Lower critical field and superfluid density in highly underdoped YBa$_2$Cu$_3$O$_{6+x}$ single crystals

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The lower critical field $H_{c1}$ for highly underdoped YBa$_2$Cu$_3$O$_{6+x}$ 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 temperature below about 0.6$T_c$, and $H_{c1}(0)$ is proportional to $T_c^{1.64 \pm 0.04}$, 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.

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

The lower critical field $H_{c1}$ is proportional to $\rho_s(0)\ln(\kappa)+0.5)/(4\pi\lambda_{ab}^2)$. (1)

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$\rho_s$ is via the lower critical field $H_{c1}=4\pi E_{em}/\Phi_0 = 4\pi(E_{em}+E_{core})/\Phi_0$, 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, $E_{em}=(\Phi_0/4\pi\lambda_{ab}^2)^2\ln(\kappa)$, where $\kappa = \lambda_{ab}/\xi_{ab}$ and $\xi_{ab}$ is the in-plane coherence length. According to Ginzburg-Landau (GL) theory, the effect of the core energy is to add 0.5 to $\ln(\kappa)$, a small correction if $\kappa$ is large. Therefore,

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$H_{c1}$, 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\textsuperscript{12} for type-II superconductors,\textsuperscript{15,14}

$B = A(H-H_{c1})^2/H^2$ \quad \text{if} \quad H_{c1} \leq H << H^∗, \quad \text{(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 $H_{c1}$. In YBCO, the onset of flux entry is not at $H_{c1}$ but rather at about $H_{c2}$, which is typically only a few $\%$ less than $H^∗$. The mixed state is therefore characterized by a regime of strong flux flow at $H_{c1} > H > H_{c2} > H^∗$.
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 non-ellipsoidal 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 \( \lambda \) 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 non-ellipsoidal 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 \( \mu \)m 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+ for perfect surfaces, prevents vortex entry until the field is far above \( H_{c1} \). Although surface roughness on the scale of \( \lambda \) 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 non-ellipsoidal 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.

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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 $\lambda_{ab}$, a separate, ortho-II phase ($\chi = 0.53$, $T_c = 56$ K) ellipsoid was also measured. The result $H_{c1}(0) = 238\pm15$ Oe yields $\lambda_{ab}(0) = 175\pm6$ nm by using $\kappa = 50^3$, in excellent agreement with the values $168\pm25$ 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 $H_{c1}(0) = 0.366T_c^{1.640.04}$ (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 work studies doping range between 0.055 and 0.064, the crucial regime where $T_c$ is falling rapidly toward zero and giving way to magnetism.
behaviour for the very underdoped sample implies that, for $T_c \leq 22$ K and $T < 0.67T_c$, $dH_\alpha/dT = (dH_\alpha/dT)(T_c)$ is independent of $T_c$. Thus $-dH_\alpha/dT \propto H_\alpha(0)/T_c \propto T_c^{-0.64}$, again suggesting $-dH_\alpha/dT$ 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 $\alpha^2\nu_F/\nu_s$, where $\nu_F$ is the Fermi velocity, $\nu_s$ is a measure of how steeply the superconducting gap opens up away from the nodes, and $\alpha$ is a renormalization of the effective charge in the quasiparticle current backflow that depletes the superfluid screening. Angle-resolved photoemission shows that $\nu_F$ is largely doping-independent and measurements of the low temperature limit of the thermal conductivity indicate that $\nu_F/\nu_s$ changes rather little in YBCO as $T_c$ approaches zero. Thus the decline of the slope $-dH_\alpha/dT$ indicates a decrease in $\alpha^2$. Within the framework of Fermi liquid theory, 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 $e\nu_F$ to $\alpha e\nu_F$. This renormalization is also expected to occur in non-Fermi liquid theories such as gauge theories, where the charge of electron-like 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 $H_{cl}$ 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 analysed using the assumption that $\alpha$ is only weakly doping and temperature dependent, $H_{cl}$ is equivalent to the phase stiffness $\rho_s$. 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 2D. 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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30. Mike Sutherland et al., Unpublished.