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Dislocation-mediated creep of highly separated vortices in *a*-axis-oriented $HgBa_2CaCu_2O_{6+\delta}$ thin films

Johan J. Åkerman*
Department of Materials Science-Tmfy-MSE, Royal Institute of Technology, S-100 44 Stockholm, Sweden
and Physics Department, University of California–San Diego, La Jolla, California 92093-0319

S. H. Yun and U. O. Karlsson
Department of Materials Physics, Royal Institute of Technology, S-100 44 Stockholm, Sweden

K. V. Rao
Department of Materials Science-Tmfy-MSE, Royal Institute of Technology, S-100 44 Stockholm, Sweden

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Using ac susceptibility, we determine the critical current density J_c and the flux creep activation energy U of an *a*-axis-oriented $HgBa_2CaCu_2O_{6+\delta}$ thin film. The critical current density at helium temperatures is found to be 4.6×10^4 A/cm^2, i.e., about two orders of magnitude smaller than for corresponding films with *c*-axis orientation. The temperature and ac field dependent activation energy is consistent with dislocation-mediated flux creep and well described by $U(T,H_{ac})=U_0(1-t^4)H_{ac}^{-1/2}$ with $t=T/T_c$, $T_c=120$ K, and $U_0=0.77$ eV Oe$^{1/2}$ for temperatures $T>45$ K and in the field range studied. The activation energy is of the same order as that found in *c*-axis-oriented films. Below $T=45$ K the activation energy is observed to decrease as thermally assisted quantum creep becomes increasingly important.

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I. INTRODUCTION

The pronounced layered nature of high-T_c superconductors (HTS) gives rise to a strong anisotropy of their superconducting properties. The fabrication and experimental study of HTS thin films of different crystallographic orientation is hence important both from a fundamental as well as an applications point of view. Due to their longer coherence length normal to the film plane, *a*-axis-oriented thin films present several advantages for superconductor-insulator-superconductor and superconductor-normal metal-superconductor trilayer Josephson junction applications.[1–4] While an overwhelming majority of experimental studies have been carried out on *c*-axis-oriented films of different materials, the rather few studies of the functional properties of *a*-axis films have been mainly limited to $YBa_2Cu_3O_{7-x}$(Y-123).[5–8] Although the fabrication of *a*-axis-oriented $(Hg_{1-x}Re_x)Ba_2CaCu_2O_{6+\delta}$ thin films has been reported,[9] it is only recently that *a*-axis films of $HgBa_2CaCu_2O_{6+\delta}$(Hg-1212) with high-T_c and good superconducting properties have been available.[10]

In this paper we present a study of the low-field vortex dynamics of an *a*-axis-oriented Hg-1212 thin film using ac susceptibility measurements of the temperature, field, and current dependent flux creep activation energy $U(T,H_{ac}J)$. Because of its limited critical current density, the film enters a fully penetrated Bean critical state for fields as low as 0.15 Oe. We have hence been able to study the dynamics of vortices that, on average, are separated by more than 10 μm. Our results are well described by a model for dislocation-mediated plastic flux creep. We also observe an additional creep mechanism possibly due to thermally assisted quantum processes up to temperatures as high as 45 K.

II. ac SUSCEPTIBILITY METHOD

The versatility of ac susceptibility measurements, as a probe for the study of flux creep, has recently been demonstrated on *c*-axis-oriented Hg-1212 and Y-123 thin films.[11,12] Thanks to the simple functional relation[13] between the critical current density J_c and the *real* part χ' of the ac susceptibility,

$$\chi' = -0.47\chi_0(J_c d/h_0)^{3/2}, \quad h_0 \gg J_c d \tag{1}$$

with $h_0 \cos \omega t$ being the external ac field, d the film thickness, and $-\chi_0$ the full screening susceptibility, it is straightforward to determine J_c as a function of temperature, ac and dc field, and frequency.

The frequency dependence of J_c is a consequence of the vortex density gradient relaxing during a time period $1/f$ of the applied ac field. This dependence is commonly analyzed within the critical state model by the expression[14]

$$J_c(T,H,f) = J_{c0}(T,H)\,g[k_B T \ln(f_0/f)/U(T,H)], \tag{2}$$

which separates the original critical current density $J_{c0}(T,H)$ before relaxation processes set in, and the actual relaxation due to thermal flux creep, expressed via a renormalization function $0<g(y)<1$. In the logarithmic approximation[15] $U(J_c)=U_0 \ln(J_{c0}/J_c)$, which is often taken as an indication of single-vortex creep,[16] the renormalization function becomes[17] $g(y)=\exp(-y)$, which together with Eq. (2) yields a power-law dependence

$$J_c(f) = J_{c0}(f/f_0)^{k_B T/U(T,H)}. \tag{3}$$

Using Eq. (1), the so-called dynamical relaxation rate[12,18] can then be calculated from the frequency dependent in-phase susceptibility

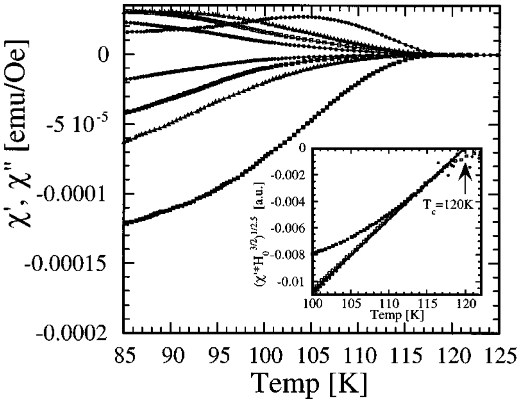

FIG. 1. χ' and χ'' vs T in ac fields $H_{\mathrm{ac}}=0.15-2$ Oe at $f=181$ Hz. The inset shows a plot of $(\chi' H_{\mathrm{ac}}^{3/2})^{1/2.5}$ vs T.

$$Q \equiv \frac{d\ln J_c}{d\ln f} = \frac{2}{3}\,\frac{d\ln|\chi'|}{d\ln f}. \tag{4}$$

Provided Eq. (3) holds, the activation energy is then given directly by $U = k_B T/Q$.

III. EXPERIMENT

The fabrication of a-axis-oriented Hg-1212 thin films has been described in detail in Ref. 10. Briefly, a quartz tube, with a rf-sputtered $\mathrm{Ba_2CaCu_2O}_x$ precursor film on a (100) $\mathrm{LaAlO_3}$ substrate, two nonreacted stoichiometric Hg:Ba:Ca:Cu=1:2:1:2 pellets and one stoichiometric Ba:Ca:Cu=2:1:2 pellet, is evacuated and heated up to 800 °C using a high ramping rate. The precursor film is annealed at 800 °C for 5 min, cooled down to 700 °C and finally quenched to room temperature.

With the exception of small traces of c-axis-oriented Hg-1212 and Hg-1223, x-ray diffraction patterns of the film studied in this paper showed mainly sharp peaks that were identified as (l00) reflections of any of the Hg-based homologs. X-ray pole figure ϕ scans showed dominantly (102) and (110) reflections of the Hg-1212 phase with a fourfold symmetry, hence indicating a single a-axis-oriented Hg-1212 phase (98% volume fraction) with the c axis in two different in-plane orientations at 90° to each other, the so-called a-c twins. The twinned microstructure was confirmed using scanning electron microscopy, where a well-connected network of on average 1-μm-wide and 10-μm-long grains could be clearly observed.[10] The film was of rectangular shape, with sides 5.4 and 1.8 mm, respectively, and had a thickness of 1 μm.

Usual sine-wave integrated ac susceptibility, measurements of χ_1' and χ_1'' were carried out in a custom-built high sensitivity ac susceptometer.[19,20] Root-mean-square ac fields, $(H_{\mathrm{ac}} = h_0/\sqrt{2})$, applied normal to the film plane, ranged from $H_{\mathrm{ac}}=0.15$ to 5 Oe at frequencies from 1.81 to 891 Hz.

IV. RESULTS AND DISCUSSION

A typical measurement at four different ac fields $H_{\mathrm{ac}}=0.15$, 0.6, 1, and 2 Oe is shown in Fig. 1. The strong diamagnetic signal, comparable to similarly sized c-axis-oriented films, indicates that the a-c twinned network acts as a continuous film rather than a collection of decoupled superconducting grains. Due to its large demagnetizing factor, a continuous film screens out a large volume above and below its plane, which leads to the large ac susceptibility response. Decoupled grains, on the other hand, would essentially only screen out their own volume, hence resulting in a much weaker signal (orders of magnitude). We can hence base our ac susceptibility analysis on Sanchez' and Clem's model for a continuous film.[13]

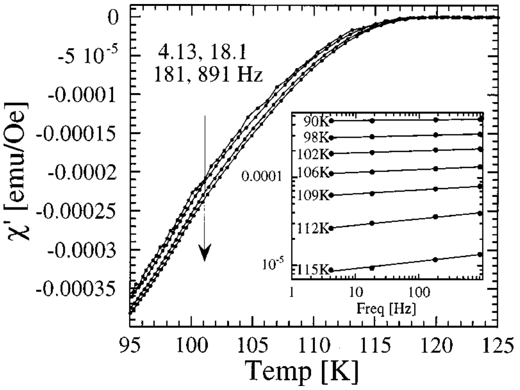

FIG. 2. χ' vs T for a number of frequencies $f=4.13-891$ Hz in an ac field of $H_{\mathrm{ac}}=0.3$ Oe. In the inset is shown a log-log plot of χ' vs f at different temperatures.

In the inset we have plotted $(\chi' H_{\mathrm{ac}}^{3/2})^{1/2.5}$ vs temperature. As discussed in Ref. 11, the overlap of all the data onto a straight line (inset) confirms the validity of Eq. (1) and suggests that, on approaching $T_c = 120$ K, the critical current density depends on temperature as $J_c(T) \propto (1-t)^{1.7}$ and $t = T/T_c$. For the smallest ac field $H_{\mathrm{ac}}=0.15$ Oe the data is seen to deviate from the straight line at about 113 K as H_{ac} becomes comparable to $J_c d$. At $T = 4.2$ K the same film has a loss maximum for $H_{\mathrm{ac}}=4$ Oe, which, using the thin disk expression[13] $J_c(T_p) = 1.46 \times H_{\mathrm{ac}}/d$, yields an estimate for the critical current density $J_c(4.2\,\mathrm{K}) = 4.6 \times 10^4\ \mathrm{A/cm^2}$. Compared to c-axis-oriented Hg-1212 thin films,[11] the critical current density hence has a similar temperature dependence but an absolute value that is about two orders of magnitude smaller. A similar strong anisotropy of the critical current density has been reported in studies on b-c twinned a-axis-oriented $\mathrm{YBa_2Cu_3O}_{7-x}$ thin films.[1,21]

It is interesting to note that there is no substantial reduction of the loss maximum with decreasing ac field, which means that the critical state model is valid even for ac fields as low as $H_{\mathrm{ac}}=0.15$ Oe. In particular, this result implies that surface barriers are not important in impeding the entrance of vortices parallel to the CuO planes and that $H_{c1,a}$ can be neglected in the field range studied. This contrasts to results obtained on c-axis-oriented Hg-1212 thin films of similar thickness where a true critical state was only realized in ac fields above 3 Oe (Ref. 11), and indicates that the anisotropy of Hg-1212 is at least $\gamma = H_{c1,c}/H_{c1,a} \approx 20$, in agreement with $\gamma = 6.7-77$ reported in the literature.[22,23]

Figure 2 shows a temperature scan where consecutive measurements of χ' are made at four different frequencies,

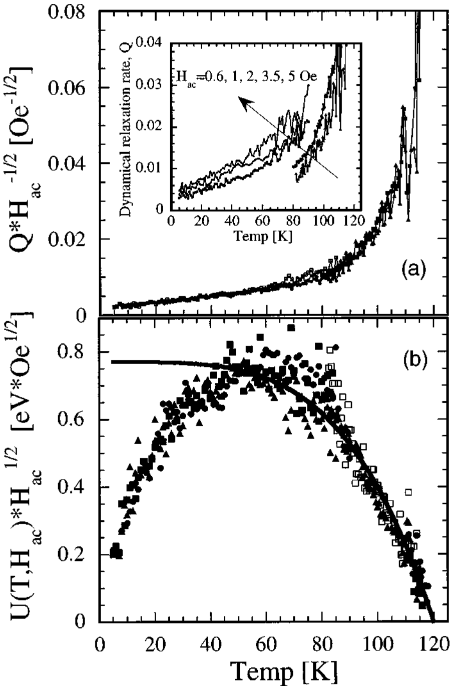

FIG. 3. (a) Dynamical relaxation rate Q plotted as $QH_{\mathrm{ac}}^{-1/2}$ vs temperature. In the inset is shown the original data for $H_{\mathrm{ac}}=0.6-5$ Oe. (b) Flux creep activation energy U plotted as $UH_{\mathrm{ac}}^{1/2}$ vs temperature, together with a fit $U(T,H_{\mathrm{ac}})=U_0(1-t^4)H_{\mathrm{ac}}^{-1/2}$ with $U_0=0.77$ eV Oe$^{1/2}$ and $T_c=120$ K.

$f=4.13$, 18.1, 181, and 891 Hz, in an ac field of $H_{\mathrm{ac}}=0.3$ Oe. The frequency dependence of the critical current density is seen as a slight increase of $|\chi'|$ with increasing frequency. To analyze this dependence in more detail, we make isothermal cuts in Fig. 2 and plot $|\chi'|$ vs f on log-log scales as seen in the inset. The straight lines are power-law fits of

$$\chi'(T,H_{\mathrm{ac}},f)=P(T,H_{\mathrm{ac}})f^{m(T,H_{\mathrm{ac}})}, \tag{5}$$

where the fitted exponent $m(T,H_{\mathrm{ac}})$ is used to calculate the dynamical relaxation rate $Q=m/1.5$, as well as the activation energy $U=1.5k_BT/m$. In the inset of Fig. 3(a), we plot Q as a function of temperature for ac fields $H_{\mathrm{ac}}=0.6-5$ Oe. Since the Earth's field is not shielded during the measurement, we only analyze data for $H_{\mathrm{ac}}\geqslant 0.6$ Oe in the following, as the field dependence has previously been found to depend solely on the ac field provided $H_{\mathrm{ac}}\geqslant H_{\mathrm{dc}}$.[12] As expected for thermally activated single-vortex creep, Q shows a monotonous increase with increasing temperature and diverges as $T_c=120$ K is approached. Q also exhibits a square-root ac field dependence, which is emphasized in Fig. 3(a) by plotting $QH_{\mathrm{ac}}^{-1/2}$ vs T. Such a field dependence is expected for dislocation-mediated flux creep where vortices propagate via local energy minima in the flux line lattice[24,25] and has been observed at considerably higher fields in several anisotropic superconductors such as $\mathrm{Bi_2Sr_2CaCu_2O_{8+x}}$ (Bi-2212) thin films, $\mathrm{Tl_2Ba_2CaCu_2O_8}$ (Tl-2212) single crystals,[26,27] and $\mathrm{Tl_{2/3}Ba_{1/3}Sr_2CaCu_2O_7}$ (Tl-1212) single crystals.[27] Initially, a half-loop is created on the vortex. It then extends so as to reach an adjacent energy minimum, typically at a distance of a fraction δ of the average lattice constant a_0, and finally stretches out until the entire vortex ends up in the new position. The energy barrier associated with this vortex motion is roughly given by the creation energy for a vortex-antivortex pair perpendicular to the original vortex line[28]

$$U=\frac{2\Phi_0^2}{4\pi\mu_0\lambda_{ab}\lambda_c}\ln(\kappa)\,\delta a_0, \tag{6}$$

where the average lattice spacing is $a_0=(4/3)^{1/4}(\Phi_0/B)^{1/2}$ for a triangular flux-line lattice, hence the square-root field dependence.

According to Eq. (6) the temperature dependence of the activation energy should be given by $\lambda^{-2}(T)=\lambda^{-2}(0)(1-t^4)$. In Fig. 3(b) we plot the activation energy as $UH_{\mathrm{ac}}^{1/2}$ vs T together with a one-parameter theoretical fit $U(T,H_{\mathrm{ac}})=U_0(1-t^4)H_{\mathrm{ac}}^{-1/2}$ with $U_0=0.77$ eV Oe$^{1/2}$. From published values of the superconducting parameters for the Hg-1212 material system, $\lambda_{ab}(0)=209$ nm (Ref. 29), $\kappa=126$ (Ref. 29), and $\gamma=67$ (Ref. 22), we get an estimate of $U_0\delta^1=27$ eV Oe$^{1/2}$, which suggests that a typical vortex jump is only about 3% of the average vortex spacing. It is noteworthy that vortices separated by up to 6 μm still cannot be regarded as isolated, since vortex-vortex interactions, typically extending about $\lambda_c=\gamma\lambda_{ab}\approx 14\,\mu$m, are important enough to yield a square-root field dependence.

The fit is valid down to about 45 K, where a clear deviation occurs and continues to the lowest temperatures. One also sees in Fig. 3(a) that the relaxation does not extrapolate to zero at $T=0$ K but rather tends towards a constant value of about 0.0035 Oe$^{1/2}$. A similar finite relaxation rate on approaching absolute zero has been observed in many other superconducting materials and is generally ascribed to quantum creep.[30–38] For Y-123 thin films, the crossover temperature from thermal to quantum creep T_{cr}, is generally found to increase with the average vortex separation, i.e., with decreasing field, in qualitative agreement with theory.[39] Y-123/Pr-123 multilayers showed evidence of so-called thermally assisted quantum creep (TAQC) up to $T_{\mathrm{cr}}\approx 15$ K in a field of 8 T and as the vortex separation was increased $(B=2\text{ T})T_{\mathrm{cr}}$ was found to increase to about 30 K.[40] Our results, obtained in very low fields, suggest $T_{\mathrm{cr}}=45$ K for highly separated vortices in a-axis-oriented Hg-1212 thin films. To our knowledge, this is the highest crossover temperature ever reported for any superconductor and illustrates that the dynamics of such highly dilute vortex ensembles can be drastically different from denser ones. The recently discovered[41] two-stage nature of the low-field thermal-to-quantum crossover in c-axis-oriented Y-123, T-1212, and Hg-1212, can also be observed in the a-axis-oriented film as a slight slope change in $U(T)$ at about $T_{\mathrm{cr}}^{*}=25$ K. We also note that TAQC in this field range is proportional to the average vortex separation a_0.

V. COMPARISON OF *a*-AXIS AND *c*-AXIS ORIENTATION

A number of interesting comparisons can be made with *c*-axis-oriented thin films of the same material system. The overall behavior of the activation energy is rather similar.[19] In particular, in both orientations, Hg-1212 thin films exhibit an inverse square-root ac field dependence, i.e., U is proportional to the average vortex separation during the ac field period. The fitted low-temperature value of $U_0 = 0.77\,\text{eV}\,\text{Oe}^{1/2}$ is also only slightly smaller than $0.91-1.2\,\text{ev}\,\text{Oe}^{1/2}$, which was found for *c*-axis-oriented films. As expected, at these low-field strengths, we hence see no increase of U_0 due to possible intrinsic pinning. TAQC of similar magnitude is also observed for both orientations, although $T_{\text{cr},a\ \text{axis}} = 45\,\text{K}$ and $T^*_{\text{cr},a\text{axis}} = 25\,\text{K}$ is significantly higher than $T_{\text{cr},c\text{axis}} = 35\,\text{K}$ and $T^*_{\text{cr},c\text{axis}} = 15\,\text{K}$.[41] We note however that $T^*_{\text{cr}}/T_{\text{cr}} = 0.53-0.55$ for both orientations. *c*-axis-oriented films have been shown to exhibit a clear flux creep maximum at a temperature T_{cm}, whereas Q for the *a*-axis-oriented film, studied in this paper, diverges on approaching T_c. As T_{cm} in *c*-axis-oriented films has been attributed to the onset of important vortex-vortex interactions, which abruptly increases the activation energy, we argue that the linear decrease of U towards zero at $T_c = 120\,\text{K}$, indicates that flux creep in the *a*-axis-oriented film remains single-vortexlike at all temperatures in the field range studied.

VI. CONCLUSION

We have carried out a detailed low-field flux creep study on an *a*-axis-oriented Hg-1212 thin film. The critical current density at helium temperature is $J_c(4.2\,\text{K}) = 5.8 \times 10^4\,\text{A/cm}^2$, i.e., approximately two orders of magnitude smaller than for *c*-axis-oriented Hg-1212 thin films. We find that the temperature and ac field dependent flux creep activation energy above $T = 45\,\text{K}$, is well described by a model for dislocation-mediated flux creep, $U(T,H_{\text{ac}}) = U_0(1-t^4)H_{\text{ac}}^{-1/2}$, with $T_c = 120\,\text{K}$ and $U_0 = 0.77\,\text{eV}\,\text{Oe}^{1/2}$. As a consequence vortices separated by about 6 μm still cannot be regarded as isolated. The low value for U_0 implies that a typical vortex jump is only a fraction 0.03 of the average lattice constant. Below $T_{\text{cr}} = 45\,\text{K}$, the apparent activation energy U is further reduced because of additional quantum creep, and even more so at the second crossover temperature $T^*_{\text{cr}} = 25\,\text{K}$. At these large vortex separations, intrinsic pinning does not seem effective.

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[*] Email address: jjonsson@ucsd.edu

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