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**DOI**

[10.1103/PhysRevLett.69.2276](https://doi.org/10.1103/PhysRevLett.69.2276)

**Publication date**

1992

**Published in**

Physical Review Letters

[Link to publication](#)

**Citation for published version (APA):**

Martinez, J. C., Brongersma, S. H., Koshelev, A., Ivlev, B., Kes, P. H., Griessen, R. P., Groot, D. G. D., Tarnavski, Z., & Menovsky, A. A. (1992). Magnetic-Anisotropy of a Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>x</sub> Single-Crystal. *Physical Review Letters*, 69(15), 2276-2279.  
<https://doi.org/10.1103/PhysRevLett.69.2276>

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## Magnetic Anisotropy of a $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ Single Crystal

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(Received 17 July 1992)

New magnetic torque measurements on a high-quality  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$  single crystal show that only a lower bound for the anisotropy parameter  $\gamma > 150$  can be given. For misorientations of the field  $H$  from the  $(a,b)$  plane larger than  $0.5^\circ$ , the results can be rescaled to magnetization  $M$  vs  $H$ . The behavior of  $M$  can be consistently described by a recent theory of Bulaevskii, Ledvij, and Kogan.

PACS numbers: 74.70.Vy, 74.60.Ec, 75.30.Gw

From numerous recent publications [1], it has become clear that the anisotropy plays a crucial role in understanding the phenomenology of high-temperature superconductors (HTS). Flux pinning, flux creep, and fluctuations in the extremely anisotropic Bi and Tl compounds clearly demonstrate two-dimensional (2D) characteristics in a large range of the field-temperature ( $B$ - $T$ ) phase diagram. The amount of anisotropy is determined by the ratio of the Josephson-coupling energy between the superconducting  $\text{CuO}_2$  double (triple) layers and the condensation energy. It can be expressed in terms of the ratio of the respective effective masses as  $\gamma = \sqrt{m_c/m_{(a,b)}}$ . For  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$  (Bi:2212) a value of  $\gamma$  between 55 and 200 is quoted [2, 3], and for  $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_x$  Gray, Kampwirth, and Farrell estimated  $\gamma = 90$  [4]. These values of  $\gamma$  were obtained by comparing experimental magnetic torque data with the anisotropic Ginzburg-Landau (AGL) expression in the London limit derived by Kogan [5]. However, the determination of  $\gamma$  and more generally the analysis of the reversible magnetic behavior in terms of AGL seems to be invalidated for strongly anisotropic HTS. Farrell *et al.* [6] showed that even for the less anisotropic  $\text{YBa}_2\text{Cu}_3\text{O}_7$  (Y:123) with  $\gamma = 7.9$  an AGL description breaks down for temperatures below  $0.8T_c$ . For lower temperatures a model of a stacking of superconducting planes coupled by the Josephson effect (Lawrence-Doniach model) gives a better agreement with the experimental results [7].

These problems in the analysis of torque and the availability of samples of better quality call for a new detailed investigation of highly anisotropic systems like Bi:2212. This Letter reports a detailed study on a high-quality single crystal of the Bi:2212 compound in the reversible regime close to  $T_c$ . It is shown that for  $\gamma$  only a lower estimate can be given which is larger than the values quoted in early works. The results for  $\theta > 0.5^\circ$  can be rescaled to magnetization  $M$  vs  $H$  curves which can be consistently described by a recent theory of Bulaevskii, Ledvij, and Kogan [8].

For this work we measured a Bi:2212 single crystal ( $2 \times 3 \times 0.1 \text{ mm}^3$ ) prepared by the traveling solvent floating zone method [9]. The transition temperature is estimated to be close to 85 K (see below). The volume  $V = 6.76 \times 10^{-10} \text{ m}^3$  is determined from the mass and the theoretical density of  $6.8 \pm 0.1 \text{ g/cm}^3$ . The latter is deduced from the composition obtained from the microprobe analysis:  $\text{Bi}_{2.15 \pm 0.05}\text{Sr}_{1.95 \pm 0.05}\text{Ca}_{1.03 \pm 0.04}\text{Cu}_2\text{O}_{8-x}$ . The single crystallinity of the sample was checked by x rays. In our better samples we observed a remarkably narrow distribution of orientations of the  $c$  axis with a half width at half maximum (HWHM)  $\varphi_0 = 0.1^\circ$ . The presence of small amounts of the phase Bi:2201 (phase with one  $\text{CuO}_2$  plane per unit cell) [10] suggests that the distribution of orientations comes from the existence of stacking faults in our single crystals. The magnetic torque is measured by means of a calibrated capacitive torque meter with a resolution of  $10^{-11} \text{ N m}$  [11]. The precision of the calibration is better than 1%. Fields up to 1.5 T, which are stabilized within  $1 \mu\text{T}$  using a Hall probe, are provided by a rotating electromagnet. The temperature is regulated by a ruthenium-oxide thermometer to better than  $\Delta T/T = 10^{-3}$ . The torque is measured as a function of the angle  $\theta$  between the  $(a,b)$  plane ( $\text{CuO}_2$  planes) and the field direction. At low angles the magnet is rotated with a constant angular velocity of  $0.1^\circ/\text{s}$ . Data sampling at a rate of 20 points per second yields an angular resolution of  $0.005^\circ$ .

The magnetic torque per unit volume  $\tau$  is given by

$$\tau = \mu_0(\mathbf{M} \times \mathbf{H}), \quad (1)$$

where  $\mathbf{H}$  is the applied field and  $\mathbf{M}$  the magnetization. In Fig. 1 typical results are presented in a plot of  $V\tau/\mu_0H$  vs  $\theta$  for a fixed field  $\mu_0H = 1.4 \text{ T}$  at various temperatures ranging between 77.4 and 84.4 K. A peak much sharper than previously reported [2, 3] is observed for  $\theta < 0.5^\circ$ . This sharp maximum decreases with increasing temperature until a smooth background remains for temperatures close to  $T_c$ . A blowup of the small-angle results at 77.4

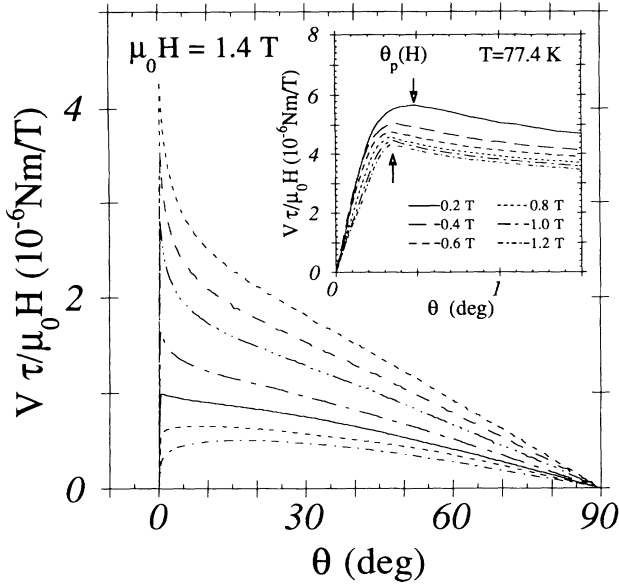


FIG. 1. Angular dependence of the scaled torque  $V\tau/\mu_0H$  of a Bi:2212 single crystal for constant field (1.4 T) and various temperatures. The different plots correspond respectively to (from top to bottom) 77.4; 78.6; 79.7; 81.5; 82.6; 83.1; and 84.4 K. The inset shows the behavior close to  $\mathbf{H} \parallel (a, b)$  at 77.4 K for fields varying from  $\mu_0H = 0.2$  to 1.2 T.

K for various fields (inset of Fig. 1) shows that the peak position  $\theta_p$  is field dependent, i.e., that it decreases from  $\theta_p = 0.5^\circ$  at 0.2 T and levels off to  $\theta_p = 0.38^\circ$  at 1.2 T. For the fields used in our experiments (above 0.1 T) no irreversibility was observed. This confirms that we are measuring the equilibrium properties.

Equation (1) can be written as  $\tau_y = \mu_0 H_x H_z (M_z/H_z - M_x/H_x)$ , where  $z$  corresponds to  $\mathbf{H} \parallel \mathbf{c}$  and  $(x, y)$  is associated with the plane  $(a, b)$ . For  $H_x \gg H_{c1,x}$  and for samples with a large demagnetizing factor  $n$ , we estimate that  $|M_z/H_z| \gg |M_x/H_x|$ . Most of the contribution to the torque is thus given by  $\mu_0 H_x M_z(H_x, H_z)$ . Moreover for angles such that  $\gamma \sin \theta \gg \cos \theta$  we can neglect the dependence on  $H_x$  so that Eq. (1) can be written in the simple form

$$\frac{\tau}{H \cos \theta} = \frac{\tau}{H_x} = \mu_0 M(H \sin \theta) = \mu_0 M(H_z). \quad (2)$$

This expression coincides with the expectation for layered superconductors in the decoupling limit (2D case). As a test for the validity of Eq. (2) we measured both  $\tau(H)$  at a fixed angle and  $\tau(\theta)$  at fixed field. After plotting the data as  $\tau/H_x$  vs  $H_z$  both kinds of measurements gave exactly the same result for  $\theta > \theta_p$ .

To further analyze the data we have to account for the geometry of our sample by computing the internal perpendicular field  $H_{i,z} = H_z - nM_z$ . The demagnetization factor  $n = 0.93$  is obtained from the ellipsoid approximation [12] and for  $M_z$  we substituted  $\tau/\mu_0 H_x$ . We obtain the results shown in Fig. 2 for  $T = 77.4$  K and various

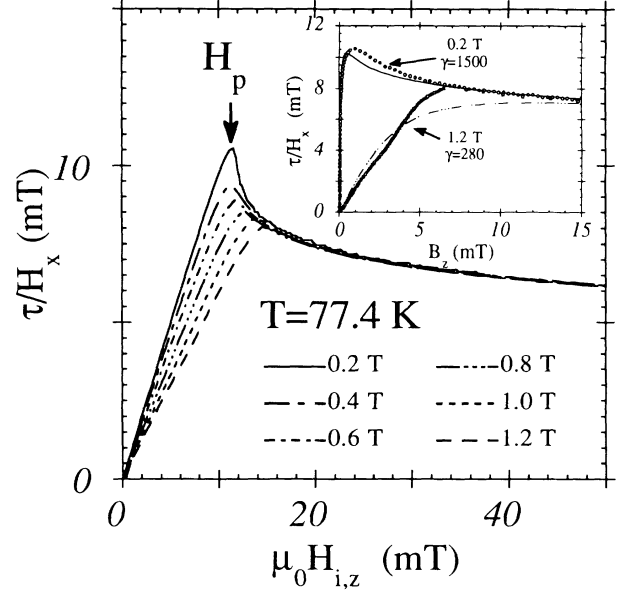


FIG. 2. Scaled torque  $\tau/\mu_0 H_x$  vs the internal perpendicular field  $\mu_0 H_{i,z}$  and the perpendicular induction  $B_z$  (inset) for different applied fields at  $T = 77.4$  K. The collapse of the data above  $H_p$  demonstrates the quasi-2D character of Bi:2212. The lines in the inset are the best fits of the AGL expression [Eq. (3)] to the data at 0.2 and 1.2 T.

applied fields between 0.2 and 1.2 T. Most striking is the collapse of all data above  $H_{p,z}$ . The universal behavior represents the reversible  $M_z(H_{i,z})$  curve as suggested by Eq. (2). We conclude that for  $\theta > 0.5^\circ$  the magnetization behaves as for a 2D superconductor.

Our data cannot be interpreted in terms of the AGL model. To show this we use the small-angle data below  $H_{p,z}$  and plot  $\tau/H_x$  vs  $B_z = \mu_0(H_{i,z} + M_z)$  for 0.2 and 1.2 T in the inset of Fig. 2. The lines correspond to an attempt to fit our results with AGL which for  $\gamma \gg 1$  can be written as [5]

$$\frac{\tau}{H \cos(\theta)} = \frac{\Phi_0}{16\pi\mu_0\lambda_{ab}^2} \frac{\gamma B_z}{B\epsilon(\theta)} \ln\left(\frac{\gamma\eta H_{c2}}{B\epsilon(\theta)}\right), \quad (3)$$

$$B\epsilon(\theta) = \sqrt{B_x^2 + \gamma^2 B_z^2}. \quad (4)$$

In Eq. (3) we consider  $\eta H_{c2} = 22$  T. This value is obtained from a least-squares fit of the high field data that is independent of  $\gamma$ . In the inset of Fig. 2 we see that the least-squares fit of  $\tau/H_x$  by Eq. (3) is not of good quality. Furthermore the fitted values of  $\gamma$  decrease from 1500 down to 280 with field increasing from 0.2 to 1.2 T. This is in contradiction with AGL since in this theory  $\gamma$  is supposed to be independent of field. We conclude therefore that the anisotropy parameter  $\gamma$  of a quasi-2D superconductor as Bi:2212 cannot be deduced from a fit of torque data to Eq. (3) as it has been done before [2, 3].

Returning to Fig. 2, it is seen that the overall behavior at 0.2 T resembles very closely the magnetization curve of an ideal type-II superconductor. At low fields ( $H_{i,z} < H_{p,z}$ )  $M_z$  varies linearly with  $H_{i,z}$  and a transverse susceptibility can be defined:

$$\chi_z = \left. \frac{\partial M_z(H_{i,x}, H_{i,z})}{\partial H_{i,z}} \right|_{H_{i,z} \rightarrow 0} \quad (5)$$

In the most general case  $\chi_z$  depends on the total applied field. However, for layered superconductors at small tilting angles, the vortices are expected to be "locked" between the layers. The perpendicular component of the field should only penetrate into the sample above some critical angle  $\theta_c \approx (1-n)H_{c1,z}/H$  by the creation of kinks (lock-in transition) [13]. In this case below  $\theta_c$ , for locked vortices  $B_z = 0$  and  $\chi_z$  should be exactly  $-1$  at all fields. This expectation is not confirmed by the present experiment. From Fig. 2 at 77.4 K, we see that  $|\chi_z(H)|$  decreases almost linearly with field from 0.99 at  $H = 0.2$  T down to 0.55 at 1.2 T.

The absence of a "lock-in" transition, as well as the field dependence of  $|\chi_z|$ , is probably due to two different factors. The lock-in transition can be destroyed by thermal fluctuations at high enough temperatures. The presence of thermally activated vortex kinks and antikinks allows the  $z$  component of the field to penetrate the sample even for  $\theta < \theta_c$ . Recently Iye, Tamegai, and Nakamura [14] used this mechanism to explain the  $(a, b)$ -plane resistivity  $\rho_{ab}$  in Bi:2212 thin films for  $\theta$  smaller than  $0.5^\circ$ .

Our results can also be influenced by the slight buckling of the  $\text{CuO}_2$  layers related to the distribution of  $c$ -axis orientations with  $\varphi_0 = 0.1^\circ$ . If  $\theta_c < \varphi_0$  the lock-in transition cannot be observed, and  $|\chi_z(H)|$  decreases with field. From  $\mu_0 H_{c1,z} \approx \mu_0 H_p \approx 10$  mT (see Fig. 2) we estimate for our sample that  $\theta_c$  is of the same magnitude as  $\varphi_0$  when  $\mu_0 H > 0.4$  T. The behavior at high

fields can thus be explained by this mechanism.

The texture will also influence the estimate of the anisotropy constant  $\gamma$ . In the absence of texture, for AGL as well as for a Lawrence-Doniach model, at high fields we can write the relation  $\theta_p \approx 1/\gamma$ . On the other hand, the distribution of orientations smooths the torque and would make the experimental value of  $\theta_p$  increase for samples of lower quality. The experimental value  $\theta_p = 0.38^\circ$  is thus an overestimate of  $1/\gamma$  and we get  $\gamma > 150$ .

We now turn to a comparison of our data with the recent theory of Bulaevskii, Ledvig, and Kogan [8] that provides detailed predictions for the magnetic behavior of layered superconductors with weak interlayer Josephson coupling. In this theory the lower critical field  $H_{c1}$  and the magnetization at intermediate values of  $B$ , i.e., for  $\mu_0 H_{c1} \gg B \gg \mu_0 H_{c2}$ , are both renormalized due to the effect of thermal fluctuations. As follows from Fig. 2, at the lowest field the magnetization resembles the classical Abrikosov behavior and from the peak position  $H_p(T)$  it is possible to make a reasonable estimate for  $H_{c1}(T)$ .

Taking into account that we are dealing with an extremely anisotropic system we can easily extract the magnetization from torque measurements at high angles (above  $2^\circ$ ). In Fig. 3 the deduced magnetization is plotted as a function of  $B_z$  for various temperatures above 77 K. At lower temperatures the magnetization follows the standard decreasing logarithmic behavior for  $B_z > 10$  mT. For some temperature close to 83.1 K the slope changes sign and we observe an increasing logarithmic behavior at higher temperatures. In Fig. 4 we show the temperature dependences of the slope  $\partial \mu_0 M / \partial \ln B$  and the peak field  $H_p(T)$  at  $\mu_0 H = 0.1$  T. One can see that both plots are linear but extrapolate to zero at different temperatures. This agrees with the theoretical predictions by Bulaevskii, Ledvig, and Kogan [8] who calculated the entropy corrections to the magnetization and penetration field. They argue that the slope  $\mu_0 \partial M / \partial \ln B$  should ex-

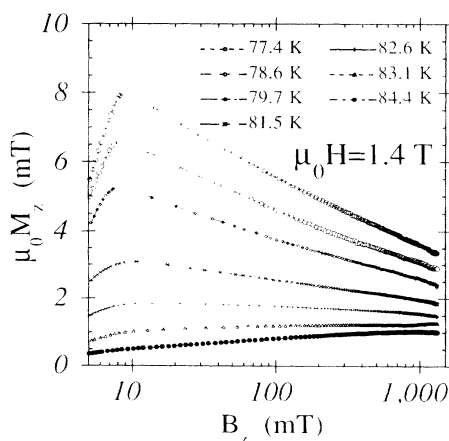


FIG. 3. Magnetization vs  $B_z$  for temperatures between 77.4 and 84.4 K. We observe a logarithmic behavior for the magnetization at  $B_z > 10$  mT.

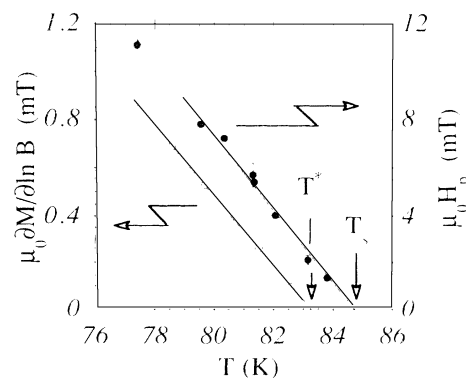


FIG. 4. Plot of  $\partial M / \partial \ln B$  (left) and  $H_p$  vs  $T$  (right) as a function of temperature. A linear behavior is observed for both quantities. The extrapolations to zero give  $T_s = 84.7$  K and  $T^* = 83.2$  K for the Kosterlitz-Thouless temperature.

trapolate to zero at the analog of the Kosterlitz-Thouless temperature for a layered superconductor  $T^*$  while  $H_{c1}$  should go to zero at a temperature  $T_s$  that is slightly smaller than the “mean field” transition temperature  $T_{c0}$ . From Fig. 4 we get  $T_s = 84.7$  K and  $T^* = 83.2$  K for the temperature of unbinding of vortex-antivortex pairs.

From the slope  $\mu_0 \partial M / \partial \ln B$  the Ginzburg-Landau penetration depth  $\lambda_{ab}^{GL}$  can be deduced [8]:

$$\mu_0 \frac{\partial M}{\partial \ln B} \approx \frac{\Phi_0}{8\pi[\lambda_{ab}^{GL}(0)]^2} \left(1 - \frac{T}{T^*}\right). \quad (6)$$

Taking into account that  $\lambda_{ab}^{GL}(0) = 0.70\lambda_{ab}(0)$  we get for the penetration depth  $\lambda_{ab}(0) = 110$  nm. From  $\lambda_{ab}$  it is possible to estimate the difference  $T_{c0} - T^*$  [15]:

$$T_{c0} - T^* = 4\pi\mu_0 k_B T_{c0}^2 \lambda_{ab}^2(0) / \Phi_0^2 s; \quad (7)$$

by taking for the distance between the superconducting planes  $s = 1.5$  nm, we get  $T_{c0} - T^* = 3.2$  K. This gives  $T_{c0} = 86.4$  K for the mean field transition temperature and a difference  $T_{c0} - T^*$  that is in good agreement with previous results obtained from transport measurements [16].

In this Letter we conclude that from AGL we cannot obtain the anisotropy  $\gamma$  of Bi:2212. Only a lower bound ( $\gamma > 150$ ) that is sample dependent can be given. Even in our better samples we could not detect any “lock-in” transition above  $T = 77$  K. This seems to be due to a distribution of the  $c$  axis with a HWHM of  $0.1^\circ$  and/or the existence of thermally activated kinks and antikinks. In this work we clearly show that for angles above  $0.5^\circ$  Bi:2212 behaves like a 2D superconductor. The magnetization can thus be deduced from the magnetic torque. From our data we observe in our field range a logarithmic behavior which agrees with a recent theory of Bulaevskii, Ledvig, and Kogan [8]. We deduce that  $T_s - T^* = 1.5$  K and  $\lambda_{ab}(T = 0) = 110$  nm.

J.C.M. would like to acknowledge clarifying discussions with L.N. Bulaevskii, A. Buzdin, and D. Feinberg. The authors thank J. Mydosh for a critical reading of the manuscript and J. Lensink for the help given in the early stages of this work. This work is part of the research

program of the “Stichting voor Fundamenteel Onderzoek der Materie,” which is financially supported by “NWO.”

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