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Low-temperature magnetic relaxation in $YBa_2Cu_3O_{7-\delta}$: Evidence for quantum tunneling of vortices

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Hall-probe measurements of an $YBa_2Cu_3O_{7-\delta}$ crystal in the range 0.1–1 K show a temperature-independent time-logarithmic magnetic relaxation. These results rule out conventional thermally activated flux creep, which predicts a $T^{2/3}$ dependence at the lowest temperatures if one takes into account the dependence of the barrier on driving force in a simple one-dimensional model. They are consistent with a phenomenological theory assuming a quantum tunneling process for vortex kink motion.

In the classical model of thermally activated flux creep,[1,2] magnetic relaxation of type-II superconductors in their critical state should disappear at zero temperature ($T=0$). Recently, however, Mitin[3] reported significant time-logarithmic relaxation down to 0.5 K in $Pb_{1.2}Mo_{6.4}S_8$ and suggested a $T=0$ vortex quantum tunneling process. However, with his next temperature point already at 2 K, he did not investigate the detailed low-temperature dependence, which is critical, we argue, for establishing quantum tunneling. Simanek[4] proposed another model based on fluctuations arising from local electron trapping and giving a more complicated T dependence.

Since small coherence lengths are expected to favor quantum tunneling of vortices, it is natural to look for $T=0$ relaxation in the high-temperature superconductors. Mota *et al.*[5] first showed evidence of low-T relaxation in these materials, though their results are hard to interpret because of the random orientations of the grains in their ceramics, because of the possibility of contributions from weak links between the grains, and because the fields were so low that a full critical state was not achieved. Various authors[6-8] have shown relaxation data down to 4.2 or even 1.6 K, which do not appear to extrapolate linearly with temperature to zero. However, as we shall show, such an extrapolation is not adequate to establish $T=0$ relaxation.

Most recently, Hamzic and co-workers[9] made a more complete study of oriented $YBa_2Cu_3O_{7-\delta}$ grains and found magnetic relaxation persisting down to 0.1 K. Although their data suggest a significant drop in relaxation rate to a finite value at $T=0$, the signal-to-noise ratio makes the precise T dependence uncertain. Another question is whether, in their ground crystallites, there could be small cracks or weak links which might dominate the low-T relaxation (though the results to be presented below appear to dispel this concern). Low-T relaxation has also been reported in Bi-Sr-Ca-Cu-O crystals.[9]

In this paper, we present new data on a well-characterized crystal of $YBa_2Cu_3O_{7-\delta}$ down to $T=0.1$ K. The results confirm the existence of strong low-temperature magnetic relaxation and show that it is essentially temperature independent as $T\rightarrow 0$. We argue that such a temperature independence is what should be expected for quantum tunneling of vortices.

Before presenting our experimental data, we describe the T dependence predicted by several theoretical models. In the standard thermally activated flux-creep model,[1,2] (TAFM) the activation energy U for flux-line hopping is assumed to vary linearly with current, i.e., $U(J)=U_0[1-(J/J_{c0})]$, where U_0 is the potential barrier in the absence of current density J, and where J_{c0} is the critical current density for which $U(J)=0$. The normalized relaxation rate for the magnetization M, or equivalently (in the critical state) for the current J, can be defined as $S\equiv d\ln(J)/d\ln(t)$, and in the TAFM model it is[10] $S_{\rm TAFM}=-[(U_0/kT)-\ln(t/\tau)]^{-1}$, where τ is a relaxation time related to the flux-line hopping attempt frequency.[7] Thus, at low-temperature $S_{\rm TAFM}(T)\propto T$. The pinning potential underlying this model is, however, rather unphysical, being a saw-tooth potential with slope discontinuities. Beasley, Labusch, and Webb[2] showed that a spatially smooth potential $U(x)$ leads instead to a nonlinear J dependence of the net barrier $U(J)$. In particular, for a sinusoidal barrier and in the limit $J_{c0}-J\ll J$, they showed that

$$U(J)=U_0[1-(J/J_{c0})]^{3/2}, \qquad (1)$$

where the $\frac{3}{2}$ power comes from the fact that the distance from the minimum to the maximum of the tilted barrier does not stay constant, as in the saw-tooth potential, but tends to zero as $J\rightarrow J_{c0}$.

Their argument can be generalized to any mathematically regular potential with an inflection point as a function of position. Expanding the potential around the inflection point at $x=0$ in a one-dimensional model, and adding the effect of a potential slope from a superimposed external current density J, we have $U(x)=c_1x(J_{c0}-J)-c_3x^3$, where the c's are constants. Determination of the extrema of this potential immediately gives Eq. (1). As shown recently by Geshkenbein and Larkin,[11] the $U(J)$ of Eq. (1), with the usual Arrhenius relation, then leads to the time-dependent current density

$$J(t)=J_{c0}\left\{1-\left[\frac{kT}{U_0}\ln\left(\frac{t}{\tau}\right)\right]^{2/3}\right\}, \tag{2}$$

which implies that at low temperatures $S_{\mathrm{TAFM}}\propto T^{2/3}$. Thus for physically meaningful pinning potentials, thermally activated flux creep predicts a vertical tangent for $S(T)$ in the limit $T\rightarrow 0$. It is certainly *not* linear as has often been assumed in the past.

However, at low enough temperature where these thermal processes are frozen out, one could expect some process such as quantum tunneling of vortices to dominate the relaxation process. According to the standard WKB approximation, we expect a tunneling frequency

$$\omega\propto\exp[-(a-bJ)], \tag{3}$$

and correspondingly a normalized quantum-tunneling relaxation rate

$$S_{\mathrm{QT}}=[\ln(t/\tau)-a]^{-1}. \tag{4}$$

Here a is a constant proportional to the square root of the difference between the barrier height and the energy of the quantum state. b is a further constant representing the lowest-order (linear-J) effect of an applied current density J, which tilts the barrier and modifies the tunneling rate. At low temperatures, as long as $S_{\mathrm{QT}}\gg S_{\mathrm{TAFM}}$, the decay rate can be shown to be T independent. At higher temperatures, $S(T)$ should cross over to the thermally activated dependence $S\propto T^{2/3}$. The crossover will be rather sharp if the activation and tunneling barriers are large. Thus, the highest-rate relaxation process dominates the magnetic relaxation; this is in contrast to transport measurements for which the voltage contributions of the two processes add linearly.

These theoretical predictions emphasize the importance of measuring the magnetic relaxation directly at ultralow temperatures rather than attempting extrapolations from a higher temperature range. This is what we undertake here, using a Hall probe technique similar to that in Ref. 9, but specially adapted[12] for the measurement of submillimeter-sized platelet crystals. A miniature In-Sb Hall probe of approximate dimensions $100\times100\times4000\ \mu m^3$, with wire contacts on the ends and sides for the current and multiple voltage leads, respectively, was sandwiched between two mica plates, with the $YBa_2Cu_3O_{7-\delta}$ crystal mounted with vacuum grease on top, centered above one pair of Hall contacts, while another pair remained far from the sample to measure the background field stability. The twinned $YBa_2Cu_3O_{7-\delta}$ crystal had dimensions $650\times390\times44\ \mu m^3$, with the c axis along the short dimension. It was prepared by a self-flux growth and oxygen annealing technique.[13] Its quality has been demonstrated by the sharp ac susceptibility and zero-field-cooled low-field dc magnetic transitions only a few tenths of a degree wide,[14] and by the dc Meissner fraction (field cooling) at low fields reaching one. The low-T zero-field J_c components are reported for a piece of the same sample in Table I of Ref. 15.

The Hall probe and sample assembly was mounted directly inside the mixing chamber of a dilution refrigerator,[16] along with a ruthenium oxide resistor for temperature measurement. The temperature could be set stably in the range 0.1–1 K by resistance heating of the chamber. The Hall-probe signal was monitored by a lock-in amplifier at 22 Hz. Fields of up to 3 T from an Oxford Instruments superconducting magnet system were applied along the c axis of the crystal, perpendicular within a few degrees to the main platelet face. The sum of signals at the same field reached in increasing and decreasing field cycles gave the Hall-probe calibration, which was linear in current but somewhat nonlinear in its field dependence. The difference of the signals, typically only one percent of the sum, gave the irreversible part of the hysteresis loop, distorted somewhat by the field-nonlinear Hall-probe characteristic. The conventional loop shape was confirmed at 5 K in a quantum design magnetometer, as shown in the inset to Fig. 1, with a low-field peak, of half width about 1 T, sitting on top of a field-independent background. The Hall-probe data indicated an increase of almost 25% in the height of the loop as the temperature dropped from 4.2 to 1 K, with no apparent change from 1 to 0.1 K within a 5% experimental scatter. The size of the signal was consistent with a field of order J_c times the crystal thickness, as expected from earlier studies[17] of the stray fields near plateletlike type-II superconductors.

Relaxation measurements were performed by first establishing a temperature, then cycling the field to 3 T, and finally descending to the operating field. We studied two such fields. The first was 0.2 T to probe the low-applied-field region where self-fields are expected to curve vortices toward the plane.[17] The value was chosen to keep the field well above the lower critical field. The second was 1.7 T to probe the region where vortices lie almost straight along the c direction. The value was chosen to be sufficiently below the maximum field to insure a well-defined critical state. Field and temperature overshoots, which can have a strong effect on relaxation,[7] appeared to be negligible within our measurement accuracy. The current for the field, operated with the superconducting switch in normal mode, was stable to several parts in 10^5, and the relaxation measurement was started immediately after the current for the desired field was established. Estimates of the effect of field changes smaller than our measured stability limit give negligibly small magnetization changes compared to those observed. Typical relaxation runs, with and without sample, are shown in Fig. 1(a), where the Hall-probe signal is plotted versus the natural logarithm of the time. A clear time-logarithmic relaxation of the sample is observed even at the lowest temperature. Signal-to-noise is limited by the stability of the lock-in amplifier itself and it becomes much better at the lower 0.2-T field, as shown in Fig. 1(b), where the background signal of the applied field is an order of magnitude lower than at 1.7 T.

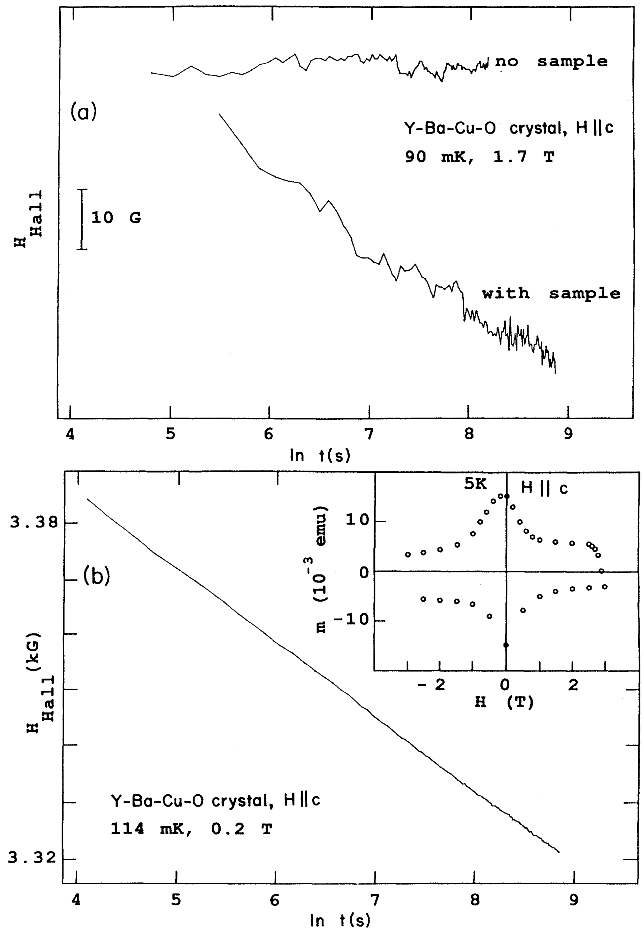

FIG. 1. Time dependence of the Hall probe signal H_{Hall} above an $YBa_2Cu_3O_{7-\delta}$ crystal in an applied field of (a) 1.7 T and (b) 0.2 T parallel to c, and the time dependence in the absence of the sample. The inset shows the SQUID magnetic hysteresis loop of the same sample at 5 K in the same field orientation.

The time-logarithmic relaxation rate, normalized by the size of the irreversible magnetization, is plotted versus temperature for the two fields in Fig. 2. These are our central results. This quantity is close to the normalized relaxation rate $S \equiv d\ln(M)/d\ln(t)$. The measurement at 0.2 T shows a rate essentially temperature-independent throughout the measured temperature range up to 1 K. The more scattered data at 1.7 T also indicates a temperature-independent behavior, with a higher average level. The normalized relaxation at 4.2 K in the same apparatus is in reasonable agreement with relaxation of the magnetic moment, determined by conventional superconducting quantum interference device (SQUID) magnetometry (with the usual precautions[18]) and shown in Fig. 3. This confirms that the Hall probe is not simply measuring some local redistribution of flux. The plateau above 20 K in the 0.2-T data, and above 5 K in the 1.7-T data has been recently explained[19] in terms of the vortex glass model. Because this model, which ignores quantum tunneling, predicts a crossover to conventional thermally activated behavior at low temperatures, it does not explain

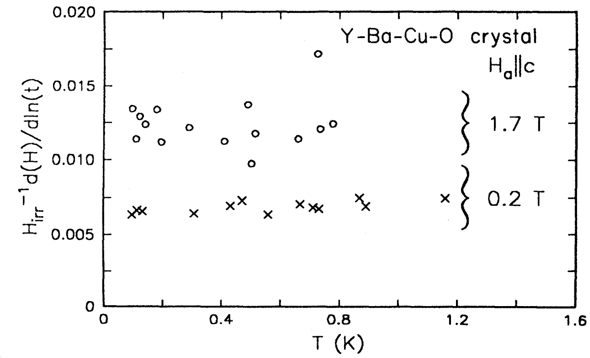

FIG. 2. Normalized relaxation rate of the Hall-probe signal H of the $YBa_2Cu_3O_{7-\delta}$ crystal vs temperature in the low-temperature range. H_{irr} is one half of the width of the Hall-probe hysteresis loop at the indicated applied field H_a.

the temperature independence at low temperatures. So the higher-temperature plateau seems to have a very different origin than the lower-temperature plateau of interest here.

Most earlier low-temperature relaxation data have been interpreted as showing a finite $T=0$ intercept and a relaxation rate S increasing linearly with T. Our more accurate results now show an essentially temperature-independent relaxation at the lowest temperatures, which agrees with our simple phenomenological theory and which therefore represents evidence for the existence of quantum tunneling of vortices. Further work is required to confirm in more detail the prediction of a crossover to $T^{2/3}$ behavior at higher temperature.

There are possible alternative explanations based on a distribution of barriers[10] extending to very low energies. Nevertheless, a distribution just such as to give a temperature-independent relaxation rate based on conventional flux creep seems implausible. Another interesting possibility is that the lowest barriers are surmounted in the presence of finite current density and contribute through a temperature-independent flux flow. Combined with a log-normal distribution of barriers, this can be shown to lead to a power-law dependence[20,21] of electric field on current density, and hence to an approximately time-

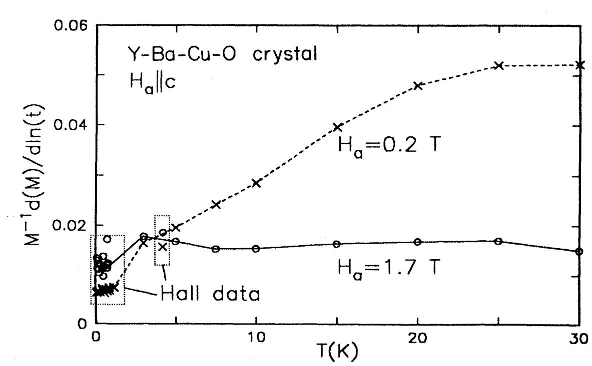

FIG. 3. Normalized relaxation rate of the magnetization M of the $YBa_2Cu_3O_{7-\delta}$ crystal, determined by SQUID magnetometry, vs temperature, compared to the Hall data (see Fig. 2) indicated in the dotted boxes.

logarithmic relaxation rate if the prefactor power in the log-normal distribution is large enough. The question here is whether such massive flux flow is compatible with the experimentally observed disappearance of flux flow below the irreversibility line at much higher temperatures in such crystals.[22]

Finally, we comment on the lower relaxation rate at 0.2 as compared to 1.7 T. A key difference between these two regions is that, as mentioned earlier, the vortices tend to curve toward the plane at low applied fields (because of self fields), while they are almost straight at high fields. This suggests that if quantum tunneling of vortices is the relevant process, then it is lower for vortices lying in the CuO_2 planes than that for vortices lying perpendicular to them. Because the flux configuration is quite complicated,[17] and because some contribution from the $\mathbf{B} \parallel c$ orientation is still present at low field, it is not possible, on the basis of the present data, to quantify the relaxation from the in-plane orientation. Nevertheless we might understand the qualitative difference by assuming that vortex motion is mediated by the motion of kinks.[23] For slightly bent vortices along the c axis, the kinks lie in the CuO_2 planes, and they move in the direction of the c axis; hence the distance between potential wells should be the greater of either the c coherence length ξ_c or the separation between CuO_2 planes. For vortices along the ab planes, the kinks lie along c and move along ab; so the longer coherence length ξ_{ab} most likely represents the nearest distance between potential wells. Since the tunneling rate in the WKB approximation is exponentially dependent on the barrier width, this suggests a mechanism whereby the relaxation might be faster for $\mathbf{B} \parallel c$ since in this orientation the kinks move the shortest distance. An alternative interpretation ignores the kinks but focuses on the larger electronic mass for the k vector perpendicular to the CuO_2 planes, and possibly a higher barrier, both of which would suppress tunneling in the WKB formalism.

In summary, large low-T magnetic relaxation gives evidence for quantum tunneling of vortices in a high-temperature superconductor, although barrier distribution models cannot be completely ruled out. Since a vortex is an extended object, and even a kink in the vortex core involves many electrons, the tunneling interpretation implies a new kind of macroscopic quantum tunneling. The tunneling probability and nature of dissipation may be quite different from the better known case[24] of Josephson junction systems and should be an interesting problem for future theoretical as well as experimental work.

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