Equilibrium Magnetization of a $Bi_2Sr_2CaCu_2O_x$ Single Crystal Measured up to 20 T

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The reversible magnetization of a high quality $Bi_2Sr_2CaCu_2O_x$ crystal has been measured by torque magnetometry at fields up to 20 T and temperatures ranging from 40 to 100 K. For fields above 7 T the superconducting diamagnetic moment follows a simple London-like behavior ($M_S \propto \ln B$). For lower fields, we observe a strong dependence of $dM_S/d\ln B$ on the magnetic field B. The results are discussed in terms of quantum and classical fluctuations of pancake vortices.

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$Bi_2Sr_2CaCu_2O_x$ is one of those rare superconductors where the reversible properties are directly accessible to the experimentalist in a large temperature and field range. An accurate analysis of the equilibrium properties is important to understand the different mechanisms that govern the behavior of high temperature superconductors (HTS). In particular, the study of the equilibrium magnetization at fields below 7 T and temperatures close to the thermodynamical critical temperature $T_{c0} \simeq 90$ K shows that classical fluctuations of vortices have a strong influence on the free energy [1–4].

Because of technical limitations, commercial SQUID magnetometers do not operate at fields higher than 7 T. Thus, the study of the reversible magnetization of HTS has been restricted to this upper limit. However, at high fields a sensitivity of the same order or better can be achieved by means of torque magnetometry. As we suggested in previous work, for highly anisotropic superconductors, it is possible to deduce the component of the magnetization perpendicular to the (a, b) plane $M_\perp(B_\perp)$ directly from torque measurements [4]. $B_\perp$ is the magnetic field along the c axis.

The objective of this work is to extend the study of the equilibrium magnetization of $Bi_2Sr_2CaCu_2O_x$ (Bi:2212) to fields up to 20 T in temperatures ranging between 40 and 100 K. We separate our discussion of the results into three main parts. In HTS the normal state presents an anisotropic paramagnetic contribution χ_V that has to be subtracted to obtain the superconducting diamagnetic contribution. We will start the discussion by explaining how χ_V is deduced. Next, we will show that the corrections to the mean field behavior due to classical thermal fluctuations cannot explain the equilibrium magnetization at low temperatures. Finally we will discuss the effect of quantum fluctuations of vortices at high fields and compare the theoretical predictions to our experimental results.

For this work we measured on a Bi:2212 single crystal ($2 \times 3 \times 0.1$ mm^3) prepared by the traveling solvent floating zone method [5]. The quality of this crystal was checked by x-ray rocking curves and by the angular dependence of the magnetic torque [4]. From those measurements we could determine that the mosaic spread of the crystal was smaller than 0.2°. We estimated the anisotropy factor $\gamma = (m_c/m_{ab})^{1/2}$ to be larger than 150.

The magnetic torque is measured by means of a compact capacitive torquemeter that can be inserted in a flow cryostat that fits in a 32 mm bore Bitter magnet. In this system static fields between 0.1 and 20 T are available. The sample and a calibration coil were fixed on a torsion wire. A current flowing through the coil compensates the torque due to the sample. The angular displacements are measured by a capacitive method. Even with the vibrations due to a water flow of 100 l/s to cool the magnet, the accuracy of the system is better than 10^{-10} N m. The temperature is regulated by a capacitive thermometer to better than $\Delta T/T = 10^{-3}$. We measured the torque as a function of the field. The (a, b) plane makes an angle $\theta = 88.5°$ with the field direction. Because of the narrow space available for the torquemeter it was not possible to change the orientation of the sample with respect to the field. The exact calibration was done by comparing the high field data with the results of rotation experiments performed in a torquemeter mounted in a rotating electromagnet (fields up to 1.2 T). All measurements have been carried out above the irreversibility line that lies below 35 K for fields above 0.1 T oriented almost parallel to the c axis.

Most of the contribution to the magnetic torque is coming from the c-axis component of the magnetization that we call $M_\perp(H_\parallel, H_\perp)$. For $\theta \gg 1/\gamma$ we can neglect the dependence on the parallel field component $H_\parallel$. In Bi:2212 this occurs at any angle above $\theta > 0.5°$ [4]. Under such conditions the torque per unit volume τ is related to the

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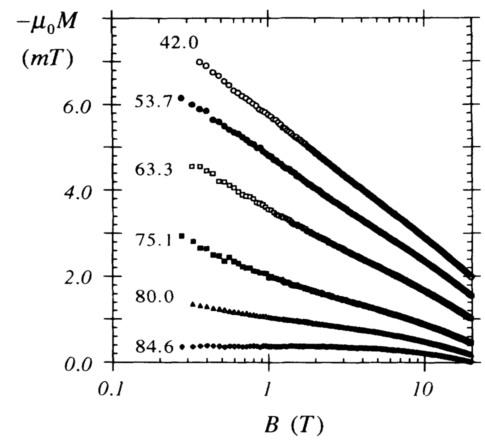

FIG. 1. Magnetization vs magnetic field B on a logarithmic scale. The different plots correspond respectively to the different temperatures indicated in the figure. These data were directly extracted from magnetic torque measurements.

perpendicular magnetization by $\tau = H \cos\theta\, M_\perp(B \sin\theta)$. Since $\theta = 88.5°$ we can assume $B \sin\theta \simeq B$.

In Fig. 1 we show $\tau/H\cos\theta = M_\perp$ as function of B for several temperatures between 42.0 and 84.6 K. The data in Fig. 1 were plotted on a semilogarithmic scale because a mean field theory, as well as a classical theory for thermal fluctuations, predicts a magnetization proportional to $\ln B$ in the regime $H_{c1} \ll H \ll H_{c2}$. The deviations from the expected logarithmic behavior become visible by plotting $dM_\perp/d\ln B$ vs B as we do in Fig. 2 for three typical temperatures between 47 and 100.6 K. We observe two different regimes. At low fields $dM_\perp/d\ln B$ decreases strongly with field while at high fields we observe a linear behavior with a slope which is practically temperature independent even above T_{c0}. Taking into account that the magnetic torque is sensitive only to the anisotropic part of magnetization, we conclude that the linear behavior is due to an anisotropic paramagnetic contribution which is mainly Van Vleck. This contribution is not affected by Cooper pairing and should indeed have a weak temperature dependence. We can consider that the magnetization is $M_\perp(B,T) = M_S(B,T) + \chi_V B$, where $M_S(B,T)$ is the superconducting diamagnetic signal and $\chi_V B$ is the paramagnetic contribution. The logarithmic slope of the magnetization is then given by

$$\frac{dM}{d\ln B} = \frac{dM_S}{d\ln B} + \chi_V B. \tag{1}$$

The linear behavior of $dM/d\ln B$ observed at high fields suggests that $dM_S/d\ln B$ is field independent for $B > 7$ T. From the linear behavior we deduce χ_V that is practically T independent and equal to $(1.5 \pm 0.2) \times 10^{-5}$ (SI). This value is in good agreement with the results of Johnston and Cho [6] obtained at temperatures above T_{c0}.

From now on we will concentrate on the superconducting contribution to the logarithmic slope. In Fig. 3 we plot $dM_S/d\ln B$ vs B. From a theory of thermal fluctuations in layered superconductors [3] it is expected that $dM_S/d\ln B$ would be given by

$$\frac{dM_S}{d\ln B} = \frac{\Phi_0}{8\pi\mu_0\lambda_{ab}^2} - \frac{k_B T}{s\Phi_0}, \tag{2}$$

where λ_{ab} is the penetration depth in the a-b plane and s the periodicity of the superconducting layers. The first term corresponds to the mean field behavior and the last is the correction due to fluctuations. This expression describes the experimental data in the vicinity of T_{c0}, but fails to explain the low temperature behavior at fields below 7 T because it predicts field independent $dM_S/d\ln B$. The corrections to the mean field behavior due to overlapping of normal cores were calculated by Hao and Clem and by Koshelev [7]. They predicted some small drop in $dM_S/d\ln B$ with B but on the scale of H_{c2} which is 1 order of magnitude larger than seen in Fig. 3.

To explain the results at lower temperatures, Bulaevskii and co-workers [8,9] proposed that quantum fluctuations of vortices affect the equilibrium properties of HTS. In the quantum approach the dynamics of a vortex

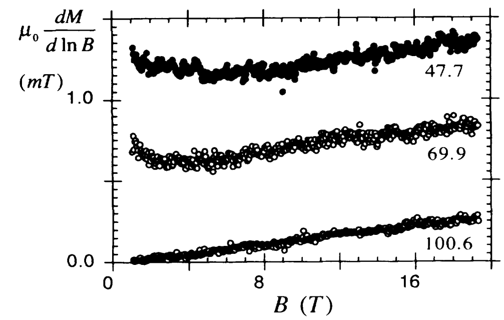

FIG. 2. $dM/d\ln B$ vs B obtained from Fig. 1 using a sliding linear fit on a window of seven data points (which correspond to a range of 0.2 T). A typical linear behavior in B is observed at high fields for 47.7, 69.9, and 100.6 K.

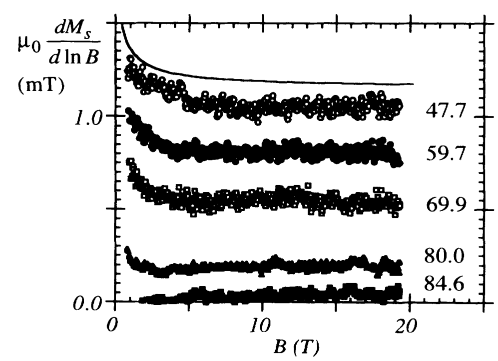

FIG. 3. $dM_S/d\ln B$ vs B for temperatures ranging from 47.7 up to 84.6 K. Here we see a clear saturation of the logarithmic slope for fields above 7 T. The full line corresponds to an estimation of $dM_S/d\ln B$ for $T = 40$ K using λ_{ab} and T_{c0} deduced from thermal fluctuations and $B_Q = 0.3$ T (see text).

lattice is important as was pointed out by Blatter and Ivlev in their treatment of quantum corrections to the melting line [10].

In a vortex lattice, individual vortices are submitted to an elastic interaction that will bring them back to the equilibrium position. Small distortions of the vortex lattice can then be treated as excitations in an oscillator. To stress the analogy with phonons the authors baptized vortex excitations as vortons. The difference with crystalline lattices comes from the fact that vortex motion is overdamped and a viscosity coefficient η has to be introduced. Up to now it was not clear if a Bardeen-Stephen model which gives $\eta_{\rm BS} = B_{c2}(\mathrm{T})\Phi_0\sigma_N$ can be used to describe the dissipation in HTS (which are in the clean limit).

By writing down the elastic modulus for distortions of the vortex lattice and using the solution of the overdamped quantum oscillator [11], Bulaevskii and coworkers deduced the contribution of quantum fluctuations to the free energy of highly anisotropic superconductors which is [8]

$$F(B,T) = \frac{B\Phi_0}{8\pi\mu_0\lambda_{ab}^2}\ln\frac{\beta B_{c2}}{B} + F_v(B,T), \tag{3}$$

where the first term in the right hand side of Eq. (3) is the mean field result. Here β is a numerical parameter of the order of 1 [7]. To calculate $dM_S/d\ln B$ the derivative of $F_v(B,T)$ with respect to elastic modulus is needed [9],

$$\frac{\partial F_v}{\partial\epsilon_i(\mathbf{k},q)} = \frac{1}{2s}\sum_i\int\frac{d\mathbf{k}}{(2\pi)^2}\int\frac{dq}{2\pi}\langle|\mathbf{u}(i,\mathbf{k},q)|^2\rangle. \tag{4}$$

The momenta along the (a,b) plane $(\mathbf{k})$ and the c axis (q) are restricted to the first Brillouin zone ($|\mathbf{k}|^2 \leq 4\pi B/\Phi_0 = K_0^2$ and $|q| \leq \pi$). The labels $i = t, l$ which appear in the elastic modulus $\epsilon_i(\mathbf{k},q)$ denote transversal and longitudinal components of the lattice distortions. The full mean squared displacement amplitude for an overdamped quantum oscillator $\langle|\mathbf{u}(i,\mathbf{k},q)|^2\rangle$ is [8,10,11]

$$\langle|\mathbf{u}(i,\mathbf{k},q)|^2\rangle = \frac{k_BT\Phi_0^2}{sB^2}\sum_m\frac{1}{\eta(\omega_m)|\omega_m| + \epsilon_i(\mathbf{k},q)}. \tag{5}$$

Here $\omega_m = 2\pi mk_BT/\hbar$ are Matsubara frequencies with m integer. At this point we can argue that vortons with energies $\hbar\omega_m$ of the order of the superconducting gap $\Delta(T)$ will result in effective excitation of quasiparticles, and the vortex as an object becomes meaningless. This suggests to introduce a cutoff in such a way that the sum over m in Eq. (5) is restricted to $\omega_m \leq \Omega \sim \Delta(T)$. In [9] a smooth cutoff was introduced by taking $\eta(\omega_m) = \eta_0[1 + (|\omega_m|/\Omega)^n]$; the simple rough cutoff $[\eta(\omega_m) = \eta_0$ and $\omega_m \leq \Omega]$ corresponds to infinite n. The latter gives field independent $dM_S/d\ln B$ at high field in accordance with experimental data shown in Fig. 3.

With the help of Eqs. (3), (4), and (5) we obtain the logarithmic slope of the magnetization. The classical expression Eq. (2) originates from the term with $m = 0$. It is valid for temperatures $\hbar\Omega < 2\pi k_BT$ or approximately $\Delta(T) \leq T$. The quantum correction (terms with nonzero m) is important at all temperatures except near T_{c0}. At low temperatures the sum can be transformed into an integral, and we get for infinite n

$$\frac{dM_S}{d\ln B} = \frac{\Phi_0}{8\pi\mu_0\lambda_{ab}^2} - \frac{\hbar\Omega}{2\pi s\Phi_0}\frac{B}{B_Q}\left[2\ln\left(1+\frac{B_Q}{B}\right) + \frac{1}{4}\ln\left(1+\frac{4B_Q}{B}\right) - \frac{B_Q}{B+B_Q}\right]. \tag{6}$$

In Eq. (6) $B_Q = \mu_0\lambda_{ab}^2\eta_0\Omega/\Phi_0$ is the field scale that characterizes the quantum corrections. Equation (6) is valid for fields above a crossover field $B_{\rm cr}$ which separates three- and two-dimensional behavior of the vortex lattice. In our case $B_{\rm cr} < 0.1$ T, and all our results are in the two-dimensional regime. For low fields the quantum correction is small, and the mean field result is valid:

$$\frac{dM_S}{d\ln B} = \frac{\Phi_0}{8\pi\mu_0\lambda_{ab}^2}, \quad B \ll B_Q. \tag{7}$$

As B increases, $dM_S/d\ln B$ drops, but in high fields saturation takes place and $dM_S/d\ln B$ becomes constant:

$$\frac{dM_S}{d\ln B} = \frac{\Phi_0}{8\pi\mu_0\lambda_{ab}^2} - \frac{\hbar\Omega}{\pi s\Phi_0}, \quad B \gg B_Q. \tag{8}$$

Comparing Eqs. (7) and (8) we see that the difference between low and high field values of $dM_S/d\ln B$ at low temperatures is determined by the cutoff frequency and interlayer spacing.

In Fig. 4 we plot the saturation values of $dM_S/d\ln B$ at high fields vs T. The analysis of this data must be separated in two regions. At high temperatures and close to T_{c0} ($T > 70$ K) we are in the classical region and the data must be fitted by Eq. (2). The temperature where the magnetization is field independent is $T^* = 84.3 \pm 0.9$ K. At T^* we obtain for our sample $M_S(T = T^*) = 0.3$ mT. Assuming that $s = 1.54$ nm (distance between two $\mathrm{CuO_2}$ double planes) we compute [3] $M_S(T = T^*) = 0.45$ mT in reasonable agreement with this value. By assuming for $1/\lambda_{ab}^2$ a linear temperature dependence we obtain a London penetration depth $\lambda_{ab}^2(T = 0) = 190 \pm 30$ nm. From our analysis we deduce that the thermodynamical critical temperature is $T_{c0} = 93$ K. Using the results obtained from classical thermal fluctuations and using Eq. (8) to fit the low temperature data we deduce the cutoff frequency, $\hbar\Omega/k_B = 460 \pm 30$ K. We used for the fit $1/\lambda_{ab}^2(T) \propto 1-(T/T_{c0})^4$ which is the temperature dependence of the penetration depth in a two fluid model. The value obtained for Ω is of the same order of magnitude as estimates of the superconducting gap, $\Delta/k_BT_c \sim 3-6.4$, from tunneling data [12].

Note that the drop in $dM_S/d\ln B$ vs B which we at-

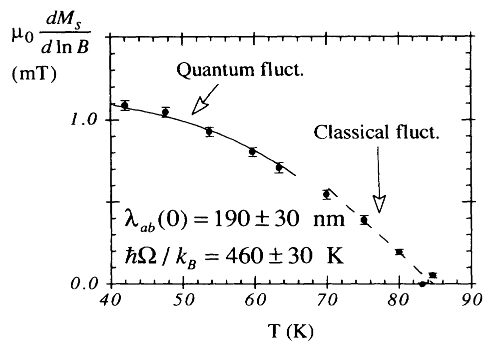

FIG. 4. $dM_S/d\ln B$ vs T for fields above 7 T (bars). The full line corresponds to a fit by quantum fluctuations using Eq. (8). In this range we used for $1/\lambda_{ab}^2$ a temperature dependence given by a two fluid model. The dashed line corresponds to the region of classical fluctuations. The fitting parameters are indicated in the figure.

tribute to quantum fluctuations is observed at $T \leq 75$ K. This is in agreement with our estimate of the region of quantum regime, $T \leq \hbar\Omega/2\pi k_B \sim 460/2\pi$.

In Ref. [9] $n = 4$ and $\hbar\Omega/k_B = 830$ K provided the best fit. However, the measurements in Ref. [9] were done in the limited field interval $B < 7$ T and do not allow an accurate determination of n and Ω.

Coming back to Fig. 3, the full line shows the result of Eq. (6). We used the values obtained for λ_{ab} and Ω and adjusted Eq. (6) to our data at $T < 60$ K by using B_Q as a fitting parameter. For the lower temperatures we obtained $B_Q \approx 0.3$ T. Since the quantum correction in Eq. (6) is temperature independent we represent, for sake of clarity, the theoretical $dM_S/d\ln B$ only for 40 K.

From the saturation field B_Q and the values obtained for $\lambda_{ab}(T)$ and Ω it is possible to estimate η_0 and compare the result with $\eta_{\rm BS}$. At 42.0 K we obtain $\eta_0 \approx 2 \times 10^{-10}$ kg/m s. By assuming that $B_{c2} \sim 60$ T at 40 K and extrapolating from the resistivity above T_c $\rho_n(T = 40$ K$) = 35$ $\mu\Omega$ cm [13] we can calculate the Bardeen-Stephen viscosity coefficient $\eta_{\rm BS} = 3.5 \times 10^{-7}$ kg/m s. We see that η_0 is 3 orders of magnitude smaller than $\eta_{\rm BS}$. This problem is not yet understood [9]. Our η_0 and B_Q are 1 order of magnitude smaller than the values determined in Ref. [9]. This discrepancy may be solved by realizing that Eq. (6) was obtained for a solid vortex lattice while the measurements were performed in the reversible regime, which corresponds to the vortex liquid state. This has been recently shown by small angle neutron scattering experiments in similar single crystals [14]. In Eq. (6) the saturation occurs into two steps related to shear and compression deformations at $|k| = K_0$. For a vortex liquid the shear contribution [i.e., the second term between brackets in Eq. (6)] should be considerably reduced and the saturation of $dM/d\ln B$ should occur at lower fields. Ignoring the shear term in Eq. (6) an equally good fit can be obtained as before, but with values $B_Q = 2$ T. By keeping Ω as well as a fitting parameter we obtain $\hbar\Omega/k_B = 600 \pm 100$ K. These values would give $\eta_0 = 1.1 \times 10^{-9}$ kg/m s.

In conclusion, the diamagnetic equilibrium magnetization is obtained at fields up to 20 T in Bi:2212. The logarithmic slope of the reversible magnetization on the magnetic field drops and then saturates for $B > 7$ T. A mean field model predicts an almost constant slope in our range of fields. Deviations from mean field behavior are explained by quantum fluctuations of vortices in the melted vortex state. The low values obtained for the viscosity coefficient η_0 were not understood until now.

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