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Pressure dependence of the superconducting transition temperature in nominal $Rb_{0.5}Cs_{2.5}C_{60}$

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We report measurements of the superconducting transition temperature in a compound with a nominal composition $Cs_{2.5}Rb_{0.5}C_{60}$ under hydrostatic pressure of up to 15.5 kbars. We observe two transitions, at temperatures T^* and T_c in this pelletized sample. We identify the lower temperature transition T^* with the onset of intergranular shielding currents. The upper transition temperature $T_c(P)$ falls on a universal $T_c(P)$ curve, defined from similar data obtained previously on K_3C_{60} and Rb_3C_{60}, after we shift the pressure axis for our data by -2.6 kbars. We identify T_c as a superconducting transition in Rb_2CsC_{60} on the basis of the compressibility analysis and published values of the lattice parameters for alkali-doped C_{60}, as well as $T_c(P=0)$ and $(dT_c/dP)_{P=0}$.

The discovery of superconductivity in the alkali-doped fullerene compounds[1-3] created a separate class of high-temperature superconductors. Superconducting transitions above 30 K have been observed in a number of compounds.[4,5] Both experimental[6-8] and theoretical[9,10] investigations arrive at the conclusion that the superconducting transitions temperature T_c, the density of states at the Fermi surface $N(E_F)$, and the face-centered-cubic lattice parameter a_0 have close to linear relationships among each other. Measurements of the superconducting transition temperature as a function of applied hydrostatic pressure, where we can tune continuously the lattice parameter, are then essential for understanding the nature of superconductivity in the doped C_{60} compounds. The route to higher transition temperatures lies in increasing the separation between the C_{60} molecules without destroying the original fcc structure of A_3C_{60} compounds. Superconductivity has been observed in Cs_2RbC_{60} at 33 K,[5] while attempts to synthesize Cs_3C_{60} have failed, presumably due to structural instability associated with the Cs atoms being too large to fit in the tetrahedral sites. Still, by increasing the Cs content further beyond Cs_2RbC_{60}, a compound may result in yet higher T_c. It is to this end that we synthesized the compound with a nominal composition $Cs_{2.5}Rb_{0.5}C_{60}$.

The nominally $Cs_{2.5}Rb_{0.5}C_{60}$ powder samples were prepared by methods described in detail elsewhere.[7] The powder was then ground, pressed into pellet, and sintered at 250 °C for 12 h. Initial x-ray-diffraction analysis indicated that a part of the sample forms in an fcc lattice with a lattice parameter value of 14.625 Å. However, dc susceptibility measurements resulted in a superconducting transition temperature of about 30.5 K. These values indicate that the superconducting compound does not fall on a monotonic progression of $T_c(a_0)$ that exists from K_3C_{60} to Rb_2CsC_{60},[4] if superconductivity is due to that part of the sample with $a_0=14.625$ Å. The questions that then arise are (1) is the observed T_c associated with material having $a_0=14.625$ Å and (2) if it is, will T_c decrease monotonically as in K- and Rb-doped samples, or will it increase to join the progression at Rb_2CsC_{60}, presumably when their lattice constants coincide?

The second motivation for undertaking pressure studies of the superconducting transition temperature in the nominal $Rb_{0.5}Cs_{2.5}C_{60}$ compound were the results of similar studies[6] performed on two different alkali-doped C_{60}: K_3C_{60} and Rb_3C_{60}. In these cases plots of the superconducting transition temperatures vs hydrostatic pressure fall on a unique curve after an appropriate relative shift along the pressure axis was made to compensate for the difference in the lattice parameters of the two materials at ambient pressure. Investigation of other superconducting doped C_{60} compounds and their high-pressure behavior would address the question of whether this curve is universal for other members of this class of materials, and possibly lend support to the theoretical ideas[9,10] that emanated from the previous studies.

To answer these questions we undertook a series of ac magnetic susceptibility experiments under hydrostatic pressure. We used a self-clamping Be-Cu pressure cell[12] and Fluorinert FC-75 as a hydrostatic pressure medium. This cell is capable of generating hydrostatic pressure of up to 20 kbar. The sample was placed in one of the two identical teflon cups that served as formers for the two coils of an astatic pair. Both cups were filled with Fluorinert and the pair was inserted into another Fluorinert-filled teflon cup. Care must be taken not to expose the sample to air, and so operation described above was performed in the He-atmosphere glove box. The pressure was determined from the inductively measured superconducting transition temperature of a piece of high purity lead that was pressurized with the sample. From the sharpness of the feature in ac susceptibility curves, we deduce that the pressure was very close to uniform over the sample volume. dc magnetization measurements have been performed on a pellet sample used

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in our experiment, and fractional diamagnetism was estimated to be 1–2 % of $1/4\pi$ at 5 K for an applied field of 10 Oe.

Figure 1 shows one of the raw data traces of ac susceptibility as a function of temperature at the nominal pressure of 1 bar. Most of such traces displayed not one, but two kinks in the curves, marked as T_c and T^* in the figure, that were separated by roughly 1.5 K. The origin of the two kinks may be the presence of two different superconducting phases in our sample. From the magnitude of the change in slope at both transitions we would then estimate the ratio of 4 to 1 of the low to high transition temperatures phases for our sample. However, similar behavior was observed for both K_3C_{60} and Rb_3C_{60} compounds,[6,11] where there was good reason to believe that only one superconducting phase was present. In those cases the lower temperature feature was attributed to the onset of intergranular coherence, with intergranular supercurrents and correspondingly higher shielding fraction. The ratios of 4:1 is also similar to those observed for these binary compounds. We therefore identify only T_c as a superconducting transition temperature for our sample.

We can plot our reduced data of $T_c(P)$ in a manner similar to that of Sparn *et al.*,[6] where the pressure data for K-doped samples were shifted along the pressure axis by 10.6 kbar to bring the data for both K- and Rb-doped C_{60} on a single curve. The resulting graph is shown in Fig. 2, where the pressure shift for T_c of our sample is -2.6 kbars, consistent with a larger unit-cell volume relative to Rb_3C_{60}. The data for both transitions join the previous data for K- and Rb-doped samples very well. That fact should be taken as an indication that superconductivity is not due to the phase with a lattice parameter of $a_0=14.625$ Å, since it falls off the common $T_c(a_0)$ trend, and therefore we should not expect it to follow the common $T_c(P)$ trend as well. From these data and the published values of the lattice parameters[4] for various

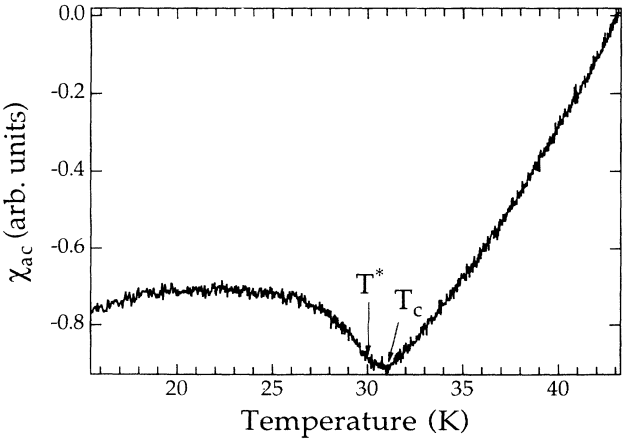

FIG. 1. ac susceptibility of nominal $Cs_{2.5}Rb_{0.5}C_{60}$ as a function of temperature for a single temperature sweep at an applied pressure of about 1 kbar. The origins of the two transitions marked as T^* and T_c are discussed in the text.

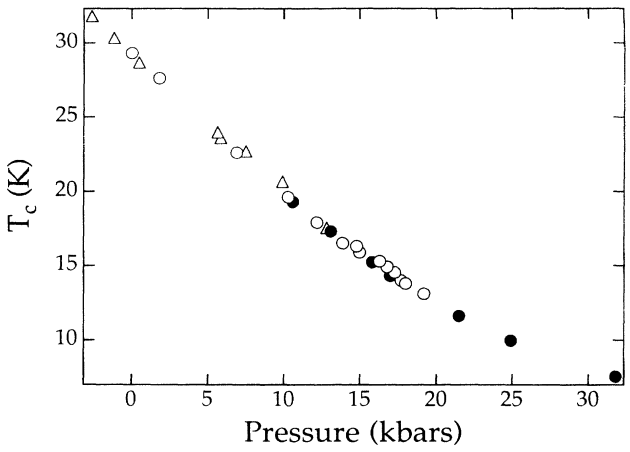

FIG. 2. Superconducting transition temperatures as a function of pressure for alkali-doped C_{60}. ●—K_3C_{60}, data taken from Ref. 6, shifted by 10.6 kbars; ○—Rb_3C_{60}, data taken from Ref. 6; △—T_c, shifted by -2.6 kbars.

alkali-metal dopants of C_{60}, we can derive the linear compressibility to aid us in identifying the superconducting phase in the compound. If we take the higher temperature transition T_c as that due to Rb_2CsC_{60}, we obtain the compressibility of 1.5×10^{-2} GPa^{-1} using the values[4] of $a_0=14.436$ Å for Rb_3C_{60} and $a_0=14.493$ Å for Rb_2CsC_{60}. Comparison with the compressibility values for K- and Rb-doped samples (1.2×10^{-2} and 1.5×10^{-2} GPa^{-1}, respectively) suggests that this identification is the correct one. The initial slope $(dT_c/dP)_{P=0}=-0.98\pm0.09$ K/kbar is virtually identical to that for Rb_3C_{60},[6] and the superconducting transition temperature $T_c(P=0)=31.8\pm0.2$ K is close to 31.3 K published previously[4] for Rb_2CsC_{60}. On the basis of the above analysis we identify the superconducting phase in our compound as Rb_2CsC_{60}. We therefore believe that structural decomposition of the nominally $Cs_{2.5}Rb_{0.5}C_{60}$ compound into $CsRb_2C_{60}$, a Cs-rich stable phase (Cs_4C_{60} and/or Cs_6C_{60}) and pure C_{60} is responsible for our observations.

In conclusion, we have investigated the superconducting transition in a compound of nominal composition $Cs_{2.5}Rb_{0.5}C_{60}$ under hydrostatic pressure of up to 15.5 kbars. We see two features in the ac susceptibility versus temperature plots and attribute the lower temperature feature at T^* as a signature of the onset of the intergranular coherence. On the basis of compressibility $(dT_c/dP)_{P=0}$, and $T_c(P=0)$ analyses, we identify the higher-temperature feature at T_c with the superconducting transition in the Rb_2CsC_{60} phase. We see a decrease with pressure of the superconducting transition temperature at the rate of -0.98 ± 0.09 K/kbar. This value is identical to that for the Rb_3C_{60}, perhaps indicating a more important role played by the smaller volume tetrahedral sites in regard to compressibility of the fcc phase of the alkali-doped C_{60}. Transition temperature T_c curves as a function of pressure can be shifted along the pressure axis to fall on the curve common with Rb- and

K-doped C_{60}. The temperature of the superconducting transition is therefore independent of the particular alkali dopant, but rather is just a function of the inter-C_{60} spacing.

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