Two-stage crossover from thermal to quantum flux creep of dilute vortex ensembles in various high-T_c superconducting thin films

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(Received 20 February 2001; published 8 August 2001)

The thermal-to-quantum flux creep crossover at low vortex densities has been studied in $YBa_2Cu_3O_7$, $TlBa_2CaCu_2O_{7-\delta}$, and $HgBa_2CaCu_2O_{6+\delta}$ thin films using ac susceptibility. The crossover temperatures T_{cr} are 10–11, 17, and 30 K, respectively. Both thermal and quantum flux creep is suppressed as the vortex density is decreased. We observe a two-stage nature in the crossover behavior which appears to be a general property of all the three materials studied.

DOI: 10.1103/PhysRevB.64.094509 PACS number(s): 74.60.Ge, 74.76.Bz

I. INTRODUCTION

Magnetic vortices in type-II superconductors are one of the few systems in which macroscopic quantum tunneling can occur. This intriguing phenomenon was first reported in high-T_c superconductors (HTS),[1] and although subsequently observed in conventional superconductors,[2–4] its study has mostly focused on optimally doped Y-123,[5–11] oxygen-deficient Y-123,[12,13] and other HTS materials such as $Y_{1-x}Pr_xBa_2Cu_3O_7$,[14] $YBa_2Cu_3O_8$,[11] $Bi_2Sr_2CaCu_2O_8$, and $Bi_2Sr_2Ca_2Cu_3O_{10}$,[15–17] $Tl_2Ba_2CaCu_2O_8$,[18–20] $Tl_2Ba_2Ca_2Cu_3O_{10}$,[21] $Hg_{0.8}Tl_{0.2}Ba_2CaCu_3O_8$,[22] and $YBa_2Cu_3O_7/PrBa_2Cu_3O_7$ multilayers.[23,24] However, to the best of our knowledge, quantum creep has previously not been studied in either $TlBa_2CaCu_2O_{7-\delta}$ (Tl-1212) or $HgBa_2CaCu_2O_{6+\delta}$ (Hg-1212), which are two of the materials investigated in this work. These material systems are of particularly high technological importance due to their high critical temperature, high critical current density, and moderate anisotropy.[25,26]

The low-temperature flux creep behavior can be very different for single crystals and thin films of the same material.[27] The crossover temperature T_{cr}, below which quantum creep effects can be observed, is generally higher for thin films. Theory also predicts a higher T_{cr} in the case of single-vortex creep (low fields) compared to creep of flux bundles, since the tunneling probability decreases with the size of the tunneling object.[28] Thin films generally also show a strong suppression of quantum creep with decreasing field, whereas a similar anomaly is not observed for single crystals. This difference has been ascribed to differing characteristic pinning sites of thin films and single crystals. In particular it has been argued that the suppression of quantum creep as $B \rightarrow 0$ is due to the presence of a limited number of strong pinning sites that dominate the pinning at low vortex densities. Hence, we expect both an increase in T_{cr} and a decrease in overall creep in our thin film samples with highly dilute vortex ensembles.

In this work we determine the thermal-to-quantum crossover temperatures for Y-123, Tl- and Hg-1212 thin films at very low fields (7–140 Oe). While we find $T_{cr}=10-11$ K for Y-123 thin films, in good agreement with previously published high-field values, T_{cr} is found to be as high as 17 and 30 K for Tl- and Hg-1212 thin films respectively. Furthermore, the crossover seems to proceed in two distinct steps, and consequently a second temperature T_{cr}^* can be defined for all three materials. We also find that quantum creep is suppressed down to the lowest accessible fields.

II. THERMAL AND QUANTUM CREEP

The theory of collective flux creep[29,30] (CFC) predicts a nonlinear current dependence of the flux creep activation energy,

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

where J_{c0} is the original critical current density before flux creep sets in, J_c is the measured decaying critical current density and μ is an exponent describing the degree of nonlinearity, with values between 1/7 and 5/2 depending on the actual collective flux creep regime. The concept of a temperature- and field-dependent μ exponent has been successfully applied in the analysis of flux creep in Y-123.[31–35] It is also well known that a simpler, logarithmic current dependence,[36] $U(J_c)=U_0\ln(J_{c0}/J_c)$, which formally corresponds to $\mu=0$, is often a very good description of experi-

mental data for very anisotropic materials such as, e.g., $Tl_2Ba_2CaCu_2O_{8+x}$ (Tl-2212) and $Bi_2Sr_2CaCu_2O_{8+x}$ (Bi-2212).[37–41] We have previously shown[42–44] that $\mu=0$ also is a good approximation for Hg-1212 and Tl-1212 thin films.

In a sinusoidally varying magnetic field, $h_0\cos 2\pi ft$, the momentary activation energy can be expressed as $U(J_c,T,H)=Ck_BT$ with $C=\ln(v_0/\pi Rf)$, v_0 being an attempt velocity and R the radius of the sample.[45] Substituting for U in Eq. (1) yields the frequency dependence of the critical current density

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

By defining the dynamical relaxation rate $Q=d\ln J_c/d\ln f$, and substituting Eq. (2) for J_c, one gets for the so-called *effective* activation energy[46]

$$U_{\rm eff}\equiv\frac{k_BT}{Q}=U_0+\mu k_BCT. \tag{3}$$

In the case of $\mu=0$, applicable to the Tl- and Hg-1212 films below, T/Q is a direct measure of the activation energy U_0/k_B in units of kelvin. At low temperature, neglecting quantum creep, we hence expect a constant temperature independent value of T/Q. For $\mu>0$, typical for Y-123 thin films, T/Q contains an additional temperature dependent term which introduces a small linear increase of T/Q when plotted vs temperature.

Quantum creep effectively introduces a second relaxation path for the screening current, $Q_{qc}(T)$. The measured relaxation can, for simplicity, be regarded as the sum of quantum and thermal contributions $Q(T)=Q_{\rm th}(T)+Q_{\rm qc}(T)$, where $Q_{\rm qc}(T)$ has a finite value at $T=0$ and is reduced to zero as thermal fluctuations take over. T/Q will consequently experience a gradual decrease as quantum creep sets in and from a plot of T/Q vs T it is thus possible to define a crossover temperature $T_{\rm cr}$ below which quantum creep is observable.[12,27] It should be noted that defined in this way, $T_{\rm cr}$ marks the *onset* of quantum creep effects, which is the focus of our work. Alternatively one may define the crossover point as the temperature where quantum creep completely *dominates* the vortex dynamics and Q becomes essentially constant, which happens typically below 1 K.[5,16,22]

III. FLUX CREEP AS MEASURED BY ac SUSCEPTIBILITY

The large-amplitude ac susceptibility technique is well suited for low-field flux creep studies in superconducting thin films.[42,47,48] For a thin disk sample in a sufficiently large ac field, the critical current density and the measured in-phase susceptibility are related by

$$J_c=\left[\frac{-\chi'}{1.33\chi_0}\right]^{2/3}\frac{2h_0}{d}, \tag{4}$$

where $-\chi_0$ is the full screening susceptibility in the Meissner state, which can be measured at low ac field amplitude, and d is the sample thickness.[49] The dynamical relaxation rate can hence be determined as a function of temperature, ac and dc field from

$$Q(T,H)=\frac{2}{3}\frac{d\ln|\chi'(T,H,f)|}{d\ln f}. \tag{5}$$

A noncircular sample shape will introduce a small multiplicative term in Eq. (4) that will not survive the logarithmic differentiation in Eq. (5), which hence remains valid for any thin film sample. All films investigated in this work were of rectangular shape with lateral dimensions of a few mm.

It should be pointed out that the above equations are entirely based on a Bean critical state formulation of the ac response, which is an adequate description for *macroscopic* vortex displacement due to large ac amplitudes and low frequencies. Our analysis does not include other possible contributions to the frequency dependence, such as, e.g., vortex viscosity and inertia, normal fluid contributions and changes in the Labusch pinning parameter, which are relevant in the case of very weak ac field amplitudes at rf frequencies superimposed onto a large dc field background, for which *microscopic* vortex displacements govern the ac response.[50–53] However, the validity of Eq. (4) was confirmed following the procedure in Ref. 42 and the applicability of the critical state formalism was further checked by noting that the ratio between the loss maximum value and the full screening susceptibility, $\chi''_{\rm max}/\chi_0$, was always in the range 0.24–0.27, which is typical for a critical state with weak flux creep and clearly different from 0.4 which is found in the nonhysteretic regime.[51,54,55]

IV. EXPERIMENT

Two 50 nm thick *c*-axis-oriented Y-123 thin films, one on a $SrTiO_3$ substrate (Y1) and another on a $LaAlO_3$ substrate (Y2), were prepared, in the same deposition run, by pulsed laser ablation at 750 °C in 200 mTorr O_2. The deposition was followed by an *in situ* anneal in 600 Torr O_2 atmosphere at 430 °C for 30 min. $T_{c0}=89.5$ K of both films was determined from usual four-contact resistivity measurements. $J_c(5\ {\rm K})=1.2\times10^7$ A/cm^2 (Y1) and 1.2×10^6 A/cm^2 (Y2) was determined from the ac field amplitude that positioned the loss maximum at 5 K.[49]

The 200 nm thick Tl-1212 film (T1) was grown on a $LaAlO_3(100)$ substrate using a two-step method reported in detail elsewhere.[56] Briefly, off-axis rf sputter deposition of an amorphous Ba-Ca-Cu-O precursor film was followed by a high-temperature thallination anneal for 10 min at 825 °C in 630 Torr O_2. X-ray diffraction found these films to be phase-pure and *c*-axis-oriented. $T_c=88$ K and $J_c(5\ {\rm K})=1\times10^7$ A/cm^2 was determined using a commercial SQUID magnetometer.

The Hg-1212 thin films were fabricated using a similar two-step method involving the deposition of 400 nm thick Hg-free precursor films on $SrTiO_3$ substrates, followed by annealing in a Hg-vapor atmosphere at 820 °C for 30 min. The higher-quality sample, H1, with $T_c=120$ K and $J_c(5\ {\rm K})=5\times10^6$ A/cm^2 showed mainly (00*l*) lines of the

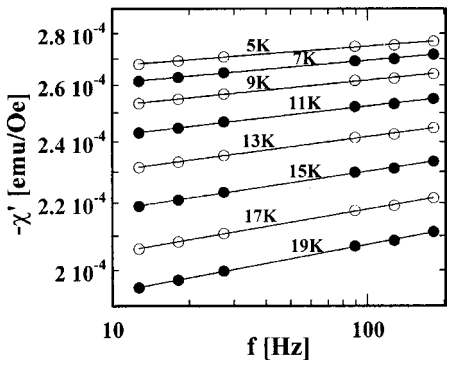

FIG. 1. Log-log plot of $-\chi'$ vs frequency for sample Y1 in an ac field of 60 Oe and at $T=5$, 7, 9, 11, 13, 15, 17, and 19 K. The straight lines are power law fits to the data.

c-axis-oriented Hg-1212 phase with minute traces of *c*-axis Hg-1223. Sample H2 $[T_c=110\,\mathrm{K}, J_c(5\,\mathrm{K})=5\times10^6\,\mathrm{A/cm^2}]$ showed additional, relatively strong, unidentified impurity lines.

Fundamental frequency sine-wave integrated in-phase ac susceptibility measurements, $\chi_1'(T,H_{\mathrm{ac}},f)$, were carried out using a custom built high-sensitivity ac susceptometer with a three-coil mutual inductance bridge and a background subtraction scheme.[57] ac fields, applied normal to the film plane, with frequencies between 12.7 and 181 Hz, ranged from $H_{\mathrm{ac}}=7-140$ Oe rms ($H_{\mathrm{ac}}=2^{-1/2}h_0$).

V. RESULTS AND DISCUSSION

The frequency dependence of χ' for sample Y1 at different temperatures is shown in Fig. 1. The straight lines are fits to a power law $\chi'\propto f^m$ from which $Q=m/1.5$ is determined.[47] The good quality of the fit indicates that possible self-heating due to hysteretic losses in the samples is negligible. The hysteretic losses deposited in the thin film being essentially proportional to the frequency, self-heating would be more than an order of magnitude stronger at 181 Hz than at 12.7 Hz leading to a frequency dependent slope with negative curvature in Fig. 1. We therefore conclude that any generated heat cannot give rise to the observed relaxation at low temperatures.[27,58,59]

The low-temperature dependence of the effective activation energy T/Q determined in different ac fields is shown for the five samples in Fig. 2. The straight lines emphasize the temperature dependence without the influence of quantum creep. The slow linear increase for the two Y-123 thin films corresponds to μ values of 0.25 and 1, respectively, as discussed in Ref. 60. The temperature independent behavior of the Tl- and Hg-1212 samples corresponds to $\mu=0$.

When the temperature is decreased a clear deviation from the straight lines is observed as T/Q sharply drops, indicative of the onset of quantum vortex tunneling. We define a crossover temperature, T_{cr}, from the point of the deviation and find $T_{\mathrm{cr}}=10-11$ K for the Y-123 films, $T_{\mathrm{cr}}=17$ K for the Tl-1212 film and $T_{\mathrm{cr}}=30$ K for the Hg-1212 films. No detectable field dependence of T_{cr} is observed in the investigated field range. The field independence of T_{cr} is strong evidence that the observed drop in T/Q reflects a true change

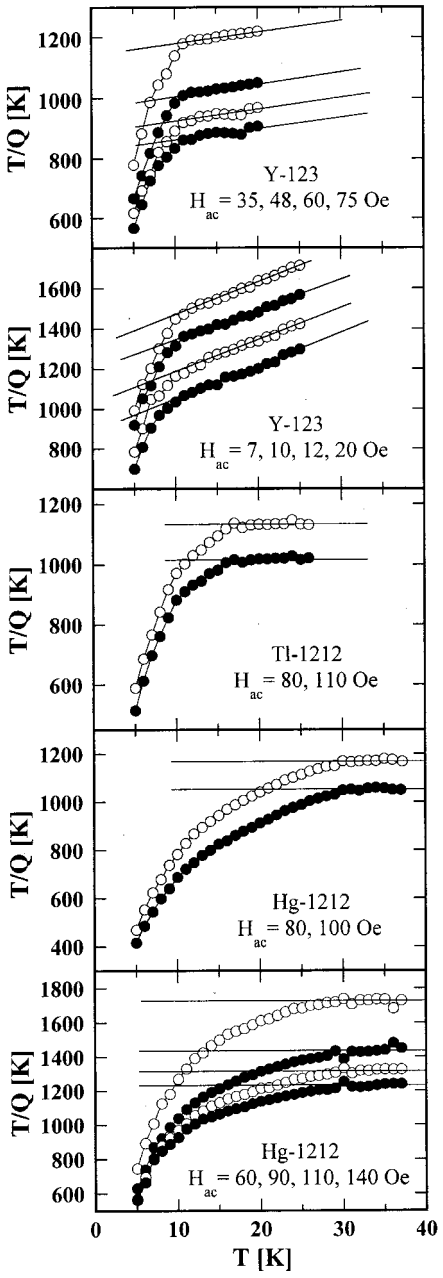

FIG. 2. Effective activation energy T/Q vs temperature and ac field for the five samples Y1, Y2, T1, H1, and H2. In each figure, the applied field is lowest for the top curve and increases between each following curve beneath it.

in the flux creep mechanism and is not caused by possible incomplete flux penetration due to the low fields used. Artifacts from incomplete flux penetration would set in at a certain value of H_{ac}/J_c and should therefore be strongly field dependent. We have made the same measurements in even weaker ac fields for which the incomplete flux penetration leads to an artificially *reduced* relaxation rate, i.e., it shows up as a sharp *increase* in T/Q below a certain field-dependent temperature and not as the drop seen in Fig. 2.

Our low-field results confirm the well-established crossover temperature for optimally doped Y-123 thin films. It is noteworthy that T_{cr} is essentially independent of the critical

current density of the Y-123 thin films, which indicates that the pinning strength does not control the crossover. T_{cr} is considerably higher for the Tl- and Hg-1212 samples, and to the best of our knowledge $T_{cr}=30$ K is the highest crossover temperature ever reported for a single *c*-axis oriented thin film, although comparable to values obtained for Y-123/P-123 multilayers.[24]

For the scaled crossover temperature T_{cr}/T_c we find 0.11, 0.19, and 0.25–0.27 for Y-123, Tl-, and Hg-1212, respectively, i.e., Y-123 is clearly less susceptible to quantum creep than Tl- and Hg-1212. It was recently shown that the irreversibility line of Tl-1212 essentially coincides with that for Hg-1212, when plotted against reduced temperature $(1\text{-}T/T_c)$, indicating that the two materials have about the same anisotropy and similar vortex pinning and creep behavior.[26] In particular, in a field of 4 T the irreversibility temperature was about 20 and 30 K for Tl- and Hg-1212, respectively, i.e., the same general temperature shift as we find for T_{cr}. It has also been shown[43,44] that low-field thermal creep proceeds via dislocations in the dilute vortex ensemble in Tl- and Hg-1212 thin films and hence has a plastic nature in these materials. In Y-123, on the other hand, the dilute vortex lattice exhibits collective flux creep via elastic deformations down to the lowest fields.[60] The question whether plastic creep might be more susceptible to quantum processes than elastic creep hence deserves more theoretical and experimental attention.

A careful look at the data in Fig. 2 reveals a second characteristic temperature, $T_{cr}^*=7$, 10, and 16 K, for the three materials, respectively, where the decrease of T/Q becomes significantly steeper. This two-stage crossover is better appreciated in Fig. 3 where we have plotted the temperature derivative of the data in Figs. 2(a), 2(c), and 2(d) normalized with the relaxation rate at T_{cr}. The data for Y1 and T1 show a distinct plateau between T_{cr} and T_{cr}^*, whereas the H1 film exhibits a more gradual slope change in this interval. We find $T_{cr}^*/T_{cr}=0.7$, 0.59, and 0.53 for the three materials. Although we do not have any good explanation for the mechanism behind this second crossover temperature, it appears to be a general feature of all the high-T_c thin films studied in this work, and should warrant more theoretical and experimental investigation.

We finally turn to the field dependence of the low-temperature creep. It is clear from the data in Fig. 2 that all samples exhibit a monotonic increase in the relaxation rate as the field is increased. Although the field dependence becomes gradually weaker at higher fields we do not observe any saturation in the field range studied. We also note that the low-field relaxation values obtained in this work are consistently smaller than what is generally observed for similar thin film samples at higher fields. The relaxation rate at T

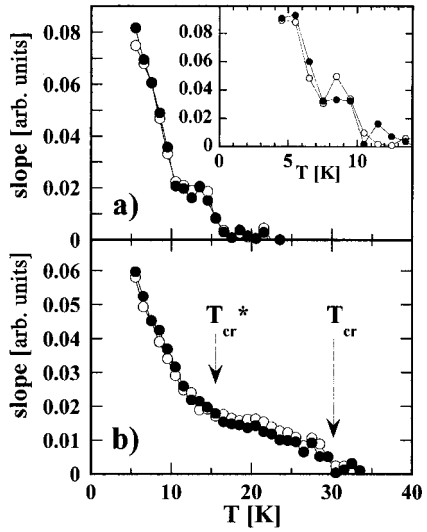

FIG. 3. Derivative of some of the T/Q data in Fig. 2. (a) T1; (b) H1. Inset: same for Y1.

$=5$ K for all samples and fields studied falls in the range $Q=0.006-0.01$, compared to $Q(T\rightarrow 0, H=1\text{ T})\sim 0.02$ of Ref. 27. There is no significant difference between different films and no important difference between the behavior above and below T_{cr} or T_{cr}^*.

VI. CONCLUSIONS

We have studied the crossover between thermal and quantum flux creep at very low vortex densities in thin film samples of Y-123, Tl-1212, and Hg-1212. The crossover temperature is found to be as high as 17 and 30 K for the two latter materials and 10–11 K for Y-123, which points to the possibility of plastic creep being more susceptible to quantum processes than is elastic creep. Both quantum and thermal flux creep are found to be suppressed as the vortex density is decreased. Our results also reveal an intriguing two-stage nature of the crossover, which might be a general property of all high-T_c materials and hence warrants more theoretical and experimental work.

ACKNOWLEDGMENTS

Fruitful discussions with B. Ivlev and J. R. Clem are gratefully acknowledged. We are indebted to S. Khartsev and A. M. Grishin for providing us with two of their Y-123 thin films. Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy under Contract No. DE-AC04-94AL85000. This research was supported by the Swedish Natural Research Council, NFR. J. J. Å. would like to thank Ivan K. Schuller for his hospitality at UCSD and for many useful discussions.

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