PHYSICAL REVIEW B VOLUME 53, NUMBER 17 1 MAY 1996-I

High-field and low-temperature magnetization and flux creep in untwinned $YBa_2Cu_3O_{7-x}$

B. Janossy,* L. Nguyen, and P. Wyder
The Grenoble High Magnetic Field Laboratory, CNRS-MPI, Boîte Postale 166, 38042 Grenoble Cedex 9, France

(Received 11 October 1995)

We report on the magnetization and flux creep in untwinned $YBa_2Cu_3O_{7-x}$ in a function of field up to 20 T and temperature between 0.4 and 4.2 K. We observe a field-dependent magnetization down to 0.4 K, exhibiting a hump at intermediate fields. The normalized relaxation rate decreases with field before going through a minimum at intermediate fields. Extrapolation to zero temperature indicates the existence of a nonzero relaxation at all magnetic fields. We compare our data to relaxation measurements on an organic superconductor. The data analyses within the framework of the collective pinning theory fail. We suggest the existence of an additional pinning mechanism in the sample.

INTRODUCTION

It is by now well established that in high-temperature superconductors (HTSC's) relaxation of the magnetization persists down to the lowest temperatures at low fields. This phenomenon has been attributed to quantum tunneling of vortices.[1] Relaxation measurements at high fields and very low temperatures are rare.[2] Seidler *et al.*'s report[3] is the only one claiming to observe the vanishing of the relaxation of the magnetization above a temperature-dependent threshold field.

The collective pinning theory[4,5] (CPT) has revived interest with the advent of HTSC's where small point defects are thought to cause pinning. The temperature dependence of the activation energy for flux creep and that of the critical current can be understood within this theory. However, the field dependence of the above two quantities is much less studied. In what follows we present the field dependence of the critical current J_c and the normalized relaxation rate s of untwinned Y-Ba-Cu-O below 4.2 K. The field dependence of these quantities is compared to those measured in the organic superconductor κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br ($\equiv$ET/(NCN)Br).[6] Although the general features of the relaxation rate are similar in both compounds, the behavior of the critical current is totally different. This strongly contradicts the predictions of the collective pinning theory where s and J_c are linked. Furthermore, the field dependence of the relaxation rate cannot be interpreted within the collective pinning theory assuming only point-defect pinning. The critical current and relaxation rate show similar field dependences as that observed at higher temperatures.[7]

EXPERIMENTAL DETAILS

Data were collected on a small ($0.5\times0.5\times0.1$ mm^3) untwinned $YBa_2Cu_3O_{7-x}$ with a sharp transition at $T=92$ K. The sample was mounted onto a high-sensitivity torque magnetometer placed into a ^{3}He cryostat. Three temperatures were selected for the investigations: 4.2, 1.3, and 0.4 K where heat regulation was not necessary. The sample was submerged into liquid ^{3}He, allowing for good thermal contact. The sample was oriented with its c axis being at ~10° away from the field direction. Measurements were taken in a 23-T resistive magnet. The field sweeps in all measurements were set to 330 Oe/s, which induced negligible heating of the sample. The magnetization and relaxation curves were taken in the same run. Torque was recorded during the field sweep. Relaxation measurements started immediately after the field stopped. Approximately 900 points were taken during the 600 s acquisition time, and then the field sweep was restarted. The steps between two relaxation experiments were larger than 1 T in order to induce sufficient change in the magnetic field to rebuild the steady state. As shown in Fig. 1, the relaxation is logarithmic and 10 min recording is sufficient to determine the normalized relaxation rate $s=M_0^{-1}dM/d\ln t$. The magnet being a resistive coil we did not have to worry about the current decay. There was no observable overshoot. The inconvenience of the resistive magnets is the fluctuation of the magnetic field. The visible fluctuations are ~10 Oe, but larger variations are possible at very short time scales. These fluctuations may be responsible for the jumps observed in some measurements.

Hysteresis curves of the magnetization for the three temperatures are shown in Fig. 2. Figure 2 is obtained from the raw torque after dividing it with the magnetic field ($M=\Gamma/H$). This procedure is not completely justified as

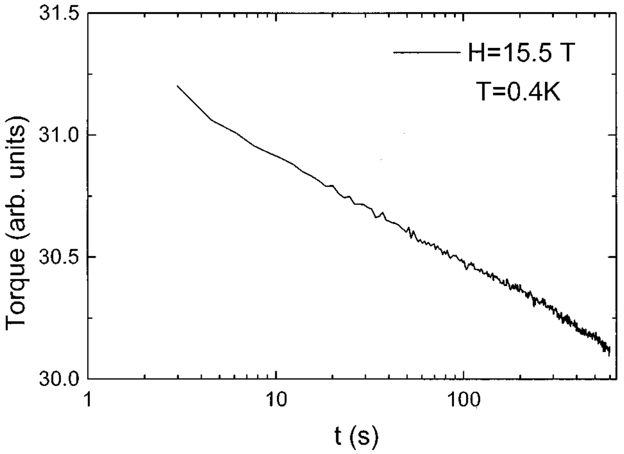

FIG. 1. Typical relaxation of the torque at $T=0.4$ K and $H=15.5$ T.

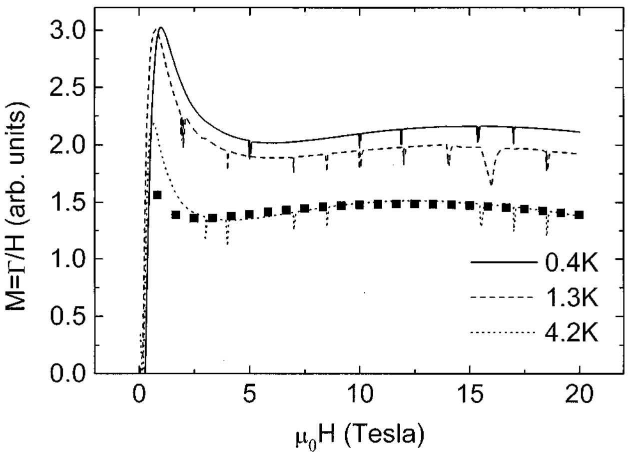

FIG. 2. Upward field sweep for 0.4, 1.3, and 4.2 K. The plot is obtained by dividing the torque with the field. At these temperatures the reversible magnetization is negligible and thus the magnetization is proportional to the critical current. Squares are a fit to Eq. (3) with $\alpha=0.15$, $\beta=3.35$, and $\mu_0 H_{\mathrm{irr}}=47$ T.

there is a non-negligible contribution to the torque coming from the magnetization along the *ab* planes. In the presence of twin planes, the torque is indeed not identical with the magnetization.[8] But in the absence of linear and planar defects, we argue that the scaling procedure proposed by Blatter *et al.*[9] is valid. The magnetizations perpendicular and parallel to the planes are linked via the anisotropy parameter and the torque is proportional to the magnetization. The jumps in the curves correspond to the restart of the magnet. After ~600 s waiting time, the magnetization is reduced due to creep. The steady state is again achieved after a change in the field of ~0.1 T. Note that the magnetization is directly proportional to the critical current within the limits of the Bean model,[10] that is, for field changes larger than the Bean penetration field H^*. In our sample $\mu_0 H^* \approx 0.4$ T was estimated from the field change necessary to reach the steady-state magnetization after reversing the field.

RESULTS

The overall features of the hysteresis curves are well known. The magnetization rapidly decays with fields up to 3–5 T and is field independent above.[11] The low-field magnetization is attributed to self-field effects due to curvature in the flux lines.[12] We observe a continuous increase of the magnetization (≡critical current) with decreasing temperature, showing that thermal activation is present at least down to 1.3 K. This is in agreement with results in the literature for Y-Ba-Cu-O where the crossover to the quantum limit that is the saturation of the magnetization occurs at ~1 K.[3,13] Figure 3 is an enlargement of the 4.2-K magnetization curve. Here we distinguish the features of the high-temperature hysteresis curves, but with characteristic fields shifted to higher values. A minimum occurs at $\mu_0 H_{\mathrm{min}}=4$ T followed by a fishtail with a maximum at $\mu_0 H_{\mathrm{max}}=12.5$ T. The same features are observed on the two other curves with H_{min} and H_{max} shifted to higher fields with decreasing temperature. In the interpretation given in Ref. 7, the minimum corresponds to a transition from single-vortex pinning to the collective-creep regime. The critical current inferred from magnetization data is affected by the decay of the magnetization during the field sweep and during the data acquisition. At the phase boundary, creep slows down, which results in an increased apparent current density. Within this interpretation, the increase in the critical current (or magnetization) with field corresponds to a dip in the relaxation rate.

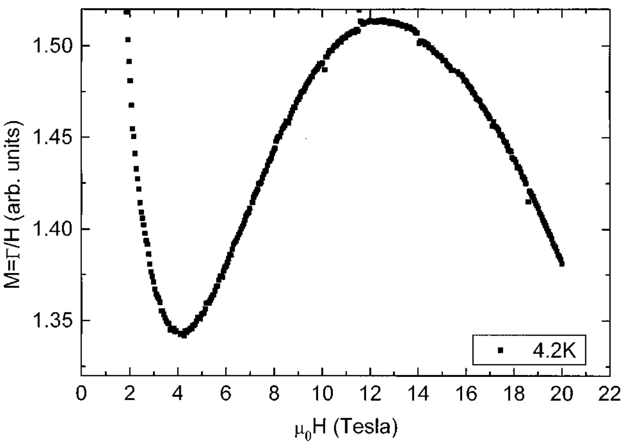

FIG. 3. Enlarged view of the hysteresis curve at 4.2 K.

The normalized relaxation rate s is shown in Fig. 4. M_0 is the value obtained from the hysteresis curve. For all three temperatures s decreases rapidly with the applied field and reaches a minimum at 8–10 T. At 4.2 K, where thermal activation is still important, the relaxation rate increases with field after reaching its minimum value. As T decreases, s tends to saturate with field at $s \approx 1.2\%$.

DISCUSSION

The collective pinning theory predicts the following field dependences of the critical current and the relaxation rate. At low fields in the single-vortex pinning limit, both J_c and the quantum creep are expected to be field independent. Above the crossover field H_{SV} in the small bundle pinning regime, both quantities are expected to decrease exponentially.[14] These predictions are not verified by our measurements. In

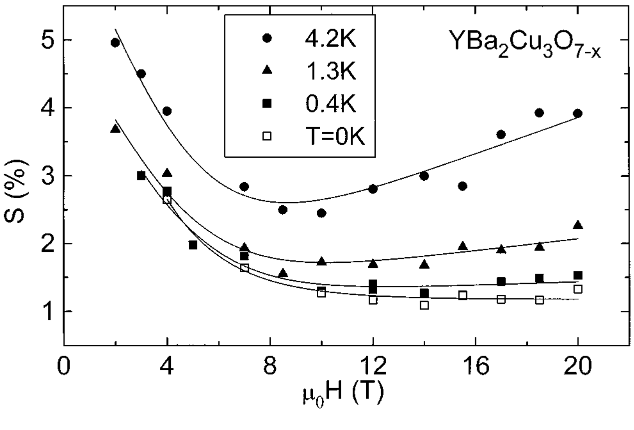

FIG. 4. Field dependence of the normalized relaxation rate s in $YBa_2Cu_3O_{7-x}$ at different temperatures. The lines are fits to Eq. (1). The parameters are given in the text.

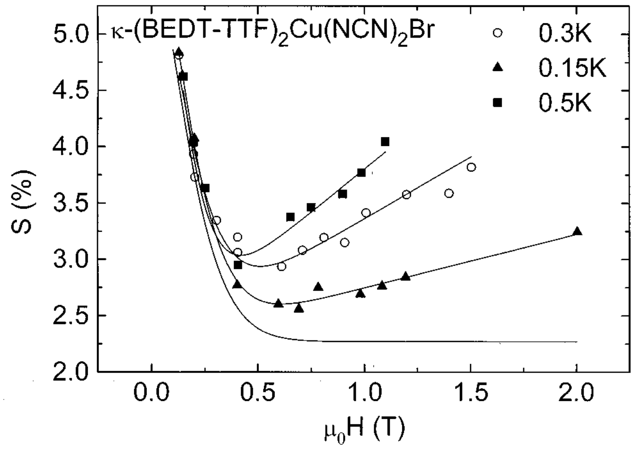

FIG. 5. Relaxation rate of ET/(NCN)Br from Ref. 6. The lines are fits to Eq. (1). The parameters are given in the text.

Fig. 4, s extrapolated to zero temperature becomes field independent only above 10 T, while below that field it strongly decreases. This result contradicts the collective pinning theory from which one expects the opposite behavior. A simple application of the CPT is again impossible when we compare our data to the organic superconductor ET/(NCN)Br. As shown in Ref. 6 in ET/(NCN)Br, the critical current decreases as $H^{-1/2}$, while the relaxation rate has the same qualitative features as s in Y-Ba-Cu-O (see Figs. 4 and 5). The minimum of the relaxation is reached in the organic compound at $\mu_0 H \approx 0.5$ T, which is roughly 20 times smaller than in Y-Ba-Cu-O. Within the CPT it is impossible to have the same $s(H)$ for different $J_c(H)$ because s is proportional to $J_c^{-1/2}$.

We can get an empirical field dependence by noticing that the low-field behavior is exponential while in the limit of zero temperature s tends towards a constant value. As thermal activation becomes important, the relaxation rate, instead of saturating, increases with field. This behavior can be described by a linear field dependence. The empirical law is then given by

$$s = A(T)\exp[-(H/H_0)^{\mu}] + B(T)H + s_0, \tag{1}$$

where $A(T)$ and $B(T)$ are temperature-dependent constants. We stress that Eq. (1) is purely empirical and may be approximate, but it contains an essential feature: It shows that the relaxation rate is the product of different types of pinning. Below we set some of the parameters of Eq. (1) to correspond to existing pinning mechanisms.

Here μ is set to $\mu = 3/2$ so that the first term of Eq. (1) corresponds to the relaxation rate for small vortex bundles. The second term is chosen such that it describes thermal activation. The relaxation rate is then $s = k_B T/U(H,T)$ where $U(H,T)$ is an effective potential. At low temperatures U depends only upon the field H. Further, we suppose that $1/U(H)$ can be expanded in the function of H. This is the case for strong pinning centers where the pinning energy decreases with increasing field. The first term of the expansion gives a relaxation rate proportional to the field. Thus for the second term of Eq. (1) we end up with $B(T)H \approx B_0 TH$, with B_0 being field and temperature independent. Finally s_0 can be interpreted as the contribution of quantum relaxation. Equation (1) then describes relaxation in the presence of different types of pinning centers.

The data in Figs. 4 and 5 are fitted to Eq. (1). We have four free parameters $[A(T),\ H_0,\ B_0$, and $s_0]$. s_0 is extracted from the $T=0$ limit, and H_0 is kept the same for the different temperatures. In this way we obtain, in the case of Y-Ba-Cu-O, $s_0 = 1.19\%$, $\mu_0 H_0 = 4.17$ T, and $B_0 = 0.031\%$ K^{-1}. A decreases with decreasing temperature and approaches $A(T \rightarrow 0) = 3.4\%$. For the organic superconductor, $s_0 = 2.27\%$, $\mu_0 H_0 = 0.345$ T, $A = 0.22\%$ (it is temperature independent), and $B_0 = 3\%$ K^{-1}. These parameters can be set independently and the accuracy is within 5%. The good quality of the fit suggests that as the field is swept different types of barriers dominate the dynamics of flux creep.

The critical current is inferred from the hysteresis loop using the Bean model, which supposes a uniform current density inside the sample. It has been known for a long time that this model is insufficient when surface pinning or strong pinning is present.[15] In HTSC's surface pinning proved to be important and we argue that bulk pinning may be better described by strong pinning than collective pinning at low temperatures. The large number of point defects necessary to obtain high-energy barriers observed in the CPT description is absent in HTSC's, which have typical mean free paths of 100–700 Å. The fact that the low-temperature relaxation rate in all layered superconductors lies in the range of 1–5 % is again contrary to CPT predictions.[1,6,16] The description of pinning can be refined by supposing the presence of additional defects such as boundaries or strong pinning centers. In this case, the Bean model is not necessarily valid as the local field distribution is strongly inhomogenous.

To interpret Figs. 2 and 3, we suppose that the measured magnetic moment is the sum of the contribution of surface currents and bulk pinning due to strong pinning centers. This model can account qualitatively for the observed magnetization curves. In Ref. 15 the pinning force for various types of defects has been calculated. It is shown that the general field dependence of the critical current is

$$J_c = C b^m (1-b)^n, \tag{2}$$

where C is a temperature-dependent constant and m and n depend on the type of pinning. For Bean-Livingstons-type barriers, one finds $m = -1/2$ and $n = 1$ and for core interaction $m = 1/2$ and $n = 1$. In the presence of two distinct pinning mechanisms separated in space, it is fair to assume that the measured magnetic moment is the sum of the two contributions proportionally weighted with their respective volume,

$$m = \alpha[b^{-0.5}(1-b)] + \beta[b^{+0.5}(1-b)]. \tag{3}$$

α and β are the weighting factors, and $b = H/H_{\mathrm{irr}}$ is the field normalized with the irreversibility field. We plotted in Fig. 6 a series of curves for different values α and β. These curves can describe qualitatively the field dependence of the critical current. Taking a ratio of 1:20—the bulk pinning being the dominant term—we reproduce the dip observed at fields around 5 T and the decrease of J_c above 14 T. Equation (3) can be quite well fitted to the high-field range where we get $H_{\mathrm{irr}} = 47$–52 T for the different temperatures. The H_{irr} value

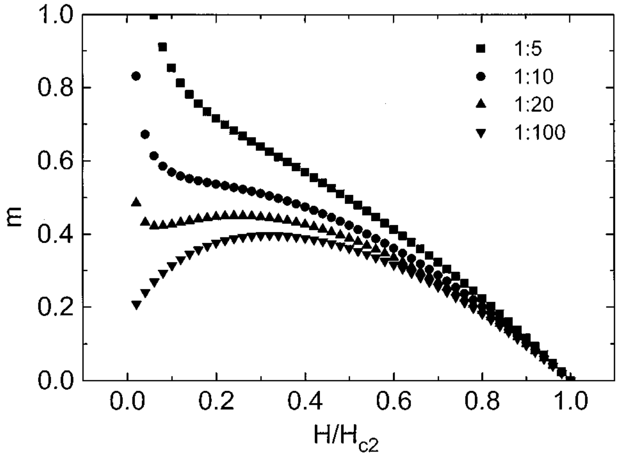

FIG. 6. Magnetic moment obtained with the use of Eq. (3) for different surface-to-bulk pinning ratios. The amount of bulk pinning is kept constant ($\beta=1$) while reducing the contribution of surface pinning.

is consistent with extrapolations from higher temperatures.[7] Within this analysis the dominant pinning mechanism is that of the bulk.

The relaxation measurements can be understood as well within this picture. With two characteristic relaxation rates s_1 and s_2 corresponding, respectively, to surface barriers and bulk pinning, the deduced relaxation rate is

$$s=\frac{\alpha s_1+\beta s_2}{\alpha+\beta}. \tag{4}$$

The increase in s could be due to the increase of s_2, while the low-field s would be dominated by the surface barriers. In the case of strong pinning, as the field is increased, the number of pinning centers available decreases and thus the barrier energy decreases (s_2 increases). The initial decrease of s could be explained if the relaxation over surface barriers was much stronger than the bulk relaxation. In this case, the decrease of the surface barrier contribution to the magnetization would lead to a decrease in the apparent relaxation rate. Equating Eq. (4) to Eq. (1), we get $s_1 \propto \exp[-(H/H_0)^{\mu}]$ and $s_2 \propto B_0 TH + s_0$.

CONCLUSION

We have studied the field dependence of the magnetization and the relaxation rate at low temperatures in $YBa_2Cu_3O_{7-x}$. We observe a peak effect near 12–15 T in the magnetization and a complex field dependence in the relaxation rate. The collective pinning theory does not describe our data. We suggest that the magnetic moment observed is the result of more than one type of pinning mechanism. Tentatively, we suppose that the combination of strong pinning and pinning by surface barriers allows for a qualitative description of both magnetization and relaxation data. Further studies are necessary in order to separate contributions to the relaxation of the magnetization due to quantum tunneling and due to thermal excitations.

ACKNOWLEDGMENTS

B.J. is indebted to D. Feinberg for enlightening discussions and to L. Fruchter for a critical reading of the manuscript.

---

*Present address: Laboratoire des Champs Magnétiques Pulsés, INSA, Complexe Scientifique de Rangueil, 31077 Toulouse Cedex, France (author to whom correspondence should be addressed).

[1] L. Fruchter, A. P. Malozemoff, I. A. Campbell, J. Sanchez, M. Konczykowski, R. Griessen, and F. Holzberg, Phys. Rev. B **43**, 8709 (1991).

[2] S. Moehlecke and Y. Kopelevich, Physica C **222**, 149 (1994).

[3] G. T. Seidler, C. S. Carillo, T. F. Rosenbaum, U. Welp, G. W. Crabtree, and V. M. Vinokur, Phys. Rev. Lett. **70**, 2814 (1993).

[4] A. Larkin and Y. Ovchinikov, J. Low Temp. Phys. **34**, 409 (1979).

[5] G. Blatter, M. V. Feigel'man, V. B. Geshkenbein, A. I. Larkin, and V. M. Vinokur, Rev. Mod. Phys. **66**, 1125 (1994).

[6] D. Prost, C. Lenoir, P. Batail, I. A. Campbell, and L. Fruchter, Phys. Rev. B **49**, 4023 (1994).

[7] L. Krusin-Elbaum, L. Civale, V. M. Vinokur, and F. Holtzberg, Phys. Rev. Lett. **69**, 2280 (1992).

[8] Twin planes give a peak in the angular-dependent torque along the *c* axis, while no such anomaly exists in conventional magnetization. See, e.g., E. M. Gyorgy *et al.*, Appl. Phys. Lett. **56**, 2465 (1990).

[9] G. Blatter, V. B. Geshkenbein, and A. I. Larkin, Phys. Rev. Lett. **68**, 875 (1992).

[10] C. P. Bean, Phys. Rev. Lett. **8**, 250 (1962).

[11] The increase of M at low fields in Fig. 2 is not relevant because this only indicates that the sample has not reached the critical state.

[12] L. W. Conner and A. P. Malozemoff, Phys. Rev. B **43**, 402 (1991).

[13] S. Uji, H. Aoki, S. Takebayashi, M. Tanaka, and M. Hashimoto, Physica C **207**, 112 (1993).

[14] V. V. Vinokur, G. Blatter, M. V. Feigelman, and V. B. Geshkebein, Physica C **185-189**, 276 (1991).

[15] A. M. Campbell and J. E. Evetts, Adv. Phys. **21**, 199 (1972).

[16] R. Griessen, J. G. Lensink, T. Scroder, and B. Dam, Cryogenics **30**, 563 (1990); S. Uji, H. Aoki, S. Takebajashi, M. Takana, and M. Hashimoto, Physica C **207**, 112 (1993); D. Prost, L. Fruchter, I. A. Campbell, N. Motohira, and M. Konczykowski, Phys. Rev. B **47**, 3457 (1993).