Lower Critical Field and Superfluid Density of Highly Underdoped $YBa_2Cu_3O_{6+x}$ Single Crystals

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The lower critical field H_{c1} for highly underdoped $YBa_2Cu_3O_{6+x}$ with T_c between 8.9 and 22 K has been determined by measurements of magnetization $M(H)$ curves with applied field parallel to the c axis. H_{c1} is linear in temperatures below about $0.6T_c$, and $H_{c1}(0)$ is proportional to $T_c^{1.64\pm0.04}$, clearly violating the proportionality between $\rho_s(0)$ and T_c. Moreover, the slope $-dH_{c1}/dT$ decreases steeply toward zero as T_c approaches zero, indicating that the effective charge of the quasiparticles vanishes as the doping is decreased toward the insulating phase.

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Cuprate superconductors have been the object of close scrutiny with many experimental probes, both long-established techniques and very new ones. One property that has been conspicuous by its absence has been measurements of the lower critical field H_{c1}, the field at which magnetic vortices first begin to enter a type-II superconductor. Reliable measurements of this property have been rare, for technical reasons that will be discussed below, and H_{c1} has never been the object of careful study in high quality crystals over a wide range of doping. In addition to the inherent value of measuring one of the key properties of a superconductor and the important place that the lower and upper critical fields hold in applications of superconductors, we will exploit H_{c1} here to give new access to the superfluid phase stiffness ρ_s at very low doping in $YBa_2Cu_3O_{6+x}$ (YBCO).

The temperature and the doping dependence of the superfluid phase stiffness ρ_s, usually determined from the in-plane London penetration depth λ_{ab} ($\rho_s \equiv 1/\lambda_{ab}{}^2$), provide many clues to the problem of high temperature superconductivity. The linear temperature dependence of ρ_s at low temperatures [1] is evidence for d-wave pairing where thermal excitation of quasiparticles near nodes in the superconducting energy gap cause ρ_s to decline linearly with temperature. $3DXY$ critical fluctuations in ρ_s near the critical temperature T_c highlight the important role that fluctuations also play in the temperature dependence of ρ_s. The most compelling feature is the typically low value of ρ_s in the underdoped regime and its tendency to rise in step with T_c as hole doping is increased [2]. This has led to the suggestion that, despite strong superconducting pairing, T_c is limited by phase fluctuations in underdoped cuprates [3–7]. Here we show that an unusual contribution from quasiparticle excitations also plays a deciding role in setting T_c.

Compared to the temperature dependence, the determination of the doping dependence of ρ_s is more difficult since it requires accurate measurements of the absolute value of λ_{ab} in different samples. Although λ_{ab} can be measured by several techniques, it has been difficult to arrive at quantitatively consistent results because of differences in samples (films, powders, and crystals) and technical drawbacks for each technique. Particularly, ρ_s in the highly underdoped region ($T_c < 0.5T_{c,\mathrm{max}}$ or $p < 0.08$) has been hardly investigated so far due to difficulties related to the properties of cuprates in this doping range. The coexistence of superconductivity and magnetism [8] renders problematic the muon spin relaxation (μSR), the technique widely used to measure λ_{ab} at higher doping. Furthermore, the large anisotropy makes λ_{ab} values obtained by techniques that measure the Meissner volume susceptible to contamination of λ_c.

An underexploited approach for evaluating ρ_s is via the lower critical field $H_{c1} = 4\pi E_1/\Phi_0 = 4\pi(E_{\mathrm{em}} + E_{\mathrm{core}})/\Phi_0$, where E_1, E_{em}, and E_{core} are the free energy of an isolated vortex, the electromagnetic energy of the supercurrent associated with the vortex, and the vortex core energy, respectively, each per unit length. For magnetic fields parallel to the c axis, $E_{\mathrm{em}} = (\Phi_0/4\pi\lambda_{ab})^2 \ln(\kappa)$, where $\kappa = \lambda_{ab}/\xi_{ab}$ and ξ_{ab} is the in-plane coherence length. According to Ginzburg-Laudau theory, the effect of the core energy is to add 0.5 to $\ln(\kappa)$, a small correction if κ is large. Therefore,

$$H_{c1} = \Phi_0[\ln(\kappa) + 0.5]/(4\pi\lambda_{ab}{}^2). \tag{1}$$

A great deal of experimental evidence indicates that κ is only weakly temperature and doping dependent [9–11]. For example, κ varies from 40 to 75 in YBCO only when T_c changes from 10 to 92 K [9], so the $\ln(\kappa)$ term is nearly a constant and H_{c1} is proportional to $1/\lambda_{ab}{}^2$.

H_{c1}, the onset of the mixed state, is conventionally determined by measuring $M(H)$ in increasing magnetic field and looking for the field of first vortex entry, characterized by the departure of the flux density $B = H + 4\pi M$

from zero. In high T_c superconductors, however, there always exists strong bulk pinning, and the entry of vortex lines is very gradual. According to the Bean critical state model [12] for type-II superconductors [13,14],

$$B = A(H - H_{c1})^2/H^* \qquad (H_{c1} \leq H \ll H^*), \quad (2)$$

where A is a constant related to sample shape and H^* is proportional to critical current density. A plot of $B^{1/2}$ vs H should yield a straight line with a threshold at H_{c1}.

An accurate determination of H_{c1} requires the use of an ellipsoid with a well-defined demagnetization factor, since the effective magnetic field on a nonellipsoidal sample is inhomogeneous. In particular, extremely high effective fields at sharp corners and edges lead to vortex entry at fields far below H_{c1}. Further complicating the issue is the Bean-Levingston (BL) surface barrier [15] that, for perfect surfaces, prevents vortex entry until the field is far above H_{c1}. Although surface roughness on the scale of λ reduces the surface barrier very effectively, measurements should be carried out reversibly by both increasing and decreasing the field to ensure the observed H_{c1} values are not artificially altered by the surface barrier. Thus far, very little effort [16] has been made to prepare ellipsoidal samples, and most reported cuprate H_{c1} data were measured using rectangular platelets and only in increasing magnetic field. The combination of nonellipsoidal samples and the BL surface barrier is likely the cause for the large discrepancies among published H_{c1} data. For example, the reported $H_{c1}(0)$ values for fields parallel to the c axis range from 180 to 8000 Oe [16,17] in optimally doped YBCO.

Here we use high purity (99.995%) YBCO crystals grown in barium zirconate crucibles [18]. A crystal with the shortest dimension 0.4 mm in the c direction was polished with 1 μm grit into a nearly ellipsoidal shape with demagnetization factor $n = 0.363$ in the c direction. The ellipsoid was set to an oxygen content $6 + x \cong 6.35$ and quenched in a sealed quartz capsule from 570 °C to an ice-water bath [19]. As quenched, the crystal had $T_c =$ 8.9 K. At this oxygen content, T_c depends on the degree of chain oxygen ordering. By leaving it at room temperature for a total of 3 weeks, allowing chain oxygen ordering to develop, T_c increased to 18 K. More annealing at room temperature under 3000 bar hydrostatic pressure increased T_c to 22 K [20]. T_c can be frozen by cooling the crystal below 0 °C, allowing us to determine H_{c1} for values of T_c between 8.9 and 22 K. This T_c range corresponds to hole doping p between 0.055 and 0.064 by the empirical relationship [21] between T_c and p.

Magnetization was measured using a quantum design SQUID magnetometer with applied field H parallel to the c direction. An optimally doped YBCO ellipsoid was placed 3 cm below the sample, such that the SQUID output contained signals from both. By fitting the SQUID output to two separated moments, the magnetizations of both crystals were obtained simultaneously. Since $H_{c1}/(1 - n)$ for the optimally doped ellipsoid is about 5 times the highest field used in this work (120 Oe), it was always in the Meissner state and its magnetization $4\pi M = -H$ served as an *in situ* calibration of the magnetometer, resulting in a fourfold improvement in precision.

The midpoint of the superconducting transition, as determined by measuring the field-cooled magnetization at 1, 2, and 3 Oe and extrapolating the data to zero field, was taken as the critical temperature T_c. The transition width ΔT_c (10%–90%) was about 1 K, independent of T_c.

After cooling the ellipsoidal crystal in zero field from above T_c to a chosen temperature, $M(H)$ was measured as the field H was first increased to several times H_{c1} and then decreased to zero. Figure 1(a) shows the plot of $B^{1/2} = (4\pi\delta M)^{1/2}$, where δM is the deviation from perfect shielding, against the effective field H_{eff}, for data taken at $T =$ 2 K when T_c was 8.9 . The data are well described by the Bean critical state model [Eq. (2)] except for a slight rounding near H_{c1}, which is expected from the superconducting transition width $\Delta T_c \cong 1$ K.

An alternative way to determine H_{c1} from $M(H)$ data is to take the derivative dM/dH. As shown in Fig. 1(b), dM/dH exhibits a kink at H_{c1} in both increasing and decreasing magnetic field data. The kink for increasing field is expected from Eq. (2). In decreasing applied field, the effective field H_{eff} on a sample is generally inhomogeneous due to inhomogeneous magnetization [12]. However, if vortex entry is limited to the sample surface

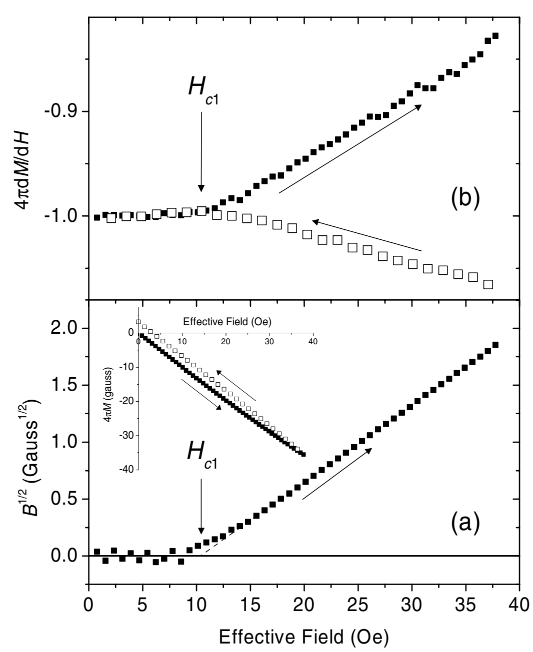

FIG. 1. Magnetization data at $T = 2$ K for $T_c = 8.9$ K. The arrows beside the data points indicate the order in which the data were taken. (a) Plot of $B^{1/2}$ vs magnetic field, showing the data well described by the Bean critical state model; inset, raw $M(H)$ data. (b) dM/dH for both increasing and decreasing field.

region ($H_{\max} \ll H^*$), as is the case in this work, the inhomogeneity of H_{eff} is small and H_{eff} is still approximately $H/(1-n)$, thus a kink in dM/dH can still be observed at H_{c1}. We emphasize that values of H_{c1} in this work were determined in both increasing and decreasing magnetic field, so they are the true thermodynamic values and are not altered by the surface barrier.

The H_{c1} data in Fig. 2 show that, below $0.6T_c$, H_{c1} is linear in temperature, indicating line nodes in the superconducting energy gap, consistent with d-wave pairing. For comparison with other techniques of measuring λ_{ab}, a separate, ortho-II phase ($x = 0.50$, $T_c = 56$ K) ellipsoid was also measured. The result $H_{c1}(0) = 238 \pm 15$ Oe yields $\lambda_{ab}(0) = 175 \pm 6$ nm by using $\kappa = 50$ [9], in excellent agreement with the values 168 ± 25 nm obtained by zero-field ESR [22] and 170 nm obtained by μSR measurements [23]. These independent measurements on similar crystals confirm that H_{c1} provides an excellent quantitative measure of $1/\lambda_{ab}{}^2$ or the phase stiffness.

In Fig. 3, $H_{c1}(0)$ and the slope $-dH_{c1}/dT$, obtained by linear fits to the data below $0.5T_c$, are plotted as functions of T_c, highlighting two key features of the data. $H_{c1}(0)$ is clearly a nonlinear function of T_c, instead following the power law $H_{c1}(0) = 0.366T_c^{1.64\pm0.04}$ (Oe). To the extent that $\ln(\kappa)$ is a constant, this implies that the relationship between critical temperature and phase stiffness is $T_c \propto \rho_s(0)^{0.61}$, clearly inconsistent with $T_c \propto \rho_s(0)$. It is worth pointing out that the reported proportionality $T_c \propto \rho_s(0)$ for underdoped cuprates [2] is a rough approximation over a wide doping range. It revealed the overall increase of T_c as $\rho_s(0)$ increases. Tallon *et al.* [24] pointed out a sublinear T_c dependence on $\rho_s(0)$ for doping p between 0.08 and 0.19. Bernhard *et al.* [25] reported that the dependence of T_c on $\rho_s(0)$ changes at 1/8 doping. The present data are for p between 0.055 and 0.064, the crucial regime where T_c is falling rapidly toward zero and giving way to magnetism.

Since $T_c \propto \rho_s(0)$ is the expected behavior if T_c is governed solely by phase fluctuations in two dimensions, the present result indicates that other factors play a key role. One candidate is the thermal excitation of nodal quasiparticles [26–28]. In this scenario, the already small phase stiffness at $T = 0$ is further depleted by the linear temperature dependence coming from excitations near the d-wave nodes, driving the phase stiffness towards zero. The nodal quasiparticles' effectiveness at depleting phase stiffness is measured by the slope $-dH_{c1}/dT \propto -d\rho_s/dT$, also plotted in Fig. 3. This slope drops steeply with decreasing doping, falling 45% from $T_c = 22$ K to $T_c = 8.9$ K and appearing to trend toward zero as T_c approaches zero. In order to further investigate $-dH_{c1}/dT$, the reduced critical field $h = H_{c1}/H_{c1}(0)$ is plotted against reduced temperature $t = T/T_c$ in Fig. 4. The scaling behavior for the very underdoped sample implies that, for $T_c \leq 22$ K and $T < 0.6T_c$, $dh/dt = (dH_{c1}/dT)[T_c/H_{c1}(0)]$ is independent of T_c. Thus $-dH_{c1}/dT \propto H_{c1}(0)/T_c \propto T_c^{0.64}$, again suggesting $-dH_{c1}/dT$ vanishes as T_c approaches zero.

In d-wave superconductors, the slope of the linear temperature dependence of the phase stiffness $-d\rho_s/dT$ is proportional to $\alpha^2 \nu_F/\nu_\Delta$, where ν_F is the Fermi velocity, ν_Δ is a measure of how steeply the superconducting gap opens up away from the nodes, and α is a renormalization of the effective charge in the quasiparticle current backflow that depletes the superfluid screening. Angle-resolved photoemission shows that ν_F is largely doping independent [29] and measurements of the low temperature limit of the thermal conductivity indicate that ν_F/ν_Δ changes rather little in YBCO as T_c approaches zero [30]. Thus the

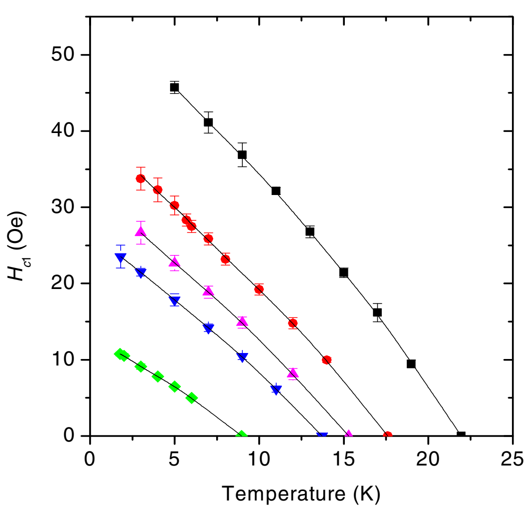

FIG. 2 (color online). H_{c1} data as a function of temperature for a YBCO ellipsoid with doping tuned by oxygen ordering. The lines are guides to the eye.

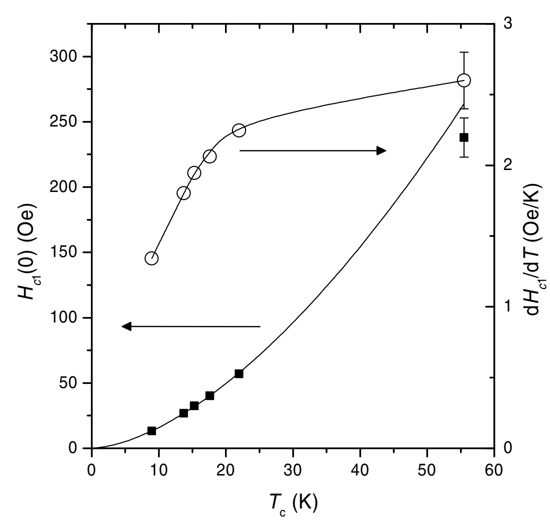

FIG. 3. $H_{c1}(0)$ (solid squares, left scale) and $-dH_{c1}/dT$ (open circles, right scale) as functions of T_c. The curve through the solid squares is the power law fit to the data of $T_c \leq 22$ K, $H_{c1}(0) = 0.366T_c^{1.64}$ (Oe).

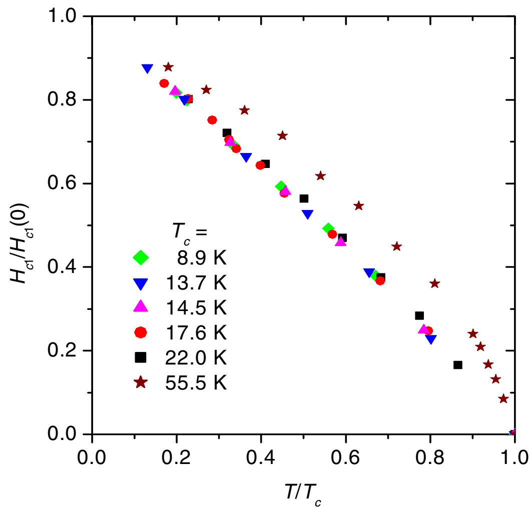

FIG. 4 (color online). Plot of $H_{c1}/H_{c1}(0)$ against reduced temperature T/T_c. All data of $T_c \leq 22$ K fall into a single curve.

decline of the slope $-dH_{c1}/dT$ indicates a decrease in α^2. Within the framework of Fermi liquid theory [31], the current renormalization is a result of the interaction between quasiparticles, which causes screening of charge and reduces the current carried by a quasiparticle from ev_F to $\alpha e v_F$. This renormalization is also expected to occur in non-Fermi liquid theories such as gauge theories based on Anderso's resonating valence bond model [32–35], where the charge of electronlike quasiparticles shrinks to zero upon underdoping. In spite of various theoretical predictions, however, there has been little experimental evidence for the quasiparticle charge renormalization in cuprates. Our data here suggest that in the very underdoped region the quasiparticle effective charge is strongly renormalized and finally vanishes as the doping is decreased towards the insulating phase.

In summary, the lower critical field H_{c1} for highly underdoped YBCO has been determined for fields parallel to the c axis, without uncertainties related to nonellipsoidal samples and the BL surface barrier. If the data are analyzed using the assumption that κ is only weakly doping and temperature dependent, H_{c1} is equivalent to the phase stiffness ρ_s. The data then show a power law relation $T_c \propto \rho_s(0)^{0.61}$, differing markedly from the linear relationship expected for a T_c governed by phase fluctuations in two dimensions. This is remarkable given the high anisotropy of this material ($\lambda_c/\lambda_{ab} \sim 100$) and the importance that phase fluctuations ought to have at such low values of phase stiffness. The low phase stiffness is further depleted by quasiparticle excitations, though this depletion weakens at very low doping in a manner suggesting the charge renormalization of the nodal quasiparticles, which reduces the effective charge to zero as the insulating part of the phase diagram is approached.

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