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Low-field vortex dynamics in various high-T_c thin films

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Abstract. We present a novel ac susceptibility technique for the study of vortex creep in superconducting thin films. With this technique we study the dynamics of dilute vortices in c-axis oriented Y-123, Hg-1212, and Tl-1212 thin films, as well as a-axis oriented Hg-1212 thin films. Results on the Hg-1212 and Tl-1212 thin films indicate that dislocation-mediated plastic flux creep of single vortices dominates at low temperatures and fields. As the temperature (or the field) is increased, the increasing vortex–vortex interactions promote a collective behavior, which can be characterized by elastic creep with a non-zero μ exponent. Also, in some of these samples effects of thermally assisted quantum creep are visible up to 45 K in some of these samples. In Y-123 thin films, creep is found to be collective down to the lowest temperatures and fields investigated, while the quantum creep persists only up to 10–11 K.

Keywords. Vortex dynamics; flux creep; high temperature superconductor; thin film; ac susceptibility.

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1. Introduction

Despite the predictions of fundamental new physics and intriguing vortex phenomena in high-T_c materials at very low fields [1–6], a vast majority of experimental vortex dynamics studies focus on the high-field properties, and only some recent investigations [7–10] have studied the low-field regime in any detail. One important reason is the lack of a fast, sensitive and accurate technique to measure flux creep at low vortex densities. Traditional relaxation measurements typically require hour-long waiting times at a single temperature, during which the applied field has to be stable within less than 0.1% to avoid perturbing the critical state under study [11]. To ease the field stability requirement, one may intentionally ramp the field up and down a value $\pm\Delta H$ about the dc field of interest and study the relaxation through the dependence of the minor loop height on the ramp rate, typically in the range 0.2–400 Oe/s [12]. ac Susceptibility has also been widely used, in particular to study the dependence of the χ'' peak temperature (T_p) on the frequency (f) [13]. However, this approach only retains a *single* data point, $T_p(f)$, out of an entire temperature scan, and the detailed analysis is further complicated by the fact that extracted values for the activation energy are averaged over the temperature range given by $T_p(f)$.

In this work we present a novel ac susceptibility technique which improves the amount of data that can be obtained for a given time up to 1–2 orders of magnitude. The technique is very sensitive, over a wide field range from 0.1 Oe to several Tesla, and lends itself to detailed studies of subtle changes in both the temperature dependence and the field dependence of flux creep. Using this technique we study the low-field vortex dynamics in Y-123, Hg-1212 and Tl-1212 thin films. While collective elastic flux creep persists down to the lowest accessible fields in Y-123, a novel regime of dilute dislocation-mediated plastic flux creep is observed in the more anisotropic materials. Whereas this regime dominates at all temperatures in *a*-axis oriented Hg-1212 thin films, collective effects appear in *c*-axis films as either the field or the temperature is increased. Onset of quantum creep is observed in all samples below a characteristic temperature ranging from 10 to 45 K.

2. Experimental

50-nm-thick *c*-axis oriented Y-123 thin films with $T_c = 89.5$ K were deposited by laser ablation on $LaAlO_3$ (Y-1), as well as $SrTiO_3$ substrates (Y-2) [10]. In these films the critical current densities at 5 K were found to be $1.2 * 10^6$ A cm^{-2} (Y-1) and $1.2 * 10^7$ A cm^{-2} (Y-2) respectively. Fabrication of 400-nm-thick *c*-axis oriented Hg-1212 thin films involved the deposition of a Hg-free precursor film on $SrTiO_3$ substrates followed by annealing in a Hg-vapour atmosphere [14]. Two samples, H-1 and H-2, with $T_c = 110$ K and 120 K respectively, were chosen for this study. In both these samples the critical current densities at 5 K were found to be $5 * 10^6$ A cm^{-2}. A similar two-step method was used for the fabrication of the 200-nm-thick *c*-axis oriented Tl-1212 film (T-1) with $T_c = 88$ K and J_c (5 K) $= 1 * 10^7$ A cm^{-2} [15]. *a*-axis oriented Hg-1212 films can be obtained by rapidly quenching the film to room temperature after the anneal in Hg-vapor [16]. The 1000-nm-thick film (H-a) studied in this work had a optimal T_c of 120 K and a typical low critical current of $4.6 * 10^4$ A cm^{-2}. Fundamental frequency sine-wave integrated in-phase ac susceptibility measurements were carried out in a custom built high-sensitivity ac susceptometer [17].

3. Results and discussions

Shortly after the problem was solved of how to apply the critical-state (CS) model [18] to the thin circular disk geometry [19,20], analytical expressions for the corresponding ac susceptibility were derived [21]. It was subsequently shown that due to the extreme demagnetizing factor of the perpendicular geometry the particular in-plane shape does not significantly influence the ac response ($< 0.2\%$) [22], which hence greatly reduces the need for patterning films prior to measurement. In the limit of large ac amplitudes, the in-phase ac susceptibility of a thin disk is given by [21] $\chi' = -0.47\chi_0(J_c d/h_0)^{3/2}$, $h_0 \gg J_c d$, with $h_0 \cos 2\pi f t$ being the external ac field, d the film thickness and $-\chi_0$ the full screening susceptibility. This simple asymptotic relation is shown in figure 1 where top portion shows the in-phase susceptibility of sample H-2 as a function of temperature and at four different ac fields, $H_{ac} = 7$, 10, 15, and 20 Oe, and bottom portion reveals a clear overlap of the data when scaled according to eq. (1), i.e., $\chi' H_{ac}^{3/2}$ vs. T. Assuming a temperature dependent critical current density, $J_c(T) \propto (1 - T/T_c)^\beta$ one expects a straight line when plotting $(\chi' H_{ac}^{3/2})^{2/3\beta}$. This is indeed observed in the inset for a choice of $\beta = 1.7$, where furthermore

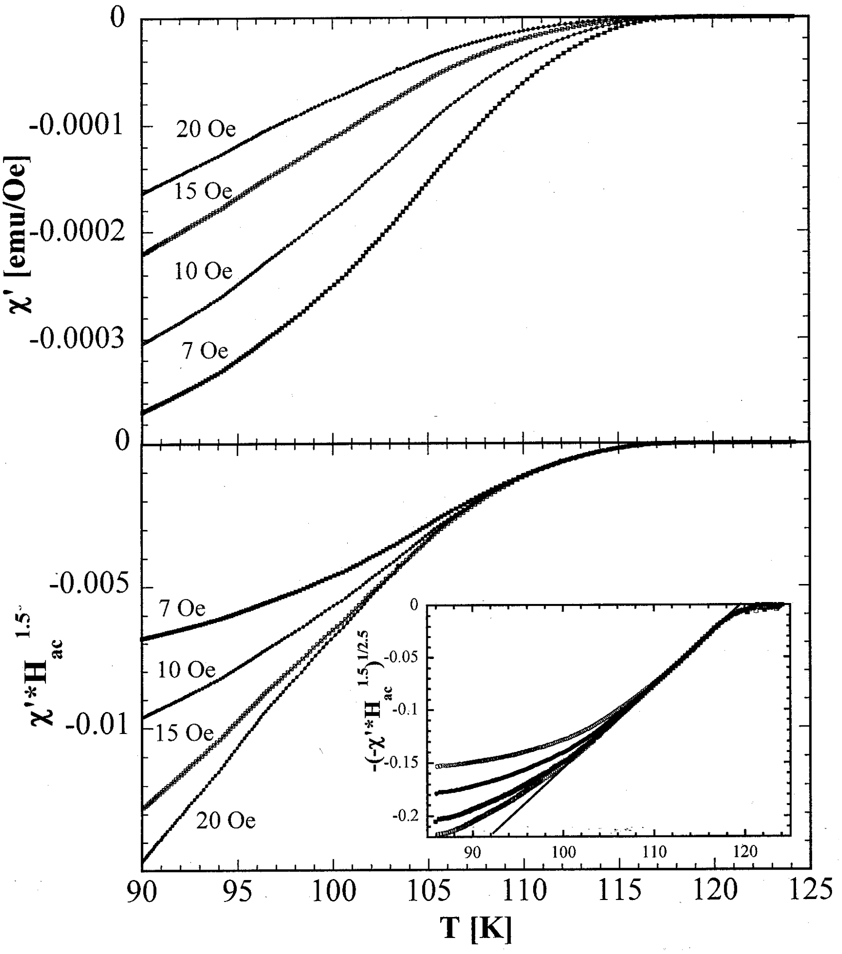

Figure 1. (**Top**) Raw data of χ' vs. T for $H_{\mathrm{ac}} = 7$, 10, 15, 20 Oe. (**Bottom**) Plotting $\chi' H_{\mathrm{ac}}^{3/2}$ vs. T collapses all data onto a single curve in a temperature range $\Delta T_{\chi'}(H_{\mathrm{ac}})$ that increases with H_{ac}. (**Inset**) Plotting $(\chi' H_{\mathrm{ac}}^{3/2})^{1/2.5}$ vs. T yields a straight line.

T_{c} can be conveniently found from the intersection of the straight line with the temperature axis. The proportionality $\chi' \propto J_{\mathrm{c}}^{3/2}$ will be used in the following to directly relate the measured ac susceptibility with the critical current density.

Flux creep effectively leads to a frequency dependent critical current density. To quantify the amount of flux creep and more easily compare frequency dependent measurements with ordinary time dependent relaxation measurements, one defines the so-called dynamical relaxation rate [23] as $Q = \mathrm{d}\ln J_{\mathrm{c}}/\mathrm{d}\ln f = (2/3)\mathrm{d}\ln|\chi'|/\mathrm{d}\ln f$. Within the collective flux theory one can show that Q is related to the effective flux creep activation, U_{eff}, and the average microscopic flux creep activation energy U_0, through $k_{\mathrm{B}}T/Q = U_{\mathrm{eff}} = U_0 + \mu C k_{\mathrm{B}} T$, where μ is an exponent with a value that measures the vortex bundling; $\mu = 1/7$ for single

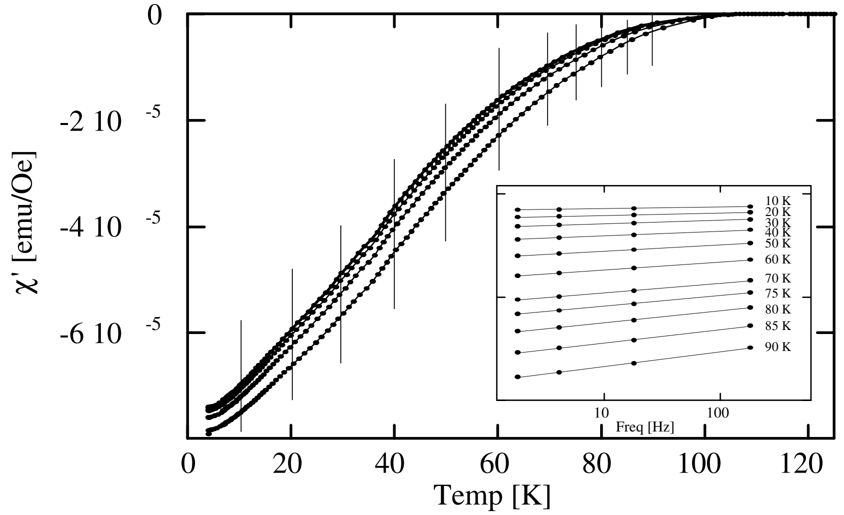

Figure 2. χ' vs. T for H-2 in $H_{\mathrm{ac}} = 7$ and $f = 1.81$, 4.13, 18.1, and 181 Hz. **Inset:** Log-log plot of χ' vs. f for the same film with data from the isothermal cuts in the main figure. Straight lines are fits to $\chi' \propto f^m$.

vortex creep, 5/2 for small bundle creep, and 7/9 for large bundle creep. For a non-zero μ 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 [24]. Experimentally, a non-zero μ is easily detected as an increase in U_{eff} with increasing T. In the case of $\mu = 0$, $k_{\mathrm{B}}T/Q$ directly yields the average microscopic activation energy U_0. A flat temperature dependence of U_{eff} is hence an indication of $\mu = 0$. In most experimental studies a drop in U_{eff} is observed below a certain temperature, which indicates the onset of quantum creep [10,12,25]. Quantum creep effectively introduces a second relaxation path that does not vanish as $T \to 0$, hence the drop in the apparent U_{eff}.

Figure 2 depicts the actual evaluation carried out experimentally. In a single warm-up scan χ' is measured at four different frequencies, $f = 1.81$, 4.13, 18.1 and 181 Hz. The effect of flux creep can be clearly seen as an increase in $|\chi'|$ with increasing f. The data taken at the isothermal cuts, marked by straight lines in figure 2, are plotted vs. f in a log-log plot in the inset. The straight lines in the inset are power-law fits, $\chi' \propto f^m$, from which $Q = m/1.5$ and $U_{\mathrm{eff}} = 1.5k_{\mathrm{B}}T/m$ are determined as a function of the field used and the temperature of the data cut.

In figure 3 we show the effective activation energy for sample Y-1 (a) and Y-2 (b). As is typical for the Y-123 system, U_{eff} *increases* quasilinearly with T at both low and intermediate temperatures, which suggests a finite μ and is consequently a strong indication of collective flux creep. The steep drop of U_{eff} at 10–11 K for both samples is characteristic of the onset of quantum creep, which eventually dominates as the temperature is further decreased. Above this temperature range, Y-1 exhibits two linearly temperature dependent regimes, from which we extract the slope $\mu C = 16.4$ and 37 respectively, independent of the ac field. A more continuous slope change is observed for Y-2, i.e. μC gradually increases from 4 to 36.5. It is noteworthy that the maximum slope value is identical for both

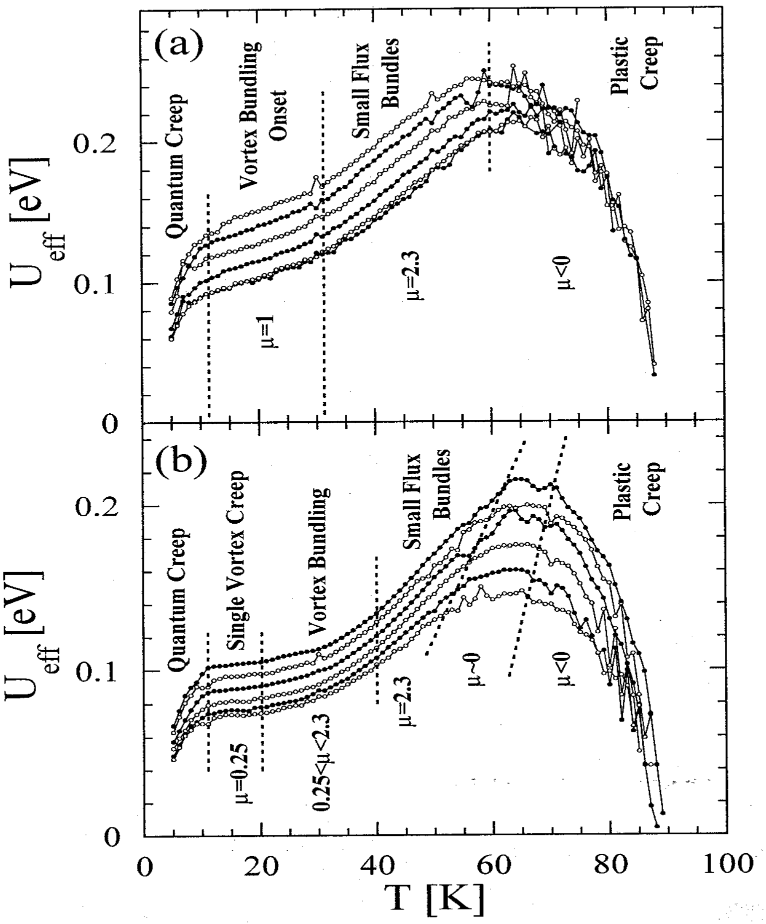

Figure 3. Effective activation energy vs. T for (**a**) Y-1 and (**b**) Y-2. The extracted μ values and the corresponding creep regimes are shown for different temperature regions.

samples. To determine μ, we use an estimate [23] for $C = \lim_{T\to 0} -(T/Q)\mathrm{d}\ln J_\mathrm{c}/\mathrm{d}T \approx 16$ evaluated in the temperature range $11 < T < 20$ K. The resulting μ for Y-2 starts out at 0.25 for $11 < T < 20$ K, increases gradually, and reaches a maximum of 2.3 above 40 K. This is in good agreement with theory, which expects $\mu = 1/7 = 0.14$ for single vortex creep and $\mu = 5/2$ for creep of small flux bundles. As an indication of larger flux bundles, μ begins to decrease at a field dependent temperature (50–63 K), and finally becomes negative at about 66–71 K, which indicates the possibility of plastic creep as the melting line and T_c is approached [26]. Similarly, Y-1 reaches $\mu = 2.3$ at about 31 K, indicative of small flux bundles. However, pure single vortex creep is never fully realized in this sample since $\mu = 1$ down to quantum creep temperatures. A similar temperature dependence has been reported, for example, in Y-123 single crystals in a 1 T field, where μ increases from about 0.7 at low temperature, reaches a maximum of about 2 at 29 K and then decreases to 0.6 at 70 K [27]. At 40 K an increasing magnetic field has a similar effect in that μ increases

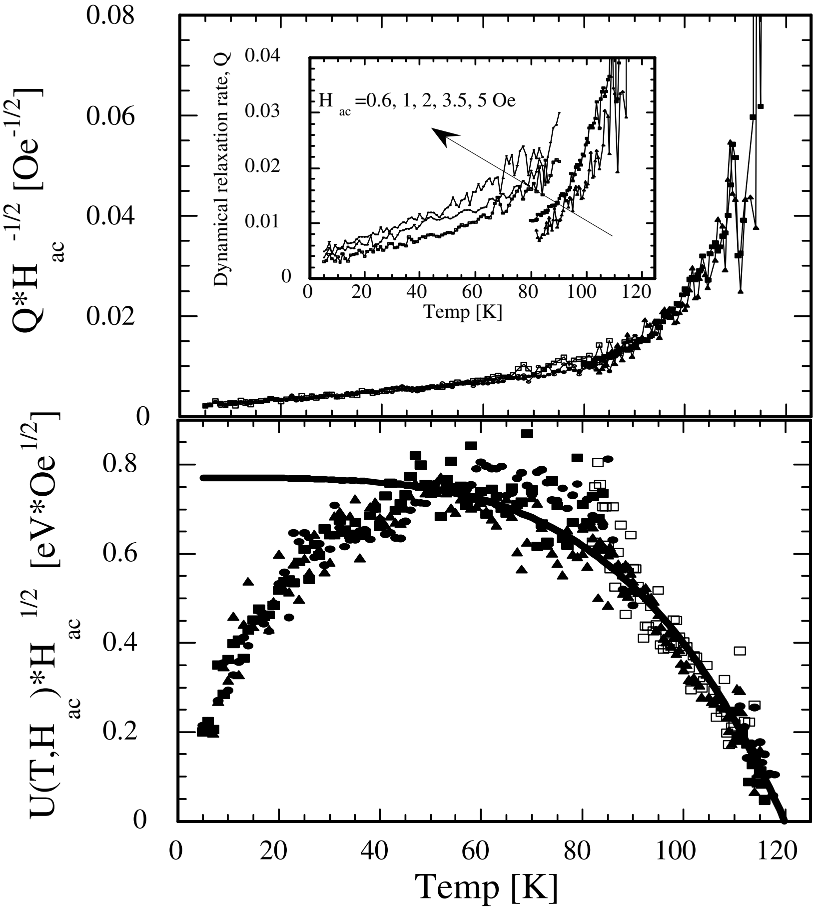

Figure 4. The inset in (top) shows the dynamical relaxation rate Q of H-a as a function of temperature. The square-root field dependence is revealed in (**top**) by the clear overlap of plots of $QH_{\mathrm{ac}}^{-1/2}$ vs. T. The corresponding flux creep activation energy can be well fitted in (**bottom**) by $U(T, H_{\mathrm{ac}}) = U_0(1 - t^4)H_{\mathrm{ac}}^{-1/2}$ with $U_0 = 0.77$ eV Oe$^{1/2}$ and $T_c = 120$ K.

from 0.16 at 0.1 T, reaches a maximum of 1.4 at 1 T, above which μ again decreases to ~ 1 [28].

We now turn to the more anisotropic materials. In the inset of figure 4(top) we show the dynamical relaxation rate of the a-axis oriented Hg-1212 sample. As expected for thermally excited creep of individual vortices, Q displays a monotonous increase with T and diverges on approaching T_c. More interestingly, Q exhibits a clear square-root ac field dependence, which is emphasized in figure 4(top) by plotting $QH_{\mathrm{ac}}^{-1/2}$ vs. T. This dependence is expected for dislocation-mediated plastic flux creep where vortices propagate by protruding vortex half-loops into local energy minima in the vortex ensemble, and has previously been observed at higher fields in other anisotropic materials such as

$Bi_2Sr_2CaCu_2O_{8+x}$ (Bi-2212) thin films, $Tl_2Ba_2CaCu_2O_8$ (Tl-2212) single crystals [29,30] and $Tl_{2/3}Ba_{1/3}Sr_2CaCu_2O_7$ (Tl-1212) single crystals [30]. The barrier limiting this vortex motion is roughly given by the creation energy of a vortex-antivortex pair perpendicular to the original vortex [31]

$$U_{\mathrm{pl}} = [2\Phi_0^2/4\pi\mu_0\lambda_{ab}\lambda_c]\ln(\kappa)\delta a_0\,. \tag{1}$$

From the temperature dependence of the penetration length one expects $U \propto (1 - t^4)$. In figure 4(bottom) we plot the activation energy as $UH_{\mathrm{ac}}^{1/2}$ vs. T together with a one-parameter fit $U(T, H_{\mathrm{ac}}) = U_0(1 - t^4)H_{\mathrm{ac}}^{-1/2}$ with $U_0 = 0.77$ eV$*$Oe$^{1/2}$. We hence conclude that *both* the temperature dependence and the field dependence are well described by eq. (1). Below about 45 K, U_{eff} deviates from the fit and drops sharply, even more so at about 25 K. As for Y-123 above, this behavior is indicative of the onset of quantum creep, and clearly displays a two-stage nature as recently reported in ref [10].

The same square-root field dependence is also found for c-axis oriented Hg-1212 (figure 3) and Tl-1212 thin films (not shown). In figure 5a one also observes a less field dependent regime for 30 Oe and below, which indicates that at about 1.2 μm vortex separation the average vortex distance no longer defines the half-loop length. Q displays a clear maximum at 95 K, above which flux creep slows down dramatically. A fit based on eq. (1), using $U_0 = 1.2$ eV Oe$^{1/2}$ and $T_{\mathrm{c}} = 110$ K agrees well with the data for 35 K $< T <$ 90 K. While the deviation below 35 K again is ascribed to quantum creep, the sharp upturn above 90 K signals the onset of collective effects in the vortex ensemble. The inset of figure 5b shows that on approaching T_{c}, this upturn is abruptly cut off at about 108 K where Q diverges and U_{eff} vanishes, possibly due to crossing over the melting line just below T_{c}. The Tl-1212 thin film showed a similar behavior, with quantum creep setting in below 17 K and a somewhat less pronounced upturn in U_{eff} as collective effects set in.

Before comparing the fitted U_0 with eq. (1) it is important to point out that its value relates to the root-mean-square ac field used in the ac susceptibility measurement. We have recently shown that U_0 has the same square-root dc field dependence, however with a 1.6 larger value, i.e., $U_{0,\mathrm{ac}} = 1.2$ eV Oe$^{1/2}$ corresponds to $U_{0,\mathrm{dc}} = 1.92$ eV Oe$^{1/2}$ [32]. From published values of the superconducting parameters for the Hg-1212 material system, $\lambda_{ab}(0) = 209$ nm [1], $\kappa = 126$ [33] and $\gamma = 67$ [2] we get an estimate of $U_0\delta^1 = 27$ eV$*$Oe$^{1/2}$, which suggests that at low fields the typical distance a vortex haluuh f-loops extends itself is only about $\delta = 0.046$ of the average vortex spacing in the a-axis sample and $\delta = 0.071$ in the c-axis samples. However, the sharp drop in U_{eff} seen in the inset of figure 5b as the melting line is crossed can be equally well fitted with eq. (1) by only changing δ from 0.071 to 0.68. At this point the average vortex jump is hence of the order of the average vortex separation as expected for the high-field/high-temperature vortex liquid.

4. Conclusion

In conclusion, we have presented a novel ac susceptibility technique to study vortex dynamics in superconducting thin films. We have shown that low-field vortex creep is surprisingly collective in Y-123 thin films, even at fields where vortices are about 2 μm apart. In the more anisotropic materials, Hg-1212 and Tl-1212, vortices creep individually at low and intermediate temperatures. A novel low-field dislocation-mediated plastic creep regime is found to dominate over a wide range of temperatures, until it is replaced

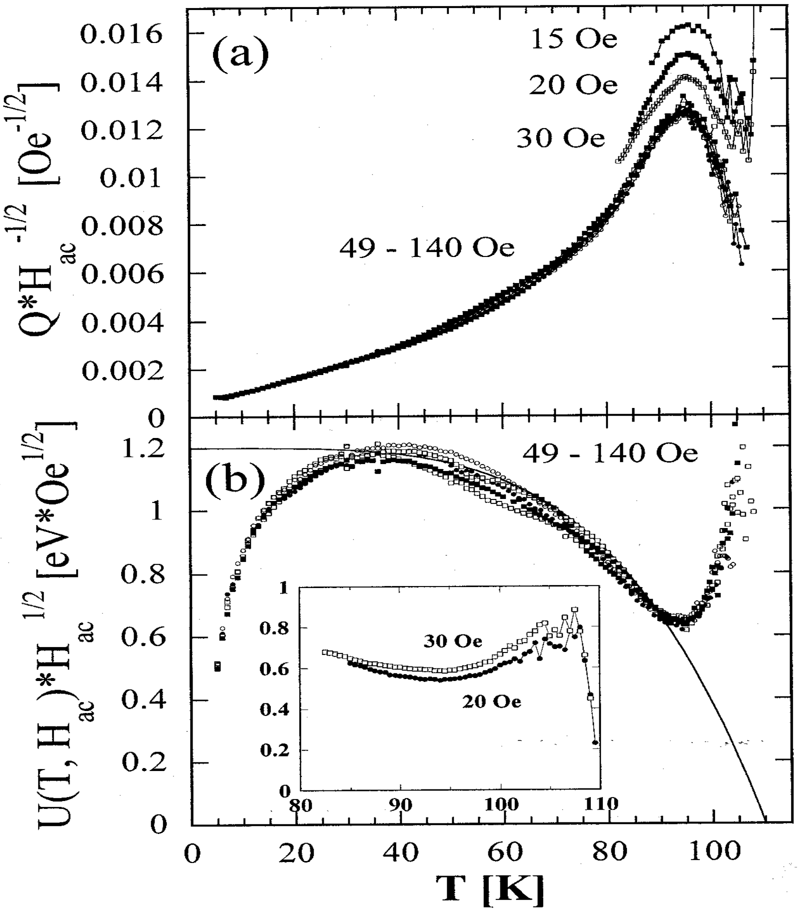

Figure 5. (**a**) $QH_{\mathrm{ac}}^{-1/2}$ vs. T for sample H-1 showing a clear overlap for fields greater than 49 Oe. A clear flux creep maximum is observed at about 95 K. (**b**) The corresponding activation energy for $H_{\mathrm{ac}} \geq 49$ Oe together with a fit to eq. (1) with $U_0 = 1.2$ eV $\mathrm{Oe}^{1/2}$ and $T_{\mathrm{c}} = 110$ K. **Inset:** Same for $H_{\mathrm{ac}} = 20$ and 30 Oe, where a sharp drop in U is observed at about 108 K.

by ordinary elastic creep as the vortex–vortex interaction increases at high temperature and high field, and creep becomes increasingly collective. Low-field quantum creep is found to become significant at relatively high temperatures in the more anisotropic materials.

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