Effective mass anisotropy of $HgBa_2Ca_3Cu_4O_{10}$ measured on a microcrystal by means of miniaturized torque magnetometry

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Torque magnetization measurements on a $HgBa_2Ca_3Cu_4O_{10}$ microcrystal of mass $m \simeq 380$ ng were performed using a miniaturized torque sensor ($120\times 90\times 4$ μm) with a very high sensitivity of $\Delta\tau \simeq 10^{-14}$ N m. In an applied field of 1 T this allows the measurement of magnetic moments as small as $m = 10^{-14}$ A m^2, which is far beyond the sensitivity of the best commercial superconducting quantum interference device magnetometers. From the angle-dependent torque measurements an effective mass anisotropy of $\gamma = \sqrt{m_c^*/m_{ab}^*} = 52(1)$ and an estimate of the in-plane coherence length of $\xi_{ab} = 1.8(5)$ nm are extracted for $HgBa_2Ca_3Cu_4O_{10}$. The large value of γ reflects the quasi-two-dimensional nature of the mercury based cuprates and demonstrates the importance of vortex fluctuations in these materials.

The class of mercury based cooper oxide superconductors[1,2] $HgBa_2Ca_{n-1}Cu_nO_{2n+2}$ [Hg-12$(n-1)n$, $1 \leqslant n \leqslant 5$] has attracted considerable interest in their superconducting properties. The critical temperature T_c varies with the number n of CuO_2 layers reaching a maximum $T_c = 133$ K for the three layer compound[2] and this material is considered as a particularly interesting candidate in view of technical applications. Here we present magnetization measurements on very small ($m < 1\,\mu$g) high-quality single crystals of Hg-1234 which have recently become available.[3] The application of a miniaturized torque magnetometer especially designed for magnetization measurements on microcrystals has allowed a *direct* determination of the effective mass anisotropy γ not previously possible in granular materials. The anisotropy $\gamma = \sqrt{m_c^*/m_{ab}^*}$, where m_c^* is the perpendicular and m_{ab}^* the in-plane effective mass of the superconducting carriers, is a measure of the effective Josephson coupling between adjacent blocks of CuO_2 layers. It plays an important role in the understanding of the phenomenology of the superconducting state in the layered cuprate materials. Thermal fluctuations of the vortex positions are greatly enhanced in highly anisotropic systems, which severely limits the technical applications of cuprate superconductors. A precise determination of this parameter is, therefore, particularly important. For the Hg-12$(n-1)n$ compounds, estimates of the anisotropy parameter γ are controversial. In grain-aligned Hg-12$(n-1)n$ compounds values of $\gamma \simeq 2$ (Ref. 4) and $\gamma \geqslant 8$ (Ref. 5) were obtained from the ratio of physical quantities related to the effective mass, as measured parallel and perpendicular to the CuO_2 layers. Other estimates of $\gamma \approx 16$ (Ref. 6) and $\gamma \approx 2-6$ (Ref. 7) were derived from magnetization measurements in randomly oriented polycrystalline Hg-12$(n-1)n$ samples, yet still comparable to the anisotropy parameter $\gamma = 5-9$ (Ref. 8) of the $YBa_2Cu_3O_{7-\delta}$ compound. On the other hand, magnetic relaxation and high-field magnetization studies reveal a pronounced two-dimensional (2D) character of the vortex system, suggesting a considerably larger anisotropy of the Hg-12$(n-1)n$ compounds.[5,9] This is also supported by the position of the irreversibility line,[10,11] which is found to fall between that of the $YBa_2Cu_3O_{7-\delta}$ ($\gamma = 5-9$) and that of the very anisotropic $Bi_2Sr_2CaCu_2O_8$ compound [$\gamma > 150$ (Ref. 12)]. From the shape of this line, analyzed in terms of a 3D-2D crossover criterion, an estimate of $\gamma \simeq 35-70$ for grain-aligned Hg-1223 was obtained.[11]

The above discrepancy between the measured values of γ and the estimates obtained from the analysis of vortex fluctuations call for a detailed and more direct measurement of the anisotropy parameter γ in the Hg-12$(n-1)n$ compounds. In this work, we present *direct* measurements of the effective-mass anisotropy on a $HgBa_2Ca_3Cu_4O_{10}$ microcrystal by means of angle-dependent torque magnetometry. For the Hg-12$(n-1)n$ compound only very tiny single crystals with dimensions of $d \ll 1$ mm are currently available[3] and the sample used in this study had a mass of only $m \simeq 380$ ng. The sensitivity of standard superconducting quantum interference device (SQUID) and torque magnetometers is far to low for magnetization measurements on such small samples. For this reason we have developed an integrated torque magnetometer with a very high sensitivity of $\Delta\tau \simeq 10^{-14}$ N m. In an applied field of 1 T this corresponds to a magnetic moment of $\Delta m \simeq 10^{-14}$ A m^2, which is three orders of magnitude smaller than the currently achieved sensitivity of the best commercial SQUID magnetometers. The ''nanotorque'' magnetometer is particularly useful for magnetization measurements on small samples with a mass $m \ll 1$ μg, such as a microcrystal taken from a granular sample. The torque sensor used in this work is based on a commercial piezoresistive

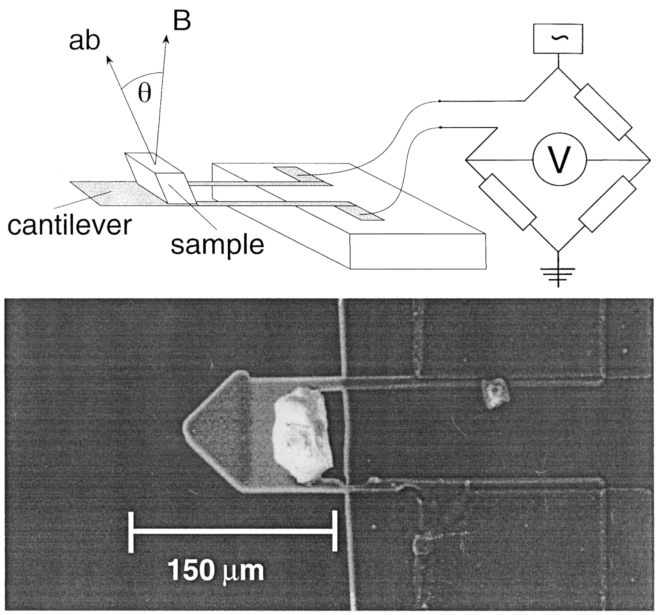

FIG. 1. Schematic figure of the miniaturized torque sensor with piezoresistive readout. The bottom is an electron micrograph of the cantilever with the sample (white).

cantilever[13] which we used for the detection of a magnetic torque $\vec{\tau}=\vec{m}\times\vec{B}$ arising from a transverse component of the magnetization $\vec{m}$. A micrograph of the sensor is shown in Fig. 1. The microfabricated silicon cantilever has a length of 120 μm, a width of 90 μm, and a thickness of 4 μm. The top surface is p doped and forms a piezoresistive path with a resistance of $R\approx 2000\ \Omega$ at 300 K. A deflection d of the cantilever results in a change of the resistance of the piezoresistive path, which can be detected using a conventional Wheatstone-bridge technique as schematically shown in Fig. 1. The resolution obtained by this technique is typically $\Delta d\simeq 0.1$ Å and is similar to that reached by optical readout systems. However, the ease of use and the absence of external deflection sensing elements of the piezoresistive cantilever makes this device very advantageous when compared to optical detection systems. For the detection of the magnetic torque the sample is carefully fixed with vacuum grease on the cantilever. In a *homogeneous* applied field a transverse component of the sample magnetization will cause a deflection of the cantilever which is proportional to the magnetic torque τ. Using an approximate force constant of 20 N/m,[13] the above cantilever yields a torque sensitivity of $\Delta\tau\simeq 10^{-14}$ N m. In an applied field of 1 T this makes it possible to measure tiny magnetic moments of $\Delta m\simeq 10^{-14}$ A m^2. The magnetic torque detection method presented here differs from the earlier proposed detection of a magnetic force produced by an *inhomogeneous* applied field (see, e.g., Ref. 14). In a homogeneous field, as used in our setup, the absence of a magnetic force allows us to discriminate the anisotropic magnetization component of the sample from the isotropic diamagnetic signal of the cantilever, which is particularly important for the measurement of very small magnetic moments. As discussed elsewhere[15] the sensitivity of the nanotorque magnetometer can further be increased with more sophisticated detection and operating techniques.

We turn now to the anisotropy measurement of Hg-1234. The Hg-1234 microcrystal used in this study as prepared by a high gas pressure synthesis technique.[3] The best samples were obtained with a PbO flux, which leads to a partial substitution of Pb for Hg. The high quality of the crystals was confirmed by transmission electron microscopy (TEM), showing the characteristic four CuO_2 layer structure. The lattice parameters were determined by x-ray diffraction which also gives additional information on the phase purity of the sample investigated. Low-field magnetization measurements on a bunch of single crystallites of Hg-1234 from the same batch (total mass $m\simeq 0.05$ mg) showed a superconductivity onset at $T=113$ K with a transition width (10–90 %) of $\Delta T_c\approx 5$ K. For the magnetic torque measurements *one* microcrystal was chosen from the above sample. The crystallite had dimensions of $60\times 80\times 10\ \ \mu m^3$ and a mass of only $m\simeq 380$ ng. The sample was fixed on the torque sensor with an approximate angle of 60° between the sample ab-plane and the cantilever surface (see Fig. 1). The torque sensor was horizontally installed in a cryostat between the poles of an electromagnet ($B<1.5$ T) and the temperature was stabilized better than 0.01 K. Angle-dependent torque measurements with an angular resolution of 0.01° were carried out by rotating the electromagnet (0.5°/sec) with respect to the sample ab plane while continuously sampling the torque signal. In the vicinity of the ab plane ($\pm 5°$) the rotation was slowed down by a factor of 10. In Fig. 2(a) the angular dependence of the torque signal is shown for $T=109.77$ K and for an applied field of $B=0.5$ T. From the symmetry of the torque signal $\tau(\theta)=-\tau(-\theta)$ the orientation of the sample ab plane with respect to the applied field was determined with an accuracy better than 0.05°. As the field rotates towards the ab direction the torque is increasing, showing a sharp maximum at $\theta_p=2.4°$ and then drops rapidly to zero for B parallel to the ab plane ($\theta=0°$). The torque was monitored for both directions of rotation, and was found to be almost perfectly reversible. Only for the field very close to the ab plane ($\theta<0.4°$) a small hysteresis curve with a pronounced peak at $\theta=0.1°$ was observed, as clearly seen in the enlarged scale in Fig. 2(b). The symmetry of the torque signal and the geometry of the experimental setup, with the ab plane not lying parallel to the cantilever, proves that the observed hysteresis curve is indeed due to the sample magnetization.

For further analysis we make use of the simplest model for the angular dependence of the magnetic torque. Based on the anisotropic London model[16,17] the angular dependence of the reversible magnetic torque τ of an anisotropic superconductor with volume V is given by

$$\tau(\theta)=\frac{\Phi_0 VB}{16\pi\mu_0\lambda_{ab}^2}\,\frac{\gamma^2-1}{\gamma}\,\frac{\sin 2\theta}{\epsilon(\theta)}\ln\left(\frac{\gamma\eta B_{c_2}^c}{B\epsilon(\theta)}\right), \qquad (1)$$

where $\epsilon(\theta)=\sqrt{\gamma^2\sin^2(\theta)+\cos^2(\theta)}$ is the angular scaling function, $B_{c_2}^c$ is the upper critical field measured along the c-axis direction, and μ_0 is the vacuum permeability. The numerical parameter η is of the order of unity and depends on the structure of the flux-line lattice. The angle θ is measured with reference to the ab plane. For a fixed applied field B the normalized torque $\tau_{\mathrm{norm}}=\tau(\theta)/\tau(\theta_p)$ depends only on

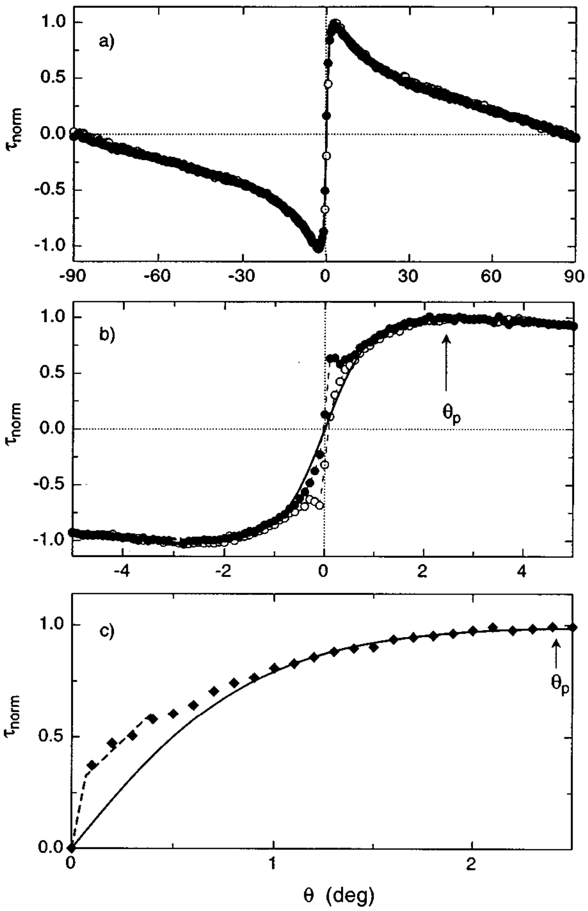

FIG. 2. (a) Angle-dependent torque of a microcrystal of $HgBa_2Ca_3Cu_4O_{10}$ ($m \simeq 380$ ng) measured at $T=109.77$ K in a field of $B=0.5$ T. The solid line is a fit of Eq. (1) to the data with an anisotropy parameter $\gamma=51.4(5)$ and an upper critical field of $\eta B_{c_2}^c=2.43(6)$ T. (b) Enlarged scale close to the ab plane. For $\theta<0.4°$ the torque becomes irreversible showing a sharp peak at $\theta=0.1°$. (c) Reversible torque in the vicinity of the ab plane. Deviations from the 3D London model (solid line) are clearly visible for $\theta<1°$. The dashed line is the *calculated* reversible torque as obtained in the framework of the Lawrence-Doniach model.

γ and $\eta B_{c_2}^c$. Equation (1) shows at a characteristic orientation angle θ_p a sharp maximum, which moves closer to the ab plane with increasing anisotropy γ. For highly anisotropic superconductors its position $\theta_p \simeq 1/\gamma$ is very sensitive to the degree of anisotropy while its dependence on $B_{c_2}^c$ is negligible. This further implies that for a reliable measurement of γ, a well defined orientation of the crystallographic c axis is required, which is only achieved by using high-quality single crystals. For the analysis of the angle-dependent torque measured on the Hg-1234 microcrystal only the reversible torque data at angles $|\theta|>0.4°$ were considered. The data were normalized with the torque signal $\tau(\theta_p)$ at the peak position. The fitting parameters are the anisotropy parameter γ and the upper critical field $\eta B_{c_2}^c$. The best fit to the data was obtained for $\gamma=51.4(5)$ and $\eta B_{c_2}^c=2.43(6)$ T. Equation (1) describes almost perfectly the full angular dependence of the experimental data [solid line in Fig. 2(a)]. Only for $|\theta|<1°$ clear deviations from the theoretical curve are observed as

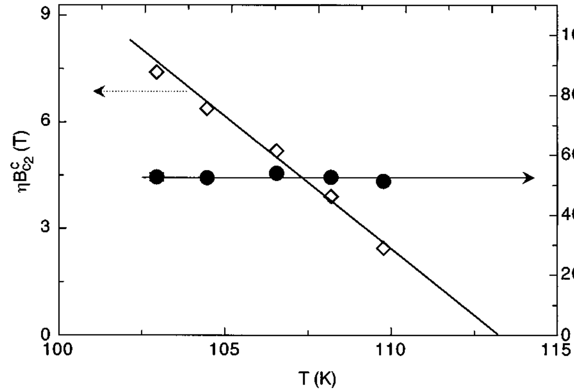

FIG. 3. Temperature dependence of the anisotropy parameter γ and the upper critical field along the c axis $\eta B_{c_2}^c$ of a $HgBa_2Ca_3Cu_4O_{10}$ microcrystal as determined by torque magnetometry. From the slope $\eta dB_{c_2}^c/dT=-0.80$ T/K an estimate of the in-plane coherence length $\xi_{ab} \simeq 1.8(5)$ nm is obtained.

shown in the enlarged scales of Figs. 2(b) and 2(c). However, the position of the maximum torque at θ_p, which is a measure of the anisotropy, falls fully within the reversible regime and is perfectly described by the fit. The quality of the fit was further tested for various strengths of the applied field (0.5 T$<B<$1.5 T) and for various temperatures (103 K$<T<$110 K). In this field and temperature regime the torque was found to be reversible at all angles $|\theta| \geq |\theta_p|$, a necessary condition for a reliable determination of the anisotropy. The results of this analysis are summarized in Fig. 3. The anisotropy γ is almost temperature independent with a mean value of $\gamma=52(1)$, while the upper critical field increases linearly with a slope of $\eta dB_{c_2}^c/dT=-0.80(6)$T with decreasing temperature. Using the formula $B_{c_2}(0) \simeq 0.7T_c|dB_{c_2}/dT|_{T_c}$ and $\eta=0.7(3)$ one obtains an estimate of the in-plane coherence length of $\xi_{ab}(0) \simeq \sqrt{\Phi_0/2\pi B_{c_2}^c(0)}=1.8(5)$ nm for Hg-1234 which is in good agreement with values ranging between 1.5 and 2.1 nm as previously reported for the Hg-1201 and Hg-1223 compounds.[5,7,9]

The large anisotropy $\gamma=52(1)$ of the Hg-1234 compound raises the question of whether the continuum London approach is appropriate for the above analysis. Even for $YBa_2Cu_3O_{7-x}$ with $\gamma \simeq 5-9$ systematic deviations from the continuum London approach were reported[18] and the discrete nature of a stack of pancake vortices has to be considered when ξ_c becomes smaller than the interlayer separation. However, these deviations are due to the scaling function $\epsilon(\theta)$ only becoming important for $\theta_0=\tan^{-1}(1/\gamma)$.[19] Using $\gamma=52$ for the Hg-1234 compound this is the case only for $\theta<\theta_0=1.1°$ as it is clearly observed in the present experiment. In the enlarged scale of Fig. 2(c) the reversible part of the torque signal (mean value of the torque data taken for both directions of rotation) is compared with the fit obtained from the London approach (solid line). For $\theta<1.0°$ systematic deviations are observed, which we now evaluate within the framework of the Lawrence-Doniach model where the discrete nature of vortices is considered.[20] For orientation angles (in our case $\theta<0.4°$) where the measured torque is about two times larger than that given by the London approach the above model predicts $\tau_{norm}=\gamma \sin\theta+1/$

$2\ln(\eta B^c_{c_2}/B)$. At even lower angles (in our case $\theta<0.1°$) the torque sharply drops to zero when the flux lines become locked between the ab plane. Putting the values of $\gamma=52$ and $\eta B^c_{c_2}=2.43$ T at 109.77 K in the above model the *calculated* torque is in excellent agreement with our data [dashed line in Fig. 2(c)]. This further corroborates the large anisotropy parameter $\gamma=52$ deduced from Eq. (1). In Eq. (1) thermal fluctuations of vortices are not considered. They become important for temperatures close to T_c and/or for magnetic fields much larger than the 3D-2D crossover field $B_{cr}=\Phi_0/(d\gamma)^2$, where d is the distance between neighboring CuO_2 blocks.[21,22] Vortex fluctuations cause a change of the shape of the angular torque curve, whereas the position of the torque maximum θ_p, which is a measure of γ, is not affected.[23] Thus, in the presence of strong vortex fluctuations, Eq. (1) will no longer describe the angular dependence of τ, but still gives a good estimate of γ. In the present experiment the excellent fit of Eq. (1) to the data indicates that vortex fluctuations are not yet relevant for $B\leqslant0.5$ T. Using $d=15$ Å and $B_{cr}=0.5$ T for the 3D-2D crossover field a lower limit of $\gamma>42$ is obtained which is in good agreement with the above London analysis.

Our value of $\gamma=52(1)$ for Hg-1234 is much larger than those measured in grain-aligned samples of related mercury based cuprates.[4,5] This discrepancy is easily understood in terms of the large angular spread ($\approx2.5°$) of the c-axis orientation present in grain-aligned samples, which prevents a reliable determination of the anisotropy. On the other hand, the present value of γ is consistent with estimates obtained from analysis on the basis of vortex fluctuations[11] and as such supports the quasi-2D character of the mercury based cuprates.

In summary, magnetization measurements on a microcrystal of $HgBa_2Ca_3Cu_4O_{10}$ were performed using a miniaturized torque magnetometer having a very high sensitivity of $\Delta\tau\simeq10^{-14}$ N m. The high-quality single crystal had a mass of only $m\simeq380$ ng. Angle-dependent torque measurements were used to extract a precise value of the anisotropy ratio $\gamma=52(1)$ and an estimate of the in-plane coherence length $\xi_{ab}=1.8(5)$ nm for $HgBa_2Ca_3Cu_4O_{10}$. The value of γ is significantly larger than those previously reported for grain-aligned samples. We further found that for $B\leqslant0.5$ T and $T<110$ K the simple anisotropic London approach is appropriate to describe the angular dependence of the torque. Only for the field applied close to the ab plane does the discrete nature of the vortex lines result in a magnetic torque which is well described by the 2D Lawrence-Doniach model. The large anisotropy of the mercury-based cuprates and the corresponding enhanced vortex fluctuations result directly from the weak interlayer coupling present in these quasi-2D layered superconductors.

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