

# Collective dynamics of a highly dilute vortex lattice in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin films

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The dynamics of highly dilute vortex ensembles in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  thin films is investigated by low-field (5–90 Oe) ac susceptibility measurements of the dynamical relaxation rate  $Q$ . In a film with relatively weak pinning, flux bundling is found to be effective at all temperatures and down to the lowest fields. In a film with stronger pinning, single-vortex creep dominates only for  $T < 20$  K, and small bundle creep is fully developed for  $T > 40$  K. The behavior of highly separated vortices is hence surprisingly collective in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ . Quantum creep is observed below 11 K, and at about 60 K the elastic vortex description breaks down as the importance of plastic creep gradually increases.

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## I. INTRODUCTION

Although the theory of collective flux creep (CFC) was originally developed for conventional low- $T_c$  superconductors in high fields,<sup>1</sup> the advent of high- $T_c$  superconductivity has greatly stimulated its further development.<sup>2</sup> The nonlogarithmic time decay of the remanent magnetization<sup>3</sup> and the peculiar universal and temperature-independent value of the relaxation rate in the  $\text{YBa}_2\text{Cu}_3\text{O}_7$  (Y-123) material system<sup>4</sup> are, e.g., two experimental observations that are successfully explained by CFC theory.

Despite the large number of published works on vortex creep in Y-123,<sup>5–9</sup> a detailed low-field study of the vortex dynamics has not yet been presented. Such a study is of great importance to understand how the dynamics changes as isolated vortices begin to interact when their average separation is reduced. We have recently demonstrated<sup>10</sup> how vortex creep in  $\text{HgBa}_2\text{CaCu}_2\text{O}_{6+\delta}$  thin films gradually changes from single-vortex dislocation-mediated creep to elastic creep of flux bundles with increasing field and/or temperature, and it is of interest to search for a similar behavior in the Y-123 system. Exotic vortex phases, such as the theoretically predicted low-field vortex liquid, could possibly also be observed in a low-field thin-film geometry.<sup>11,12</sup>

In this work we present low-field ac susceptibility measurements on two  $c$ -axis-oriented Y-123 thin films laser ablated onto  $\text{LaAlO}_3$  and  $\text{SrTiO}_3$  substrates, respectively. The frequency dependence of  $\chi'(T, H_{ac}, f)$  is used to extract the dynamical relaxation rate  $Q(T, H_{ac})$  and to calculate the effective flux creep activation energy  $U_{\text{eff}}(T, H_{ac})$ .<sup>13</sup> From an analysis within CFC theory we deduce the dynamical exponent  $\mu$ , which characterizes the effective flux creep regime. In the film with weaker pinning (on  $\text{LaAlO}_3$ ), single-vortex creep is never effective since flux bundling occurs at all temperatures. In the other film,  $\mu$  indicates a crossover from single-vortex creep below 20 K, to small bundle creep in an intermediate-temperature range, possibly followed by creep of increasingly larger flux bundles. Contrary to our expectations, flux bundling is hence surprisingly important for highly separate vortices in Y-123 thin films. On approaching  $T_c$ , a negative  $\mu$  indicates the existence of plastic creep in both films, i.e., elastic deformations are superceded by plastic deformations as being the dominant creep mechanism. At

the very lowest temperatures, below  $T = 11$  K,  $Q$  tends to a finite value, which we attribute to quantum creep.

## II. PROBING COLLECTIVE FLUX CREEP WITH AC SUSCEPTIBILITY

A key result within CFC theory is the prediction of a nonlinear current dependence of the flux creep activation energy,

$$U(J_c) = \frac{U_0}{\mu} \left[ \left( \frac{J_{c0}}{J_c} \right)^\mu - 1 \right], \quad (1)$$

where  $J_{c0}$  is the true critical current density before flux creep sets in,  $J_c$  is the decaying momentary screening current, and  $\mu$  is a exponent describing the degree of nonlinearity, acquiring different values depending on the actual collective flux creep regime:  $\frac{1}{7}$ ,  $\frac{5}{2}$ , and  $\frac{7}{9}$  for single-vortex, small bundle, and large bundle vortex creep, respectively (Fig. 1).<sup>2</sup> The nonlogarithmic time decay of the remanent magnetization is a direct result of Eq. (1) since, as  $J_c$  decreases, the activation energy  $U$  for vortex motion increases.

In a so-called subcritical state, achieved in a ramped magnetic field, the momentary activation energy can be ex-

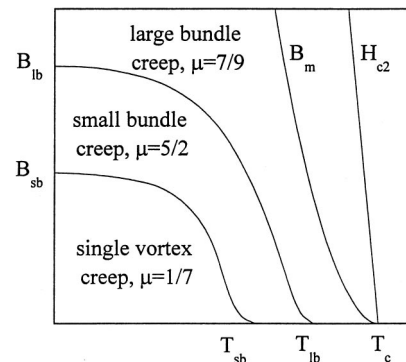


FIG. 1. Schematic phase diagram showing the different creep regimes and the corresponding values, for  $\mu$ . Crossovers between different creep regimes are expected at the  $(B_{sb}, T_{sb})$  and  $(B_{lb}, T_{lb})$  lines, respectively. Also shown are the upper branch of the melting line ( $B_m$ ) and the upper critical field ( $H_{c2}$ ).

pressed as  $U(J_c, T, H) = Ck_B T$ , where  $C = \ln(\nu_0/\pi Rf)$  in the case of a sinusoidally varying magnetic field  $h_0 \cos 2\pi ft$ ,  $\nu_0$  is an attempt velocity, and  $R$  is the radius of the sample.<sup>14</sup> Using Eq. (1), one finds the frequency dependence of the screening current,

$$J_c(T, H, f) = J_{c0}(T, H) \left( 1 + \frac{\mu C k_B T}{U_0} \right)^{-1/\mu}. \quad (2)$$

Experimentally, this frequency dependence is described by the dynamical relaxation rate,  $Q = d \ln J_c / d \ln f$ , and one gets for the so-called *effective* activation energy

$$U_{\text{eff}} \equiv \frac{k_B T}{Q} = U_0 + \mu k_B C T. \quad (3)$$

For finite  $\mu$  the second term will eventually dominate as the temperature increases, which is the reason for the characteristic plateau seen in plots of relaxation rate vs temperature for Y-123 samples.<sup>4</sup>

As recently demonstrated,<sup>10,13,15–17</sup> the field- and temperature-dependent dynamical relaxation rate  $Q$  can be conveniently determined from temperature scans of  $\chi'(T, H_{\text{ac}}, f)$  at a set of different fields and frequencies. For sufficiently large<sup>18</sup> ac fields, the critical current density and the measured in-phase susceptibility are related by  $J_c = 2(-\chi'/1.33\chi_0)^{2/3} h_0/d$ , where  $\chi_0$  is the full screening susceptibility and  $d$  is the sample thickness. The dynamical relaxation rate can hence be determined as a function of temperature and ac and dc field from

$$Q(T, H) = \frac{2}{3} \frac{d \ln |\chi'(T, H, f)|}{d \ln f}. \quad (4)$$

### III. EXPERIMENT

Two *c*-axis-oriented Y-123 thin films, one on a LaAlO<sub>3</sub> substrate (film I) and another on a SrTiO<sub>3</sub> substrate (film II), were prepared, in the same deposition run, by pulsed laser ablation at 750 °C in 200-mTorr O<sub>2</sub>, in an off-axis geometry. The deposition was followed by *in situ* annealing in 600-Torr O<sub>2</sub> atmosphere at 430 °C for 30 min. Film thickness was estimated to 50 nm from profilometry on similar 100-nm films grown for twice the amount of time at identical conditions. Only (00*l*) Y-123 lines were observed using x-ray diffraction, confirming single-phase *c*-axis-oriented samples. While no significant outgrowths were observed on film I using electron microscopy, film II revealed a fairly high surface density (11  $\mu\text{m}^{-2}$ ) of 50–200-nm large outgrowths.  $T_{c0} = 89.5$  K of both films was determined from usual four-contact resistivity measurements and the transition width, defined as  $\Delta T = T_{\rho=0.9\rho(94\text{K})} - T_{\rho=0.1\rho(94\text{K})}$ , was less than 3 K. The critical current density at 5 K was  $1.2 \times 10^{10}$  and  $1.2 \times 10^{11}$  A/m<sup>2</sup> for films I and II, respectively, determined from the ac field amplitude ( $H_{\text{ac}} = 5$  and 50 Oe rms), which positioned the loss maximum at 5 K.

Fundamental frequency sine-wave integrated in-phase ac susceptibility measurements,  $\chi'_1(T, H_{\text{ac}}, f)$ , were carried out using a home-built high-sensitivity ac susceptometer with a

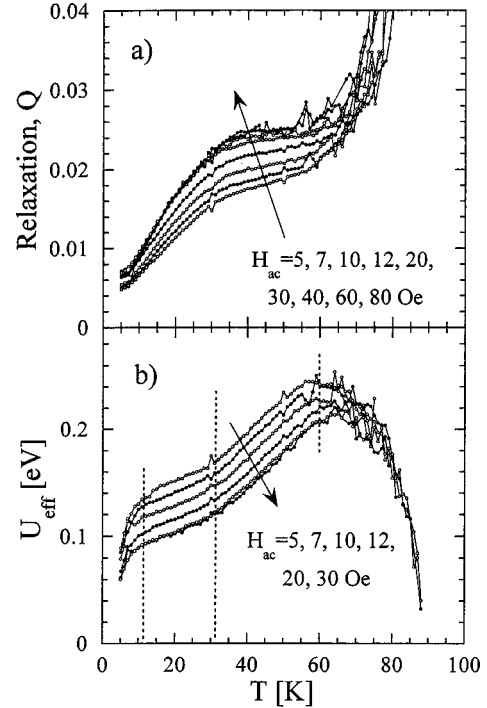


FIG. 2. (a) Dynamical relaxation rate  $Q$  vs  $T$  for film I in ac fields  $H_{\text{ac}} = 5, 7, 10, 12, 20, 30, 40, 60,$  and  $80$  Oe. (b) Effective flux creep activation energy  $U_{\text{eff}}$  vs  $T$  for  $H_{\text{ac}} = 5–30$  Oe calculated from the data in (a).

three-coil mutual inductance bridge and a background subtraction scheme.<sup>19</sup> Ac fields, applied normal to the film plane, ranged from  $H_{\text{ac}} = 5–90$  Oe rms ( $H_{\text{ac}} = 2^{-1/2} h_0$ ). Six different frequencies,  $f = 12.7, 18.1, 27.3, 89.1, 127,$  and  $181$  Hz, were used to determine the frequency dependence of  $\chi'$ .

### IV. RESULTS

In Fig. 2(a) we show the dynamical relaxation rate for film I as a function of temperature for ac fields  $H_{\text{ac}} = 5–80$  Oe. The corresponding effective activation energy is shown in Fig. 2(b) for  $H_{\text{ac}} = 5–30$  Oe. At all temperatures,  $Q$  ( $U_{\text{eff}}$ ) first increases (decreases) with field but then saturates at field-independent values for all fields 20–80 Oe. At the lowest temperatures ( $T < 11$  K),  $Q$  tends to a constant finite value with decreasing temperature and  $U_{\text{eff}}$  drops, which is indicative of quantum creep. For  $11 < T < 31$  K,  $Q$  increases with temperature and  $U_{\text{eff}}$  exhibits a remarkably linear temperature dependence. In an intermediate-temperature region ( $31 < T < 60$  K),  $Q$  depends much less on temperature, and a plateau value around  $Q = 0.025$  is reached for fields above 20 Oe.  $U_{\text{eff}}(T)$  remains linear in  $T$ , however with a distinct increase in the slope at 31 K. At about 60 K the slope of  $U_{\text{eff}}(T)$  abruptly changes sign and as  $U_{\text{eff}}$  drops to zero, on approaching  $T_c$ ,  $Q$  again increases with temperature and finally diverges. A similar behavior is found for film II in Fig. 3, with a few differences:  $Q$  does not saturate at the highest fields but instead increases monotonically for all fields, the overall temperature dependence is smoother, and the changes in the slope of  $U_{\text{eff}}(T)$  are more gradual.

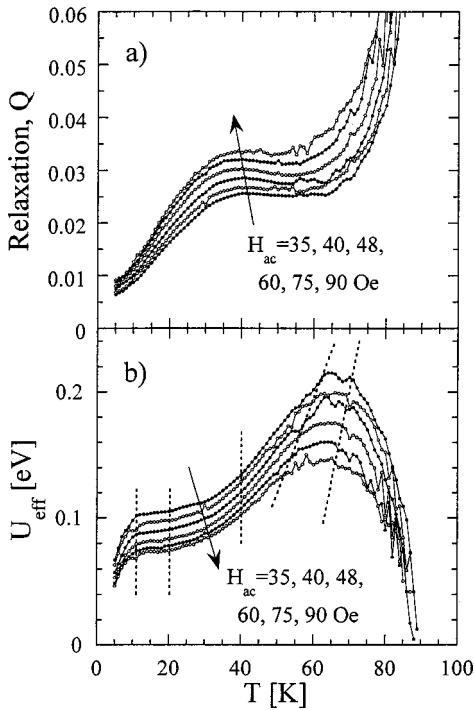


FIG. 3. (a)  $Q$  vs  $T$  for film II in ac fields  $H_{ac} = 35, 40, 48, 60, 75,$  and  $90$  Oe. (b)  $U_{eff}$  vs  $T$  calculated from the data in (a).

The general behavior of the relaxation rate for both samples is typical for the Y-123 system. In particular, the observed plateau value in the range  $Q = 0.022 - 0.026$  has been previously reported for different kinds of Y-123 samples, and is the most obvious indication of a finite value of  $\mu$ .<sup>4</sup> From the two distinct linear regions of  $U_{eff}(T)$  for film I we extract  $\mu C = 16.4$  and  $37$ , respectively, independent of ac field. The low end ( $11 < T < 20$  K) of the more continuous change in slope for film II yields  $\mu C = 4$ , and the high end ( $40 < T < 60$  K)  $\mu C = 36.5$ . The maximum value of the slope is hence identical for both samples.

As shown in Ref. 14 a value for  $C$  can be determined from the critical current density and the relaxation rate by means of  $C = \lim_{T \rightarrow 0} -(T/J_c) d \ln J_c / dT$ . Since this expression is only valid for thermally activated flux motion we estimate  $C$  from measurements in the region  $11 < T < 20$  K, where quantum creep does not seem effective. While the value  $C \approx 15 - 30$  for film I depends rather strongly on temperature, a less temperature-dependent value of  $C \approx 16$  is obtained for film II. In the following we therefore use  $C \approx 16$  for both films, which is close to previously reported values for Y-123 thin films.<sup>20</sup>

In Fig. 4 the estimated values for  $\mu$  are summarized for the two films as a phase diagram of different creep regimes. In the region  $11 < T < 20$  K the value  $\mu = 0.25$  for film II is in fair agreement with the theoretically predicted  $\frac{1}{7} = 0.14$ .<sup>2</sup> Above  $20$  K,  $\mu$  increases gradually and reaches a maximum value  $\mu = 2.3$  at a field-independent temperature of about  $40$  K. This is in good agreement with theory, which, for small bundle creep, predicts a maximum value of  $2.5$ . As a possible indication of a crossover into intermediate and large bundle creep,  $\mu$  begins to decrease at a field-dependent temperature,

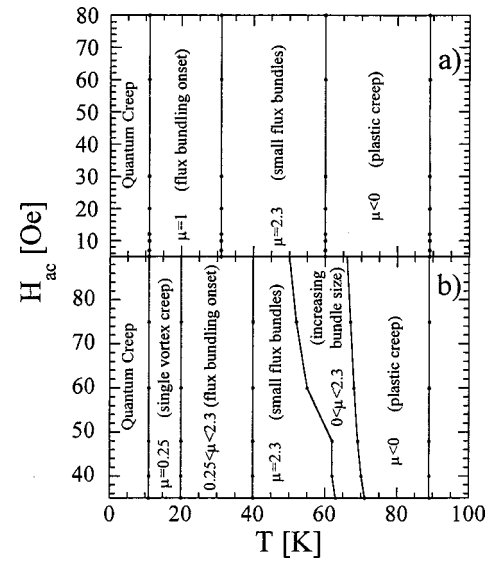


FIG. 4. Phase diagram of  $\mu$  as a function of temperature and ac field for (a) film I and (b) film II.

in the range  $50 - 63$  K, and finally becomes negative at about  $T = 66 - 71$  K. As expected, small bundle creep is hence found to persist to higher temperatures for the lower fields. A negative  $\mu$  indicates the presence of plastic creep, i.e., plastic deformations within the vortex lattice take over as the dominant mechanism for flux creep.<sup>21</sup> A similar phase diagram can be made for film I, where in particular,  $\mu$  reaches the same small bundle value  $2.3$  above  $31$  K. However, at low temperatures,  $\mu = 1$ , which suggests that flux bundling occurs down to the lowest temperatures in film I, i.e., the single-vortex creep regime is never realized. The smaller value of the critical current density indeed indicates weaker pinning, and it is hence possible that  $J_c$ , already at low temperatures, has decayed away from the single-vortex regime. Another indication of weaker pinning in film I is a general shift to lower temperatures of all thermal creep regimes.

A similar temperature dependence for  $\mu$  has been reported in a work by Thompson, Sun, and Holtzberg, where  $\mu$ , for a Y-123 single crystal in a field of  $1$  T, increases from about  $0.7$  at low temperatures, reaches a maximum of about  $2$  at  $T = 29$  K, and then decreases to about  $0.6$  at  $70$  K.<sup>3</sup> For a similar Y-123 single crystal at  $T = 40$  K, Civale *et al.* found  $\mu$  to increase with field from  $0.16$  at  $0.1$  T, to a maximum  $\mu = 1.4$  at  $1$  T above which  $\mu$  again decreases to  $\mu \approx 1$ .<sup>22</sup> In our low-field study, the small bundle regime seems dominant in a comparably large temperature window. The observed crossover temperature  $T_0 = 11$  K between thermal and quantum flux creep has also been seen in Y-123 thin films and single crystals at higher fields.<sup>20,23,24</sup>

## V. DISCUSSION

It is rather surprising that our low-field results should be so similar to results obtained in magnetic fields of the order of teslas. In a field of  $5$  Oe, vortices are on the average  $2 \mu\text{m}$  apart, whereas the penetration length of Y-123 is only  $\lambda(0)$

$=0.14 \mu\text{m}$ .<sup>25</sup> Since vortex-vortex interactions decrease exponentially with large intervortex distance, one would indeed expect vortices to creep individually at such large separations. At  $T=0$  the expected crossover field between single-vortex creep and small bundle creep,  $B_{sb}(0) \approx 5B_{c2}J_c/J_{GL}$ , with  $B_{c2} = \phi_0/2\pi\xi^2$ , the Ginzburg-Landau depairing current density  $J_{GL} = \phi_0/3^{3/2} \pi\mu_0\lambda^2\xi = 3.4 \times 10^{12} \text{ A/m}^2$ ,  $\phi_0 = 2.07 \times 10^{-15} \text{ T/m}^2$ , and  $\xi(0) = 1.5 \text{ nm}$  is about 2.6 and 26 T for films I and II, respectively. Yet, down to fields as low as 5 Oe the single-vortex creep regime seems absent in film I. The crossover temperature into fully developed small bundle creep can be found from solving

$$\frac{T_{sb}}{T_c} = \left( \frac{J_c(0)}{GiJ_{dp}(0)} \right)^{1/2} \left( 1 - \frac{T_{sb}}{T_c} \right)^{1/3}, \quad (5)$$

where the Ginzburg number  $Gi$  is of the order of  $10^{-2}$  for Y-123.<sup>2</sup> While  $T_{sb} = 31 \text{ K}$ , in agreement with the observed kink in  $U_{\text{eff}}(T)$ , is obtained using  $Gi = 0.022$ ,  $T_{sb} = 69 \text{ K}$  is in rather poor agreement with the experimental data for film II, indicating fully developed small bundle creep already at 40 K. To get  $T_{sb} = 40 \text{ K}$  for film II one would have to assume an unrealistically high  $Gi = 0.12$ .

One possible explanation for the observation of flux bundling already at very low fields could be the limited sample thickness of 50 nm, resulting in an effective penetration depth  $\lambda_{\text{eff}} = 2\lambda^2/d = 0.78 \mu\text{m}$ . As the two-dimensional situation of Pearl's vortices<sup>26</sup> is approached ( $d \ll \lambda$ ), the vortex interactions also become long range, as the essentially exponential decay with distance  $\exp(-r/\lambda)/r^{1/2}$  gradually changes into an inverse dependence  $r^{-1}$ . It might be that the combined effect of a longer penetration depth and the long-range nature of vortex-vortex interactions can drastically reduce  $B_{sb}$  and possibly also  $T_{sb}$ . Further low-field studies on even thinner samples might clarify this point.

The main differences in  $U_{\text{eff}}(T, H)$  between the two films—saturation with field and smoothness of the temperature dependencies—are likely to originate from the different defect structures of the two films. The observed decrease of  $U_{\text{eff}}$  with vortex density indicates the presence of a limited number of strong pinning sites that are the first to get occupied at low fields.<sup>24</sup> As the vortex density increases the average pinning energy decreases since the vortices that arrive at a later stage have to fill up the weaker pinning sites. The existence of saturation in film I indicates that all strong pin-

ning sites have been occupied and that flux creep is only due to vortices that are pinned with approximately the same weaker energy. The distinct kink in  $U_{\text{eff}}(T)$  of film I at 31 K also indicates that the entire vortex ensemble changes nature at the same temperature, i.e., the ensemble is relatively uniform. The absence of saturation in film II, on the other hand, indicates that this sample has a larger number and a wider distribution of strong pinning sites and at the fields used not all of them have yet been occupied. The characteristic field of 225 Oe, associated with the outgrowth density in film II, is indeed larger than any field used in this study. The smoother temperature dependence of film II also points to a more heterogeneous vortex ensemble, possibly linked to the size dispersion of the outgrowths. On the other hand, as it is well known that the number of other defects, such as edge and screw dislocations, correlates strongly with both  $J_c$  and general pinning properties,<sup>27</sup> it is unlikely that the outgrowth density alone should govern the detailed low-field vortex behavior. It is, however, beyond the scope of this work to determine the respective contribution of each defect type to the overall pinning strength.

## VI. CONCLUSIONS

We have carried out a detailed ac susceptibility study of the dynamics of highly dilute vortex ensembles in Y-123 thin films. The frequency dependence of the ac susceptibility response has been used to determine the dynamical relaxation rate, the effective activation energy, and the dynamical exponent  $\mu$  in ac fields  $H_{\text{ac}} = 5 - 90 \text{ Oe}$  and in a temperature range  $T = 5 - 89.5 \text{ K}$ . Our results suggest that quantum creep is effective up to  $T = 11 \text{ K}$ . Above 11 K thermally activated collective flux creep successfully describes the vortex dynamics, with small bundles ( $\mu = 2.3$ ) dominating at intermediate temperatures. As  $T_c$  is approached, a negative value for  $\mu$  is extracted, indicative of plastic flux creep. Indications of single-vortex creep ( $\mu = 0.25$ ) are only observed at the lowest temperatures and only in the film with a high critical current density. Flux bundling can hence be effective over a much wider field range than was previously known, even down to fields as low as 5 Oe.

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