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Diamagnetism of $YBa_2Cu_3O_{6+x}$ crystals above T_c: Evidence for Gaussian fluctuations

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The magnetization of three high-quality single crystals of $YBa_2Cu_3O_{6+x}$, from slightly overdoped to heavily underdoped, has been measured using torque magnetometry. Striking effects in the angular dependence of the torque for the two underdoped crystals, a few degrees above the superconducting transition temperature (T_c), are described well by the theory of Gaussian superconducting fluctuations using a single adjustable parameter. The data at higher temperatures (T) are consistent with a strong cutoff in the fluctuations for $T \gtrsim 1.1T_c$. Numerical estimates suggest that inelastic scattering could be responsible for this cutoff.

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Cuprate superconductors show much stronger thermodynamic fluctuations than classical ones because of their higher transition temperatures (T_c), shorter Ginzburg-Landau (GL) coherence lengths, and quasi-two-dimensional layered structures with weakly interacting CuO_2 planes.[1,2] Observations of diamagnetism[3] and large Nernst coefficients over a broad temperature (T) range well above T_c for several types of cuprate[4,5] are intriguing.[6] They are often cited as evidence for preformed Cooper pairs without the long-range phase coherence needed for superconductivity. In contrast, in Ref. 7 it is argued that phase and amplitude fluctuations set in simultaneously. However, the fluctuations are still considered to be strong in that the mean-field transition temperature T_c^{MF}, obtained by applying entropy and free energy balance considerations to heat capacity data, is substantially larger than T_c, especially for underdoped cuprates. In standard GL theory the coefficient of the $|\psi|^2$ term in the free energy, where ψ is the order parameter, changes sign at $T_c^{\mathrm{MF}_1}$, as explained in Ref. 8. If $|\psi|^4$ and higher order terms are neglected, $T_c^{\mathrm{MF}_1}$ can be obtained from a Gaussian fluctuation (GF) analysis of the magnetic susceptibility and other physical properties.[1]

One difficulty in this area is separating the fluctuation (FL) contribution to a given property from the normal state (N) background. Recently this has been dealt with for the in-plane electrical conductivity $\sigma_{ab}(T)$ of $YBa_2Cu_3O_{6+x}$ crystals by applying very high magnetic fields (B).[9] When analyzed using GF theory, $\sigma_{ab}^{\mathrm{FL}}(T)$ was found to cut off even more rapidly above $T \gtrsim 1.1T_c$ than previously thought.[10,11] It was also strongly reduced at high B and the fields needed to suppress $\sigma_{ab}^{\mathrm{FL}}(T)$ extrapolated to zero between 120 and 140 K depending on x, which tends to support a vortex or Kosterlitz-Thouless scenario. Therefore questions such as the applicability of GF theory versus a phase fluctuation or mobile vortex scenario and the extent to which T_c is suppressed below $T_c^{\mathrm{MF}_1}$ by strong critical fluctuations are still being discussed. They are of general interest because superconducting fluctuations could limit the maximum T_c that can be obtained in a given class of material,[7] and, moreover,[9] the fluctuation cutoff could be linked in some way to the pairing mechanism.

Here we report torque magnetometry data measured[12] from T_c to 300 K for tiny $YBa_2Cu_3O_{6+x}$ (YBCO) single crystals from overdoped (OD) to heavily underdoped (UD). These were grown in nonreactive $BaZrO_3$ crucibles from high-purity (5N) starting materials. Evidence for the quality of the UD crystals includes extremely sharp x-ray peaks,[13] and substantial mean free paths from quantum oscillation measurements.[14] The OD89 crystal is from another preparation batch which had narrow superconducting transitions and a maximum T_c of 93.8 K.[15] We analyze the results using GF theory which, unlike some other approaches, predicts the *magnitude* of the observed effects as well as their T dependence. We show that it gives excellent single-parameter fits to the striking angular dependence of the torque, which has previously been attributed to the presence of a very large magnetic field scale.[3] We also show that inelastic scattering is a plausible mechanism for cutting off the fluctuations at higher T and a possible alternative to strong fluctuations for limiting T_c.

Although measurements of the London penetration depth[16] below T_c and thermal expansion[17] above and below T_c for optimally doped (OP) YBCO crystals give evidence for critical fluctuations described by the three-dimensional (3D) XY model, up to ± 10 K from T_c, we argue later that these do not alter our overall picture.

A crystal with magnetization M in an applied magnetic field B attached to a piezoresistive cantilever causes a change in electrical resistance proportional to the torque density $\tau \equiv \underline{M} \times \underline{B}$. If B is parallel to the c axis of a cuprate crystal, then in the low-field limit the contribution to M in the c-axis direction from Gaussian fluctuations (M_c^{FL}) is given by[2]

$$M_c^{\mathrm{FL}}(T) = -\frac{\pi k_B T B}{3\Phi_0^2}\frac{\xi_{ab}^2(T)}{s\sqrt{1+[2\xi_{ab}(T)/\gamma s)]^2}}. \quad (1)$$

Here $\gamma = \xi_{ab}(T)/\xi_c(T)$ is the anisotropy, defined as the ratio of the T-dependent coherence lengths $\parallel$ and $\perp$ to the layers, i.e., $\xi_{ab,c}(T) = \xi_{ab,c}(0)/\epsilon^{1/2}$ with $\epsilon = \ln(T/T_c^{\mathrm{MF}_1})$.[2,9] The distance between the CuO_2 bilayers is taken as $s =$ 1.17 nm, and Φ_0 and k_B are the pair flux quantum and Boltzmann's constant, respectively. For $B \perp c$ the fluctuation magnetization is negligibly small.

As the angle θ between the applied field and CuO_2 planes is altered, $\tau(\theta)$ will vary as $\tau(\theta) = \frac{1}{2}\chi_D(T)B^2 \sin 2\theta$, as long as $M \propto B$. Thus, fits to $\tau(\theta) \propto B^2 \sin 2\theta$ give $\chi_D(T) \equiv \chi_c(T) -$

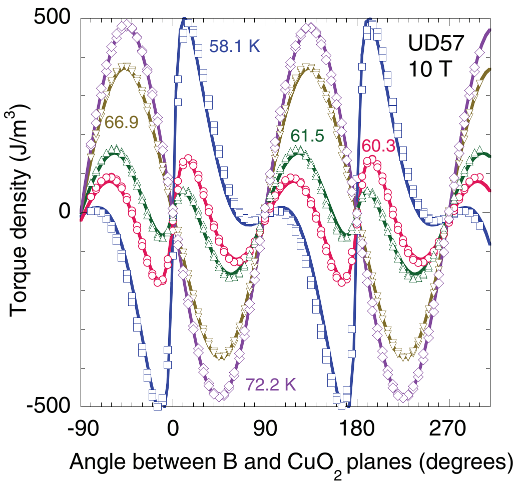

FIG. 1. (Color online) Angular dependence of the torque density for the UD57 $YBa_2Cu_3O_{6.5}$ crystal in 10 T at $T = 58.1$, 60.3, 61.5, 66.9, and 72.2 K after correcting for a fixed instrumental offset of 10° and subtracting the gravitational term (Ref. 12). The solid lines show single-parameter fits to the formula for 2D GF derived from Eq. (2) plus $\chi_D^N(T)$ shown in Fig. 2(a). Note the $\sin 2\theta$ behavior at higher T.

$\chi_{ab}(T)$, which is the susceptibility anisotropy. Figure 1 shows torque data for UD57 up to 15 K above the low-field T_c of 57 K. Much of our data, including the two curves for UD57 in Fig. 1 at higher T, follow a $\sin 2\theta$ dependence very closely, however, there are striking deviations at lower T arising from nonlinearity in $M(B)$ that we discuss later.

Figure 2(a) shows $\chi_D(T)$ obtained from $\sin 2\theta$ fits for three doping levels at high enough T so that M remains $\propto B$. The solid lines for OD89 and UD57 are fits up to 300 K that include $\chi_c^{\mathrm{FL}}(T)$ from Eq. (1), with the strong cutoff described below, plus the normal state background anisotropy $\chi_D^N(T)$ which arises from the g-factor anisotropy of the Pauli paramagnetism.[18] For UD crystals the T dependence of $\chi_D^N(T)$ is caused by the pseudogap (see Ref. 19), plus a smaller contribution from the electron pocket[18] observed in high-field quantum oscillation studies.[20] We used the same pseudogap energies ($k_B T^*$) and other parameters defining $\chi_D^N(T)$ as in our recent work on larger single crystals,[18] e.g., $T^* = 435$ K for UD57. OD89 has no pseudogap and presumably no pockets, so we represent the weak variation of $\chi_D^N(T)$ with T by the second order polynomial shown in Fig. 2(a).

Figures 2(b)–2(d) show plots of $1/|\chi_c^{\mathrm{FL}}(T)|$ vs T where $\chi_c^{\mathrm{FL}}(T) \equiv \chi_D(T) - \chi_D^N(T)$. The short-dashed lines for UD22 and UD57 in Figs. 2(b) and 2(c) show the contribution from Eq. (1) in the 2D limit ($\gamma \to \infty$) with the two adjustable parameters $T_c^{\mathrm{MF}_1}$ and $\xi_{ab}(0)$ given in Table I. The solid lines show the effect of the same type of cutoff used in previous studies of the the conductivity $\sigma_{ab}^{\mathrm{FL}}(T,B)$, as summarized in Ref. 21. For OD89 we use the full 2D-3D form of Eq. (1) with $\xi_{ab}(0) = 1.06$ nm and $\gamma = 5$,[22] shown by the short-dashed line, with the solid line again including the cutoff.[21] The high quality of these fits could be somewhat fortuitous in view of our neglect of any charge density wave (CDW),[19] but other subtraction procedures give similar values of $1/|\chi_c^{\mathrm{FL}}(T)|$. Heat capacity studies give a very similar value $\xi_{ab}(0) = 1.12$ nm for OD88 YBCO (Ref. 24) while our values for UD57 and UD22

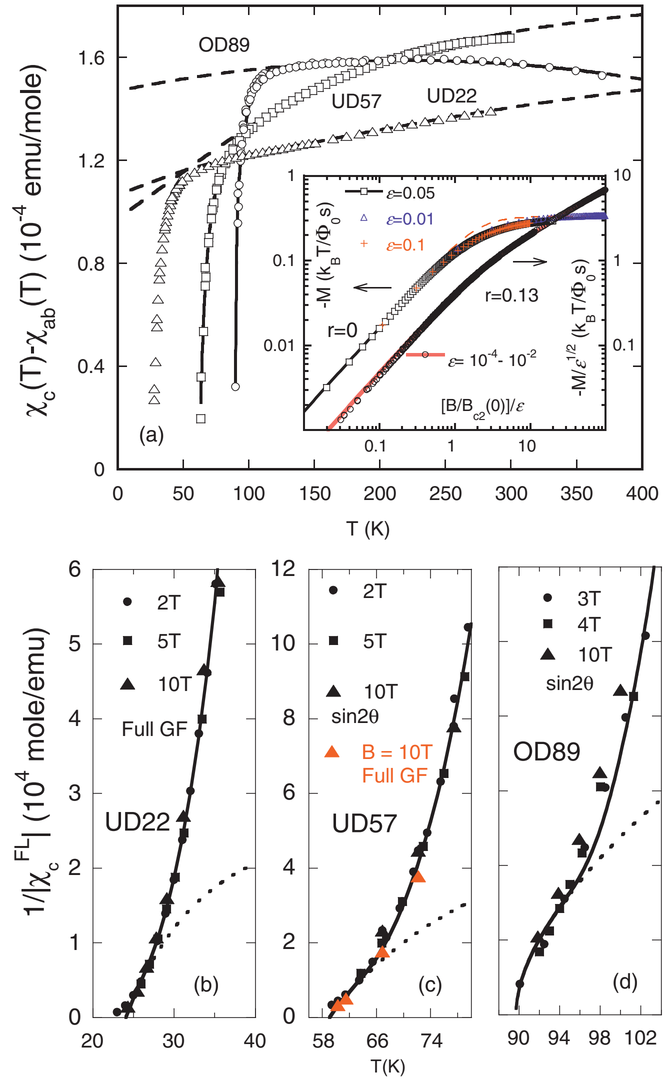

FIG. 2. (Color online) (a) Main: $\chi_D(T)$ for the three crystals; solid lines show fits to $\chi_c^{\mathrm{FL}}(T) + \chi_D^N(T)$ for OD89 and UD57, and dashed lines show $\chi_D^N(T)$. Inset: Symbols show M calculated for various values of ϵ, using Eq. (2), when the anisotropy parameter $r \equiv [2\xi_c(0)/s]^2 = 0$. For $r = 0.13$ symbols show M given by the 2D-3D form of Eq. (2), which contains r and an extra integral (Ref. 2). The lines show formulas used (Ref. 23) to represent these values of M when fitting $\tau(\theta)$ data. (b)–(d) Plots of $1/|\chi_c^{\mathrm{FL}}(T)|$ vs T for the three crystals. GF fits based on Eq. (1) are shown by short dashed lines, without a cutoff, and by solid lines, with a strong cutoff (Ref. 21). Red triangles for UD57 show $\xi_{ab}(0)^2/\epsilon$ obtained by fitting $\tau(\theta)$ to the full 2D GF formula when $M(B)$ is nonlinear, and converted to $1/|\chi_c^{\mathrm{FL}}(T)|$ using Eq. (1). For UD22 the full GF formula was used for all the points shown in (b).

agree with previous work[9,25] for the same T_c values. For UD57, setting $\gamma = 45$,[26] rather than the 2D limit of Eq. (1) ($\gamma \to \infty$), has no significant effect.

As the critical region is approached from above T_c the exponent of $\xi_{ab}(T)$ is expected to change from the MF value of $-1/2$ to the 3D XY value of $-2/3$.[1] It is very likely that this will also apply to strongly 2D materials, including UD57, since heat capacity data above and below T_c (Ref. 27) do show the $\ln|\epsilon|$ terms associated with the 3D XY model. We have

TABLE I. Summary of results.

Sample	T_c[a] (K)	$T_c^{MF_1}$ (K)	$\xi_{ab}(0)$ (nm)	$0.59\tilde{B}_{c2}(0)$[b] (T)	$\Delta(0)$[c] (K)
OD89	89.4	89.7	1.06 ± 0.1	173	448
UD57	56.5	59	2.02 ± 0.1	48	234
UD22	21.6	24	4.5 ± 0.5	10	105

[a]T_c defined by sharp onsets of superconducting quantum interference device (SQUID) signal at 10 G and torque data at ±50 G.
[b]2D clean limit formula (Ref. 2) for $B_{c2}(0)$.
[c]From the BCS relation $\xi_{ab}(0) = \frac{\hbar v_F}{\pi\Delta(0)}$, which may not hold exactly for d-wave pairing, with $v_F = 2 \times 10^7$ cm/s.

addressed this by repeating our GF fits in Figs. 2(b) and 2(c) with $\epsilon \geqslant 0.20$ (UD22) or 0.15 (UD57) without altering the cutoff.[21] The only significant change is that $\xi_{ab}(0)$ becomes 15% larger for UD57. For OD89, fits with $T_c^{MF_1} = 90$ K and $\epsilon \geqslant 0.05$ do not alter $\xi_{ab}(0)$ within the quoted error. This is expected since the width of the critical region for OD89 is much smaller than for OP YBCO (Refs. 16,17) because of the extra 3D coupling from the highly conducting CuO chains.[24]

Figure 3 shows plots of $\tau/B\cos\theta$ vs $B\sin\theta$ at fixed T for UD57. We use this representation of the data and mks units, A/m, for comparison with Ref. 3. If $\chi_D^N(T)$ is subtracted, which has not been done for Fig. 3, then since M_{ab}^{FL} is small, this would be the same as plotting M_c^{FL} vs $B \parallel c$. Near T_c there is clear nonlinearity which is remarkably consistent with GF in the 2D limit, for which the free energy density at all B is[2]

$$F = \frac{k_B T}{2\pi\xi_{ab}^2 s}\left\{b\ln\left[\Gamma\left(\frac{1}{2}+\frac{\epsilon}{2b}\right)\Big/\sqrt{2\pi}\right]+\frac{\epsilon}{2}\ln(b)\right\} \quad (2)$$

using the standard Γ function, with $b = B/\tilde{B}_{c2}(0)$, where $\tilde{B}_{c2}(0) = \Phi_0/2\pi\xi_{ab}(0)^2$, and as before, $\epsilon = \ln[T/T_c^{MF_1}(B = 0)]$. The magnetization $M = -\partial F/\partial B$ obtained by numerical differentiation of Eq. (2) for three typical values of ϵ is shown in the inset to Fig. 2(a). M scales with b/ϵ to within a few percent and for $0.01 < \epsilon < 1$ can be adequately represented by the simple formula $-bk_BT/[\Phi_0 s(3b+6\epsilon)]$, which has a single unknown parameter $\xi_{ab}(0)^2/\epsilon$. We note that GF formulas will be approximately valid in the crossover region to 3D *XY* behavior,[1] because to first order the main effect is the change in the exponent of $\xi_{ab}(T)$.

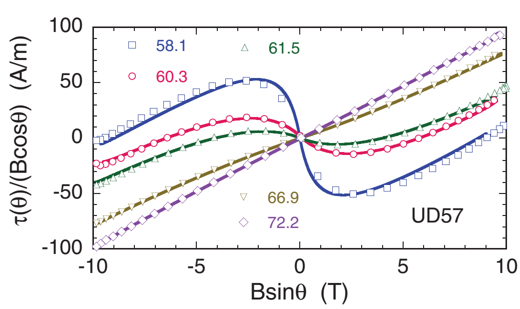

FIG. 3. (Color online) Magnetic field dependence of the magnetization obtained from the torque data for UD57 at $T = 58.1$, 60.3, 61.5, 66.9, and 72.2 K. Solid lines show fits to the 2D GF formula for M plus the same normal state contribution used in Figs. 1, 2(a), and 2(c).

Figures 1 and 3 show that this formula fits our data for UD57 very well and importantly, as shown by the red triangles in Fig. 2(c), the corresponding values of $1/\chi_c^{FL}(T)$ obtained via Eq. (1) agree well with points from $\sin 2\theta$ fits at lower B or higher T. For OD89 strong deviations from $\sin 2\theta$ behavior only occur within ~1 K of T_c and these[28] are not properly described by GF theory. For UD22 there were small jumps in $\tau(\theta)$ at $\theta = 0$ between 35 and 26 K of size $M_c = 0.01 - 0.03 k_B T/(3\Phi_0 s)$ that were fitted by including an extra contribution from Eq. (2) in the $\epsilon \ll b$ limit. This is ascribed to small regions, 1%–3% of the total volume, with higher T_c (Ref. 29) that are not detected in low-field measurements of T_c because they are much smaller than the London penetration depth. Figure 2(b) shows that the values of $\xi_{ab}(0)^2/\epsilon$ [or equivalently $1/\chi_c^{FL}(T)$] obtained from full GF fits to $\tau(\theta)$ data at 2, 5, and 10 T agree well, which supports this conclusion.

The good description of our data by this GF analysis suggests that the high critical fields proposed in Refs. 3–5 for $0.01 < \epsilon \lesssim 0.2$ are *not* associated with vortex-like excitations. In the present picture 2D GF give $M_c^{FL} \simeq -0.33 k_B T/\Phi_0 s = -0.112$ emu/cm^3 or -112 A/m at 60 K for $B \gtrsim \phi_0/[2\pi\xi_{ab}(T)^2]$. We expect this to be suppressed for $B \gtrsim B_{c2}(0)$ where the magnetic length becomes smaller than $\xi_{ab}(0)$ and the slow spatial variation approximation of GL theory breaks down. However, it may also fall when $\epsilon \gtrsim 0.1$ because of the GF cutoff discussed below. So in the first approximation the high fields are $\simeq B_{c2}(0)$. Precise analysis of these effects at very high fields might need to allow for small changes in $\chi_D^N(T)$ with B that depend on the ratio of the Zeeman energy to the pseudogap. We note that the present results are consistent with a recent study of B_{c2} for YBCO (Ref. 30) and that recent torque magnetometry data[31] for $HgBa_2CuO_{4+x}$ and other single-layer cuprates show similar exponential attenuation factors to those for YBCO.[9,21]

An intriguing question about the present results and those of Ref. 9 is the origin of the strong cutoff in the GF above ~$1.1T_c$. If the weakly T-dependent $\chi_D^N(T)$ behavior for OD89 shown in Fig. 2(a) is correct, then our $\chi_D^{FL}(T)$ data and $\sigma_{ab}^{FL}(T)$ (Ref. 9) both decay as $\exp[-(T - 1.08T_c)/T_0]$ above $T \sim 1.08T_c$ with $T_0 \sim 9$ K. If instead $\chi_D^N(T)$ were constant below 200 K, then our $\chi_D^{FL}(T)$ data would give $T_0 \sim 25$ K, a slower decay than Ref. 9. In either case the presence of this cutoff for OD YBCO rules out explanations connected with the mean distance between carriers. This is much less than $\xi_{ab}(0)$ for hole concentrations of $\simeq 1.2$ per CuO_2 unit, the value found directly from quantum oscillation studies of OD $Tl_2Ba_2CuO_{6+x}$ crystals.[32]

Assuming there are no unsuspected effects caused by d-wave pairing, one hypothesis is that the GF and possibly T_c itself are suppressed by inelastic scattering processes. In a quasi-2D Fermi liquid the inelastic mean free path l_{in} can be found from the T dependence of the electrical resistivity and the circumference of the Fermi surface. For OD YBCO the measured a-axis resistivity[25] gives $l_{in} = 2.5(100/T)$ nm, but values for UD samples are less certain because of the pseudogap. The BCS relation $\xi_{ab}(0) = \hbar v_F/\pi\Delta(0)$, where $\Delta(0)$ is the superconducting energy gap at $T = 0$, implies that, irrespective of the value of the Fermi velocity v_F, the usual pair-breaking condition for significant inelastic scattering,

$\hbar/\tau_{\rm in} \gtrsim \Delta(0)$, is equivalent to $l_{\rm in} \lesssim \pi\xi_{ab}(0)$. Taking $\xi_{ab}(0)$ from Table I and the above value of $l_{\rm in}$ shows that this is satisfied at 100 K for OD YBCO. So some suppression of GF and indeed T_c by inelastic scattering is entirely plausible. If T_c is suppressed, then $\Delta(T)$ will fall more quickly than BCS theory as T_c is approached from below, which would affect the analysis of Ref. 7.

Another possibility[9] which might account for the observations is that the pairing strength itself falls sharply outside the GL region, for example, when the in-plane coherence length becomes comparable to, or less than, the correlation length of spin fluctuations. From Figs. 2(b)–2(d) we can read off the values of T where the solid and dashed lines differ by (say) a factor of 2. At these points $\xi_{ab}(T) \equiv \xi_{ab}(0)/\ln(T/T_c^{\rm MF_1}) = 15.6$, 9.5, and 7.9 nm for UD22, UD57, and OD89, respectively. Neutron scattering studies[33,34] typically give a full width at half maximum of $0.17\frac{2\pi}{a}$ for the scattering intensity from spin fluctuations. Although this does vary with composition and scattering energy, it corresponds to a correlation length[35] of just over six lattice constants a, or 2.5 nm, similar to $\xi_{ab}(0)$ but much smaller than the $\xi_{ab}(T)$ values for which $\chi_c^{\rm FL}$ is reduced by a factor of 2. It remains to be seen whether theory could account for this.

In these two pictures the effective T_c describing the strength of the GF would fall for $T > 1.1T_c$ either because of inelastic scattering or because of a weakening of the pairing interaction. If it could be shown theoretically that $\tilde{B}_{c2}(0)$ falls in a similar way, this would account naturally for the fact[9] that the magnetic fields needed to destroy the GF fall to zero in the temperature range 120–140 K, where the fluctuations become very small. In summary, Gaussian superconducting fluctuations, plus a strong cutoff that seems to be linked to a reduction in the effective value of T_c, provide a good description of the diamagnetism of our superconducting cuprate crystals above T_c.

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[12] The crystal is glued to the end of a commercial piezolever with its CuO_2 planes parallel to the flat surface of the lever. A dummy lever compensates background magnetoresistance signals, using a three-lead Wheatstone bridge circuit driven by a floating 77 Hz current source. The chip is mounted on a single-axis rotation stage inside a ^{4}He cryomagnetic system providing stable temperatures from 1.4 K up to 400 K and fields up to 15 T. The bridge signal arising from the gravitational torque on the crystal when the sample stage is rotated in zero magnetic field gives the T-dependent sensitivity of the piezolever. Because the masses of the glue and the lever are much less than that of the crystal, the calibration constant relating the out-of-balance bridge signal to the angular-dependent torque density $\tau(\theta)$ in J/m^3 or $\chi_D(T)$ (Ref. 37) only depends on the distance between the center of mass of the crystal and the base of the lever at the silicon chip, measured to $\pm 5\%$.

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certainly responsible for the pocket. However, an unpublished analysis by J. R. Cooper and J. W. Loram (2012) of heat capacity data for UD67YBCO shows that CDW order sets in when the pseudogap is already formed. It probably causes gradual changes $\sim\pm25\%$ of the pocket contribution to $\chi_D^N(T)$ (Ref. 18), or $\pm0.035 \times 10^{-4}$ emu/mol over a T interval $\sim$30 K.

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