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# Electrical resistivity of $K_3C_{60}$

O. Klein and G. Grüner
*Department of Physics, University of California, Los Angeles, Los Angeles, California 90024*

S.-M. Huang, J. B. Wiley, and R. B. Kaner
*Department of Chemistry, University of California, Los Angeles, Los Angeles, California 90024*
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We have measured the electrical resistivity $\rho$ of $K_3C_{60}$ prepared in a pressed pellet form, by employing a contactless technique operating at microwave frequency (60 GHz). The temperature dependence of $\rho$ can be fit by the functional form $\rho(T) = a + bT^2$ and the magnitude of the resistivity at $T_c$, $\rho(20\ \mathrm{K})$, is on the order of 0.5 m$\Omega$ cm. We also find that the resistivity drops by almost an order of magnitude between 300 and 20 K.

Although the normal-state magnetic properties of the alkali-metal-doped fullerenes have been well studied,[1–4] the transport properties remain largely unexplored. Recently, Palstra *et al.*[5] looked at the temperature dependence of various transport parameters on thin-films of $K_3C_{60}$. In particular they measured the dc resistivity and found an upturn at 250 $K$ with $\rho(T)$ increasing as $T$ decreased, reaching 3 m$\Omega$ cm at $T_c$. This feature was interpreted as the signature of a localization of carriers, the inferred mean free path being of the order of the interatomic distance. Another set of experiments by Xiang *et al.*[6] on a $K_3C_{60}$ single crystal shows a metallic temperature dependence to the dc resistivity. The variation of $\rho(T)$ between room temperature and $T_c$ was within a factor of 2, with $\rho$ equal to approximately 2.5 m$\Omega$ cm just above the superconducting transition. However, dc transport measurements are hindered by sample inhomogeneities and the available data are known to be affected by limited crystallinity in the case of thin films[5] or uncontrolled distribution of the alkali atoms in the case of slowly doped $C_{60}$ crystals.[6] Moreover, there has not been any precise evaluation of $\rho$ from dc data, as the exact value depends on the sample geometry.

In this paper, we report the measurement of high-frequency resistivity using a contactless technique, where a pressed pellet of $K_3C_{60}$ forms one wall of a microwave resonator. The $K_3C_{60}$ compound was produced from the solid-state reaction of a mixture of $C_{60}$ powder and K vapor. This process has been described in detail elsewhere.[7] The sample quality was monitored by measuring the fractional shielding diamagnetism relative to a Nb bulk reference. In the inset to Fig. 1, we display the temperature dependence of the shielding diamagnetism for a selected 10 mg of the synthesized powder, showing that at least 50% of this sample was composed of the superconducting phase. All together, we have synthesized 80 mg of the compound (with at least 30% of material composed of the superconducting phase). The powder was then pressed in a pellet of 8 mm diameter and 1 mm height under a pressure of 40 bars. The pellet was subsequently annealed for 12 h in a furnace at 200 °C.

The pellet was used to replace one of the walls of a cylindrical copper cavity resonating in the $TE_{011}$ mode at microwave frequencies (60 GHz). By measuring the temperature dependence of the bandwidth ($\Gamma$) and characteristic frequency ($f$) of the resonator, we can evaluate the electrical properties of the sample.[8] During the process, the pellet was *never* exposed to air, and all the sample mounting was performed in a He filled drybox. In a separate run we calibrated our apparatus using a disk of bulk Nb of 99.99% purity.

The variation of the bandwidth induced by the specimen ($\Delta\Gamma$) is proportional to the so-called surface resistance if the electromagnetic energy is screened by the sample (skin-depth regime). This regime is applicable if the height of the pellet is greater than the skin depth $\delta$ in the normal state and the penetration depth $\lambda$ in the superconducting state. This assumption can be checked

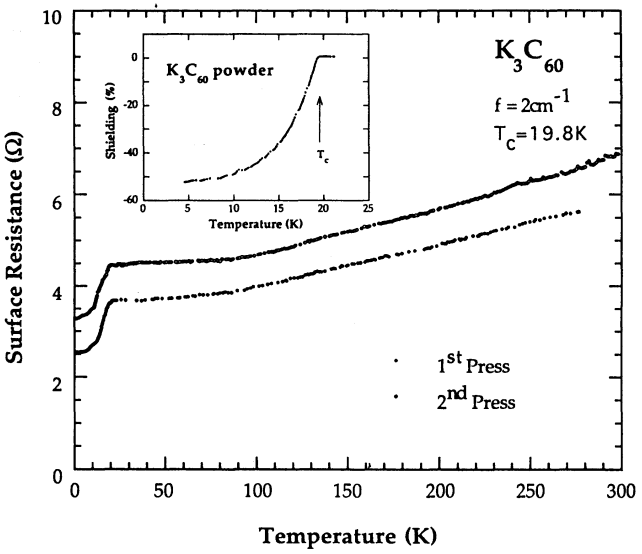


FIG. 1. Temperature dependence of the surface resistance $R_s$ of $K_3C_{60}$. The open circles represent a separate run after the sample was ground up and pressed a second time. Inset: Temperature dependence of the shielding diamagnetic fraction for a selected 10 mg of the powder sample.

 

experimentally by measuring the change in the characteristic frequency of the resonator before and after the introduction of the sample. Since the characteristic frequency of the cavity depends on the total volume of the resonator (skin depth included), we can estimate an upper bound for the effective skin depth in the pellet ($< 10$ $\mu$m) and check that the value is well below the height of the sample. It is important to note that we cannot make a precise evaluation of $\delta$ this way as the exact value of $f$ depends on the fine mounting geometry of the sample, and furthermore the measurement accuracy of the characteristic frequency is poor (because the bandwidth of the loaded cavity is very large) and we cannot extract from $f(T)$ any precise information below the corresponding 10 $\mu$m scale. We have displayed in Fig. 1 the temperature dependence of the surface resistance $R_s$ of the sample:

$$\Delta\Gamma/2f = \xi R_s, \tag{1}$$

where $\xi$ is the resonator constant and depends only on the geometrical characteristics of the cavity. Two different measurements are displayed, where in the second run (open circles) we have reground the sample and repressed it following the same procedure as above (the first sample preparation).

We note that several features are of importance: first a spuriously large residual contribution [$R_s^{\rm res}$ defined as the value of $R_s(1.2\ {\rm K}) \approx 3\ \Omega$] is observed due most probably to a leakage of the electromagnetic energy through the contact of the copper cavity body with the pellet, although in principle the chosen resonance mode should be insensitive to this effect (the magnetic field vanishes at the corners of the cavity and no eddy current should be flowing between the end plate and the body). The magnitude of $R_s^{\rm res}$ varies strongly on the full preparation process;[9] the value has changed by almost 0.8 $\Omega$ upon regrinding. However, the relative temperature dependence of the surface resistance, $R_s(T) - R_s^{\rm res}$, is unchanged as is evident from the figure, suggesting that the spurious contribution is very likely temperature *independent.*[10] The second important observation is that the temperature dependence of $R_s$ is linear above 100 K and can be fit by a straight line $R_s(T > 100\ {\rm K}) \approx a + bT$, where $a$ is sample dependent. We also observe that $R_s(T)$ saturates at low temperature ($20 < T < 100$ K). Next, we remark that the temperature of transition, $T_c$=19.8 K, is in good agreement with the value inferred by magnetic susceptibility measurements.[7] In the superconducting phase, the surface resistance drops sharply implying a high quality superconducting compound.

For a bulk material, the surface resistance is related to the electrical resistivity through the formula

$$R_s = \sqrt{\mu_0 \rho \omega / 2}, \tag{2}$$

where $f = \omega/2\pi$ and $\mu_0$ is the permeability of the free space. In order to extract the intrinsic part, we have plotted in Fig. 2 $[R_s(T) - R_s^{\rm res}]^2$ as a function of the temperature, and we believe that the observed dependence represents the intrinsic resistivity of the metallic phase. We have fit the temperature dependence of the resistivity with the functional form $\rho(T) = a + bT^2$. Both sets of

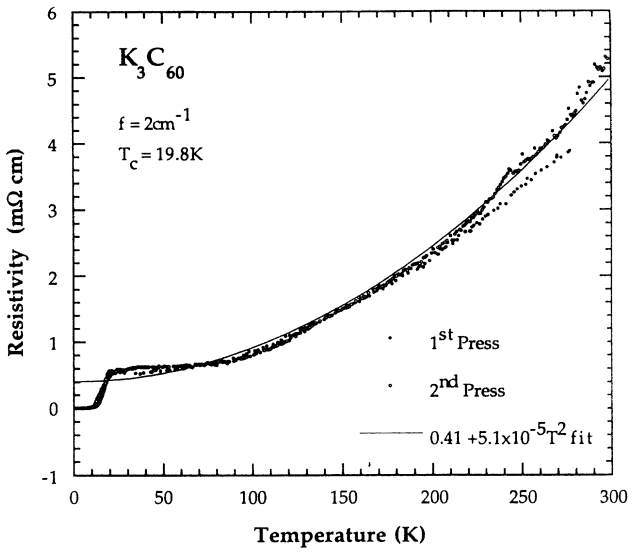


FIG. 2. Temperature dependence of the intrinsic resistivity. The solid lines represents a fit of the data using the functional form $\rho(T) = a + bT^2$.

data agree within 5%, and we think that this represents the overall uncertainty of the measurement. The resistivity drops by more than an order of magnitude between 300 and 20 K, displaying the strongest temperature dependence observed to date. From the resistivity at $T_c$ we can compute the skin depth $\delta$:

$$\delta = \sqrt{2/\mu_0 \omega \sigma} = 4.6\ \mu{\rm m}, \tag{3}$$

where $\sigma = 1/\rho$ is the conductivity. The skin depth is much smaller than the sample size, but certainly comparable to the grain size. Also, the surface of the pressed pellets have a nice mirror finish and therefore no corrections due to surface roughness are needed in the *normal state*, where the electromagnetic radiation probes the sample in a depth scale of at least several microns (skin depth).

Our results indicate that the sample is close to the clean limit. The plasma frequency can then be inferred from the penetration depth measurement[4] $\lambda(0)$=4800 Å by using the Pippard formula (valid in the London limit), which includes finite mean-free-path corrections:[11]

$$\lambda = \lambda_L \sqrt{1 + \xi(0)/\ell}, \tag{4}$$

where $\lambda_L = c/\omega_p$. If we assume that $\xi(0) \approx \ell$, then Eq. (4) leads to a plasma frequency $\omega_p/2\pi = 1.3 \times 10^{14}\ {\rm sec}^{-1}$, very close to the value measured by electron energy loss spectroscopy[12] ($1.8 \times 10^{14}\ {\rm sec}^{-1}$). Using the measured resistivity we can evaluate the scattering rate $1/\tau = 2.5 \times 10^{13}\ {\rm sec}^{-1}$, assuming a simple Drude response. From magnetic measurements[1] we obtain the Fermi velocity of $v_F = 5 \times 10^6$ cm sec$^{-1}$, leading to a mean free path $\ell = v_F \tau = 20$ Å. The mean free path is then comparable to the coherence length $\xi(0) = 26$ Å evaluated from critical field measurements,[3] indicating that the sample is in the intermediate limit [$\ell/\pi\xi(0) \approx 1$]. It is useful to note

that band calculations[13,14] lead to a Fermi velocity $v_F = 1.8 \times 10^7$ cm sec$^{-1}$, approximately four times larger than the previously quoted value, and thus a mean free path four times larger.

In conclusion, we have measured the normal-state electrical resistivity of $K_3C_{60}$ using a contactless microwave technique. The magnitude of the resistivity at the transition temperature is 0.5 m$\Omega$ cm. The temperature dependence of the resistivity can be fit by the functional form: $\rho(T) = a + bT^2$. Our results are in contrast with the upturn observed in $\rho(T)$ for thin films[5] but in qualitative agreement with earlier results on single crystals;[6] it should be noted, however, that we observe a much stronger temperature dependence (the resistivity drops by almost an order of magnitude between 300 and 20 K) than $\rho(T)$ observed in the single crystal.

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