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Observation of Metallic Conductivity and Sharp Superconducting Transition at 19 K in Potassium-Doped Fulleride, C_{60}, Single Crystal

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The first observation of a "metallic" conductivity behavior by resistivity measurements is reported for a potassium-doped C_{60} single crystal. The superconducting transition takes place very sharply with about one degree temperature width, that is, the onset of T_c is 20 K and the resistivity becomes null at 19 K. The details of the doping process are described.

The discovery of superconductivity of potassium-doped C_{60}[1)] has greatly accelerated the study on solid state C_{60}. In the first report by Hebard et al., thin films of K-doped C_{60} as well as powders were proved to show superconducting transition around 18 K.[1)] Elaborated studies for alkali metal-doped C_{60} polycrystalline powders have shown the superconducting transitions for K_3C_{60}, Rb_3C_{60}, and $Cs_2Rb_1C_{60}$ at 19.3, 30, and 33 K, respectively, by magnetic susceptibility measurements.[2,3)] In this communication we show the first observation of metallic conducting behavior for a K-doped C_{60} single crystal and a very sharp superconducting transition at 19 K through the resistivity measurement.

Single crystals of C_{60} were obtained from CS_2 solution as tiny black rods or plates. The typical size was $500\times100\times20$ μm^3. X-Ray diffraction analysis has proved that the crystal has a different crystal system of orthorhombic from the fcc system which is known for crystals obtained from benzene solution or vacuum deposition.[4)] The lattice constants of the orthorhombic crystal are a=25.001(6), b=25.582(7), and c=10.003(3) Å (Z=8). The density is 1.50 g cm^{-3} which is substantially smaller than 1.65 g cm^{-3} of fcc crystal. The details of structural analysis will appear elsewhere.[5)]

The doping of potassium to a crystal was carried out in a glass vessel under the vacuum of $\approx 10^{-6}$ Torr (Fig. 1(a)). A crystal was mounted on four gold wires (20 μm thick) spun on a BN bed and fixed by gold paint as shown in Fig. 1(b). The resistivity change on doping was continuously recorded in situ detection with four-probe method.

After completion of doping indicated by minimum resistivity, the glass vessel was disconnected from the vacuum line and transferred in a glove box which was filled with 1 atm. argon gas after evacuation to $\approx 2\times10^{-2}$ Torr. Immediately after taking the mounted crystal out of the vessel under the argon atmosphere, it was put into a tiny glass bottle, which was capped and sealed with a piece of potassium metal. Then, the bottle was set to a sample port on the insert of a cryostat and sealed in a cylinder by indium seal in the glove box under argon atmosphere. The insert was set in a liquid helium Dewer and the temperature dependence of the doped crystal resistivity was measured by dc or ac four-probe method.

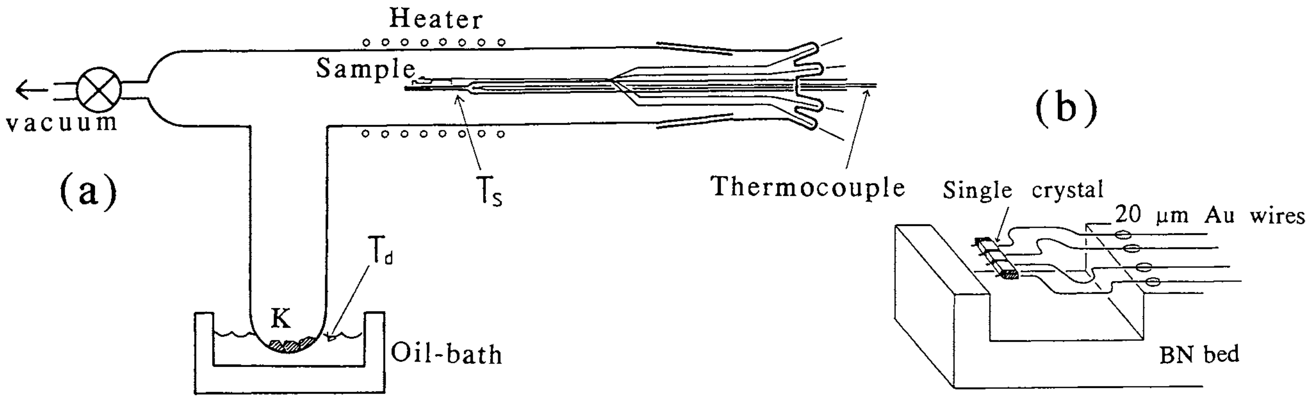

Fig. 1. Schematic representation of the glass vessel (a), and the sample part (b) for potassium doping.

We have tried to measure three crystals so far. The resistivity of a neat crystal is $\geq 10^8$ Ω cm at room temperature. The doping conditions and corresponding results for each crystal are summarized in Table 1. The crystal #1 showed rather low resistivity at room temperature, which increased with decreasing temperature. Observed temperature dependence data were much scattered and very noisy, but the "average resistivity" seemed to drop around 20 K as shown in Fig. 2. Most part of the crystal may be in superconducting state below ≈20 K. The crystal #2 which was doped very slowly with very long doping time. However, the final resistivity was much higher than #1, and its temperature dependence was much more insulating as shown in Fig. 3. A small drop in resistivity at 18 K was observed, but it never became zero above 4.2 K.

The crystal #3 was doped rather quickly in shortest doping time among three crystals. It showed the lowest resistivity at room temperature and "metallic" behavior with decreasing temperature (Fig. 4(a)). The metallic

Table 1. Doping Conditions and the Resistivities after Doping

	T_s/°C	T_d/°C	Doping time/h	ρ(r.t)/mΩ cm	ρ(20 K)/mΩ cm
Crystal #1	160-210	90-140	54	43	117
Crystal #2	190-200	60-95	234	600	80,000
Crystal #3	200-220	60-130	28[a)]	10	7

a) For the crystal #3, annealing at 220-230 °C was applied for 12 h after doping.

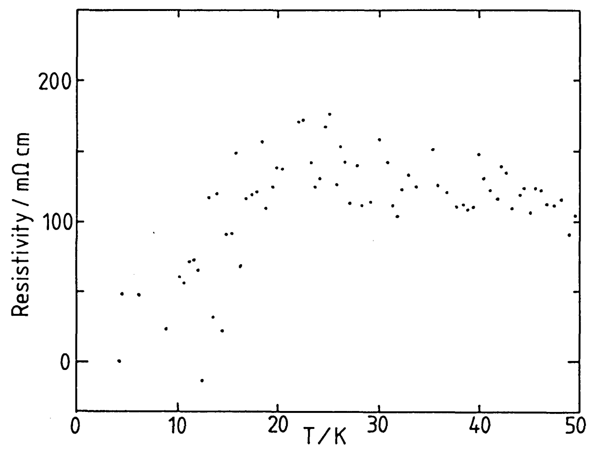

Fig. 2. Temperature dependence of the electrical resistivity of the crystal #1.

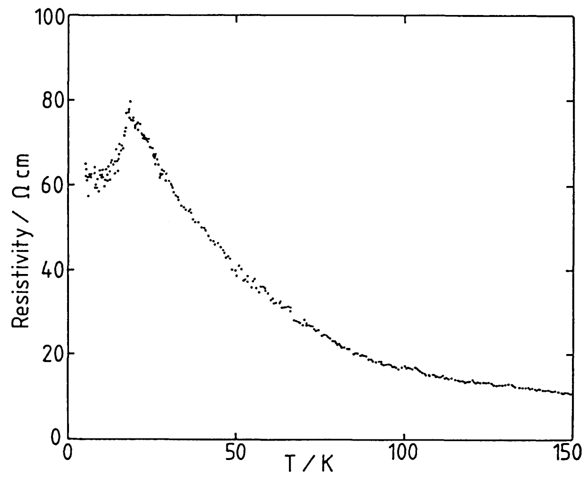

Fig. 3. Temperature dependence of the electrical resistivity of the crystal #2.

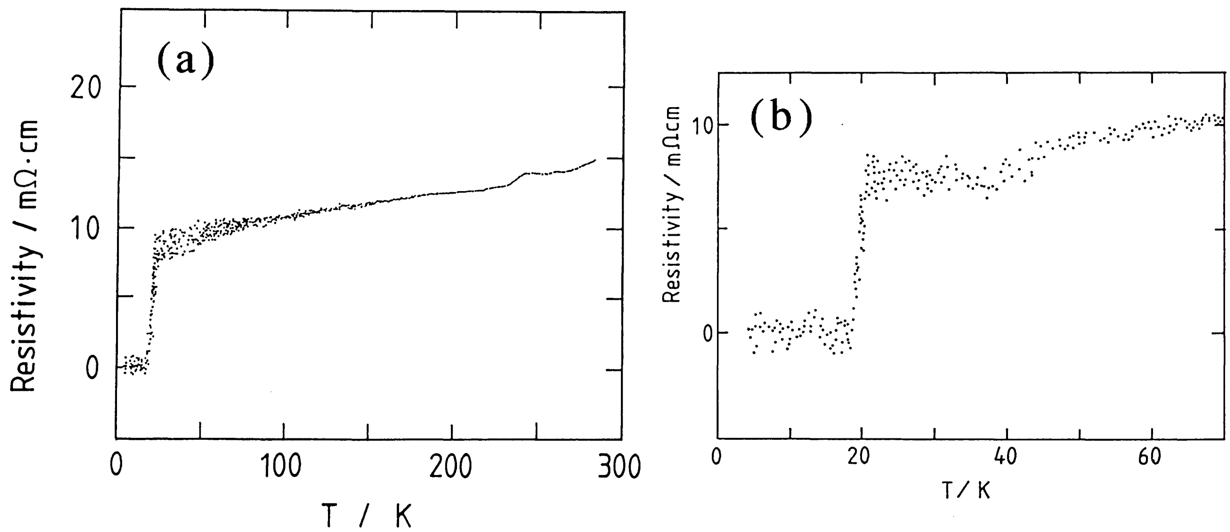

Fig. 4. Temperature dependence of the electrical resistivity of the crystal #3 (a), and the low temperature region (b).

behavior in resistivity change was observed for the first time as far as we know on the literatures reported so far. Moreover, a very sharp and clear superconductivity transition took place at 20 K (T_c onset) and the resistivity became zero at 19 K (Fig. 4(b)). The resistivity data are much scattered below 40 K even with ac measurement (40 Hz). The origin of the noise is not clear yet.

In summary, we observed a metallic conduction of a K-doped C_{60} single crystal by a resistivity measurement for the first time, and confirmed a sharp and clear superconductivity transition at 19-20 K.

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