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1/f-noise-power measurements of copper oxide superconductors in the normal and superconducting states

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The first measurements of the 1/f-noise spectrum in copper oxide superconductors are presented. The key features of our results are (1) no noise is found in the superconducting state, (2) in the normal state the noise is large, comparable in magnitude to that in metal-insulator composites, and (3) the temperature dependencies of the noise and the resistivity are opposite, unlike either metals or semiconductors. Implications for possible conduction models are considered.

Much of the recent interest in copper oxide superconductors stems from the many practical applications conceived for them, from levitated trains to pixel-sized sensors. One sensor use, based on the large temperature derivative of resistance near T_c, would be as a radiation detector or bolometer. In the superconducting state, they could be used in superconducting quantum interference devices (SQUID's). Despite much activity investigating the properties of these materials, there have been few studies of fluctuation phenomena in them. One quantity of interest in these superconductors, for both theoretical and practical reasons, is the spectrum of excess electrical noise power, the noise above the frequency-independent "white" thermal noise.

In metallic conductors, the excess noise power is usually so small that it is measurable only in thin-film samples. However, we find significant noise-power spectra in bulk composite samples of the nominal 90-K superconductors $Y_1Ba_2Cu_3O_{7-\delta}$ and $Er_1Ba_2Cu_3O_{7-\delta}$ from T_c to room temperature. The frequency dependence of the excess noise power is in the range generally referred to as 1/f noise. However, both the very large magnitude of the noise and its temperature dependence are qualitatively different from that expected in a metal. We discuss several possible explanations for the unusual magnitude and temperature dependence of this noise.

Powders were made by both solid-state reaction and coprecipitation methods,[1] and pressed into bars of average length 2.5 cm and cross section 0.2 cm^2. The superconducting transition temperatures varied from 81 to 90 K, with widths of 2–4 K. The dc resistivities of our samples compare favorably with those reported elsewhere in both composites and single crystals.[2–4] Above T_c, the resistance is approximately linear in temperature. Typical 100-K (300-K) resistivities in Y-Ba-Cu-O were 0.74 (1.35) $m\Omega$ cm, and in Er-Ba-Cu-O 7.5 (12.8) $m\Omega$ cm. Scanning electron microscope (SEM) photographs indicate the samples are mixtures of small and large grains, with linear dimensions on the order of 3–4 and 10–20 μm, respectively. X-ray spectra show the grains have the single-phase 1-2-3 structure.

Electrical leads were attached by evaporating 2000 Å of nichrome and 2000 Å of gold onto masked areas of the samples; silver epoxy with copper wires embedded was then applied. To minimize contact resistance and noise, the current contacts completely covered the two end faces of each sample; the voltage contacts were placed on a third side, approximately 1.4 cm apart. Contact resistances were less than 0.1 Ω.

Electrical noise-power spectra were measured by standard 4-terminal techniques. A sample was clamped to a cylindrical copper block, on which a heater was symmetrically wound, by a strip of copper with a thermometer attached, electrically isolated from the copper by thin sheets of Mylar; this block was inside a vacuum can immersed in a liquid-nitrogen bath, and weakly thermally coupled to the bath by a nylon post. Noise was detected by passing battery-generated dc currents of 60–700 mA through the sample, with a large ballast resistance (at least 1000 times the sample resistance) in series to minimize contact noise. The voltage signal was dc filtered through a 1.1-F capacitance and passed through a low-noise transformer, preamplifier, and low-pass filter before detection by an HP 3582A spectrum analyzer, which performed rms averages of fast-Fourier transforms of up to 256 samples of the input signal. The sample cell and all noise circuit electronics, other than the spectrum analyzer, were located in a vibration isolated mumetal box; all electronics inside the box were powered by batteries.

Noise-power spectra were measured by cooling a sample to 77 K, connecting the current source, and slowly increasing heater power while monitoring the signal with the spectrum analyzer at various temperatures. Figure 1 shows the typical behavior of the total noise power near T_c. The noise in all runs exhibited a sharp peak at the same temperature as the peak in the measured bulk property $(dR/dT)^2$; the peaks were generally of comparable width. The absence of detectable contact noise in the normal state was verified by changing the ballast resistance while maintaining a constant voltage across the sample and observing the power spectrum to be unchanged.[5]

Two types of noise spectra were observed. In low-noise samples the spectrum became frequency independent at a frequency well within the frequency range measured (0.4–102 Hz). In high-noise samples frequency dependent noise was observed over the entire frequency range. SEM photographs showed that, while the size and number of large grains in all samples were comparable, the high-noise samples had a significantly larger number of small grains than did the low-noise samples, and the shapes of

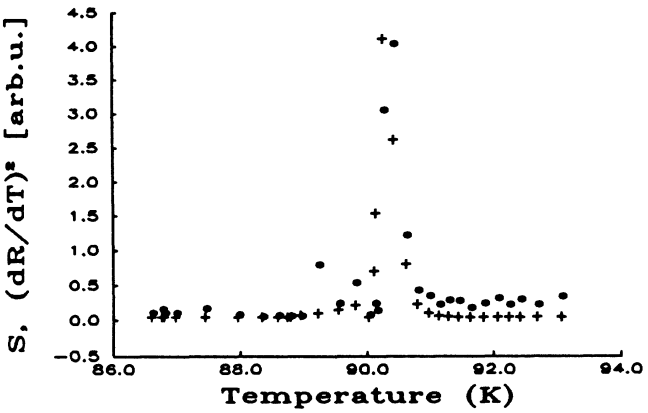

FIG. 1. Excess noise power at 6 Hz (●) and square of the resistance derivative (+) near T_c in $Y_1Ba_2Cu_3O_{7-\delta}$, plotted with arbitrary units.

the large grains in the high-noise samples were more irregular than in the low-noise samples. Longer sintering and annealing times resulted in samples with fewer small grains, and thus less noise, than those treated for shorter periods.

The measured noise spectrum was assumed to be the sum of thermal noise, amplifier noise, and excess noise from the sample. The thermal and amplifier noises are both independent of frequency in the range studied. The excess noise power $S_{exc}(f)$ was extracted from the measured total noise power spectra by two methods. For the low-noise data, the observed background (thermal + amplifier) noise was directly subtracted from the spectrum to give the excess noise. For the high-noise data, the amplifier noise was obtained by subtracting the thermal noise $4k_BTR$ from the measured noise of a copper wire (assumed free of excess noise); the excess noise in a sample was then found by subtracting the thermal and amplifier noises. Figure 2 shows a typical frequency spectrum of the excess noise of a high-noise sample in the normal state, along with that measured while superconducting.

In the superconducting state, within our sensitivity of 10^{-19} V², *no* $1/f$ noise could be measured, relative to a copper-wire resistor, although a few scattered nonreproducible peaks occurred at higher frequencies. Since in the normal state, the measured excess noise power scaled accurately as the square of the voltage V, we express the excess noise power in the form

$$S_{exc}(f) = C(T)V^2/f^{\alpha(T)}, \quad (1)$$

where $C(T)$ and $\alpha(T)$ are determined from straight line fits to $\log_{10}(S_{exc}) - \log_{10}(f)$ plots at each temperature (Fig. 2).

Figure 3 shows the temperature dependence of typical normalized excess noise spectra $S_{exc}(6\text{ Hz})/V^2$ for two samples. As indicated by Fig. 1, the sharp peak at T_c is not an artifact of dividing by a small voltage near the transition. These results are unusual in that the excess noise measured in metals normally *increases* with increasing temperature below room temperature, as does the resistivity; in semiconductors, both the noise and resistivity decrease with increasing temperature. For comparison, the noise of a 1.8-Ω carbon resistor was measured, yielding a value of $\log_{10}[S_{exc}\ (6\text{ Hz})/V^2] = -14.50$.

The parameters $C(T)$ and $\alpha(T)$ for $T > T_c$ were evaluated from spectra measured during higher precision runs. The exponent $\alpha(T)$ ranged from 0.78 to 1.21, in the range generally referred to as $1/f$ noise, and decreased with decreasing temperature, although considerable scatter was evident. A typical result for $1/C$ is shown in Fig. 4; $1/C$ varied approximately linearly with temperature far above T_c, while closer to T_c it decreased more rapidly toward zero near the transition.

The large noise power we observe is presumably due to intrinsic resistance fluctuations in our samples, which may arise from fluctuations in the number of carriers, as in a semiconductor, or from mobility or scattering fluctuations, as in metals. In the latter case, recent evidence indicates that resistance fluctuations probably arise from motion of defects, although the microscopic details remain unclear. One way of parametrizing $1/f$ noise, due to Hooge,[6] is $S(f) = \gamma V^2/Nf^{\alpha}$, where N is the number of scattering centers and α is a number close to 1. The quantity γ is found to be of order 10^{-1}–10^{-5} in metals. For

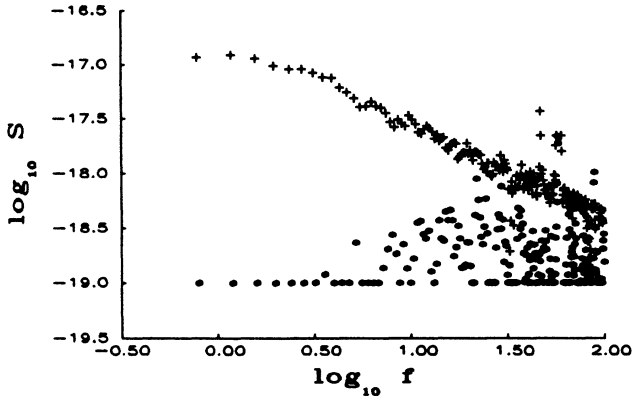

FIG. 2. Noise-power spectra of $Er_1Ba_2Cu_3O_{7-\delta}$ in the superconducting (●) and normal (+) states. These spectra were measured at 83.5 and 148.7 K, respectively. −19 is the maximum sensitivity of measurements.

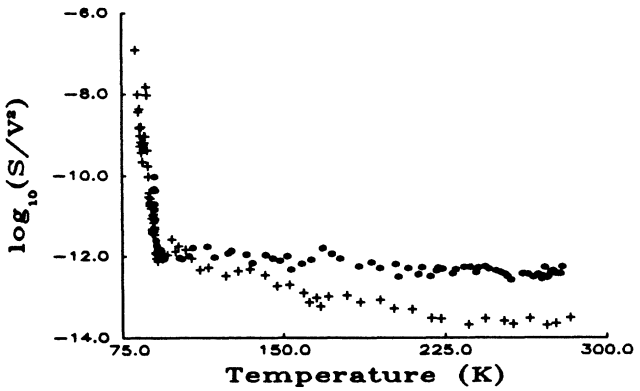

FIG. 3. Typical temperature dependence of the normalized noise spectra S/V^2 at 6 Hz in $Y_1Ba_2Cu_3O_{7-\delta}$ (●) and $Er_1Ba_2Cu_3O_{7-\delta}$ (+).

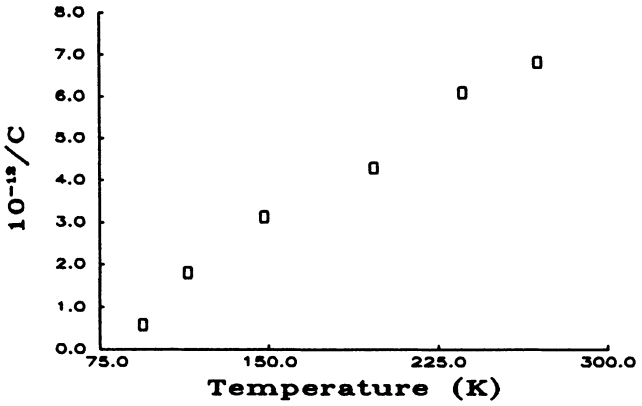

FIG. 4. Temperature dependence of the quantity $1/C$, defined in Eq. (1), for an $Er_1Ba_2Cu_3O_{7-\delta}$ sample.

ordinary metals N is comparable, within a few orders of magnitude, to the number of charge carriers N_c. An estimate of N_c from the free-electron gas model, using the mean free path of 22 Å measured[7] in Y-Ba-Cu-O and our measured sample resistivity, gives $N_c = 3.3\times10^{18}$. Using this picture, the large noise power in our samples implies a very large value of $\gamma \approx 10^5$–10^7 at 300 K. Thus in Y-Ba-Cu-O there is a discrepancy of about eight orders of magnitude in the value normally obtained for γ in metals.

Another way of characterizing the data involves a thermal fluctuation model[8] in which the noise power spectrum is given by $S(f) = V^2\beta^2 k_B T^2/fC_v A$, where $\beta = (1/R)dR/dT$ is the temperature coefficient of resistance, C_v the heat capacity, and A a geometric factor of order unity. The predicted noise power from this model is usually found to be comparable to observed values in metals both near room temperature as well as near T_c, where β is very large. As shown in Fig. 1, S and β peak sharply at the same temperature confirming the relevance of thermal fluctuations. However, using the measured value of the specific heat,[9] the predicted magnitude of the noise at 180 K is about 10^{11} smaller than our measured noise, very similar to the large underestimate found from the Hooge equation.

The fact that the measured noise is approximately ten orders of magnitude larger than would be estimated from conventional models suggests several distinct possibilities for the origin of the noise. One source of the noise could be the large anisotropy of the copper oxide material and hence would be an intrinsic noise process, present even in a single crystal. Another possible noise source could arise from the composite nature of the material.

The intrinsic noise process has as its origin activated hopping between Cu-O planes arising from the large resistivity anisotropy observed[10] in Y-Ba-Cu-O. In this picture, noise-free metallic conduction occurs within these planes, while semiconductorlike conduction takes place between planes by a thermally activated hopping process. The reduced number of carriers involved in the interlayer conduction process results in an enhanced contribution to the noise. The puzzling inverse temperature dependencies of the noise and the resistivity are due to the fact that the resistivity is dominated by the low-resistance (intralayer) links in a polycrystalline sample, while the noise arises mainly from the high-resistance (interlayer) conduction processes.

A second alternative source of the large noise is a composite effect which would be absent in a single crystal. In this picture, noise-free propagation occurs within grains, but conduction through boundaries involves noisy hopping processes. The boundaries could be extrinsic grain boundaries or internal twinning defects. It has been previously noted[11–14] that the excess noise in granular composites greatly exceeds the noise in homogeneous materials of similar resistances. Scaling inversely by the sample volume, Mantese *et al.*[11] measure a normalized noise power $S(10\text{ Hz})/V^2$ of 7×10^{-12} in a sample of our size at 300 K comparable to our measured values of 10^{-13}. They explain[15] their noise by tunneling processes which, in a two-component effective medium approach, give a much larger relative contribution to resistance fluctuations (noise) than to the resistance.

In our system, however, the grains appear to be separated by low-resistance tunneling barriers with linear I-V characteristics at low current; the barriers are of *SNS* character rather than *SIS*. Bulk superconductivity then occurs due to proximity coupling of the grains through *SNS* junctions. The observed resistivity is a complicated function of the separate metallic and boundary resistivities; thus measurements of the bulk resistivity may not accurately reflect metallic properties within the grains. This picture suggests that the noise is produced in a volume much smaller than the bulk of the sample. It has been observed[16] that the structure and composition of the grain boundaries differ from the bulk of the grains. The increase in noise with decreasing temperature in these materials could be due to exponentially activated semiconductorlike tunneling barrier resistances at these inhomogeneous boundaries.

Several observations can be made in regard to these two pictures. The measured ratio of the *ab* plane and *c* axis conductivities is observed to be less than 100 at the transition; it is difficult to see how an anisotropy of this size would account for an enhancement of 10^8 relative to the magnitude of noise in ordinary metals. Secondly, the noise decrease in larger grain size samples is inconsistent with an intrinsic noise mechanism but consistent with a granular picture. If the noise arises from internal twinning boundaries, the noise would vary with the density of defects, not necessarily with the grain size, although the latter two could be correlated.

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