Nonlinear effective activation energy and field dependence of the critical current density in $HgBa_2Ca_2Cu_3O_{8+\delta}$

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The effective activation energy of $HgBa_2Ca_2Cu_3O_{8+\delta}$ superconductor was extracted from the magnetic relaxation measurements below 30 K, and a power-law dependence on current density was found. We also obtained the effective activation energy from the whole magnetic hysteresis (M-H) loops plotted at constant field-sweep rate. The data sets for the M-H loops at different temperatures overlap each other, and all fall on the smooth $U_{\rm eff}(J,0)$ curve based solely on the magnetization relaxation measurements. This indicates that the field dependence of magnetic critical current density in high-temperature superconductors is determined by the flux creep via nonlinear effective activation energy at low temperatures.

Among many striking properties of high-T_c superconductors (HTSC's), giant flux creep is an important one. Understanding this feature has both theoretical and practical interest. Considerable work and intensive study have already been devoted to this problem, yielding many models for the detailed description of magnetic relaxation.[1-4] Among these models, the vortex-glass (VG) model of Fisher[1] predicts that random pinning forces from impurities induce a phase transition from a vortex-fluid phase into a vortex-glass phase, in which the vortices are frozen into some disordered pattern. In the vortex-glass state, vortices are not movable; therefore there is a true resistance-free state at finite temperatures. Collective-pinning (CP)[2] theory gives a similar picture. In particular, the two theories give the same power-law form for the dependence of activation energy $U_{\rm eff}(J)$ on current density J. Up to date, many magnetic measurements[5-7] are in support of the power law $U_{\rm eff}(J)$ relationship; however, electric measurements did not favor these theories until Bishop *et al.*[8] recently reported direct measurements of I-V characteristics of thin Y-Ba-Ca-O films using a superconducting quantum interference device (SQUID) picovoltmeter system. With improved voltage sensitivities of 10^{-10} V, the experimental window was expanded considerably. Their results strongly supported the VG-CP model.

Recently, superconductivity in $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ is particularly noteworthy due to the high critical temperature (T_c). Besides T_c, a high value of critical current density is needed for large-scale applications. Aiming at understanding the flux-pinning properties, some works are devoted to the magnetic characterization of the members of single Hg-O layer superconductors. Welp *et al.*[9,10] and Umezawa *et al.*[11] determined the irreversibility line of polycrystalline $HgBa_2CuO_{4+\delta}$, $HgBa_2CaCu_2O_{6+\delta}$, and $HgBa_2Ca_2Cu_3O_{8+\delta}$ using measurements of magnetic hysteresis. They found that the position of this line in the $H_{\rm irr}$ vs t ($t=T/T_c$) plane for $HgBa_2CuO_{4+\delta}$ is higher than that of double-layer and triple-layer Bi-Sr-Ca-Cu-O, but lower than that of $YBa_2Cu_3O_{7-\delta}$(Y-123). Welp *et al.*[10] made a systematic comparative study of the magnetization of these three compounds, which have similar exponential temperature dependence of critical currents, suggesting a similar flux dynamics. The position of the irreversibility lines was explained on the basis of the separation distance between the superconducting CuO_2 blocks. Schwartz *et al.*[12] reported an enhancement of magnetic J_c in neutron-irradiated polycrystalline $HgBa_2CuO_{4+\delta}$(Hg-1201), and concluded that it is not intrinsically limited to low current density and may become a technologically important compound. Schilling *et al.*[13] from magnetization-loop measurements and the broadening of the resistive transition in external fields H, estimated the location of the irreversibility boundary $H_{\rm irr}(T)$ in the magnetic phase diagram of $HgBa_2Ca_2Cu_3O_{8+\delta}$(Hg-1223). Their estimates of the critical current densities J_c in a magnetic field for $T=10$ and 75 K suggested that the intragrain critical current

densities are of comparable order of magnitude to those routinely observed in $YBa_2Cu_2O_7$ at these temperatures.

It is obvious that more detailed work on the characterization of flux dynamics in Hg-based superconductors and its comparison with that of other families of HTSC's is needed. In this work, we report magnetic studies on a polycrystalline Hg-1223 sample. Following the method suggested by Maley *et al.*,[3] a smooth curve of $U_{\text{eff}}(J,0)$ based on the magnetization relaxation measurements was obtained as well as the fitting parameter C. An empirical power-law field dependence of

$$U_{\text{eff}}(J,T,H)=kG(T)U_{\text{eff}}(J,0,H=1\text{ T})/H^n$$

on critical current was introduced to extract the activation energy. By adjusting the exponent n, all data sets from 0.6 to 6 T fall nearly on a single curve of the power-law fitting of activation energy based on the magnetic relaxations, using the same value C. This indicates that the field dependence of magnetic critical current density is determined by the flux creep via a nonlinear relationship between effective activation energy and current density.

The Hg-Ba-Ca-Cu-O polycrystalline samples in this work were prepared by the method of solid-state reaction with a detailed description in Ref. 14. Based on the x-ray diffraction (XRD) pattern, resistivity, dc and ac magnetic susceptibility measurement, we showed that $HgBa_2Ca_2Cu_3O_{8+\delta}$ is the major superconducting phase. The dimension of the sample is $4.0\times0.8\times1.5$ mm^3 and the weight is 14.7 mg. T_c is about 129 K, corresponding to the zero-resistivity temperature.[14]

The magnetic measurements reported in this work were performed on a vibrating-sample magnetometer (Oxford 3001) in the temperature range from 4.2 to 110 K. The magnetic hysteresis loops were recorded at a constant field-sweep rate of 100 G/s. The measurement of each M-H loop consists of cooling the sample from above T_c to a given temperature, cycling the field from zero to a maximum, usually 7 T, down to zero, reversing the field, sweeping the field to a minimum, and finally back to zero. The measurement of magnetic relaxation follows subsequently. The field was first increased to a settled field of 1 T at a rate of 100 G/s. After an initial time of 60 s, the magnetic signal was recorded every 10 s over a period of 2000 s.

Figure 1 shows typical magnetic hysteresis loops of the Hg-1223 sample at 4.2, 8, 10, 15, 30, and 40 K. At these temperatures the magnetic hysteresis (the width of the M-H loops) decreases with the applied field, but remains nonzero at maximum field of 7 T. However, at higher temperatures, even much lower than T_c, say 80 K, the magnetic hysteresis diminishes at a moderate field of 3 T. By using a J_c criterion of 10^4 A/cm^2, we determine the irreversibility line as illustrated in Fig. 2. The position of this line is lower than that of the $YBa_2Cu_3O_7$ system in the H_{irr} vs reduced temperature $t(t=T/T_c)$ plane, indicating that the flux pinning is still rather weak in the Hg-based system.[10,11]

The magnetic-momentum decays of the Hg-1223 sample at 1 T follow roughly a logarithmic behavior, as

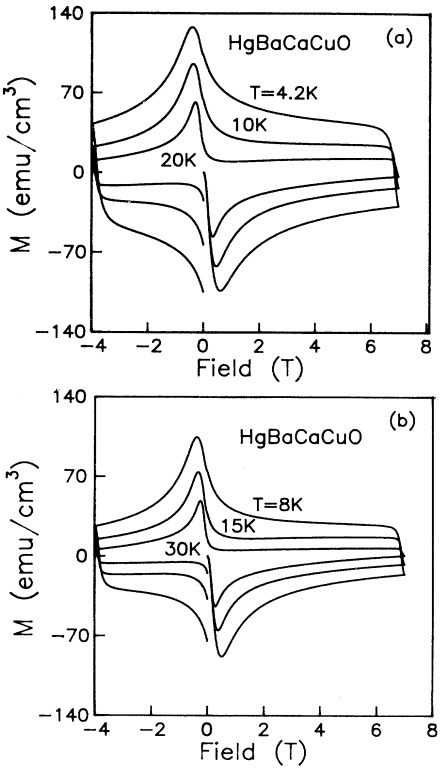

FIG. 1. Magnetization loops of the Hg-1223 sample at 4.2, 8, 10, 15, 20, and 40 K.

shown in Fig. 3. Note an initial time of 60 s is added to the data file, the normalized flux-creep rate $S=(-\partial M/\partial t)(1/M_{\text{irr}})$ where M_{irr} is the irreversible component in the M-H loops at $H=1$ T is also quite large, ranging from 0.028 at 4.2 K to 0.079 at 60 K. These values are comparable to those of Y-Ba-Cu-O. It can also be noted from Fig. 4 that a plateau of S-T exists, consistent with many previous reports.[16]

Using the Bean critical-state model, one can estimate

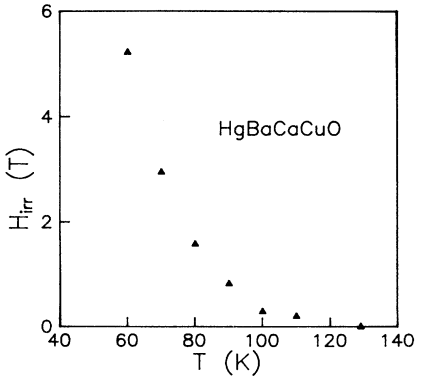

FIG. 2. Irreversibility field (H_{irr}) plotted against temperature T for the Hg-1223 sample. The criterion of J_c is 10^4 A/cm^2. In the field range from 0.5 to 5.2 T, T depends on H_{irr} logarithmically, $T=87.1-16.1\ln H_{\text{irr}}$.

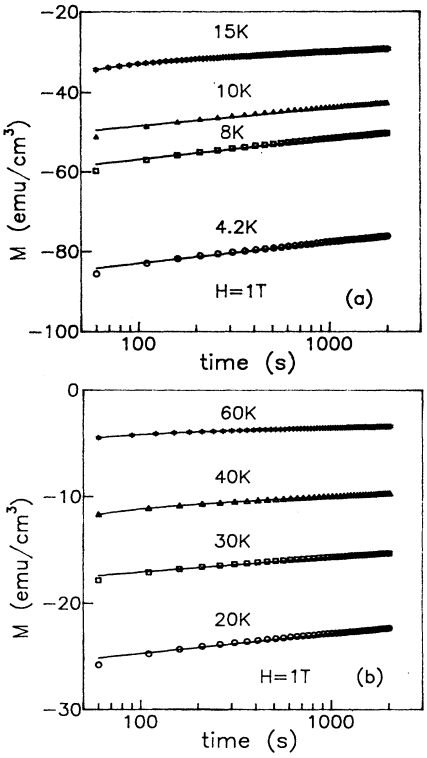

FIG. 3. Magnetization relaxations of the Hg-1223 sample at temperatures of 4.2, 8, 10, 15, 20, 30, 40, and 60 K and field of 1 T display a logarithmic decay (solid lines).

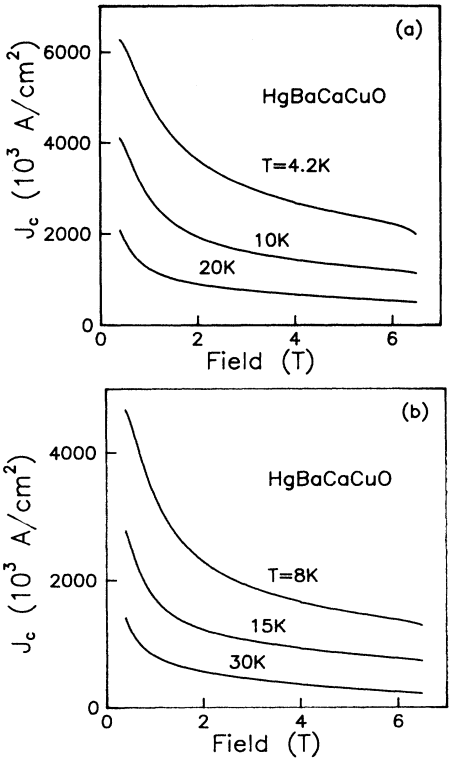

FIG. 5. Field dependence of critical current densities for the Hg-1223 sample.

the intragranular critical current density in the sample. $J_c = 30\Delta M/d$ in practical units, where ΔM (emu/cm^3) is the difference of the upper and the lower branches in the M-H loops, and $d \approx 10$ μm is the average grain diameter based on scanning electron micrographs of this sample.[14] The maximum fields in this work are large enough to break the weak links between the grains in the sample, so J_c calculated in this way is the intragranular critical current density. Though the estimated J_c is only correct in the order of magnitude, the comparison of its values at different fields and temperatures is meaningful. In Fig. 5 we show the field dependence of critical current density at temperatures up to 30 K; further discussion on this dependence will be given at the end of this paper. The critical current density decreases quasiexponentially with temperature, as shown in Fig. 6 for fields of 1 and 2 T, where the solid lines are exponential fitting.

We also need to check the error in applying the Bean model, that is, the assumption that the critical current density is uniform in the sample. By estimating the rela-

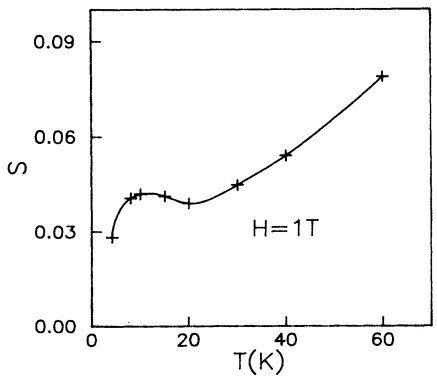

FIG. 4. Normalized flux-creep rate S vs T. The solid line (cubic spline fitting) is a guide to the eye.

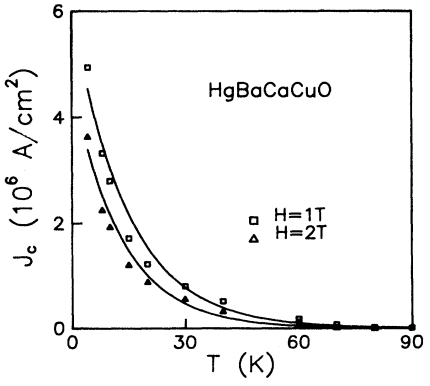

FIG. 6. $J_c(T)$ of the Hg-1223 sample at 1 and 2 T, estimated from the Bean model, displays a drastic drop with temperature. The solid line is an exponential fit with form e^{-T/T_0}, where $T_0 \approx 15$ and 13 K for $H = 1$ and 2 T, respectively.

tive change of flux density in the grains from the J_c-H shown in Fig. 5, we found that the error introduced by using the Bean model is less than 5%;[15] therefore the field dependence of the critical current density estimated in this way deviates only slightly from the real case.

Since the effective activation energy depends on the current density nonlinearly, we should follow the method proposed by Maley *et al.*[3] and McHenry *et al.*[17] to extract $U_{\rm eff}(J,T=0\ {\rm K},H=1\ {\rm T})$. Starting from the flux conservation equation and Arrhenius law for flux hopping, the effective activation energies $U_{\rm eff}(J,T,H)$ can be derived[15] as

$$\frac{U_{\rm eff}(J,T,H)}{T}=\left[C-\ln\left|\frac{\dot{H}}{4\pi}+\frac{dM}{dt}\right|\right] \quad (1)$$

with $C=\ln(Ba\omega_0/\pi d)$, where ω_0 is a characteristic attempt frequency of vortices, and a is the average hopping distance of vortices. In the measurement of isothermal magnetization relaxation at constant field, $\dot{H}=0$, and the above equation is reduced to the expression used in some reports.[3,6,7,17] C is treated as a constant determined by fitting the magnetic relaxation data to obtain the smoothest continuous curve of $U_{\rm eff}(J,T=0,H=1\ {\rm T})$ versus J. The temperature dependence of $U_{\rm eff}(J,T,H=1\ {\rm T})$ is accounted for in the function $G(T)$,[17] usually of the form $\{1-[T/T_{\rm irr}(H)]^2\}^{3/2}$, [$T_{\rm irr}(H)$ is the irreversible temperature]. For the Hg-1223 sample in this work $T_{\rm irr}(H=1\ {\rm T})=87$ K which is interpolated from the irreversibility line in Fig. 3. It should be emphasized that the construction of the $U_{\rm eff}(J,0)$ curve with several segments at different temperatures is only valid at low temperature, where the depinning critical current density J_{c0} can be assumed as a constant, but not valid at temperatures too low, typically below 1 K, where the quantum tunneling of flux may dominate the flux creep.[18,19] Therefore in this work we will confine ourselves to the analysis of data between 4.2 and 30 K [30 K is about one-third of $T_{\rm irr}(H=1\ {\rm T})$]. We presented the extracted $U_{\rm eff}(J,T=0,H=1\ {\rm T})$ vs J relationship in Fig. 7(a), and in a current density window of more than one decade $U_{\rm eff}(J,0,H=1\ {\rm T})$ follows a power-law dependence on J. A double-logarithmic plot [Fig. 7(b)] of $U_{\rm eff}(J,0,H=1\ {\rm T})$ vs J makes this dependence clearer, the solid line is the best fit to $J^{-\mu}$ with $\mu\approx 1.03$, close to the value of $\frac{7}{9}$ in the regime of flu, creep in large vortex bundles predicted by three-dimensional (3D) collective-creep theory.[2] We also calculated the effective activation energies at $H=1$ T corresponding to the M-H loops plotted at constant field-sweep rate $\dot{H}$. These data points all fall on the power-law fitting curve of $U_{\rm eff}(J,0,H=1\ {\rm T})$ determined from the magnetic relaxations, which manifests that the temperature dependence of so-called magnetic critical current density is determined by flux creep via the $U_{\rm eff}(J)$ relationship at low temperatures. This point has been clarified by McHenry *et al.*[17] and Ji *et al.*[15] It is then quite natural to consider how the field dependence of magnetic critical current density is influenced by flux creep, so as the second step in this work, we constructed $U_{\rm eff}(J,0,H=1\ {\rm T})$ vs J from the magnetic hysteresis loops at low temperatures by adjusting n in the empirical power-law field dependence of effective activation energy

$$U_{\rm eff}(J,T,H)=kG(T)U_{\rm eff}(J,0,H=1\ {\rm T})/H^n\ .$$

The above fitted value of C was used. Similar to the previous treatment, we would use the measurement results at low temperatures and fields not too high. Tentatively, we assume J_{c0} is weakly dependent on the field below the maximum, 7 T below 30 K in this work. Furthermore, we exclude the data at the central peak and in the field reversal process, for the Bean model is no longer valid in these regimes. We substituted the applied field H for the flux density B, and disregarded the demagnetization effect.[15]

In practice we used only the data from 0.6 to 6 T. The results of such an analysis are shown in Fig. 8. It can be seen that the data sets at different temperatures overlap each other at equal J, and additionally the data sets at six

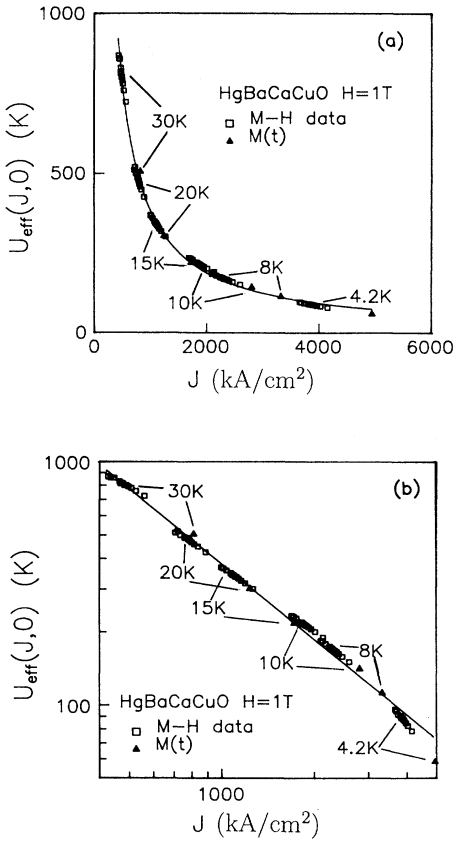

FIG. 7(a) The effective activation energies $U_{\rm eff}(J,0,H=1\ {\rm T})$ (□) extracted from the magnetic relaxation fits a power law (solid line is the result of best fitting). The data points marked with (▲) are those based on the width of magnetic hysteresis loops at $H=1$ T. The fitting parameter $C=16$. (b) A double-logarithmic plot of $U_{\rm eff}(J,0,H=1\ {\rm T})$ indicating a power-law dependence on the current density.

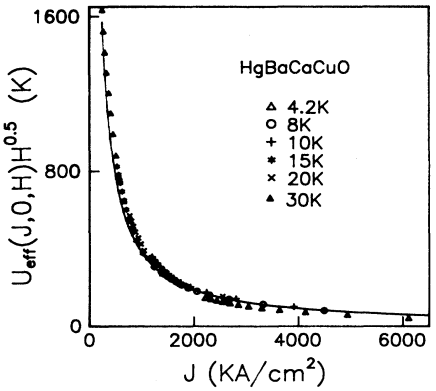

FIG. 8. Current density dependence of effective activation energy $U_{\rm eff}(J,0,H=1\ {\rm T})$ deduced from the magnetic hysteresis loops. $U_{\rm eff}(J,T,H)=kG(T)U_{\rm eff}(J,0,H=1\ {\rm T})/{\rm H}^n$, with the fitting parameter $n=0.5$. The solid line is the power-law fitting curve in Fig 7(a).

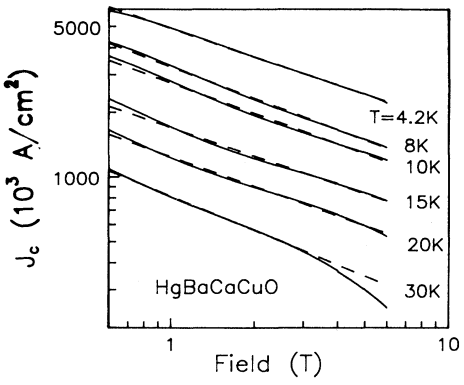

FIG. 9. Double-logarithmic plot magnetic critical current J_c as a function of field at several temperatures. The dashed lines are best fits with power law $J(H)=P(T)H^{-Q}$, where $Q=0.44,0.49,0.47,0.44,0.46$, and 0.53 for data of $T=4.2,8,10,15,20$, and 30 K, respectively; namely, the slopes of the lines in the figure are approximately equal.

temperatures 4.2, 8, 10, 15, 20, and 30 K all collapse on the power-law fitting line in Fig. 7. Note that the term $dM/dt=(dM/dH)(dH/dt)$ in Eq. (1) is much less than dH/dt, and its neglect in the calculation of the effective activation energy at $H=1$ T from the M-H loops introduces an error of 4% at most; therefore we neglected this term in extracting the effective energies from the whole magnetic hysteresis loops.

The consistency of $U_{\rm eff}(J,0,H=1\ {\rm T})$ determined by two different methods leads to an inevitable conclusion: the field dependence of magnetically measured critical current density is determined only by the giant flux creep at low temperature. This is the central point of this work. Moreover, this consistency would imply a power-law field dependence of critical current density, because $U_{\rm eff}(J,T,H)H^n/G(T)\propto J^{-\mu}$ and $U_{\rm eff}(J,T,H)/T=C-\ln\dot{H}/4\pi$ can be combined to yield $J\propto H^{-n/\mu}[T/G(T)]^{-1/\mu}$. Since $G(T)=\{1-[T/T_{\rm irr}(H)]^2\}^{3/2}$, in which the irreversibility field decreases exponentially with temperature in the field range studied (Fig. 2), the magnetic critical current density depends on the applied field approximately in a power law.

By replotting Fig. 5, we demonstrated this dependence clearly (Fig. 9). The dashed lines are fits to the expression $J(H)=P(T)H^{-Q}$. The coefficient $P(T)$ is approximately proportional to $[T/G(T)]^{-1/\mu}$, as shown in Fig. 10, where $T_{\rm irr}(H)$ in $G(T)=\{1-[T/T_{\rm irr}(H)]^2\}^{3/2}$ is set to 87 K corresponding to a field of 1 T. This arbitrariness would not influence the results, especially at low temperatures, since $G(T)$ is of order unity in the temperature range studied and $T_{\rm irr}(H)$ is slowly varying with field. However, there does exist deviation from power-law dependence for the data set $T=30$ K, and this deviation may arise from the field dependence of $G(T)$, so we only fitted the data from 0.6 through 3 T to the expression of $J(H)=P(T)J^{-Q}$ in order to find $P(T)$. The facts that the magnetic critical current density follows a roughly power-law dependence on the applied field with nearly equal exponents and that $P(T)$ scales with $[T/G(T)]^{-1/\mu}$ provide further supports to the above conclusion.

One may ask if the field dependence of the depinning critical current density $J_{c0}(H)$, e.g., power-law $J_{c0}(H)$, also contributes considerably to the field dependence of magnetically determined $J_c(H)$, but has been included in the field dependence of effective activation energy so that we can still obtain the consistency of activation energies by two means. In our viewpoint, $J_{c0}(H)$ may depend on the field in a power law with close exponents from 4.2 to 30 K, but it is unlikely that they have close coefficients at higher temperature too. Therefore we tend to believe that flux creep is the predominant factor of the field dependence of magnetic J_c of Hg-1223 below 30 K.

At higher temperatures, the real critical current density J_{c0} should exhibit substantial change with the applied

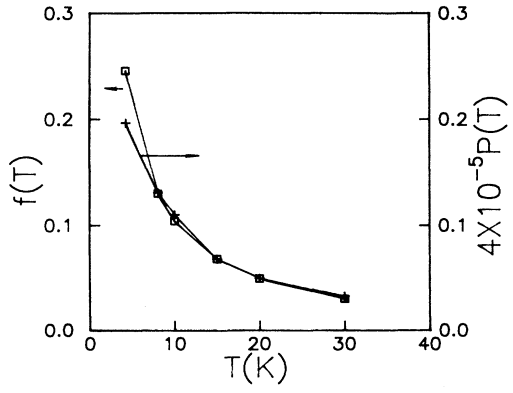

FIG. 10. Scale of $P(T)$ with $f(T)=[T/(G(T)]^{-1/\mu}$, where $G(T)=\{1-[T/T_{\rm irr}(H)]^2\}^{3/2}$, $T_{\rm irr}(H=1\ {\rm T})=87$ K and $\mu=1.03$. $P(T)$ is the coefficient in $J(H)=P(T)H^{-Q}$, in which $J(H)$ and H are in the units of kA/cm^2 and T, respectively.

field; therefore the shape of magnetic hysteresis loops is determined by a combined effect of $J_{c0}(H)$ and giant flux creep. A direct determination of the flux-pinning mechanism from the scaling of the flux-pinning force density F_p as discussed in detail for conventional superconductors by Dew-Hughes[20] should not be suitable for high-temperature superconductors. However, the scaling of F_p vs H is reported in all systems of high-temperature superconductors, including the newly discovered Hg-based system.[10,11] This can be illustrated in the following way. By assuming the validity of a power-law $U(J)$ relationship, the combination of

$$U(J,T,H)H^n/G(T) \propto [J/J_{c0}(H)]^{-\mu}$$

and $U(J,T,H) \propto T$ at constant field-sweep rate yields an expression $J' \propto H^{-n/\mu} J_{c0}(H)$. That is to say, if the flux-pinning force calculated from $J_{c0}(H)$ satisfies the scaling law, $F_p(H)$ from $J(H)$ also obeys the scaling. The approximation of C as a constant is reasonable because of its logarithmic dependence on field B. Of course, one can use the generalized inversion scheme proposed by Schnack *et al.*[19] to extract $J_{c0}(H)$, upon which to base further discussion.

In conclusion, magnetic hysteresis and relaxation measurements have been performed on one $HgBa_2Ca_2Cu_3O_{8+\delta}$ sample. The current density dependence of effective activation energy was first extracted from the magnetic relaxation below 30 K. We also obtained $U_{\rm eff}(J,0,H=1\ {\rm T})$ vs J from the magnetic hysteresis loops by adjusting the exponent n in the power-law field dependence of $U_{\rm eff}(J,T,H)$. These data sets overlap each other and fall on a single curve, and moreover they are consistent with the $U_{\rm eff}(J,0,H=1\ {\rm T})$ vs J calculated from the magnetic relaxation, which we think is a strong support to the conclusion that the giant flux creep determines the field dependence of magnetically measured critical current density at low temperatures via the nonlinear effective activation energy $U_{\rm eff}(J)$.

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