JOURNAL DE PHYSIQUE
Colloque C8, Supplément au n° 12, Tome 49, décembre 1988

THE EFFECT OF FLUX PINNING AND FLUX CREEP ON MAGNETIC MEASUREMENTS OF SINGLE CRYSTAL $Y_1Ba_2Cu_3O_{7-x}$

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Abstract. – A series of noncontact magnetic measurements on high-quality single crystals of $Y_1Ba_2Cu_3O_{7-x}$ indicate that the superconducting properties are those of a conventional anisotropic superconductor. However, the magnetic properties are profoundly affected by flux pinning. We develop a classical, thermally activated flux flow model which we use to extract H_{C1} from the decay of the remnant magnetization at low temperature and to explain the logarithmic frequency dependence and the curvature seen in the apparent H_{C2} near T_C.

Introduction

In the new Cu-O superconductors, measurements on high quality single crystals are beginning to clarify the phenomenology of the superconducting state. These measurements generally indicae a similarity to previously known low temperature anisotropic superconductors; however, several puzzling and novel features have appeared. At low temperatures, even in the highest quality crystals, flux pinning has a profound effect on attempts to estimate H_{C1} from magnetization data [1-5]. Estimates of $H_{C2}(T)$ from ac susceptibility [6, 7] and resistance transitions [8-10] have given lower results than measurements of magnetization [11, 12] and heat capacity [13].

In this paper we review some early measurements of the anisotropic critical current in single crystals [1] and more recent measurements of the time dependant magnetization that are used to extract H_{C1} values in the presence of flux pinning [14]. We present new data on the field and frequency dependence of the ac susceptibility transitions near T_C and develop a classical flux creep interpretation which allows us to explain some of the puzzling details of previous attempts to measure H_{C2}. We find that flux creep phenomena dominate all aspects of the magnetic behavior of these new superconductors even in their bulk crystalline form.

The $Y_1Ba_2Cu_3O_{7-x}$ single crystals used in these studies were grown in two ways [1, 15] from $BaCuO_2$-CuO-$Y_1Ba_2Cu_3O_{7-x}$ compositions. The crystals were annealed in 1 atmosphere of oxygen at 400-450 C to obtain super-conducting transitions at 85-93 K. The crystals were of two general shapes, block like, with typical dimensions 400 μm by 400 μm by 100 μm and thin platelets typically 1 000 μm by 500 μ by 25 μm. Measurements of a single crystal in a scanning X-ray diffractometer showed it to be well transformed into the orthorhombic structure and twinned. TEM studies indicated that the twin domains were approximately 1 000 Å in width.

Magnetization measurements

Magnetization hysteresis loops, such as those shown in figure 1 for one of the crystals at 45 K, are traditionally used to estimate H_{C1} and provide a noncontact method for determining the critical currents density from the magnetic moment resulting from the induced screening currents. Loops were measured with the crystal mounted with the c axis parallel (Fig. 1a) and perpendiculat (Fig. 1b) to the magnetic field as illustrated in the insets. The differences in the scale and in the shape of the magnetization for the two orientations are striking. The critical current density, estimated using the Bean model for the critical state [16] is $J_C^{\parallel} = 2.9 \times 10^6$ A/cm^2 and $J_C^{\perp} = 4.20 \times 10^5$ A/cm^2. In our nomenclature, $J_C^{\parallel}$ is the critical current induced by fields applied parallel to the c axis such that the induced screening currents actually flow perpendicular to the c axis (in the Cu-O planes), and $J_C^{\perp}$ is the critical current determined from fields applied perpendicular to the c axis such that the currents flow perpendicular to the Cu-O planes. $H_{C1}^{\parallel}$ and $H_{C1}^{\perp}$, the lower critical fields, for a crystal oriented such that the applied field is parallel and perpendicular to the c axis, respectively, were estimated from the point in the initial part of each loop at which the departure from linearity begins. Because of pinning effects the precise field where the magnetization departs from linearity is difficult to identify, and because of demagnetization effects at the corners, the field of first measurable departure needs to be corrected to obtain H_{C1}. We earlier reported a significant deviation at an applied field, (corrected for demagnetization), of 5 200 Oe for H parallel to the c axis and 530 Oe for H perpendicular to the c axis. The strong pinning when the field is applied along the

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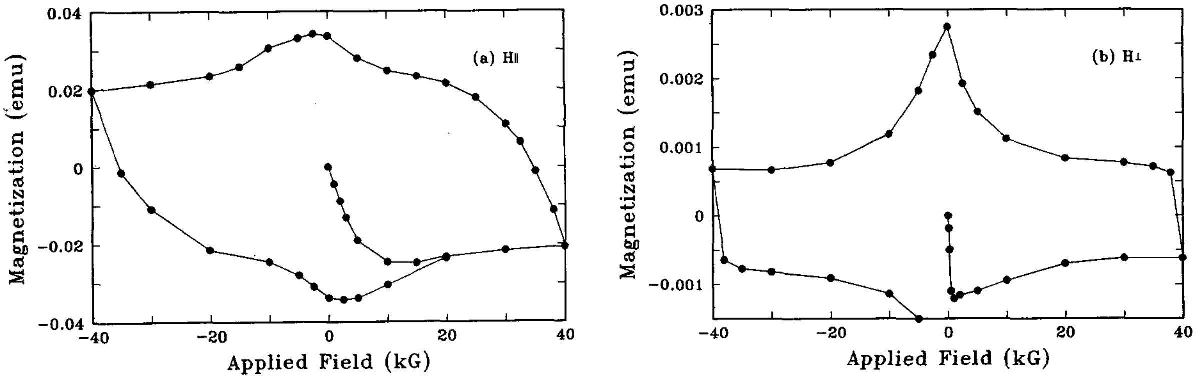

Fig. 1. – Magnetization hysteresis loops at 4.5 K for an annealed single crystal with the magnetic field oriented (a) parallel to the c axis and (b) prpendicular to the c axis

c-axis is evident in the continued rise in the magnetization up to an applied field of 10 kG in figure 1a. Measurements by McGuire *et al.* on higher quality crystals reported $H^{\parallel}_{C1} = 4\ 000$ Oe and $H^{\perp}_{C1} = 200$ Oe [2]. The crystals have large demagnetization corrections; for fields parallel to the c-axis, approximating the crystal as a oblate ellipsoid, the actual field seen by a sample is about twice the applied field for the thicker crystals and for the thinner crystals the demagnetization enhancement can be a factor of 10 or more. For fields parallel to the Cu-O planes, the demagnetization correction is much smaller, about 1.33. There is also evidence for a surface layer where the superconducting properties are suppressed [7]. We have also observed that often the twin structure near the corners is different from the bulk. Both of these effects suggest that measurements which are critically dependent on flux penetration at the surface, such as searches for the first sign of flux penetration to determine H_{C1}, might easily lead to incorrect results.

We have recently developed a new technique to determine H_{C1} [14]. This technique uses the logarithmic time dependance of the magnetization first noted in high T_C materials by Müller *et al.* [17] in ceramics and later by Worthington *et al.* in single crystals [18]. This decay in the magnetization [17-19] has been interpreted either as conventional flux creep or in terms of a superconductive glass state. Glassy like effects due to Josephson coupling are to be expected in ceramic samples where the grain boundaries provide natural tunnel junctions. However, in single crystals, the demonstration of a conventional flux lattice [20] and the consistent explanation of the low field magnetization results [21] are convincing evidence for conventional flux creep. However the size of the flux creep is vastly greater near T_C than in conventional superconductors because of the unprecedented high temperature and the low pinning energy.

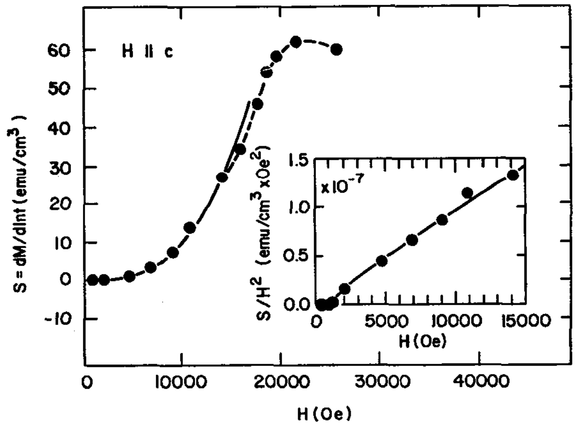

Fig. 2. – Relaxation rate of the zero-field cooled magnetization as a function of field (correct for demagnetization) for fields parallel to the orthorhombic c-axis of a $Y_1Ba_2Cu_3O_{7-x}$ crystal. Inset: relaxation rate $S = \mathrm{d}M\ /\ \mathrm{d}\ln t$ normalized by H^2 as a function of field. Solid lines are a fit to equation (1) with $n = 1$. Dotted line is a guide for the eye.

Figure 2 shows a plot of the logarithmic magnetization decay rate as a function of applied field (corrected for demagnetization effects) for the applied field parallel to the c axis at 6 K. To explain this relaxation Anderson [22] suggested a flux creep model in which flux lines in the critical stae hop over potential barriers U_0. We have extended this model to include a field dependence of the critical current and the possibility of only partial flux penetration. Our model predicts

$$\frac{\mathrm{d}M}{\mathrm{d}\ln(t)} = \frac{C}{D}\left[H^{n+2} - H_{C1}^{n+2}\right]\frac{kT}{U_0} \qquad (1)$$

for $H_{C1} \leq H \leq H^*$. Here C is a constant that depends on the maximum value of the critical current, D is the sample diameter and n describes the field dependence of the critical current $J_C \sim B^{-n}.H^*$ is the field at which currents first flow through the entire volume of the sample. A qualitative fit to this equation for $n = 1$ is shown in the inset in figure 2. The value of $H^{\parallel}_{C1}$ is 950 ± 100 Oe. The value extracted by this technique for fields applied perpendicular to the c axis $H^{\perp}_{C1} = 230 \pm 50$ Oe. We have studied two crystals, produced by different methods and with large differences in demagnetization corrections that have resulted in consistent H_{C1} values. The value for $H^{\parallel}_{C1}$ is smaller than those reported in references [1-3] but is consistent with the results of references [4, 5].

This relaxation technique has the advantage that it eliminaes the need to detect the first variation from a large linear background. Furthermore the theory of equation (1) provides a consistent analysis to extract H_{C1} from flux penetration into the bulk of the material; in this way it reduces the uncertainties from corner effects when trying to define H_{C1} as an onset of nonlinearity, torque or relaxation.

Upper critital field measurements

We have developed an ac susceptibility measurement technique in an attempt to probe the upper critical field as a function of temperature and crystal orientation [6, 7]. We use a small coil of copper wire, 0.7 mm id by 0.3 mm high glued to a small silicon chip. A crystal is placed in the center of the coil and held to the silicon with thermal grease. This mounting technique gives accurate alignment and maintains good thermal contact with the crystal. The grease also provides protection for the crystal and makes handling much easier. The crystal-coil assembly is glued to a copper block containing silicon diode, carbon glass, platinum and capacitance thermometers. The inductance and the series resistance of the coil were measured at 1 MHz with a Hewlett-Packard 4 194 A impedance analyser. After subtraction of the temperature dependent background measured with the coil empty, the inductance is proportional to the susceptibility, χ' and the series resistance is proportional to the ac losses in the sample, χ''. The ac field from the coil is $\sim$ 1 Oe. In field the transition broadens somewhat but remains sharp enough to easily determine a transition emperature as either the midpoint of the inductance drop or the resistance peak (which occur at the same temperature). This technique can be used to measure this transition temperature, as a function of field using ac fields at 10^5 to 10^8 Hz. Figure 3 shows the transition for a crystal in zero external field at 1 MHz. The quality of the crystals is judged by the sharpness of the ac susceptibility transition. Only crystals with sharp (less than 200 mK), single transitions have been used in these studies. We have recently extended the frequency range downward by changing the technique. A second coil is placed around the small coil containing the crystal and used to apply the ac field. A lock-in amplifier is used to measure the induced voltage in the small coil. This technique is useable from 100 to 10^5 Hz and has the advantage that only a negligible current flows in the inner coil so that there are no resistive losses in the copper that cause a temperature dependent background. The lock-in amplifier also has better noise performance at low frequency. These techniques have been used to measure the critical temperature, as defined by the peak in the losses as a function of field and frequency. Figure 4 shows new data of the apparent critical field *vs.* temperature, for the applied field parallel the c-axis at several measurement frequencies. Figure 5 shows the apparent critical temperature at 1 kG external field as a function of the frequency of the ac field for a different crystal.

Anisotropic Ginzbutg-Landau theory predicts that the critical field along the c-axis should be linear near T_C. The curvature which is evident in figure 4 is observed in both the resistively determined H_{C2} measurements [8-10] and those determined by ac susceptibility [6, 7]. This curvature has been variously attributed to dimensionality effects or non-mean-field critical fluctuations [10]. The latter interpretation is undermined by specific heat measurements [13] which indicate a critical region only a degree or so wide, while the curvature apparent in figure 4 persists more than 20 K below

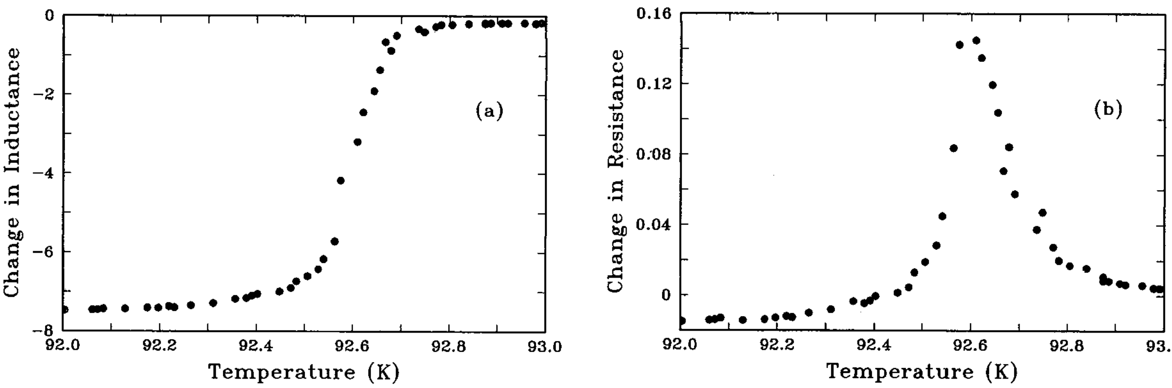

Fig. 3. – Temperature dependence of the inductive (a) and resistive (b) components of the ac susceptibility measured at 1 MHz for a $Y_1Ba_2Cu_3O_{7-x}$ single crystal.

T_C. Even more puzzling is the apparent frequency dependence of the transition temperature evident in the three sets of data in figure 4 and shown explicitly at one field in figure 5. It is also of concern [23] that the values of dH_{C2}/dT determined by ac susceptibility and resistivity are significantly lower than those measured by dc magnetization [11, 12] and specific heat transitions [13].

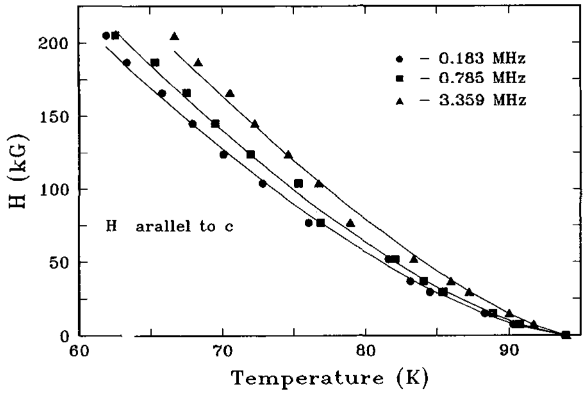

Fig. 4. – Temperature dependence of the critical field determined by ac susceptibility at three different frequencies for the field oriented parallel to the c axis.

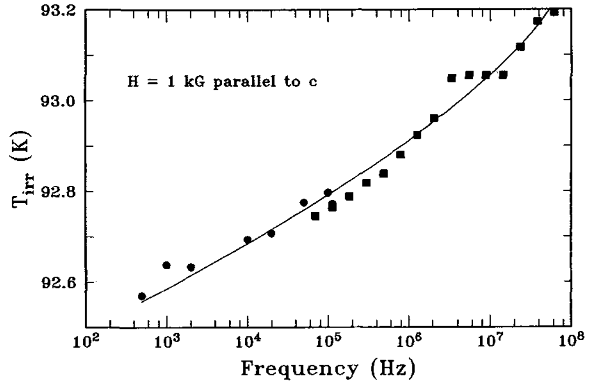

Fig. 5. – The frequency dependance of the transition temperature of a $Y_1Ba_2Cu_3O_{7-x}$ single crystal determined by ac susceptibility for an applied field of 1 kG along the c axis. The solid line is a qualitative fit to equation (4) ignoring the $\sqrt{1-t}$ term inside the logarithm.

To understand these effects it is important to understand exactly what is being measured in this technique. As the sample is cooled through the superconducting transition, the flux becomes quantized and a small diamagnetic (Meissner) response, linear in $(T_C - T)$ is expected. Athyreya *et al.* [12] have reported a $(T_C - T)^2$ behavior for the reversible magnetization near T_C which is not understood; in any case the magnitude of the reversible magnetization is below the resolution of our instrumentation. At some lower temperature, which we choose to call T_{irr}, when the superconductive state supports sufficient critical current to shield the interior of the crystal from the ac field, a large change in inductance is observed. In addition the ac field induces viscous losses due to motion of the flux line cores until the surface critical current shields the flux lines from the ac field. This, we believe, produces the sharp peak in the series resistance. The maximum in the losses, and the maximum rate of change in the inductive component is expected to occur when ac field, δh, just penetrates to the center of the sample. In a simple critical state model, this implies that $J_C = c\delta h / 2\pi D$ where aivc is the speed of light and D is the sample diameter. This analysis implies that the H_{C2} *vs.* T curves are really irreversibility lines measured at constant J_C and therefore the true H_{C2} line lies somewhere above these lines.

We analyse this irreversibility line in terms of thermally activated flux flow first proposed by Anderson and Kim [22]. The thermal activation of vortices from pinning sites of energy U, reduces the critical current, J_C, from its value in the absence of thermal fluctuation J_{C0}, making it frequency, as well as temperature and field, dependent. This effect is particularly pronounced because of the low pinning energies and the high transition temperatures in these new materials. Our previous treatments of this effect [19, 23] neglected an important point, noted by Dew-Hughes [24], that because of the unprecedentedly high T_C of these new materials, both backward as well as forward hopping of vortices must be considered.

The net hopping frequency of vortices in the presence of a driving force due to the current is [24]

$$f = 2f_0 e^{-U/kT} \sinh\left(\frac{\Delta W}{kT}\right). \qquad (2)$$

Where ΔW is the work done moving the flux lines against the driving force and f_0 is the escape attempt frequency of the vortex, typically of order 10^{12} Hz. Near T_C, $\Delta W \le kT$ so that we can approximate $\sinh(x) \sim x$ and get

$$f = 2f_0 \frac{\Delta W}{kT} e^{-U/kT}. \qquad (3)$$

At fields where there are many more flux lines than pinning sites, we can write $\Delta W = J_C U / J_{C0}$ [24]. To estimate the size of this effect near T_C we associate the creep rate f, with the frequency of the measurement since if the flux moves in response to the ac field we will not observe a transition. We choose the Anderson-Kim estimate for the pinning energy $U = p\left(H_C^2 / 8\pi\right) V$. Here p is a numerical factor which describes the fraction of the condensation energy available as pinning energy, H_C is the thermodynamic critical field, and V the activation volume. near T_C, we assume the Ginzburg-Landau form $H_C = 1.73\ H_C(0)(1-t)$ where $H_C(0)$

is the critical field at zero temperature and $t = T / T_C$ is the reduced temperature. We estimate the volume as $1.3\, a_0^2 \xi_z$ where $a_0 = 1.07\,(\Phi_0 / B)^{1/2}$ is the flux line spacing and ξ_z is the coherence length along the field direction, the c axis in these experiments. Using the standard clean limit formula for the divergence of ξ_z near T_C we get

$$(1-t)^{3/2} = \frac{8\pi k T_C B}{3.3 P H_C(0)^2 \Phi_0 \xi_z(0)} \times \ln\left(\frac{2.3 \Phi_0^{3/2} \xi_z(0) f_0 J}{B^{1/2} (1-t)^{1/2} c f k T_C}\right). \quad (4)$$

From our earlier discussion, we estimate the current density necessary to shield a 1 Oe field to be ~ 50 A/cm^2 and using $\xi_z = 4$ Å and $H_C = 10^4$ Oe this reduces to

$$(1-t)^{3/2} = \frac{1.2 \times 10^{-7} B}{p} \ln\left(\frac{3.5 \times 10^{-3} f_0}{B^{1/2} (1-t)^{1/2} f}\right). \quad (5)$$

We can compare this prediction to our data for the reversibility line as a function of field and frequency. Using the data in figure 4 at 183 kHz, a fit to $(1-t)^n = aB$ yields, $n = 1.51$ and $a = 1.1 \times 10^{-6}$ Oe^{-1}. The value inside the logarithm is dominated by f / f_0. To compare with the data in figure 4 at 183 kHz, we estimate the value of the logarithmic term to be about 7 (we have ignored the field and temperature dependence inside the logarithm). This results in an effective value for p of about 1 which agrees well that implies by the low temperature estimate for the pinning energy for magnetic relaxation data [14]. This means that the pinning energy is approximately equal to the condensation energy times the activation volume.

Figure 5 shows the transition temperature, which we call T_{irr}, the irreversibility temperature, as a function of frequency. The four data points near 5 MHz are affected by a resonance in the cable-coil circuit which drastically changes the current in this frequency range. Equation (4) predicts an approximately linear dependence on ln (f) at low frequency with an increased dependance dut to the 2/3 power at higher frequency. The line in figure 5 is a qualitative fit to equation (4) with $f_0 = 1.5 \times 10^{10}$ and $p \sim 3$. Again we have ignored th $\sqrt{1-t}$ term inside the logarithm which has only a very small effect.

There are many alternate models for flux creep. We have chosen this particular model because of its simple form and because the resulting temperature and field dependence near T_C agrees with our experimental data. There are other models that may be applicable. In particular, we explore the possibility of a distribution of pinning energies in a related paper [25].

Conclusions

The analysis sketched above has important implications for experiments that attempt to measure H_{C2}. Techniques that rely on the existence of a finite critical current, such as ac susceptibility and resistivity transitions do not in fact measure H_{C2} but rather an irreversibility transition measured at constant critical current. What do these results imply for the true H_{C2}'s and the coherence lengths deduced from them? One cannot simply extrapolate the irreversibility to higher frequencies to obtain H_{C2}, although the true H_{C2} lies above the irreversibility line and so the coherence length is smaller than previously reported [6, 7]. Reversible magnetization measurements are needed and have been reported by Fang *et al.* [11] but show a puzzling downward curvature (the opposite tendency to that of Fig. 4). This has been attributed to the effects of twin boundaries, but more work needs to be done before definitive coherence lengths can be extracted.

The expression we have chosen for the pinning energy is general and not dependent on the mechanism responsible for the pinning, and therefore does not provide any information on the mechanism. One clue, however, comes from the fact that the low temperature critical current in high quality single crystals [1, 2] is approximately the same as in aligned films [26] where there are many more possible mechanisms. This suggests that the mechanism might be intrinsic to the material. Several possible pinning mechanisms have been suggested including twin planes [27] and randomly distributed oxygen vacancies that must occur because of the non-integral number of oxygens per unit cell [28]. However, independent of the pinning mechanism, the low value of the deduced pinning energy U will set an important limitation on attempts to increase critical current densities at high temperatures in these superconductors.

Acknowledgements

The high field data was taken while one of us (TKW) was a Guest Scientist at the Francis Bitter National Magnet Laboratory, which is supported at M.I.T. by the National Science Foundation.

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