Magnetization of underdoped YBa$_2$Cu$_3$O$_y$ above the irreversibility field

Jing Fei Yu, 1,* B. J. Ramshaw, 2 I. Kokanović, 3, 4 K. A. Modic, 5 N. Harrison, 2 James Day, 5 Ruixing Liang, 5, 6 W. N. Hardy, 5, 6 D. A. Bonn, 5, 6 A. McCollam, 7, S. R. Julian, 5, 6 and J. R. Cooper 5

1 Department of Physics, University of Toronto, Toronto, Ontario M5S 1A7, Canada
2 Los Alamos National Laboratory, Mail Stop E536, Los Alamos, New Mexico 87545, USA
3 Cavendish Laboratory, University of Cambridge, Cambridge CB3 0HE, United Kingdom
4 Department of Physics, Faculty of Science, University of Zagreb, P.O. Box 331, Zagreb, Croatia
5 Department of Physics and Astronomy, University of British Columbia, Vancouver, British Columbia V6T 1Z1, Canada
6 Canadian Institute for Advanced Research, 180 Dundas St. W, Toronto, Ontario M5S 1Z8, Canada
7 High Field Magnet Laboratory (HFML-EMFL), Radboud University, 6525 ED Nijmegen, The Netherlands

(Received 18 September 2015; revised manuscript received 1 November 2015; published 23 November 2015)

Torque magnetization measurements on YBa$_2$Cu$_3$O$_y$ (YBCO) at doping $y = 6.67$ $(p = 0.12)$, in dc fields ($B$) up to 33 T and temperatures down to 4.5 K, show that weak diamagnetism persists above the extrapolated irreversibility field $H_{irr} (T = 0) \approx 24$ T. The differential susceptibility $dM/dB$, however, is more rapidly suppressed for $B \gtrsim 16$ T than expected from the properties of the low field superconducting state, and saturates at a low value for fields $B \gtrsim 24$ T. In addition, torque measurements on a $p = 0.11$ YBCO crystal in pulsed field up to 65 T and temperatures down to 8 K show similar behavior, with no additional features at higher fields. We offer two candidate scenarios to explain these observations: (a) superconductivity survives but is heavily suppressed at high field by competition with charge-density-wave (CDW) order; (b) static superconductivity disappears near 24 T and is followed by a region of fluctuating superconductivity, which causes $dM/dB$ to saturate at high field. The diamagnetic signal observed above 50 T for the $p = 0.11$ crystal at 40 K and below may be caused by changes in the normal state susceptibility rather than bulk or fluctuating superconductivity. There will be orbital (Landau) diamagnetism from electron pockets and possibly a reduction in spin susceptibility caused by the stronger three-dimensional ordered CDW.

DOI: 10.1103/PhysRevB.92.180509

The possible existence of bulk superconductivity as $T \rightarrow 0$ K above the irreversibility field ($H_{irr}$) [1] in the cuprates has been a long-standing question. Not only is this problem important for our understanding of the cuprates, but also because there is still debate [2, 3] about whether Cooper pairs persist in the region of the field-temperature plane where quantum oscillations are seen [4].

Many experimental efforts have been made to address this issue [5–8]. Diamagnetism has consistently been reported using torque magnetometry at high fields in many families of cuprates and it is argued that this observation shows the persistence of Cooper pairs above $H_{irr}$ [5]. For YBa$_2$Cu$_3$O$_5$ (YBCO), resistivity measurements have established $H_{irr} (T = 0)$ to be below 30 T for fields along the $c$ axis for dopings between $p = 0.11$ (OII) and $p = 0.12$ (OIII) [9]. Moreover, x-ray [10–12], NMR [13], and sound velocity measurements [14] have demonstrated the existence of static charge-density-wave (CDW) order that competes with superconductivity: Reference [12] shows a distinct long-range three-dimensional (3D) order that emerges at high field and continues to grow at 28 T for an OVIII crystal, consistent with that first observed in NMR studies [13]. The CDW is strongest and the suppression of $H_{c2}$ is largest at $p = 0.125$ for YBCO [11, 15].

Recent thermal conductivity measurements by Grissonnanche et al. [7] show a sharp transition precisely at the extrapolated $H_{irr} (T = 0) \approx 22$ T for OII YBCO. They have interpreted this feature (henceforth referred to as $H_K$) as a signature of $H_{c2}$, arguing that the end of the rapid rise in thermal conductivity at 22 T reflects a corresponding increase in the mean free path as a result of the sudden disappearance of vortices at $H_{c2}$. On a crystal with same doping, Marcenat et al. [16] show that the specific heat saturates at a field $H_{sp}$, $H_{sp} (T)$ lies above $H_{irr} (T)$, but they extrapolate to the same value at $T = 0$ K [16]. In contrast, torque measurements by Yu et al. [6] on a crystal with the same doping suggested diamagnetism persisting to fields much higher than $H_K$. The debate is thus still open.

To resolve this problem, we conducted torque magnetometry measurements of magnetization ($M$) on two $p = 0.12$ (OVIII, $T_c = 65$ K) crystals in dc fields and one $p = 0.11$ (OII, $T_c = 60$ K) crystal in pulsed fields. The crystals were mounted on piezoresistive cantilevers and placed on a rotating platform, with the CuO$_2$ planes parallel to the surface of the lever. dc field sweeps, first from 0 to 10 T and later from 0 to 33 T, were performed with the $c$ axis of the OVIII crystal at a small angle $\theta$ from the field. The magnetoresistance of the levers was eliminated by subtracting data from the complementary angle ($-\theta$) (see Supplemental Material for raw data [17]). Similar procedures were used for the OII crystal in pulsed magnetic fields up to 65 T. For strongly anisotropic superconductors, where out-of-plane screening currents can be neglected, the torque $\tau$ per unit volume $V$ at an angle $\theta$ from field $B$ is given by [18] $\tau/V = \frac{1}{2} \chi_D (T) B^2 \sin 2 \theta + M_B \sin \theta$. Here $\chi_D (T) = \chi_c (T) - \chi_{ab} (T)$ is the anisotropic susceptibility in the normal state and $M_B$ is the magnetization from in-plane screening currents for a field of $B \cos \theta$ along the $c$ axis. This is a good approximation when $M_c \gg \chi_B$ or when the superconducting gap and $M_c$ are both small.
A key challenge with magnetization measurements in the cuprates is the separation of the normal state from the superconducting contributions, because superconducting fluctuations are thought to contribute to \( \chi(T) \) even at temperatures far above \( T_c \) [18], while \( \chi^{\text{normal}} \) is temperature dependent to well below \( T_c \). We follow the procedure outlined in Refs. [18,19] and interpret \( \chi(T) \) in the normal state of underdoped YBCO as arising from the pseudogap and \( g \)-factor anisotropy, plus a superconducting fluctuation term that sets in below 120 K. Neglecting isotropic Curie and core susceptibility terms, which do not contribute to \( \tau \), the total normal state contribution to \( \chi(T) \) is [19]

\[
\chi^{\text{normal}}_D(T) = \chi^{\text{PG}}_D(T) + \chi^{\text{VV}}_D + \chi^{\text{R}}_D(T),
\]

where \( \chi^{\text{VV}}_D \) is the \( T \)-independent Van Vleck susceptibility, \( \chi^{\text{PG}}_D(T) \) is the pseudogap contribution assuming a V-shaped density of states [20], and \( \chi^{\text{R}}_D(T) \) is thought to arise from an electron pocket or Fermi arcs in the region 0.0184 < \( p < 0.135 \). Specifically, \( \chi^{\text{PG}}_D = A[1 - y^{-1}\ln\cosh(y)] \), where \( A = N_0\mu_B^2/2k_B T \), \( y = E_F/2k_B T \), \( E_F = k_B T^* \) and \( T^* \) is the pseudogap temperature, and \( \chi^{\text{R}}_D(T) = \chi^{\text{R}}(0)[1 - \exp(-T_F/T)] \) where \( T_F \) is the Fermi temperature. The fit is shown in Fig. 1, along with a linear model for the normal state \( \chi \) used in Ref. [6]. Both fits agree well with the data for \( T \gtrsim 120 \) K.

Our background is almost twice as small as that of the linear model; it is about \( 150 \) A/m at 30 T.

In Figs. 2(a) and 3(a), we show \( M_c \) vs \( B_z \) curves at selected temperatures for the OVIII and OII crystals, obtained by subtracting \( M_{BG} = \chi_{BG} B_z \), where \( \chi_{BG} \) is the blue line in Fig. 1, and \( B_z \) is the field projected along the crystalline \( c \) axis. For the OVIII crystal, at \( T = 103 \) K, we see that \( M_c \) is almost zero. At 58 K, just below \( T_c \), we see significant diamagnetism that gradually tends to about \( -130 \) A/m at high field. Figure 2(a) shows that the crystal remains weakly diamagnetic down to 4.5 K in fields up to 33 T. Similar behavior was found for the OII crystal in pulsed fields. As shown in Fig. 3(a), \( M_c \) is still diamagnetic at the highest field \( B_z = 63 \) T, but has a small value — about \( -90 \) A/m at 8 K. Our results differ from those of Yu et al. [6]: Our normal state susceptibility is larger than theirs by approximately 8 A/m/T, and after background subtraction, at 10 K and 20 T we find \( M_c \) to be up to four times larger (see Supplemental Material for details on the calibration procedure); at 30 T we find about \( -200 \) A/m for OII and OVIII rather than their value of \( -75 \) A/m. Our estimated uncertainty in \( \chi_D(0) \) corresponds to \( \pm 32 \) A/m in \( M_c \) at 33 T and \( \pm 61 \) A/m at 63 T.

Although the weak diamagnetic signal persists to higher fields, we are able to see a signature in our differential susceptibility \( dM/dB \) at fields comparable to \( H_K \) (22 T) found by thermal conductivity [7]. In each curve of Figs. 2(b) and 3(b), \( dM/dB \) decreases linearly, up to a field we call \( H_d = 24 \) T, which is close to the extrapolated \( H_m(T = 0) \). This is consistent with the feature at \( H_K \) found by thermal conductivity.
MAGNETIZATION OF UNDERDOPED YBa$_2$Cu$_3$O$_{\ldots}$

FIG. 3. (Color online) (a) Magnetization ($M_r$) of the OII crystal measured in pulsed magnetic field up to $B_z = 63$ T, where $M_r = M_{obs} - M_{BG}$, $M_{BG} = \chi_D B_z$, and $\chi_D$ is the blue line in Fig. 1. For clarity only the falling-field sweeps are shown. Diamagnetism is present though extremely weak at high field (inset). The small offset for $0 < H / H_c < 2$, has the mean field property of saturating toward a constant value, but this is very small and requires field dependence [22] for $\lambda(T = 0)$, but results of tunneling experiments on Bi-2212 imply thermally induced pair breaking near the nodes [23], indicating a weaker field dependence at higher $T$. Thus, for simplicity, we assume a negligible field dependence of $\lambda$. We also assume [21] $\alpha = 0.77$ and $\beta = 1.44$ for $0.02 < H / H_{c2} < 0.3$, in reasonable agreement with later works [24,25], and we fit the low field magnetization and obtain an estimate of $H_{c2}(T)$, shown in Fig. 4. Since our GL values of $H_{c2}$ join smoothly to $H_d$, it is possible to interpret $H_d$ as the low temperature GL-type $H_{c2}$.

When $H / H_{c2} > 0.3$, and again using cgs units for an $s$-wave superconductor, the magnetization is expected to obey $4\pi M = (H - H_{c2})/(2\kappa^2 - 1)\beta_A$, where $\kappa$ is the GL parameter and $\beta_A = 1.16$ is the Abrikosov parameter [26,27]. Figures 2(b) and 3(b) show that for $B > 28$ T, $dM/dB$ has the mean field property of saturating toward a constant value, but this is very small and requires $\kappa \approx 50$, a value far greater than $\kappa = 50$ given in Ref. [7]. This means that our high field $dM/dB$ is nearly ten times smaller than would be expected. This may be due to the field dependent charge-density-wave (CDW) order within the vortex liquid region [11,12]. The CDW competes with superconductivity and is partially suppressed at low field. As increasing field suppresses superconductivity, the CDW order is gradually restored [14]. The presence of a relatively strong CDW would increase $\lambda$ and thus increase $\kappa$, as illustrated in Fig. 5. A linear region in $M_r(B)$ can also be seen in Fig. 2(a), for $T = 20$ K and $T = 16$ K and $B \leq 17$ T, with $\kappa = 41$, and in Fig. 3(a), for $T = 20$ K and $B \leq 17$ T, with $\kappa = 50$. These linear regions are not present above 20 K, where $M_r(B)$ is likely to be smeared out by thermal fluctuations. As shown in Fig. 5, for the OVIII crystal, the low field $M_r$ extrapolates to zero around 24 T, consistent with our GL-type $H_{c2}$. To summarize, clear linear regions, with slopes corresponding to the expected values of $\kappa$, have been observed for both doping levels below 20 K.

The value of $H_{c2}(0) \approx H_d \approx 24$ T obtained from these GL analyses may refer to a low field, unreconstructed Fermi
order that sets in above 15 T [12]. The change required would be 1.36 A/m/T in $\chi_d(0)$. This is consistent with the significant decrease in diamagnetism between 40 and 50 K shown in the inset of Fig. 3(a), the region where the 3D CDW seen at high fields goes away [12].

The above discussion highlights the importance of competing CDW and superconductivity instabilities [11,33]. Little is known about the size of the CDW energy gap, or the MF behavior expected for a $d$-wave superconductor just below $H_{c2}$ as $T \to 0$ K. Therefore the linear $H$ dependence of $dM/dB$ we observe below $H_g$ might be a fundamental property of a $d$-wave superconductor. In other words, because of Volovik-type pair breaking effects, the MF transition at $H_{c2}$ could have a discontinuity in $d^2M/dB^2$, rather than in $dM/dB$, which is the standard MF result for the second order transition in a conventional $s$-wave superconductor.

In summary, we observe diamagnetism in OVIII YBCO at fields up to 33 T and OII YBCO at fields up to 65 T using torque magnetometry. The analysis uses a different model for the high temperature normal state susceptibility that gives a smaller correction at low temperature compared with earlier models [6]. We also find that $dM/dB$ departs from a linear lower field behavior at fields $H_g \approx H_{c2}(0) \approx 24$ T, and approaches a constant value at higher fields. We propose two candidate scenarios: a competing order scenario where a fully fledged CDW at high field mostly suppresses the superconductivity so that the diamagnetism at high field could be attributed to bulk superconductivity; or a fluctuation picture in which for $H > H_g$, the system crosses over to superconducting fluctuation behavior. The diamagnetism at 65 T for the OII crystal could arise from the orbital susceptibility of carrier pockets and a reduction in spin susceptibility associated with the stronger 3D CDW order. It would be of interest to develop $d$-wave expressions for the MF magnetization and for the fluctuation contribution in the low temperature, high field regime, for comparison with our data. This could settle the debate over the existence of the high field vortex liquid region.

We thank G. Grissonnanche for useful discussions. This work was generously supported by NSERC under Grant No. RGPGP 170825-13 and No. RGPIN-2014-04554 and CIFAR of Canada, Canada Research Chair, EPSRC (UK) under Grant No. EP/K016709/1, Croatian Science Foundation project (No. 6216), and the Croatian Research Council, MZOS NEWFEL-PRO project No. 19. We thank HFML-RU, a member of the European Magnetic Field Laboratory. The work at LANL was funded by the Department of Energy Basic Energy Sciences program “Science at 100 T.” The NHMFL facility is funded by the Department of Energy, the State of Florida, and the National Science Foundation (NSF) Cooperative Agreement No. DMR-1157490.

[1] For ease of comparison with Refs. [7] and [6], we use the same units (Tesla) and notation (e.g., $H_{in}$ and $H_{c2}$) throughout this Rapid Communication.


