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Low-frequency noise in the normal state of thin-film high-temperature superconductors

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The excess low-frequency (1/*f*) noise of $YBa_2Cu_3O_{7-\delta}$ thin-film microbridges has been studied in the normal state. The normalized noise-power spectral density, S_V/V^2, shows quantitatively different behavior depending on the film-substrate lattice mismatch and the oxygen content. The noise behavior is largely attributable to the hopping of basal-plane oxygen vacancies subject to strain-dependent activation barriers. Both electromigration biases and oxygen plasma annealing can be effective techniques for lowering these noise levels, but in different ways that point to the origin of the noise. The low-temperature, ~100 K, noise behavior, indicates that even films with high critical current densities have a substantial density of oxygen-defect states with very low activation energies for hopping.

Many undesirable properties of $YBa_2Cu_3O_{7-\delta}$ (YBCO) and other cuprate superconductors are related to their high level of structural and chemical defects, of which the oxygen vacancy is particularly pervasive and unstable.[1,2] A significant aspect of these unstable defects is the wide distribution of activation energies for their fluctuation between metastable positions and for their displacement. Experimental studies of anelastic creep and elastic aftereffect have revealed a broad relaxation-time spectrum for bulk YBCO samples in response to an applied stress.[3] The existence of such broadly distributed relaxation rates was also inferred from the "stretched exponential" time dependence of the microbridge resistance upon the application of an electromigration force.[2] Via suitable coupling mechanisms, the same processes can also be expected to manifest themselves in low-frequency resistance fluctuations, i.e., 1/*f* noise, since such noise has been shown to typically arise from ensembles of fluctuators having a broad distribution of activation energies.[4,5] Indeed, due to the sensitivity of 1/*f* noise in a given bandwidth to the fluctuations of defects with a narrow range of activation energies, temperature-dependent noise measurements can be quite effective for examining the energy distribution of the defect fluctuations,[6] particularly in thin-film systems, and thus for gaining insight into the microscopic origin of this distribution and of the defects themselves.

Several groups[7,8] have reported 1/*f* noise measurements in various cuprate high-T_c superconductors. In the normal state the magnitude of the noise power was found to be much higher than that of a typical metal, by as much as five orders of magnitude. A model in which conductance fluctuations originate from localized carrier traps in the insulating boundaries of substructures was proposed[7] to account for the observed high noise levels in YBCO single crystals. More recently Liu *et al.*[8] measured 1/*f* noise in oxygen-deficient $YBa_2Cu_3O_x$ films and found a nonmonotonic behavior in the normalized noise spectral density as the oxygen content was being varied, with a minimum at $x=6.5$. This behavior was attributed to the more stable character of the composition which allows, in the ideal case, the formation of the ortho II phase in which oxygens in the basal plane (of a conventional $YBa_2Cu_3O_x$ unit cell) are ordered to form alternating rows of vacant and occupied Cu-O chains along the *b* axis.

Here we report 1/*f* noise measurement on uniform, relatively high quality thin-film microbridges composed of $YBa_2Cu_3O_{7-d}$ deposited on MgO and $SrTiO_3$ substrates by pulsed laser ablation. The critical currents for these microbridges were all in excess of 10^6 A/cm^2 at 77 K. X-ray diffraction study of the unpatterned film showed that the samples were primarily *c*-axis oriented with the in-plane *ab* axes aligned with the substrate orientation. The bridges with widths of 1, 2, and 5 μm and a typical length/width ratio of 2.5 to 1 were patterned using standard photolithography. Their noise power spectrum was measured using a variable dc bias and standard low-frequency spectrum analyzer equipment.

For the uniform, grain-aligned microbridges of $YBa_2Cu_3O_{7-d}$ we find that the normal-state 1/*f* noise power spectral density scales with the square of the bias current, indicating that the noise is dominated by conductance fluctuations. The 1/*f* power spectral density also scales with volume, though with much more scatter, which is attributable to the experimental uncertainty in the volume. To characterize and compare the noise behavior we use Hooge's empirical relation $S_V(f)=\gamma V_{dc}^2/N_c f$, which is appropriate for independent resistive fluctuation sources. Here V_{dc} is the dc bias voltage, N_c the total number of carriers, f the frequency at which the spectral density is measured (typically 10 Hz), and γ is a numerical factor which is characteristic of the system being measured.

The upper panel of Fig. 1 shows the noise spectral density after normalization for two microbridges of YBCO/$SrTiO_3$ and YBCO/MgO, denoted as sample A and sample B, respectively. The volumes of both samples are 3×10^{-18} m^3. Taking the density of carriers to be 5.75×10^{27}/m^3 for the YBCO film[8] yields γ values of 5.75×10^{-3} and 2.3×10^{-2} for YBCO/$SrTiO_3$ and YBCO/MgO, respectively, at room temperature. This is about a factor of 1000 times smaller than the smallest g value of this material reported so far,[8] and is close to the range that is typically seen in polycrystalline metal films. This suggests that we are probing fluctuations that are not dominated by the sources which are extrin-

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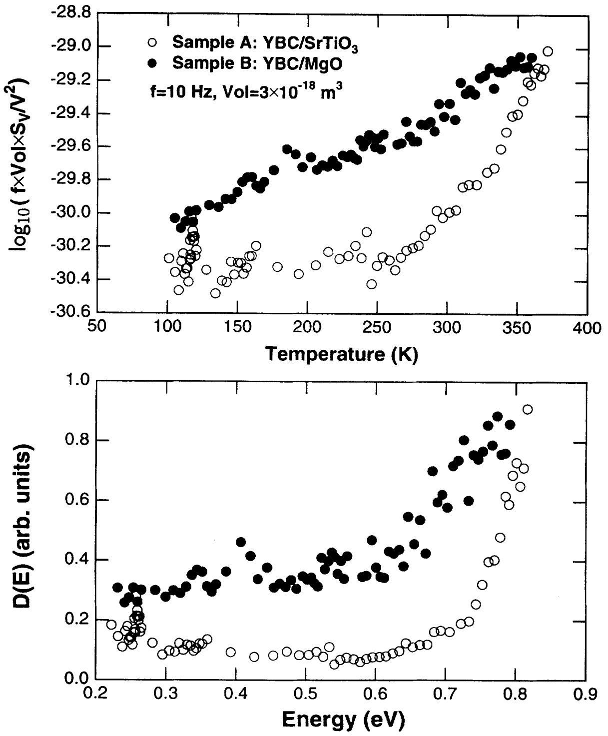

FIG. 1. Upper panel: normalized noise power density vs temperature for two YBCO microbridges. The volume of the microbridge is 3×10^{-18} m^3 for both samples. Lower panel: energy distribution of fluctuating states for these samples as calculated using the Dutta-Horn model.

sic to high quality grain aligned thin films. Focusing on the temperature dependence, we note that the two samples show markedly different behavior. For YBCO/SrTiO$_3$ S_V/V^2 is temperature independent to within about a factor of 3 (5 dB) from 100 to 250 K. This was followed by a 20-fold increase in noise between 250 and 375 K. Conversely, for YBCO/MgO the S_V/V^2 increases continuously between 95 and 375 K, beginning and ending at about the same magnitudes as for YBCO/SrTiO$_3$. In some cases the variation is almost linear, starting right above T_c as seen from the data of sample B in the upper panel, Fig. 1.

The Dutta-Horn[6] model can be applied to analyze $1/f$ noise sources in the situations where the fluctuations result from thermally activated processes with a broad distribution of activation energies with respect to k_BT. The variation of noise power density with temperature may then be used to calculate the energy distribution of the density of the fluctuating states $D(E)$. The lower panel of Fig. 1 displays the result of such a calculation for these two samples. The energy abscissa is calculated using $E=-k_BT\ln(\omega t_0)$, where $\omega=2\pi f$, f being the frequency at which the spectrum is taken, and τ_0 is the inverse of the characteristic attempt frequency. We assume τ_0 to be the same for all activated fluctuators, and from internal friction measurements we take it to be 1.2×10^{-13} s.[3] We note from the lower panel of Fig. 1 that this analysis results in a pronounced increase in the density of states for energies greater than 0.8 eV, particularly for YBCO/SrTiO$_3$ samples. Although we do not have data for energies much greater than that, being limited by an upper temperature beyond which oxygen loss or electromigration-assisted oxygen vacancy reordering may take place, the available data are indicative of a peak in the density at about 1 eV. The one electron-volt energy is particularly appealing, since the mass transport technique[9] measured an activation energy of 0.93 eV for oxygen diffusion. Internal friction and anelastic creep and elastic aftereffect experiments,[3,10] which measure the mechanical energy loss in solids due to atomic relaxations, also gave rather consistently an activation energy ~1 eV for the thermally activated process involved. It is well accepted that the O long-range diffusion and the anelastic relaxation effects are the result of basal plane oxygen hopping, in particular, the O(1)–O(5) jumps (our designation of oxygen sites follows that of Ref. 15).

In order for the structure in the noise data to be attributable to the same origin as the peaks in the relaxation experiments, some coupling mechanism is required. This may be understood simply in the framework of the charge transfer model,[11] in which not only the concentration but also the ordering of oxygen vacancies in the basal plane controls the doping in the CuO$_2$ sheets, and therefore directly affects the local carrier densities. In this picture the $1/f$ noise is a direct consequence of fluctuations in local carrier densities, which in turn is caused by oxygen vacancy hopping and reordering in the basal plane of YBCO structures.

This analysis points to the explanation for the broadening of the distribution of the fluctuating states in the case of YBCO/MgO. There is substantially a larger lattice mismatch between the c-axis YBCO and the MgO substrate than that between YBCO and the SrTiO$_3$ substrate, $>9\%$ vs $\sim1\%$. Consequently the inhomogeneous strain field which arises from the extended defects that accommodate the lattice mismatch is more severe in films grown on MgO than those grown on SrTiO$_3$. This is consistent with the difference in local atomic order as indicated by the ion-channeling signal for YBCO films on the two different substrates even when similar levels of critical current densities are obtained.[12] The relatively larger strain field in the YBCO/MgO system then results in a more nonuniform distribution of the oxygen vacancies than in the YBCO/SrTiO$_3$, hence the much broader distribution of activation barriers for oxygen vacancy fluctuations.

As a further confirmation of the link between stress and oxygen vacancy hopping we performed an electromigration experiment on a YBCO/MgO sample. Electromigration at low current levels has been found[2] to substantially improve critical current densities, apparently due to the much improved ordering of oxygen vacancies in the microbridge.[13] The sample was biased with progressively higher current densities for 10 h intervals. It was then cooled immediately following the removal of the electromigration current and the $1/f$ noise measured. As is demonstrated in Fig. 2 the noise of a YBCO/MgO sample could be modified in this way to the seemingly less stressed state of the YBCO/SrTiO$_3$ samples. However, this bias-induced reduction in $1/f$ noise levels is not permanent; when left at room temperature, the samples would resume their original noise behavior within a few hours. We conclude that while the electromigration force can displace basal oxygen atoms to more stable, more ordered positions, when the force is removed inhomogeneous

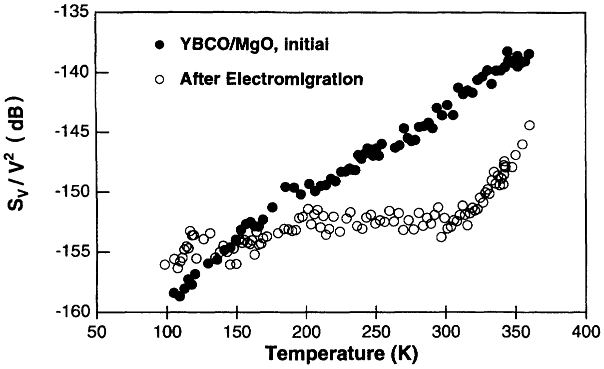

FIG. 2. Noise power density vs temperature for a YBCO/MgO sample before and immediately after electromigration. Note that the noise behavior afterwards resembles that of YBCO/SrTiO samples.

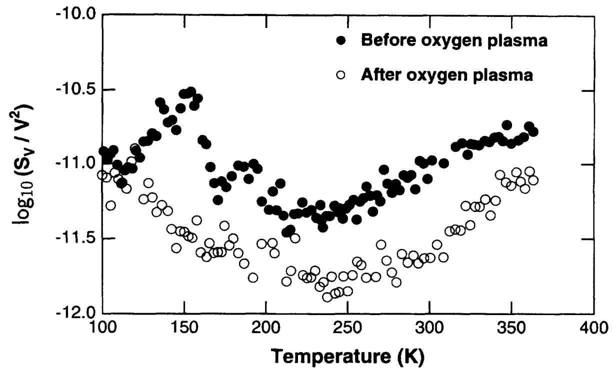

FIG. 4. The nonmonotonic noise vs temperature behavior of a YBCO/MgO microbridge containing a high-angle grain boundary. The oxygen plasma treatment removed the low-T peak at about 150 K.

stresses arising from structural defects in the film act to slowly return these atoms back to unstable positions.

A different type of change in the YBCO noise behavior can be induced by subjecting the film to a room-temperature oxygen plasma treatment. Such treatment has been shown to increase the critical current density and T_c of YBCO thin films and weak links,[14] presumably by increasing the basal oxygen concentration. In Fig. 3 we show the noise behavior of a YBCO/MgO sample before and after undergoing a plasma treatment which had the effect of reducing the normal-state resistance by ~10% and increasing the critical current at 77 K by ~20%. Here the effect on the noise is to reduce the level at all temperatures, an effect that persists for at least several weeks after the treatment. We conclude that the oxygen plasma increases the oxygen stoichiometry, perhaps restoring oxygen that was lost during the processing of the film. As a result, the number of fluctuating oxygen vacancies, and hence the resultant noise is reduced, but the energy distribution of the fluctuating elements is not substantially altered by this plasma treatment.

An important observation to be made concerning the lower panel of Fig. 1 is the leveling off of the density of fluctuating states at a substantial, nonzero level at low temperatures for both types of samples. In fact the noise spectral density S_V/V^2 itself, although it generally decreases slowly with decreasing T, is still quite high at $T \sim 100$ K. This low-temperature YBCO $1/f$ noise may have serious technological implications as it could be responsible for phase noise in thin-film resonators, with the phase noise arising from local fluctuations in supercurrent density, and for $1/f$ noise in the critical current and magnetic pinning of SQUID devices. The existence of this noise in normal YBCO films is indicative of the presence in both types of samples of atomic fluctuators with very low activation energies. Moreover, the low-temperature behavior appears to be associated with some fluctuations of the basal oxygen, perhaps arising from regions of high oxygen deficiency.

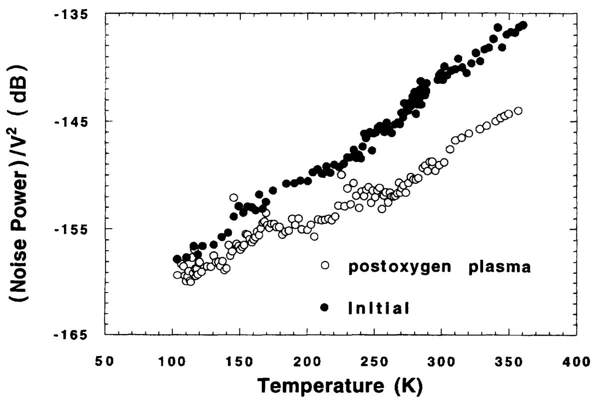

FIG. 3. Noise power density vs temperature for a YBCO/MgO sample before and after oxygen plasma treatment.

This conclusion is supported the normal-state noise behavior of microbridges which are bisected by high-angle grain boundaries. Such microbridges generally exhibit a Josephson weak-link behavior in superconducting state, a behavior that is rather definitely associated with oxygen deficiency and disorder in the vicinity of the grain boundary.

Figure 4 shows the noise spectral density of such a microbridge before and after an oxygen plasma treatment. Before the treatment the $1/f$ noise behavior of this sample is nonmonotonic, with a noise peak at ~150 K, in contrast to the uniform samples discussed previously. It has been suggested[10] that the increased oxygen deficiency results in a reduced activation barrier for oxygen self-diffusion. While this suggestion was not corroborated by the oxygen tracer diffusion measurement,[9,10] low-energy hopping modes have been observed in internal friction measurements of oxygen-depleted YBCO.[15] The reduced activation barrier of such hopping modes, when combined with a strain-induced broadening of the energy distribution for these modes, would lead directly to enhanced low-temperature $1/f$ noise levels.

We note, however, that if the low-temperature noise arises from basal oxygen hopping in regions of high oxygen deficiency, there is likely to be a different mechanism that couples the atomic fluctuations to the resistance. In oxygen-depleted, nonmetallic YBCO regions, transport is expected to proceed by resonant tunneling or hopping via localized states.[16] This type of conduction is controlled by a few localized states with optimal configurations,[17,18] and will exhibit particularly strong fluctuating characteristics if the dopant oxygen atoms are unstable.

Whatever the coupling mechanism the fundamental role of the basal oxygen in the low T, as well as high T, noise behavior is demonstrated by the effects of oxygen plasma treatments or electromigration biases on the low-temperature grain boundary noise. For example, in Fig. 4 the oxygen plasma treatment is seen to have removed the noise peak at ~ 150 K in this sample, as well as reduced the overall low-temperature noise level by a factor of 2. This $1/f$ noise reduction was in conjunction with a decrease of weak link resistance R_n from 7 to 4.5 Ω, and an increase of weak link critical I_c from 85 to 136 μA (measured at 4.2 K). Similar noise reductions, albeit temporary ones, have been obtained by electromigration biases.

In conclusion, we have examined the normal-state $1/f$ noise behavior in the grain-aligned YBCO microbridges and in YBCO microbridges containing high-angle grain boundaries. Much lower noise levels have been found than previously reported. The temperature dependence and effect of oxygen plasma treatment demonstrate that basal plane oxygen instabilities are responsible for this noise in both types of samples. The substrate dependence and the effect of electromigration biases indicates that the distribution of the activation energies for this process is strongly influenced by the stress-induced local strain field in the thin films. All samples showed a similar low-temperature background noise level, possibly due to some inherent structural defects which substantially reduce the activation energy of oxygen displacement below the nominal 1 eV value. Some of the anomalous low-temperature noise behavior of grain-boundary weak links may be attributable to particularly low-energy modes of basal plane oxygen hopping, which possibly arise in regions of low oxygen content. Both electromigration and oxygen plasma annealing were found to reduce $1/f$ noise levels. The beneficial effect of the former was not stable, but the latter procedure results in noise reductions that appear to be stable for at least a period of more than one month.

The relative instability of the basal oxygen, and the apparent sensitivity of the hopping rate to local stress, results in substantial low-T noise effects that can impact high-T_c Josephson devices and microwave resonators. While this study has focused on YBCO films, similar studies of $1/f$ noise in TBCCO films on $SrTiO_3$ have revealed very similar results, including the low-T noise plateau. Thus this noise behavior is a problem that may well be generic to many cuprate superconductor systems, given the high mobility of the dopant atoms in these systems.

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