Asymmetry of sweep rate and magnetic relaxation effects in YBCO thin films

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Abstract

We report magnetic field sweep rate and magnetic relaxation effects in YBCO thin films (thickness \( t < 450 \) nm) at various temperatures. It is observed that for the \( H \parallel c \) configuration and at relatively high temperatures \((T > 73 \) K) there is a pronounced asymmetry to both the field sweep rate and relaxation effects. Both the sweep rate and relaxation effects are much more pronounced for increasing fields as compared to decreasing fields. The sweep rate asymmetry is more evident for slow sweep rates and higher fields, consistent with increased relaxation and lowered activation barriers for increasing fields. On the other hand, low temperature measurements down to 35 K do not reveal any asymmetry in either effect. The dependence on temperature and orientation leads us to conclude that the asymmetry originates in the unique characteristics of the critical state for very thin films \((t \ll \lambda\), where \( \lambda \) is the penetration depth) in perpendicular applied fields and the effects of the field line curvature therein. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

The magnetic behavior of high-\( T_c \) superconductors has revealed many unique features, particularly in relation to magnetic relaxation. The lowered values of the pinning energy and the consequent rapid and large magnetization changes during the course of relaxation measurements are well understood features \([1–3]\). Some further aspects, such as, e.g. the effects of surface and geometrical barriers \([4–8]\) can lead to an asymmetry in the relaxation rates between increasing and decreasing fields. Experimental and theoretical studies on the role of surface barriers in relaxation have been reported \([8–10a,b]\). In particular, Brandt \([8]\) has reviewed the role of various types of real and apparent surface and edge barriers on flux dynamics and the asymmetry effects associated with them. Burlachkov \([9]\), for longitudinal geometry, has shown that a Bean–Livingstone type surface barrier can also result in an asymmetry of the relaxation rates with the rate for flux entry being much larger than for exit. Weir et al. \([11]\) observed pronounced asymmetry effects in YBCO only at low temperatures with, however, the rate of entry being slower than for exit. It is to be noted that asymmetry of relaxation was also discussed in the early work of Beasley et al. \([12]\). This asymmetry effect, which was calculated and measured within the context of longitudinal geometry and bulk pinning, was seen to be very small. It has also been discussed \([9]\) that in
the presence of both bulk and surface pinning it is the weaker of the two mechanisms which determines the rate of relaxation, and a relaxation rate crossover has been reported [10a,b,11]. The analysis of the critical state response in very thin films \((t \ll \lambda)\) for the perpendicular geometry \([5,7]\), \((H \parallel c\text{-axis})\) has shown however that pronounced asymmetry effects can develop in the magnetic relaxation without invoking the concept of surface barriers. This asymmetry was shown to develop as a consequence of current patterns on field reversal extending over much of the thin film and driving it into a subcritical state with \(J < J_c\). This is to be compared to the case for the bulk materials where the sub-critical state only extends to the depth to which the field change has penetrated. Experiments [13] showed an asymmetry of the rates of relaxation consistent with the work of Zeldov et al. [5] when a transport current is made to flow through a sample in the remnant state. Thus while the main idea of a critical state balance defined by \(J = J_c\) with stationary vortices continues to retain its usefulness even in perpendicular thin films with \(t \ll \lambda\), the new features which emerge can be related to the role of field line curvature and its associated currents. The main source of driving currents in the perpendicular case derives from the field line curvature. It is also appreciated that magnetic relaxation for thin films in the \(H \parallel c\text{-axis}\) configuration is susceptible to a high degree to any slight overshoot of the field and can lead to erroneous conclusions about asymmetry. Schnack et al. [1] have given examples of such behavior in simulation studies. We note further that while there have been some studies of the asymmetry of relaxation effects, there has been no study of the closely related effect, viz. the magnetic field sweep rate dependence of the magnetization in very thin films in perpendicular geometry. Since both these effects, viz. time and sweep rate dependence of magnetization, are in general dependant on the activation barriers [1,14,15], we may well expect that the asymmetry of the barriers would be manifest in the sweep rate dependence as well. If so, it would be a more convenient method of measuring asymmetry effects in thin films due to the aforementioned problems in relaxation measurements. We have thus conducted both sweep rate and relaxation experiments in thin films in perpendicular orientation at various temperatures, to identify and relate any asymmetry in these effects and to discuss their origin.

2. Experiment

The experiments were conducted on thin films of the \(Y_{1-x}Ba_xCu_3O_y\) composition prepared at Technical University Munich by laser ablation on yttria and yttria-stabilized zirconia substrates. The films were annealed to achieve an optimization of the oxygen content. All the films used were verified to be single phase, as evident by the sharpness of the superconducting transition, and for the orientation of the \(c\text{-axis}\). The \(c\text{-axis}\) lay normal to the plane of the films. The film thicknesses of the two samples \(F_1\) and \(F_2\) reported in this work were 450 and 300 nm, respectively. Initial studies were conducted on films of area \(10 \times 9 \text{ mm}^2\) while later studies were repeated for samples which had been reduced to \(5 \times 6 \text{ mm}^2\). No qualitative differences were observed in the magnetic response of the big and the small samples. Most of the data being reported is thus for the small size samples that corresponded better with the optimum sizes for the magnetometers. The critical temperature of the sample \(F_1\) was 89.5 K while that of \(F_2\) was 87 K. All the magnetic measurements were performed using commercial Vibrating Sample Magnetometers and electromagnets at Quaid-I-Azam University (QAU) and at the Ghulam Ishaque Khan Institute of Technology (GIK).

2.1. Sweep rate effects

In Fig. 1 we present the data for the magnetization at 77 K for the sample \(F_1\) at various sweep rates ranging from 16.66 to 169.8 Oe/s, for both the increasing and decreasing field branches. The sample was oriented parallel to the \(c\text{-axis}\) \((\theta = 0)\). All the data being reported is for the zero-field-cooled condition and was repeated several times for several sizes and both thicknesses. All the data on the sweep rate effect were fully reproducible over several months of experimentation. The (applied) field for full penetration of the sample \(H^*\) at 77 K was determined by careful magnetization rotation tests reported elsewhere [16] and was obtained as 38 and
Fig. 1. Magnetization $M(H)$ loops for sample F1 at $T = 77$ K, at different field sweep rates, i.e. 16.9, 33.8, 67.6 and 169 Oe/s for the field parallel to the $c$-axis ($\theta = 0$). Loops are displaced outwards successively with sweep rates. Arrows indicate direction of field sweep. Large sweep rate asymmetry is evident.

25 Oe, respectively, for the two films. These small values of the applied fields may of course correspond to much larger effective fields due to the strong demagnetization factor effects [17a,b] at low fields. Thus all the measurements to be discussed are in a field range $H > 500$ Oe, much higher than the penetration fields for these samples. Referring now to Fig. 1 we note that there is a very pronounced asymmetry of the sweep rate effect between the increasing and decreasing field branches. While the increasing field branch shows a pronounced effect, with the magnetization increasing with the field sweep rate as is generally seen [18a,b,c], there is very little effect for the decreasing field branch for both the samples shown. The same pattern is repeated in the negative side of the hysteresis loops (not shown). We carefully tested the sample holders to eliminate the possibility of any unusual background effects and find no background contributions. It is also apparent from the hysteresis loops that their shapes correspond to the usual bulk pinning dominated magnetization and not that of surface pinning. For surface pinning dominated loops one expects the magnetization in decreasing fields to be close to zero. This is evidently not the case for our samples. What is noticeable, particularly for slow sweep rates is that the absolute value of magnetization in the decreasing field branch is larger. This is suggestive of more rapid flux penetration (increasing $H$) than of expulsion (decreasing $H$). We also studied the sweep rate dependence at angles other than the $c$-axis. In these measurements the sample was arranged to be tilted with respect to the applied field. In the data of Fig. 2 the sweep rate dependence of $M$ at $\theta = 60^\circ$ is shown. It is apparent on comparison with Fig. 1 that there is still an asymmetry of the sweep rate effect but it is significantly less pronounced. The differences in the sweep rate dependence for the two orientations are quantified later and confirm that the sweep asymmetry is much less for $H$ deviating far from the $c$-axis.

The sweep rate dependence of the magnetization ($H \parallel c$) for the increasing and decreasing field branches is represented for the applied fields of 1 and 2 kOe on a log–log plot in Fig. 3. The smaller slopes (d ln $M$/d ln $H$) for the field decreasing...
Fig. 2. Magnetization $M(H)$ loops at different field sweep rates as in Fig. 1, for applied field making an angle $\theta = 60^\circ$ with the $c$-axis.

branch are evident. Detailed analysis of sweep rate dependence of $M$ is in general model dependent in terms of the dependence of the activation barrier for flux motion, $U$, on the current density $J$, i.e. $U(J)$. It

Fig. 3. Variation of $\ln M$ vs. $\ln H$ for sample F at two different dc fields for field increasing and field decreasing branches. $\Box$ represents the applied field of 1 kOe and $\triangle$ represents the applied field of 2 kOe. Filled symbols are for the field increasing branch and hollow symbols are for the field decreasing branch ($T = 77$ K, $H \parallel c$).
has however been shown [1,14,15] that for such frequently encountered situations in high-$T_c$ materials, as, e.g. linear, power law or logarithmic $U(J)$ dependence, the sweep rate dependence of $M$ (i.e. $\frac{d \ln M}{d \ln H}$) yields the ratio of thermal activation energies, $\varepsilon(J) = KT/U(J)$. In the absence of any similar model to quantify the sweep rate effects in ultra thin films we shall use the value of the logarithmic derivative $C = (d \ln M/d \ln H)$ as an indicator of the parameter $\varepsilon(J) = KT/U(J)$. This interpretation of $C$ will be seen to be consistent with the magnetic relaxation effects we observe, as well as the discussion in Section 3. Taken in this spirit, the asymmetry of $C$ between the two branches reflects the asymmetry of the barriers $U(J)$ for flux entry and exit. The higher the value of the logarithmic derivative $C$, the lower the value of the activation barriers, for a given field and temperature. At $T = 77 \text{ K}$, for the increasing field branch, the value of $C$ for the fields 1, 2 and 3 kOe are found from the data of Fig. 3 to be equal to 0.08, 0.15 and 0.26, respectively. The corresponding values for the decreasing branch are 0.029, 0.0166 and 0.0169, respectively. The higher values of $C$ in the field increasing branch clearly suggest a lower value of the barriers for flux entry. This asymmetry, we shall see later, is also indicated by the slower rates of relaxation in the field decreasing branch. The sweep rate constant $C$, for the sample F2 for the fields of 1, 2 and 3 kOe for the increasing field branch was found to be 0.09, 0.15 and 0.26, respectively. For the decreasing field branch these values were determined to be 0.023, 0.032 and 0.02, respectively. In general we find, as the above values for $C$ reflect, that the asymmetry of the sweep rate effects becomes more pronounced for higher fields.

The sweep rate dependence of magnetization for the loops obtained at $\theta = 60^\circ$ shows, as mentioned earlier, a significant decrease in asymmetry. For a field of 1 kOe the sweep rate constant $C$ was obtained as 0.06 and 0.04, while for $H = 2$ kOe the values are 0.17 and 0.037, for increasing and decreasing field branches, respectively. By comparing the ratios of sweep rate constants for the increasing ($C_{\text{inc}}$) and decreasing ($C_{\text{dec}}$) field branches, i.e. ($C_{\text{inc}}/C_{\text{dec}}$), for the two orientations, one can see the effect very clearly. For the orientation ($\theta = 60^\circ$) we obtain ($C_{\text{inc}}/C_{\text{dec}}$), as 1.5 and 4.59 for applied fields of 1 kOe and 2 kOe, respectively. On the other hand, for $H \parallel c$-axis, ($C_{\text{inc}}/C_{\text{dec}}$) for the same fields is 2.76 and 9.37, respectively. Similar behavior is observed at other fields. The smaller values of the ratios for $\theta = 60^\circ$ suggests that the sweep rate effect asymmetry is reduced for the field orientations away from the $c$-axis.

### 2.2. Relaxation measurements

We conducted magnetic relaxation measurements for both the samples in the same, i.e. zero-field-cooled condition after once cycling the field and bringing the sample fully into the critical state. The measurements on the same samples were made at two different laboratories QAU and GIK, with all the $T < 77 \text{ K}$ work being at GIK. The magnetization was measured at every 1 s interval after raising the field up to the desired value at a fixed rate (typically 66 Oe/s) and allowing the field to stabilize. The data at long times was seen to be highly sensitive to even the smallest field drift and hence only the short-time data ($t < 150 \text{ s}$) is being reported. For these times the field was perfectly constant to within our QAU resolution of about 2 Oe. To ensure reproducibility the data were obtained several times under the same conditions. The data for both branches are shown in the inset of Fig. 4 as also are the fit to the usual logarithmic form [2].

$$M = M_0[1 - S \ln(1 + t/\tau)].$$

where $M_0$, $S$ and $\tau$ are left as fit parameters. The differences in the relaxation effects are noticeable. From this data, for an applied field of 1 kOe, we obtain the rate constant $S = 0.08$ for the increasing field branch and $S = 0.04$ for the decreasing field branch. The value of the time constant $\tau$ was determined to be in the range 0.1–0.5 s. Thus the asymmetry of the relaxation rate for the increasing and decreasing branches is quite apparent. As also suggested by the sweep rate measurements, the smaller values of $S$ in the decreasing field branch show that flux creep rate in the decreasing field branch is much slower than for the field increasing branch.

Similar data on the sample F1 at 73 K were also obtained at the GIK magnetometer where the field...
stability was ensured for longer periods. The data are shown in Fig. 4. The data shown initiate 30 s after the field hold has begun, which was a typical field stabilization time for this instrument. The variation was still very clearly logarithmic over the entire time span. From this data, taken up to a time of 600 s, we extract the relaxation rate \( S \) at 1 kOe to be 0.104 for the field increasing branch and 0.0170 for the decreasing field branch. Thus for \( T = 73 \) K and a longer time period, the values of \( S \) and their asymmetry are larger but consistent with those obtained from the short time data of 77 K. We also note that there is no change of slope of the relaxation curve over this time span \( (t < 600 \text{ s}) \). Hence this longer time data, shows no effects which can be related to the crossover of flux creep time scales, as noted for example by Weir et al. [11]. This indicates that whatever the pinning mechanism which determines the flux creep process, it continues to be the dominant process at least up to the time of 600 s at a temperature of 73 K. We note that the values of the relaxation rate constant for the increasing field branch are on the relatively high side compared to typical values for the YBCO compounds, for these temperatures. This suggests that the unusual asymmetry of relaxation at these temperatures may be related to the rapid relaxation.

2.3. Low-temperature measurements

In our low-temperature measurements we attempted to identify the relation of surface pinning effects, if any, to the asymmetry. It is expected that if there are both bulk and surface pinning mechanisms then the relaxation would be dominated by the faster of the two [9]. The weaker pinning mechanism would, in other words, determine the rate of relax-
Fig. 5. Magnetization $M(H)$ loops for sample F at $T = 35$ K, at different field sweep rates, i.e. 16.66, 66.6 and 169 Oe/s for the field parallel to c-axis ($\theta = 0$). Outer loops are faster loops. No asymmetry is evident.

ation. With decreasing temperatures bulk pinning increases while surface pinning remains the same [11]. Hence if surface pinning is responsible for the asymmetry, one should observe a greater degree of asymmetry in the relaxation.

The low-$T$ studies were carried out on the same piece of the sample F used in the above discussed studies. Data were taken in the range $35$ K $< T < 75$ K at GIK. The field sweep rate dependence was measured for fields up to 10 kOe as described earlier. Clear asymmetry of the rate dependence with large values of $C$ for increasing field branch could be seen at $T = 75$ K, while the asymmetry was negligible at 50 K and 35 K (Fig. 5). The loops retain the bulk pinning dominated shape. The value of the sweep rate constant for $T = 50$ K range between $C = 0.041$ and 0.054 as the field $H$ changes from 1 to 4 kOe for both the increasing and decreasing branches. Similarly for $T = 35$ K the values of $C$ for the two branches range from 0.033 to 0.049. The differences between the increasing and decreasing branches are within the experimental error.

Magnetic relaxation measurements were conducted at temperatures of 65 and 50 K for an applied field of 1 kOe with the same procedure as discussed for the 73 K data. At $T = 50$ K the relaxation rate constant $S$ has a value equal to 0.024 for the increasing field branch and 0.025 for the decreasing field branch. Similarly for $T = 65$ K, the corresponding values of $S$ are 0.018 and 0.019. The values of the relaxation rate constant are thus very close to each other for both these temperatures. The relaxation rate apparently goes down very significantly between 73 and 65 K for the increasing field branch. The rates are slightly less at 50 K where, still, there is no asymmetry. Thus the asymmetry both in $C$ and $S$ appears only for $T$ close to $T_c$.

3. Discussion and conclusions

Initially we summarize our main observations. We observe clearly a faster creep rate as well as enhanced field sweep rate effects in the increasing field branch for higher temperatures, $T > 73$ K. However for low temperatures no asymmetry is present in either effect. It is also clear that the sweep rate asymmetry is definitely more pronounced for the field applied along the c-axis. We also find no indications of a crossover in relaxation rates in our time window and the hysteresis loops are clearly bulk pinning dominated. The data are consistent with the notion of effectively larger activation barriers for flux exit than for entry. The discussion in the Section
2 has identified that a surface barrier effect of the type identified in Ref. [9] is not consistent with many of our observations. Basically, if surface barrier mechanisms were to be present then the asymmetry effects should have been further enhanced at low temperatures. As discussed in the preceding section, this is expected since the manifest increase in the bulk pinning at low temperatures would lead to the creep being dominated by the weaker, surface pinning [11]. The observation that the asymmetry effects are negligible at low temperatures suggests that surface barrier effects are not the source of the asymmetry. Furthermore, and in the same context, the shape of our hysteresis loops do not correspond to those expected for a system where the surface pinning is dominant. Equally important, it is understood [8] that the various types of real edge and surface barriers effective in the transverse geometry lead to a delay in the penetration of flux but not the exit. Thus the asymmetry with such barriers create would lead to a slower rate of flux entry as compared to the rate of expulsion. This is similar to what Weir et al. [11] have observed. We obviously find the converse of this, viz. that the rates of flux entry are more rapid as compared to those for exit.

We therefore refer to the analysis of critical state behavior in very thin films for perpendicular geometry [5,7] to find an explanation of our observations. We do not have conclusive evidence to support the critical state analysis of thin films in perpendicular geometry [5,7], for such a test would have to be based on measurements of local field and current density profiles. However we believe that most of our observations relating to the asymmetry of the observed effects find a natural explanation within this model. The main features of these analyses are that for thin films with thickness \( t \ll \lambda \), and for perpendicular applied fields, the longitudinal Bean state due to the flux density gradient is in general absent and the driving force derives from the curvature of the field lines, rather than from the flux density gradient. The largeness of the curvature terms is a consequence of the large value of the derivatives of the form \( dH_y/dx \) where \( H_y \) is the longitudinal component of the fields (coming from the shielding current profiles) and \( x \) is the direction transverse to the film plane. In this situation reversal of the field or current change direction (e.g. increasing to decreasing) in the film has the effect of suppressing the current density below the value of \( J_c \) almost everywhere. In the Bean model applied to thick samples, in contrast, the magnitude of the current equals \( J_c \) everywhere within. As pointed out by these authors, [5,7] this effect, viz. suppression of the current density, is expected to drastically slow down the relaxation rate on field reversal. These features can be seen in the critical state field and current density profiles from Zeldov et al. [5] and Brandt and Indebom [7]. Fig. 6 illustrates the respective profiles for increasing fields \( H_y \) of different magnitudes, while Fig. 7 illustrates the same for decreasing fields, (from Ref. [7]). The fields and currents in the figures are scaled as \( H_y/H_x \) and \( J/J_c \) where \( H_x \) corresponds to the characteristic field \( H_x = J_c d \), \( J_c \) the critical current density and \( d \) the film thickness. Comparing the two figures it is clear that for the increasing fields (Fig. 6) and \( H_y/H_x \geq 1 \) the current density \( J \) in the film is equal to 1 over much of the sample. For the decreasing fields (Fig. 7), on the other hand, as the field \( H_y \) decreases (e.g. from 2 to 1 in the figure), the current density falls below \( J_c \) over almost the entire sample. As emphasized in Refs. [5,7] this drives the thin film into a subcritical state \( J < J_c \) on field reversal with consequently very little relaxation. The asymmetry of relaxation

![Fig. 6. Current density and magnetic field profiles in a superconducting strip of width 2a in a perpendicular magnetic field \( H_y \) which increased from zero (virgin state). The depicted profiles are for \( H_y/H_x = 0.5, 1, 1.5, 2, 2.5 \) (from Ref. [7]).](image-url)
between field increasing and decreasing branches is thus a manifest feature of the critical state behavior of thin films in perpendicular geometry. As the figures illustrate, in the above geometry the driving currents in the sample can assume a value less than $J_c$ even with the assumption of a Bean-like, constant critical current density $J_c$. While the sweep rate effect for thin films is not analyzed in these models we think that it follows the same dynamics, i.e. flux entry during the field increasing branches is rapid while flux expulsion during field decreasing branch is slow. Thus for example in the field decreasing branch, the slowing down of the sweep rate leads to no significant increase in the flux expulsion and, concomitantly, to any decrease in the values of $M$.

We further note that the reduced asymmetry of the sweep rate effect which we observe at large angles is consistent with the above discussion. For fields applied at an angle to the $c$-axis, as, e.g. in the data in Fig. 2, there is a large component parallel to the surface. Consequently the longitudinal Bean state and the effects of the flux density gradient are no longer negligible, and the creep effects in both branches should be the same, as is observed in parallel geometry. Finally, we believe that the pronounced asymmetry effects at higher temperatures only, and stronger effects at higher fields are both due to the weakening of pinning in these conditions, viz. with higher temperature and fields. The lowered bulk pinning at higher temperatures and fields leads to pronounced relaxation effects in general. Thus any asymmetry of relaxation related effects would also be more readily observable. Finally, theoretical work in the direction of obtaining an explicit sweep rate dependence to the magnetization of thin films in perpendicular geometry would be very useful for further experimental work in this direction.

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