

Field-modulated microwave surface resistance in a single-crystal $\text{Tl}_2\text{Ca}_2\text{BaCu}_2\text{O}_8$ superconductor

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(Received 19 August 1991; revised manuscript received 18 November 1991)

We report magnetic-field-modulated microwave surface resistance in a single-crystal $\text{Tl}_2\text{Ca}_2\text{BaCu}_2\text{O}_8$ superconductor. At a fixed temperature, as the applied magnetic field is varied, the observed signal has two features: an intense sharp peak at low field and a broad maximum at high field. The peak field defined for the broad maximum does not have the same temperature dependence as the irreversibility line determined from dc magnetization measurement. The results are analyzed on the basis of recent theoretical models. It is found that the flux-flow resistivity $\rho_f \propto B$ for $B < B_{\text{peak}}$ and $\rho_f = \text{const}$ for $B > B_{\text{peak}}$. Scaling of the data at different T and B gives a universal function for the surface resistance.

Since the discovery of the high-temperature oxide superconductors, extensive theoretical and experimental effort has been directed toward understanding the onset of resistance when magnetic fields are applied below T_c . Standard dc and low-frequency ac measurements point to the existence of an irreversibility line along which resistivity returns.¹ It is unclear whether this line represents the activation of the flux-line lattice over pinning barriers (thermally assisted flux flow) or a genuine melting transition. Studies of the frequency dependence of the irreversibility line may help distinguish between these possibilities. We report here the results of modulated microwave surface impedance studies, analyzed in the context of a recent calculation by Coffey and Clem.² That model properly treats the change in effective penetration depth induced by Lorentz-force modulation of the flux lattice via microwave surface currents. Microwave studies eliminate the need for electrical contacts and permit measurements to be carried out at much lower current densities than required for conventional measurements. Possible non-Ohmic response, local heating, and other contact-related problems are thereby eliminated. Further, microwave applications are already under development, making an understanding of the microwave surface impedance of critical importance.

Although extensive microwave data have been reported for polycrystals, single crystals, and films of the Y-Ba-Cu-O and Bi-Sr-Ca-Cu-O systems, there are few data on the thallium system. Because single-crystal Tl superconductors have typically only about 10% the volume of typical Bi-Sr-Ca-Cu-O crystals, those microwave measurements that have been performed have used polycrystalline Tl-Ba-Ca-Cu-O samples. Unaligned polycrystalline Tl-Ba-Ca-Cu-O samples usually show a $g=2$ resonance^{3,4} due to Cu^{2+} impurities, which disappears below T_c and is replaced by a nonresonant, low-field peak, presumably

due to weak intergranular links. These data have been hard to interpret. The present work reports the temperature, modulation frequency, and angular dependence of the modulated microwave surface resistance of a single-crystal $\text{Tl}_2\text{Ca}_2\text{BaCu}_2\text{O}_8$ (Tl 2:2:1:2) superconductor. No $g=2$ resonance was observed at any temperature, indicating good sample quality.

Sample preparation has been reported elsewhere.⁵ The experiment was carried out on a Varian TE₁₀₂ reflection cavity with a loaded Q of about 3800 operating at a frequency of 9.3 GHz. An APD flow-through system and a TRI Research temperature controller were used to control the temperature of the helium flow. The Dewar is made of concentric quartz tubes, within which the sample is mounted on a thin sapphire plate. In a flow-through system such as this, the sample temperature is very different from the control temperature. To measure the temperature of the sample, a thermocouple was mounted on one end of the sapphire plate, located just outside the cavity. The thermocouple was then calibrated using a calibrated LakeShore Ga-Al-As diode sensor.

The sample was located in the region of maximum magnetic field and zero electric field, accomplished by maximizing the signal. The microwave magnetic field was always perpendicular to the applied magnetic field. While it is desirable to measure the surface impedance directly through the change in the reflected power as the applied field is varied, the small Tl 2:2:1:2 single crystal requires that field modulation be employed to detect the derivative signal with respect to the applied field. The field modulation used in this experiment is 8 G peak to peak. The signal was found to be linear with a modulation field up to 16 G peak to peak. The samples used in this experiment are two Tl 2:2:1:2 single crystals, each $0.3 \times 0.3 \times 0.4 \text{ mm}^3$, which come from a larger one. The sample was characterized by magnetization measurement

on a Quantum Design superconducting quantum interference device (SQUID) magnetometer to check the sharpness of the transition width. In a field of 5 G, T_c , defined as the onset temperature, is 102 K with $\Delta T_c(10\%-90\%) \approx 8$ K.

Shown in Fig. 1 is the magnetic-field dependence of the microwave signal at $T = 79$ K at various angles θ , defined as the angle between the applied magnetic field and c axis of the Tl 2:2:1:2 crystal. The rotation axis is parallel to the microwave magnetic field. The overall shape of the modulated surface resistance is qualitatively similar to that observed for Bi-Sr-Ca-Cu-O and Y-Ba-Cu-O crystals.⁶ For $0^\circ < \theta < 60^\circ$, the microwave signal is independent of θ . The peak position moves to higher field and the intensity decreases only when θ is close to 90° . This behavior is consistent with the results from torque measurements on Tl 2:2:1:2 films, which exhibit giant anisotropy.⁷

The variation of the microwave signal with temperature is shown in Fig. 2 for $\theta = 0^\circ$. At a fixed temperature, the signal shows a very sharp peak at low field and a broad maximum at high field. The nonresonant, low-field peak is much stronger in the Tl crystal than in the Y-Ba-Cu-O and Bi-Sr-Ca-Cu-O crystals.⁶ The low-field peak position occurs at an applied field of about 9 G. In polycrystalline samples the low-field dissipation is usually attributed to flux lines moving in and out between grains randomly coupled by Josephson weak links. As it depends on sample preparation and grain sizes,⁸ it is not intrinsic to the materials. Here the low-field peak probably occurs when magnetic-flux lines suddenly penetrate the sample as the applied field crosses H_{c1} . In the remainder of this paper, we focus on the high-field peak.

The broad peak observed at higher field is asymmetric and occurs in a narrow temperature window. For Y-Ba-Cu-O and Bi-Sr-Ca-Cu-O single crystals, this restricted temperature window occurs for $0.8T_c \leq T \leq T_c$.⁶ For the

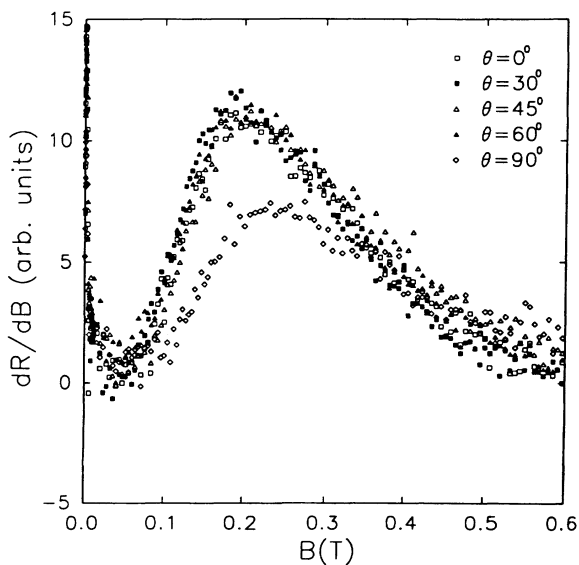


FIG. 1. Modulated microwave signals at different orientations: θ is the angle between the applied magnetic field and c axis of the $\text{Tl}_2\text{Ca}_2\text{BaCu}_2\text{O}_8$ crystal.

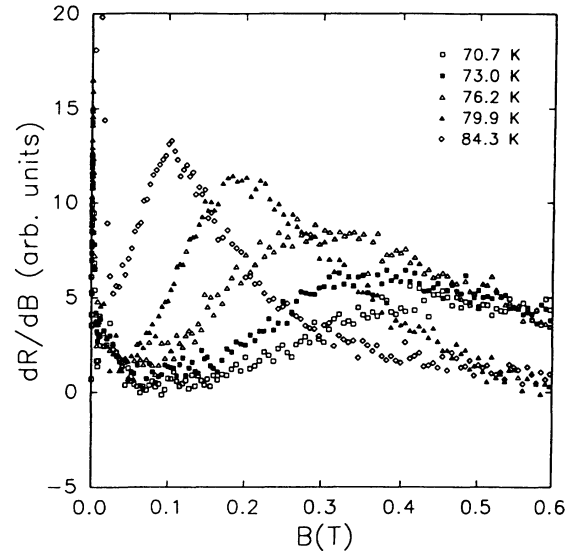


FIG. 2. Modulated microwave signals at various temperatures for a $\text{Tl}_2\text{Ca}_2\text{BaCu}_2\text{O}_8$ crystal at $\theta = 0^\circ$.

Tl single-crystal superconductor used in this experiment, the temperature window occurs between 60 and about 85 K. For $T > 85$ K the broad peak merges with the low-field peak. The low-field peak intensifies and then disappears above T_c . The broad-peak magnetic field B_{peak} , defined at the maximum of the signal at $T = 76.2$ K, is larger than that observed in the Bi-Sr-Ca-Cu-O single crystals at the same temperature.⁶

The irreversibility line is determined from dc magnetization measurements. Shown in the inset of Fig. 3 is a plot of the magnetization curves for a field of 0.45 T. The temperature at which the zero-field-cooled (ZFC) and field-cooled (FC) curves do not coincide is called the irreversible temperature T_{irr} . In Fig. 3 we plot $B(T_{\text{irr}})$ determined from magnetization curves similar to the inset, along with $B_{\text{peak}}(T)$. The solid lines are power-law fits: $B_{\text{irr}} \propto (1 - T/T_c)^{5.6}$ and $B_{\text{peak}} \propto (1 - T/T_c)^{2.6}$, where T_c is taken as the superconducting onset temperature 102 K. This shows that for Tl 2:2:1:2 crystal, B_{irr} and B_{peak} do not share the same temperature dependence, while for Y-Ba-Cu-O and Bi-Sr-Ca-Cu-O systems, B_{irr} and B_{peak} coincide.^{6,9,10}

For the curve taken at $T = 84.3$ K in Fig. 2, the tail approaches a constant value at a field of about 0.6 T. If we take $B_{c2}(T=0) \approx 100$ T and $B_{c2}(T) = B_{c2}(0)(1 - t^2)/(1 + t^2)$, where $t = T/T_c$, then $B_{c2}(84.3 \text{ K}) \approx 18.8$ T, which is much larger than 0.6 T. We conclude that the modulated surface resistance saturates at fields much smaller than B_{c2} , but considerably larger than B_{irr} at the same temperature. Saturation behavior was also observed in the Bi-Sr-Ca-Cu-O superconductor.⁶ However, as we describe below, B_{peak} depends logarithmically on modulation frequency, tending toward B_{irr} as the frequency is reduced. In the rest of this paper, we take $T_c = 102$ K, the superconducting onset temperature.

The microwave surface impedance of superconductors

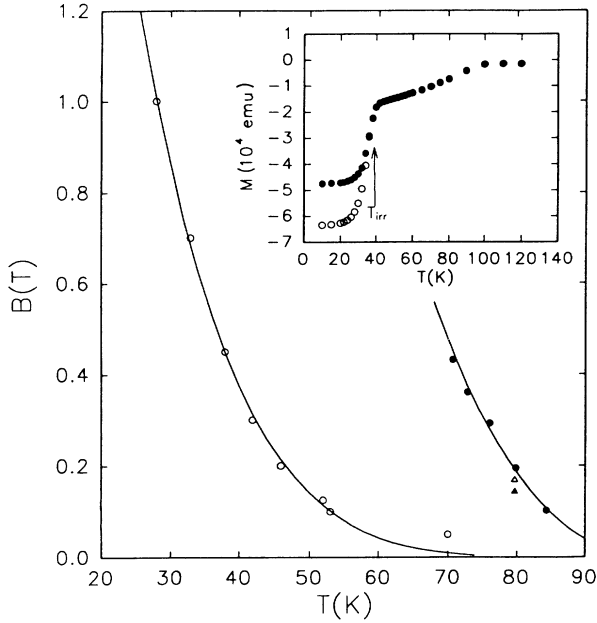


FIG. 3. Temperature dependence of the irreversibility field of a $\text{Tl}_2\text{Ca}_2\text{BaCu}_2\text{O}_8$ crystal at $\theta=0^\circ$. Open circles are determined from dc magnetization measurements. Microwave measurements of B_{peak} at various modulation field frequencies are 100 kHz (\bullet), 50 kHz (\triangle), and 30 kHz (\blacktriangle). The lines through the data are fits to $B \propto (1 - T/T_c)^\alpha$, where $T_c = 102$ K. Inset is a plot of the Meissner (FC) and shielding (ZFC) curves for a field of 0.45 T along the c axis of a $\text{Tl}_2\text{Ca}_2\text{BaCu}_2\text{O}_8$ crystal.

was first treated by London in the context of the two-fluid model.¹¹ Subsequently, Caroli and Maki,¹² Hu and Thompson,¹³ and Gorkov and Kopnin¹⁴ undertook a detailed treatment of the vortex state of type-II superconductors in the dirty limit.¹²⁻¹⁴ Even in the dirty-limit approximation, only the immediate regions of $B \rightarrow 0$ and $B \rightarrow B_{c2}$ were treated in detail. Recent analyses of high-temperature superconductors^{15,16} have reverted to the phenomenological London approach, modified by the addition of a flux-flow-induced electric field. However, while the two-fluid model correctly approaches the normal-state limit at T_c and the perfect superconducting reactance at $T=0$, the phenomenological approach that adds flux-flow and superconducting impedances in series does not give the correct limits. In particular, these models do not revert to the normal-state surface impedance $Z = (1+i)\mu_0\omega\delta_n/2$ as $T \rightarrow T_c$ or $B \rightarrow B_{c2}$, where δ_n is the normal-state skin depth. An appropriate model for the surface impedance must correctly include the dynamics of the flux line, while taking account of thermally activated flux flow (TAFF), flux creep, and the possibility of a melting line.

In an attempt to explain the field-dependent dc resistivity, Tinkham combined Ambegaokar and Halperin's model of the single overdamped Josephson junction with the thermally activated depinning model described by Yeshurun and Malozemoff.^{9,17,18} He found that the resistance $R = R_n/I_0^2(\nu)$, where $I_0(\nu)$ is the modified Bessel function, $\nu = A(1-t)^{3/2}/2B$, $A = UB_{c2}(0)/k_B T$, and U

is the activation energy barrier. In this model the temperature width of transition scales as $\Delta T \propto B^{2/3}$ and dR/dB has a temperature-dependent maximum as a function of B .

More recently, Coffey and Clem (CC) calculated the microwave surface impedance over a wide range of frequencies ω , magnetic induction fields B , and temperatures T .² The theory takes into account the effects of pinning and flux creep. Their expression for the flux-flow portion of the complex penetration depth differs from Tinkham's result in that only the fraction B/B_{c2} of the flux lattice contributes (similar to the Bardeen-Stephen approach). The theory reduces to the London two-fluid model as the vortex density goes to zero and has the correct limiting form as the system approaches the normal state. The theory is valid for $B/\mu_0 \gg H_{c1}$ and the intervortex spacing $a_0 \ll \lambda$, the weak-field penetration depth. They consider the microwave magnetic field \mathbf{b} to be parallel to the applied magnetic field, in which case the Lorentz force produced by \mathbf{b} causes the flux lines to shake back and forth. However, the present experiment has \mathbf{b} perpendicular to the applied magnetic field. In this configuration the flux lines bend because they have a finite shear modulus. In either case the rf penetration depth is greatly increased by the motion of the flux lattice. It turns out that when \mathbf{b} is perpendicular to the applied magnetic field, the same expressions hold as when \mathbf{b} is parallel to the applied field, but with different flux-line moduli.¹⁹

The surface resistance R_s is expressed in terms of an effective complex penetration depth $\tilde{\lambda}$:

$$R_s = -\mu_0\omega \text{Im}\tilde{\lambda}, \quad (1)$$

where ω is 2π times the experimental frequency, and

$$\tilde{\lambda}(\omega, B, T) = \left[\frac{\lambda^2(B, T) - \frac{1}{2}i\delta_v^2(\omega, B, T)}{1 + 2i\lambda^2(B, T)/\delta_{\text{NF}}^2(\omega, B, T)} \right]^{1/2}. \quad (2)$$

Here λ is the superconducting penetration depth, δ_{NF} is the normal-fluid skin depth, and δ_v is the complex effective skin depth arising from vortex motion. Taking $\lambda(0) \approx 1500$ Å, $T = 80$ K, and $B = 0.6$ T, we find

$$\begin{aligned} \lambda(B, T) &= \lambda(0) / \{(1-t^4)[1 - B/B_{c2}(T)]\}^{1/2} \\ &\approx 0.19 \mu\text{m}, \end{aligned}$$

where t is the reduced temperature. In the superconducting state, both δ_v and δ_{NF} are always larger than the normal-state microwave skin depth, about $1 \mu\text{m}$, so that $\lambda^2 \ll \delta_v^2$ and $\lambda^2 \ll \delta_{\text{NF}}^2$. Thus, to the first approximation, we can ignore λ^2 , in which case Eq. (2) reduces to

$$\tilde{\lambda}(\omega, B, T) = \left[-\frac{1}{2}i\delta_v^2(\omega, B, T) \right]^{1/2}. \quad (3)$$

Following Coffey and Clem, we have

$$\delta_v^2(\omega, B, T) = \frac{2\rho_f}{\mu_0\omega} \left[\frac{\varepsilon + i(1-\varepsilon)\omega\tau}{1 + (\omega\tau)^2} \right], \quad (4)$$

and $\omega\tau$ is given by

$$\omega\tau = \frac{\omega\eta}{\kappa_p} \left[\frac{I_0^2(\nu) - 1}{I_0(\nu)I_1(\nu)} \right] \approx \frac{\omega\eta}{\kappa_p}, \quad (5)$$

where $\eta = \phi_0 B_{c2} / \rho_n$ is the flux-flow viscosity, κ_p is the restoring force constant of pinning, and $\varepsilon = 1 / I_0^2(\nu)$. The flux-flow fraction ρ_f is taken to be $\rho_n(T)B / B_{c2}(T)$, following Bardeen and Stephen.²⁰ Taking $B_{c2}(T=80 \text{ K}) \approx 24 \text{ T}$, $\phi_0 = 2 \times 10^{-15} \text{ T m}^2$, $\rho_n \approx 2 \times 10^{-6} \Omega \text{ m}$, and $\kappa_p = 2 \times 10^4 \text{ N/m}^2$, $\omega\tau \approx 0.07$, which is small, and so we only keep first-order terms. The surface resistance is then

$$R_s = \left[\frac{\mu_0 \omega \rho_f}{2} \left(\omega\tau + \frac{1 - \omega\tau}{I_0^2(\nu)} \right) \right]^{1/2}. \quad (6)$$

Field modulation at 100 kHz was used to detect dR_s/dB , the derivative of R_s with respect to the applied field. Here B is the induction field. In this paper we neglect the demagnetization factor so that the applied magnetic field $\mu_0 H = B$. Differentiating Eq. (4) with respect to B gives

$$\frac{dR_s}{dB} = \frac{D}{2\sqrt{BP}} \left[P + (1 - \omega\tau) \frac{2\nu I_1(\nu)}{I_0^3(\nu)} \right], \quad (7)$$

where $P = \omega\tau + (1 - \omega\tau) / I_0^2(\nu)$ and $D = [\mu_0 \omega \rho_n(T) / 2B_{c2}(T)]^{1/2}$ is a constant for a given T .

Shown in Fig. 4 are the fits of the CC theory and Tinkham modification ($\rho_f = \rho_n$) to the experimental data at $T = 76.2$ and 79.9 K with $\nu = A(1-t)^{2.6} / 2B$. Clearly, setting $\rho_f = \text{const}$ fits the data better at high fields and the CC model ($\rho_f \propto B$) at low fields. Table I gives the fitting parameters of the two temperatures. The energy barrier

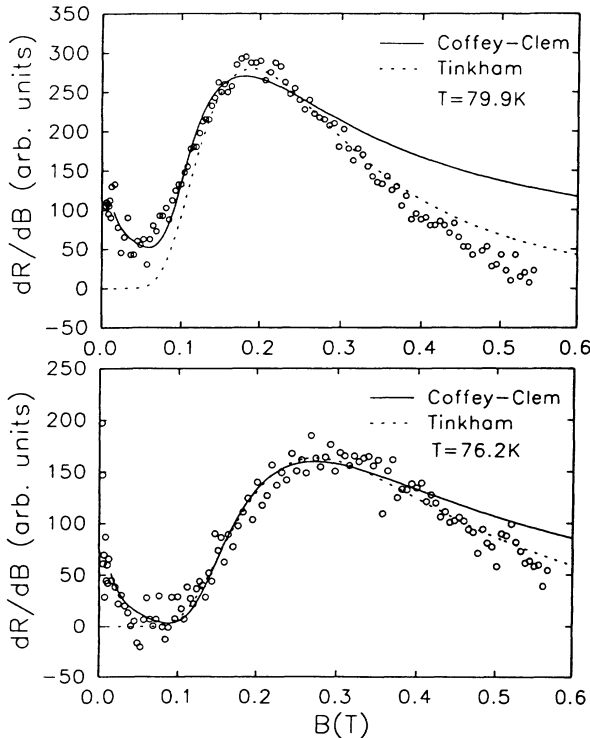


FIG. 4. Modulated microwave signals at $\theta = 0^\circ$ for $T = 76.2$ and 79.9 K . Solid line is a fit with Eq. (7) of Ref. 2, and dotted line is a fit to the result of Ref. 18.

TABLE I. Tl 2:2:1:2 fitting parameters.

T (K)	A (T)		U (meV)	
	Ref. 18	Ref. 2	Ref. 18	Ref. 2
76.2	35.9	39.6	2.35	2.60
79.9	37.4	39.5	2.58	2.72

height U was calculated from $A = UB_{c2}(0) / k_B T$ using $B_{c2}(0) = 100 \text{ T}$.⁹ The best fit of the CC model to the data at the same temperature was obtained with $\omega\tau \approx 0.0265$.

The data suggest that the flow resistance ρ_f is not given by the simple Bardeen-Stephen resistivity. Indeed, recent dc resistivity measurements by Chien *et al.* on microtwinning single crystals of $\text{YBa}_2\text{Cu}_3\text{O}_7$ do not follow the simple Bardeen-Stephen flux-flow resistivity at any field or temperature.²¹ A calculation of ρ_f / ρ_n by Hu and Thompson found that $\rho_f / \rho_n < B / B_{c2}$ for all B , increasing as $0.33B / B_{c2}$ at low fields.¹³ This result does not improve the CC fit to the data. A plausible model is that $\rho_f \propto B$ for $B \leq B_{\text{peak}}$ and $\rho_f \approx \text{const}$ for $B \geq B_{\text{peak}}$. This in turn suggests that B_{peak} has some significance as a melting line and is not simply a crossover line. This effect is most pronounced in Tl 2:2:1:2, where the irreversibility line falls far below $H_{c2}(T)$.

The CC model assumes a single activation energy and does not consider collective effects. However, the apparent saturation of ρ_f for fields larger than B_{peak} argues for collective freezing rather than simple TAFF. One indicator of glassy behavior is the frequency dependence of the irreversibility line, discussed by Malozemoff *et al.*¹ In the above analysis the line of peaks in Fig. 3 is given by $\nu = \nu_p$, the value that maximizes Eq. (6). However, ν must be related to the maximum-energy barrier that can be crossed at frequency scale f , the modulation frequency in the experiment, resulting in a new definition of that time line as

$$A' \frac{(1-t)^{2.6}}{B \ln(f_0/f)} = \nu_p. \quad (8)$$

Here f_0 is an attempt frequency and A' is related to the 100-kHz values in Table I through $A' = A \ln[f_0 / (100 \text{ kHz})]$. Data taken at a fixed temperature show a systematic reduction of B_{peak} with modulation frequency, as shown in Fig. 4. In Fig. 5 we plot $1/B_{\text{peak}}$ vs $\ln(f)$ for $T = 79.7 \text{ K}$. The line corresponds to $f_0 \approx 4 \text{ MHz}$. This low attempt frequency is perhaps indicative of the shallow pinning wells that appear to characterize the flux phase of Tl 2:2:1:2.

In a recent work, Brandt calculated the surface impedance within linear-response theory.²² The theory accounts for the boundary conditions (image vortices), diffuse driving force, nonlocal elasticity of the vortex lattice, flux flow, and thermally activated flux creep. The calculation was performed for a small perturbing ac field, and the result was generalized to account for flux flow and flux creep. For $\omega\tau_0$ small the resulting surface resistance R_s , to first order, does not have a maximum in dR_s/dB when we assume $U \propto 1/B$. This is in contrast to the experimental data, which show a broad maximum in

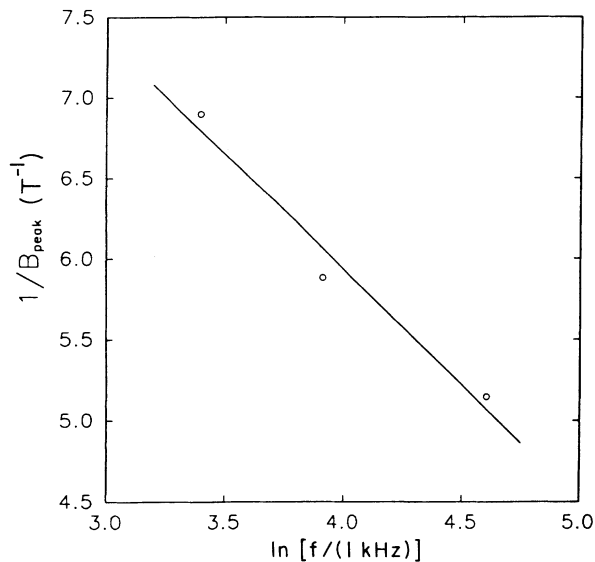


FIG. 5. Dependence of B_{peak} on modulation frequency at $\theta=0^\circ$.

dR_s/dB .

Zuo *et al.* showed previously that $(dR_s/dB)B_{\text{peak}}$ is a universal function of B/B_{peak} .⁶ A test of this is shown in Fig. 6 at four different temperatures. Within experimental errors the data scale quite well. In Eq. (6) note that when we set $\rho_f = \text{const}$, dR_s/dB has this scaling property, and that when we set $\rho_f \propto B$, Eq. (7) has $(dR_s/dB)\sqrt{B_{\text{peak}}}$ as a universal function of B/B_{peak} .

In summary, we have measured the modulated microwave surface resistance in a single-crystal $\text{Tl}_2\text{Ca}_2\text{BaCu}_2\text{O}_8$ superconductor. Because of the frequency dependence of the measurements, the irreversibility line determined from dc magnetization measurement does not have the same temperature dependence as B_{peak} . The Coffey-Clem model fits the data quite well for $B \leq B_{\text{peak}}$ and the Tinkham model for $B \geq B_{\text{peak}}$. We sug-

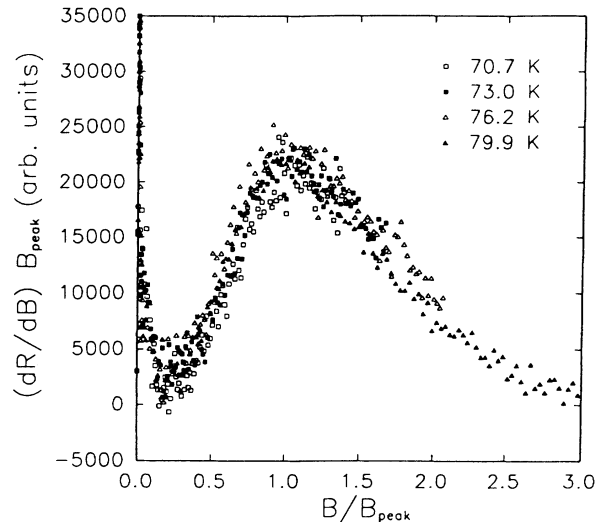


FIG. 6. Modulated microwave signal plotted with the x axis scaled to B/B_{peak} and the y axis scaled to $(dR/dB)B_{\text{peak}}$ at four different temperatures.

gest that $\rho_f \propto B$ for $B \leq B_{\text{peak}}$ and $\rho_f \approx \text{const}$ for $B \geq B_{\text{peak}}$. These theories must account for the fact that the surface resistance saturates at B much smaller than B_{c2} but larger than B_{irr} . The Tl 2:2:1:2 and Bi-Sr-Ca-Cu-O single crystals have the same scaling property.

We would like to thank M. W. Coffey and J. Clem for useful discussions and for providing reports of their work prior to publication. We also thank E. H. Brandt for making his results available to us prior to publication. One of us (N.H.T.) would like to thank K. Ghiron, J. Shi, and R.-C. Yu for helpful discussions, K. Ghiron for help with using the SQUID, and K. O'Hara for use of his fitting program. This work has been supported by the National Science Foundation (NSF) Grant No. STC 88-09854 through the Science and Technology Center for Superconductivity and NSF Grant No. DMR 89-20538.

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