

# Nonmonotonic temperature dependence of the experimentally determined vortex-creep activation energy in disordered high-temperature superconductors

L. Miu\* and S. Popa

*National Institute of Materials Physics, Bucharest-Magurele, MG-7, Romania*

T. Noji and Y. Koike

*Department of Applied Physics, Tohoku University, Aramaki Aoba, Aoba-ku, Sendai 980-77, Japan*

D. Miu

*National Institute of Laser, Plasma, and Radiation Physics, Bucharest-Magurele, MG-6, Romania*

S. Diaz and G. Chouteau

*Grenoble High Magnetic Field Laboratory, B. P. 166, 38042 Grenoble, Cedex 9, France*

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The temperature  $T$  variation of the normalized vortex-creep activation energy  $U^*$  determined in standard magnetization relaxation experiments for  $\text{Pb}_2\text{Sr}_2\text{Y}_{0.53}\text{Ca}_{0.47}\text{Cu}_3\text{O}_{8+\delta}$  single crystals with random point disorder exhibits a maximum, which moves to lower- $T$  values by increasing the external magnetic field oriented parallel to the  $c$  axis. The nonmonotonic  $U^*(T)$  dependence is related to the change of the vortex pinning barriers involved in the creep process across the order-disorder transition in the vortex system (accompanied by the occurrence of the second magnetization peak), in a dynamic scenario. The decrease of  $U^*$  with decreasing  $T$  in the low- $T$  region is caused by the shift of the current density  $J$  range probed in standard magnetization measurements toward the critical current density, and the significant  $U^*(J)$  variation in the elastic-creep domain. The dynamic approach is confirmed by the behavior of highly disordered top-seeded melt-grown  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  crystals at low  $T$ , for which no second magnetization peak appears, and  $U^*$  does not depend on  $T$ .

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Flux-creep measurements remain essential for the study of high-temperature superconductors (HTSC's) in connection to their practical applications, since the thermal activation of vortex motion cannot be avoided.<sup>1</sup> In order to reduce the intrinsic ambiguity of flux creep measurements,<sup>2</sup> many magnetization relaxation studies on HTSC's (see, for example, Refs. 3–7) focused on the analysis of the normalized vortex-creep activation energy  $U^* = T|d \ln(t)/d \ln(M_{\text{irr}})|$ , where  $M_{\text{irr}}$  is the absolute value of the irreversible magnetization and  $t$  is the relaxation time. However, even in this approach, the experimentally determined  $U^*$  appeared to be inconsistent with the expected  $T$  and magnetic field variation of the true pinning potential, as discussed in Ref. 3.

The nonmonotonic  $U^*(T)$  dependence observed for disordered HTSC's was attributed to pinning barrier distribution,<sup>8</sup> collective pinning behavior,<sup>9</sup> or to a crossover from bulk pinning to surface barriers.<sup>10</sup> Alternatively, the peculiar decrease of  $U^*$  with decreasing  $T$  in the low- $T$  region, leading to a weakly  $T$  dependent normalized relaxation rate  $S = |d \ln(M_{\text{irr}})/d \ln(t)|$ , was associated with a large contribution of vortex tunneling<sup>11</sup> (quantum vortex creep), characterized by a  $T$  independent  $S$ .<sup>1</sup>

In this work we discuss the  $U^*(T)$  variation determined in standard magnetization relaxation experiments for  $\text{Pb}_2\text{Sr}_2\text{Y}_{0.53}\text{Ca}_{0.47}\text{Cu}_3\text{O}_{8+\delta}$  single crystals with random point disorder in terms of classic vortex creep, by considering the change in the creep process across the second magnetization peak (SMP)<sup>12</sup> (accompanying the order-disorder transition in

the vortex system<sup>13</sup>), in a dynamic scenario. This approach is supported by the fact that in the case of highly disordered top-seeded melt-grown  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  single-grain samples, showing no SMP at low  $T$ ,  $U^*$  does not depend on  $T$  in the low- $T$  region. In this framework, we explain the nonmonotonic  $U^*(T)$  variation, the unexpected increase of the second magnetization peak field with decreasing  $T$  in the low- $T$  domain, as well as the appearance of a weakly  $T$  dependent  $S$  up to relatively high  $T$  values.

The investigated specimens are  $1 \times 0.5 \times 0.1 \text{ mm}^3$   $\text{Pb}_2\text{Sr}_2\text{Y}_{0.53}\text{Ca}_{0.47}\text{Cu}_3\text{O}_{8+\delta}$  single crystals (PSYCCO) grown by the  $\text{PbO-NaCl}$  flux method<sup>14</sup> (with the critical temperature  $T_c \approx 76 \text{ K}$  and the anisotropy factor  $\gamma \approx 5$ ), and relatively large ( $3 \times 3 \times 1.5 \text{ mm}^3$ ) fully oxygenated top-seeded melt-grown  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  crystals (TSMG YBCO), with  $T_c \approx 91 \text{ K}$  and  $\gamma = 8-10$ . The quenched disorder in PSYCCO single crystals is essentially random point disorder.<sup>14</sup> Additionally, the TSMG YBCO crystals contain  $\approx 15\%$  nonsuperconducting  $\text{Y}_2\text{BaCuO}_5$  particles ( $\approx 10 \mu\text{m}$  mean size), twins, and many growth defects. The normal state resistivity of both kinds of samples is of the order of  $1 \text{ m}\Omega \text{ cm}$ . The magnetization  $M$  (identified with the irreversible magnetization) was measured in zero-field cooling conditions and increasing external magnetic field  $H$  (oriented parallel to the  $c$  axis, i.e., along the smallest sample dimension), using a commercial Quantum Design SQUID magnetometer and/or a magnetometer with extraction. In magnetization relaxation measurements the relaxation time was considered to be zero when

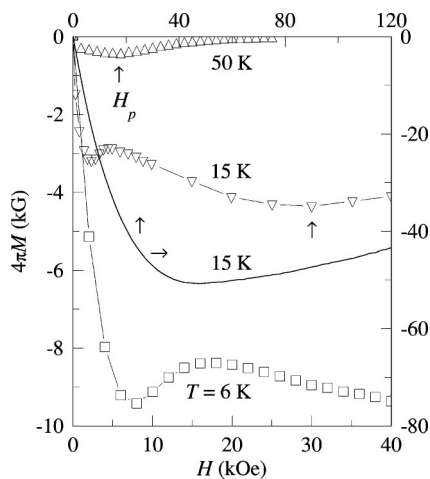
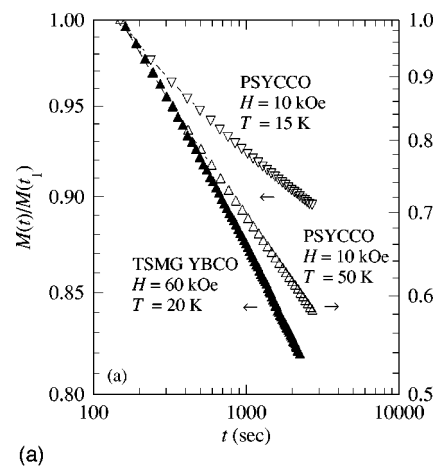


FIG. 1. Dc magnetization curves  $M(H)$  of  $\text{Pb}_2\text{Sr}_2\text{Y}_{0.53}\text{Ca}_{0.47}\text{Cu}_3\text{O}_{8+\delta}$  single crystals (PSYCCO) with random quenched disorder exhibiting the SMP over a large  $T$  interval (symbol, with the SMP field  $H_p$  indicated by an arrow), and of highly disordered top-seeded melt-grown  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  crystals (TSMG YBCO), showing no SMP at low  $T$  (continuous line).

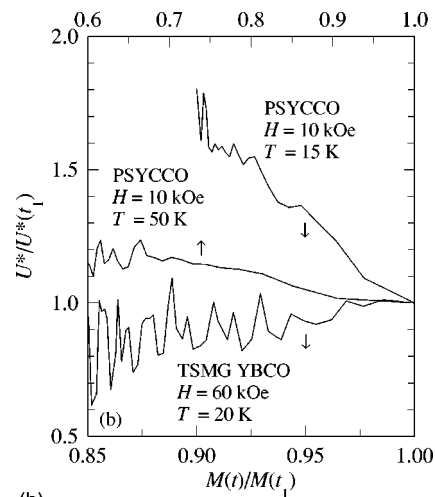
the magnet charging was finished, and the first data point was taken at  $t_1 = 150$  s.

PSYCCO single crystals exhibit a pronounced SMP over a large  $T$  interval,<sup>14</sup> as illustrated in Fig. 1. As known, the SMP represents the signature of the transition between the low-field quasiordered vortex phase (the Bragg glass, stable against dislocation formation) and a disordered vortex phase at higher  $H$ , where there is a better accommodation of vortices to the pinning centers and dislocations proliferate.<sup>12,13</sup> When the thermal energy is small, the order-disorder transition in the vortex system roughly occurs when the pinning energy generated by the quenched disorder overcomes the elastic energy of the vortex system. In this context, the relatively strong  $T$  variation of the peak field  $H_p$  even in the low- $T$  range (see Fig. 1) may be surprising,<sup>15</sup> since for  $T/T_c \leq 1/3$  the pinning energy is practically  $T$  independent.<sup>16</sup> The increase of the characteristic fields for the SMP with decreasing  $T$  in the low- $T$  range when bulk pinning is dominant was observed for  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  single crystals, as well, and was connected to the reduction of the actual pinning energy by the macroscopic current induced during magnetization measurements.<sup>17</sup> In contrast, for our highly disordered TSMG YBCO samples at low  $T$  no SMP appears, as can be seen in Fig. 1. The absence of the SMP seems to be caused by the presence of various, strong pinning centers, such as  $\text{Y}_2\text{CaCuO}_5$  particles and twin boundaries, when no ordered or quasiordered vortex structure can exist, at least for  $H$  above the field of first full penetration.

Characteristic magnetization relaxation curves  $M(t)$  for the two samples are shown in Fig. 2(a). For a better comparison,  $M(t)$  was normalized to the first measured value  $M(t_1)$ . For TSMG YBCO the  $M(t)$  data in log-log scales is close to a straight line, as proved for  $H = 80$  and  $100$  kOe at  $T = 20$  K, as well. PSYCCO single crystals behave similarly for  $H > H_p(T)$ . However, even in the representation from Fig. 2(a) the relaxation curve for PSYCCO at  $T = 15$  K and for



(a)



(b)

FIG. 2. (a) Characteristic magnetization relaxation curves  $M(t)$  of TSMG YBCO crystals and PSYCCO single crystals (log-log scales). (b) The same data plotted as  $U^* = -Td \ln(t)/d \ln(|M|)$  versus  $|M|$ . For a better comparison,  $M(t)$  and  $U^*$  were normalized to their first determined values  $M(t_1)$  and  $U^*(t_1)$ , respectively, with  $t_1 = 150$  s.

$H = 10$  kOe exhibits an upward curvature. It means that for  $H$  between the onset of the SMP and  $H_p$  (see Fig. 1) the logarithmic model<sup>18</sup> for the current density  $J(\propto |M|)$  variation of the actual vortex-creep activation energy  $U$  [ $U = U_0 \ln(J_c/J)$ , where  $J_c$  is the critical-current density and the pinning barrier  $U_0$  does not depend on  $J$ ] may represent a crude approximation only. This can be seen in Fig. 2(b), as well, where the data from Fig. 2(a) has been plotted as  $U^* = -Td \ln(t)/d \ln(|M|)$  versus  $|M|$ . For TSMG YBCO and PSYCCO in  $H > H_p(T)$  the  $U^*(J)$  variation is weak, whereas for PSYCCO at  $H \leq H_p(T)$  there is a significant increase of  $U^*$  with decreasing  $J$ . [The slight decrease of  $U^*$  with decreasing  $J$  appearing for TSMG YBCO in Fig. 2(b) is mainly due to the increase of the magnetic induction inside the sample during magnetization relaxation.]

Using the approximation  $U(J) = U_0 \ln(J_c/J)$  for both samples in a limited relaxation time window (which is the case of standard magnetization measurements), an averaged  $U^*$  was first determined as  $U^* = -T\Delta \ln(t)/\Delta \ln(|M|)$ , by

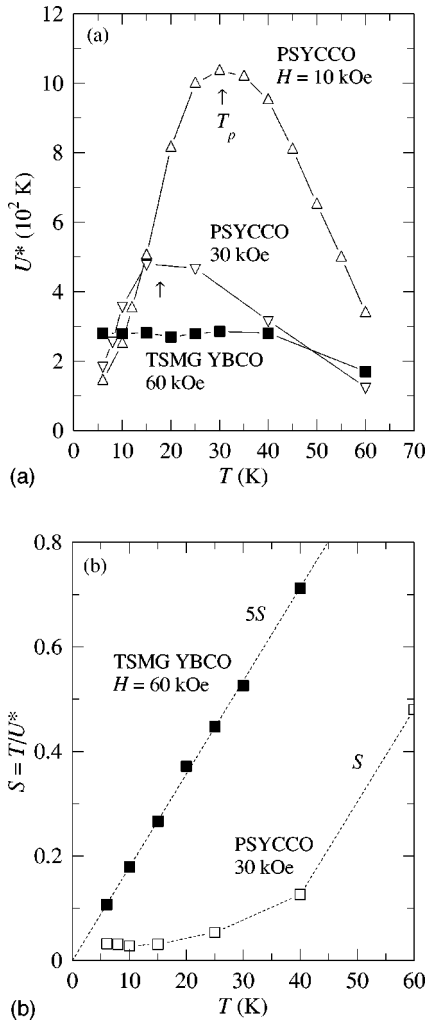


FIG. 3. (a) Characteristic  $T$  dependence of the normalized vortex-creep activation energy  $U^* = -T\Delta \ln(t)/\Delta \ln(|M|)$  for PSYCCO single crystals and TSMG YBCO crystals. In the case of PSYCCO  $U^*(T)$  exhibits a maximum at  $T = T_p$  (indicated by an arrow), and  $T_p$  decreases with increasing  $H$ , whereas for TSMG YBCO  $U^*$  is practically  $T$  independent below  $\approx 40$  K. (b)  $T$  variation of the related normalized relaxation rate  $S = T/U^*$  for PSYCCO and TSMG YBCO.

considering the slope of the relaxation curve in double logarithmic scales for  $500 \text{ s} \leq t \leq 2000 \text{ s}$ . The resulting  $U^*(T)$  is illustrated in Fig. 3(a), and the  $T$  dependence of the related normalized relaxation rate  $S = T/U^*$  is plotted in Fig. 3(b).

The first observation in Fig. 3(a) is that  $U^*(T)$  for PSYCCO exhibits a maximum. The decrease of  $U^*$  with decreasing  $T$  in the low- $T$  region cannot be explained by the logarithmic  $U(J)$  variation, since with  $U(J) = U_0 \ln(J_c/J)$  and  $J \propto |M|$  in the general vortex-creep equation<sup>19</sup> [ $U = T \ln(t/t_0)$ , where  $t_0$  is the macroscopic time scale for creep] one obtains  $U^*(T) = -Td \ln(t)/d \ln(|M|) = U_0(T)$ , which should be constant for  $T \leq 30$  K, where all the superconductor parameters for PSYCCO are weakly  $T$  dependent. On the other hand, a logarithmic  $U(J)$  would be in agreement with the constant  $U^*(T)$  determined for TSMG YBCO at  $T \leq 40$  K [Fig. 3(a)].

The second observation in Fig. 3(a) is that the temperature

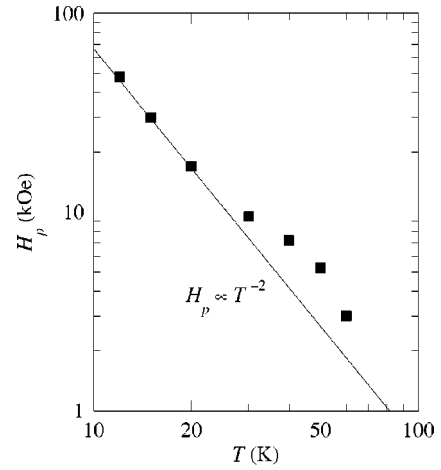


FIG. 4. The increase of  $H_p$  with decreasing  $T$  in the low- $T$  domain, where it takes the form  $H_p \propto T^{-2}$  (the continuous line).

$T_p$  for the  $U^*(T)$  maximum shifts to lower- $T$  values with increasing  $H$ , and  $T_p(H)$  appears to follow the  $H_p(T)$  variation plotted in Fig. 4. This, and the absence of the  $U^*(T)$  maximum in the case of TSMG YBCO, with no SMP at low  $T$ , suggest that the nonmonotonic  $U^*(T)$  variation can be related to the change in the pinning behavior across the SMP line.

As known, across the SMP there is a crossover between elastic creep at low  $H$  (in the Bragg glass domain) and plastic vortex creep for  $H \geq H_p$ .<sup>20,21</sup> The pinning barriers involving the plastic deformation of the vortex system have a weak intrinsic  $J$  variation, whereas the elastic (collective) pinning barriers  $U_{el}$  diverge with decreasing  $J$  [ $U_{el} \approx U_c (J_c/J)^\mu$ ,<sup>9</sup> where  $U_c$  is the collective pinning barrier, and  $\mu > 0$  is the collective pinning exponent]. This implies a change in the  $J$  dependence of the actual activation energy  $U$  upon crossing the  $H_p(T)$  line.

The analysis of  $U(J)$  is a complex problem,<sup>2</sup> since besides the intrinsic  $U(J)$  dependence given by the pinning barriers involved in the creep process, there exists an extrinsic  $U(J)$  nonlinearity, mainly caused by the barrier distribution.<sup>8</sup> However, it is well known from the study of classical superconductors<sup>22</sup> that extrinsic effects roughly lead to power-law shaped voltage-current characteristics, which means that in the presence of extrinsic effects the linear  $U(J)$  decrease,<sup>23</sup> for example, approximately takes a form  $\propto \ln(J_c/J)$ . It can then be assumed<sup>21</sup> that  $U(J) \approx U_{int}(J) \ln(J_c/J)$ , where  $U_{int}(J)$  represents the intrinsic  $J$  variation of the actual activation energy. For plastic barriers  $U_{int}(J) \approx \text{constant}$ , whereas for elastic pinning  $U_{int} = U_{el} \approx U_c (J_c/J)^\mu$ , when  $J$  is far enough from  $J_c$ . Thus, for TSMG YBCO in the whole  $H$  range of interest, as well as PSYCCO in  $H \geq H_p$ ,

$$U(J) \approx U_0 \ln(J_c/J). \quad (1)$$

In the case of PSYCCO in  $H < H_p$  one should consider  $U(J) \approx U_c (J_c/J)^\mu \ln(J_c/J)$ . However, the barrier distribution is expected to be more important above  $H_p$ , where vortices

accommodate to the pinning centers. When  $\mu$  is not small, and  $J$  is well below  $J_c$ , one can then use

$$U(J) \approx U_c(J_c/J)^\mu \approx U_{el}. \quad (2)$$

The  $U(J)$  dependence becomes relevant for any experiment based on a fixed relaxation time window over a large  $T$  interval, as discussed in Ref. 24. In standard zero-field-cooling magnetization measurements,  $H$  is changed with a constant step or a constant rate, and  $M$  is registered after a certain constant relaxation time interval  $t_1$ . In these conditions, with decreasing  $T$  in the low- $T$  range the probed current density  $J(t_1) \propto |M(t_1)|$  is progressively shifted toward  $J_c$ , because the overall relaxation in the time interval from  $t_0$  to  $t_1$  becomes smaller.

The behavior of  $U^*(J)$  for the two samples in the low- $T$  region is obtained with Eqs. (1) and (2) and  $J \propto |M|$  in the creep relation. The result is  $U^*(J) \approx U_0 \approx \text{constant}$  for TSMG YBCO and PSYCCO at  $T \geq T_p(H)$ , and  $U^*(J) \approx \mu U_c(J_c/J)^\mu \approx \mu U_{el}(J)$  for PSYCCO at  $T < T_p(H)$ , with a significant  $J$  variation, in agreement with Fig. 2(b). In the present context, the unexpected  $T$  variation of the peak field  $H_p$  at low  $T$  illustrated in Fig. 4 can easily be explained. The elastic creep-plastic creep crossover across the SMP<sup>20,21</sup> implies that at  $H_p(t_1)$  one has  $U^*(\text{plastic creep}) = U^*(\text{elastic creep})$ , which leads to  $U_0(T, H) = \mu U_{el}(T, H) = \mu T \ln(t_1/t_0)$ . In the case of random point disorder  $U_0 \propto H^{-1/2}$ ,<sup>1,25</sup> and  $U^*(H)$  in the elastic creep domain is weak [Fig. 3(a)]. Neglecting the  $T$  variation of  $U_0$  and  $\mu$  at low  $T$ , when the  $t_0(T, H)$  dependence is not very strong one obtains  $H_p(T) \propto T^{-2}$ , in agreement with the experimental results from Fig. 4.

For the  $U^*(T)$  variation at low  $T$ , where the intrinsic  $T$  dependence of the pinning potential is weak,  $U^*(T)$

$\approx U_0(T) \approx \text{constant}$  for TSMG YBCO, whereas for PSYCCO at  $T < T_p(H)$ , in the conditions of a fixed relaxation time window,  $U^*(T) \approx \mu U_{el} \propto T$ . The  $U^*(T)$  maximum from Fig. 3(a) results from the fact that at  $T < T_p(H)$  the creep is elastic, and the above “ $U^*(J)$  effect” is essential, leading to the decrease of  $U^*$  with decreasing  $T$ . For  $T > T_p(H)$  the  $U^*(J)$  variation is small (plastic creep), whereas the intrinsic  $U_0(T)$  decrease with increasing  $T$  (linear in  $T$  at high  $T$ ) is dominant. Consequently,  $S(T)$  will exhibit the “classic” behavior in the case of TSMG YBCO, as expected, whereas for PSYCCO the above discussed decrease of  $U^*$  with decreasing  $T$  generates a weak  $S(T)$  variation [Fig. 3(b)], resembling quantum vortex creep.

In summary, the nonmonotonous  $U^*(T)$  dependence observed for disordered HTSC’s appears to be related to the change of the vortex pinning barriers involved in the creep process across the order-disorder transition in the vortex system. The relatively strong  $U^*(J)$  variation in the case of (elastic) collective pinning and the finite relaxation time window in standard magnetization relaxation experiments lead to the decrease of  $U^*$  with decreasing  $T$  in the low- $T$  range. As a result, the normalized creep rate exhibits a weak  $T$  variation up to relatively high- $T$  values, resembling quantum vortex creep, and the SMP field increases with decreasing  $T$  in the low- $T$  region. The presented dynamic approach is supported by the behavior of TSMG YBCO samples, showing no SMP at low  $T$ , for which  $U^*$  does not depend on  $T$  in the low- $T$  domain.

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\*Electronic mail: elmiu@alpha1.infm.ro

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