Anisotropy of $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ single crystals studied by torque magnetometry

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The coherence-length anisotropy ratio γ of $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ superconducting single crystals ($x \leq 0.22$) has been measured by the equilibrium-torque magnetometry method. The experiments were performed at temperatures several K below the onset T_c of superconductivity in magnetic fields of 1–2 T. $\gamma=9.3\pm0.3$ for two pure $YBa_2Cu_3O_{7-\delta}$ single crystals, which is 20% larger than that of previously published results. The γ value of $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ increases with increasing Fe concentration to $\gamma=29\pm1.5$ for $x=0.22$. The direct torque from the localized magnetic moments of Fe was not observed within the experimental sensitivity. The fundamental reason for the anisotropy increase is not clear.

I. INTRODUCTION

One of the key features of the high-T_c cuprate superconductors is their high anisotropy compared with conventional three-dimensional (3D) BCS superconductors. This, along with other factors, leads to the conclusion that the CuO_2 planes play an essential role in the superconductivity and there are weak interlayer couplings between them. The degree of anisotropy is usually expressed by the dimensionless parameter γ, which is defined as the ratio of the superconducting coherence lengths parallel and perpendicular to the basal plane (*ab* plane). In the 3D Ginzburg-Landau model, $\gamma=(m_c/m_a)^{1/2}$, where m_c and m_a are the effective masses along the *c* axis and the *ab* plane, respectively.

Ways of measuring γ include resistive, magnetic, and torque methods. Among all these possibilities, the equilibrium torque method offers the most unambiguous way to measure γ directly. An anisotropic superconductor experiences a torque $\tau=M\times H$ in a magnetic field H. The angular dependence of $\tau(\theta)$ gives a direct measure of γ of the superconductor where θ is the angle between the *c* axis and the applied H. This line of thinking has been developed theoretically by Kogan and Clem[1,2] and more recently by Hao and Clem[3] and Blatter, Geshkenbein, and Larkin.[4]

For pure $YBa_2Cu_3O_{7-\delta}$ crystals, recent equilibrium torque measurements gave a γ value close to 7.5.[5,6] As a comparison, previous anisotropy measurements of H_{C2} by dc magnetization method indicated $\gamma\approx5$ (Ref. 7) and resistivity measurements gave $\rho_c/\rho_a\approx40$ (Ref. 8) for pure $YBa_2Cu_3O_{7-\delta}$.

In regard to the general physical properties of the $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ system, earlier work provided a great deal of information. The solubility limit of x in $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ was determined to be approximately 0.5.[9,10] The magnetic Fe^{+3} ions mainly go into the Cu(1) chain sites at low concentrations and substitution for Cu(2) plane sites starts to prevail at higher Fe concentrations.[10–12] The structure of the $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ is orthorhombic with $x<\sim0.08$ and is tetragonal with $x>0.08$.[9,10] Lan *et al.*[13] measured the resistivities of the $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ single crystals and found a metallic behavior for $x<0.25$. The superconducting T_c decreases steadily with Fe doping and no abrupt change was observed across the orthorhombic-tetragonal transition.[11] The Fe^{3+} ions form localized magnetic moments with $\mu_{eff}=3.4\mu_B$ per ion, from the Curie-Weiss law of the normal state susceptibility[9] and the Mössbauer data.[10] The *c*-axis lattice constant increases up to ~11.68 Å at low doping level and then decreases after the orthorhombic-tetragonal transition.[9]

In this paper, we present anisotropy measurements of high-quality $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ single crystals by the equilibrium torque magnetometry method. Six different doped samples with x ranging from 0 to 0.22 were studied and we found that the γ value increases with increasing Fe doping. More details will be discussed in the following sections.

II. EXPERIMENTAL DETAILS

A rotating magnetic torque magnetometer similar to that of Iye *et al.*'s[14] was constructed. The magnetometer consists of two parts: a rotatable sample probe inside a fixed outer vacuum jacket. The magnetometer is in a dewar between the pole faces of a 2 T electromagnet. The sample sits on a thin Be-Cu plate which serves as a spring and the capacitance between this spring and a fixed electrode separated by a distance d is measured. The total capacitance is in the range of 0.25–0.35 pF. The torque τ causes a deviation Δd of the Be-Cu spring. For small Δd, τ is proportional to Δd. Since the capacitance change ΔC is proportional to $\Delta d/d(d+\Delta d)$, for a small deviation Δd the torque τ is proportional to ΔC

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TABLE I. Doping concentration x versus onset T_c (K) for our $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ samples.

x	0	0.015	0.04	0.13	0.17	0.22
T_c	92.5	91.5	90	78	72	64

with small corrections. A Be-Cu spring with a thickness of approximately 0.04 mm was selected to make it sensitive to detect the small torque signals with a maximum capacitance change of less than 1% of the total capacitance during the experiments. With these conditions, linearity between the capacitance change and the torque signal is obeyed within 1%. Due to some magnetic impurities in the thin Be-Cu spring, some sinusoidal torque background is observed at 77 K. For our small samples, the background problem is the largest systematic error in our torque magnetometry. Numerical subtraction of the background is needed for the data analysis and it will be discussed in more detail in the next section.

The sample probe was rotated by a computer-controlled stepping motor with a maximum angular resolution of 0.009°. The rotation was done at 1 or 0.1 degree intervals and the stepping rate was between 0.5 and 0.8 steps per second. A transverse Hall sensor installed on the probe was used to monitor the angular position of the probe. Liquid nitrogen was used to cool the jacket of the probe and a mechanical pump was used to lower the vapor pressure of the liquid nitrogen so that temperatures between 55 and 77 K could be used. On the copper sample chamber, a heater connected to a temperature controller was used to stabilize the temperature to within ±30 mK.

High-quality $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ single crystals were grown by a modified self-flux technique at the University of California at Davis.[15,16] The samples were oxygen annealed at 420 °C for one week in a flowing oxygen environment to obtain an optimum oxygen content. The samples were smooth and shiny with typical dimensions of approximately 40 μm×0.7mm×0.7mm. The masses were between 0.1 and 0.3 mg. The orthorhombic samples were microtwinned. The determination of x was made by energy-dispersive x-ray analysis (EDX)[16] and by the comparison of the onset T_c (10% magnetic susceptibility drop) to the previously published results.[9] Due to the low amount of Fe and the uncertainty of the oxygen concentration, the uncertainty of both methods was about 10%. This is the largest uncertainty in our experiments. The concentration x and the onset T_c of our $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ crystals were given in Table I.

The torque experiments were performed at 2–5 K below the onset T_c in a field of 1.5 or 2 T. The sample probe was rotated through 190° in both directions while the computer recorded the capacitance. The temperature and field ranges were selected so that the flux dynamics were in the reversible regime in our experimental time scale ($\sim$1 s).

III. RESULTS

Based on the 3D anisotropic London model, in the field range $H_{C2} > H \gg H_{C1}$, the angular dependence of the equilibrium torque per unit volume is given by[2]

$$\tau(\theta)=\frac{\phi_0 H}{64\pi^2\lambda^2}\frac{\gamma^2-1}{\gamma^{1/3}}\frac{\sin(2\theta)}{\epsilon(\theta)}\ln\left[\frac{\gamma\eta H_{C2\parallel}}{H\epsilon(\theta)}\right], \quad (1)$$

where $\epsilon(\theta)=(\sin^2\theta+\gamma^2\cos^2\theta)^{1/2}$, θ is the angle between the applied field H and the c axis, $\gamma=\xi_a/\xi_c$, λ is the average penetration depth, $H_{C2\parallel}$ is the upper critical field along the c axis, and η is a constant of the order of unity which is dependent on the flux lattice structure. Demagnetization effects are negligible for $H \gg H_{C1}$. Due to an uncertainty in determining the exact value of θ during experiments, a small angular offset $\Delta\theta$ was left as a fitting parameter. Thus, four fitting parameters were used: γ, $\eta H_{C2\parallel}$, the amplitude and $\Delta\theta$. Equation (1) has been verified by experiments with $YBa_2Cu_3O_{7-\delta}$ (Refs. 5, 6, and 17) and $Bi_2Sr_2CaCu_2O_8$.[18]

Figure 1 shows the $\tau(\theta)$ curves for three different samples. In order to show each data without overlapping, arbitrary units of torque were chosen for these three data sets. The solid lines are the least χ^2 fit of Eq. (1) to the data. A careful look at the data shows that in the angular range $0<\theta<\theta_{max}$ [θ_{max} is the angle at which $\tau(\theta)$ is maximum], the curvature of $\tau(\theta)$ is negative, i.e., $d^2\tau/d\theta^2<0$. This feature is different from that observed by Janossy, Hergt, and Fruchter[5] and Beck *et al.*[6] where a positive curvature was observed. A small $\eta H_{C2\parallel}/H$ value for our temperature and field ranges makes the curvature of Eq. (1) negative.

A problem encountered during the experiments was that the Be-Cu spring was slightly magnetic and gave a sinusoidal background which could be as large as 10% of the maximum torque signal from our smallest samples. The subtraction of this background was never perfect and caused the largest uncertainty in the fitting procedures. To minimize the background effect, the angular range chosen for the fit was either 0° < θ < 180° with 180 points

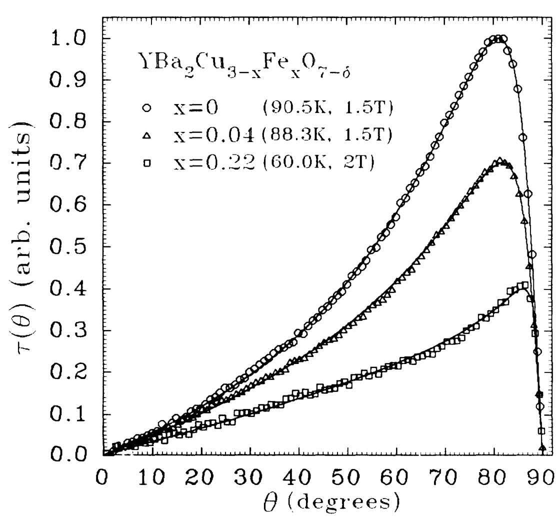

FIG. 1. Typical $\tau(\theta)$ curves for $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ single crystals with x = 0, 0.04, and 0.22 at the temperatures and fields specified on the graph. The solid curves are the least χ^2 fit of Eq. (1) to the data points.

or $80° < \theta < 100°$ with 100–200 points. Both fittings gave consistent γ values to within 3–10 % for data at an identical temperature and field.

The magnitude of the fitting parameter $\Delta\theta$ was always less than 0.05° for the same background subtraction and was a negligible factor for $\gamma < 30$. The angular correction from the deviation of the spring caused approximately a 1% increase of the γ value.

The fitting parameter $\eta H_{C2\parallel}$ in the logarithmic term usually was within a factor of 2 of the predicted value of $\eta H_{C2\parallel}(T)$ for $\eta = 1$ and $dH_{C2\parallel}/dT = -1.5T/K$ at $x = 0$. The slope $dH_{C2\parallel}/dT$ decreased as x increased. We did not try to determine this parameter because the uncertainty of the background dominated the analysis for this parameter.

The normalized $\tau(\theta)$ curves for three different samples are plotted in Fig. 2 to show $\tau(\theta)$ more clearly in the angular range $80° < \theta < 90°$. Data were taken every 0.1° or 0.2° angle. The solid curves are the least χ^2 fit for $80° < \theta < 100°$ with $\gamma = 9.56$, 18.9, and 29.1 for these particular three data sets.

For pure $YBa_2Cu_3O_{7-\delta}$, we measured $\gamma = 9.3 \pm 0.3$ for two samples with $T_c > 92.5$ K. The value is 20% larger than the ones reported earlier by Beck *et al.*[6] ($\gamma = 7.7 \pm 0.2$) and by Janossy, Hergt, and Fruchter[5] ($\gamma = 7.5 \pm 0.1$). This discrepancy is beyond any possible systematic error in our experiments and the higher value is attributed to a higher sample quality.

Figure 3 shows a plot of γ versus x for our $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ single crystals. γ steadily increases to 29 ± 2 for $x = 0.22$. The error bars represent the overall uncertainty from different data and different fitting procedures. The dotted curve is a guide to the eye.

No temperature dependence of γ was observed in our narrow temperature range. This is expected theoretically in the 3D Ginzburg-Landau model.

The possibility of a torque from the magnetic Fe^{+3} ions was examined carefully. This torque would be indicated if $\tau(\theta)$ is inconsistent with Eq. (1) or there is a temperature insensitive ferromagnetic hysteresis in $\tau(\theta)$. This evidence was not observed because for all the $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ samples no torque was observed above T_c and, a few Kelvin below T_c in the reversible regime, the $\tau(\theta)$ data were consistent with Eq. (1). These observations indicated that the Fe^{+3} magnetic moments contributed no torque within our experimental sensitivity. Previous studies of the $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ system also indicated no magnetic ordering of the Fe^{+3} ions from the localized magnetic moments.[9,10]

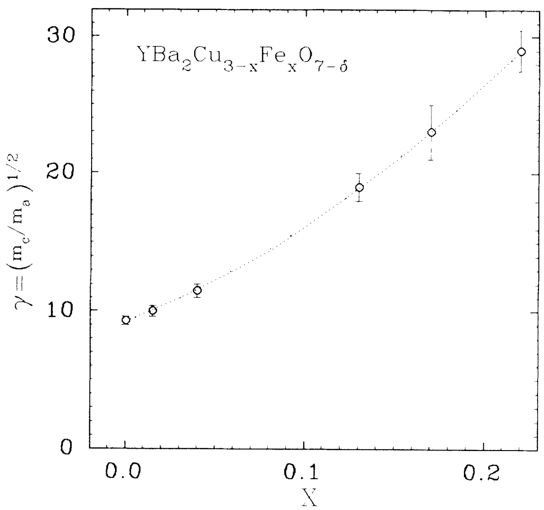

FIG. 3. γ versus x for $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ single crystals. Vertical error bars represent the uncertainties for different data sets and different fitting procedures. The dotted curve is a guide to the eye.

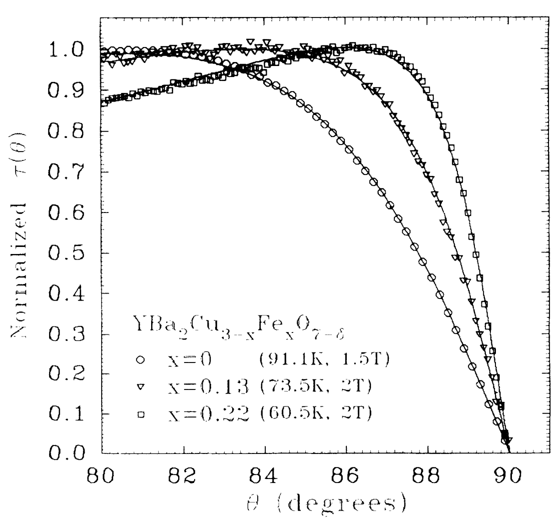

FIG. 2. Normalized $\tau(\theta)$ for field directions within 10° of the *ab* plane. The data were taken in 0.1° or 0.2° intervals. The γ values are the best fit values for $80° < \theta < 100°$ and they are 9.56, 18.9, and 29.1 for $x = 0$, 0.13, and 0.22, respectively.

IV. DISCUSSION

Superconducting anisotropy was an insignificant detail in the 3D BCS superconductors. After the discovery of the high-T_c cuprate superconductors, it is well known that the CuO_2 planes play an essential role in the novel superconductivity and there are weak interlayer couplings between them. Study of the anisotropy of the new quasi-two-dimensional superconductors becomes an important issue.

At the present time, no theory can calculate the anisotropy of the high-T_c materials, let alone the effect of doping on the anisotropy. A direct measurement of the anisotropy of the doped high-T_c superconductors such as $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ may provide important information on the superconducting mechanisms and the physical properties of such a system.

For $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ crystals, the *c*-axis lattice constant increases with Fe doping before the orthorhombic to tetragonal transition ($x < \sim 0.08$), which could possibly *explain* the increase of γ value since the interlayer couplings would be lowered. However, this argument fails at higher doping levels since γ still increases as the *c*-axis lattice constant decreases. Another possible explanation is that a change in the Cu(2) valence may lead to an anisotropy change. It is well known that for oxygen deficient $YBa_2Cu_3O_{7-\delta}$, the Cu(2) ion valence decreases[10] and the anisotropy increases.[19] For the $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ system, the substitution of the Fe^{+3} ions *may* decrease the valence of Cu ions by charge balance. However, the charge balance is complicated by the oxygen increase[10,12] with increasing x. Thus the link between anisotropy and Cu(2) valence cannot be easily established.

Based on our anisotropy measurements, we would like to make a few comments on the previous experiments with the $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ system. Previous anisotropy measurements of the $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ single crystals by H_{C2} measurements perpendicular and parallel to the c axis gave a constant γ value close to 4.5 regardless of the Fe concentration.[13] This is probably due to the large experimental uncertainties. The same group also measured the critical current density J_c of these samples and found that at the 5 K and 1 T, J_c decreases upon Fe doping.[20] Such measurements seemingly contradict the observation that the twinning and disorder increase upon Fe doping for $x<0.1$,[9] which should lead to higher flux pinning and higher J_c. This possibly could be reconciled by the increase of γ which leads to a smaller tilt modulus of the flux lattice[21] and tends to reduce the overall pinning strengths.

V. CONCLUSIONS

The $\tau(\theta)$ data of the $YBa_2Cu_{3-x}Fe_xO_{7-\delta}$ single crystals were measured in the reversible regime and were consistent with Eq. (1). $\gamma=9.3\pm0.3$ for pure $YBa_2Cu_3O_{7-\delta}$ crystals, which is 20% higher than that of previously reported values. γ increases monotonically up to 29 ± 2 for $x=0.22$. No temperature dependence of γ was observed, which is in agreement with the 3D Ginzburg-Landau model. No torque directly from the Fe^{+3} magnetic moments was observed. The fundamental reason for the increase of γ with increasing x is not understood.

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