Thickness dependence of the irreversibility line in $YBa_2Cu_3O_7$ epitaxial thin films

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We report a determination of the irreversibility line of epitaxial *c*-axis-oriented $YBa_2Cu_3O_7$ thin films with different thicknesses obtained by irreversible magnetization measurements. While close to T_c the irreversibility line is obtained by direct measurements, a universal relation is found at lower temperatures which allows a determination of the irreversibility line down to 4.2 K. This line is found to shift towards large values when the film thickness d is increased and saturates for d larger than the penetration depth $\lambda_{ab}(0)$. The results are discussed in the framework of flux creep in quasi-two-dimensional flux-line-lattice and vortex-lattice melting models.

I. INTRODUCTION

At the present time, it is well established that in the phase diagram, magnetic field H versus temperature T, of high-T_c superconductors such as $YBa_2Cu_3O_7$ in the mixed state, there is a so-called irreversibility line $H_{irr}(T)$ above which the critical current falls to zero. However, the position of this line depends on the type of measurement and on the experimental accuracy. Different models in which $H_{irr}(T)$ is determined by vortex-glass transition, vortex-lattice melting, depinning or flux creep, have been proposed (see, for example, Ref. 1 and references therein). In most experimental articles one can find the results of $H_{irr}(T)$ measurements close to T_c only. This is due to the fact that at lower temperatures $H_{irr}(T)$ lies well above the available magnetic fields. Interpretation of the experimental results close to T_c turns out to be difficult, because in the high-temperature range ($T>70$ K) both the coherence length $\xi(T)$ and the penetration depth $\lambda(T)$ strongly vary with temperature. The temperature dependence of $H_{irr}(T)$ is therefore very similar in the different models. For a given field, the onset of irreversibility occurs at a temperature $T_{irr}(H)$ which scales with the magnetic field as

$$H_{irr} \propto [1-T_{irr}(H)/T_c(0)]^{\nu} .$$

In many cases, the exponent ν is equal to $\frac{3}{2}$. But larger values have been reported in the literature.[2,3] The value $\nu=\frac{3}{2}$ may be well explained both in the vortex-lattice melting[4,5] and flux-creep models.[6]

In this work we report a study of $H_{irr}(T)$ obtained by irreversible magnetization measurements down to low temperatures ($T=4.2$ K) for epitaxial $YBa_2Cu_3O_7$ thin films with different thicknesses. $H_{irr}(T)$ is obtained close to T_c by direct dc magnetization measurements and for temperatures below ~ 60 K by a scaling method. We will show that, well below T_c, the $H_{irr}(T)$ dependence may be expressed in the form

$$H(T,d)=A(d)F(T) ,$$

where d is the film thickness and $F(T)$ a sample independent universal function. The numerical coefficient A is found to be independent of thickness above approximately 1500 Å. This value is close to the penetration depth $\lambda_{ab}(0)$; A increases with increasing d for $d<\lambda_{ab}(0)$. Temperature and thickness dependences will be compared to the flux-lattice melting[5] and flux-creep[6,7] models.

II. EXPERIMENTAL TECHNIQUES

Epitaxial $YBa_2Cu_3O_7$ thin films have been prepared by pulsed laser deposition on single-crystalline MgO(100) substrates heated at 730 °C in an ambient oxygen pressure of 0.2 mbar. After deposition, the oxygen pressure was increased to 1000 mbar and the sample was slowly cooled down to room temperature. The film thickness was varied by changing the number of applied laser pulses. The film preparation is described in more detail in Ref. 8. The film surface is 2×2 mm^2 and the thickness is in the range 200–4000 Å. The films have been characterized by scanning tunneling microscopy (STM) and some of them by grazing incidence x-ray diffraction and x-ray reflectometry.[9] The surface topography of the films, imaged by STM under atmospheric pressure and at room temperature with constant current mode (0.2 nA and 0.6 V) and Pt-Ir tip, reveals, for film thickness less than 500 Å, small islands with a diameter of about 1000 Å. Thicker films show well-developed islands and step structure. Many of these islands can be identified as screw dislocations. X-ray reflectometry gives the accurate thickness for films with $d<1000$ Å and reveals that the surface roughness is in the range 10–20 Å, much smaller than d for all the films. The c axis of the films is oriented perpendicular to the substrate. The transition temperature T_c and the transition width ΔT_c (10–90 %), obtained by dc resistance measurements in zero magnetic field, are given in Table I. It should be noted that T_c is found

TABLE I. Properties of the investigated thin films (see text for detail). The film thickness d has been obtained from x-ray reflectometry for $d < 1000$ Å and from the number of laser pulses above.

Thickness d (Å)	245	375	856	2000	3000	4000
T_c (K)	75	83	87	88	89	89
ΔT_c (K)	6.3	2.7	1.2	1.3	1	1.2
T_1 (K) (see text)	5	20	56	60	60	60
T_2 (K) (see text)	40	55	65	70	70	70

significantly lower than 89 K only for the thinnest films.

The magnetic properties have been studied by means of a homebuilt high-sensitivity (magnetic moment of 10^{-6} emu) vibrating sample magnetometer in fields up to 60 kOe. In order to study the irreversibility line, we measured the magnetization hysteresis loops in the transverse magnetic field $H \| c$, varied from -60 to 60 kOe, with a constant sweeping rate $v \approx 30$ Oe s^{-1}.

III. EXPERIMENTAL RESULTS

The irreversible magnetization $M_{\rm irr}(H)$, defined as the half difference between the measured magnetization on the H ascending and H descending branches of the major hysteresis loops, is shown in Fig. 1 for a 2000-Å film in the temperature range 4.2–85 K. At constant temperature the irreversible magnetization decreases with increasing magnetic field and vanishes at a field defined as $H_{\rm irr}(T)$. Close to T_c, $H_{\rm irr}(T)$ is experimentally defined as the field for which the difference between the upper and lower branch of the hysteresis loop is smaller than 10 emu cm^{-3}.

Below a temperature T_1, the value of $H_{\rm irr}$ exceeds the available magnetic fields of 60 kOe. The directly measurable irreversibility line is therefore limited to temperatures above the value T_1 for each film. However, we found, as in Ref. 10, that the normalized irreversible magnetization m is a universal function of a normalized field H/H^*:

$$m \equiv \frac{M_{\rm irr}(H,T)}{M_{\rm irr}(0,T)} = m\left[\frac{H}{H^*}\right] . \tag{1}$$

The universal dependence given by Eq. (1) is found at temperatures lower than T_2. The value of T_2 is higher than T_1 for all the films we have measured. Thus, there is a temperature interval in which we are able to determine the irreversibility line directly ($T > T_1$) and in which the magnetization follows a universal function ($T < T_2$). In this interval, $T_1 < T < T_2$, the values of H^* and $H_{\rm irr}$ coincide. For temperatures lower than T_1, H^* is chosen such that the normalized irreversible magnetization coincides with the one measured in the interval $T_1 < T < T_2$. The universal dependence $m(H/H^*)$ for films of different thicknesses is shown in Fig. 2. One may see that for each film, the dependence of m on H/H^* corresponds to a unique universal line, for all temperatures $T < T_2$. Thus, we believe that at $T < T_1$, H^* represents the field at which the irreversible magnetization would fall to zero if the available magnetic field was high enough. Therefore, we assume in the following that the scaling parameter H^* is the irreversibility field $H_{\rm irr}$ at temperatures lower than T_1.

From Fig. 2, one may assume that in fields close to H^* the dependence of the normalized irreversible magnetization is approximately linear:

$$m \propto (1 - H/H^*) . \tag{2}$$

Therefore, the determination of H^* does not seem to depend on the experimental criterion.

It is also clear from Fig. 2 that the universal line does not depend on the film thickness for $d \geq 2000$ Å. Since the London penetration depth $\lambda_{ab}(0)$ is 1500 Å, this may indicate that there is no thickness dependence for $d > \lambda_{ab}(0)$. For films with $d < \lambda_{ab}(0)$ the universal dependence changes when the thickness is decreased.

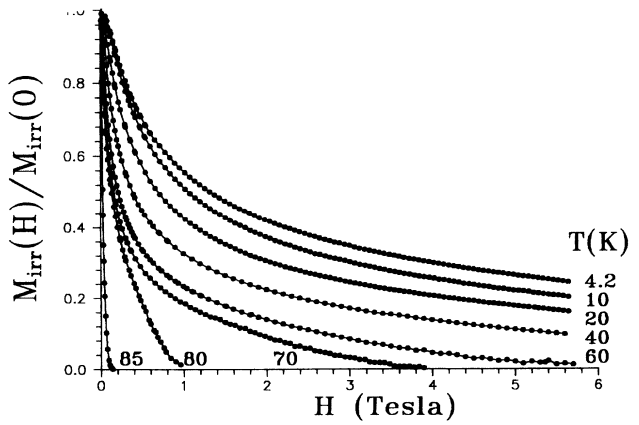

FIG. 1. Normalized irreversible magnetization as a function of magnetic field at different temperatures; film thickness 2000 Å.

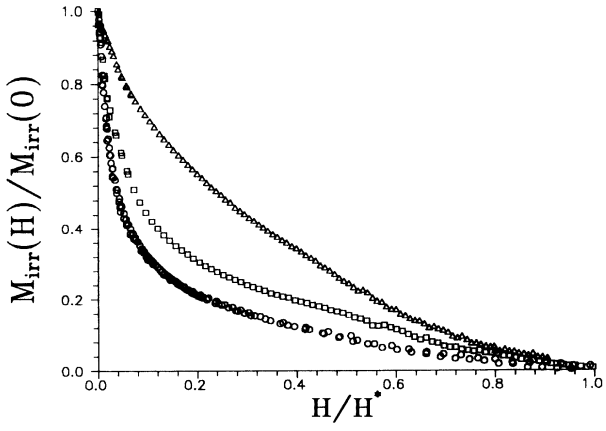

FIG. 2. Universal relation for $M_{\rm irr}(H)/M_{\rm irr}(0)$ vs H/H^* for films with different thicknesses: (○) 2000 Å, 3000 Å, 4000 Å; (□) 856 Å; (△) 375 Å, 245 Å. Each curve represents results from different measurement temperatures below T_2 (see text).

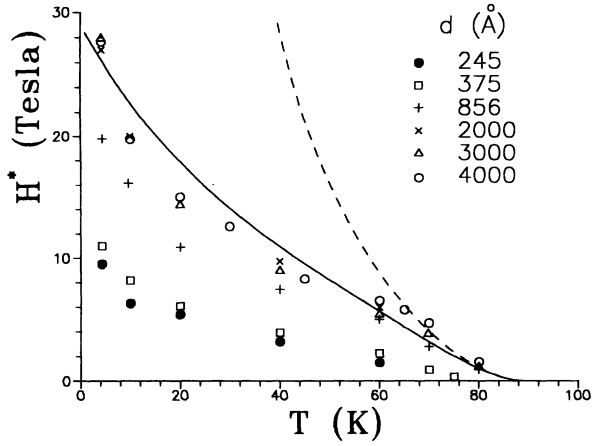

FIG. 3. Temperature dependence of the irreversibility line for films with different thicknesses d. The dashed line is a fit according to the melting model [Eqs. (9) and (10)]. The solid line is a fit according to quantum melting model [Eq. (12)].

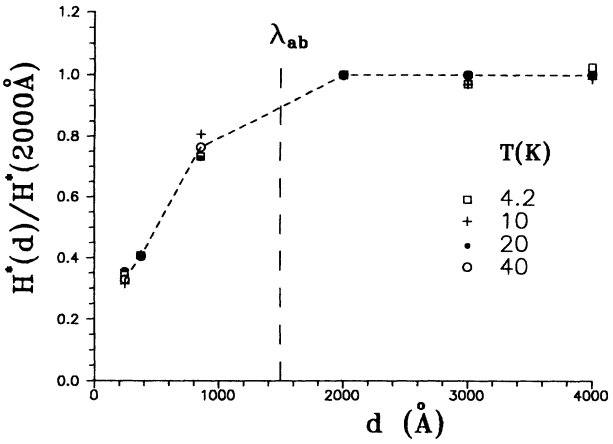

FIG. 5. Thickness dependence of the irreversibility field normalized to its value obtained on a 2000-Å film, for different temperatures $T=4.2$, 10, 20, 40 K.

Figure 3 shows the curves of H^* vs T for films with different thicknesses. These results are consistent with those reported in Ref. 11 close to T_c. We found, as illustrated in Fig. 4, that for all films, the temperature dependence of the ratio $H^*(T)/H^*(4.2\text{ K})$ is nearly the same except very close to T_c. At fixed temperature, in the range 4.2–60 K, the absolute value of H^* increases with d and saturates for $d > \lambda_{ab}(0)$, as shown in Fig. 5.

IV. DISCUSSION

In the critical state, the field B inside the film is $H+h$, where H is the applied field and h is the self-field created by the induced current. The Maxwell equation may be written as follows:

$$j_c = \frac{\partial h_r}{\partial z} - \frac{\partial h_z}{\partial r} , \tag{3}$$

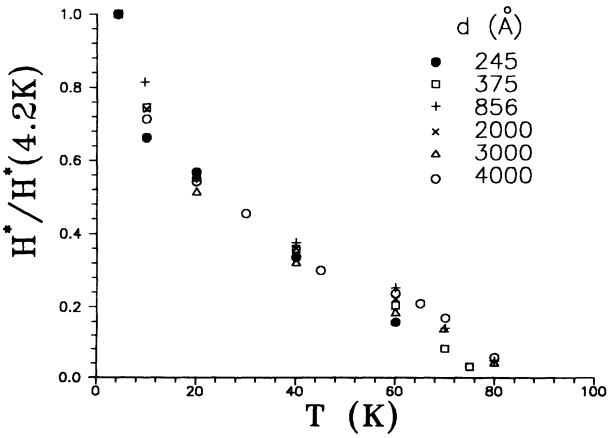

FIG. 4. Temperature dependence of the irreversibility field normalized to its value at $T=4.2$ K for films with different thicknesses.

where j_c is the critical current density, h_r and h_z are, respectively, the radial and axial components of h with $z \| c$. In the case of a thin film, in a transverse field, h_r and h_z are of the same order of magnitude, $h_r, h_z \sim h$ (Ref. 12). Thus, the first term in Eq. (3), corresponding to the vortex tilt, is of the order of h/d, and the second one, corresponding to the vortex density gradient, $\sim h/r$. Taking into account that $r/d \sim 10^4$, we may neglect the second term and consider that the critical state is related to the tilt of the vortices, their density being practically constant along r. It follows from Eq. (3) that the magnitude of the self-field h is about $j_c d$.

For $H < h$, the field inside the film is inhomogeneous and distributed in a quite complicated way.[12] Thus, due to the dependence of j_c on B, j_c will be significantly inhomogeneous and will depend both on the r and z coordinates. In our case, h does not exceed 100 Oe. Therefore, except in and close to the remanent state, h is much smaller than H on the hysteresis loop. In this case, one may consider the field inside the film to be H and the current to be homogeneous throughout the film: $j_c(B) = j_c(H)$. In the critical state model[13] the half-height of the magnetic hysteresis loop (the irreversible magnetization) is

$$M_{\text{irr}}(H) = \tfrac{2}{3} j_c(H) r , \tag{4}$$

where r is the equivalent radius of the film. Therefore, from the field dependence of M_{irr} one can obtain the field dependence of j_c. However, such a procedure is correct only when M_{irr} constant T and H does not change with time.

Pinning of vortices in type-II superconductors is usually described by an effective pinning potential U. If U is finite, which seems to be true in the case of thin films, M_{irr} at constant field and temperature may decrease with time (flux creep). This decrease may be characterized by a relaxation time τ_r so that $M_{\text{irr}}(\tau_r) \approx 0$. The value of τ_r depends on the ratio $U/k_B T$:

$$\tau_r(H,T) \sim \exp(U/k_B T) \ . \tag{5}$$

On the other hand, we can define a characteristic time for the experiment, τ_e. Suppose that we cool the film below T_c in a magnetic field $H \gg j_c(H)d$. At constant temperature, the magnetic field is increased with a sweeping rate $v = dH/dt$. The critical state in the sample will be completely established in a time $\approx h/v$, where h is the full penetration field for the vortices. This field is reached when $j_c(H)$ flows in the whole volume, so that $h \approx j_c(H)d$. Thus, the characteristic time of the experiment is given by

$$\tau_e(H,T) \sim \frac{j_c(H)d}{v} \ . \tag{6}$$

The experimental determination of $M_{\rm irr}$ depends on a competition between τ_e and τ_r. It is clear that, for fields smaller than $H^*(T)$ (when $M_{\rm irr} \neq 0$), the characteristic time of the experiment, τ_e, is much shorter than the relaxation time τ_r. The increase of the field at constant temperature leads to a decrease of U and therefore of τ_r. The measured value $M_{\rm irr}(H,T)$ reaches zero when $\tau_r < \tau_e$. The experimental $H^*(T)$ line corresponds then to the condition

$$\frac{\tau_r}{\tau_e} = \alpha < 1 \ . \tag{7}$$

We should note that τ_r in our experiments is of the order of 1s, so the condition given by Eq. (7) is fulfilled for large values of U.

A. Thickness dependence of H^*

If one assumes a logarithmic dependence of $M_{\rm irr}$ on t, one may fit the universal $m(H/H^*)$ and $H^*(T)$ experimental lines, choosing the appropriate $U(H,T)$ values, as was done in Ref. 10. From our point of view, such a procedure requires strong assumptions and we will not go beyond a qualitative explanation. First of all we shall discuss the thickness dependence $H^*(d)$, which seems to be the most interesting result of these experiments.

According to Eq. (6) τ_e is proportional to the thickness d, so that H^* which corresponds to the condition given by Eq. (7) should decrease with increasing d. Experimentally we found the opposite $H^*(d)$ dependence (see Fig. 5). That means that for the films with $d < \lambda_{ab}(0)$, the value of U significantly increases with increasing d.

Measuring the resistance of multilayers, the authors of Ref. 14 found that for isolated $YBa_2Cu_3O_7$ films with thickness in the range $24 < d < 240$ Å, the activation energy U is proportional to d and logarithmically decreases with H:

$$U/d = -\gamma \ln(H/\beta) \ , \tag{8}$$

where γ and β are parameters. Qualitatively this result is in a good agreement with our data. The increase of d leads to the increase of U and according to Eqs. (5)–(7) shifts H^* to larger values. The authors of Ref. 14 reached the conclusion that the dependence given by Eq. (8) is due to the creation of dislocation pairs in a quasi-2D flux-line lattice. It is known that such a quasi-2D approach is valid in the case of logarithmic vortex-vortex interaction, which takes place for the films with $d < \lambda_{ab}$ (Ref. 15). There is also another condition $\lambda^2/d > \rho$, where ρ is the Kosterlitz-Thouless correlation length, which is the characteristic dislocation pair size. Apart from the vicinity of the Kosterlitz-Thouless transition temperature, ρ is small ($\rho < \lambda$); thus, the condition $\lambda^2/d > \rho$ is always fulfilled for films with $d < \lambda$. Thus, the strong dependence of U on d should vanish for the films with $d \geq \lambda_{ab}$ and the $H^*(T)$ line should be approximately the same for all thicknesses larger than $\lambda_{ab}(0)$.

Finally, we may state that the experimentally observed increase of H^* with d and the further saturation of H^* for $d > \lambda_{ab}(0)$ give a strong support to the suggestion that for $d < \lambda_{ab}(0)$ the value of U corresponds to the creation of dislocation pairs in a quasi-2D flux-line lattice.

The discussion of the temperature dependence of H^* for thin $[d < \lambda_{ab}(0)]$ films seems to be quite difficult, because it requires assumptions about the $U(T)$ and $j_c(H,T)$ dependences. For thick $[d > \lambda_{ab}(0)]$ films, the temperature dependences of U and of H^* correspond to the 3D bulk case. Several recent experimental results, obtained on $YBa_2Cu_3O_7$ single crystals,[16,17] indicate the existence of a phase transition line $H_m(T)$ below the line $U = 0$. This suggests a possible mechanism for the temperature dependence of H^*, in the framework of the vortex-lattice melting model.

B. Vortex-lattice melting model

One may assume that in the bulk case, at $H = 0$, U is quite large so that $\tau_r \gg \tau_e$. This condition is kept with increasing H up to a phase transition, characterized by a line $H_m(T)$. In the case of a first-order transition, there is an abrupt drop of U on this line. In the case of a second-order transition, U depends smoothly on H, but the main, quite abrupt, decrease of U occurs in the vicinity of the phase transition line $H_m(T)$. Thus, we may assume that above $H_m(T)$, $\tau_r < \tau_e$ and $M_{\rm irr} = 0$. Thus, the experimental $H^*(T)$ line would correspond to $H_m(T)$.

Let us compare the curve $H^*(T)$ for thick films with the melting line given by the Lindeman criterion:

$$\langle u^2 \rangle = c_L^2 a^2 \ . \tag{9}$$

Here $\langle u^2 \rangle$ is the vortex mean-square displacement due to fluctuations, $a^2 \sim \Phi_0/B$ is the square of the vortex-lattice spacing and $c_L \approx 0.1$–0.3 is the Lindeman coefficient.

If we consider thermal fluctuations only,[18]

$$\langle u^2 \rangle = \langle u_{\rm th}^2 \rangle = \frac{k_B T 4\pi_{ab}^2 \varepsilon}{\Phi_0^{3/2} B^{1/2}} \ , \tag{10}$$

where $\varepsilon = (\lambda_c/\lambda_{ab})$ for $YBa_2Cu_3O_7$. The $H_m(T)$ dependence, calculated from Eqs. (9) and (10) with $c_L = 0.29$ is shown by a dashed line in Fig. 3. This temperature dependence differs significantly from our experimental data. It is in good agreement with the experimental results close to T_c, but at low temperatures the difference is more than one order of magnitude. It is impossible to fit

the experimental data with different values of c_L, because the temperature dependences of the experimental $H^*(T)$ and calculated $H_m(T)$ lines are significantly different. Thus, vortex-lattice melting cannot account for the experimental temperature dependence of H^* in the whole temperature range if we consider only classical thermal fluctuations.

Recently Blatter and Ivlev[5] have shown that one should also take into account zero-point quantum fluctuations of vortices, $\langle u_q^2 \rangle$, so that

$$\langle u^2 \rangle = \langle u_{\rm th}^2 \rangle + \langle u_q^2 \rangle \ . \tag{11}$$

The amplitude of zero-point fluctuations was estimated[5] to be of the order of the coherence length: $\langle u_q^2 \rangle \approx Q\xi^2$, where Q is a constant of the order of unity. Thus, from Eqs. (9)–(11) it follows:

$$H_m^{1/2} = \frac{(A^2 + 4Qc_L^2\Phi_0/\xi^2)^{1/2} - A}{2Q} \ , \tag{12}$$

where

$$A = \frac{k_B T 4\pi\lambda_{ab}^2 \varepsilon}{\Phi_0^{3/2}\xi^2} \ .$$

The solid line shown in Fig. 3 was calculated with $Q=2.9$ and $c_L=0.29$. One may see that this line is in good agreement with the experimental results for $d > \lambda_{ab}(0)$. Thus, in the vortex melting model, the experimental dependence of H^* on T for thick films indicates that quantum corrections have to be taken into account.

V. CONCLUSION

We have measured the irreversible magnetization $M_{\rm irr}$ of $YBa_2Cu_3O_7$ thin films in a transverse magnetic field. We have found that in a large temperature range the normalized magnetization $m \equiv M_{\rm irr}(H,T)/M_{\rm irr}(0,T)$ is a universal function of a normalized magnetic field H/H^* where H^* corresponds to the condition $M_{\rm irr}(H^*)=0$. The $m(H/H^*)$ function does not depend on the thickness of the film, for $d > \lambda_{ab}(0)$. The irreversibility field $H^*(T)$ does not depend on d for $d > \lambda_{ab}(0)$ and decreases with film thickness for $d < \lambda_{ab}(0)$.

We believe that $H^*(T)$ is not a characteristic line on the phase diagram of the vortices and corresponds to a certain relation between the characteristic time of the experiment and the relaxation time. The decrease of $H^*(T)$ with thickness for $d < \lambda_{ab}(0)$ may be qualitatively explained in the framework of a quasi-2D flux-line lattice, with an activation energy U for flux creep due to the creation of vortex-lattice dislocation pairs.

The temperature dependence of H^* for thick films may be well described by melting, taking into account zero-point quantum fluctuations of vortices. However, it cannot be considered as a proof of vortex-lattice melting. Moreover, it is not clear why the dependence of H^* on T is the same for all films.

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