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High-temperature reverse ac Josephson effect in $YBa_2Cu_3O_7$

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The reverse ac Josephson effect in single-phase polycrystalline $YBa_2Cu_3O_7$ has been measured in the range $80 < T < 360$ K. The temperature dependence was found to be of the form $V = ae^{bT}$, and the effect persisted up to 300 K. We also show that the partial superconductivity indicated by the reverse ac Josephson effect is not a product of superconducting fluctuations from the 90-K supeconductivity phase of $YBa_2Cu_3O_7$. It is argued that the partial superconductivity is the property of small regions of inhomogeneous crystalline structure.

INTRODUCTION

Since its discovery in 1987 (Ref. 1) the ceramic superconductor $YBa_2Cu_3O_7$ has been extensively studied. A number of these investigations report partial superconductivity[2-7] in $YBa_2Cu_3O_7$ at temperatures well above its nominal critical temperature of 90 K. More specifically, in 1987 Chen *et al.*[2] reported reverse ac Josephson-effect measurements in bulk multiphase samples of Y-Ba-Cu-O. This was soon followed by a similar report by Gupta *et al.*[3] who had made similar measurements on 30-μm-thick films of $YBa_2Cu_3O_7$. In both these reports the reverse ac Josephson effect was measurable up to 240 K. The limited data available and the use of multiphase samples have made the analysis of the origin of the partial superconductivity difficult. The all important question of whether the partial superconductivity is due to superconducting fluctuations of the 90-K phase or an unidentified material with a critical temperature well above 90 K has remained unanswered. In this paper reverse ac Josephson-effect data are presented which shows that the partial superconductivity persists up to 300 K in single-phase polycrystalline $YBa_2Cu_3O_7$. The results also show that in single phase $YBa_2Cu_3O_7$ the temperature dependence of the reverse ac Josephson effect is reproducible. From the temperature dependence of the reverse ac Josephson-effect data it is argued that the partial superconductivity measured at 300 K is not the product of superconducting fluctuations from the 90-K phase. A possible source for the superconductivity is suggested.

THE EXPERIMENTS

The reverse ac Josephson effect was measured in $YBa_2Cu_3O_7$ in the temperature range $80\ \mathrm{K} < T < 360$ K. Experiments were performed with ac currents in the 1–65-MHz (rf) frequency ranges. The dc voltage generated in the sample was measured across the voltage leads of the sample (Fig. 1) using a digital meter.

The sample was mounted on a glass substrate on which four silver leads had been made by vacuum deposition. Electrical contact between the sample and the silver leads was obtained using Silver Print.[8] The sample was mounted in a double Dewar cryogenic cooling system. The temperature dependence of the reverse ac Josephson effect was measured with the rf supplied by either (A) a current source across the current leads of the sample or by (B) an antenna placed in the vicinity of the sample (approximately 5 cm away). The second method ensured that the dc voltages measured did not originate from a dc offset of the rf generator.

In all cases when the temperature was varied care was taken to measure the Seebeck voltage (thermopower) across the sample before applying the ac bias to measure the induced dc voltage, V_{dc}. To minimize the Seebeck signal the temperature was varied slowly (2–3 h) from 300 to 80 K. Three types of experiments were performed:

(1) temperature dependence of V_{dc} for a constant frequency (ω) and a constant generator output (V_{ac});

(2) the ac power dependence of V_{dc} at constant frequency and temperature;

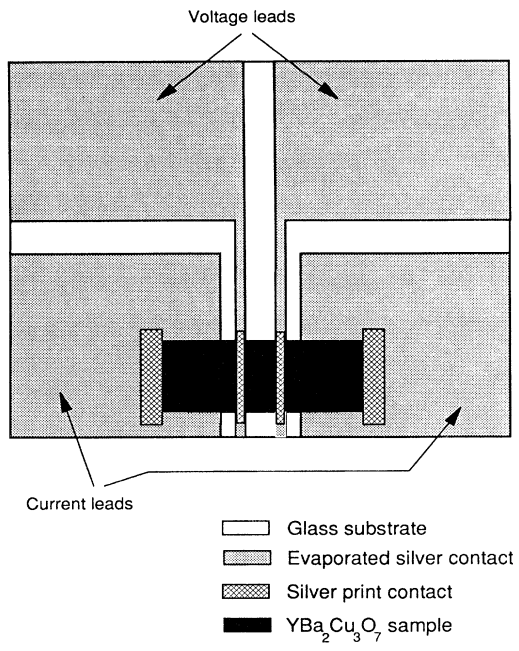

FIG. 1. $YBa_2Cu_3O_7$ sample mounted on a glass substrate for reverse ac Josephson-effect measurements.

(3) the frequency dependence of V_{dc} at constant ac generator output and constant temperature.

The samples used in the experiments were rectangular pieces (1 mm×3mm in area and up to 15 mm long) cut from pellets 2 cm in diameter. The pellets were prepared by solid state reaction[9] of stoichiometric proportions of Y, Ba, and Cu from the oxide or carbonate of each element. X-ray powder diffraction and electron microscope microdiffraction showed the samples to be basically single-phase polycrystalline $YBa_2CU_3O_7$. By basically we mean that no minority phases were seen by x-ray diffraction but that electron microscopy reveals crystalline defects. The predominant defects were twins. Room temperature resistivity of the two samples used in this investigation was 4400 $\mu\Omega$ cm.

RESULTS

The temperature dependence of the reverse ac Josephson effect in $YBa_2Cu_3O_7$ was investigated using rf in the temperature range $80\text{ K} < T < 360\text{ K}$. The temperature dependence of V_{dc} was measured for several rf frequencies (5.0, 14.5, 6.6 Mhz) and no significant difference was found between the V_{dc} versus T curves at each frequency. Furthermore, no difference was found between the V_{dc} versus T curves acquired with either the current source or the antenna methods of supplying the ac.

Between 100 and 250 K the reverse ac Josephson-effect dc voltage, V_{dc}, decreases exponentially with increasing temperature (Figs. 2 and 3). The best fits to a V_{dc} versus T curve require two or three separate exponential curves of the form $V_{\text{dc}} = ae^{bT}$. Figures 2 and 3 show typical V_{dc} versus T curves requiring two or three exponentials to fit the data between 100 and 250 K. As the temperature varies, V_{dc} jumps randomly between the different exponential fits. A few points lie between the fitted exponentials. At the temperatures where V_{dc} does not lie on an exponential, the voltage changes across the sample are too rapid for the digital multimeter to follow (the digital multimeter needs $\frac{1}{3}$ sec to measure a voltage value) and the final value displayed by the digital multimeter is a time-averaged value of the potential across the sample. The data from the five different V_{dc} versus T curves can all be fitted using only two values of b: $b = -0.0195 \pm 0.0015\ \text{K}^{-1}$ or $b = -0.0110 \pm 0.0015$

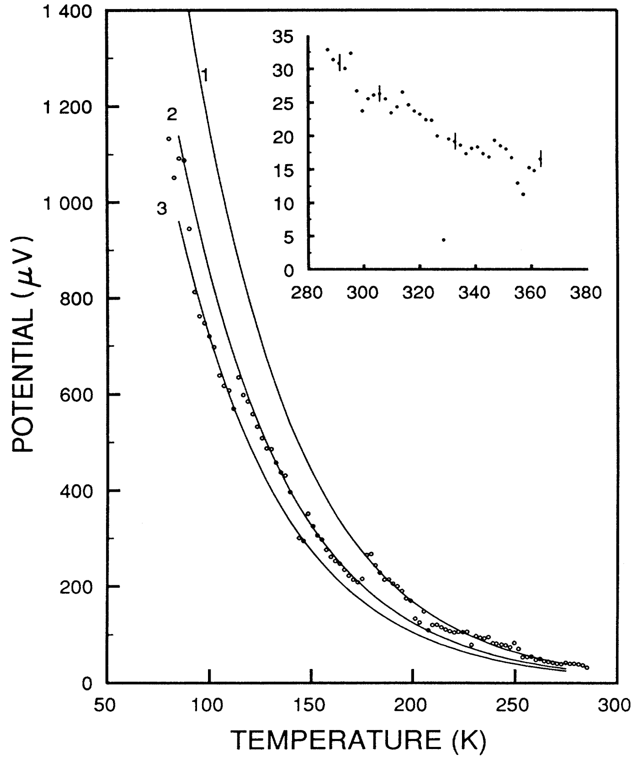

FIG. 2. Temperature dependence of the reverse ac Josephson-effect voltage for an ac bias of 14.5 MHz. The solid lines are exponential fits to the data. For curve 1, $a = 4900\ \mu\text{V}$ and $b = -0.01925\ \text{K}^{-1}$, for curve 2, $a = 5850\ \mu\text{V}$ and $b = -0.01925\ \text{K}^{-1}$, and for curve 3, $a = 7870\ \mu\text{V}$ and $b = -0.01925\ \text{K}^{-1}$. The inset shows reverse ac Josephson-effect voltage versus temperature for the high-temperature range 280–360 K.

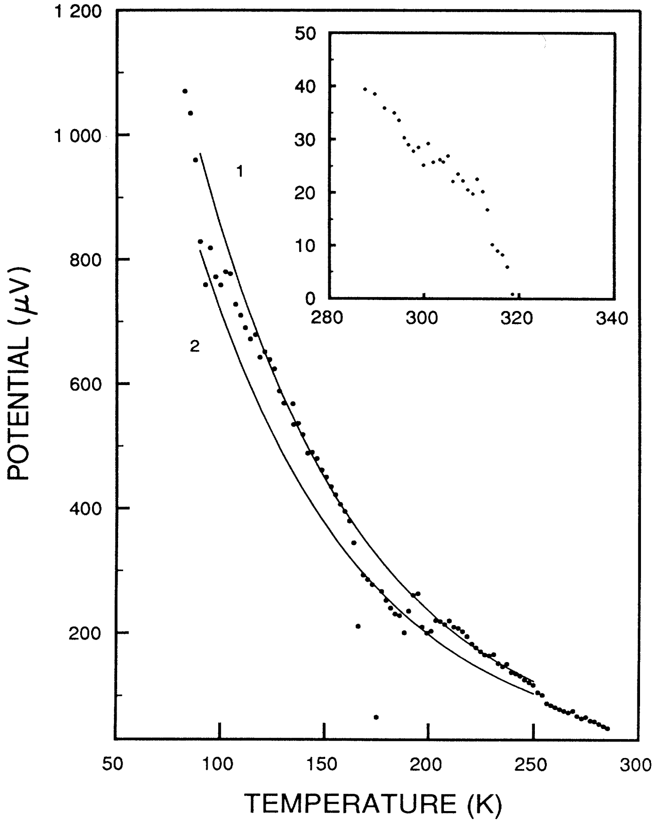

FIG. 3. Temperature dependence of the reverse ac Josephson-effect voltage for an ac bias of 12.31 MHz. The solid lines are exponential fits to the data. For curve 1, $a = 2800\ \mu\text{V}$ and $b = -0.0123\ \text{K}^{-1}$ and for curve 2, $a = 3100\ \mu\text{V}$ and $b = -0.0121\ \text{K}^{-1}$. The inset shows reverse ac Josephson-effect voltage versus temperature for the high-temperature range 280–320 K.

K^{-1}. Thus the exponential fit parameter b is a reproducible parameter which can be used to demonstrate the reproducibility of the temperature dependence of the reverse ac Josephson effect in $YBa_2Cu_3O_7$.

Above 250 K, V_{dc} continues to decrease with increasing temperature, although an exponential fit is not possible for $T > 250$ K. In all five experiments V_{dc} was clearly nonzero at 300 K. The temperature at which V_{dc} becomes zero or less than the measurement error is different for each curve and lies between 320 and 350 K.

The temperature dependence of the reverse ac Josephson effect was also measured with microwaves as the ac source (Fig. 4). For this experiment the sample was placed in a microwave guide[9] and V_{dc} was measured by modulating the microwave power at 1000 Hz and measuring the amplitude modulation of V_{dc} with an oscilloscope. Figure 4 shows V_{dc} versus T for two different microwave frequencies. Again V_{dc} is nonzero at 300 K and the data between 100 and 250 K can be fitted to the equation $V_{dc} = ae^{bT}$. Once more the data can be fitted with $b = -0.0110$ K^{-1} which falls within the range of values used to fit the data obtained with rf ac bias.

The frequency dependence (0–60 MHz) of V_{dc} was measured at room temperature. Figure 5(a) shows the frequency dependence of V_{dc} at 300 K when V_{dc} is measured using a digital multimeter. To eliminate the possibility that V_{dc} originated from a frequency dependent dc offset of the ac generator output, the experiment was repeated using a lock-in amplifier [Fig. 5(b)]. The important feature of the V_{dc} versus frequency curves is the change in polarity of the dc voltage. The change in polarity of V_{dc} with frequency is a feature of the reverse ac Josephson effect which allows the reverse ac Josephson effect to be differentiated from rectification effects. At frequencies larger than 30 MHz and up to 60 MHz, V_{dc} is zero.

Figure 6 shows the rf power dependence of V_{dc} in $YBa_2Cu_3O_7$ at room temperature as measured with the lock-in amplifier and digital multimeter at two different rf frequencies. Again the important feature is the reversal of polarity of V_{dc} with power. Figure 6 also shows a reversal in polarity between the two test frequencies at which the V_{dc} versus V_{ac} curves were taken. This is in agreement with the frequency dependence data of Fig. 5. The ac power dependence of the reverse ac Josephson effect at microwave frequencies was also measured at 296 and 86 K (Fig. 4). No reversal of sign of V_{dc} occurred with changing microwave power.

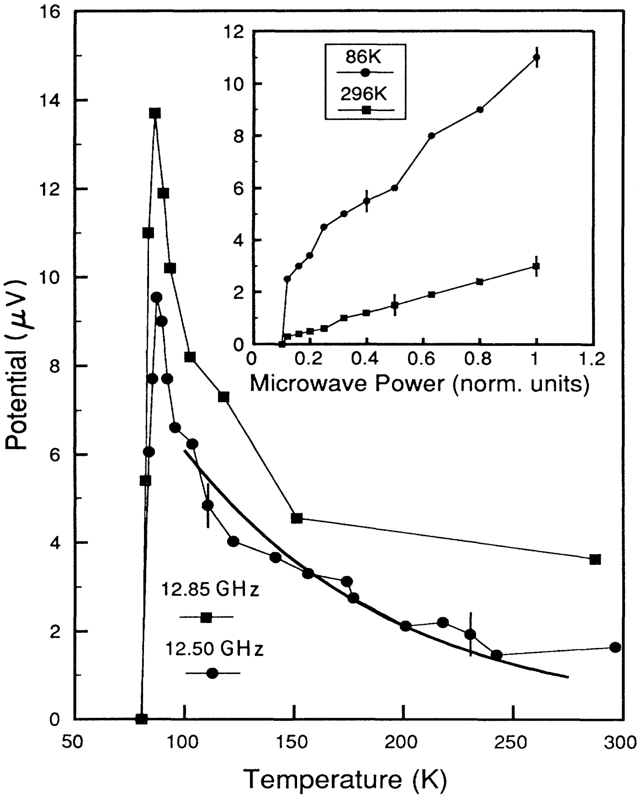

FIG. 4. Temperature dependence of the reverse ac Josephson-effect voltage with microwaves as the ac bias. The thick line is an exponential fit using $b = -0.011$ K^{-1}. The microwave power dependence of V_{dc} is shown in the inset for $T = 86$ K and $T = 296$ K.

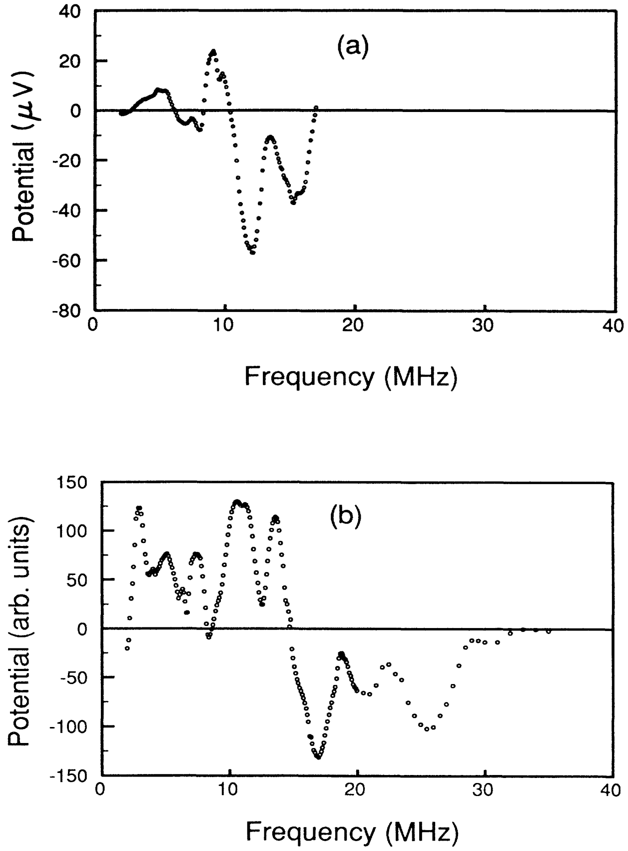

FIG. 5. Frequency dependence of V_{dc} in $YBa_2Cu_3O_7$ at 300 K as measured with (a) a digital multimeter and (b) a lock-in amplifier.

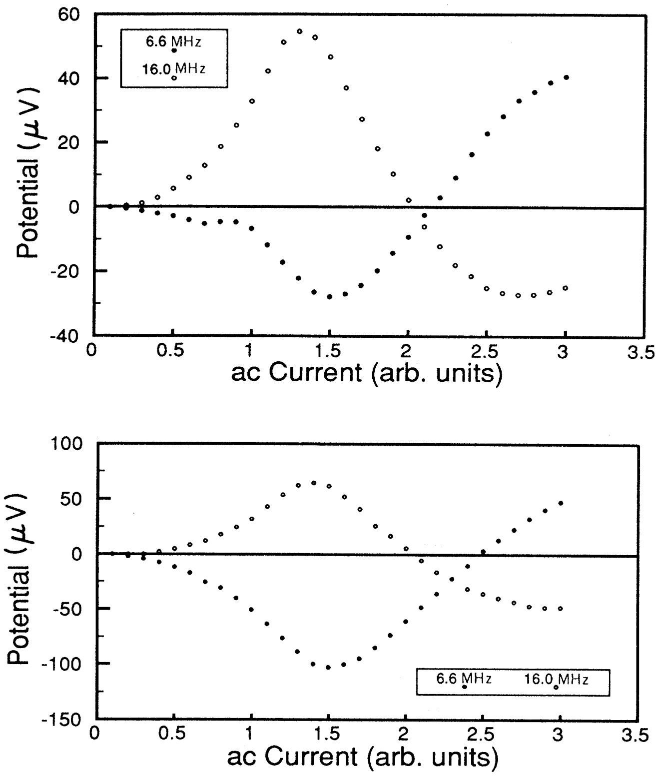

FIG. 6. AC power dependence of V_{dc} in $YBa_2Cu_3O_7$ at 300 K as measured with lock-in amplifier. The two sets of data were obtained from the same sample with different sets of contacts to demonstrate the repeatability of the measurement.

DISCUSSION

The first report of measurements of induced dc voltages by ac currents in bulk samples of $YBa_2Cu_3O_7$ was published by Chen *et al.*[2] In this paper Chen reports that V_{dc}:

(A) increases as T approaches T_c and is nonzero up to 240 K; (B) is measured for frequencies of 14.5 MHz, 5 MHz, 8.28 GHz, and 8.25 GHz and changes polarity with frequency;

(C) is ac power dependent (for microwave and RF) with polarity changes;

(D) varies between samples from a few microvolts to a few millivolts.

The samples which produced the largest reverse ac Josephson-effect signal were identified by the above authors as being fabricated intentionally to be multiphase. They report that their single-phase material had measurable V_{dc} only at T near T_c. Chen *et al.* conclude that in multiphase samples some grains of unknown composition (supposed to be different than $YBa_2Cu_3O_7$) are superconducting at $T > T_c$. Some of the superconducting grains are in close proximity and superconducting weak lines are formed. The ac current induced V_{dc} is a manifestation of the reverse ac Josephson effect in the weak links. In Ref. 2 the temperature dependence of V_{dc} results vary greatly from sample to sample. Some of the data displayed a temperature dependence similar to the exponential dependence reported in this paper.

Gupta *et al.*[3] also report reverse ac Josephson-effect measurements on $YBa_2Cu_3O_7$ (resistivity at 300 K larger than 4000 $\mu\Omega$ cm) for temperatures up to 230 K. In that paper V_{dc} is measured at 9.8723 GHz and has an exponential temperature dependence between 100 and 220 K (we were able to obtain fits to Gupta's data using $a = 1050\text{-}\mu V$ and $b = -0.354\ K^{-1}$). Gupta *et al.* show a linear dependence of V_{dc} with microwave power at 77 K, this is in agreement with the data presented here (Fig. 4).

For high resistivity multiphase samples, nonsuperconducting phases interfere with the reverse ac Josephson-effect measurements at $T > T_c$. Because of this the results are difficult to reproduce in multiphase samples. The presence of multiple phases will also make it difficult to determine which phase or phases are responsible for the reverse ac Josephson-effect signal.

In the Chen and Gupta papers the samples are multiphase, and the authors suggest that for $T > T_c$ the weak links form at the interface between superconducting grains (of phase other than $YBa_2Cu_3O_7$). Given that the reverse ac Josephson effect is nonzero in single phase $YBa_2Cu_3O_7$ it is highly unlikely that there is a sufficient quantity of any minor phase in the sample to form grains with a structure other than $YBa_2Cu_3O_7$. We propose that at $T > T_c$ the regions of superconductivity are probably not superconducting grains but smaller localized regions, many of which can coexist within one grain. Some regions of superconductivity are in close enough proximity to form weak links which can exhibit the reverse ac Josephson effect. Munger and Smith[9] have investigated the effect of magnetic fields on the current-voltage curves of single phase $YBa_2Cu_3O_7$ for $T_c < T < 300$ K. They conclude that in that temperature range the average area of the weak links is of the order of $(1000\ \text{Å})^2$.

The 1972 Chen *et al.*[10] explained the reverse ac Josephson effect and showed that

$$V_{dc} = n\frac{\hbar\omega}{q} = -RI_J J_n\left[\frac{qV_{ac}}{\hbar\omega}\right]\sin\phi_n\ , \tag{1}$$

where R is the resistance of the external measuring circuit, I_J the critical current of the device, J_n the Bessel functions of the nth order, and n is an integer. In Eq. (1) the only temperature-dependent term is the critical current of the weak link, I_J. The temperature dependence of I_J is determined by the type of device under investigation (Dayem bridge, point contact, S-N-S weak link, etc.) and the properties of the superconducting material. By studying the temperature dependence of V_{dc} it should be possible to obtain information about the type of weak link from which the reverse ac Josephson originates. From the device type the origin of the partial superconductivity at 300 K can then be inferred.

The three possible sources for the partial superconductivity are (a) superconducting fluctuations from the 90-K superconductor, (b) a small amount of superconducting material with critical temperature greater than or equal to 300 K, and (c) a small amount of material with critical temperature well above T_c but less than 300 K.

If a new superconductor with a critical temperature between T_c and 300 K is responsible for the partial superconductivity in $YBa_2Cu_2O_7$ for $T > T_c$, there should be some evidence of a phase transition at the critical temperature of this material. There is no evidence of a superconducting phase transition in any of the five V_{dc} versus T curves or in the resistance versus temperature data.[9]

To identify the source of the partial superconductivity at $T > T_c$ it is then necessary to distinguish between superconducting fluctuations from the 90-K superconductivity or a new material with a critical temperature greater than 300 K. We conclude that the partial superconductivity is not a measurement of superconducting fluctuations from the 90-K superconductivity for the following reasons.

Measurements of the temperature dependence of the excess conductivity, and diamagnetic susceptibility have been done on $YBa_2Cu_3O_7$.[11-16] The analysis of the temperature dependence data agrees very well with the dirty limit (short coherence lengths) of the theories of fluctuations[17-19] developed for explaining the data from BCS superconductors. The best fits to the $YBa_2Cu_3O_7$ data are obtained using the Lawrence-Doniach[18] (L-D) model which was developed to calculate the effect of fluctuations on the transport properties of layered superconductors. The conclusion that $YBa_2Cu_3O_7$ is a layered superconductor is in agreement with the crystallographic[20-23] data which has demonstrated that $YBa_2Cu_3O_7$ has a layered structure, and coherence length estimates.[11,24] As for the low temperature superconductors the highest temperature at which any evidence of fluctuations can be detected in $YBa_2Cu_2O_7$ is $\approx 2T_c$, which for $YBa_2Cu_2O_7$ is approximately 200 K. Data from Friedman, presented in Ref. 11, shows that above the 200-K fluctuation contributions to the excess conductivity go to zero. This is well below 300 K where we find that V_{dc} is nonzero. Furthermore Kulik[25] has calculated the temperature dependence of the critical current for a weak link formed using superconducting fluctuations. He found that

$$I_J \propto \frac{T_c}{T-T_c}$$

which is not equivalent to the exponential temperature dependence found in our data. We therefore conclude that the partial superconductivity is the result of a small amount of material with a critical temperature greater than or equal to 300 K.

After examining all the possible types of weak links the only device which showed an exponential-like temperature dependence of I_J was the superconductor–normal metal—superconductor (S-N-S) weak link, where the normal metal has a superconducting critical temperature lower than that of the superconducting electrode. The exact form of the temperature dependence of I_J (Ref. 26) is

$$I_J \propto (T-T_S)^2 e^{-d_n K_n}$$
$$\text{with } K_n = \left[1+\frac{2}{\ln(T/T_n)}\right]^{-1} \left[\frac{2\pi k_B T}{hD_n}\right]^{1/2}, \quad (2)$$

where T_n and T_s are the critical temperatures of the normal metal and superconductor, respectively, d_n the thickness of the metal, and D_n the diffusion coefficient of the metal. This equation cannot be easily simplified to an exponential and too many parameters are unknown for a fit of the data with Eq. (2). To show the correspondence between this equation and the exponential fit of the data, values of I_J were calculated using Eq. (2) for 100 K $< T <$ 250 K and an exponential was fitted to the calculated points. Figure 7 is a plot of the calculated I_J's normalized to $I_J(T=T_n)$ with the exponential fit superimposed. In the calculation the following values were given to the different parameters: $T_n = 90$ K, $T_s = 350$ K, and $d_n(2\pi k_B/hD_n)^{1/2} = 1$. The last product is equivalent to assuming that the barrier thickness is of approximately the same size as the distance which superconducting pairs can travel into the normal metal. The correlation of the fit to the calculated points is better than 0.999. This shows that the weak links in $YBa_2Cu_3O_7$ are S-N-S weak links and that superconducting fluctuations of the 90-K

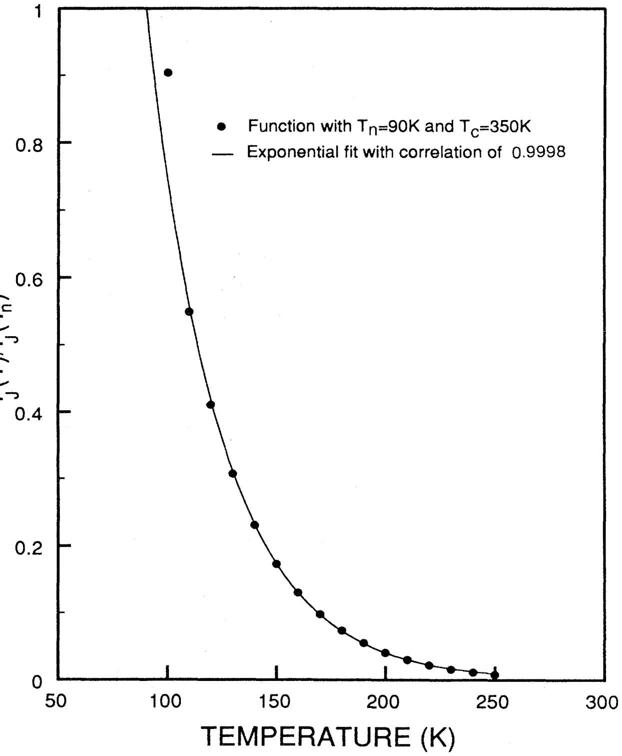

FIG. 7. Exponential fit to the temperature dependence of the critical current of a S-N-S device when calculated using Eq. (2). The normal metal has a critical temperature well below the critical temperature of the superconductor. The critical current is normalized to the critical current of the device at $T = T_n$.

phase are not responsible for the partial superconductivity at $T > T_c$.

Crystallographic investigations[9] of samples taken from the same pellets as the samples used in this investigation have shown that the samples are basically single phase. Thus it is expected that the superconducting material responsible for the partial superconductivity is not a new ordered phase of Y-Ba-Cu-O. Therefore the idea that the weak links are formed at interface between superconducting grains[2,3] does not apply for the samples used in this project. Furthermore, in Ref. 9 it was calculated that the average area of contact between areas of superconductivity forming weak links is of the order of 10^6 Å^2. This is much smaller than the size of weak link expected from the grain boundaries of 30-μm diameter superconducting grains. It has been demonstrated by microdiffraction studies[27,28] that even single crystals of $YBa_2Cu_3O_7$ have inhomogeneous regions spanning distances of the order of 1000 Å. The crystals contain small deviations from the average $YBa_2Cu_3O_7$ structure over that distance. Within that distance the crystal structure changes continuously creating a small region of disordered crystalline structure. In these and other crystallographic studies[20,23] it has also been shown that $YBa_2Cu_3O_7$ is heavily twinned and that the twinning distance is of the order of 1000 Å.

Based on these arguments the authors suggest that the partial superconductivity is a property of the inhomogeneous material in regions where the crystalline structure deviates from the ideal $YBa_2Cu_3O_7$ structure. This might occur in the inhomogeneous regions discussed above or in the vicinity of crystalline defects (for example, twins).

The switching between exponential curves (Figs. 2 and 3) can also be explained using Eq. (1). From this equation it can be seen that a number of V_{dc} values can be measured for a particular ac bias. Each different V_{dc} will correspond to solving Eq. (1) for a different value of the integer n with V_{dc} an integer multiple of $h\omega/q$. The temperature dependence of V_{dc} will still follow the temperature dependence of I_J. All of the solutions which satisfy Eq. (1) are equivalent so that the device can alternate randomly between the different solutions. This type of behavior has been observed experimentally in Josephson junctions.[10] In $YBa_2Cu_3O_7$ each of the two exponentials represents an equilibrium state for the multiple weak link system. The ratio of the b values is $(1.95/1.10) \approx 2$, which would indicate that the array is switching between the $n=1$ and $n=2$ states of the system. At any temperature there might be more than one equally probable state for the array and the array oscillates between the possible voltage states.

Jumps between exponentials of same b values (but different a values) can possibly be the result of structural instabilities in the granular structure. As the temperature changes structural changes due to temperature gradient stresses unlocks some parts of the array which are very weakly linked to other portions of the array. In parts of the sample the coherence between the weak links is broken down (some weak links are disconnected from the rest of the sample) and V_{dc} is reduced. As the temperature changes temperature gradient stresses change and the areas of the samples which act coherently change. In this way V_{dc} can jump several times from exponential to exponential with changing temperature. Another possible explanation for the jumps between an exponential with the same b values is that $YBa_2Cu_3O_7$ undergoes some kind of electronic transition. In such situations the transition occurs because the new electronic state is energetically favorable for lower temperatures. After such a transition occurs it is not energetically favorable for the system to return to the original electronic configuration at any temperature below the transition temperature. In the V_{dc} versus T data, V_{dc} sometimes jumps from one exponential to a second and back to the first as the temperature is lowered. These types of jumps are not characteristic of electronic transitions but are characteristic of structural instabilities.

Alternative explanations for ac to dc conversion in granular materials have been proposed.[29,30] Ikegawa *et al.*[29] suggest that the ac to dc conversion is the result of driving a nonlinear current-voltage characteristic with an ac bias containing harmonics of the fundamental frequency. They argued that in order to obtain a dc potential in the presence of an ac bias then $(d^2V/dI^2)_{I_{dc}=0} \neq 0$. They also show that to obtain a change in the polarity of V_{dc} it is necessary to change the phase between the fundamental and the second harmonic, and to have $(d^3V/dI^3)_{I_{dc}=0} \neq 0$. First we do not believe this model would apply to the data reported in this paper as the current-voltage characteristics of our samples are ohmic at small currents so that $(d^2V/dI^2)_{I_{dc}=0} = (d^3V/dI^3)_{I_{dc}=0} = 0$. Secondly, our results show that at rf frequencies V_{dc} will change polarity with I_{ac}. This is not predicted by the model of Ikegawa *et al.*

A different method of ac to dc conversion has been discussed by Gerber and Deutscher[30] who showed that ac to dc conversion can occur in percolated films of lead or aluminum of average loop size 3000 and 1500 Å, respectively. They interpreted their results in terms of the Aharonhov-Bohm effect. In this model quantum coherence occurs when the phase-coherence length of the electrons is comparable to the average diameter of the rings in the percolated film. This effect will be masked by the presence of any superconducting material which has a much longer coherence length. As our samples do not have a percolated ring structure comparable to the phase-coherence length of normal electrons this model could not be applied to the data presented in this paper.

CONCLUSIONS

In this paper it has been shown that a dc voltage appears across $YBa_2Cu_3O_7$ samples if an ac bias is applied. AC frequency and power dependence of the dc voltage confirm that the reverse ac Josephson effect is the source of the dc voltage. This suggests that single phase $YBa_2Cu_3O_7$ is partially superconducting at temperatures well above its nominal critical temperature. All samples tested had a measurable reverse ac Josephson effect at 300 K. It is shown that the partial superconductivity at 300 K is not the result of superconducting fluctuations from the 90-K superconductor.

Using the exponential temperature dependence of V_{dc} it is shown that the 300-K partial superconductivity is due to a small amount of material with a critical temperature greater than 300 K. It is argued that the superconductivity is a property of small regions of disordered structure. Such regions could originate in the vicinity of crystalline defects such as twins.

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