Vanishing Magnetization Relaxation in the High Field Quantum Limit in YBa₂Cu₃O_{7-s}

G. T. Seidler, C. S. Carrillo, and T. F. Rosenbaum

NSF Science and Technology Center for Superconductivity and The James Franck Institute and Department of Physics, The University of Chicago, Chicago, Illinois 60637

U. Welp, G. W. Crabtree, and V. M. Vinokur

NSF Science and Technology Center for Superconductivity

and Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439

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We have investigated the magnetic response of untwinned single crystals of YBa₂Cu₂O_{7- δ} at millikelvin temperatures using a Bi thin film magnetometer of micron dimensions. Below T = 0.8 K, the magnetization relaxation rate S crosses over from thermally activated to quantum behavior. Above a sharply defined and strongly temperature-dependent threshold field, S disappears altogether. In concert with the vanishing magnetization relaxation, discrete steps appear in the magnetic hysteresis B(H), each of which corresponds to the "stick-slip" motion of 10³ vortices under the magnetometer.

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Pronounced low-temperature magnetization relaxation of superconductors has been reported for many different materials [1,2]. It is a general characteristic of all such measurements that nonzero relaxation rates are observed at the lowest temperatures of the respective studies, and that the relaxation rates extrapolate to a finite value at T=0. This behavior is incompatible with a thermally activated relaxation mechanism and has been taken as evidence for quantum tunneling of vortices in bulk superconductors.

The enhanced flux flow characteristics of the hightemperature superconductors make them especially attractive materials for investigating vortex dynamics [3]. In addition to the usual Abrikosov lattice [4], there appears to be a persistent vortex liquid state [5] as well as a frozen vortex glass [6,7]. The combination of a small superconducting coherence length and a high normal-state resistivity may also lead to large dynamical effects in the quantum limit [8].

We report here a detailed study of the magnetic hysteresis and the magnetization relaxation rate as T approaches zero for largely untwinned single crystals of YBa₂Cu₃O_{7- δ} with field parallel to the crystallographic c axis. In agreement with previous investigations [2], we observe a temperature-independent relaxation rate at fields of order 20 kOe. In contradistinction to those results, however, we find that the low-temperature vortex dynamics are suddenly quenched above a temperaturedependent threshold magnetic field. Concurrent with the disappearance of magnetization relaxation is a change in the form of the hysteresis curve B(H) from smooth to discontinuous, exhibiting many plateaus and steps with changing H. The present experiments reveal this unusual response because they probe higher fields at millikelvin temperatures than before, as well as sampling the hysteretic behavior continuously rather than episodically.

The bismuth gaussmeters used in this experiment were prepared photolithographically on a 0.3 mm sapphire substrate. The contact pads consisted of 100 nm of Au on top of 10 nm of Cr, defining an active area of 2 μ m×10 μ m (see lower inset to Fig. 3). With this size active region, a unit change of the number of vortices under the active region correspond to a change in field of 1 Oe. The thermally evaporated 500 nm thick bismuth films were granular with a grain size of 0.5 μ m and a Hall coefficient approximately 10% of the bulk Bi value.

Calibration of a gaussmeter without a sample in the top-loading dilution refrigerator showed only weak deviations from a linear Hall response. The response was temperature independent to 80 kOe for temperatures below 1 K and had significant temperature dependence at 3 K only for fields above 60 kOe. The resistance offset in zero field from mixing of the longitudinal resistive component due to slight contract misalignment was about 2 Ω , and was easily removed from the roughly 1 Ω/kOe response of the gaussmeter. The Hall probe response was smooth and nonhysteretic in applied field at all temperatures.

The Hall probe was measured by an ac lock-in technique at 16 Hz with a 3 s time constant and an rms current of 0.1 μ A. No change in the magnetization relaxation or hysteresis measurements was observed upon changing the measuring frequency by an order of magnitude, or by decreasing the measuring current by a factor of 3. This ensured that the weak ac magnetic field generated by the current in the Hall probe was neither affecting the sample response nor contributing to selfheating. Under typical experimental conditions the device resolution was 0.8 Oe.

The performance of the superconducting magnet is always a major limiting factor in the ways that the experimental parameter space can be accessed. Our magnet was always ramped in current controlled mode and exhibited no overshoot of the target field at the level of our 0.8 Oe sensitivity. The magnet ramping was linear in time at 8 Oe/s. Once in persistent mode, the logarithmic creep of flux into the superconducting magnet itself had a maximum rate of 2 Oe per decade in time at 75 kOe; this slow creep rate does not affect our conclusions. The magnet always relaxes in the same direction as it has been most recently ramped, prohibiting an artificial suppression of the true relaxation. No change in the magnetization relaxion rate was observed at 0.1 K by doubling or halving the magnet ramp rate.

A thin layer of Apiezon N grease separated the $20 \ \mu m^2$ active region of the gaussmeter from the YBa₂Cu₃O_{7- δ} surface. The samples were positioned so that a large untwinned region of the crystal being studied lay above the active region of the gaussmeter, at least 0.3 mm away from any edge. We report complete results on a $0.9 \times 1.4 \times 0.07 \ mm^3 \ YBa_2Cu_3O_{7-\delta}$ single crystal grown by a conventional flux method in a gold crucible and detwinned by applying uniaxial stress at elevated temperature [9]. Additional crystals exhibited the same general behavior, but with differences in the exact magnitude and field dependence of the response. The crystals had superconducting transition temperatures of 92 K, with inductive transition widths of less than 0.2 K.

In the magnetization relaxation measurements, the magnet was ramped from at least 30 kOe below the target field for final H > 30 kOe and, otherwise, was ramped from zero to the target. The presence of a complete Bean critical state in the sample was verified by the independence of the relaxation rate upon ramping upward from even lower starting fields [10]. The magnet was quickly switched to persistent mode after initial current stabilization, and the relaxation rate was then evaluated over the range $t = 10^2$ to 6×10^3 s. In all runs, including several extended to over 4×10^5 s, the data showed no deviations from log(t) behavior.

We plot in Fig. 1 the temperature dependence of the normalized relaxation rate $S = (1/M_0) dM/d \ln(t)$, where M(t) is the magnetization as a function of time and M_0



FIG. 1. The normalized magnetization relaxation rate S vs temperature T at two magnetic fields $H\parallel c$. At low H, S crosses over from a quantum (T independent) to a thermally activated regime above T=0.8 K, seen more clearly in the larger temperature range of the inset. At H=65 kOe, S suddenly disappears with decreasing T.

is the initial magnetization, for magnetic fields of 20 and 65 kOe. At H = 20 kOe the relaxation is temperature independent below 0.8 K, as has been seen [2] previously in a heavily twinned single crystal of YBa₂Cu₃O_{7- δ}. Such behavior has been ascribed to quantum tunneling of superconducting vortices. In the inset to Fig. 1, we show the temperature dependence of S up to 4 K. The crossover from quantum tunneling to thermal activation on increasing temperature becomes evident above T = 0.8 K.

Contrary to all previous investigations of vortex dynamics in high-temperature superconductors at very low temperatures, S(T) evaluated at 65 kOe shows a sharply defined threshold temperature below which we find no evidence for flux creep in the sample. At 65 kOe, this threshold temperature is approximately 300 mK, and the entire width of the step in S is less than 30 mK wide. The corresponding graph for S as a function of H is plotted in Fig. 2 for T=0.1 and 0.3 K. At both temperatures, the data show a well-defined threshold field above which no flux creep is observed. The width of the step in S at T=0.1 K is less than 300 Oe. Thus, there is a section of the H-T plane at low T and high H where the magnetization relaxation rate vanishes.

The absence of measurable magnetization relaxation necessarily implies a low or zero amplitude for the spectrum of relaxation times from very short times out to at least times of the order of 10^3 s. No general quantitative relationship exists between the spectrum of relaxation times for a system relaxing from metastable equilibrium and the steady-state response for the same system when forced from one local minimum in the free energy surface to another. However, the sudden disappearance of the fast relaxation modes which we observe should have unusual consequences for hysteresis measurements [i.e., the determination of B(H)] in the high field low-temperature regime.

We show in Fig. 3 hysteresis data taken at T=325 mK. The data shown are a series of half loops with pro-



FIG. 2. Analogous plot to Fig. 1 as a function of field at two temperatures. The magnetization relaxation rate is essentially constant until it abruptly vanishes at a temperature-dependent magnetic field.



FIG. 3. A series of nested hysteresis loops at T=0.325 K. Steps reproducibly appear above a threshold magnetic field H_{up} (H_{down}) on increasing (decreasing) field. Top inset magnifies the step structure for 42 < H < 47 kOe. Lower inset is a schematic of the Bi thin-film magnetometer.

gressively higher maximum fields which reproducibly overlay. Full hysteresis loops which sweep positive and negative in applied field are completely symmetric. The offset in B arises from remnant field trapped in the sample at the end of a previous hysteresis loop.

At high field on the increasing branch we find a crossover from smooth B(H) to a more complicated functional dependence with striking steps and plateaus. These steps lie within an uninterrupted extension of the B(H) curve and indicate a departure from the critical state. The steplike structure continues to much lower field on the decreasing branch, and we define two fields H_{up} and H_{down} as the indicated threshold fields. We present an enlargement of the region from H = 42 to 47 kOe in the upper inset of Fig. 3. The tailing in the steps is consistent with dynamics on a time scale much faster than 1 s being smeared by the 3 s time constant of our measurement apparatus. However, the gentle slope of the plateaus is not instrumental; at lower temperatures the plateaus become flatter, while at higher temperatures the plateaus become noticeably rounder and also the frequency of steps decreases. All crystals show similar features at low T and high H. Although steps in B become slightly more frequent with increasing H, neither a 1/H, 1/B, nor $1/B^{1/2}$ dependence describes their occurrence. We note that the typical step size of 1 kOe corresponds to a change of 10³ in the number of vortices under the active region of the gaussmeter.

We present the temperature dependence of H_{up} and H_{down} in Fig. 4. Solid lines are least-squares fits by $H_{up/down} = H_0 + AT^n$, with $n = 2.5 \pm 0.1$ and 2.6 ± 0.1 , respectively. By comparison, we also show where S goes to zero for T = 0.1 and 0.3 K (see Fig. 2). Clearly, there is a strong correlation between the disappearance of magnetization relaxation on increasing field and the qualitative change in B(H) occurring on increasing field at H_{up} .



FIG. 4. Temperature dependence of the threshold fields for step formation in B(H) and the disappearance of the magnetization relaxation rate S. Solid lines are least-squares fits approximately following a $T^{5/2}$ power law.

the response of the system in different ways is strong evidence for the intrinsic nature of the observed suppression of the quantum tunneling rate. We point out that the nontrivial temperature dependence and the splitting of the threshold fields exclude simple explanations based on matching of the vortex lattice spacing with some length set by pinning defects in the crystal.

Our B(H) data are reminiscent of measurements of flux penetration into hollow superconducting tubes. The observed "flux jumps" for the tube geometry occurred both at low fields prior to the establishment of the high field critical state and at high fields near H_{c2} , again removing the system from the critical state. The jumps were attributed to a breakdown of pinning from thermal instabilities caused by internal local heating during ramping of the external field [11].

Two major problems mitigate against the model of thermal instabilities explaining our B(H) data. First, the occurrence of thermal instabilities should be sensitively dependent on the ramp rate of the applied field. We find, however, that B(H) was unchanged by halving the magnet ramp rate at T=0.1 K. Second, instabilities should occur in identical regions in B on the increasing and decreasing branches of a hysteresis loop, as was observed in the experiments on tubular samples. In contrast, we have shown in Fig. 4 that not only are H_{up} and H_{down} nondegenerate, but they respond to temperature changes in markedly different fashions.

Given the difficulties in explaining the high field structure in B(H) as due solely to thermal instabilities, we point out that purely dynamical instabilities also may play a role [12]. Classical damped systems sometimes exhibit dynamical instabilities at small values of driving rate which disappear at higher rates where the system instead undergoes smooth motion. For example, this behavior is observed in the stick-slip "earthquakes" of the Burridge-Knopoff model [13] and in rotating-drum sandpile experiments [14], where small rotation speeds give avalanches and high rotation speeds give smooth flow of the granules. The velocity of vortices in a superconducting sample in the critical state exposed to a steadily changing external field *decreases* with increasing field. Hence, instabilities are expected only at high fields when of dynamical origin, as opposed to the two instability regimes observed in the tube experiments. Moreover, the analog to an independent H_{up} and H_{down} is seen in the sandpile experiment, where the critical velocity to be exceeded to achieve smooth flow is dependent on the recent history of the system [15]. We are still left, however, with the difficulty of explaining the insensitivity of the threshold fields to magnet ramp rate.

Regardless of the nature of the instabilities, a number of consequences of their presence need to be addressed. As instabilities prevent the superconductor from reaching the critical state, the absence of any measurable flux creep (be it thermal or quantum) follows immediately from the increased energy barriers. Nonetheless, we note that at both T=0.1 and 0.3 K the flux creep vanishes approximately 4 to 6 kOe prior to the first observed instabilities (see Fig. 4). The disappearance of tunneling might then be the first signature of some fundamental change in the superconducting state which leads to a greater proclivity for instabilities, marking the occurrence of an unusual quantum to classical crossover in the dynamics. The sharp demarcation between regimes occurs with decreasing temperature and increasing field, that is, upon moving deeper into the quantum limit.

Increased vortex-vortex interactions would cause both the quenching of quantum tunneling and stiffening of the vortex lattice, amplifying the tendency of the flux lattice to magnify small instabilities. Stronger interactions leading to the formation of vortex bundles is a viable possibility, with the approximately 1000 vortex change under the gaussmeter with a step in B(H) what one expects for a single vortex bundle [16]. On the other hand, the abruptness of the disappearance of the magnetization relaxation seems contrary to the present view of vortex bundles forming from a gradual increase in vortex-vortex interactions and wave-function overlap [17]. We point out as well that increased correlations resulting from a Bose condensation of vortices into a pinned zero-momentum superfluid are sufficient to explain the decreased tunneling rate and the onset of instabilities. The present data, however, do not address directly the strength of the vortex-vortex interaction.

In conclusion, we report that the flux creep present in $YBa_2Cu_3O_{7-\delta}$ at low temperatures is suddenly and se-

verely suppressed in high magnetic fields. In the regime where the relaxation disappears, hysteresis loops cease to be smooth, instead showing a complicated pattern of steps and plateaus. The data indicate a transition from quantum to classical dynamics of unknown origin deep in the quantum limit.

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