

## Equilibrium magnetization in heavy-ion-irradiated $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ probed by torque and SQUID magnetometry

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Measurements of the magnetization and torque on heavy-ion-irradiated  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  single crystals show that the equilibrium distribution of pancake vortices only depends on the applied field component parallel to the  $c$  axis, for fields both above and below the dose equivalent field and also for various irradiation angles with respect to the  $c$  axis. This observation is in clear contrast with the irreversible magnetization, which shows a large anisotropy with respect to the field alignment with the columnar defects. [S0163-1829(98)51526-3]

The introduction of amorphous columnar defects in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  (Bi-2212) by swift heavy-ion irradiation is well known to drastically enhance the pinning properties of this material at high temperatures. Moreover, the columns give rise to a pronounced angular dependence of the irreversible pinning properties (at temperatures  $>50$  K) when the field direction is rotated with respect to that of the columnar defects,<sup>1,2</sup> i.e., shielding and sustainable transport currents being stronger when the magnetic field is aligned along the irradiation direction. This observation in a highly anisotropic layered superconductor such as Bi-2212 indicates that vortices can behave as well-connected lines in this material,<sup>1,2</sup> in contrast to previous low-temperature experiments showing isotropic pinning.<sup>3</sup> The latter measurements had supported the common notion that vortices in Bi-2212 should be effectively decoupled into pancake vortices (i.e., the two-dimensional intersections of the vortices with the  $\text{CuO}_2$  planes).<sup>4,5</sup>

Two types of explanations have been put forward to explain the crossover between an isotropic pinning enhancement by columnar defects at low temperature and the anisotropic pinning at higher  $T$ : (i) the anisotropy is the result of a thermodynamic effect or (ii) it is a nonequilibrium effect, the result of a change in vortex dynamics as function of temperature. In the first case (i), the disappearance of the angular dependence of the magnetization as temperature is lowered is due to a redistribution of the pancake vortices over the columnar defects driven by the increasing pinning strength of the columns. This scenario supposes that at high temperatures vortex lines (stacks of pancakes) are adjusted to the columnar defects when the field is oriented along the irradiation direction, but form kinks of free pancakes when

the field is applied at an angle with respect to the irradiation direction. At low temperatures, the pancake vortices would adjust into a “locked” state in which they are mutually aligned on the columnar defects irrespective of the field angle. Two possible “locked” states could occur: either the pancakes belonging to the same vortex occupy the *same* columnar track, or they are spread over different tracks in order to produce a local field of the same orientation as the applied field.<sup>6</sup> Alternatively (ii), a crossover in vortex dynamics implies that at low temperature the critical nucleus for vortex activation from the columnar defects consists of a single pancake vortex, whereas at higher temperature only the activation of strings of pancakes can initiate vortex motion on large length scales. This is because the current necessary to delocalize a stack of pancakes, i.e., to make it move through the forest of columnar tracks, drops below the current necessary for the growth of the activation nucleus consisting of a single pancake.<sup>7</sup> Whereas the activation of a single pancake is an isotropic process,<sup>8</sup> that does not depend on the relative inclinations of field and columnar tracks, the activation of a string of pancakes is expected to be an anisotropic process.

In this paper we directly probe the distribution of pancake vortices over the columnar defects as function of angle by measuring the reversible torque of heavy-ion-irradiated Bi-2212 single crystals. Torque magnetometry has already proven itself to be a powerful tool for measuring the equilibrium properties and for studying the dimensionality of the vortex system.<sup>9</sup> By comparing our results with superconducting quantum interference device (SQUID) measurements, it is shown that the occupation of columnar tracks by pancake vortices only depends on the field component parallel to the sample  $c$  axis, i.e., the distribution of pancakes depends only

on the pancake density and not on the field direction. It means that the total pinning energy increases, and hence, that free pancakes are removed, not created, as the field is tilted away from the irradiation direction. By implication, the anisotropic pinning due to columnar defects must be a nonequilibrium effect.

The experiments are performed on two Bi-2212 single crystals grown by the traveling solvent floating zone technique<sup>10</sup> (typical dimensions:  $2 \times 2 \text{ mm}^2 \times 40 \text{ }\mu\text{m}$ ). A post annealing at  $800 \text{ }^\circ\text{C}$  in air is done in order to ensure a homogeneous oxygen content. The critical temperature  $T_c$  of both samples is equal to  $90 \text{ K}$ . The irradiation was performed at GANIL (Caen, France) with a beam of  $6 \text{ GeV}$  Pb ions (which traverse the entire specimen) with a density of  $n_d = 2.5 \times 10^{10} \text{ cm}^{-2}$ , corresponding to a dose equivalent field  $B_\phi = \Phi_0 n_d = 0.5 \text{ T}$  ( $\Phi_0$  is the flux quantum). For one crystal the beam was aligned with the  $c$  axis, whereas for the second sample the beam was oriented at an angle of  $60^\circ$  with respect to the crystal  $c$  axis. In this configuration the columnar defect density is reduced by a factor of 2, leading to an “effective”  $B_\phi$  of  $0.25 \text{ T}$ . On the other hand, the total volume occupied by the columns remains the same due to the increase of their lengths by a factor of 2 as a consequence of the large incident angle of the heavy ions. All samples were checked using magneto-optical technique for visualization of the flux penetration, and all exhibited homogeneous magnetic behavior.

We probed the equilibrium magnetization with the aid of (i) a (noncompensated) capacitive torquemeter and (ii) a SQUID magnetometer. The torque magnetometer is home built and consists of two equal phosphor-bronze capacitor plates (one holding the specimen) coupled in a Wheatstone bridge configuration. The setup is equipped with an external resistive magnet which can be rotated (maximum field  $H_a = 0.95 \text{ T}$ ; the angular resolution is better than  $0.001^\circ$ ). In the experiments described here we have fixed the field and rotated it from  $\Theta = 90^\circ$  to  $-90^\circ$  with respect to the  $ab$  plane of the crystal. At each angle, the torque per unit volume  $\vec{\tau} = \mu_0 \vec{M} \times \vec{H}_a$  is measured. Isothermal field sweeps in the more conventional configuration with the field along the  $c$  axis (where the torque is zero and the magnetization can thus not be obtained from torque data) were also performed, using a Quantum Design SQUID MPMS-5S magnetometer. A quartz tube sample holder with a temperature independent background of about  $-2 \times 10^{-5} \text{ emu}$  (at  $5 \text{ T}$ ) was used; our data were corrected for the background signal by subtraction of a measurement taken at  $110 \text{ K}$ .

For the analysis of the torque data, we follow the same procedure as used previously by Martinez and co-workers.<sup>9,11</sup> The torque is defined as:  $\tau = \mu_0 (M_z H_x - M_x H_z)$ , where  $M_z$  corresponds to the magnetization along the  $c$  axis of the crystal and  $M_x$  is the magnetization perpendicular to it.  $H_{x,z}$  are the applied field component perpendicular and parallel to the  $c$  axis, respectively. The Bi-2212 single crystals under consideration have such a large uniaxial magnetic and geometric anisotropy that the magnetic moment is oriented uniquely along the  $c$  direction. The contribution  $-M_x H_z$  can therefore be neglected; the  $z$  component of the magnetic moment can be obtained by simply dividing

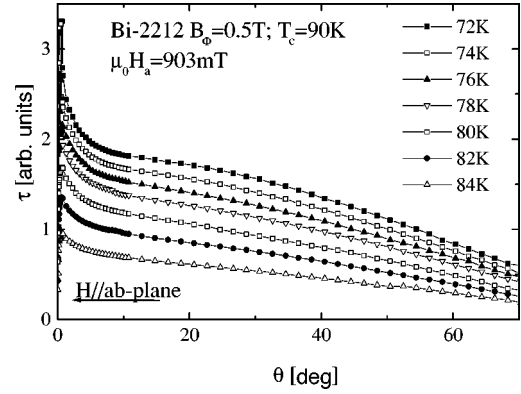


FIG. 1. Untreated voltage data from the torque magnetometer, for  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  irradiated with  $6 \text{ GeV}$  Pb, with dose-equivalent field  $B_\phi = 0.5 \text{ T}$ . Temperatures and field are indicated.

the torque by the field component parallel to the sample,  $H_x = H_a \cos \Theta$ , where  $\Theta$  is the angle between the field and the  $ab$  plane.<sup>9</sup>

Figure 1 shows the raw data of a torque experiment at  $76 \text{ K}$  (applied fields are indicated). No irreversibility is observed except for very small angles where the field orientation approaches the  $ab$  plane. The behavior of the irradiated specimen is rather similar to that of unirradiated Bi-2212;<sup>9</sup> however, the curves exhibit a slightly more convex shape which we shall analyze below in more detail. The typical width of the sharp peak near the  $ab$  plane is less than  $0.5^\circ$ , indicating little  $c$ -axis misorientation within the sample.

In order to test whether the magnetization depends only on the perpendicular field component  $H_z \equiv H_a \sin \Theta$  or also on  $H_x$ , we plot  $M_z \equiv \tau / H_x$  versus  $H_z$  in Fig. 2 for fixed temperature and various fields. A universal behavior is obeyed for all applied fields except for angles close to the  $ab$  plane (small  $H_z$ ) and angles close to the  $c$  axis (large  $H_z$ ). The latter deviation is an artefact caused by the fact that for  $H_a \parallel c$  the  $1/\cos \Theta$  term diverges. This deviation depends sensitively on the background correction used; its artefactual origin is clear because the magnetization at a given value of  $H_z$  is always equal to that at the same value of  $H$  applied along the  $c$  axis. The shape of the “universal” reversible torque curve is not sensitively influenced by the background offset. The second deviation at low angles (small  $H_z$ ), which

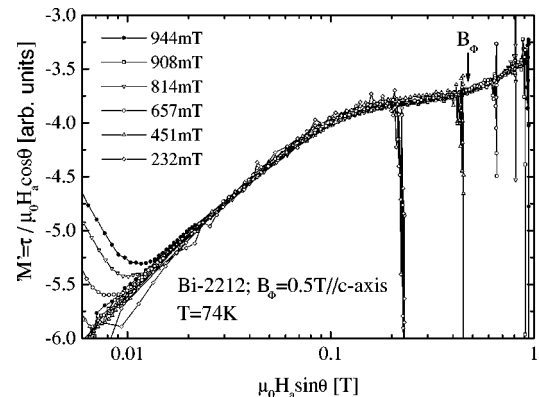


FIG. 2. Plot of the perpendicular magnetic moment  $M_z = \tau / H_a \cos \Theta$  derived from torque data, versus the perpendicular magnetic field  $H_z$ , for the same sample as in Fig. 1.

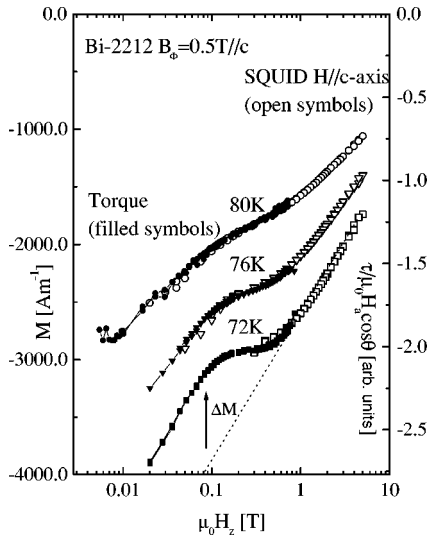


FIG. 3. Perpendicular magnetization  $M_z$  versus perpendicular field  $H_z$ , as determined by torque magnetometry (filled symbols), and by SQUID measurements (open symbols), at  $T=72$  K ( $\square$ ), 76 K ( $\nabla$ ), and 80 K ( $\circ$ ). For the sake of clarity the SQUID data are cut off where irreversibility sets in; note that we have also omitted the “diverging” torque data.

is accompanied by irreversibility, has been previously addressed as a surface-barrier effect, or as a consequence of a lock-in transition.<sup>12</sup>

The universal behavior depicted in Fig. 2 is one of the central results of this paper since it reflects the angular response of the vortex system: for the same value of  $H_z$  applied at different angles, the equilibrium magnetic moment is the same. This means that the magnetization  $M_z$  only depends on the perpendicular field component, i.e., on the density of pancake vortices in the  $ab$  plane. Note that this behavior is observed over the whole field range, from  $B \ll B_\phi$ , where the magnetization is determined by vortices localized on columns, to high fields  $B \gg B_\phi$ , where the magnetization is determined by free vortices. This is depicted in Fig. 3, where we have scaled the SQUID measurements onto the torque data. Although the measurement geometry is totally different for both methods, a striking match is apparent. This illustrates that the proportion of pancake vortices localized on the columnar defects with respect to the free pancakes also only depends on  $H_z$ , regardless of the field angle. As the field is tilted away from the  $c$ -axis direction, which is also the direction of the columns, the perpendicular field component  $H_z$  and hence the number of pancake vortices decreases monotonically. In addition, it follows from the field dependence of the reversible magnetization,<sup>13</sup> which corresponds to the free-energy change per added (or removed) vortex, that in the nonmonotonic part of the  $M(H)$  curve that the fraction of *free* pancakes swiftly decreases with decreasing  $H_z$ . Hence the pancake vortices are optimally adjusted to the columnar pins at all investigated temperatures. Extracting the zero-temperature pinning energy from the reversible magnetization<sup>13</sup> (from these data we obtain 1200 K), one finds that this does not depend on the angle between field and columnar defects, and therefore our results do not display the anisotropy found in transport properties.<sup>1,2,6,14,15</sup> It is interesting to note that the reversible

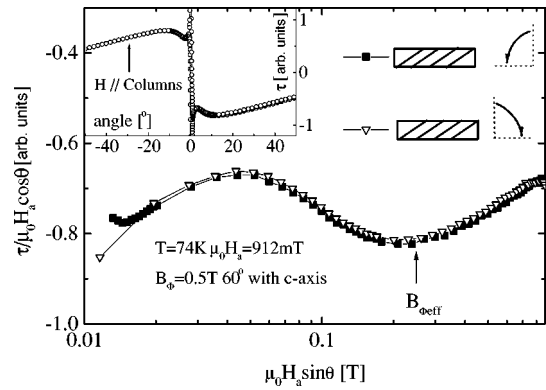


FIG. 4. Perpendicular magnetization  $M_z$ , extracted from torque data, versus the perpendicular field  $H_z$ , for the Bi-2212 crystal irradiated under an angle of  $60^\circ$  with respect to the  $c$  axis. The reversible magnetization is the same for field orientations close to, and nearly perpendicular to, the irradiation direction. Inset: Raw data used for the same temperature and field. It reveals that no features appear when the field becomes parallel to the columns ( $\Theta=30^\circ$ ).

magnetization can be measured to much lower fields (or temperatures) using the torque magnetometer than using the SQUID, even though the effective time scale in the torque experiments is considerably smaller than in the SQUID experiments. Since the pinning energy itself does not display the anisotropy related to field alignment with the columnar defects, this must be a direct consequence of the fact that the vortex creep rate is considerably enhanced when the field is *not* aligned with the irradiation direction (as in the torque experiment), with respect to the case where the field is aligned with the columns (as in the SQUID experiment). The anisotropy in the irreversible magnetization and transport measurements<sup>1,2,6,14,15</sup> therefore must be a consequence of the anisotropy in vortex dynamics only.

In Fig. 4, two magnetization curves (obtained from torque data), for the sample irradiated at  $60^\circ$  from the  $c$  axis, are depicted. One curve was obtained from a scan over the angular range  $0 < \Theta < 90^\circ$ , where the field becomes parallel to the columns during the scan, whereas the other was obtained from the angular scan over the range  $-90^\circ < \Theta < 0$  in which the field will be perpendicular to the column direction at a certain  $\Theta$  (depicted in the inset of Fig. 4). Very little difference is observed between the two measurements, confirming the fact that the pancake distribution in the reversible state is only dependent on the magnitude of  $H_z$ .<sup>16</sup> It should also be noted that in none of our reversible torque measurements was any special feature for precise alignment of the applied field with the irradiation direction ( $\Theta=30^\circ$ ) observed. We have thus been unable to find any evidence for a lock-in phenomenon where the pancakes belonging to the same vortex align to the same column.

The above results demonstrate that in Bi-2212, pinning of vortices by columnar defects can be described in terms of the pinning of two-dimensional pancake vortices at all field inclinations with respect to the columnar defect direction. At low magnetic fields, where all pancakes are localized on a columnar defect, an optimally adjusted state exists at all temperatures, in which the pancakes benefit maximally from the energy gain in the random potential due to the columns. As

has been shown by Koshelev *et al.*, pancakes belonging to the same stack do not necessarily have to occupy the same columnar defect,<sup>7</sup> although they should globally line up along the direction of the applied field. While the equilibrium magnetization is insensitive to the alignment of pancake vortices, we can compare its results to Josephson plasma resonance (JPR) experiments that do sense the pancake alignment.<sup>17,18</sup> These experiments directly probe the Josephson critical current along the  $c$  axis, itself determined by the relative phase shifts of the order parameter between layers and therefore by the correlations in pancake vortex distribution between the  $\text{CuO}_2$  layers. In turn, the JPR cannot sense how many vortices are trapped on a columnar defect; its measurement is thus complimentary to that of the reversible magnetization. Comparing the two experiments, we find that at low fields  $B \lesssim \frac{1}{5}B_\phi$  and high temperatures, there is a single JPR peak, the angular dependence of which is such that it occurs at fixed  $H_z$  (the field component parallel to the  $c$  axis and to the column direction). From the magnetization curves in Fig. 3, we find that at the same fields ( $B_z < 0.1$  T, at all angles) all pancakes are trapped by a columnar defect. It means that there exists a high-temperature ‘‘vortex liquid’’ state, in which pancakes are randomly positioned over column sites, and the  $c$ -axis phase correlation is progressively destroyed as more pancakes are added. At lower temperatures however, a second, high-field JPR peak

appears (at  $B \gg \frac{1}{5}B_\phi$ ), the position of which is nearly insensitive to the field angle. In contrast, the reversible torque measurements indicate that in this regime the pancake arrangement does depend solely on  $H_z$ . This indicates that in the field range larger than  $B \sim \frac{1}{5}B_\phi$ , all pancakes (irrespective of the fact of whether they are located off or on column) are lined up in a coherent way; an alignment which is induced by the available sites between the columnar defects. The onset of pancake alignment at  $B \sim \frac{1}{5}B_\phi$  occurs at a field where nearly all pancakes occupy a columnar defect, therefore any change in the reversible magnetization related to such a recoupling transition should be small.

In conclusion, we have established that the pancake distribution in heavy-ion irradiated  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  only depends on the field component parallel to the  $c$  axis of the crystal, and not on the field direction with respect to the columns. Furthermore, the creep rate is much more rapid for magnetic fields applied at an angle with respect to the columnar defect orientation. The apparent anisotropy in pinning measured in irreversible magnetization and transport measurements is a dynamic effect, with no direct relation to the equilibrium configuration of pancakes on the columnar defects.

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