High-field magnetization of $HgBa_2Ca_2Cu_3O_{8+\delta}$: Fluctuations, scaling, and the crossing point

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Fluctuation-induced diamagnetism is observed well above T_c in high-field torque magnetization measurements on the cuprate superconductor $HgBa_2Ca_2Cu_3O_{8+\delta}$. Universal scaling behavior anticipated by fluctuation theories is not observed in the high-field, high-temperature regime. A crossing point, where M reaches a field-independent value at a temperature which characterizes the interlayer separation, disappears in the fluctuation regime above $\sim H_{c2}$, suggesting that deviations from the lowest Landau level description of Ginzburg-Landau theory appear at high fields.

Superconductors are known to exhibit diamagnetic susceptibility above the critical temperature T_c due to thermodynamic fluctuations. In the first reports of fluctuation magnetization 30 years ago, Gollub *et al.* noted clear deviations from behavior expected from Ginzburg-Landau (GL) theory.[1] It was recognized soon thereafter that the magnetization data exhibited universal scaling behavior.[2] This scaling was then partly explained using corrections to GL theory which account for short-wavelength fluctuations,[3] an approach improved upon by incorporating nonlocal effects.[4,5]

More recently, the dependence of the magnetization M on applied magnetic field H in layered cuprate superconductors has been shown to vanish at a characteristic temperature $T^* < T_c$, as a result of a balance between the entropic effects of thermal fluctuations and the free energy of the equilibrium vortex lattice.[6–8] Within GL theory, with only the lowest Landau level occupied, the magnitude M^* of the magnetization at T^* provides a measure of the distance s between superconducting layers, i.e., $M^* \sim T^*/s$, as shown by several groups.[9–11] Associated with this crossing point, fluctuation scaling formulas have been derived as functions of field and temperature, allowing one to discern the dimensionality (two or three dimensions) of the superconductivity in the material under study.[12,13] The only fitting parameter in the scaling laws is the field-dependent transition temperature $T_c(H)$, with $M/(TH)^\alpha$ scaling as $[T - T_c(H)]/(TH)^\alpha$, where α is a dimensionality exponent. An important application of this scaling is the extraction of a mean-field upper critical field $H_{c2}(T)$ via $T_c(H)$ from magnetization data, a feat not possible in anisotropic superconductors from conventional analyses of $M(T)$ or $M(H)$ plots, which yield a ''T_c'' which increases with field.[6]

The majority of published magnetization data on the cuprates have been taken in relatively low (for the cuprates) magnetic field, typically below 5 T. As mentioned above, fluctuation scaling appears to work well in such fields, including its prediction of the crossing point (M^*, T^*). We show below, however, that for the layered cuprate superconductor $HgBa_2Ca_2Cu_3O_{8+\delta}$, the fluctuation-induced diamagnetism continues to increase above T_c and H_{c2}, leading to a disappearance of the crossing point in fields greater than $H_0 \sim H_{c2}(T^*)$. As a result, the scaled magnetization no longer follows the theoretical prediction of Ref. 4 when plotted versus scaled magnetic field. However, it appears that $M(T,H)$ continues to scale with the temperature, with the caveat that the two-dimensional (2D) and 3D functions above seem to work equally well, and thus do not allow for a clear identification of the dimensionality of the superconductivity. These facts question the high-field applicability of the scaling laws which led to the prediction of the crossing point, as well as the suitability of the lowest Landau level approximation in this system.

A single-crystal $HgBa_2Ca_2Cu_3O_{8+\delta}$ sample measuring $0.4 \times 0.25 \times 0.14\,mm^3$ was attached with grease to the free end of a silicon microelectromechanical systems (MEMS) cantilever magnetometer,[14] with the crystal c axis tilted at an angle of 20° from the plane of the cantilever (see Fig. 1). The transition temperature of the sample was $T_c = 132$ K, as determined by superconducting quantum interference device (SQUID) magnetometry at 500 Oe. This T_c implies that the

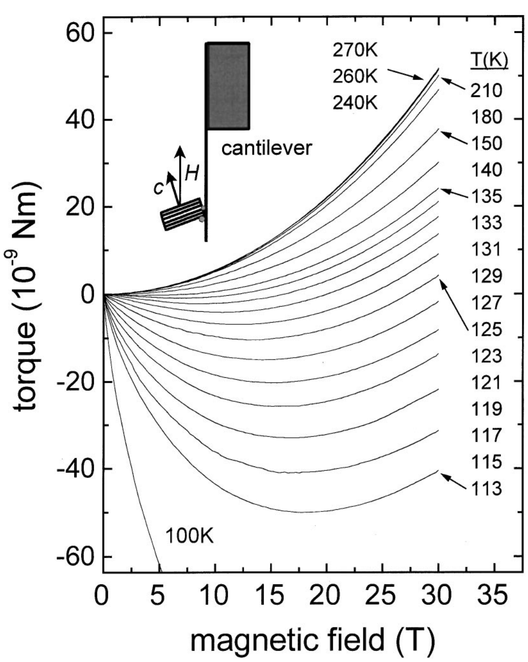

FIG. 1. Torque versus magnetic field at various temperatures. At high T, the torque is independent of T, and results from a constant paramagnetic, normal-state susceptibility. Below 240 K, superconductivity-induced diamagnetism sets in. The sample/magnetometer configuration is shown.

oxygen content of the sample is near optimal doping. X-ray analysis[15] of crystals from the same source reveals little evidence for minority phases, but does suggest that overdoping is common, with excess oxygen ($\delta \sim 0.1$) restricted to the basal plane. The magnetometer was mounted with the plane of the cantilever parallel to the applied magnetic field produced in a 30-T magnet. The application of field produced an anisotropic magnetization in the layered material, which in turn generated a torque seen by the cantilever. The system temperature was regulated in helium exchange gas with a calibrated Cernox resistor, which in the employed temperature range has a magnetoresistance corresponding to a maximum $\Delta T/T$ of 0.02% (20–30 mK). This mode of temperature regulation was found to be slightly better than that afforded by a capacitive sensor, which tends to drift with time. Isothermal field sweeps and fixed field temperature sweeps were taken. To within our instrumental noise level ($\sim$10 aF in resistive magnets, corresponding to less than 10^{-11} Nm), the capacitance of the silicon cantilever showed no field dependence. A small temperature dependence of the empty cantilever device ($\sim$6 ppm/K) was subtracted from the temperature sweep data. The cantilever was calibrated *in situ* to $\sim$1% accuracy by means of a fixed current passing through a coil deposited on the cantilever surface.[14]

We show in Fig. 1 torque versus magnetic field for various temperatures from 100 to 270 K. Between 250 and 270 K, the torque is independent of temperature. This is taken to represent the normal-state response. It varies identically as H^2, meaning the perpendicular magnetization τ/H varies linearly with field, such that the normal-state susceptibility $\Delta\chi \sim \tau/H^2$ is field and temperature independent. Starting at 240 K, a deviation from the normal-state background is detected. This diamagnetic deviation increases in magnitude as temperature is reduced, especially near and below T_c ($B=0$). The torque is reversible over the entire H-T regime shown (sizable irreversibility was seen to set in below $\sim$85 K). In order to analyze the magnetization associated with the superconducting state, we then subtract the high-T, normal-state torque from all the data in Fig. 1. This, of course, assumes that the normal-state contribution remains independent of temperature at all temperatures, an important assumption which can only be fully verified by completely suppressing superconductivity. In a separate experiment at 20 T, the torque was found to be independent of temperature between 295 and 330 K. We next divide the net torque $\tau(H,T) - \tau(H, 270\text{ K})$ by the applied field $\mu_0 H$ and the sine of the tilt angle to get the component of the magnetic moment $m(H,T)$ along the crystal c axis. This convention leaves open the question of the dimensionality of the superconductivity until after the scaling (below), since if the material is 2D, one expects the moment along c to dominate, and if it is more properly described as 3D, only the magnitude of m, and not its dependence on H or T, are affected. That is, so close to the c axis, the a-b plane moment (and torque) will be small compared to the c-axis signal.

The resulting moment versus field curves are shown in Fig. 2, from 100 to 240 K, on a semilogarithmic scale (lower panel). The overall low-field behavior is as expected, with $-m(H)$ increasing with field in the high-temperature weak fluctuation regime,[16,17] and decreasing linearly in log H in the vortex liquid state below T_c.[18] There is a temperature regime near 126 ± 1 K where the magnetization appears to be independent of field, especially below $\sim$5 T. In previous studies on Hg-1223 to 3 T, such a ''crossing point'' was indeed observed at $T^* = 127$ K.[19] However, upon close observation, the magnetization at the high-field end in Fig. 2 can be seen to increase in magnitude at all temperatures above $\sim$120 K. In fact, $-m(H)$ at 127 K increases by more than 20% at 30 T over its zero field value (see upper panel in Fig. 2). These data imply that a crossing point does not exist at high field, such that the field dependence of the fluctuation magnetization does not saturate as anticipated. This in turn suggests that an imbalance arises in strong magnetic fields between thermal fluctuations and the elasticity of the fluctuating vortices.

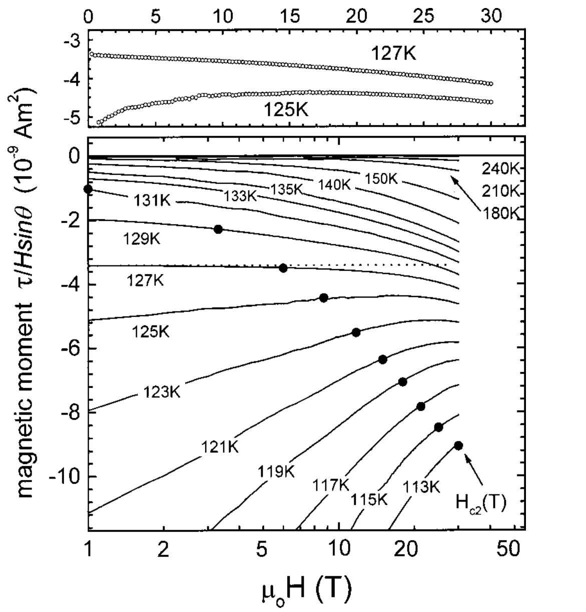

FIG. 2. Fluctuation-induced diamagnetism obtained from torque data of Fig. 1, after subtraction of the normal-state contribution. The dashed line at 127 K shows that the magnetization becomes field dependent above $\sim$5 T. The solid circles indicate the field positions at each temperature of $H_{c2}(T)$, as obtained from the 3D scaling in Fig. 4. Top panel shows $m(H)$ in vicinity of T^*.

The loss of the crossing point at high field is shown more clearly when we make a cross plot of the data in Fig. 2 in constant fields. This is shown in Fig. 3, a plot of the volume magnetization $M = m/V$ versus temperature for fields between 2 and 30 T. On the grand scale in the main figure, it appears as if there exists a single point where all the $M(T)$ curves cross, near 126 K. However, as seen in the inset, the low-field traces cross each other at $T_{max} \sim 126.5$ K, while for higher fields, the crossing temperature systematically decreases to $T_{min} \sim 122$ K. With regard to an issue of experimental uncertainty, and the potential for an artifactual origin to the loss of the crossing point, we point out the following. For the 30-T curve to be able to cross the lower field curves at T_{max} in Fig. 3, instead of at the observed T_{min}, it must be moved up by $\sim$60 A/m. This corresponds to a torque change of 6.6×10^{-9} Nm which, referring to Fig. 1, would mean that the ''real'' torque trace at 127 K should end up somewhere between the present 129 and 131 K traces. We estimate the uncertainty in the absolute torque to be less than 1×10^{-11} Nm,[14] only $\sim$1% of that required by this example.

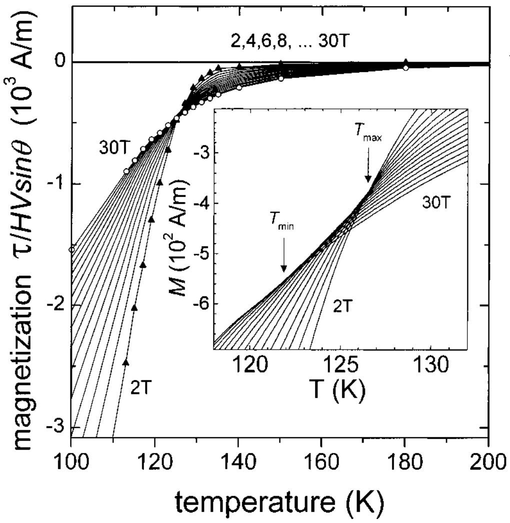

FIG. 3. Magnetization versus temperature at fixed field from 2 to 30 T, in 2-T steps, showing apparent ''crossing point'' near T^* ~ 126 K. Inset details the crossing region, and shows that T^* actually varies with field.

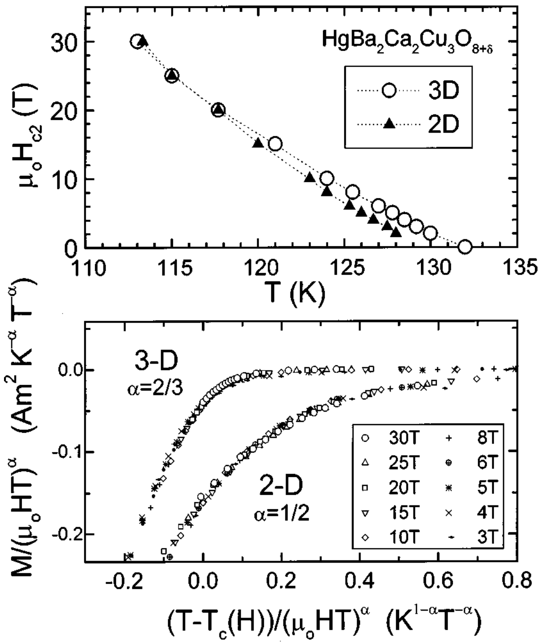

FIG. 4. Lower: 3D and 2D scaling plots of magnetization data above and below T_c, using scaling equation in the text. Both functions provide reasonable scaling fits. Upper: $H_{c2}(T)$ datum points used in the scaling plots.

Moreover, if a finite temperature dependence to the normal-state background susceptibility was to be responsible, it would mean that this was a paramagnetic susceptibility which increased with increasing temperature. This contrasts with typical paramagnetic dependencies, which are either T^0 (Pauli, Van Vleck) or T^{-1} (Curie). In fact, if a magnetic-impurity Curie(-Weiss) contribution was present, it would mean that the diamagnetism is even larger than shown in Figs. 1 and 2, and the crossing point in Fig. 3 is even less certain. Thus we believe the high-field deviation from a field-independent magnetization in Fig. 2, and the resulting loss of the crossing point in Fig. 3, to be real. In a related publication, Xue *et al.*[20] showed that the crossing point ratio T^*/M^* for the Hg-based cuprates does not scale linearly with interlayer distance s as predicted, and instead is independent of s. This led them to suggest that the theoretical description of the role of fluctuations in quasi-2D superconductors, which led to the crossing point idea, may need to be modified.[20] Junod *et al.*[21] have shown that anisotropic 3D superconductors such as YBCO have a crossing point in $M/H^{1/2}$ vs T rather than in M. In the present case, we find some improvement using this function, but a spread in crossing temperatures remains, with $T_{max}-T_{min}\sim 2.4$ K.

The absolute value of the perpendicular magnetization $\tau/HV = M\sin\theta$ in the vicinity of this crossing regime is close to values measured for Hg-1223 in Refs. 19 and 22, $-M^*\approx 150$ A/m. In our case, the apparent $-M^*$ increases from $\sim$130 A/m below 5 T to almost 180 A/m at 30 T. According to Koshelev's refinement[11] of the Bulaevskii-Ledvig-Kogan theory, $M^* = 0.346 k_B T^*/s\phi_0$, where k_B and ϕ_0 are Boltzmann's constant and the flux quantum, respectively. Use of this equation and values from Fig. 3 yields a superconducting layer thickness s between 2.3 nm (2 T) and 1.5 nm (30 T), values to be compared with a unit-cell parameter along the c axis of 1.6 nm. Employing the crossing point hypothesis therefore implies a reduction of s in high magnetic field, an aspect that to our knowledge has not been addressed theoretically.

The magnetization (and other properties derived from the free energy) in the fluctuation regime has been shown theoretically to exhibit scaling behavior as a function of magnetic field and temperature.[3,4,12,23] Essentially, two scaling formulas have been derived, one for 3D superconductors, and one for quasi-2D.[8,9,12] Both have the quantity $M/(TH)^\alpha$ scaling as $[T\text{-}T_c(H)]/(TH)^\alpha$, where α is $\frac{2}{3}$ in 3D and $\frac{1}{2}$ in the quasi-2D case.[13] Thus, by so scaling magnetization data, one should in principle be able to determine the effective dimensionality of a superconductor. This scaling procedure has been successfully applied to a number of anisotropic cuprate superconductors, including YBCO,[7] Bi-2212,[8,9] Hg-1212,[24] and some BEDT-TTF molecular superconductors.[25,26]

In the present case, we were able to scale our magnetization data to both the 3D and 2D formulas with fits of essentially equal quality, as shown in Fig. 4. However, different sets of fitting parameters $T_c(H)$ were used in the two cases. These parameters are plotted as H_{c2} versus T in the upper panel. For the scaling plots, we found that for both values of dimensionality exponent α, the scaling seemed to work well over broad ranges of temperature and field (i.e., for abscissa from -1.0 to $+1.5$, though we show a smaller range in the figure to emphasize the closeness of the 2D and 3D fits). Hence, on the basis of this scaling comparison, we are simply not able to determine whether the superconductivity is of a 2D or a 3D nature. One might conclude that the dimensionality of Hg-1223 is best described as somewhere between the two. We note that both scalings yield slight positive curvature in H_{c2} at the field end.

Another measure of dimensionality in a layered superconductor is provided by the anisotropy parameter $\gamma = \xi_\parallel/\xi_\perp$, the ratio of the superconducting coherence lengths parallel and perpendicular to the layers. For YBCO, $\gamma\sim 7$, and so it is not surprising that it scales as 3D.[7] A similar case arises for Hg-1212, $\gamma \leqslant 7.7$.[25] (BEDT-TTF)$_2$Cu[N(CN)$_2$]Br had an anisotropy ratio of $\gamma\sim 25$, and was reported to be well de-

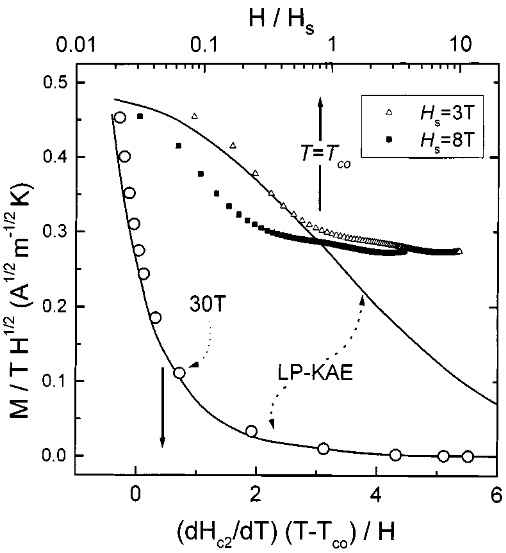

FIG. 5. Lower: Comparison of T dependence of the scaled magnetization at T_c with LP-KAE model. Upper: Scaled H dependence with two scaling fields H_s (see text).

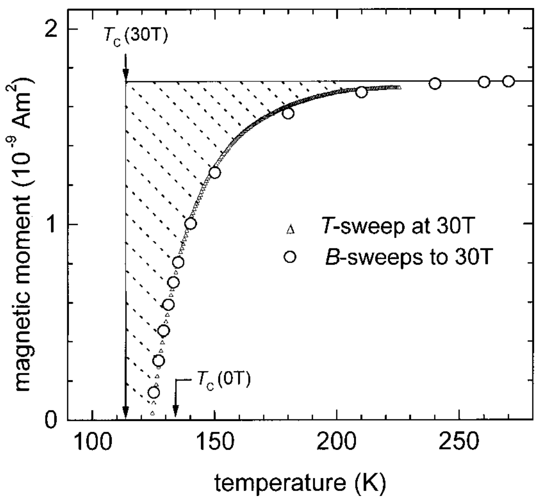

FIG. 6. This plot shows the extent of the measurable fluctuation regime above T_c in a 30-T magnetic field. The total magnetic moment (i.e., without removal of normal-state contribution) is plotted vs T, from both fixed temperature field sweeps from Fig. 1 and a fixed-field (30-T) temperature sweep. The hatched region shows that a diamagnetic signal persists to above 220 K.

scribed as 2D.[26] In Ref. 25, a similar γ of 30 was found, but neither the 2D nor the 3D scaling worked particularly well. For Bi-2212, reports of γ range from 55 (Ref. 27) up to 1500,[28] such that a 2D scaling should be appropriate, as was observed.[8] However, one cannot tell from the literature to what degree the 3D scale works for this system, as only 2D scaling plots were shown.[8] The material in the present study, Hg-1223, was reported in Ref. 19 to have $\gamma = 52$. Based on the above information, this should be large enough to scale as 2D, and not as 3D. We are thus faced with a contradiction of sorts in the present work. Either Hg-1223 ($\gamma \sim 50$) is both quasi-2D and quasi-3D (i.e., borderline), or the fluctuation scaling approach is an insufficiently resolute method to determine dimensionality. Support for the latter hypothesis is found in similar results on $(BEDT\text{-}TTF)_2Cu(NCS)_2$, with a reported anisotropy ratio greater than 100, but where 2D and 3D scalings perform equally well, in fields to 5 T.[25]

Whatever the dimensionality of the superconductivity, it is quite clear that the fluctuation regime can be very large, reaching close to twice T_c in some of the classical[29] and other cuprate[30] superconductors, for example. In the present case, field-induced diamagnetism is observed above 200 K at high field, approaching perhaps 240 K or beyond at 30 T. This is shown in Fig. 5, where we plot $m(T)$ at 30 T. The circles are taken from the field sweep data in Fig. 1 at 30 T, after dividing by the field, without any background or normal-state corrections. The solid line is the direct magnetic moment from a temperature sweep in constant field, where the moment signal is obtained from a feedback current provided to the nulling/calibration loops on the cantilever surface (i.e., $m = IA_{\text{loop}}$). We were only able to obtain reliable and reproducible data in this manner up to ~225 K, above which we could not maintain thermal equilibrium in the sample chamber without exceedingly long sweep times in the high field. The horizontal line across the top represents our assumed temperature-independent normal-state magnetization, while the vertical line signifies the transition temperature at 30 T from Fig. 4. The hatched region therefore provides a view of the fluctuation magnetization regime above T_c, which extends beyond 220 K, or $\geqslant 1.7 T_c$. This increase of the temperature span is characteristic of the weak fluctuation regime,[16,28] such that 30 T should be considered a weak field for this material.

We compare in Fig. 6 our measured fluctuation magnetization at 30 T with that predicted by the high-field limiting case of a model calculation by Lee and Payne (LP) (Ref. 4) and Kurkijarvi, Ambegaokar, and Wilkins (KAE),[5] which is based on the work of Schmidt,[15] Schmid,[16] and Patton, Ambegaokar, and Wilkins.[3] The LP-KAE model accounts for high-energy (short-wavelength) fluctuations and nonlocality due to the magnitude of B, in contrast to the Landau-Ginzburg phenomenological treatment,[31] and starts with a free energy derived from a clean limit microscopic theory.[32] It appears that the temperature dependence of M at 30 T (lower curve in Fig. 5) can be fairly well described by this model. This supports the idea that fluctuation theories which go beyond low-field Landau-Ginzburg, such as those in Refs. 3–5, may be necessary to explain magnetization data in strong magnetic fields as employed here. Using the LP-KAE model, Gollub, Beasley, and Tinkham[2] have shown that the quantity $M/TH^{1/2}$ in Fig. 6 also exhibits universal field dependence, when H is scaled by a scaling field H_s, characterizing the field at which deviations from GL theory become appreciable. In the theory, $H_s = 0.083\phi_0/2\pi\xi_0^2$, with the coherence length ξ_0 obtained from $T_c dH_{c2}/dT|_{T_c} = \phi_0/2\pi(0.74)^2\xi_0^2$. Using our $T_c = 132$ K and a critical-field slope from Fig. 4 of 1.35 T/K, we obtain $\xi_0 \sim 1.9$ nm, yielding a scaling field of $H_s = 8$ T. As seen in Fig. 6 (top), the magnetization fails to scale in the predicted manner at any field for this H_s. The dirty-limit model of Maki and Takayama[33] does not fully solve the problem. If we consider that the coherence length itself is anisotropic, a fact not directly addressed in the above theories, we can adjust ξ_0 to obtain a better fit. The curve with $H_s = 3$ T ($\xi_0 \sim 3$ nm) provides agreement at low field, but again fails at high fields, $H > H_s$. This is a reflection of the anomalous increase of M

with H even well above T_c (Fig. 2).

We have taken the critical temperature used as the adjustable parameter in the 3D scaling fit in Fig. 4, inverted it to get $H_{c2}(T)$, and plotted the resulting phase points as closed circles in Fig. 2. We note that the magnetization at fields lower than these points, and below 127 K, the temperature at which there does appear to be a field-independent magnetization, and therefore a crossing point, varies approximately linearly in $\log H$, as predicted by theory for the vortex state. At higher fields, i.e., above H_{c2}, $m(\log H)$ develops curvature, tending to an increase of diamagnetism, as in the fluctuation state above 127 K. This suggests a correlation between $H_{c2}(T^*\sim 127\,\mathrm{K})$ and the disappearance of the crossing point. Note that at 127 K, the magnetization continues to decrease (i.e., increasing diamagnetism) out to the maximum field, which is of order $5H_{c2}$. It would be interesting to follow the curves in Fig. 2 to much higher fields, to observe the inevitable turnaround in $m(H)$ toward decreasing diamagnetism, as all remnants of fluctuations become quenched.

In summary, we have measured the magnetization of a single crystal of Hg-1223 up to 30 T, in the vicinity of and well above T_c. We found that a crossing point (M^*,T^*), anticipated for low dimensional superconductors and associated with superconducting fluctuations, does not explicitly exist in magnetic fields above $H_{c2}(T^*_{\mathrm{low\ field}})\sim 5\,\mathrm{T}$ in $HgBa_2Ca_2Cu_3O_{8+d}$ $(\gamma\sim 50)$. The data seem to adequately scale in manners prescribed by both 2D and 3D fluctuation theories, such that one cannot readily discern the dimensionality of the superconductivity. These results suggest that modifications to the models describing vortex fluctuations in quasi-2D superconductors are necessary. The upper critical field $H_{c2}(T)$ was derived from the data using the scaling equations. Fluctuation-induced diamagnetism was observed beyond 220 K at 30 T.

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