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Nonlinear magnetization of Y-Ba-Cu-O crystals

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We investigate the nonlinear magnetization $M(B)$ of an $YBa_2Cu_3O_{7-x}$ crystal in applied superimposed dc and ac magnetic fields B up to about 80 G. $M[B(t)]$ contains both even and odd harmonics of the applied fundamental audio frequency in a restricted temperature range of 1-2 K below T_c. Harmonic amplitudes oscillate with both temperature and dc field. We propose a simple dynamical model based on the observed "flux creep" resistance in which temperature and field appear only in the ratio $(1-T/T_c)^{3/2}/B$. The model is in qualitative and semiquatitative agreement with the data.

The markedly nonlinear magnetization $M(B)$ of polycrystalline Y-Ba-Cu-O samples[1,2] was recently added to the list of novel magnetic-field-induced properties of the high-T_c materials which includes the field-induced broadening of the transition,[3-5] irreversible magnetization,[6] hysteresis,[7] and unusual nonresonant microwave absorption.[8] Efforts to understand some of these phenomena in the theoretical framework of the thermally activated flux creep were triggered by the interpretation of the "irreversibility line" $1-T_{irr}/T_c \propto B^{2/3}$ by Yeshurun and Malozemoff.[6] Recent phenomenological models by Tinkham[9] and Palstra *et al.*[3] as well as an analytical critical state model by Hagen, Griessen, and Salomons[10] show that the resistive transition may also be described in the framework of flux creep theory.

The nonlinear $M(B)$ of $YBa_2Cu_3O_{7-\delta}$ polycrystalline and powder samples, as measured by harmonics in $M[B(t)]$ at 77 K, has been attributed to Josephson junctions between grains,[1,11] rather than to flux creep, primarily because the amplitudes of harmonics of M oscillate periodically with B_{dc}, reminiscent of conducting loops containing Josephson junctions.[11] An alternative interpretation[2] of the oscillations with B_{dc} is a variation of the Bean critical state model,[12] in which the field penetration is nonuniform, and results in similar semiquantitative agreement with the data.

The present study focuses on $M(B)$ in a high-quality $YBa_2Cu_3O_{7-\delta}$ crystal for comparison with polycrystalline materials. Our main results are the following: (1) harmonics above the fundamental exist only in the temperature region of the field-broadened transition measured by ac susceptibility, (2) the amplitudes of the higher harmonics oscillate as a function of *temperature* as well as B_{dc}/B_{ac}, and (3) the oscillations can be understood in terms of a highly idealized model in which the nonlinearities arise from the measured magnetoresistance of the material.[3,4] The key feature of the model, which is based on Tinkham's scaling model[9] of the magnetoresistance, is that T and B enter only in the ratio $(1-T/T_c)^{3/2}/B$, so that oscillations with B are naturally associated with oscillations with T.

Our apparatus is based on the extensively used "two coil" method. It consists of a solenoidal primary coil that generates both dc and ac fields, and a secondary comprising two counterwound flat spiral coils placed coaxially with each other and with the primary. The $YBa_2Cu_3O_{7-\delta}$ crystal lies on one of the two pickup coils so that the c axis of the crystal is parallel to the applied fields. The voltage $V(t)$ induced in the secondary is proportional to the time derivative of the magnetization

$$V(t) \propto \frac{dM}{dt} = \omega B_{ac} \sum_{n=1}^{\infty} n[-\chi_n' \sin(n\omega t) + \chi_n'' \cos(n\omega t)] , \quad (1)$$

where the magnetic field is $B = B_{dc} + B_{ac}\cos\omega t$. While one can integrate the signal to obtain[2] $M(B)$, conventionally one studies the amplitudes of the nth harmonics, $V_n \propto n\omega[\chi_n'^2 + \chi_n''^2]^{1/2}$, to focus on deviations from linear response. V_n is measured with a lock-in amplifier with a systematic uncertainty of about 20% imposed by the geometry of our setup.

The crystal was prepared in our laboratory using standard flux-rich techniques.[13] It is flat and rectangular with rough dimensions of $1\times1\times0.1$ mm^3 and its orthorhombic c axis lies along the shortest edge. It has a very sharp transition, with $T_c \approx 90.5$ K and width $\delta T_c < 0.3$ K at $B_{ac} = 4$ G and $B_{dc} < 10$ mG. In all our measurements the sample is cooled at $B_{dc} < 10$ mG, fields are applied, and data are taken while warming slowly. In our estimation for the applied fields we used a demagnetizing factor of 0.8.

We measured V_n from $n=1$ to 5 as a function of T, dc field $0 \le B_{dc} \le 50$ G, ac field $4 \le B_{ac} \le 50$ G and frequency 1 kHz $\le f \le$ 10.1 kHz. V_n is proportional to f in this frequency range. Figures 1 and 2 show V_1 through V_5 at $B_{ac} = 50$ G, $B_{dc} = 7.5$ G, $f = 10.1$ kHz. In Fig. 1(a), the in-phase $V_1' \propto \chi_1'(T)$ and the out-of-phase $V_1'' \propto \chi_1''(T)$ components of the fundamental lock-in voltage indicate the transition broadening. Within a 20% uncertainty, the amplitude of V_1' below 89 K corresponds to complete expulsion of the ac field.

Figures 1(b), 1(c), 2(a), and 2(b) show that V_2, V_3, V_4, and V_5 are nonzero only in the resistive transition where the material is still dissipative, i.e., where V_1'' is nonzero. This result is central to our interpretation. Furthermore, as shown in the figures, V_2 through V_5 oscillate with T and the number of oscillations increases with n. These

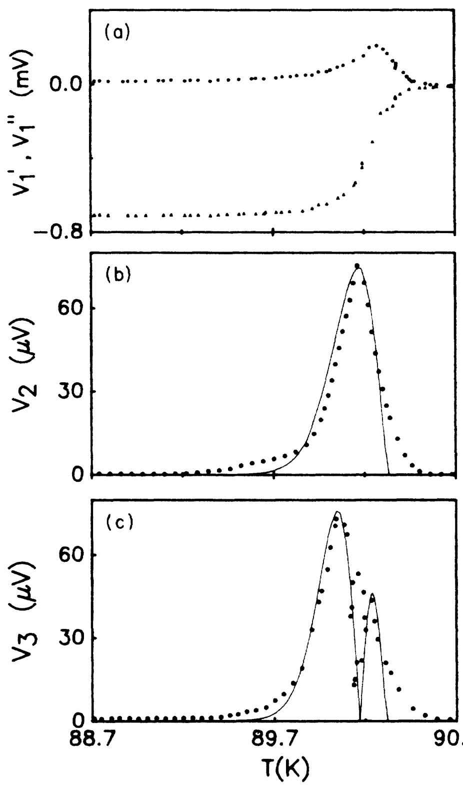

FIG. 1. (a) In-phase V_1' (triangles) and out-of-phase V_1'' (circles) component at $f=10.1$ kHz. (b) Second-harmonic-amplitude data and fit (solid line) with $A=9\times10^5$ G, $w=6$. (c) Third-harmonic-amplitude data and fit (solid line) with $A=9\times10^5$ G, $w=5$.

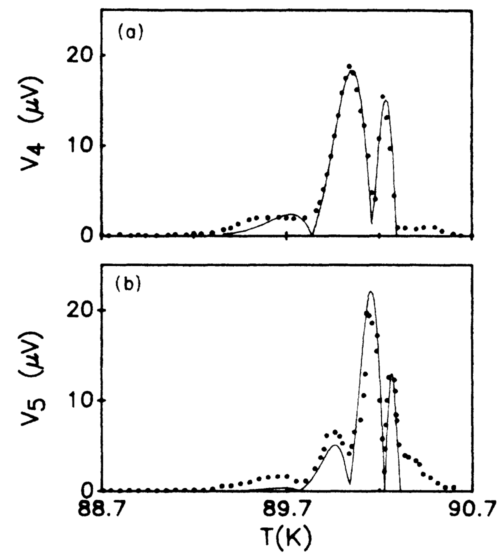

FIG. 2. (a) Fourth-harmonic-amplitude data and fit (solid line) with $A=7\times10^5$ G, $w=6$. (b) Fifth-harmonic-amplitude data and fit (solid line) with $A=9\times10^5$ G, $w=6$.

surprising oscillations have not been reported in ceramic or powder samples.

Figure 3 shows for comparison V_2 vs B_{dc}/B_{ac} with the node at $B_{dc}=0$ reported and discussed for ceramic samples.[1,2] The asymmetry with field in Fig. 3 is an artifact of a small temperature drift during the measurement. We have also observed amplitude oscillations for $n=3$, 4, and 5. For all n, oscillations of V_n vs B_{dc}/B_{ac} are qualitatively similar to results on polycrystals in that they are symmetric about $B_{dc}=0$, the even harmonics have a zero at $B_{dc}=0$ where the odd harmonics have a maximum, and the amplitude of the peaks decreases for large B_{dc}.

We interpret these data as a consequence of the magnetoresistance of crystals near T_c. In flux creep theory, a resistance appears in a current-carrying conductor as a result of thermally activated flux motion biased by the driving force on the flux lines caused by the current. For $YBa_2Cu_3O_{7-\delta}$, Tinkham[9] showed on physical grounds why a good approximation for the dc resistance R is

$$R=R_n/\{I_0[A(1-T/T_c)^{3/2}/2B]\}^2\,, \qquad (2)$$

where R_n is the normal-state resistance, I_0 is a modified Bessel function, and B is the applied dc magnetic field. The parameter A is proportional to the Ginzburg-Landau depairing critical current density at $T=0$, and it controls the width of the resistive transition in a field. A central feature of the model is that the activation energy for flux motion is proportional to the ratio $(1-T/T_c)^{3/2}/B$.

In our experiment, where B oscillates, we assume that R is given by Eq. (2), at least for small frequencies. Unfortunately, the solution for eddy currents in a conductor with such a nonlinear magnetoresistance is unknown. We turn to a highly idealized discrete-circuit model, which we believe contains the essential physics. Calculated curves are shown in Figs. 1–3 as solid lines.

In this model the shielding current $I(t)$ circles the sample in a single closed loop of inductance L feeling a resistance R due to flux creep. The equation for the current I in the loop is taken to be

$$L\,dI/dt+RI=d\Phi/dt=-B_{ac}S\omega\sin\omega t\,, \qquad (3)$$

where Φ is the flux through the surface S of the loop and $B_{ac}\cos\omega t$ is the applied ac field. The form of $R(t)$ in this

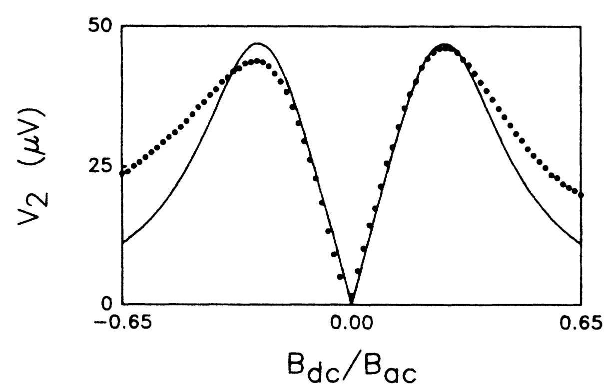

FIG. 3. Field dependence of second harmonic at $T=90.1$ K. The solid line is a fit with $A=9\times10^5$ G, $w=5$.

model is

$$R(t)=R_n/\{I_0[A(1-T/T_c)^{3/2}/2\,|\,wB_{dc}+B_{ac}\cos\omega t\,|\,]\}^2 . \qquad (4)$$

The parameter w is needed to fit the dependence of V_n on B_{dc}/B_{ac}. It is probably related to the different penetration depths of the dc and ac fields into the crystal, but the connection is vague. w affects only the periodicity of oscillations in V_n vs B_{dc}/B_{ac}, not their existence or amplitudes.

The measured signal is $V(t)\propto dM/dt \equiv L\,dI/dt$, and nonlinearities arise from the time dependence of R modulating $I(t)$. We note than an immediate consequence of the model is that harmonics are restricted to the temperature region of the broadened transition, since the same function $R[B(t)]$ governs the appearance of the harmonics and the transition broadening. Furthermore, since the activation energy in the model scales the field with the temperature as $B\propto(1-T/T_c)^{3/2}$, the oscillations of V_n vs B_{dc} are naturally connected to oscillations versus T.

While it is possible to solve Eq. (3) analytically in terms of integrals of $R(t)$, it is more useful to work with an approximate solution valid for $\omega L/R \gg 1$. Our assumption that the screening currents circle the crystal at a single loop could be valid only in the temperature range where the resistance is small and the pinning force is large, so that the currents are at the edge of the crystal, the inductance is relatively large, and $\omega L/R \gg 1$ is reasonable. This limit breaks down close to T_c, probably above the peak in V_1'' at 90.3 K, where the fields sweep through the crystal in the absence of strong pinning and L gets smaller, while R becomes large. However, at these high temperatures, the scaling model of magnetoresistance also breaks down, since it describes an infinitely sharp transition in zero field and the measured transition is 0.25 K wide. Hence, a comparison between the data and the model makes sense only below 90.3 K, where $\omega L/R \gg 1$.

The approximate solution to Eqs. (3) and (4) is

$$I(t)=(B_{ac}S/L)\cos\omega t-(B_{ac}S/L^2\omega)R(t)\sin\omega t . \qquad (5)$$

In keeping with the simplicity of the model, we neglect any dependence of L and S on f and T. For $n>1$, noting that $R(\omega t)$ is symmetric about π,

$$V_n=-an\omega\int_0^{2\pi}\sin(n\omega t)R(\omega t)\sin(\omega t)d(\omega t) , \qquad (6)$$

where a is a coupling constant.

The solid lines in Figs. 1(b), 1(c), 2(a), and 2(b) show the calculated T dependence of V_n for $2\le n\le 5$ and $B_{dc}/B_{ac}=7.5$ G/50 G. We used $A=9\times10^5$ G, $w=6$, and $T_c=90.3$ K for all the fits, except $A=7\times10^5$ G for $n=4$ and $w=5$ for $n=3$. The small variations in A and w may be attributed to systematic uncertainties in the experiment. We note that for $n=3$, the fitted coupling constant a is double the value for other harmonics. This probably reflects the crudeness of the model, which may entirely neglect other possible sources of nonlinearities. As expected the fits for all harmonics are poor very close to T_c, but are quite good at lower temperatures. With similar success we fit the temperature dependence data of V_n in the entire range of fields we examined. We conclude that the model contains the essential physics, namely the scaling equivalence of B and T, despite its simplicity. It is not clear that any other model in the literature can explain our data for V_n vs T as well.

There is a simple connection implicit in the model between the oscillations of V_n vs T and V_n vs B_{dc}/B_{ac}. As B_{dc}/B_{ac} increases, the peaks of V_n grow and move to lower T. Thus, seen at fixed T, V_n oscillates between successive peak values and zero. In agreement with crystal and polycrystalline data, the model predicts a node for all even harmonics and a maximum for the odd, at $B_{dc}=0$. A fit of the field dependence of the second harmonic at $T=90.1$ K, with $A=9\times10^5$ G and $w=5$, is shown in Fig. 3.

To test the applicability of the model to polycrystalline samples we need information about the field *and* the temperature dependence of higher harmonics, but such data are not currently available. As noted by Tinkham,[9] the weaker coupling between grains in these samples reduces the value of the critical current densities[9] by a factor of 100, broadens the resistive transition in a field, and consequently decreases the parameter A by the same factor of 100. If our model is valid for polycrystals with the different microscopic structure, the change in A will not alter the qualitative predictions of the model, namely the oscillations of V_n with temperature and fields.

Taken together, our data and analysis demonstrate that the qualitative features observed in harmonics V_n vs B_{dc}/B_{ac} and V_n vs T, measured on crystals can be explained in terms of a nonlinear "flux creep" resistance. Despite the simplicity of the analysis, our results show the scaling between T and B in a clear fashion. Similar measurements would be interesting in other high-T_c superconductors like the Bi- and Tl-based ones with the different defect structure and interplanar coupling, where the activation potential has a more complicated B dependence[14] than that described in Tinkham's model.

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