## 1/f Noise in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> Superconducting Bicrystal Grain-Boundary Junctions

M. Kawasaki, P. Chaudhari, and A. Gupta

IBM Research Division, T. J. Watson Research Center, Yorktown Heights, New York 10598

(Received 24 May 1991)

We have measured 1/f noise of single-grain-boundary junctions in superconducting YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> epitaxial films. The origin of 1/f noise is identified and the data are used to rule out recent suggestions for new mechanisms which generate 1/f noise. Using ozone annealing, both 1/f noise and critical current density of the grain boundary were found to improve. However, the persistence of both noise and Josephson coupling suggest that oxygen deficiency at the boundary is not the sole cause of the anomalous coupling in these materials. We speculate that the depression of the order parameter is connected with the cores of the dislocations forming an array.

## PACS numbers: 74.60.Ge, 74.40.+k, 74.70.Vy

One of the outstanding unsolved problems in the hightemperature cuprate superconductors is concerned with understanding the microscopic origin of the unusual superconducting behavior of grain boundaries. What is known is that they are weak links [1]; the critical current across the boundary is limited by flux flow for small angles of misorientation and by Josephson coupling for large angles [2-8]. The weak-link behavior is reproducible. It presents difficulties in the fabrication of superconducting wires and an opportunity to produce superconducting quantum interference devices (SQUIDs). The latter have been built by several groups [7-11] and, in fact, the energy sensitivity, determined by the white noise level at 77 K, was recently shown [8] to be  $3 \times 10^{-31}$ J/Hz. This value is approximately 450 times the Planck's constant and satisfactory for almost all conceivable applications.

In an attempt to further narrow down the possible mechanisms responsible for the weak-link behavior of the grain boundary, we have been studying 1/f noise generated by the boundary. Our rationale for this approach stems from the observation that 1/f noise from conventional junctions is well understood in terms of trapping of carriers and their subsequent release subject to a distribution of activation energies or trapping times [12,13]. Hence if we could identify the source of 1/f noise in the grain boundaries to be similar to that of conventional junctions, we could then apply many of the concepts developed in that field. Furthermore, if we could perturb the structure of the boundary in a reproducible way, it might help in guiding us to the microstructural element responsible not only for noise, but also for the weak-link behavior. In addition to this straightforward approach, we were also interested in designing experiments that would enable us to compare the magnitude of 1/f noise we measure with recent theoretical proposals [14-17].

The details of growing bicrystals, depositing epitaxial films, and patterning have been described in earlier publications [3,7,8]. The data we present here were obtained on a 25° grain-boundary junction of width 10  $\mu$ m. Following measurements of noise and other transport properties, the bicrystal films were annealed in ozone in an attempt to disturb the oxygen level at the boundary.

The samples were annealed in ozone at 100 °C for approximately half an hour. This temperature was found by trial and error, in a separate study of bicrystal films, to result in changes of the grain-boundary properties without any measurable changes in the bulk of the film. Ozone annealing was carried out by exposing the films to the ultraviolet radiation from a low-pressure mercury lamp in the presence of 1 atm pressure of oxygen. Ozone rather than oxygen was used to provide a large chemical potential or driving force for diffusion of oxygen into the grain boundary [18]. We note that the effects of ozone annealing largely disappear within a few days when the sample is left at ambient conditions.

There can be at least two sources of 1/f noise in a superconductor containing a Josephson-coupled weak link. One of these can be associated, quite generally, with flux motion and the other, with noise arising from scattering by imperfections in the Josephson-coupling region. It is not readily possible to separate these two sources if a single Josephson-coupled weak link is studied. If the flux line (or flux bundle) moves about its mean position, the change in magnetic field results in a change of phase across the junction and hence in the critical current. Fluctuations in the critical current arising from flux motion cannot be distinguished from those caused by changes in the junction transmission induced by scattering or trapping of carriers.

This distinction is of particular interest because a recent theoretical paper predicts that 1/f noise from flux motion in short-coherence-length materials may be high [14]. This noise is generated by universal conductance fluctuations. They are present in the high-temperature superconductors due to the motion of flux lines, which have a small core diameter defined by the short coherence length. As the film and junction are in series, the noise cannot be simply partitioned experimentally between junction or flux-motion-induced 1/f noise.

We have resolved this experimental difficulty following earlier work [7,8,19] by studying noise in SQUIDs, which have two junctions in a closed loop, rather than a single junction. Any flux change in the loop associated with motion of flux lines is picked up by the SQUID. Measurements show that flux motion does indeed contribute to 1/f noise but dominates only at temperatures close to the superconducting transition temperature [7,8]. The data we report here are only for those temperatures for which flux noise is not dominant. Hence, we arrive at our first conclusion: Universal conductance fluctuations are not the dominant source of 1/f noise in the hightemperature superconductors except, perhaps [20], close to  $T_c$ .

In our SQUID papers [7,8] we have already shown that the voltage noise power spectrum follows a  $1/f^a$ dependence, where  $\alpha \simeq 1$ . Because of recent theoretical interest on the effect of driving current on the exponent  $\alpha$ , we have measured the voltage noise power spectrum as a function of the bias current to values which are several times the critical current. We do not find the exponent  $\alpha$ to change. In the absence of such a change we shall assume that models of 1/f noise based on self-organized criticality [17] do not apply to our situation. We have already ruled out the possibility of flux motion. Accordingly, we interpret our observations within the framework of a distribution of trapping times at the junction [17,18]. Before we can do this, we need to establish the following: (1) The 1/f noise arises from critical current fluctuations; (2) these critical current fluctuations are from fluctuations in the junction transmission, i.e., the resistance of the junction also produces 1/f noise; and (3) the magnitude of the critical current and junction transmission fluctuations are correlated.

Voltage noise measured as a function of bias current for a grain-boundary junction is shown in Fig. 1. The noise increases rapidly as the bias current increases, reaches a maximum, and then decreases to a shallow minimum before increasing again. This type of variation of noise with bias current was observed in all of the measurements. It appears to be a universal behavior of the bicrystal boundaries in the YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> superconductors.

We explain this behavior by assuming that there are two sources of fluctuations which produce 1/f noise. These are critical current and junction resistance fluctuations. Although the two types of fluctuations are used as independent variables in fitting the data, we shall argue that they arise from the same source and are, in fact, not independent; the critical current fluctuations are due to fluctuations in the resistance of the junction. Hence conventional interpretation of the data should be valid.

We have shown [21] that the bicrystal grain boundary can be described by a resistively shunted junction (RSJ) with thermal noise arising from phase slippage across the boundary [22]. This model is now used to calculate the effect of a critical current fluctuation,  $\delta I_0$ , and junction resistance fluctuations,  $\delta R_n$ , in generating noise as a function of the bias current that models 1/f noise at a given frequency. This is done by allowing the amplitudes of  $\delta I_0$  and  $\delta R_n$  to vary until a fit to the voltage noise power spectrum is obtained. An example of such a calculation is shown in Fig. 1(a). In Fig. 1(b) the measured





FIG. 1. (a) Voltage noise as a function of bias current for the as-deposited sample measured at 77 K. Calculated results based on the RSJ model are also shown. The dotted line is calculated by assuming only critical current fluctuations are present and the dashed line by assuming only resistance fluctuations. The solid line assumes both critical current and resistance fluctuations are present. Data are for f=100 Hz. (b) Voltage noise as a function of dynamic resistance for the same data set as in (a).

and calculated voltage noise are plotted as a function of the dynamic resistance,  $R_d \equiv dv/dI$ . The calculated values reproduce the linear relationship seen in the experimental data. This dependence is seen for both the asdeposited and ozone-annealed samples measured at various temperatures (Fig. 2). The normalized noise value,  $S_v/R_d^2$ , which corresponds to current noise, determined from Fig. 2 is plotted as a function of critical current in Fig. 3. Within the scatter of the data, we find  $S_v$  is proportional to  $I_0^2$ . Since the maximum dynamic resistance depends on the critical current, we can combine the two. From numerical evaluation we find that the maximum dynamic resistance is proportional to  $\gamma^{0.3}$  for  $\gamma$  ranging between 3 and 60. Here  $\gamma$  is defined as  $\hbar I_0/2ek_BT$ , where  $\hbar$  is Planck's constant, e is the charge of the electron, and  $k_B$  is Boltzmann's constant. Combining this with the critical current dependence of noise we find that  $S_{c}(I=I_{0}) \propto I_{0}^{2.6}$ . Indeed this dependence is observed in



FIG. 2. Voltage noise as a function of dynamic resistance measured at various temperatures for both the as-deposited and ozone-annealed samples. Data are for f = 100 Hz.

the data reported here (see the variation in the peak value of noise as a function of temperature shown in Fig. 4) as well as for the temperature dependence of noise of submicron SQUIDs in the literature [8]. We conclude from an analysis of the data shown in Figs. 1-4 that the assumption of critical current fluctuations dominating noise at or just below the critical current is valid. We now verify the second part of the assumption: Resistance fluctuations of the junction dominate noise at large bias currents.

One of the characteristic experimental observations of grain-boundary junctions in the YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> superconductors is the lack of temperature dependence of the junction resistance  $R_n$  [3,21]. This implies that the voltage noise power spectrum of the junctions at large bias currents must be independent of temperature and, in fact, fall on a single curve, which is only a function of the bias current. Indeed this is observed, as shown in Fig. 4. The noise is quadratic with the bias current I and confirms that at these currents 1/f noise is dominated by resistance fluctuation of the junction.

Using the experimental data we can estimate the magnitude of  $(\delta I_0)^2$  and  $(\delta R_n)^2$ . We find that  $|\delta I_0/I_0|$  and  $|\delta R_n/R_n|$  for the sample on which the data shown in Fig. 1 were obtained to be  $1.0 \times 10^{-5}$  Hz<sup>-1/2</sup> and  $4.0 \times 10^{-6}$  $Hz^{-1/2}$ , respectively. Using the scaling relation of critical current with resistance for bicrystals [23,24], we find that the ratio of  $\delta R_n/\delta I_0$  is -0.4. This is remarkably close to the experimental value from noise data and suggests that the microscopic mechanism responsible for the fluctuation in critical current is the same as that for fluctuation in resistance of the grain-boundary junction. The grain boundary must therefore contain charge trapping sites. The density of such sites is significantly larger than in conventional superconductors. For example, the cross section of the smallest junction we have examined [8] is comparable in area to that of the smallest junction made of conventional metals [13] in which discrete trapping



FIG. 3. The current noise, determined from linear relationship shown in Fig. 2, as a function of critical current. Data are for f = 100 Hz.

sites could be observed.

Following an ozone anneal, the critical current of the grain boundary increases and its resistance decreases. As can be seen from Figs. 2-4 ozone annealing also decreases the amplitude of noise in both the fluctuation of critical current, from  $1.0 \times 10^{-5}$  to  $5.0 \times 10^{-6}$  Hz<sup>-1/2</sup>, and the fluctuation of resistance, from  $4.0 \times 10^{-6}$  to  $2.0 \times 10^{-6}$  Hz<sup>-1/2</sup>, respectively. Here we note that the ratio of the fluctuations,  $|\delta R_n/R_n|/|\delta I_0/I_0|$ , remained 0.4. The decrease in noise is larger than the value corresponding to a decrease in  $R_n^2$  for a given  $I_0$ . Ozone annealing therefore also reduces the sites at the grain boundary which give rise to fluctuations in resistance. We conclude



FIG. 4. Voltage noise as a function of bias currents. Resistance fluctuation is estimated from the linear relationship at large bias current. The peak values of  $S_r$ , observed as a function of critical current, scale as  $S_r(I=I_0) \propto I_0^{2.6}$ . Data are for f=100 Hz.

that in the YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> superconductor, the weak-link behavior of the grain boundary is, at least in part, connected with oxygen deficiency. However, since we could not eliminate the weak-link behavior completely, we conclude that oxygen deficiency is not the sole cause for the weak-link behavior. We speculate that the cause of the weak link is due to the dislocations at the boundary. More specifically, we believe that it is the core of the dislocation that suppresses the order parameter. This region is normal in much the same way as the core of a vortex. The order parameter is therefore periodic in one dimension along the boundary. As the distance between the dislocations decreases, the maximum amplitude of the order parameter decreases until Josephson coupling across the boundary dominates [25]. Dislocation cores in the high-temperature superconductors introduce charge imbalance. If the lines contain kinks, as normally expected, a distribution of charge trapping sites is to be anticipated and hence 1/f noise. Oxygen anneal eliminates some of the charge imbalance but cannot be expected to eliminate all of it.

In summary, we have shown that 1/f noise from grain boundaries arises from critical current fluctuations when the bias current is near the critical current and from junction resistance fluctuations for large currents. The critical current fluctuations arise from resistance fluctuations. These findings suggest that recent models [14-17] for 1/f noise are not dominant in these junctions. Rather, conventional models of charge trapping and detrapping are valid descriptions.

Ozone annealing improves both the noise characteristics and critical current density. These experiments establish that oxygen deficiency at the boundary is present but is not the microscopic origin of the weak-link behavior.

- P. Chaudhari, J. Mannhart, D. Dimos, C. C. Tsuei, C. C. Chi, M. M. Oprysko, and M. Scheuermann, Phys. Rev. Lett. 60, 1653 (1988).
- [2] D. Dimos, P. Chaudhari, J. Mannhart, and F. K. LeGoues, Phys. Rev. Lett. 61, 219 (1988); J. Mannhart, P. Chaudhari, D. Dimos, C. C. Tsuei, and T. R. McGuire, Phys. Rev. Lett. 61, 2476 (1988).
- [3] D. Dimos, P. Chaudhari, and J. Mannhart, Phys. Rev. B 41, 4038 (1990).
- [4] B. H. Moeckly, S. E. Russek, D. K. Lathrop, R. A. Buhrmann, J. Li, and J. W. Mayer, Appl. Phys. Lett. 57, 1687 (1990); see also Appl. Phys. Lett. 57, 2951 (1990).

- [5] N. Tomita, Y. Takahashi, and Y. Ishida, Jpn. J. Appl. Phys. 29, L30 (1990).
- [6] S. E. Babcock, X. Y. Cai, D. L. Kaiser, and D. C. Larbalestier, Nature (London) 347, 167 (1990).
- [7] R. Gross, P. Chaudhari, M. Kawasaki, M. B. Ketchen, and A. Gupta, Appl. Phys. Lett. 57, 727 (1990); see also Physica (Amsterdam) 170C, 315 (1990).
- [8] M. Kawasaki, P. Chaudhari, T. Newman, and A. Gupta, Appl. Phys. Lett. 58, 2555 (1991).
- [9] K. Char, M. S. Colclough, S. M. Garrison, N. Newman, and G. Zaharchuk (unpublished).
- [10] B. Oh, R. H. Koch, W. J. Gallagher, R. P. Robertazzi, and W. Eidelloth (unpublished).
- [11] Z. G. Ivanov, P. A. Nilsson, D. Winkler, J. A. Alarco, G. Brorsson, T. Claeson, E. A. Stepantsov, and A. Ya. Tzalenchuk (unpublished).
- [12] P. Dutta and P. M. Horn, Rev. Mod. Phys. 53, 497 (1981).
- [13] C. T. Rodgers and R. A. Buhrmann, Phys. Rev. Lett. 53, 1272 (1984).
- [14] L. Wang, Y. Zhu, H. L. Zhao, and S. Feng, Phys. Rev. Lett. 64, 3094 (1990).
- [15] M. Johnson, M. J. Ferrari, F. C. Wellstood, and J. Clarke, Phys. Rev. Lett. 66, 1799 (1991).
- [16] L. Wang, Y. Zhu, H. L. Zhao, and S. Feng, Phys. Rev. Lett. 66, 1800 (1991).
- [17] H. J. Jensen, Phys. Rev. Lett. 64, 3103 (1990).
- [18] M. Kawasaki, S. Nagata, K. Takeuchi, and H. Koinuma, Jpn. J. Appl. Phys. 27, 2227 (1988).
- [19] R. H. Koch, John Clarke, W. M. Goubau, J. M. Martinis, C. M. Pegrum, and D. J. Van Harlingen, J. Low Temp. Phys. 51, 207 (1983).
- [20] After our paper was submitted we have learned that M. J. Ferrari, F. C. Wellstood, J. J. Kingston, and John Clarke, Phys. Rev. Lett. 67, 1346 (1991), have concluded, independent of our work, that their data do not support the universal conduction fluctuation models at all temperatures.
- [21] R. Gross, P. Chaudhari, D. Dimos, A. Gupta, and G. Koren, Phys. Rev. Lett. 64, 228 (1990).
- [22] V. Ambegoekar and B. Halperin, Phys. Rev. Lett. 22, 1364 (1969).
- [23] R. Gross, P. Chaudhari, M. Kawasaki, and A. Gupta, Phys. Rev. B 42, 10735 (1990).
- [24] S. E. Russek, D. K. Lathrop, R. A. Buhrmann, and D. H. Shin, Appl. Phys. Lett. 57, 1155 (1990).
- [25] P. Chaudhari, in Proceedings of the Toshiba International School of Superconductivity, Kyoto, Japan, 1991 (Springer-Verlag, Berlin, to be published); in Proceedings of the Third International Conference on Mechanisms and Materials—High Temperature Superconductivity, Kanazawa, Japan, 1991 [Jpn. J. Appl. Phys. (to be published)].