PHYSICAL REVIEW B VOLUME 55, NUMBER 18 1 MAY 1997-II

Current-voltage characteristics of intrinsic $Bi_2Sr_2CaCu_2O_8$ Josephson-junction stacks and an unconventional temperature dependence of the magnitude of the order parameter

Mikitaka Itoh, Shin-ichi Karimoto, Kazuichi Namekawa, and Minoru Suzuki
NTT Basic Research Laboratories, Nippon Telegraph and Telephone Corporation, 162 Tokai, Ibaraki 319-11, Japan
(Received 6 February 1997)

Very thin stacks of five intrinsic Josephson junctions have been fabricated on the surface of $Bi_2Sr_2CaCu_2O_8$ single crystals. The current-voltage characteristics of these stacks are in good agreement with those of a superconductor-insulator-superconductor tunnel junction made of a two-dimensional d-wave superconductor. This strongly suggests that there is no gap suppression caused by the nonequilibrium heating effect, which is usually significant in stacked junctions of this kind. From these results, we obtain a gap value of 57 meV at 4.2 K for the $Bi_2Sr_2CaCu_2O_8$ system. Furthermore, it is definitely found that the gap magnitude exhibits significant temperature dependence even at low temperatures, which is unlike the BCS behavior. [S0163-1829(97)50322-5]

The superconducting order parameter is one of the most fundamental physical properties as regards understanding high-T_c superconductivity. Recent experimental results strongly support the view that the order parameter of the high-T_c cuprates has d-wave symmetry,[1–5] i.e., $\Delta(\theta)=\Delta_0\cos(2\theta)$, where $\Delta(\theta)$ is the order parameter and $\theta=\tan^{-1}(k_y/k_x)$. The magnitude of Δ_0, however, differs from experiment to experiment, ranging from 20 to 100 meV in the case of scanning tunneling spectroscopic (STS) analyses.[6–9] Furthermore the temperature (T) dependence of the gap magnitude is unclear since the current-voltage (I-V) characteristics are smeared at high temperatures due to the superconductor-insulator-normal metal (SIN) junction structure.

Since the order parameter symmetry of the high-T_c superconductors is most likely to be d-wave with line nodes, the T dependence of the maximum order parameter amplitude $\Delta_0(T)$ is expected to be different from the BCS theory. At present, however, $\Delta_0(T)$ for the high-T_c superconductors is not clear. For example, in the theories thus far proposed for the T dependence of the Josephson current for a d-wave superconductor junction, BCS T dependence is assumed for $\Delta_0(T)$.[10,11] In order to understand the exact T dependence of these quantities, we need to know $\Delta_0(T)$. Therefore, it is important to know the exact T dependence of the gap magnitude as well as the gap magnitude itself.

The intrinsic Josephson junctions naturally formed in the layer-structured $Bi_2Sr_2CaCu_2O_8$ and $Tl_2Ba_2CaCu_2O_8$ systems show tunneling characteristics similar to those of conventional superconductor-insulator-superconductor (SIS) tunnel junctions.[12,13] Therefore, it is reasonable to suggest that the intrinsic Josephson junctions provide the most reliable gap values for the present. In the observed I-V characteristics of the intrinsic Josephson junctions, however, the gap magnitude becomes considerably ambiguous due to a significant gap suppression, which is caused by the nonequilibrium superconductivity effect or Joule heating.[14,15]

Recently, Yurgens *et al.*[16] reported a value of $2\Delta_0=20-26$ meV for the $Bi_2Sr_2CaCu_2O_8$ system based on measurements of stacks of single and nine intrinsic Josephson junctions. These values are rather small compared with other experimental results. The authors ascribe these reduced values to the proximity effect arising from the conductive barrier layers. This also causes the strong T dependence of $\Delta_0(T)$ which they observed near T_c. However, the lack of a clear normal tunneling region in the I-V characteristics strongly suggests gap suppression due to Joule heating as well as to the nonequilibrium heating effect. A comparison of these I-V characteristics with the numerical calculation[17] also indicates a deviation from the genuine characteristics expected for a d-wave superconductor SIS tunnel junction. Therefore, for the determination of $\Delta_0(T)$ using intrinsic Josephson junctions, it is necessary to observe the I-V characteristics of very thin stacks in which there is almost no heating.

We have fabricated very thin stacks, approximately 7.5 nm thick, of intrinsic Josephson junctions on the surface of $Bi_2Sr_2CaCu_2O_8$ single crystals. The thickness corresponds to five CuO_2 bilayers, i.e., five tunnel Josephson junctions connected in series. The I-V characteristics observed in these stacks are in good agreement with calculated quasiparticle I-V curves for a d-wave superconductor SIS junction with a slight leakage of conductance and an almost unsuppressed energy gap, providing an unambiguous Δ_0 value and its T dependence. The obtained value of $2\Delta_0=57$ meV is much larger than the value estimated by Yurgens *et al.*[16] Furthermore, we have found that $\Delta_0(T)$ remains T dependent at low temperatures; $\Delta_0(T)$ increases as T approaches 0 K, which is quite unconventional and should be reflected in the T dependence of various properties of high-T_c superconductors.

The $Bi_2Sr_2CaCu_2O_8$ single crystals we investigated were grown by the self-flux method using a platinum crucible.[18] We annealed the crystals at 845 °C for 80 h and 600 °C for 80 h, and then cleaved small laminar crystals from them which were typically 1 mm × 1 mm × 50 μm in

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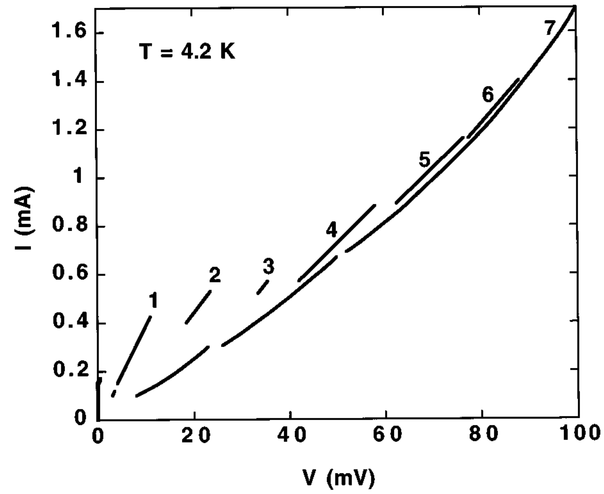

FIG. 1. Current-voltage characteristics for a 7.5 nm thick stack (five double CuO_2 layers) at 4.2 K.

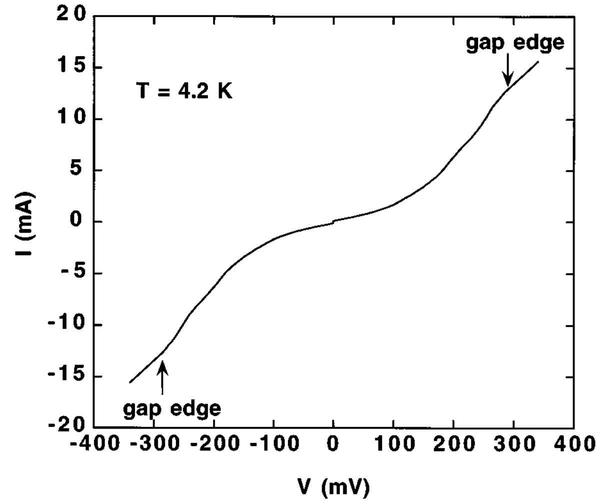

FIG. 2. Current-voltage characteristics over an extended range for the 7.5 nm thick stack in Fig. 1 at 4.2 K.

size. Then, a 50 nm thick Au film was evaporated on the crystals and annealed at 650 °C for 1 h in O_2 gas to reduce the contact resistance. These crystals were glued on sapphire substrates. The 7.5 nm thick stacks were fabricated using standard photolithographic and Ar milling techniques. The upper electrode wiring consisted of a 450 nm Au film. We employed a three terminal configuration, which functioned to facilitate direct heat flow into the upper Au electrode. The dimension and thickness of the stacks were 20 μm $\times$ 20 μm and 7.5 nm, respectively. Other fabrication details are described elsewhere.[15]

Before measuring the *I*-*V* characteristics of the intrinsic Josephson junctions, we measured the resistivity along the c-axis ρ_c as a function of temperature. At room temperature, $\rho_c = 107\ \Omega$ cm, which is larger than reported in previous studies.[15,19] From 250 K down to a T_c of 80 K, ρ_c exhibited a semiconductive T dependence. These characteristics indicate that the crystals employed are in the underdoped regime. The contact resistance was less than 1 Ω and negligible in the *I*-*V* curve measurements.

Figure 1 shows typical *I*-*V* characteristics observed for a 7.5 nm thick stack at 4.2 K, in which seven voltage steps can be seen. The maximum Josephson currents I_c of the constituent junctions range from 0.15 to 1.4 mA (40 to 350 A/ cm^2). This I_c distribution is probably due to a variation in the barrier layer conductance, which tends to vary depending on the oxygen content. The first five resistive branches are separated with a voltage interval V_j of approximately 10 mV. The sixth and seventh voltage branches are very close with a separation of less than 1 mV, strongly indicating that the bottom junction is resistively shunted with a small resistance of less than 0.5 Ω caused by the ion milling process. Therefore, it is safe to regard this stack as having five effective junctions. The characteristics of other stacks are always accompanied by resistive branches of this kind.

The I_c value of this stack is much smaller than previously reported values.[12,13,15,19] The reduced I_c value is likely to be due to an increase in tunneling resistance R_N, which is caused by oxygen desorption from the crystal surface during the process. The smaller voltage interval of $V_j \sim 10$ mV results from the smaller I_c values. This is easily understood when we take into account the shape of the resistive branches, for which the d-wave order parameter is responsible, as shown later.

We can also see several jumps in the *I*-*V* curve in the voltage state. Similar jumps have often been observed in the *I*-*V* curves of $Bi_2Sr_2CaCu_2O_8$ intrinsic Josephson junctions.[20] We presume that these jumps are of the same origin as those observed by Schlenga *et al.*[20]

Figure 2 shows the *I*-*V* characteristics of the same stack over an extended range. The gap structure can be clearly seen at $V = 287$ mV. Above 287 mV, the linear normal region is also seen and this is in contrast to the characteristics observed by Yurgens *et al.*[16] For our evaluation of $2\Delta_0$, we assume that each junction in the stack has an identical R_N value. This assumption seems reasonable for at least three or four junctions in the stack when we take the I_c distribution into account. We obtained a gap value $2\Delta_0$ of 57 meV for a single junction by using the junction number $N = 5$. This value is almost equal to the $2\Delta_0$ values obtained by STS analyses.[7,9]

With thick stacks, the *I*-*V* curves are always accompanied by negative resistance or distortion induced by the gap suppression due to the nonequilibrium superconductivity effect or Joule heating.[14,15] By contrast, the *I*-*V* curve in Fig. 2 has no negative resistance. In light of the previously reported numerical calculation results, the curve even appears to have little distortion caused by gap suppression. In order to confirm this almost complete absence of gap suppression, the *I*-*V* curve is compared with the numerical calculation in Fig. 3, based on the d-wave superconductor SIS junction model, which provides good numerical fits to the characteristics for thicker stacks.[15,17]

We calculated the *I*-*V* curve for a d-wave SIS junction using the following equations:

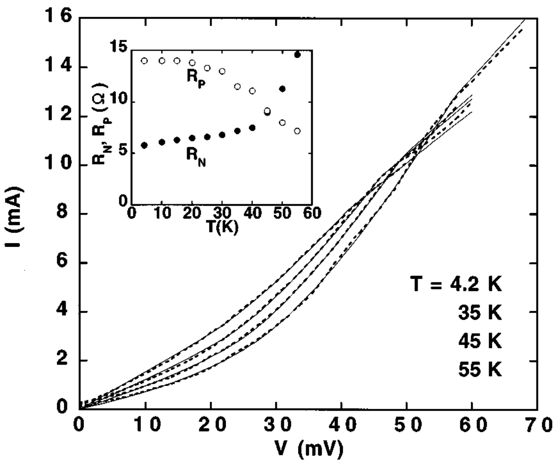

FIG. 3. A series of calculated *I-V* curves (solid lines) and experimental results (broken lines) at various temperatures. The inset shows the normal tunneling resistance R_N (solid circles) and the parallel resistance R_p (open circles) used for the fits.

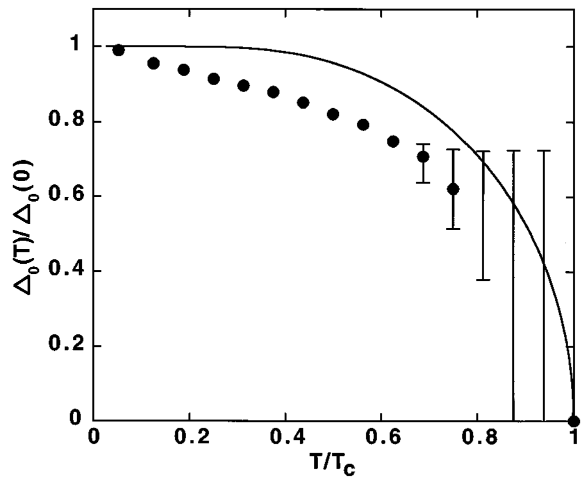

FIG. 4. Temperature dependence of the gap magnitude estimated from the gap edge (solid circles). The data is normalized with $2\Delta_0(0)=58$ meV. The solid line is the BCS order parameter.

$$I(V)=\int_{-\infty}^{\infty} N(E)N(E-eV)\{f(E-eV)-f(E)\}dE, \quad (1)$$

$$N(E)=\mathrm{Re}\int_0^{2\pi}\frac{1}{2\pi}\left[\frac{E}{\sqrt{E^2-\Delta(\theta)^2}}\right]d\theta, \quad (2)$$

where $N(E)$ is the normalized quasiparticle density of states and $f(E)$ is the Fermi function. For the numerical fitting, we assumed a finite parallel leakage resistance R_p. The details of the numerical calculation have already been reported.[17]

Figure 3 compares the calculated *I-V* curves (solid lines) with the experimental results (broken lines) at various temperatures. For the experimental curve observed at 4.2 K, the best fit is obtained when $R_N=5.8\ \Omega$ and $R_p=14\ \Omega$. It is obvious that the experimental *I-V* characteristics with a large subgap conductance are reproduced by the calculation based on this 2D pure *d*-wave superconductor model. It should be noted that the above calculation does not include the gap suppression due to the nonequilibrium heating effect. Therefore, the excellent agreement between the calculated curves and the experimental results indicates that the gap suppression due to the nonequilibrium effect and the heating effect is negligible. This result implies that the nonequilibrium superconductivity effect in layered superconductors is significantly enhanced when the layer number is large as indicated previously.[15] The most important consequence of these numerical fits is that we can estimate $\Delta_0(T)$ in the absence of gap suppression arising from the nonequilibrium heating effect, which is probably T dependent in an as yet unknown way.

Figure 4 shows the T dependence of $2\Delta_0(T)$ normalized with an extrapolated value of $2\Delta_0(0)$ = 58 meV. In Fig. 4, there are no values $\Delta_0(T)/\Delta_0(0)$ values in the T region near T_c since the *I-V* curve becomes smoother near T_c and the gap edge is indiscernible. Thus, numerical fits in this T range failed to provide definite values for $2\Delta_0$. Therefore, only possible ranges for Δ_0 are indicated with error bars based on parameters which give reasonable fits.

Figure 4 clearly shows that $2\Delta_0$ for the Bi system has a T dependence different from that of the BCS theory which is depicted with a solid line. The most significant difference is the absence of $\Delta_0(T)$ saturation when $T/T_c=t\rightarrow 0$. In the BCS theory, the order parameter is assumed to be **k** and E independent, which leads to the $\Delta(T)$ saturation when $t\rightarrow 0$. For *d*-wave superconductors, Δ is **k** and E dependent in the self-consistent equation, resulting in a T dependence different from the BCS theory. However, the explicit form for $\Delta_0(T)$ is not clear at present because of analytical complexities. The present result shown in Fig. 4 is the first experimental data for $\Delta_0(T)$ in a *d*-wave superconductor, provided $Bi_2Sr_2CaCu_2O_8$ has *d*-wave pair symmetry.[19,21]

In the inset in Fig. 3, the R_N and R_P values used for the fits are plotted as a function of T. Below T_c, R_N is almost T independent, which agrees with our previous work.[15] Above 30 K, however, R_N increases with increasing T by a factor of 2, and this is accompanied by a significant decrease in R_p. This suggests that the tunneling behavior near T_c cannot be explained solely in terms of a simple SIS junction. The origin of the increase in R_N remains to be determined. The behavior of the normal tunneling part in the *I-V* curve should be understood in connection with a pseudogap recently found in this system.[19]

In conclusion, we have fabricated very thin stacks of $Bi_2Sr_2CaCu_2O_8$ intrinsic Josephson junctions. The *I-V* characteristics of the stacks are in good agreement with those of *d*-wave SIS junctions. A numerical fit provides a value of $2\Delta_0=57$ meV at 4.2 K for the $Bi_2Sr_2CaCu_2O_8$ systems. Furthermore, $2\Delta_0$ increases as $t\rightarrow 0$, which is different from the behavior predicted by the BCS theory. The present result for $2\Delta_0(T)$ is considered to provide experimental evidence for the temperature dependence of the maximum magnitude of the *d*-wave order parameter.

The authors would like to thank Dr. T. Makino for his encouragement and interest in this work. They also thank Dr. S. Kubo, Dr. K. Miyahara, and K. Tsuru for useful discussions.

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