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Strong shift of the irreversibility line in high-T_c superconductors upon vortex shaking with an oscillating magnetic field

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Torque magnetometry is a powerful method for probing superconducting anisotropy in the mixed state. In order to use the three-dimensional anisotropic London model to analyze torque data, the vortex lattice must be in a reversible state, a state normally restricted to a narrow range close to the upper critical-field boundary $H_{c2}(T)$ because of large pinning effects that set in at lower temperatures T. We show that the application of an additional oscillating magnetic field perpendicular to the main field B leads to a fast depinning of the vortex lattice. This vortex-shaking process dramatically extends the reversible domain in the (H,T) phase diagram, and thus the range in which torque investigations can be made. [S0163-1829(98)51934-0]

Torque magnetometry has proven to be a useful method for determining superconducting parameters such as the effective mass anisotropy γ, the in-plane penetration depth λ_{ab}, and the in-plane coherence length ξ_{ab} in high-T_c cuprates. These parameters are derived by fitting the measured angular-dependent reversible torque with a theoretical expression derived by Kogan for a three-dimensional (3D) anisotropic superconductor in the mixed state.[1–4] The use of the London model, which provides a good approximation of the Ginzburg-Landau approach for large κ values, assumes a thermodynamic equilibrium of the mixed state. Unfortunately, in most superconductors, only a narrow reversible range in the field-temperature (H,T) phase diagram below the upper critical field $H_{c2}(T)$ can be explored, owing to the onset of irreversibility by pinning effects at lower T. Moreover, close to T_c, thermal fluctuations become dominant, and modify the torque signal significantly. Thus they must be taken into account in any analysis of the torque data.[5,6] One of the parameters governing the width of the fluctuation range is the amplitude of the effective mass anisotropy γ.[7] In some cases, a continuous crossover occurs between the fluctuation and flux-pinning regimes, thus hiding the reversible flux liquid range and rendering an estimation of γ almost impossible. Other constraints on the determination of γ close to T_c can be the smallness of the torque signal, the influence of a magnetic background of the sample itself, and/or a broad superconducting transition width ΔT_c due to inhomogeneities.[5] Hence it is vital that the reversible regime be extended as much as possible if one wants to investigate the evolution of the superconducting parameters at lower temperatures.

In the past decade, a tremendous effort has been made to investigate the pinning properties of the flux-line lattice (FLL) in high-T_c superconductors (HTS's) and to understand the nature of the irreversibility (IL) or melting line, using techniques such as dc magnetization, ac susceptibility, vibrating reed, torque, magneto-optics, and specific-heat measurements.[8–13] In the magnetic hysteresis loops $M(H,T)$ of many HTS single crystals, an anomalous peak or fishtail effect has been observed. In $YBa_2Cu_3O_{7-\delta}$ (YBCO) a possible origin of this effect is the presence of an inhomogeneous distribution of oxygen-deficient regions, which act as pinning centers. This mechanism is corroborated by the absence of the fishtail effect in well-oxygenated YBCO (123) samples.[14] The peak effect is also related to the pinning dynamics, and depends on the relaxation processes. As relaxation effects and measuring time play an important role in the determination of both the irreversibility line and the peak effect, it is of course tempting to check the influence an additional oscillating field has on them.

Already in the early 1970s, it was observed that small oscillating (ac) fields influence the depinning current density,[15] produce spectacular changes on the peak effect,[16] and can strongly modify the transport properties of type-II superconductors in the flux-flow state.[17] In the HTS $Bi_2Sr_2CaCu_2O_8$, the perturbation of irreversibility by an external stray ac field was reported by Farrell *et al.*[11] The theory of the penetration of magnetic ac fields into type-II superconductors or, equivalently, of periodic vortex tilting in vibrating-reed experiments[18] was established by Brandt *et al.*[19]

The main idea of our experiment is to shake the vortices out of their pinning potential and to force their relaxation by applying an additional ac magnetic field perpendicular to the main field $\vec{B}$. This technique allows us to create or extend the reversible regime in strongly pinned superconductors, in which so far no reversible torque curves could be obtained. Thereby we observe dramatic changes of the IL in the (H,T) phase diagram that depend on the amplitude and, to some extent, on the frequencies of the ac field.

Our experiment was performed with two different torquemeters, one based on a miniature piezoresistive cantilever,[20,21] the other on a macroscopic capacitive lever.[22] An NMR electromagnet generates the horizontal main field B up to 1.5 T, and can be rotated about a vertical axis at angular velocities ranging from 1° to 500°/min. The additional ac field b_{ac} up to 70 mT is generated by a vertical solenoid fixed between the poles of the NMR magnet. The stainless-steel He-flow cryostat that partially screens b_{ac} is rigidly mounted and mechanically decoupled from the solenoid to prevent vibrations. To achieve an optimal torque resolution, it is important that all vibrations be damped and stable sensors be used. We observe that the efficiency of the vortex shaking increases with the amplitude and frequency of the ripple field, but that the coil inductivity and the eddy currents set an upper limit to the frequency. Experimentally, an optimum between 500 Hz and 1 kHz has been found. In our case it is for reasons of sensor stability not possible to shake the FLL and simultaneously measure the torque signal. Therefore, we perform the angular- or field-dependent measurements in three phases. First we sweep the angle or field, then apply the ripple field b_{ac} to shake and relax the FLL, and finally acquire the data. This cycle is referred to as a ‘‘sweep-shake-measure’’ (SSM) sequence. Five parameters can be adjusted: the amplitude and frequency of b_{ac}, and the sweeping, shaking, and measuring times denoted by $t_1/t_2/t_3$, respectively. The first two parameters and t_2 are chosen according to the pinning-force intensity, while t_1 and t_3 control the measurement resolution. With this technique, it is possible to achieve reversible torque measurements at much lower temperatures than before.

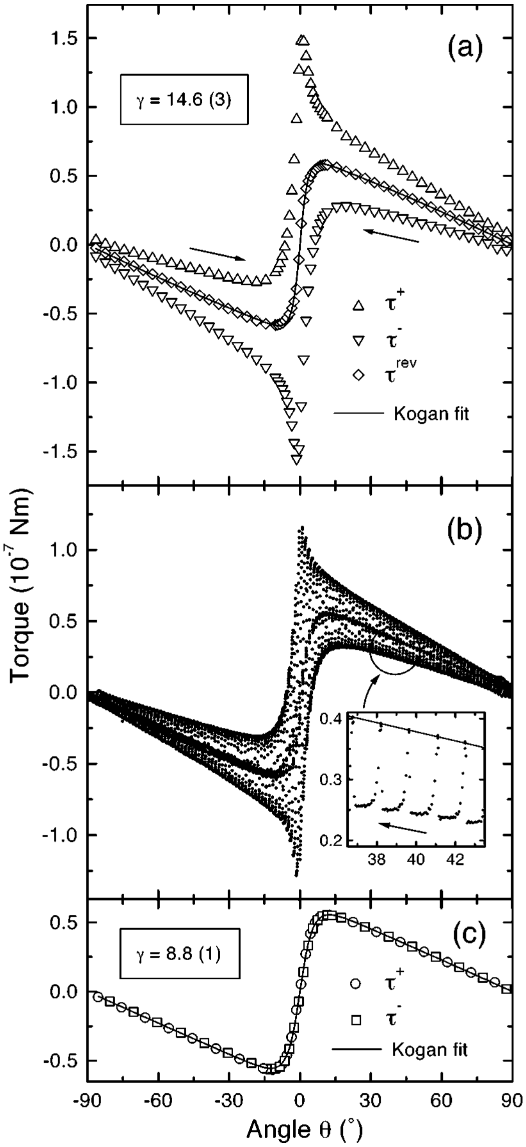

FIG. 1. Angular-dependent torque measurements in the ac plane of an untwinned single crystal of $YBa_2Cu_3O_{7-\delta}$. For $\vec{B}$ parallel to the ab plane of the sample, $\theta=0°$. (a) Torque for clockwise (τ^+) and counterclockwise (τ^-) rotations of $\vec{B}$ obtained without FLL shaking at $T=91.2$ K, $B=0.1$ T, and $d\theta/dt=110°/\mathrm{min}$. The solid line represents the fit to $\tau_{\mathrm{rev}}(\theta)\simeq\frac{1}{2}[\tau^+(\theta)+\tau^-(\theta)]$ with Kogan's expression, and leads to $\gamma\simeq14.6(3)$. (b) Similar measurements as in (a), but with vortex shaking. Details of relaxation sequences are shown in the inset. (c) The fit after FLL shaking gives $\gamma=8.8(1)$. The curves are fully reversible in this case.

We have investigated a high-quality YBCO (123) untwinned single crystal[14] grown in a $BaZrO_3$ crucible and containing no metallic impurities such as Al, Au, or Sn, which act as pinning centers.[4] The detwinning procedure was done by annealing the crystal under uniaxial pressure of 1 GPa in flowing O_2 at 500 °C for 30 min. This sample has a T_c onset of 93.5 K. A surprisingly large pinning remained in the crystal, which can be explained by the presence of oxygen clusters or separations into phases with $O_{6.5}$ and O_7 content.[14] In YBCO (123), the reversible regime at fields below 1.5 T is quite narrow. An example of an angular-dependent measurement in the irreversible regime is shown in Fig. 1(a), with an ac-anisotropy curve taken at 0.1 T and 91.2 K by rotating the field in the ac plane in absence of a ripple field (no FLL shaking). The reversible torque, given in good approximation by $\tau_{\mathrm{rev}}(\theta)\simeq\frac{1}{2}[\tau^+(\theta)+\tau^-(\theta)]$, can be fitted by the well-known expression that Kogan *et al.*[2] derived from a 3D anisotropic London model. By doing so, an unrealistically high γ value of 14.6(3) is obtained, which is explained by the large irreversibility that peaks at $\theta\simeq0$, i.e., for $\vec{B}$ close to the ab plane of the sample. The above expression for $\tau_{\mathrm{rev}}(\theta)$ is not suitable in this case. By now applying the shaking procedure to the crystal at the same field and temperature, a fully reversible curve is obtained as shown in Fig. 1(b). Here the five parameters of the SSM sequence are $f=500$ Hz, $b_{ac}\cong7.5$ mT, with a time cycle of 8s/4s/4s. Data have been acquired every 250 ms in order to follow each relaxation process closely as shown in greater detail in the inset of Fig. 1(b). However, for the final analysis, only the data points in the measurement phase of the SSM sequence are considered. Now the fit with Kogan's expression [see Fig. 1(c)] yields a realistic γ value of 8.8(1) for YBCO (123), in agreement with values reported earlier in the literature.[23,24] In torque magnetometry the IL is usually derived from the onset field where the upward and downward $\tau(B)$ curves, measured at a

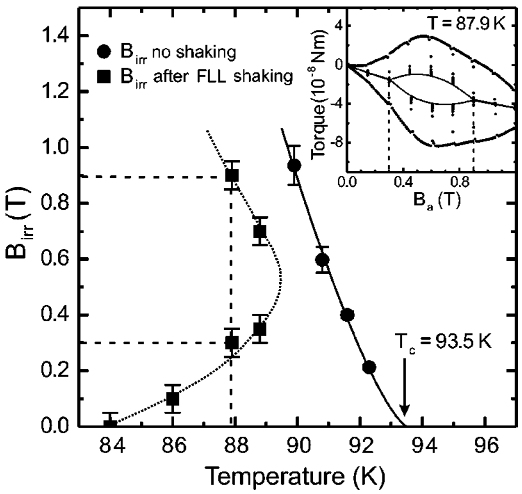

FIG. 2. Irreversibility lines extracted from field-dependent measurements (inset) for $YBa_2Cu_3O_{7-\delta}$ with and without FLL shaking. The field $\vec{B}$ is in the ac plane with $\theta=-81°$, and swept at $dB/dt=20$ mT/s. The solid line corresponds to a power law [(Eq. (1)] with exponent $n=\frac{4}{3}$. The dotted line is a guide to the eye. $\mu_0H_{c1}(T)$ is too small ($\simeq 15$ mT at 83 K) to be plotted here. Inset: the solid line is a spline to the torque after the relaxation process.

given angle and temperature, begin to overlap. An example of a $\tau(B)$ curve measured with the FLL shaking process is shown in the inset of Fig. 2. The external envelope represents the virgin hysteresis loops (data point); the full line is the spline to the points corresponding to the relaxed state of the FFL after a SSM sequence (Fig. 2). Here the field $\vec{B}$, applied at $\theta=-81°$ ($\theta=0°$ corresponds to the a direction), was swept at a rate $dB/dt=20$ mT/s. The SSM parameters are $f=581$ Hz, $b_{\rm ac}\cong 1.5$ mT, with a time cycle of 5s/8s/2s, while all other experimental parameters were kept constant. Clearly the hysteresis loop has collapsed in the low- and high-field range, yielding two well-defined field onsets marked by the vertical dashed lines. From these values it is possible to reconstruct the respective IL's before (solid line) and after (dotted line) the relaxation process. They are drastically different from each other. The ''untreated'' IL exhibits the well-known dependence that can be fitted with the power law[9]

$$B_{\rm irr}(T)=B_{\rm irr}(0)(1-T/T_c)^n \tag{1}$$

and exponent $n=\frac{4}{3}$. After the forced FLL relaxation, the IL becomes reentrant. Different orientations and/or sweep rates of the magnetic field were observed to modify details of the curve but not the pronounced ''nose'' structure.

To check whether the reversible torque obtained by shaking the FLL effectively corresponds to a thermodynamic equilibrium of the superconducting state, we have performed similar measurements on a material exhibiting very weak pinning. A good candidate is $Y_2Ba_4Cu_8O_{16}$ (248), which has a fixed stoichiometry, no oxygen vacancies, and can be grown in high-quality single crystals.[25] In the reversible regime, we verified that the superconducting parameters ξ_{ab}, λ_{ab}, and γ, obtained with and without FLL shaking, are identical within experimental error. The second check was to measure the torque angular dependence of a ''clean'' 248 crystal and compare it with that of a 248 crystal that was slightly doped with Al and thus more irreversible.[4] By analyzing the $\tau_{\rm rev}(\theta)$ curves of both samples measured with forced relaxation, we found the superconducting parameters to be in excellent agreement in the temperature and field ranges in which the ''clean'' sample was previously reversible and the Al-doped one fully irreversible. In fact, the SSM sequences in the undoped 248 sample ($T_c\cong 79$ K) were able to shift the irreversibility point at 0.4 T from 59 to 24 K, i.e., a drop of 35 K. The validity and strength of the FLL shaking technique are thus demonstrated. In particular, the influence of possible heating effects due to the ac field can be neglected. The extension of irreversibility to lower temperature opens larger windows on the effects of dimensional crossover. $\tau_{\rm rev}(\theta)$ is indeed found to deviate significantly from Kogan's expression for a 3D anisotropic superconductor at low temperature, in particular for field orientations close to the ab plane. The most plausible explanation of this deviation, apart from demagnetization effects, is the crossover from 3D to 2D behavior.[24,26]

We have observed similar dramatic FLL relaxation enhancements and shifts of the IL into a reentrant behavior by an additional ac field also in other crystals such as twinned YBCO (123) or $(La_{1-x}Sr_x)_2CuO_4$. The method works in the presence of various types of pinning centers such as twin boundaries, oxygen clusters, metallic dopants, precipitates, or dislocations. The reentrance observed here depends on the experimental parameters and hence is not intrinsic to the material itself, in contrast to the reentrant melting line of the FLL predicted theoretically by Blatter *et al.*[7] for highly anisotropic superconductors. In the latter case, the width of the vortex-liquid phase close to $H_{c1}(T)$ is very narrow and difficult to see experimentally. A recent study of the peak effect in $NbSe_2$ by ac susceptibility $\chi_{\rm ac}$ (Ref. 27) claimed the occurrence of a reentrant feature in the field and temperature regime close to T_c, but this was, in our opinion, wrongly attributed to the intrinsic melting-line reentrance. A collapse of the irreversibility line derived by $\chi_{\rm ac}$ was also reported earlier by Krusin-Elbaum *et al.*[28] close to T_c and interpreted as a thermal softening of the vortex core pinning.

Why are the effects of a perpendicular ac field so important? Part of answer is the enhanced depinning process due to the tilting of the vortices generated by surface screening currents; another is the existence of a distribution of pinning centers of different strengths that is described by the functional form $F_p=J_cB\propto b^n(1-b)^m$ for the pinning-force density. Here b is the normalized field H/H^*, where H^* can be chosen as H_{c2} or H_{irr}. The ac perturbation increases the depinning probability, and forces the relaxation primarily of weakly anchored vortices in the low- and high-field regions, which results in a dramatic modification of the shape of $F_p(b)$. Assuming an elastic pin-vortices interaction, the changes of the irreversibility line are expected to be governed by the elastic moduli c_{11}, c_{44}, and c_{66}, which are field dependent. During the FLL shaking, we observe an exponential relaxation of the magnetization M over more than four orders of magnitude towards its equilibrium value $M_{\rm eq}$, with a limitation given by the instrumental resolution. A typical example of $M(t)-M_{\rm eq}$ during one SSM cycle is displayed in Fig. 3. M increases as B is swept to its new value during time t_1, and then relaxes during the shaking time t_2. From the slope of the decay line in the log-lin plot, a depinning

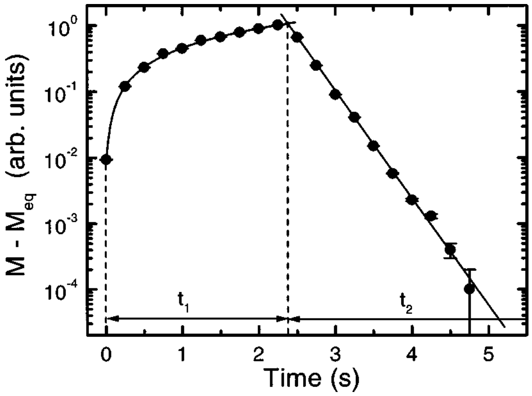

FIG. 3. Time dependence of the magnetization M within one SSM cycle: sweep of the main field B during time t_1, followed by the FLL shaking process with the ac field b_{ac} during time t_2. The relaxation is exponential over four orders of magnitude as the straight line fitted to the data points in the log-lin plot shows.

probability of 0.47% per cycle is derived. This relaxation process is obviously different from the thermally activated flux creep, which leads to a ln(t) dependence, and needs further investigation. The occurrence of the ''nose'' shape in the reentrant irreversibility line must in some way be related to the field dependence of the shear modulus c_{66}, which is proportional to b at low field and decreases as $(1-b)^2$ at high fields.[29,30]

In summary, we have shown that vortex shaking through an additional perpendicular ac field is a powerful technique to modify the irreversibility properties of the mixed states in HTS's. The strong relaxation of the magnetization induced by this process shifts and dramatically changes the shape of the irreversibility line derived from the field-dependent torque curves $\tau(B)$. The extension of the reversible region in the (H,T) phase diagram allows us to study the intrinsic anisotropy properties of the HTS's via the angular dependence $\tau_{\rm rev}(\theta)$ better. Some superconducting samples contain so much pinning that, without this technique, they simply would not be measurable at any temperature. This approach should shed more light on the problem of pinning in anisotropic material, and open the road to new theoretical explanations.

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