effect of nonequilibrium current injection in coupled arrays of tunnel junctions is usually manifested in a heavy backbending of the $I$-$V$ curves in the region of the sum-gap voltage.[20] Figure 1 shows an example of such a backbending. The last branch of the $I$-$V$ dependence, which corresponds to all nine junctions in the quasiparticle state has a discontinuity at a current $I^*=5.6$ mA and the dynamic resistance is negative beyond this current. The sum-gap voltage decreases continuously for $I^*\leq I<12$ mA (shown partly). Stacks with more junctions show even more pronounced decreases (up to 60% of the maximal value) of the sum-gap voltages.[3] However, a contribution to the gap suppression from simple heating may not be excluded due to higher energy dissipation in this case. The single-junction sample shows similar behavior, as is seen in Fig. 2. An additional source of quasiparticles in this case is due to two extra junctions underneath the current leads for this sample. They do not show up in the $I$-$V$ dependence due to the four probe measurements.

The injection of quasiparticles certainly may be important in stacked tunnel junctions, particularly at high current values. However, one may strongly question the reduced $V_g$ being solely due to the injection of quasiparticles as different samples with quite different critical current densities have nearly the same $V_g$. Furthermore, one would not expect such a nonequilibrium phenomenon to decrease $V_g$ to roughly half its expected value in many different samples with varying number of junctions and within large temperature intervals.

(ii) Large gap anisotropy at different parts of the Fermi surface is known to exist in conventional superconductors.[21] One could expect that the same situation takes place in much more anisotropic high-$T_c$ compounds which would suggest that there is a certain value of the gap parameter along the

$c$-axis direction. Many groups report on large in-plane gap anisotropy.[22,23] In recent ARPES experiments it has also been observed that the gap parameter along the $\Gamma - X[(110)]$ direction (the Bi-O-Bi direction in real space) is about 10 meV and is strongly temperature dependent much like $\Delta_c$ in our experiments.[23] The gap parameter along the $\Gamma - \overline{M}[(100)]$ direction ($\approx 16$ meV) remains at full value up to $0.85T_c$, and the anisotropy ratio of the gap parameter, thus, increases with temperature.[23] These observations seem to conform with several models, which assume that interlayer coupling is of key importance for high-temperature superconductors,[24,25] but it is not clear how the in-plane gap anisotropy might influence the c-axis superconducting properties seen in our experiments. In addition, several claims have been made for a distinct $c$-axis gap parameter of $\approx 12$ meV from tunneling experiments.[26]

While the gap anisotropy model can provide a plausible picture, its validity is ultimately subject to the actual electronic structure and the exact shape of the Fermi surface of the Bi2212, which is not firmly established.[27] Without going into detail, we just mention the difficulty in using the model.

The observation of the IJ effects in Bi2212 implicitly assumes that every double $CuO_2$ bilayer (plus, possibly, surrounding Bi-O single layers) acts very much like an independent superconductor with a three-dimensional Fermi surface, which, strictly speaking, is hard to define in half a unit cell. We think that it is not correct to use the concept of Fermi surface to describe the $c$-axis superconducting properties of Bi2212 and more work should be done to clarify this issue.

In summary we have measured the temperature-dependent $c$-axis gap voltage $V_g \equiv 2\Delta_c(T)/e$ of intrinsic Josephson tunnel junctions in Bi2212 single crystals. The gap parameter $\Delta_c(T) \approx 10-13$ meV at low temperature, which is approximately half the expected value. $\Delta_c(T)$ deviates strongly from the BCS temperature dependence. These observations agree well with a multilayer model of the $c$-axis transport in Bi2212, assuming a proximity-induced superconductivity of the Bi-O layers. We believe that our experiments will help to find a proper model to explain high-temperature superconductivity and will stimulate further experimental and theoretical investigations.

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