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Microwave surface resistance and vortices in high-T_c superconductors: Observation of flux pinning and flux creep

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History dependence and logarithmic time dependence of the microwave surface resistance have been observed in polycrystalline $YBa_2Cu_3O_{7-x}$ in a magnetic field. We interpret our data in terms of granularity and use the model of Gittleman and Rosenblum to relate the surface resistance to the number of vortices in the sample. At a given field, we find that the surface resistance of a zero-field-cooled sample is higher than that of a field-cooled sample, suggesting that the former contains more intergranular vortices. It is also observed that once the external dc field is removed, flux creep out of the sample results in a logarithmic time decay of the surface resistance, from which we estimate a thermal activation energy at 77 K of about 0.1 eV.

We report history dependence and logarithmic time dependence of the high-frequency electromagnetic absorption of polycrystalline $YBa_2Cu_3O_{7-x}$ high-temperature superconductors in a magnetic field. Extending models of fluxon response,[1,2] we relate the measured surface resistance to the number of vortices in the sample and to the fraction of their length passing through high loss, intergranular regions. Our data show that this fraction is dependent on the magnetic history of the sample, and a logarithmic time dependence of the magnetic-field-induced high-frequency loss after the field is removed is attributed to thermally activated creep of fluxons pinned in individual grains.

The motion of fluxons in type-II superconductors is governed generally by their viscous drag and the restoring force from pinning sites. Here, we discuss the case where the external dc magnetic field is perpendicular to the ac current induced in the sample. Gittleman and Rosenblum[1] have shown that at sufficiently high frequencies, viscous drag is dominant and limits the motion of a vortex to oscillations about its equilibrium pinning position small enough that the restoring force is negligible. The vortex thus dissipates energy as if it were unpinned. In this high-frequency regime, where our experiment is performed, the sample dissipates energy proportional to the number of vortices it contains, and inversely proportional to the coefficient of viscous drag.[1] Specifically, the flux-flow resistivity is given by

$$\rho_H = \frac{n\phi_0^2}{c^2\eta} = \frac{\phi_0 B}{c^2\eta}, \tag{1}$$

where $n = B/\phi_0$ is the vortex number density, $\phi_0 = hc/2e$ is the flux quantum, and η is the coefficient of the viscous drag.

On the other hand, the complex conductivity $\sigma_s(\omega)$ for the superconductor in zero external field can be written as[3] $\sigma_s(\omega) = \sigma_1(\omega) - i\sigma_2(\omega)$. For $\hbar\omega \ll 2\Delta$, $\sigma_1(\omega)$ falls to zero exponentially at low temperature, and $\sigma_2(\omega) = c^2/4\pi\omega\lambda^2$, where λ is the penetration depth, describes the accelerative supercurrent. At temperatures not close to T_c and $\hbar\omega \ll 2\Delta$, $\sigma_1(\omega) \ll \sigma_2(\omega)$; hence the resistivity can be simplified to

$$\rho_s(\omega) = \frac{1}{\sigma_s(\omega)} \approx \frac{\sigma_1}{\sigma_2^2} + i\frac{1}{\sigma_2}. \tag{2}$$

Since ρ_H adds to the total dissipation, it should be in series with $\rho_s(\omega)$. For temperatures not close to T_c, $\hbar\omega \ll 2\Delta$, and nonzero fields, ρ_H is much larger than σ_1/σ_2^2. Thus the complex resistivity for a bulk sample in dc magnetic fields in approximately given by

$$\rho = \frac{\phi_0 B_{\rm eff}}{\eta c^2} + i\frac{4\pi\omega\lambda^2}{c^2}, \tag{3}$$

where $B_{\rm eff}$ is the effective flux density responding to microwave fields. Following Portis *et al.*,[2] one can then obtain the surface resistance for a bulk sample,

$$R_s = X_0\{[-1 + (1 + 4B_{\rm eff}^2/B_0^2)^{1/2}]/2\}^{1/2}, \tag{4}$$

where $X_0 = 4\pi\omega\lambda/c^2$ is the surface reactance at $B_{\rm eff} = 0$, and $B_0 = 8\pi\omega\eta\lambda^2/\phi_0$ is the characteristic value of $B_{\rm eff}$ above which the dissipation process dominates the impedance.

Portis *et al.* interpreted the effective density $B_{\rm eff} = fB$ (with f estimated to be of the order of 0.1 for small fields) as the density of free or weakly pinned fluxons. However, the argument of Gittleman and Rosenblum[1] implies that all vortices dissipate energy as if they were unpinned at sufficiently high frequency. An alternative interpretation of $B_{\rm eff}$ is the following: since the viscosity for fluxons between the grains is orders of magnitude smaller than that for fluxons in the grain[4] (because of the high intergranular resistance), one can neglect the loss due to dissipation *in* the grains; hence, while taking the viscosity parameter η as the one for the intergranular vortices, we interpret the fraction f as the fraction of the total length of all vortices in the sample which fall in intergranular regions. As shown in Fig. 1, we define two types of vortices according to their pinning characteristics: (i) *grain-pinned vortices* with density n_g are those which are pinned by pinning centers within the grains; (ii) *grain-boundary vortices* with density n_j are those which never pass through grains.

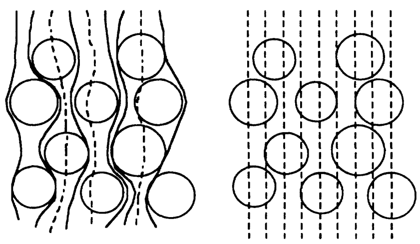

FIG. 1. Figurative representation of flux lines for ZFC and FC samples in a granular sample where grains are shown as circles. Two types of vortices may exist: grain-boundary vortices (solid lines) and grain-pinned vortices (dashed lines). (Only a representative sampling of vortices is shown.)

If the sample size is larger than the grain size, a *grain-pinned vortex* will nevertheless have a fraction x (on average) lying in intergranular regions. Hence the effective flux density is

$$B_{\text{eff}}=(n_j+xn_g)\phi_0\,. \tag{5}$$

As a zero-order approximation, we expect x to be a constant determined by the morphology of the sample, i.e., its total intergranular volume versus the total sample volume. Experimentally, we find n_j and n_g to be field, history, and time dependent.

The samples used in our experiments are bulk and powder samples of polycrystalline $YBa_2Cu_3O_{7-x}$ obtained commercially and through solid-state reaction synthesis in our laboratory. The sample is affixed to the removable end plate of a cylindrical Cu cavity resonating at 15 GHz in the TE_{011} mode, and the surface impedance of the sample is measured via a perturbation method.[5] A copper solenoid applies a magnetic field of up to 300 G along the axis of the cavity, normal to the surface of the disk-shaped sample of 8 mm diameter and 2 mm thickness. The microwave currents induced in the sample flow in a circular path with a magnitude dependent on the radial position.[5] The cavity is coupled, via a circular iris located off axis in the fixed end plate, to a waveguide which extends out of the cryostat, where, through a waveguide-coaxial adapter and a coaxial cable, it connects to an HP 8510B network analyzer. Using a calibration technique, network impedances up to the position of the adapter are removed from the measurement, making our data insensitive to that part of the microwave circuit. The network analyzer is then used to measure the power reflected from the cavity at resonance, from which we compute the sample surface resistance as follows:

$$1/\beta=(1\pm\sqrt{p})/(1\mp\sqrt{p})=\gamma_s R_s+\gamma_c R_c\,. \tag{6}$$

Here β is the ratio of the cavity Q to the coupling Q, $p=P_r/P_i$ is the ratio of the power reflected from the cavity at resonance to the incident power, R_s and R_c are surface resistances for sample and cavity, respectively, and the γ's are geometrical factors. The upper and lower signs are for the undercoupled ($\beta<1$) and overcoupled ($\beta>1$) situations, respectively. In the temperature range of our measurement, the surface resistance of the cavity is almost constant, so that $\Delta(1/\beta)\propto\Delta R_s$. Therefore, by measuring the change of $1/\beta$, we measure the change of the surface resistance of the sample, apart from a constant scale factor.

The magnetic history dependence of the surface resistance as a function of temperature is shown in Fig. 2. It is well known that, for any type-II superconductor above H_{c1}, cooling in zero field and then applying a magnetic field (so-called zero-field-cooled or ZFC sample) is very different from cooling in the magnetic field (field-cooled or FC sample). With the FC process, fluxons can be trapped in the sample, causing the sample to be less diamagnetic. Therefore the induced flux density B measured in the magnetization measurement is always lower for the ZFC sample than the FC sample, i.e., a ZFC sample contains fewer vortices than the FC sample. Since the microwave dissipation is proportional to the number of vortices in the sample, we might expect it to have a similar history dependence. Our experimental results show the opposite behavior. As shown in Fig. 2, the surface resistance for the ZFC sample is higher than that of the FC sample. We propose that, while the magnetization measurement measures the total number of vortices of the sample, all with equal weight, the microwave surface resistance measurement measures the effective intergranular flux density B_{eff} as discussed earlier. Our data simply indicate that the ZFC sample contains more vortices between grains than the FC sample, i.e., B_{eff} is higher for the ZFC sample even if B is lower. For the ZFC sample, grains are more diamagnetic, resulting in a stronger demagnetization effect between grains than in the FC sample. This leads to higher flux concentration at the grain boundaries, and hence more *grain-boundary* vortices n_j. Generally speaking, as shown in Fig. 1, the distribution of vortices in the FC sample is more uniform, while in the ZFC sample, more vortices are concentrated in the intergranular regions. As expected, the difference between ZFC and FC data diminishes at higher field.

We have also measured the time dependence of the surface resistance when the external field is changed. After an initial instantaneous drop when the external dc field is suddenly removed, the surface resistance decreases logarithmically in time.[6] We attribute this logarithmic de-

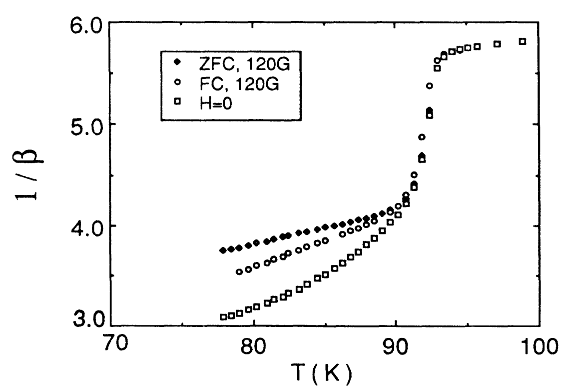

FIG. 2. Surface resistance changes (arbitrary units) as a function of temperature for zero field, and for ZFC and FC samples with the external field of 120 G.

crease to flux creep. A simple theory of flux creep yields a logarithmic time dependence for the magnetization,[7]

$$M(t)=M_0\left[1-\frac{kT}{U}\ln(t/\tau)\right], \qquad (7)$$

where M_0 is measured right after the initial drop when the external field is removed, U is the thermal activation energy, and $\tau(\ll 1\ \mathrm{s})$ is a characteristic time. Assuming the flux-flow dissipation process dominates the surface impedance, i.e., $B_{\mathrm{eff}}\gg B_0$ in Eq. (4), then the surface resistance is proportional to $B_{\mathrm{eff}}^{1/2}$, and also assuming B_{eff} is proportional to B (to be justified later)

$$R_s(t)=R_s(t=0^+)\left[1-\frac{kT}{U}\ln(t/\tau)\right]^{1/2}$$

$$\approx R_s(t=0^+)\left[1-\frac{kT}{2U}\ln(t/\tau)\right]. \qquad (8)$$

Here $R_s(t=0^+)$ is the value measured immediately after the field is removed. Figure 3 shows data from a ZFC sample which was cooled to 78 K in zero magnetic field, after which we applied a field of 120 G and then removed the field at time $t=0$. The logarithmic fitting gives

$$dR_s/d(\ln t)\propto d(1/\beta)/d(\ln t)=0.0046 .$$

Using the first data point taken at 6 s after the field is turned off

$$R_s(t=0^+)\propto[1/\beta(t=0^+)-1/\beta(H=0)]=0.14 ,$$

and we obtain the thermal activation energy $U=0.1$ eV.

Is this the creep of grain-pinned vortices or grain-boundary vortices? A comparison of our ZFC and FC samples reveals the answer for this question. While the values of activation energy obtained from ZFC and FC samples are about the same, the initial drop of the surface resistance is very different. The following is a set of data taken on a sample at 77 K in a field of 65 G, prepared through ZFC and FC processes: (a) ZFC sample, $R_s(H=65\ \mathrm{G})$ is 0.43, $R_s(t=0^+)$ is 0.09; (b) FC sample, $R_s(H=65\ \mathrm{G})$ is 0.31, $R_s(t=0^+)$ is 0.22; here all R_s values are measured in arbitrary units with the same scaling constant and R_s for zero field is taken to be zero. Note that the initial drop of R_s for the ZFC sample after the field is removed accounts for about 80% of the total surface resistance when the field is on; while for the FC sample, this drop is only 30%. Considering the possibilities allowed by the critical state model,[8] and referring to Fig. 1, we propose the following explanation of the data. In the initial drop at $t=0^+$, all *grain-boundary vortices* have left the sample, as have some of the *grain-pinned vortices,* in order to establish a new critical state for individual grains. The subsequent flux creep is solely due to the *grain-pinned vortices* creeping out of the individual grains. Because $n_j=0$ after the initial drop, $B_{\mathrm{eff}}=xn_g\phi_0$ is proportional to the residual flux density which justifies the assumption made in the derivation of Eq. (8).

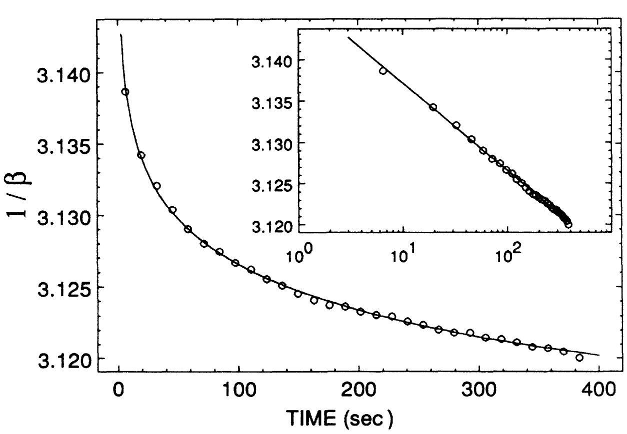

FIG. 3. Circles: Measured surface resistance change (arbitrary units) at 77 K vs time after the external field of 120 G is removed at $t=0$. Line: Logarithmic fitting with $d(1/\beta)/d(\ln t)=0.0046$. Inset: same data plotted on logarithmic time scale.

One difficulty in determining the intrinsic activation energy might arise from the small fields used in our experiment. If the initially applied field to a ZFC sample is smaller than the penetration field H^* for grains, once that field is removed, due to the shape of the flux profile in the critical state, only half of the vortices are creeping out of the sample, while the other half are creeping into the interior of the grains causing no reduction of the microwave loss. In this case, the activation energy U should be 0.05 eV instead of 0.1 eV. Either value is consistent with the range of values found in magnetic relaxation and resistive transition studies.

Flux creep can also be studied by changing the external field in other ways, for example, applying a magnetic field to a ZFC sample and leaving it on. In the magnetization measurement, the flux density B has an instantaneous increase followed by logarithmic increase as a function of time. However, when we apply a field (usually about 100 G) to a ZFC sample, we observe that after the instantaneous increase (establishing the critical state), the surface resistance increases slowly with time only for the first 10 s or so; then it *decreases* slowly with time. To explain this phenomenon, we think two processes are involved: (i) the vortices creep into the intergranular region from outside the sample, and (ii) the grain boundary vortices creep into the grains. The first process increases the surface resistance, while the second process decreases the surface resistance because vortices in the grains are less dissipative than those in the intergranular region. Our result seems to suggest that the second process is dominant for the surface resistance for the long-time scale. This is also consistent with the history-dependence data as shown in Fig. 2, since the FC sample, corresponding to the equilibrium state toward which the creep is progressing, possesses a lower value of surface resistance. Overall, the method described in the earlier paragraphs, i.e., measuring the time dependence after the field is removed, seems to be a simpler way to determine an activation energy from the microwave measurement.

In conclusion, we have observed a logarithmic decay of surface resistance which is related to flux creep. The observed history dependence of microwave losses can be understood in the framework of a model which takes account of granularity and the large difference between the viscosities for flux lines within and between the grains. These time-dependent data and the magnetic hysteresis of surface resistance can all be qualitatively described by our model. Microwave measurements appear to provide a unique tool to distinguish vortices in different environments and can be used to test theoretical models for granular superconductors.

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