# Resistive Upper Critical Field of High-$T_c$ Single Crystals of $Bi_2Sr_2CaCu_2O_8$

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The upper critical field $H_{c2}(T)$ of $Bi_2Sr_2CaCu_2O_8$ crystals with $T_c > 92$ K has been measured from the out-of-plane resistivity in magnetic fields ($\mathbf{H} \perp ab$) up to 15 T. By the use of the empirical procedure the $H_{c2}(T)$ curve has been extrapolated up to $H_{c2} \simeq 230$ T and $T/T_c \simeq 0.35$ which is independent of the choice of the $R/R_N$ ratio. We found that $H_{c2}(T)$ does not follow the conventional theory with or without fluctuations but is consistent with the prediction based on the Bose-Einstein condensation of charged bosons formed above $T_c$. Our results together with the heat capacity measurements provide an evidence for the possibility of $2e$ Bose liquid ground state of high-$T_c$ oxides.

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One of the striking superconducting properties of high-$T_c$ oxides is their unusually high values of the upper critical field and its temperature dependence. In many superconducting oxides, including the cubic ones, an upward curvature of $H_{c2}(T)$ near $T_c$ has been observed [1,2]. For example, measurements for overdoped Tl-Ba-Cu-O [3] and Bi-Sr-Cu-O [4] with low $T_c \sim 20$ K have revealed a startling increase of $H_{c2}(T)$ down to millikelvin temperatures. Alexandrov [5] has proposed an explanation for this phenomenon based on the Bose-Einstein condensation (BEC) of charged bosons. However, in the highest $T_c$ cuprates, the in-plane superconducting transition is known to display pronounced broadening in a magnetic field, with the top of the transition having a much weaker field dependence than in the region near the bottom [6–8]. This together with the high values of $H_{c2}$ have made an experimental determination of $H_{c2}$ very difficult in materials with $T_c > 60$ K, with the consequence that widely varying values of $H_{c2}(0)$ have been estimated based on different models. The out-of-plane resistive transition is known to show a different behavior in a magnetic field. An increasingly pronounced maximum (peak) developed below $T_c$ and shifted to lower temperature rather than broadened with increasing field [9]. By modeling the $c$-axis conduction with a stack of Josephson junctions in series, Gray and Kim [10] have obtained an unusually large energy gap with large fluctuations. Consequently, no attempts have been made so far to determine $H_{c2}(T)$ from these observations.

We report in this Letter the out-of-plane resistivity of $Bi_2Sr_2CaCu_2O_8$ (BSCCO-2212) crystals measured in perpendicular fields up to 15 T ($\mathbf{H} \parallel I \parallel c$). We propose a procedure for extrapolating the values of the resistive upper critical field $H_{c2}(T)$ which is independent of the background normal resistance and the choice of $R/R_N$ values over a wide temperature range. In contrast to the predictions of conventional theory, this $H_{c2}(T)$ shows a negative curvature just below $T_c$ followed by a positive curvature at lower temperatures with no sign of saturation down to $0.35T_c$. We have also determined the irreversibility field $H_{\rm irr}(T)$ ($\parallel c$). This has a qualitatively different temperature dependence. We propose a quantitative explanation of $H_{c2}(T)$ based on the theory of BEC in the magnetic field [5].

BSCCO-2212 single crystals have been grown by the self-flux method during solid-state reaction and have $\sim(7-10)\%$ Y substitution for Ca [11]. For the present measurements, three homogeneous single crystals have been selected with $T_{c0}$ between 92 and 95 K and in-plane dimensions $870 \times 270$, $340 \times 150$, and $800 \times 350$ $\mu$m$^2$.

Figure 1(A) shows the typical out-of-plane resistive transitions in magnetic fields up to 15 T ($H \parallel I \parallel c$). In agreement with Refs. [9,12] no significant $c$-axis magnetoresistance at temperatures well above the peak is observed. We have analyzed our data based on three assumptions. The first is that the normal state resistance $R_N(T)$ is independent of the field and can be represented by a single function obtained by extrapolating the $R(T)$ curves from temperatures well above the transition [see Fig. 1(A)]. After dividing the $R(T)$ curves taken at different fields by $R_N(T)$, the resultant curves [Fig. 1(B)] show a nearly parallel shift of the transition, particularly at high fields. We believe these "normalized" resistance curves provide a good insight to the effects of field on the superconducting properties. Not surprisingly, a plot of the fields $H^*(T)$ [13] as a function of reduced temperature $\tau = 1 - T/T^*$ for different $R/R_N$ values will give rise to a series of similar curves as shown in Fig. 2 where $T^*$ is defined as the temperature of the corresponding $R/R_N$ value in zero field.

It is apparent that $H^*(\tau)$ obtained for different $R/R_N$ values has a similar shape which is made up of two regions: up to $\sim 6K$ from $T^*$ all $H^*(\tau)$ obey a simple relationship $H^*(\tau) \sim \tau^\nu$, where $\nu$ is found to be about 0.6. This unusual relationship is not consistent with

  

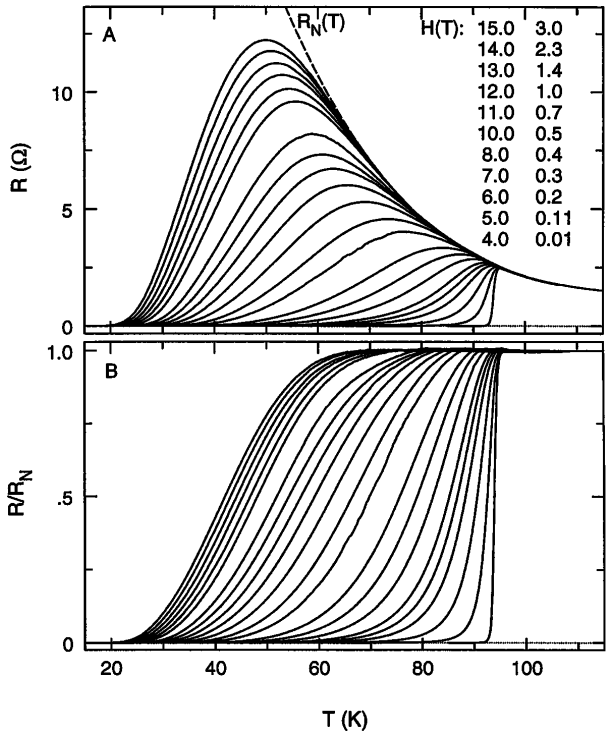


FIG. 1. (A) $c$-axis resistive transition in BCCSO-2212 crystal in a magnetic field up to 15 T. $R_N(T)$ is shown by dotted line. (B) (A) normalized by $R_N(T)$.

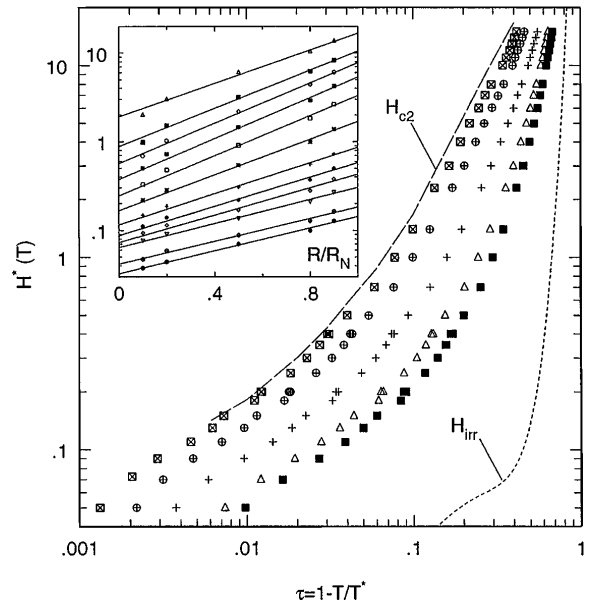


FIG. 2. $H^*(\tau)$ determined at the level of $R/R_N = 0.1, 0.2, 0.5, 0.8, 0.9$ (from the bottom). The same level is applied to determine $T^*$ from the zero field transition. Long dashed line represents the asymptotic behavior of $H^*$ as $R \to R_N$; the irreversibility field determined as $H^*$ at $R/R_N = 10^{-4}$ is shown by the short dashed line. Inset: scaling of $H^*$ with the ratio $R/R_N$ at fixed temperature.

conventional theory and we propose an explanation below. However, the value of $\nu$ was rather sample dependent and our measurements gave an accuracy of $\sim$20%. On the other hand, for $\tau > 0.07$, $H^*(\tau)$ takes on an upward curvature with $\nu > 1$. In this region, data taken from different crystals agreed with one another to better than 15%.

One of our objectives is to determine $H_{c2}$. The behavior of $H^*$ at a given $\tau$ in Fig. 2 for different $R/R_N$ values leads us to our second assumption, namely,

$$H^*(\tau) \propto \exp[s(\tau)R/R_N]. \quad (1)$$

This empirical expression for $H^*$ holds well for $R/R_N = 0.1-0.9$ and $s(\tau)$ is of order 2 as shown by solid lines in the inset of Fig. 2. The variation in $s(\tau)$ (slopes of the lines) reflects the small changes in the shape of $R/R_N$ vs $T$ curves in Fig. 1(B). Our third assumption is to use Eq. (1) and estimate $H_{c2}(\tau)$ as an asymptotic of $H^*(\tau)$ when $R \to R_N$, which are the intercepts on the field axis on the right side of the inset to Fig. 2. The extrapolated $H_{c2}(T)$ is shown in Fig. 2 by a dashed curve representing the upper limit of $H^*(T)$ and in Fig. 3 for two extremes of $s(\tau)$. This process has the advantage of removing the uncertainty in determining $H_{c2}$ due to fluctuations as $R(T)$ approaches $R_N(T)$ near the top of the transition and provides us with good insight into the experimental uncertainty in $H_{c2}$. The empirical expression, Eq. (1), describes the dependence of the resistivity on the magnetic field at a fixed temperature in the transition region, where the resistivity is controlled by the flux motion. It also gives the value of the magnetic field $H_p \sim H_{\rm irr} < H_{c2}$ at which vortices are pinned ($R = 0$). We have estimated the irreversibility curve $H_{\rm irr}(T)$ by taking the $H^*$ values at arbitrary small $R/R_N$ of $10^{-4}$.

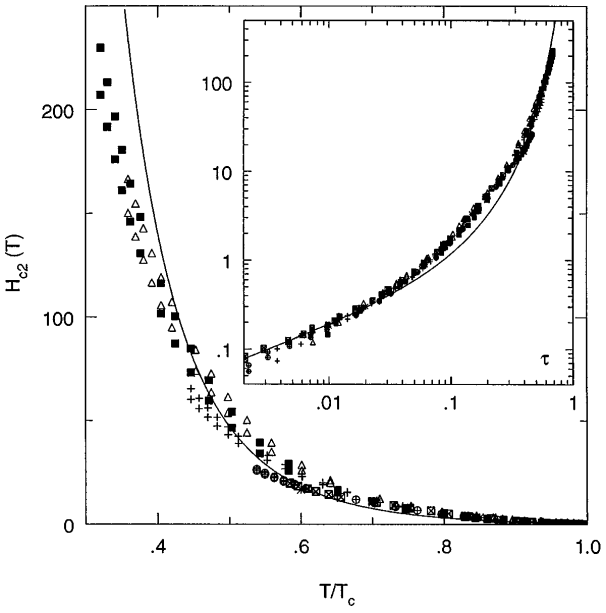


FIG. 3. $H_{c2}(T)$ as obtained by scaling the data of Fig. 2 by use of Eq. (1) to the limit $R \to R_N$. Theoretical fit with Eqs. (9) and (12) and $H_0 = 0.25$ T is shown by the solid line. Inset presents the same $H_{c2}(\tau)$ in log-log plot.

This is shown in Fig. 2. It is clear that $H_{c2}(T)$ is both quantitatively as well as qualitatively different from $H_{\rm irr}(T)$. In a similar study, Hellerqvist *et al.* [14] has described the whole process in terms of macroscopic correlation of vortices in the $c$ direction and we hope to provide evidence through the present investigation that $H_{c2}$ is independent of the vortex dynamics consideration.

On the other hand, our measured $H_{c2}(T)$ does not follow the conventional model, which predicts a zero or positive curvature very close to $T_c$ and negative at lower temperatures. We observe just the opposite behavior, which we believe is related to a very small coherence volume in high-$T_c$ oxides. The coherence volume estimated from the heat capacity measurements near $T_c$ is comparable with or even less than the unit cell volume [15]. This favors a charged $2e$ Bose liquid as a plausible microscopic model of the ground state [16]. Here we extend the theory of $H_{c2}(T)$ of the charged Bose liquid [5] to the region $\omega_c\tau < 1$, where $\omega_c$ is the cyclotron frequency and $\tau$ a scattering time. We expect that this regime is realized in our samples near $T_c$. $H_{c2}(T)$ is determined as the field in which a nonzero solution of the linearized stationary equation for the macroscopic condensate wave function $\psi_0(\mathbf{r})$ first appears [5]

$$\Big[-\frac{1}{2m_{ij}}[\nabla - 2ie\mathbf{A}(\mathbf{r})]_i[\nabla - 2ie\mathbf{A}(\mathbf{r})]_j + U_{\rm sc}(\mathbf{r}) - \mu\Big]\psi_0(\mathbf{r}) = 0\,, \qquad (2)$$

where $\mathbf{A}(\mathbf{r})$, $U_{\rm sc}(\mathbf{r})$, and $\mu$ are the vector, scattering, and chemical potentials, respectively, and $m_{ij}^{-1}$ is the inverse mass tensor ($\hbar = c = 1$). The definition of $H_{c2}$ is identical to that of the upper critical field of the BCS superconductor. The difference is due to the chemical potential. In the Bose liquid $\mu$ is determined by conservation of the number of bosons $n$ in

$$\int f_B N(\epsilon, H) d\epsilon = n\,, \qquad (3)$$

where $N(\epsilon, H)$ is the density of states (DOS) of the Hamiltonian Eq. (2) and $f_B = 1/\{\exp[(\epsilon - \mu)/T] - 1\}$ is the Bose-Einstein probability function. A first nontrivial extended solution to Eq. (2) appears at $H = H_{c2}$ when $\mu$ coincides with the energy of the lowest extended state $\mu = E_c$. The DOS is determined by the imaginary part of Green's function $G_\alpha(\epsilon) = [\epsilon_\alpha - \epsilon - \Sigma_\alpha(\epsilon)]^{-1}$. In the "noncrossing" approximation for any elastic scattering the self-energy is given by

$$\Sigma_\alpha(\epsilon) = \sum_\beta \frac{|M_{\alpha,\beta}|^2}{\epsilon_\beta - \epsilon - \Sigma_\beta(\epsilon)}\,. \qquad (4)$$

Here $|M_{\alpha,\beta}|^2$ is the matrix element square of the scattering potential for states $\alpha, \beta$ of a free particle in a magnetic field. As it does not depend on the quantum numbers $\alpha, \beta$ for a short-range impurity scattering, $|M_{\alpha,\beta}|^2 = M^2 = \text{const}$. The same is almost true for the acoustic phonon scattering if the sound velocity is small compared with the thermal velocity of carriers resulting in elastic scattering. However, in this case the scattering rate is proportional to temperature and then $M^2 \sim T$. Introducing the DOS of a noninteracting system for $\mathbf{H} \parallel c$

$$N_0(\epsilon, H) = \frac{\sqrt{2eHm_c}}{4\pi^2}\,\mathrm{Re}\sum_{k=0}^{\infty}\frac{1}{\sqrt{\epsilon - \omega_c(k + 1/2)}}\,, \qquad (5)$$

one can write

$$\Sigma(\epsilon) = M^2\int d\epsilon'\,\frac{N_0(\epsilon', H)}{\epsilon' - \epsilon - \Sigma(\epsilon)}\,, \qquad (6)$$

where $\omega_c = 2eH/m$ and $m$, $m_c$ are the in-plane and out-of-plane effective mass, respectively. The integral in Eq. (6) yields in dimensionless form

$$\sigma(x) = \frac{1}{\sqrt{h}}\sum_{k=0}^{k_{\rm max}\gg 1}[k - x - \sigma(x)]^{-1/2}, \qquad (7)$$

where $\sigma(x) = \Sigma(\epsilon)/\omega_c$, $x = \epsilon/\omega_c - 1/2$ and $h = H/H_0$ with $H_0 = m_c m^3 M^4/16\pi^2 e$. The cutoff parameter $k_{\rm max}$ eliminates the divergent term, appearing as a result of the approximation for the matrix element in Eq. (4). It drops out of the final results. Near the edge of the continuum spectrum $x_c \equiv E_c/\omega_c$ one can expand $\sigma(x)$ in powers of $x - x_c$ to obtain

$$\sigma(x) \simeq -\delta - x + i\left[\frac{8h^{1/2}(x - x_c)}{3\sum_{k=0}^{\infty}(k + \delta)^{-5/2}}\right]^{1/2} \qquad (8)$$

for $x - x_c < h$. Here $\delta = -x_c - \sigma(x_c)$ is determined by the condition that $x_c$ has a minimum as a function of $\delta$. The use of Eq. (7) yields the following equation for $\delta$:

$$\sum_{k=0}^{\infty}(k + \delta)^{-3/2} = 2h^{1/2}. \qquad (9)$$

Near the edge one obtains

$$N(\epsilon, H) = \frac{\omega_c \mathcal{T}\sigma(x)}{\pi M^2} = \frac{h^{5/4}}{\pi^2}\left[\frac{2eH_0 m m_c(x - x_c)}{3\sum_{k=0}^{\infty}(k + \delta)^{-5/2}}\right]^{1/2}. \qquad (10)$$

In the classical region far away from the edge ($x - x_c \gg h$) one can apply Poisson's formula to the sum in Eq. (7) with the following result for the DOS:

$$N(\epsilon, H) = \frac{1}{\pi^2}[ehH_0 m m_c (x - x_c)]^{1/2}. \quad (11)$$

Replacing the sums by integrals in Eqs. (9) and (10), we estimate $\delta \simeq 1/h$. Then Eq. (10) also describes the classical limit, Eq. (11). Therefore one can apply Eq. (10) to the whole integration to obtain the sum rule, Eq. (3). Substituting Eq. (10) into the sum rule, Eq. (3), with $\mu = \omega_c x_c + 1/2$ we find for the critical curve $h(t)$

$$h = \left[ \frac{3}{2t^3} \sum_{k=0}^{\infty} (k + \delta)^{-5/2} \right]^{2/3}, \quad (12)$$

where $t \equiv T/T_c$. The numerical solution of this equation together with Eq. (9) is shown in Fig. 3. The inequality $\delta \gg 1$ is satisfied very close to $T_c$. Then one can apply Poisson's formula to the sums in Eqs. (9) and (12). This gives

$$h \sim \tau^{0.5}, \quad (13)$$

which is in agreement with our observation, Fig. 2. Farther below $T_c$ the theory predicts an upward curvature with $h \sim T^{-4.5}$ for $T \to 0$.

There is therefore a remarkable agreement with experiment in the whole temperature region studied. It should be emphasized that we have not used any fitting parameters, as shown in Fig. 3. At low temperatures the Anderson localization of bosons can be important [5]. Acoustic phonon scattering contributes also making the scaling parameter $H_0$ temperature dependent. This temperature dependence has little effect on $H_{c2}(T)$ near $T_c$. However, taking into account both the impurity and acoustic phonon scattering and the Anderson localization one can improve the fit at low temperatures. Further comparison between theory and experiment now requires more data to be obtained in the lower temperature region.

As we have mentioned above the conventional theory of $H_{c2}(T)$ clearly contradicts our observations. However, Ovchinnikov and Kresin [17] have recently suggested an explanation of the unusual shape of $H_{c2}(T)$ of overdoped low-temperature oxides in terms of the conventional theory with a strong spin-flip scattering. They argue that unusual features of overdoped high-$T_c$ oxides can be understood as a result of a strong magnetic pair-breaking if the spin-flip mean free path $l_s$ is shorter than the coherence length $\xi_0$, which is less than the mean free path $l$ in our samples. The inequality $l_s < \xi_0 < l$ seems difficult to realize in our samples because of a very short coherence length and the absence of any significant Curie-like contribution to the normal state magnetic susceptibility. Moreover, the observed temperature dependence of $H_{c2}(T)$ in the close vicinity of $T_c$ does not follow the prediction of the mean-field theory with or without magnetic scattering. Of course, the determination of $H_{c2}(T)$ from the resistivity should be supplemented by independent measurements. From our experience and that of others we know that the magnetization measurements reveal the unusual shape of the critical magnetic field in the copper-based oxides. However, at present it is difficult to say how close the magnetically determined critical field is to the real $H_{c2}$. The specific heat measurements in a magnetic field [18] reveal the critical behavior similar to that of $^4$He in agreement with our approach.

In conclusion, the upper critical field has been determined from the $c$-axis resistivity of high-$T_c$ BSCCO-2212 single crystals. It has an unusual temperature dependence which cannot be explained by conventional theory. The critical exponent $\nu$ is less than unity just below $T_c$ while $\nu > 1$ farther away. An explanation is proposed based on the Bose-Einstein condensation of charged bosons formed above $T_c$.

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