Magnetization relaxation in YBCO films with improved supercurrent transport properties

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Abstract. The relaxation of the irreversible magnetization in optimally doped YBCO films with natural and artificial pinning centres was measured in zero-field cooling conditions using SQUID magnetometry. The external magnetic field $H$ was oriented along the $c$ axis. An appropriate method for the determination of the characteristic vortex pinning energy from the normalized vortex-creep activation energy is discussed. This is based on the existence of a crossover elastic (collective) vortex creep at low temperatures $T$ — plastic vortex creep at high $T$, caused by the $T$ dependent macroscopic currents induced in the sample during magnetization measurements.

1. Introduction

YBa2Cu3O7−δ (YBCO) still represents one of the best choices for large scale applications of high-temperature superconductors (HTS), such as the fabrication of superconducting coated conductors [1], and the increase of vortex pinning through artificial routes is a field of intense activity [2-7]. It was found that BaZrO3 inclusions in YBCO thin films, for example, significantly enhance vortex pinning for the external magnetic field $H$ parallel to the $c$ axis [2]. The pinning behavior in HTS is widely investigated using standard magnetization relaxation measurements, with a fixed relaxation time window.

As known [8], the vortex pinning energy barrier would easily be extracted from the relaxation of the irreversible magnetization $M(t)$, where $t$ is the relaxation time, if the current density $J$ dependence of the vortex-creep activation energy $U$ would be linear [9], $U(J) = U_0(1 - J/J_C)$, where $U_0$ is the
pinning barrier in static conditions (no current) and \( J_c \) is the critical current density. The above linear \( U(J) \) is equivalent with \( M \) decreasing logarithmically in time, \( M(t) = M(t_0)\left(1 - S_0 \ln\left(t/t_0\right)\right) \), where \( S_0 = T/U_0 \) is the magnetization relaxation rate in the linear model and \( t_0 \) is the time scale for creep \([10]\). This leads to the determination of \( U_0 = -TM(t_0)\Delta \ln(t)/\Delta M \), where \( t_0 \) is substituted by a plausible low value and \( M(t_0) \) is extracted by extrapolation. However, the \( U(J) \) relation is usually nonlinear \([11]\), and in order to reduce the intrinsic ambiguity of flux creep experiments a normalized vortex-creep activation energy \( U^*(J) = -Td\ln(t)/d\ln\left(\left|M\right|\right) \) was considered \([8]\). When the magnetization relaxation is weak, one determines a normalized activation energy averaged over the relaxation time window, \( U^* = -T\Delta \ln(t)/\Delta \ln\left(\left|M\right|\right) \), and this quantity was expected to supply information about the effective pinning energy barrier.

Here we analyze magnetization relaxation data obtained for YBCO films with improved supercurrent transport properties. A crossover elastic (collective) creep at low \( T \) (high \( J \)) – plastic creep at high \( T \) (low \( J \)) was observed, generated by the \( T \) dependent macroscopic currents induced in the sample during standard magnetization measurements. This crossover manifests itself through a maximum in the \( T(J) \) variation of \( U^* \), and allows an accurate determination of the characteristic vortex pinning energy.

2. Experimental details
The investigated specimens were rectangular (~2.1×2.3 mm\(^2\)) optimally doped YBCO films prepared by pulsed laser deposition (PLD) on (100) oriented MgO substrates with SrTiO\(_3\) (STO) buffers, using a Lambda Physik KrF excimer laser (\( \lambda = 248 \text{ nm} \)) at an energy of 340 mJ/pulse and an oxygen partial pressure of 200 mTorr. A STO target was ablated to form the STO buffer layer. Prior to the STO deposition the MgO substrates were annealed in air at 700 °C for 30 min. The deposition of 120 nm thick STO buffers was conducted at a substrate temperature of 730 °C. As a reference, pure YBCO films were grown on STO buffers at 800 °C.

To form \( \text{Y}_2\text{O}_3 \) (YO) doped and \( \text{BaZrO}_3 \) (BZO) doped YBCO films, a “surface modified method” was used \([3]\). The same YBCO target was modified: a thin (0.5 mm thick) and narrow (area of about 2 ÷ 2.5 % of the surface area of the YBCO target) YO (or YSZ) sector was cut from YO (or YSZ) single crystal, respectively, and stuck on the YBCO target by silver paint. With these surface-modified targets, doped YBCO films were grown using the same procedure as that for the reference sample. Afterwards all the films were cooled to room temperature within an hour in 500 Torr of oxygen.

Below we refer to experimental results obtained for a “pure” ~310 nm thick film (YBCO), a ~294 nm thick film with YO inclusions (YBCO YO), and for a ~317 nm thick film containing BZO nanorods, resulting from the reaction between ZrO\(_2\) and YBCO while fabricating the film \([3]\), and preferentially oriented along the \( c \) axis (YBCO BZO). In YBCO YO round-shaped YO inclusions of a mean diameter of 6~8 nm were randomly distributed (with a mean spacing of about 12 nm) and dispersed throughout the entire film thickness. In YBCO BZO the defect diameter was estimated to be on the order of 5 nm, whereas the mean spacing is of ~30 nm, leading to a matching field \( B_s \approx 1.9 \text{ T} \).

The dc magnetization measurements were performed using a commercial Quantum Design Magnetic Property Measurement System, with \( H \) always applied in zero-field cooling conditions and oriented along the \( c \) axis. For magnetization relaxation measurements the superconducting magnet was in persistent mode. The relaxation time \( t \) was considered to be zero when magnet charging was finished, and the first data point was registered at \( t_1 \approx 100 \text{ s} \).

The inductively determined critical temperature \( T_c \) is ~90.3 K for YBCO, and decreases to ~88.5 K in the case of YBCO BZO, as illustrated in the inset to figure 1, where \( m \) is the measured magnetic moment. The critical current density \( J_c \) (affected by thermally activated vortex motion) was extracted from the difference \( \Delta m \) between the \( m \) values obtained for increasing and decreasing \( H \). When pinning is strong, \( \Delta m \approx 2m \), and the measured \( m \) can be identified with the irreversible magnetic moment.
Figure 1. Main panel: Magnetic field $H$ variation of the critical current density $J_c$ (affected by thermally activated vortex motion) determined with equation (1) for the YBCO film and for the film with BZO nanorods (YBCO BZO) at $T = 20$ K. The nanorod inclusion leads to the increase of $J_c$, while the critical temperature $T_c$ decreases to ~88.5 K, as illustrated in the inset, where $m$ is the magnetic moment measured on warming in $H = 10$ Oe.

For our rectangular samples we used a Bean relation [12,13] for practical units,

$$J_c = \frac{40|m|}{dLl^2(1-1/3L)},$$  (1)

where $L$ is the sample length, $l$ - the width, and $d$ - the thickness. The above relation supplies $J_c$ in A/cm$^2$ if $m$ is in emu and all dimensions are in cm. As shown in the main panel of figure 1, $J_c$ of YBCO BZO is approximately two times higher than that of YBCO at $T = 20$ K. For the same conditions, $J_c$ of YBCO YO lies in between.

3. Results and discussion

Figure 2 illustrates, as an example, the irreversible volume magnetization $M$ vs. ln($t$) for YBCO BZO ($H = 10$ kOe). Due to a relatively small overall magnetization relaxation in the relaxation time window $t_w$, the plots are almost linear.

Figure 2. Characteristic magnetization relaxation curves, $|M|$ vs. ln($t$), for the YBCO BZO film ($H = 10$ kOe).

Figure 3. Temperature variation of the magnetization relaxation rate $S_0$ and of the corresponding pinning barrier $U_0 = T/S_0$ determined with the linear $U(J)$ model.

Figure 3. Temperature variation of the magnetization relaxation rate $S_0$ and of the corresponding pinning barrier $U_0 = T/S_0$ determined with the linear $U(J)$ model.
Since the plots in figure 2 are practically linear, it is tempting to consider a linear \( U(J) \) and to determine the averaged magnetization relaxation rate \( S_0 = -[\Delta M/\Delta M(t_0)]/\Delta t \) and the corresponding barrier \( U_0 = T/S_0 \). Figure 3 shows the \( T \) dependence of \( S_0 \) and \( U_0 \) extracted from the relaxation curves from figure 2, by taking \( t_0 = 10^{-3} \text{ s} \). As can be seen, \( U_0 \) decreases with decreasing \( T \) in the low-\( T \) range (where the \( T \) variation of the superconductor parameters is weak), which indicates that \( U_0 \) does not represent the true pinning barrier. This behavior appears for all plausible \( t_0 \) values, and can easily be explained if a nonlinear \( U(J) \) is considered [11]. At the same time, \( S_0(T) \) shows a maximum at low \( T \).

In the case of a nonlinear \( U(J) \), the use of the normalized vortex-creep activation energy \( U^*(J) \) was proposed, related, as shown above, to the inverse of the local slope of the magnetization relaxation curves in double logarithmic scales. Figure 4 illustrates the relaxation curves from figure 2 in a double logarithmic plot, where we added some other data at higher temperatures. Again, due to a relatively small overall magnetization relaxation in the conditions of strong pinning and to the usually limited range, allowing the determination of the averaged \( U^* = -T\Delta \ln(\ln(|M|)/\Delta t) \).

Figure 5 shows the resulting \( T \) variation of \( U^* \) for YBCO BZO at several field values below and above \( B_c \). The notable feature is the appearance of a maximum in \( U^*(T) \) at a crossover temperature \( T_{cr} \), which decreases with increasing \( H \). YBCO and YBCO YO behave similarly. The fact that \( U^* \) decreases with decreasing \( T \) in the low-\( T \) range suggests that \( U^* \) does not generally represent a measure of the pinning energy barrier. The \( U^*(T) \) maximum has been reported for HTS with weak quenched disorder, such as \( \text{Tl}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_8 + \delta \) thin films [14], \( \text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_2\text{O}_8 + \delta \) single crystals [15], and high-quality YBCO films with a smaller \( J_c \) obtained by high-pressure dc sputtering [16], as well as for YBCO films obtained by PLD, where strong vortex pinning on growth induced linear defects is present [17]. The difference is that in the latter case \( T_{cr} \) is significantly higher.

At the same time, the low-\( T \) shoulder in \( U^*(T) \) appearing in figure 5 for \( H \) up to \(~40\) kOe is absent in films with weaker pinning [16]. The existence of this shoulder leads to a peculiar increase of \( U^* \) with increasing \( H \) for \( T \sim 15–40 \) K. The anomalous \( U^*(H) \) increase was observed by us for YBCO, as well. However, in this case the effect is maximum around 20 K, and is approximately two times smaller than that observed for YBCO BZO.

![Figure 4](image-url) Figure 4. Magnetization relaxation curves from figure 2 in double logarithmic scales, where we added the data at higher temperatures, \( T = 70 \) K and 75 K. Due to the relatively small overall magnetization relaxation, the plots are practically linear.

![Figure 5](image-url) Figure 5. Temperature variation of the normalized vortex-creep activation energy \( U^* \) resulting from the data plotted in figure 4. \( U^*(T) \) exhibits a maximum at a crossover temperature \( T_{cr} \) (indicated by an arrow), which shifts to a lower \( T \) value with increasing \( H \).
The $U^*(T)$ maximum has been interpreted through a crossover elastic (collective) vortex creep at low $T$ – plastic vortex creep at high $T$ [14,15]. The meaning of $U^*$ can be obtained by considering the actual vortex-creep activation energy $U(J)$, which can be expressed [18] as

$$U(J) = (U_c/p)((J_c/J)^p - 1),$$  \hspace{1cm} (2)

where $U_c$ is a characteristic pinning energy, $J_c$ is the critical current density for the ideal creep-free case, whereas the exponent $p$ is identified with the (positive) collective pinning exponent $\mu$ of the order unity in the case of elastic (collective) vortex creep [19], and $p < 0$ for plastic creep. The value $p = -1$ corresponds to the classic Kim-Anderson model [9], whereas a value $p = -0.5$ often appears [16,20].

With equation (2) and considering $J \propto |M|$ as explicit variable, one can derive $U^*(J) = -Td \ln(|M|)/d \ln(t)$ using the general creep relation [21] $U = T \ln(t/t_0)$. Assuming a small change of $t_0$, for the elastic-creep domain one obtains

$$U^*(J) = U_c(J_c/J)^\mu.$$  \hspace{1cm} (3)

In the plastic creep regime $U^*$ has an opposite variation with $J$,

$$U^*(J) = U_c(J_c/J)^p.$$  \hspace{1cm} (4)

Since for collective creep the pinning centers do not accommodate vortices, the characteristic pinning energy for elastic creep $U_{cc}$ can be much lower than the characteristic pinning energy for plastic creep $U_{cp}$.

With a fixed $t_w$ and taking $T$ as explicit variable, for $T$ well below $T_c$ the above equations lead to

$$U^*(T) \approx U_{cc} + \mu T \ln(t_w/t_0)$$  \hspace{1cm} (5)

for elastic creep, and

$$U^*(T) \approx U_{cp} - |p| T \ln(t_w/t_0)$$  \hspace{1cm} (6)

in the case of plastic creep. [For simplicity, we have considered the same $t_0$ for elastic and plastic creep.] Thus, the maximum in $U^*(T)$ from figure 5 indicates a creep crossover at $T_{cr}$. In the approximation $t_0 = \text{constant}$ and $U_{cp} \gg U_{cc}$, equations (5) and (6) lead to

$$T_{cr} \approx \frac{U_{cp}}{[(\mu + |p|)\ln(t_w/t_0)]},$$  \hspace{1cm} (7)

where $\mu (p)$ is the creep exponent determined for $T$ significantly below (above) $T_{cr}$, respectively. Relation (7) describes the shift of the $U^*(T)$ maximum to lower $T$ values with increasing $H$, through the $U_{cp}(H)$ decrease.

It is clear from equations (2), (3) and (4) that $U^*$ does not represent a measure of the effective pinning energy barrier, which is $U$. However, the change of the creep exponent across the $U^*(T)$ maximum suggests a method to compare the pinning efficiency of various HTS. Simply, when the creep exponent changes sign the curvature of the $M(t)$ curves in the representation from figure 4 changes sign, which means that at $T = T_{cr}$ the relaxation curve in double logarithmic scales should be a straight line. In this situation, $U(J) = U_c \ln(J_c/J)$, and $U^* = U_c$. Thus, $U^*$ represents a measure of characteristic pinning energy at $T = T_{cr}$ only.
Because below ~60 K. The deviation at high temperature and measurements is to consider the energy balance relation \(26\) for dynamic conditions \(27\) (finite temperature). For HTS, with or without correlated disorder \(17,31-35\). It is worthy to note that efficiency means a high \(Tc\) or strong (like in thin films \(29\)). At high \(H\), \(J\) is much lower than the true \(Jc\), and \(E_p\) becomes higher than \(E_{el}\). In these conditions dislocations proliferate, the pinning centers accommodate vortices, and one expects vortex creep in a plastically pinned vortex assembly at low drives. In a crude approximation, one can consider \(E_p = E_{el}\) at \(T = T_{cr}\) and \(E_p \propto U\). Using the general vortex-creep relation and \(E_{el} \propto H^{-1/2}\), for a fixed \(T_{cr}\) it results \(T_{cr} \propto H^{1/2}\). As can be seen in the main panel of figure 6, where we plotted \(T_{cr}\) (resulting from the \(U^*(H)\) data from figure 5) vs. \(H\), the above relation is fulfilled below ~60 K. The deviation at high \(T\) reflects a stronger influence of thermal fluctuations, and, because \(J\) is low, a crossover toward Bose-glass behavior \(30\) is possible, as indicated by the fact that below \(B_g\) the \(U^*(T)\) maximum at high \(T\) is sharper.

Since for HTS with reduced pinning the \(U^*(T)\) maximum at high \(H\) appears at \(T_{cr} \ll T_c\), it was concluded \(15\) that it is generated by the \(T\) dependent macroscopic currents induced in the specimen, controlling the effective pinning energy. Indeed, plotted vs. \(J\) [extracted from \(m(t), T\)], for example, with equation \(1\), \(U^*\) shows a maximum. Figure 7 illustrates \(U^*\) vs. \(1/J\) for YBCO BZO in a field \(H = 49\) kOe, for which \(T_{cr} \approx 48\) K (see figure 5). The slope of \(U^*(1/J)\) in a double logarithmic plot supplies the creep exponent. As can be seen, in the collective creep regime at high \(J\) (low \(T\)) the obtained \(\mu\) exponent is in the range delivered by the theory \(19\), whereas in the plastic creep regime at low \(J\) (high \(T\)) the resulting \(p\) is close to the value ~0.5 proposed in \(20\).

Finally, we discuss the origin of the shoulder in \(U^*(T)\) from figure 5, which appears to be specific to strongly disordered HTS, with or without correlated disorder \(17,31-35\). It is worthy to note that this is located deeply in the elastic creep regime, where vortex localization on the correlated disorder is weak, and it disappears well above \(B_g\).
Figure 7. The normalized vortex-creep activation energy $U^*$ vs. $1/J$ for YBCO BZO at $H = 49$ kOe. The current density $J$ was obtained from $|m(t_1, T)|$ with equation (1), for $T$ between 10 K and 70 K (step of 5 K). The slope of the $U^*(1/J)$ dependence in double logarithmic scales (dotted lines) supplies the creep exponent, positive in the elastic creep regime, and negative for plastic creep [see equations (3) and (4)].

In figure 8 we plotted $U^*$ vs. $1/J$ for $H = 40$ kOe and 10 kOe, where $J$ was obtained from the relaxation data sets for $T$ between 2 K and 60 K, using equation (1) with $|m|$ mediated over the relaxation time window. The arrows indicate $T = 20$ K. The mean collective creep exponent $\mu$ values resulting from the linear fit (continuous lines) are discussed in terms of pure collective creep above 20 K.

The first observation is the occurrence of unreasonably high “$\mu$-values” (of the order of 10) for $T$ below ~10 K. In our opinion [16], this is caused by the presence of micro flux jumps [36]. Here we note a simple way to detect the presence of thermo-magnetic instabilities. After the last point on a low-$T$ relaxation data set was taken (at $H = 10$ kOe, for example), $H$ was increased to 12 kOe (with the option “no overshoot”, always used in our relaxation experiments), and $m$ was measured again. The $H$ sweeping rate was of ~20 Oe/s, and the above field change is highly sufficient (as checked) to inverse the Bean profile, due to the strong demagnetization effects in thin film specimens. It was found that for $T < 20$ K the moment $m(t_1, 10$ kOe) measured after decreasing $H$ is significantly lower than $|m(t_1, 10$ kOe)| corresponding to the first point of the relaxation data set, signaling thermo-magnetic instabilities. At $T = 5$ K the difference of the absolute values represents ~25 %, and becomes almost negligible (~3 %) at 20 K. The use of pure thermally activated flux creep regime over $t_0$ becomes inappropriate below ~20 K ($H = 10$ kOe). While the thermo-magnetic instabilities are expected to occur mainly between $t_0$ and $t_1$ (where $J$ is closer to $J_c$), when $T$ is low enough ($T \leq 10$ K) micro flux jumps are present in $t_n$, as well. This can explain the strong depression of $U^*$ with decreasing $T$. 

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In figure 8 we plotted $U^*$ vs. $1/J$ for YBCO BZO at $H = 40$ kOe [where the $U^*(T)$ shoulder almost disappears, see figure 5], and $H = 10$ kOe, where a pronounced shoulder occurs for $T \sim 25-30$ K, leading to maximum in the $T$ variation of the normalized magnetization relaxation rate $S = T/U^*$ [31-35]. Here $J$ was obtained from the relaxation data set, using equation (1) with $|m|$ mediated over $t_n$. For $T \ll T_c$ the main effect of the thermal energy is to change the probed $J$ interval, and the slope of the $U^*(1/J)$ dependence in double logarithmic scales is $\mu$, as noted above.
The above discussion about the creep crossover at $T_{cr}$ assures that for YBCO BZO, for example, pure thermally activated vortex creep can be considered in the whole investigated $H$ range at $T \geq 20$ K. [The $H$ variation of the $T$ value below which the micro flux jumps become important necessitates further investigation. In the case of our thin films, it decreases with $H$.] However, the increasing role of the pinning strength distribution on approaching $T_{cr}$ leads to a supplemental restriction of the $T$ interval, to $T << T_{cr}$. It can be seen in figure 8 that for $H = 40$ kOe the collective creep exponent $\mu$ does not change too much just above the $T$ range of thermo-magnetic instabilities. As predicted by equation (5), $U^*$ is linear in $T$ (see figure 5), and $U_{ce}$ is small. The situation is quite different for $H = 10$ kOe, where $\mu$ increases continuously roughly from 0.3 to 1 with increasing $T$ from 20 K to 40 K. For a quantitative analysis, we compare the mean $\mu$ values in the interval from 20 K to 30 K for the two $H$ values. For $H = 40$ kOe the obtained $\mu$ is two times higher than for $H = 10$ kOe. At the same time, the linear fit of $U^*(T)$ from figure 5 with equation (5) in the same $T$ range supplies $U_{ce}(10$ kOe)~200 K, which is approximately four times higher than $U_{ce}(40$ kOe). The reduction of $U_{ce}$ with increasing $H$ was also reported in [17]. With the above $U_{ce}$ and averaged $\mu$ values equation (5) can account for the increase of $U^*(25$ K) by $\sim 40$ % when $H$ increases from 10 kOe to 40 kOe, as can be seen in figure 5.

For the two fields (10 kOe and 40 kOe) the determined $\mu$ values in the case of YBCO BZO for $T$ between 20 K and 40 K lie in the interval $\sim 0.3$–1.2. This is in the range of the $\mu(J/J_c)$ variation from the original collective pinning theory [19]. The theory predicts $\mu = 1/7$ for $J$ very close to $J_c$, and, with increasing $T$ (decreasing $J$), $\mu$ should increase toward 3/2. As discussed above, in the case of standard magnetization relaxation measurements an important point is what is happening in the time interval between $t_0$ and $t_1$, determining the $J/J_c$ interval probed in $t_w$. A relatively large $U_{ce}$ at $H = 10$ kOe means a reduced overall relaxation from $t_0$ and $t_1$. Consequently, for a given $T$ just above the low-$T$ domain where thermo-magnetic instabilities set in, the $J$ interval probed over $t_w$ is not far from $J_c$, where $\mu$ is low, leading to a reduced $U^*$ [see equation (5)]. At a higher $T$ the probed $J/J_c$ is lower, $\mu$ increases, and $U^*$ is higher. Due to a smaller $U_{ce}$ at $H = 40$ kOe, the overall relaxation between $t_0$ and $t_1$ is more pronounced for the same $T$ range. In these conditions, over $t_w$ one probes the lower $J/J_c$ (higher $\mu$) domain only, and, consequently, the $U^*(T)$ shoulder disappears.

4. Conclusions

In summary, the analysis of magnetization relaxation in YBCO films with improved supercurrent transport properties reveals a crossover elastic creep at low $T$ – plastic creep at high $T$ generated by the $T$ dependent macroscopic currents induced in the sample during magnetization measurements. This crossover appears to be a general behavior of HTS, with or without correlated disorder. The crossover temperature increases with increasing pinning, and a method to compare the efficiency of various pinning systems using standard magnetization relaxation data is proposed. It is also shown that the anomalous $T$ variation of the normalized vortex-creep activation energy at low $T$ is related to the variation of the classic collective creep exponent in the dynamic conditions specific to standard magnetization relaxation measurements and to the occurrence of micro flux jumps in the low-$T$ limit.

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