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Field and temperature dependence of the mean penetration rate of fluxons in the mixed state of high- T_c superconductors

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Abstract. — The time decay of the zero field cooled diamagnetic magnetization of superconducting Bi(Pb)₂Sr₂Ca₂Cu₃O_y and YBa₂Cu₃O₇₋₈ ceramics has been measured and analyzed in terms of a mean activation energy \overline{E} . We find $\overline{E} \propto H_c(T)[H^{-1} - H_0^{-1}]$ where $H_c(T)$ is the thermodynamic critical field, H the applied one and H_0 a constant. The proportionality coefficient as well as the characteristic field H_0 depend sensitively on the level of diamagnetic susceptibility $P_f = -4 \pi M_f H$ at which experiments are performed. Our experiments show below 10 K the existence of an anomaly on the mean activation energy. Different possible alternatives to explain this phenomenon are considered, the most probable being a (thermally assisted) quantum tunneling of vortex lines.

Much attention has been recently given to dynamical (as and/or quasi-static) experiments on high- T_c superconductors. These experiments [1-16], usually interpreted in terms of giant flux creep [1-24], show that thermal activation acting on thin vortices pinned by point defects is capable to strongly reduce the critical current in the mixed state of these materials. Beyond this practical reason this subject deserves further studies for the following reasons :

— The recent literature tends to suggest that the concept of mean activation energy is not reliable (see e.g. Ref. [16]). In this paper we show that, on the contrary, this concept leads to very coherent results, if the level of diamagnetic susceptibility at which the experiments are performed, is kept constant.

— At the moment several attempts are made in order to distinguish between different creep models [5, 7, 9, 10, 17-21]. In particular the standard Anderson-Kim model [17-19] leads to a logarithmic time dependence of the magnetization $m(t) \approx \operatorname{Ln} t$ in the short time limit, and to an exponential one $m(t) \approx \exp - (t/\tau)$ in the long time one, whereas in the vortex-grass model [21-24] $m(t) \approx 1/(\ln t)^{\mu}$, $\mu > 0$, only for long time periods. An interpolation between the Anderson-Kim short time limit and the vortex-glass long time limit has also been proposed in order to interpret a large variety of magnetic creep experiments [5]. In most other approaches the authors assume that the Anderson-Kim model works and they use it as a basis to deal with experimental data. Conclusions are controversial. — In c-axis oriented YBa₂Cu₃O_{7- δ}, Shi *et al.* [10] find that $S = dm/d \ln t$ increases with the applied field H up to 2 kOe and then decreases in higher fields. In the same type of samples Campbell *et al.* [9] show that S constantly decreases with H between 0 and 40 kOe. In Bi₂Sr₂Ca_{0.8}Cu₂O_{8- δ} single crystals, Palstra *et al.* [6] obtain different regimes of field variations of their activation energy U_0 in which U_0 is always a decreasing function of the field (between 1 and 10² kOe). A more sophisticated way to extract $U_0(H)$ from their experiments led Inui *et al.* [7] to the conclusion that $U_0(H)$ continuously decreases in the same range of fields. Beyond the internal contradiction showed for each type of sample (Y or Bi systems), the Anderson-Kim relation, $dm/d \ln t \propto kT/U_0$, also shows a striking contradiction when comparing the conclusions obtained in Y and Bi samples.

We believe we have found a way to clarify this situation. First of all we do not presuppose the validity of any flux-creep model. We simply measure the temperature and field dependences of a characteristic time τ_f associated with the decay of the magnetization from initial value $M_{\text{ini}} \approx H/4 \pi$ to a final value $M_f = P_f H/4 \pi$ where $0 \leq P_f \leq 1$.

In our experiments we have measured the time dependence of the magnetization M, after a fast increase of the applied field from H = 0 to $H = H_m$, the measuring field. The initial value of the magnetization, is $M_{\rm ml} \approx H/4 \pi$ (no vortex has yet penetrated in the sample). We chose a final value $M_{\rm f}$ such as $M_{\rm f} = P_{\rm f} H/4 \pi$ where $P_{\rm f}$ is a dimensionless number smaller then or equal to 1 (Fig. 1). The choice of $P_{\rm f}$ is arbitrary but is must remain the same value for a given set of experiments (i.e. an experiment where we compare the effects of different values of H and T). Other sets of experiments with different values of $P_{\rm f}$ have also been performed. We shall show later that the results can be affected by changes in the values of $P_{\rm f}$.



Fig. 1. — (a) Zero field cooled magnetization M and (b) diamagnetic susceptibility $P = -4 \pi M'/H$ in Bi(Pb)SrCaCuO as a function of the applied field at various temperatures. The arrows show the effect of holding the applied magnetic field constant for 10 000 seconds. The time variations of M and P at constant fields are reproduced in each insert.

The mean relaxation time τ which defines the evolution of the magnetization from $M_i = H/4 \pi$ to $M_f = H \cdot P_f/4 \pi$ is given by :

$$\frac{1}{\tau} = \frac{\mathrm{d}(P/P_0)}{\mathrm{d}t} \bigg|_{P = P_f} \tag{1}$$

where P_0 is the initial susceptibility ($P_0 \approx 1$), for $t = \tau_0$, the attempt time. By measuring P(t) in the vicinity of P_f it is possible to determine dP/dt at $P = P_f$ and therefore to determine the mean relaxation time τ .

Expression (1) can be understood as follows. Taking into account the distribution of field dependent energy barriers n(E(H)), an analogy with magnetic systems shows that the decay of the normalized irreversible susceptibility should be given by ([28] and therein Refs.):

$$(1/2 P_0)(\mathrm{d}P/\mathrm{d}t) = k_\mathrm{B} T \int_0^\infty n(E(H)) \,\mathrm{e}^{-\lambda t} \,\mathrm{d}\lambda \tag{2}$$

where $\lambda = 1/\tau = 1/\tau_0 \exp(-E(H)/k_B T)$. Since n(E(H)) varies slowly in comparison to $e^{-\lambda t}$, equation (2) may be approximated as:

$$(1/2 P_0)(\mathrm{d}P/\mathrm{d}t) = k_\mathrm{B} Tn(\bar{E}(H)) \int_0^\infty \mathrm{e}^{-\lambda t} \mathrm{d}\lambda = k_\mathrm{B} Tn(\bar{E}(H))/t ,$$

Taking this expression for $P = P_{f}$, we get :

$$1/\tau = (1/2 P_0)(dP/dt)_{P=P_f} = 1/\tau_0 \exp(-\bar{E}(H))/k_{\rm B}T$$
(3)

where E(H) and τ are respectively the mean activation energy and the corresponding mean relaxation time associated with the relaxation of the susceptibility of level $P = P_f$. Note that τ could also have been defined by $1/\tau = (1/2 P_0)(dP/dt)_{P=P_f}$. In this case one would have to introduce the effective attempt time $\tau'_0 = \tau_0/k_B Tn(E)$.

We have shown that the field and temperature dependences are exactly the same for our two samples, YBa₂Cu₃O_{7-d} and Bi(Pb)₂Sr₂Ca₂Cu₃O_y superconductors. These samples were wellcharacterized disk-shaped YBa₂Cu₃O_{7-d} and Bi(Pb)₂Sr₂Ca₂Cu₃O_y ceramics. The magnetization experiments have been performed on a SQUID magnetometer after zero-field cooling at various temperatures and applied fields (Fig. 1a). The normalized diamagnetic susceptibility $P = 4 \pi M/H$ has been corrected for a factor of 10 %, the volume fraction of the superconducting phases, determined by X-ray diffraction having been found nearly equal to 0.90 for both samples [35, 36]. In the presence of a constant magnetic field the magnetization M decays with time (inset of Fig. 1). Figure 2a shows log (dP/dt) versus H, for different temperatures in the Bi-sample. Due to the narrowness of our measuring time window and to our constraint according to which the level of final susceptibility P_f must be the same for all the experiments, we cannot use very different field values at a given temperature. It is then difficult to determine the functional dependence E(H) from only figure 2a. However, if one assumed that $E(H) \propto (H_0 - H)^{\alpha}$, $\alpha > 0$, this plot should be linear down to fields large enough so that $\tau \approx \tau_0$. It is clear that this cannot be the case because linear extrapolations of « measured segments » of $[\ln (1/\tau)]^{1/\alpha}$ versus $(H_0 - H)$ at each temperature do not all intercept the same point, and even if one forced such an intercept to hold overcoming errors bars, then one would find a τ_0 value much larger than τ which is nonsense. Following a procedure used for disordered ferromagnets (25-28) we have assumed that E(H) might be proportional to 1/H. If one plots the same data versus the reciprocal magnetic field 1/H we observe that the



Fig. 2. — Magnetic field dependence of the penetration rate of flux dP/dt in Bi(Pb)SrCaCuO at various temperatures. (a) Semilog plot of P as a function of H for $P_f = 0.5$. (b) and (c) show the semilog plots of P as a function of 1/H. Each plot gives a set of convergent straight lines for $P_f = 0.5$ (b) and for $P_f = 0.089$ (c).

linear extrapolations of log $(1/\tau)$ determined at each temperature give a convergent set of straight lines intercepting at $H_0 = 9.1$ kOe and $1/\tau_0 = 2.5 \times 10^{11} \text{ s}^{-1}$ (Fig. 2b). Very similar results are obtained with the Y-sample with $H_0 = 2.9$ kOe and $1/\tau_0 = 4 \times 10^{12} \text{ s}^{-1}$ Figure 2b clearly shows that $\bar{E}(H, T) = (1/H - 1/H_0) g(T)$. The function g(T) is then determined from the plot of $g(T) = -T d \ln (\tau/\tau_0)/d(1/H)$ versus T (Fig. 3). In the same figure we have compared the obtained data point to the trial function $g(T) = g(0)[1 - (T/T_c)^2]^{\alpha}$ for our two samples ($T_c = 108$ K and $T_c = 91$ K for the Bi and Y ceramics). Above 10 K the agreement with the value $\alpha = 1$ is surprisingly good. Another trial function such as $(1 - T/T_c)^{3/2}$ gives a less good fit (note that we have not studied in details the region very close to T_c).

In the temperature range $0.1 < T/T_c < 0.8$, our experiments can be summarized by the following mean activation energy :

$$\overline{E}(H,T) = g(0) \left[1 - \left(\frac{T}{T_c}\right)^2 \right] \left[\frac{1}{H} - \frac{1}{H_0}\right]$$
(4)

with the following parameters for $P_{\rm f} = 0.5$:

Bi-sample ; $T_c = 108$ K, g(0) = 480 K kOe, $H_0 = 9.1$ kOe

Y-sample ; $T_c = 91$ K, $g(0) = 1\ 100$ K, $H_0 = 2.9$ kOe.

These parameters; H_0 and g(0) vary strongly with the provided values of P_f which characterizes the mixed (vortex) state of superconductors. Some experiments have been



Fig. 3. — Temperature dependence of $-[T \cdot d \ln (\tau/\tau_0)/d(1/H)]$ for two samples of Bi(Pb)SrCaCuO and YBaCuO for $P_f = 0.5$ (a) and for different values of P_f (b). Solid lines show the temperature variation of the function $g(T) = g(0)[1 - (T/T_c)^2]$, where T_c is the critical temperature.

performed for different values of $P_{\rm f}$, in the Bi-system. It is interesting to note that the field and temperature dependences on the energy barrier remain unchanged when $P_{\rm f}$ is different : $E \propto (1/H - 1/H_0)$ and $E \sim [1 - (T/T_c)^2]$ as shown in figures 2c and 3b. The variations of these parameters, $g(0, P_{\rm f})$ and $H_0(P_{\rm f})$ with $P_{\rm f}$ are given in figure 4.

This is the main result of this paper. It shows a very coherent behavior for two different high- T_c superconductors and for different vortex states. Furthermore the field dependence we



Fig. 4. — Variations of $g(0, P_f)$ and $H_0(P_f)$ with P_f . $H_0(0)$ obtained appropriately by an extrapolation, signifies $H_{c2}(0)$ in which the vortices penetrate completely in the sample.

obtained $\overline{E} \propto 1/H$ for $H \ll H_0$ has been suggested by Yeshurun and Malozemoff [1] and Tinkham [20]; $U_0 \propto 1/B$ and provided $P_f = Cst$, the flux density in the sample B is proportional to the applied field H. We also see here the importance of our choice of a constant level of diamagnetic susceptibility in the experiments. Concerning the thermal variations, we have found $\bar{E}(T) \propto T$ below a cross-over temperature $T_{\rm cr}$ and $\bar{E}(T) \sim [1 - (T/T_{\rm c})]^2 \sim$ $H_{\rm c}(T)$ above this temperature, where $H_{\rm c}(T)$ is thermodynamic critical field. This last result can be understood as follows. The pinning energy per vortex in unit length resulting from a local lowering of the vortex-core energy, is proportional to $H_c^2 \xi^2 \sim H_c$. $H_c \lambda \xi \sim \phi_0 H_c(T)$ (we have assumed __the proportionality $\lambda(T) \sim \xi(T)$ and the Ginzburg-Landau relation $\phi_0 = 2 \sqrt{2} \pi H_c \xi \lambda$ [33]). This variation is obeyed only above $T_{cr} = 6$ K in Bi-sample and $T_{cr} = 8$ K in Y-sample. Below T_{cr} , E(T) varies linearly with the temperature. A similar variation has been obtained in oriented particles of $YBa_2Cu_3O_{7-\delta}$ by Campbell et al. [9] and Xu et al. [34]. A detailed analysis of this variation led us to eliminate the influence of a sharp low energy distribution of relaxation times as well as the possibility of a cross-over between a single vortex regime and a vortex-glass one [21-23]. Following the same analysis as for disordered ferromagnets [25-28, 37] we define an effective temperature T^* which forces the relaxation time to follow the Arrhenius law :

$$1/\tau = 1/\tau_0 \exp\left(-E(H)/k_{\rm B}T^*\right).$$
(5)

At high temperatures i.e. at temperatures $T > T_{cr}$, we must have $T^* \equiv T$ and at low temperatures the anomaly occurring at T_{cr} , $T^* = T_{const}$. The variation of T_{cr} versus T can be obtained by the plot of

$$T^* = g(T/T_c) \frac{d \ln (\tau/\tau_0)}{d(1/H)}$$
(6)

as shown in figure 5. This plot clearly verifies this assumption. We get $T^* = 6$ K and 8 K respectively for the Bi and Y-samples. Writing the activation energy $E \approx BHV^*$ we get at low temperatures and as an example in H = 1 kOe an activation volume $V^* = 7 \times 10^{-20}$ cm³ showing that non-activated events are coherent over sizes of the order of $(40 \text{ Å})^3 \approx 10^4$ atoms. Note that at higher temperatures activation volumes are of the same order of magnitude showing that local irreversible jumps are rather of the size of vortex-cores than of that of vortex



Fig. 5. — Temperature dependence of $T/T^* = -[T/g(T)] d \ln (\tau/\tau_0)/d(1/H)$. The low and high temperature limits are indicated by the straight lines intercepting at T = 6 K and 8 K, for Bi(Pb)SrCaCuO and YBaCuO respectively. The insert shows the effective temperature T^* versus temperature T. The arrow the cross-over temperatures from thermal and quantum regimes.

separation. The plot of T^* versus T (Fig. 5) is suggestive of a thermal activation to a nonactivation relaxation regime very similar to that observed in the motion of magnetic domain walls in ferromagnetic systems [25-28, 37]. In the case of ferromagnetic systems the nonthermal process of relaxation is due to the quantum tunneling of magnetic domain walls in the weak dissipation regime. In the present case it results from the quantum tunneling of vortices in the strong dissipation regime. Such a phenomenon is very similar to the case of macroscopic quantum tunneling in Josephson junctions [30-32].

In conclusion we have shown that the dynamics of vortices in Bi and Y-samples of high- $T_{\rm c}$ superconductors can be described in a very coherent way by using the mean relaxation time τ defined at a constant level of diamagnetic susceptibility P_f . The mean energy barrier E which is associated with this relaxation time is found to be proportional to the reciprocal magnetic field in both non-oriented ceramics, $E \sim (1/H - 1/H_0) \sim (1/B - 1/B_0)$. Moreover, if P_f is not kept constant, the energy barrier depends strongly on this quantity (see Fig. 4). This result shows that magnetic relaxation experiments must be performed at a given level of $P_{\rm f}$ (or of magnetization in magnetic systems) otherwise the derived energy barrier E would be a mixture of the H and P_f dependences, and therefore the uncontrolled $E(P_f)$ variations would be attributed to field variations. The temperature dependence of the energy barrier is found to be the same for both samples above a cross-over temperature $T_c = 6$ K and 8 K, $E \sim [1 - (T/T_c)^2]$. Below their cross-over temperatures both samples show a change in the mechanism of relaxation : the relaxation is no longer thermally activated but independent of temperature. This is attributed to the quantum tunneling of vortices (and more particularly across sample surface energy barriers). This phenomenon is very similar to the quantum tunneling of the magnetization in ferromagnets [26-28, 37]. The volume associated with each elementary process is of the order of 10^4 Å. We have to note that in such a macroscopic system, as is the case in bulk ferromagnets [28, 38], the dynamics at low temperatures could strongly be enhanced by weak sample heating resulting from the restoration of vortex potential energy into kinetic energy especially since we are here in the strong dissipation limit.

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