# Evidence for Mean Free Path Fluctuation Induced Pinning in $YBa_2Cu_3O_7$ and $YBa_2Cu_4O_8$ Films

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The critical current $j_c$ and the pinning energy $U_c$ have been determined for three types of yttrium-based superconducting films from current $j_s$ and dynamic relaxation $Q = d\ln j_s/d\ln(dB/dt)$ data by means of the generalized inversion scheme. For $B < 2$ T and $T < 80$ K the temperature dependence of $j_c$ and $U_c$ for all films is found to be in excellent agreement with a model of single vortices pinned by randomly distributed weak pinning centers via spatial fluctuations of the charge carrier mean free path. Pinning due to spatial fluctuations of $T_c$ is not observed.

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In type II superconductors pinning results from spatial variations of the Ginzburg-Landau (GL) coefficient $\alpha$ associated with disorder in the transition temperature $T_c$ and/or from spatial variations in the charge carrier mean free path $l$ near lattice defects [1-5]. These two sorts of pinning are usually called $\delta T_c$ pinning and $\delta l$ pinning [6]. In $\delta T_c$ pinning spatial variations of $T_c$ lead to spatial modulations of the linear and quadratic terms of the GL free energy functional $\alpha|\Psi|^2+(\beta/2)|\Psi|^4$ while in $\delta l$ pinning variations of the mean free path only affect the $|\nabla\Psi|^2$ term through variation of the two components $m$ and $M$ of the effective mass tensor.

For example, for a single vortex along the $c$ axis of a superconductor containing randomly distributed weak pinning centers Blatter *et al.* [1] showed that all quantities characterizing the collective pinning of a single vortex can be expressed in terms of a disorder parameter $\delta$, i.e.,

$$L_c = \xi\delta^{-1/3}\varepsilon^{4/3},$$
$$U_c = \mu_0^{-1}B_c^2\xi^3\delta^{1/3}\varepsilon^{2/3}, \qquad (1)$$
$$j_c = j_0\delta^{2/3}\varepsilon^{-2/3},$$

where $\xi$ is the GL coherence length, $B_c$ the thermodynamic critical field ($B_c = \Phi_0/2\sqrt{2}\pi\lambda\xi$ with $\Phi_0 = h/2e$ as flux quantum), $j_0 = 4B_c/3\sqrt{6}\mu_0\lambda$ the depairing current, $j_c$ the true critical current, and $\varepsilon$ is the mass anisotropy ratio $\varepsilon^2 = m/M < 1$. The collective pinning length $L_c$, the critical current $j_c$, and the collective pinning energy $U_c$ are related via $U_c = j_c\Phi_0L_c\xi$. In the absence of disorder (and, consequently, of pinning) the relations in Eq. (1) lead to $U_c = 0$, $j_c = 0$, and $L_c = \infty$. For $\delta T_c$ pinning the disorder parameter designated by $\delta_\alpha$ is proportional to $\xi$ while for $\delta l$ pinning the corresponding disorder parameter varies as $\delta_m \propto \xi^{-3}$ [1]. For both types of pinning one can, however, derive the following $\delta$-independent relation:

$$U_c(T,B) = (B_c^2\xi^3\varepsilon/\mu_0 j_0^{1/2})j_c^{1/2}(T,B) \qquad (2)$$

for single vortex pinning [see Eq. (1)]. Similar, $\delta$-independent relations for small and large vortex (super)bundles can also be derived [7].

In their monumental review article Blatter *et al.* [1] conclude, however, that in high-$T_c$ superconductors it has not been possible so far to prove experimentally which of the two pinning mechanisms ($\delta T_c$ or $\delta l$ pinning) is the dominant one. The purpose of this Letter is to demonstrate that for stoichiometric yttrium-based high-$T_c$ superconducting thin films $\delta l$ pinning is the only important pinning mechanism below $\sim 80$ K in moderate magnetic fields ($B < 2$ T). As explained below this conclusion is reached without using any explicit model for the disorder parameter $\delta$. Moreover, the independence of the $\delta l$-pinning mechanism on the substrates and growth conditions of the films suggests that the source of $\delta l$ pinning is related to randomly distributed oxygen vacancies [8] which are likely to be present in all studied films.

The samples investigated in this work are of three different types: (i) $YBa_2Cu_3O_7$ films of 180 nm thickness obtained by molecular-beam epitaxy (MBE) codeposition of pure Y and Cu from electron guns and $BaF_2$ from a Knudsen cell and postannealed in wet oxygen in order to remove the fluorine [9]. From x-ray diffraction patterns it is found that the FWHH of the (005) peak is 0.28°. These films have thus a good crystallinity with the $c$ axis perpendicular to the film plane. The critical temperature is $T_c \cong 91$ K and the transition width $\Delta T_c = 0.5$ K. The substrates are made of $SrTiO_3$ as in (ii). (ii) $YBa_2Cu_4O_8$ films of 100 nm obtained with a deposition technique [10] similar to that in (i), with $T_c = 78$ K and $\Delta T_c = 0.5$ K. (iii) $YBa_2Cu_3O_7$ films of 450 nm thickness deposited onto MgO(100) substrates using *in situ* 90° off-axis sputtering [11]. X-ray diffraction as well as Rutherford backscattering channeling measurements show that the films are grown epitaxially on the MgO(100) substrates, with a channeling minimum yield smaller than 7%. The layers are purely $c$ axis oriented and have narrow rocking curves [FWHH(005) $\le 0.3°$]. The as-prepared films have a critical temperature $T_c \cong 89$ K and $\Delta T_c < 2$ K.

The superconducting current $j_s$ induced in the films when the external magnetic field is swept at a constant


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rate $dB/dt$ was determined by means of a sensitive capacitance torque magnetometer similar to that described in Refs. [12] and [13]. The torquemeter is mounted in the continuous flow cryostat of a 7 T Oxford Instrument cryomagnetic system. The temperature is stabilized to within 0.1 K. For all measurements the external field is applied at a finite angle close to the $c$-axis direction since the torque vanishes when $B$ is exactly parallel to the $c$ axis. (For the analysis it is, however, completely justified to consider $B$ as being parallel to the $c$ axis.) The dynamic relaxation rate $Q$ [defined as $Q = d\ln j_s / d\ln(dB/dt)$], which is essentially equal to the conventional relaxation rate $-d\ln j/d\ln t$, is determined from the variation of $j_s$ as a function of the sweep rate [14,15]. At low temperatures it is experimentally found that $j_s$ depends linearly on $\ln(dB/dt)$ over a large range of sweep rates, $10^{-7}$ T/s $\leq dB/dt \leq 4\times10^{-2}$ T/s. At higher temperatures a clear curvature develops. In Figs. 1(a) and 1(b) we show the temperature dependence of $j_s$ and $Q$ at 0.5 T for the three different films. The spread in the absolute values of $j_s$ reflects the microstructural differences between films grown under different conditions and/or on different substrates. Similar data were obtained at 1 and 2 T.

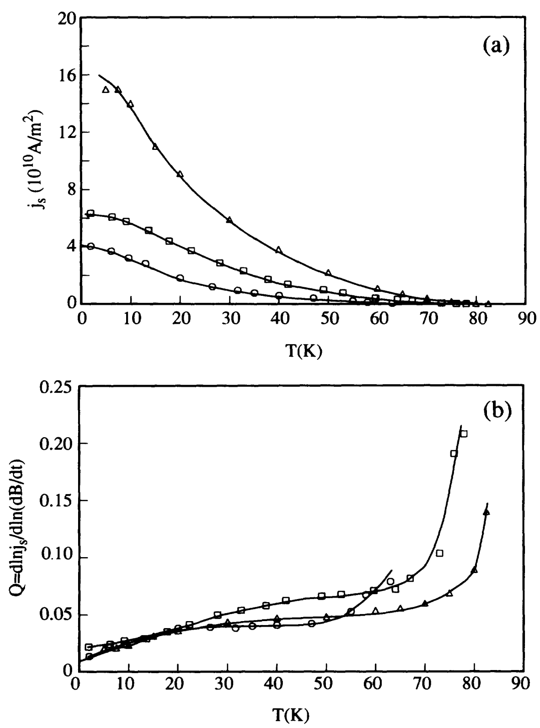


FIG. 1. Experimental data for (a) the superconducting current density $j_s$ and (b) the dynamic relaxation rate $Q \equiv d\ln j_s/d\ln(dB/dt)$ as a function of temperature at $B=0.5$ T. Both $j_s$ and $Q$ are determined at $dB/dt = 4\times10^{-2}$ T/s. The symbols correspond to three different films: (Δ) $YBa_2Cu_3O_7$ magnetron sputtered; (□) MBE codeposited $YBa_2Cu_3O_7$ with postannealing and (○) MBE codeposited $YBa_2Cu_4O_8$ with postannealing. The finite value of $Q$ at $T=0$ is due to quantum creep [16]. The lines are guides to the eye.

In the absence of dissipation $j_c(T,B)$ is equal to $j_s(T,B)$ and can directly be used for a comparison with theory. In the presence of the giant flux creep characteristic of high-$T_c$ superconductors $j_s$ differs, however, markedly from the true critical current $j_c$ (which corresponds to a vanishing activation energy for thermally activated vortex motion) and $Q$ is not simply related to the activation energy $U(j,T,B)$ which governs the hopping probability ($\propto \exp[-U(j,T,B)/kT]$) of a vortex or vortex (super)bundle. As shown recently by Schnack *et al.* [16], this is not a major difficulty since it is possible to determine reliably $j_c(T,B)$ and $U(j,T,B)$ over a large portion of the $(j,T)$ plane (at constant $B$) by means of the so-called generalized inversion scheme (GIS). The GIS which is described in detail in Ref. [16] is much more general than all the methods of analysis proposed until now [17] because it assumes only that

$$U(j,T,B) = g(T,B) f(j/j_c(T,B),B) \quad (3)$$

and

$$g(T,B) = \left[\frac{j_c(T,B)}{j_c(0,B)}\right]^p G(T). \quad (4)$$

As $j_c = j_c(T,B)$ is defined by the condition $U(j_c,T,B) = 0$, we have necessarily $f(1,B) = 0$. Since only the product $g\times f$ is physically relevant, both $g$ and $f$ are determined up to a multiplicative constant. We set arbitrarily $g(0,B) = 1$. The thermal function $G(T)$ depends on the dimensionality of the superconductor under investigation and on the vortex pinning and creep regime. For a collectively pinned single vortex (SV) one finds $p = 1/2$ and [18]

$$G_{SV}(T) = (1-t^2)^{-1/4}(1+t^2)^{5/4} \quad (5)$$

as follows directly from Eq. (2) with $B_c \propto (1-t^2)$, $\xi \propto [(1+t^2)/(1-t^2)]^{1/2}$, and $\lambda \propto (1-t^4)^{-1/2}$ with $t = T/T_c$. For the collective creep of small vortex superbundles (SB) in three dimensions Wen *et al.* [7] found $p = -3/2$ and

$$G_{SB}(t) = (1-t^2)^{3/4}(1+t^2)^{17/4}. \quad (6)$$

Finally for large vortex superbundles (LB) $p = -1/2$ and

$$G_{LB}(t) = (1-t^2)^{-3/4}(1+t^2)^{7/4}. \quad (7)$$

Both Eqs. (6) and (7) apply to the case of intermediate and large fields when the vortex-vortex interaction plays an essential role. In order to avoid excessive compressional energy increases during a creep process vortex bundles form elongated superbundles containing $(C_{11}/C_{66})^{1/2}$ elementary bundles [19]. This has been taken into account in deriving Eqs. (6) and (7).

We show now that at $B = 0.5$ T all the data in Figs. 1(a) and 1(b) are in the single vortex regime over the whole temperature range. To illustrate how this conclusion can be drawn we consider in Fig. 2 the $j_c(T)$ curves obtained by means of the GIS, from the data for the MBE codeposited $YBa_2Cu_3O_7$ (YBCO) film for the three values $p = -3/2$, $-1/2$, and $1/2$ predicted by theory

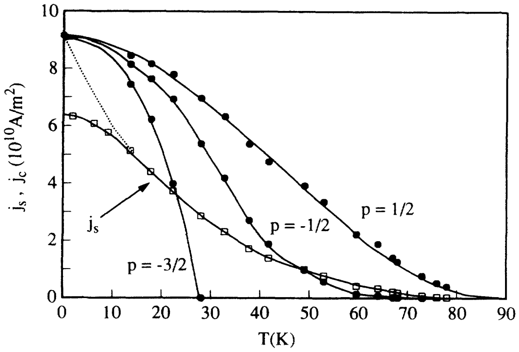


FIG. 2. Critical current density $j_c(T)$ values (●) obtained point by point by means of the generalized inversion scheme (GIS) of Schnack *et al.* [16] from the MBE codeposited $YBa_2Cu_3O_7$ film data in Fig. 1 (□) for three different values of the parameter $p$ in Eq. (4): $p=1/2$ corresponds to the single vortex regime, $p=-1/2$ to the small superbundle regime, and $p=-3/2$ to the large superbundle regime. The curves are guides to the eye. The dotted line indicates how $j_s$ would vary with temperature in the absence of quantum creep [16]. Similar curves are obtained for the other films.

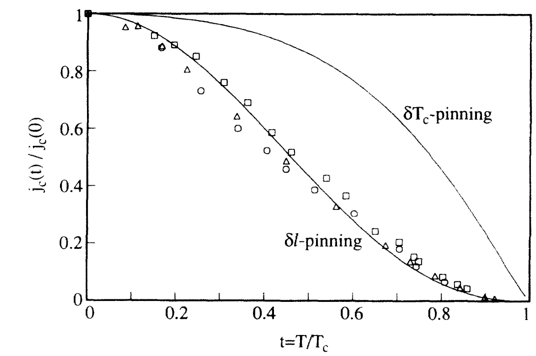


FIG. 3. Normalized temperature dependence of the critical current density for three samples at $B=0.5$ T. The symbols are the same as in Figs. 1(a) and 1(b). The "$\delta l$-pinning" line corresponds to Eq. (8) and the "$\delta T_c$-pinning" line to Eq. (10).

(see above). Both for $p=-3/2$ and $-1/2$ the critical current $j_c$ drops below the measured current $j_s$ above a certain temperature. This would imply that above this temperature the vortex system does not creep but flows. This is in contradiction with the nearly logarithmic dependence of $j_s$ on $dB/dt$ observed at these temperatures. Consequently, $p=1/2$ is the only physically acceptable value [20]. Very similar curves are obtained for the data of the two other types of films (MBE postannealed $YBa_2Cu_4O_8$ and magnetron sputtered $YBa_2Cu_3O_7$). Another evidence in favor of the single vortex regime is the weak field dependence of $j_c$. Up to $B=7$ T we found for all three types of samples that $j_c$ decreases with increasing $B$ approximately as $B^{-\alpha}$ with $\alpha\approx 0.3$. This is much weaker than the exponential decay $j_c \propto \exp[-2B^{3/2}]$ or $j_c \propto B^{-3}$ predicted by theory in the small bundle and large bundle regimes, respectively [1,19,21]. A further argument is that on theoretical grounds [1] it is expected that the single vortex regime is realized as long as $B < B_{SB} \cong 5(j_c/j_0)B_{c2}$. With $j_c=10^{11}$ A/m$^2$, $j_0 \cong B_c/\mu_0\lambda = 10^{13}$ A/m$^2$, and $B_{c2}=50$ T one obtains $B_{SB}\cong 2.5$ T. An additional piece of evidence for $p=1/2$ has also been found from measurements of $U_c$ and $j_c$ in a series of oxygen deficient $YBa_2Cu_3O_x$ films with $6.35 \le x \le 7$, which lead to $U_c(x) \propto [j_c(x)]^{0.51}$ [22].

In Fig. 3 we show the critical current as a function of temperature as determined point by point from the data in Figs. 1(a) and 1(b) by means of the GIS with $p=1/2$. To demonstrate the great similarity between the critical currents of the three samples the $j_c$'s are scaled to their respective values at $T=0$ and the temperature is normalized to $T_c$. Since the GIS makes it possible to reconstruct $U$ as a function of $j$ and $T$ for a given external field, it is, in particular, possible to determine the functions $g$ and $f$ in Eqs. (3) and (4). For the purpose of the present work we focus our attention on $g(T, B=0.5\ \mathrm{T})$ which is plotted as a function of the reduced temperature $T/T_c$ in Fig. 4 for the same three samples as in Fig. 3. The fact that the data points in Figs. 3 and 4 merge into a narrow band is rather remarkable since the films are of clearly different types.

At this point it is very important to stress that the data points in Figs. 3 and 4 have been obtained *without any assumptions about the type of pinning.* The temperature dependences in the thermal functions $G(T)$ entering the GIS [Eqs. (5) to (7)] are only related to the $T$ dependence of equilibrium thermodynamic parameters such as $B_c$, $\lambda$, and $\xi$. The disorder parameter $\delta$ does not enter at all the GIS.

Having emphasized this we compare now the data in Figs. 3 and 4 to the prediction of specific theoretical models. As all the data are in the single vortex regime, $j_c$ and $U_c$ can directly be calculated from the relations in Eq. (1) once a theoretical model has been chosen for $\delta$. For $\delta l$ pinning the disorder parameter is $\delta_m \propto \xi^{-3}$ and one obtains from Eq. (1)

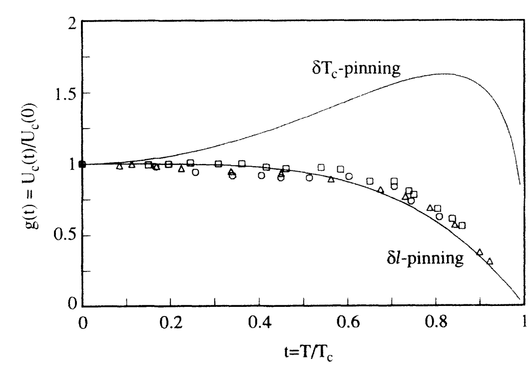


FIG. 4. Normalized temperature dependence of the $g(t)$ function introduced in the generalized inversion scheme [Eqs. (3) and (4)] for three different samples at $B=0.5$ T. This function has the same temperature dependence as the characteristic pinning energy $U_c$ [see Ref. [1]). The symbols are the same as in Figs. 1(a) and 1(b). The "$\delta l$-pinning" line corresponds to Eq. (9) and the "$\delta T_c$-pinning" line to Eq. (11).

$$j_c(t)/j_c(0)=(1-t^2)^{5/2}(1+t^2)^{-1/2}, \quad (8)$$

$$g(t)=1-t^4. \quad (9)$$

For the $g(t)$ function we have used the fact that $g(T,B)$ introduced in the GIS corresponds to $U_c(T,B)/U_c(0,B)$. The lines corresponding to Eqs. (3) and (4) are indicated as "$\delta l$ pinning" in Figs. 3 and 4. The central result of this Letter is the remarkably good agreement found between the $j_c(t)/j_c(0)$ and $g(t)$ data and the corresponding "$\delta l$-pinning" lines in these figures. In sharp contrast, the "$\delta T_c$-pinning" lines in Figs. 3 and 4 obtained with $\delta_a \propto \xi$, i.e.,

$$j_c(t)/j_c(0)=(1-t^2)^{7/6}(1+t^2)^{5/6}, \quad (10)$$

$$g(t)=(1-t^2)^{1/3}(1+t^2)^{5/3}, \quad (11)$$

are in total disagreement with the experimental data. This implies that the relevant disorder parameter varies as $\xi^{-3}$ and not as $\xi$. This conclusion also provides us with a direct explanation of the seemingly large spread in values for $L_c$ reported in literature. From resistivity measurements on YBCO/PrBCO multilayers with increasing YBCO layer thickness Brunner *et al.* [23] concluded that $L_c \cong 40$ nm in bulk $YBa_2Cu_3O_7$ for $B \leq 6$ T at $T/T_c \cong 0.9$. On the other hand, van Dalen *et al.* [24] concluded from their study of quantum creep at *low temperatures* in similar samples that $L_c \cong 4$ nm. This order of magnitude difference between $L_c$ ($t=0$) and $L_c$ ($t=0.9$) is easily understood on the basis of Eq. (1) where for $\delta l$ pinning $L_c \propto \xi^2 \propto (1+t^2)/(1-t^2)$ and consequently $L_c(t=0.9)/L_c(t=0)=9.5$. For $\delta T_c$ pinning $L_c \propto \xi^{2/3}$ and $L_c(t=0.9)/L_c(t=0)=2.1$ which is much too small.

Finally a rough estimate of the relative importance of $\delta T_c$ pinning and $\delta l$ pinning can be made by noting that the reduced temperature at which both disorder parameters $\delta_a$ and $\delta_m$ are equal is certainly larger than 0.9 as no significant deviations from the "$\delta l$-pinning" line are observed in Fig. 3 below $t=0.9$. Since $\delta_a/\delta_m \propto \xi^4, \delta_a/\delta_m=1$ at $t \geq 0.9$ implies that $\delta_a(t=0)/\delta_m(t=0) \leq 10^{-2}$.

In conclusion, we have found strong evidence for the importance of $\delta l$ pinning, i.e., pinning related to spatial variations of the charge carrier mean free path in stoichiometric yttrium-based superconductors. Over the temperature range considered in this work $\delta T_c$ pinning is at least 2 orders of magnitude weaker. This seems to be generally valid since it has been found in MBE codeposited and magnetron sputtered $YBa_2Cu_3O_7$ as well as in $YBa_2Cu_4O_8$ films.

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