

Crossover from thermal to quantum flux creep in various high- T_c superconducting thin films

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AUG 17 2000

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Magnetic vortices in type-II superconductors are one of few systems in which macroscopic quantum tunneling can occur. This intriguing phenomenon was first observed in the high- T_c superconductor (HTS) $\text{YBa}_2\text{Cu}_3\text{O}_7^1$ (Y-123), and although subsequently observed in conventional superconductors^{2,3,4}, its study has mostly focused on optimally doped Y-123^{5,6,7,8,9,10,11}, oxygen-deficient Y-123¹², and other HTS materials such as $\text{Y}_{1-x}\text{Pr}_x\text{Ba}_2\text{Cu}_3\text{O}_7^{13}$, $\text{YBa}_2\text{Cu}_3\text{O}_8^{11}$, $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ and $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}^{14,15,16}$, $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_8^{17,18,19}$, $\text{Tl}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}^{20}$, and $\text{YBa}_2\text{Cu}_3\text{O}_7/\text{PrBa}_2\text{Cu}_3\text{O}_7$ multilayers^{21,22}. However, to the best of our knowledge, quantum creep has previously not been studied in neither $\text{TlBa}_2\text{CaCu}_2\text{O}_8$ nor $\text{HgBa}_2\text{CaCu}_2\text{O}_8$, which are two of the materials investigated in this work.

It was recently pointed out that the low-temperature flux creep behavior can be very different for single crystals and thin films of the same material.²³ The crossover temperature T_{cr} , below which quantum creep effects can be observed, is generally much higher for thin films than for single crystals. Thin films generally also show a strong suppression of quantum creep with decreasing field, whereas a similar anomaly has not been observed for single crystals. Both differences have been ascribed to differing characteristic pinning sites of thin films and single crystals. In particular it was 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.

In this work we determine the crossover temperatures for Y-123, Tl-1212 and Hg-1212 thin films in very low fields (7-140 Oe). While we find $T_{cr}=10$ -11K 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-1212 and Hg-1212 thin films respectively. The crossover seems furthermore to proceed in two distinct steps and a second temperature T_{cr}^* can consequently be defined for all three materials. We also find that quantum creep is suppressed down to the lowest accessible fields.

Flux creep theory

A key result within the Collective Flux Creep theory (CFC)^{24,25} is the prediction of a nonlinear current dependence of the flux creep activation energy,

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

where, J_c is the true critical current density before flux creep sets in, J_s is the decaying momentary screening current and μ is a exponent describing the degree of nonlinearity, acquiring different values depending on the actual collective flux creep regime: 1/7, 5/2 and 7/9 for single vortex, small bundle and large bundle vortex creep respectively. It is also well known²⁶ that a simpler, logarithmic current dependence, $U(J_s) = U_0 \ln(J_c/J_s)$, which formally corresponds to $\mu=0$, often is a very good description of experimental data.

In a linearly ramped external magnetic field, the momentary activation energy can be expressed as

$$U(J_s, T, H) = C k_B T \quad (2)$$

$$C = \ln \left[\frac{2v_0 H}{R(dH/dt)} \right] \quad (3)$$

where v_0 is an attempt velocity and R is the radius of the sample.²⁷ In our case of a sinusoidally varying magnetic field, $h_0 \cos \omega t$, Eq.3 is used in the form

$$C = \ln \left(\frac{v_0 H}{\pi R f} \right) \quad (4)$$

which, together with Eq.1, yields the frequency dependence of the screening current

$$J_s(T, H, f) = J_c(T, H) \left(1 + \frac{\mu C k_B T}{U_0} \right)^{-1/\mu} \quad (5)$$

By defining the dynamical relaxation rate

$$Q = \frac{d \ln J_s}{d \ln f} \quad (6)$$

one gets for the so-called *effective* activation energy

$$U_{eff} \equiv \frac{k_B T}{Q} = U_0 + \mu k_B C T. \quad (7)$$

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 be naively regarded as the sum of quantum and thermal contributions $Q(T) = Q_{th}(T) + Q_{qc}(T)$, where $Q_{qc}(T)$ 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 hence possible to define a crossover temperature below which quantum creep is observable.

Q from ac susceptibility measurements

The ac susceptibility technique is uniquely well suited for low-field flux creep studies in superconducting thin films. As recently demonstrated^{28,29}, the field and temperature dependent dynamical relaxation rate Q can be conveniently determined from temperature scans of $\chi'(T, H, f)$ at a set of different fields and frequencies. For a thin disk sample in a sufficiently large ac field, $h_0 \cos \omega t$, the critical current density and the measured in-phase susceptibility are related by

$$J_c = \left[\frac{-\chi'}{1.33\chi_0} \right]^{2/3} \frac{2h_0}{d}, \quad (8)$$

where χ_0 is the full screening susceptibility, determined at low ac fields, and d is the sample thickness.³⁰ The dynamical relaxation rate can hence be determined as a function of temperature, ac and dc field from

$$Q(T, H) = \frac{2}{3} \frac{d \ln |\chi'(T, H, f)|}{d \ln f}. \quad (9)$$

A noncircular sample shape will introduce a small multiplicative term in Eq.8 that will not survive the logarithmic differentiation in Eq.9, which hence remains valid for any thin film sample.

Experimental

Two c-axis oriented Y-123 thin films, one on a SrTiO₃ substrate (Y-1) and another on a LaAlO₃ substrate (Y-2), with nominal thickness of 200 nm, were prepared, in the same deposition run, by pulsed laser ablation at 750°C in 200 mTorr O₂. The deposition was followed by an in-situ anneal in 600 Torr O₂ atmosphere at 430°C for 30 minutes. $T_{c0}=89.5\text{K}$ of both films was determined from usual four-contact resistivity measurements.

The 200 nm thick Tl-1212 film was grown on a LaAlO₃(100) substrate using a two-step method reported in detail elsewhere³¹. 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 minutes at 825°C in 630 Torr O₂. X-ray diffraction found these films to be phase-pure and c-axis oriented. $T_c=88\text{K}$ and $J_c(5\text{K})=1 \times 10^7\text{ A/cm}^2$ 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 SrTiO₃ substrates, followed by annealing in a Hg-vapor atmosphere at 820°C for 30 minutes. The higher-quality sample, Hg-1, with $T_c=120\text{K}$, showed mainly (00*l*) lines of the c-axis oriented Hg-1212 phase with minute traces of c-axis Hg-1223. Sample Hg-2 ($T_c=110\text{K}$) showed additional, relatively strong, unidentified impurity lines.

Fundamental frequency sine-wave integrated in-phase ac susceptibility measurements, $\chi_1'(T, H_{ac}, f)$, were carried out using a home-built high-sensitivity ac susceptometer with a three-coil mutual inductance bridge and a background subtraction scheme.³² ac fields, applied normal to the film plane, with frequencies between 12.7 and 181 Hz, ranged from $H_{ac}=7-140\text{ Oe rms}$ ($H_{ac}=2^{-1/2}h_0$).

Results and discussion

The frequency dependence of χ' for sample Y-1 at different temperatures is shown in fig. 1. The straight lines are fits to a power law $\chi' \propto f^m$ from which $Q=m/1.5$ can be determined.²⁸ The good quality of the fit indicates that possible self-heating due to hysteretic losses in the samples is too small to be of any problem. 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 hence conclude that any generated heat can not be at the origin of the observed relaxation at low temperatures.^{23,33,34}

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.35. The temperature independent behaviour of the Tl-1212 and Hg-1212 samples on the other hand 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 $T_{cr}=10-11\text{K}$ for the Y-123 films, $T_{cr}=17\text{K}$ for the Tl-1212 film and $T_{cr}=30\text{K}$ for the Hg-1212 films. No detectable field dependence of T_{cr} is observed in the field range studied. Our low-field results hence confirms the well-established crossover temperature for optimally doped Y-123 thin films. T_{cr} is however considerably higher for the Tl-1212 and Hg-1212 samples, and to the best of our knowledge $T_{cr}=30\text{K}$ is the highest crossover temperature ever reported for a single thin film, although comparable to values obtained for Y-123/P-123 multilayers²¹. A careful look at the data reveals a second characteristic temperature, T_{cr}^* , where the decrease of T/Q becomes significantly steeper. This two-stage crossover is better appreciated in a plot of the temperature derivative of the data in fig.2a,c&d normalized with the relaxation rate at T_{cr} (fig.3). The data for Y-123 and Tl-1212 show a distinct plateau between T_{cr} and T_{cr}^* , whereas the Hg-1212 film exhibits a more gradual slope change in this interval. 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 (fig.4). 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 $T=5\text{K}$ for all samples and fields studied falls in the range $Q=0.006-0.01$, compared to $Q(T>0, H=1\text{T})\sim 0.02$ of ref.23. There is no

significant difference between different films and no important difference between the behavior above and below T_{cr} or T_{cr}^* .

Acknowledgment

Fruitful discussions with B. Ivlev are gratefully acknowledged. Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of energy under Contract DE-AC04-94AL85000. This research is supported by the Swedish Natural Research Council, NFR.

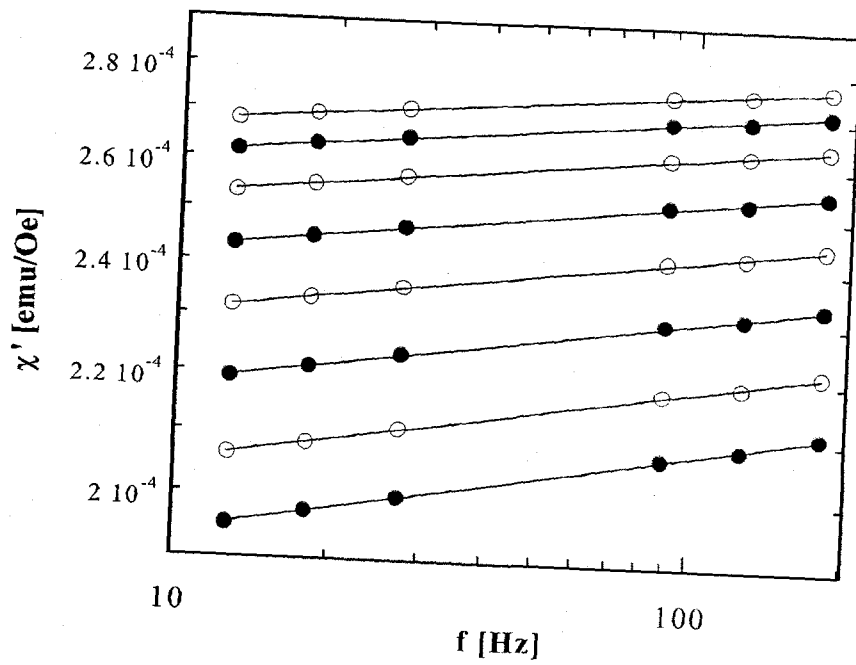


Fig. 1 log-log plot of $-\chi'$ vs frequency for sample Y-1 at $T=5, 7, 9, 11, 13, 15, 17$ and 19K . The straight lines are power law fits to the data.

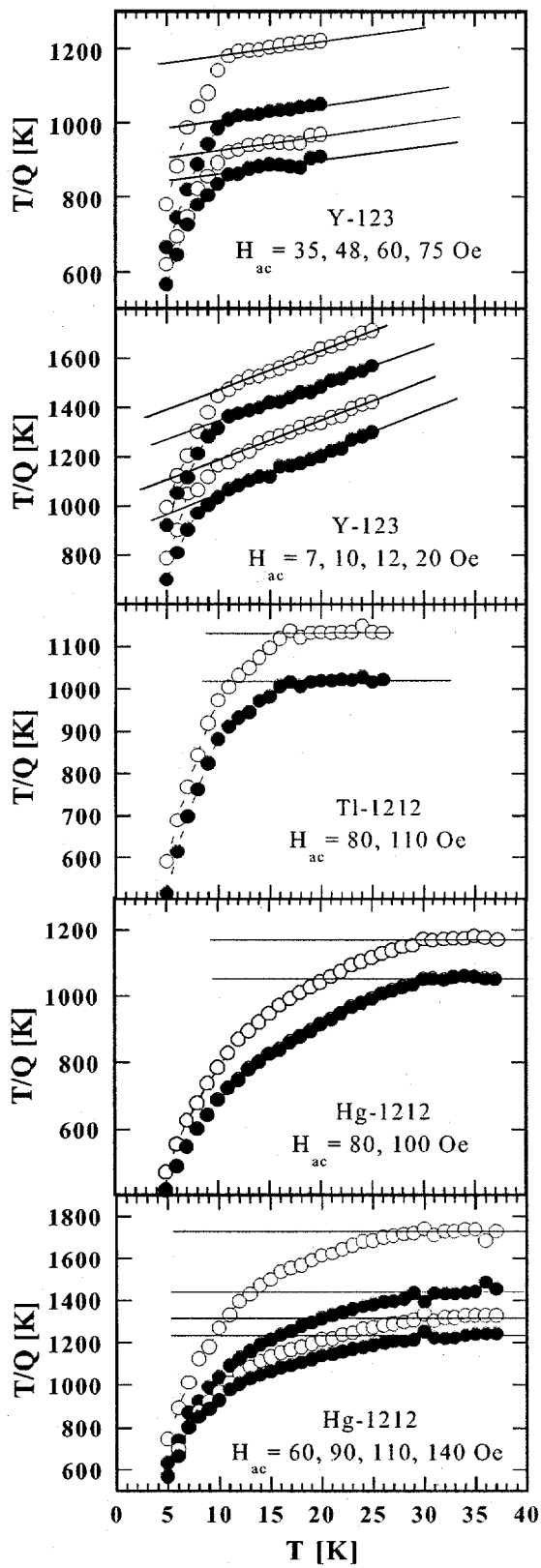


Fig. 2 Effective activation energy vs temperature and ac field for the five samples.

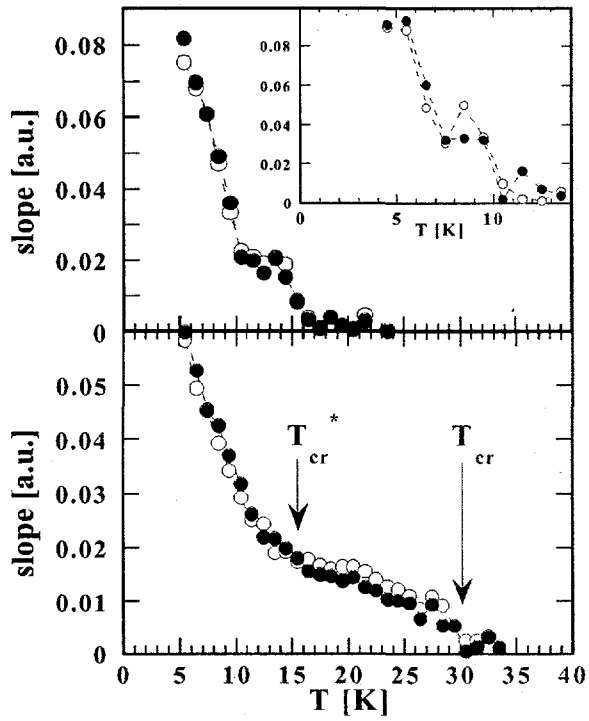


Fig. 3

Derivative of the T/Q data in fig.2 for a) Tl-1212, b) Hg-1212
 Inset: same for Y-123.

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