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DETERMINATION OF SUPERCONDUCTING AND NORMAL STATE PARAMETERS OF SINGLE CRYSTAL K_3C_{60}

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We report magnetroresistance measurements on superconducting single crystals of K_3C_{60}. An analysis of the experimental data yields superconducting state and normal state parameters, including the upper critical field $H_{c2}(T)$, coherence length ξ, penetration depth λ, scattering time τ, mean free path ℓ, and zero temperature resistivity $\rho(0)$. Some of the intrinsic single crystal parameters are significinatly different from those extracted from thin film and powder specimens.

The surprisingly high transition temperatures of recently discovered fulleride superconductors [1] have generated tremendous theoretical and experimental interest. Various models of superconductivity have been proposed including novel electronic mechanisms[2] and conventional phonon-mediated mechanisms with emphasis on either intramolecular[3-5] or intermolecular[6] modes. In order to evaluate different theoretical models, it is desirable to establish intrinsic parameter values for both the normal and superconducting states of the material. Although a number of experiments have been performed on granular thin films and pressed powder of the doped fullerenes, very few measurements exist for single crystal specimens.

In this communication, we report on magnetoresistance experiments on superconducting single crystal K_3C_{60} in fields up to 7.3 T. Our measurements yield directly the upper critical field $H_{c2}(T)$ between T_c = 19.7 K and 13 K. Further analysis allows us to determine intrinsic superconducting state parameters including $H_{c2}(0)$, the coherence length ξ and penetration depth λ, and parameters of the normal state including the scattering time τ, mean free path ℓ, and zero-temperature resistivity $\rho(0)$. These parameters provide a self-consistent picture of the superconducting state within Eliashberg theory.

K_3C_{60} single crystals were prepared using a sublimation growth/intercalation doping method described in detail elsewhere.[7] X-ray diffraction of the crystals prior to doping confirmed the well-known fcc crystal structure. Electrical measurements were made using van der Pauw and in-line four-probe contact geometries employing gold leads silver painted to evaporated silver pads. The sample temperature was monitored using a carbon-glass resistance thermometer.

Figure 1 shows the temperature dependence of the resistivity of a K_3C_{60} single crystal near the superconducting transition temperature T_c for different applied magnetic fields. The inset shows the zero field resistivity over an extended temperature range. Above $T_c(H = 0)$ = 19.7 K (transition midpoint), the resistivity displays a metallic temperature dependence. The transition to the superconducting state is sharp, $\Delta T \approx 250$ mK for zero field. For each magnetic field, the field-cooled data were taken with the same cooling rate of about 2 mK/s. The applied dc current was kept small to insure negligible joule heating effects. Although the transition width broadens slightly under high magnetic field, no evidence for excessive broadening near T_c is observed in contrast to the behavior of high-T_c copper oxide superconductors.[8] The sharpness of the transition in K_3C_{60}, even in high magnetic fields, allows a clear identification of $H_{c2}(T)$.

Figure 2 shows the temperature dependence of H_{c2} where we have used the resistivity drop midpoints to define the T-H critical line. A linear fit to $H_{c2}(T)$ yields dH_{c2}/dT =

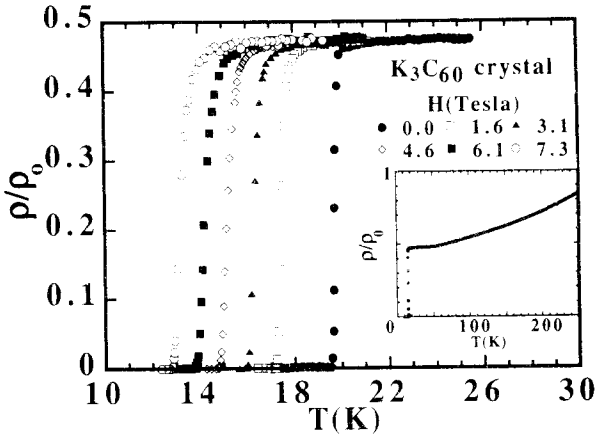

Fig.1. Normalized resistivity of K_3C_{60} single crystal near T_c for different applied H field. ρ_0 is the resistivity at room temperature. Inset: zero field temperature dependence of normalized resistivity.

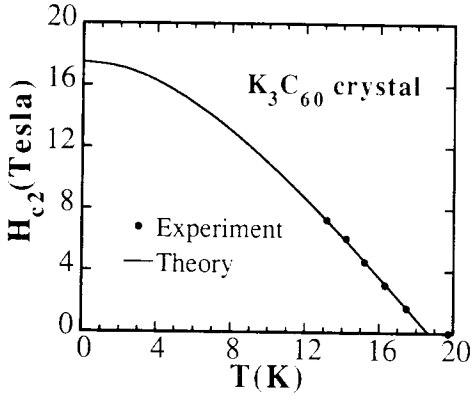

Fig.2. Temperature dependence of H_{c2}; solid dots: experimental data; solid line: theoretical calculation (see text).

-1.34 T/K. The experimentally determined $H_{c2}(T)$ relationship can be used to determine characteristic parameters for K_3C_{60}. Because it is not immediately clear whether K_3C_{60} is in the clean or dirty limit, we perform an analysis valid for arbitrary mean free path.

We first evaluate the scattering time τ using the experimentally determined $H_{c2}(T)$. H_{c2} is related to τ through the expression[10]

$$\frac{1+\lambda}{\lambda-\mu^*} = 2\pi\frac{T}{T_c}\sum_{i=0}^{N_c}\frac{1}{\chi_i^{-1}-(2\tau^*)^{-1}} \qquad (1)$$

where

$$N_c = \frac{1}{2}\left(\frac{\bar{\omega}}{\pi T}+1\right) \qquad (2)$$

and

$$\chi_i = \frac{2}{\sqrt{\alpha^*}}\int_0^\infty e^{-q^2}\tan^{-1}\left(\frac{q\sqrt{\alpha^*}}{(2i+1)\pi\frac{T}{T_c}+\frac{1}{2\tau^*}}\right)dq \qquad (3)$$

$H_{c2}(T)$ enters through the expression

$$\alpha^* = \frac{1}{2}e H_{c2}^* v_f^{*2} \quad , \qquad (4)$$

with the renormalized quantities defined as $H_{c2}^* = H_{c2}/(1+\lambda)T_c$, $v_f^* = v_f/\sqrt{(1+\lambda)}T_c$, and $\tau^* = \tau(1+\lambda)T_c$. The electron-phonon coupling constant λ is determined by the relation $T_c = 1.13\,\bar{\omega}\exp[-(1+\lambda)/(\lambda-\mu^*)]$. These expressions derive from a two-square well analysis of the Eliashberg theory. Pauli limiting has been neglected in this analysis--an approximation[10] consistent with the relatively small value of $(dH_{c2}/dT)[1/(1+\lambda)]$.

Equation (3) and associated relations allow a direct calculation of τ from H_{c2} if the coulomb repulsion μ^*, average phonon frequency $\bar{\omega}$, and Fermi velocity v_f are known. We assume μ^* to be in the range $0.1 \le \mu^* \le 0.3$, and use a value of $v_f = 1.8 \times 10^7$ cm/sec from a band structure calculation.[11] The average phonon frequency is estimated within three different electron-phonon models of the superconductivity for alkali fullerides,[3-5] which yield $\bar{\omega} \sim 500$ K and $\lambda \sim 1.0$ for the model of Jishi *et al.*,[4] $\bar{\omega} \sim 1000$ K and $\lambda \sim 0.7$ for the model of Schlüter *et al.*,[5] and $\bar{\omega} \sim 2000$ K and $\lambda \sim 0.5$ for the model of Varma *et al.*[3] These models yield very similar results. For $\mu^* = 0.2$, the models of Schlüter *et al.*[5] and Jishi *et al.*[4] yield $\tau \approx 1.7 \times 10^{-14}$ sec whereas the model of Varma *et al.*[3] yields $\tau \approx 1.6 \times 10^{-14}$ sec. Assuming $\mu^* = 0.1(0.3)$ gives scattering times roughly 0.2×10^{-14} sec smaller (larger).

Taking into account the uncertainties in the analysis, we estimate the zero temperature scattering time to be in the range $\tau = 1.7 \pm 0.5 \times 10^{-14}$ sec. With $\ell = v_f\tau$, we obtain a mean free path $\ell = 31 \pm 8$ Å which indicates that K_3C_{60} is in neither the clean nor dirty limit. We can use Eq. (3) to calculate the full temperature dependence of $H_{c2}(T)$; this is shown for the model of Jishi *et al.* as the solid curve in Fig. 2. Other models[3,5] yield similar results. The extrapolated value $H_{c2}(0) = 17.5$ T. We note that this value agrees well with that obtained directly from the Werthamer-Helfand-Hohenberg formula[12]

$$H_{c2}(0) = 0.69\left(\frac{dH_{c2}}{dT}\right)T_c \quad , \qquad (5)$$

which (using $dH_{c2}/dT = -1.34$ Tesla/K and $T_c = 18.7$K, a linearly extrapolated value) yields $H_{c2}(0) = 17.3$T.

Using $H_{c2}(0) = 17.5$T and the standard relation $H_{c2}(0) = \Phi_0/2\pi\xi^2$, we obtain the zero temperature coherence length $\xi(0) = 45$ Å. This value is significantly larger than the values obtained on polycrystalline samples (~26 Å).[13] In this regard, we note that granular thin-film samples also yield a coherence length of ~26 Å.[14] The short coherence length of the thin-film samples is attributed to the increase in H_{c2} which is observed in samples where the grain size approches the coherence length.[15] In support of this hypothesis, we remark that the experimentally observed H_{c2} (T) curve for K_3C_{60} is dependent on sample quality, even for single crystal specimens. For single crystal samples with transition width broader than 0.3 K, the upper critical field is a function of the transition width ranging from 28 T to 55 T for samples with ΔT_c ranging from 0.4 to 1.0 K. For these samples, the superconducting fluctuation conductivities were found to be of zero dimension. This suggests that the grain size in these inferior samples is comparable to the coherence length, presumably due to the effects of inhomogeneity or defects. Therefore, we believe that the higher upper critical field measured in these samples is not intrinsic (see Ref. 9). The H_{c2} result discussed in this paper was obtained in our best sample which has a pure three-dimensional fluctuation effect, and the homogeneous domain size (> 0.6 μm) determined by the fluctuation measurement is much larger than the coherence length.

The calculation of $H_{c2}(0)$ is sensitive to v_f, with τ varying roughly as the inverse v_f^2. The band structure calculation which yields v_f assumes orientational order of the

C_{60} molecules. This assumption is at variance with experimental results[16] which suggest a random occupation of two orientations. A tight-binding calculation[17] indicates that this disorder will smear out the peaks in the density of states. However, the density of states at the Fermi level is roughly the same for the ordered and disordered systems. This result suggests (but does not guarantee) that the actual v_f is close to that calculated for the oriented system.

Knowledge of the scattering time and the plasma frequency allows an independent estimation of the resistivity $\rho = 4\pi/\tau\omega_p^2$. The band structure calculation[11] yields a plasma frequency of 1.2 eV, close to the free electron value of 1.4 eV assuming three electrons per C_{60} and an effective mass of $m^* \sim 3\ m_e$, the value which brings the free electron Fermi velocity into agreement with the band structure result.[11] Using a plasma frequency of 1.2 eV and $\tau = 1.7 \pm 0.5 \times 10^{-14}$ sec, we estimate the zero temperature resistivity $\rho(0)$ to be of order 0.18 ± 0.06 mΩ–cm. This value is in excellent agreement with $\rho(0) = 0.12$ mΩ-cm determined indirectly from paraconductivity measurements.[18] Although contact geometries make a direct determination of $\rho(0)$ difficult, our most reliable results so far yield $\rho(0) \approx 0.5$ mΩ-cm which is of the same order as the calculated value and significantly lower than $\rho(0)$ extrapolated from thin film data.[14]

We briefly consider the sensitivity of the calculation of $\rho(0)$ to errors in the bare plasma frequency. The plasma frequency relates the Fermi velocity by $\omega_p \sim \sqrt{\langle N(0) v_f^2 \rangle}$. Therefore, a smaller ω_p may imply a smaller v_f, which yields a longer scattering time τ. The changes in ω_p and τ tend to cancel out in the calculation of the resistivity. This result suggests that the calculated resistivity is relatively insensitive to the uncertainties in ω_p and v_f. The mean free path, the product of τ and v_f, behaves similarly.

Assuming a plasma frequency of 1.2 eV, we obtain a London penetration depth of $\lambda_L = c/\omega_p = 1560$ Å. Using Ginzburg-Landau theory[19] with $\tau = 1.7 \pm 0.5 \times 10^{-14}$ sec, we obtain $\lambda(0) \approx 2400 \pm 300$ Å. The Ginzburg-Landau theory can also be applied to derive the clean limit coherence length $\xi_0 \approx 135 \pm 20$ Å. This value is slightly larger than that obtained from Allen's formula,[19]

$$\xi_0 = \sqrt{\frac{7}{4}\zeta(3)}\ \frac{\hbar v_f}{2\pi k_B T_c (1+\lambda)} \qquad (6)$$

which yields $\xi_0 \approx 80$ - 110 Å for an electron-phonon coupling constant $\lambda \approx 0.5$ - 1.0, the relevant range for the models studied.

To summarize our results, we list in Table 1 the macroscopic parameters relevant to the normal and superconducting states of K_3C_{60}, extracted from our single crystal studies and related analysis. We note a caveat in the Ginzburg-Landau results in that real systems deviate somewhat from the Ginzburg-Landau temperature dependence. The quoted errors in $\lambda(0)$ and ξ_0 arise from uncertainties in τ and do not take into account possible deviations from the simple version of Ginzburg-Landau theory. Our analysis ignores interball electron-electron correlations, which may be significant for this narrow band system. We also remark that our analysis assumes metallic conduction, i.e. $k_f \ell > 1$. Although K_3C_{60} crystals satisfy this criterion, the relatively high normal state resistivity suggests the possibility of non-conventional scattering mechanisms.

Table 1. Macroscopic superconducting state and normal state parameters of K_3C_{60}.

Parameter	Value
T_c	19.7 K[a]
dH_{c2}/dT	-1.34 Tesla/K[a]
$H_{c2}(0)$	17.5 Tesla[a]
$\xi(0)$	45 Å[a]
ξ_0	130 ± 15 Å,[b] 95 ± 15 Å[c]
$\lambda(0)$	2400 ± 300 Å[b]
λ_L	1560 Å[d]
κ	53 ± 7[a]
τ	$1.7 \pm 0.5 \times 10^{-14}$ sec[a]
ℓ	31 ± 7 Å[a]
$\rho(0)$	0.18 ± 0.06 mΩ-cm[a]

[a]this work
[b]this work within Ginzburg-Landau theory
[c]from Eq. (6)
[d]from Ref. 10

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