Microwave Conductivity of Fullerene Intercalation Compounds: Mott Transition Driven by the Orthorhombic Distortion

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Superconductivity of A_3C_{60} (A=K and Rb) has been believed to be successfully explained by the BCS theory base on the band picture including a highly symmetric property of C_{60} molecules [1]. However, the intercalation of neutral ammonia molecules was found to turn the superconductivity into the antiferromagnetic ordered state [2, 3]. This suggests the breakdown of the simple band picture and the importance of the electronic correlation, such as an occurrence of the Mott-Hubbard metal-insulator transition in fullerene systems.

Unfortunately, the electric conduction of the ammonia intercalated compounds has not been investigated enough, because they are obtained only in the powder form at present. In particular, since the ammonia content is easily affected in the palletizing processes, the dc resistivity and the optical reflectivity have not yet been explored.

As shown in Fig. 1, by using a microwave cavity perturbation technique, we succeeded in studying the electric conduction of some ammoniated fullerides, including both superconductors (K_3C_{60}, $(NH_3)_xNaRb_2C_{60}$) and antiferromagnets, $(NH_3)K_{3-x}Rb_xC_{60}$ [4]. As shown in Fig. 2, it was found that the electric conduction of $(NH_3)K_3C_{60}$ was basically insulating below room temperature, showing a sharp contrast to the behavior of K_3C_{60}, by comparing the microwave loss, $\Delta(1/2Q)$, at the microwave magnetic field ($\boldsymbol{H}_\omega$) with that at the microwave electric field ($\boldsymbol{E}_\omega$).

Furthermore, we confirmed that the conductivity of the antiferromagnet $(NH_3)K_{3-x}Rb_xC_{60}$ at 200K is already 3-4 orders of magnitude smaller than those of superconductors, K_3C_{60} and $(NH_3)_xNaRb_2C_{60}$, as shown in Fig. 3. The striking difference in the conductivity between $(NH_3)K_3C_{60}$ and $(NH_3)_xNaRb_2C_{60}$ seems to be never explained by the increase of the effective volume per C_{60} molecule. Thus, the present results strongly suggest that the Mott-Hubbard transition in the A_3C_{60} systems is driven by a reduction of lattice symmetry from face-centered-cubic (fcc) to face-centered-orthorhombic (fco), rather than by the magnetic ordering.

According to a recent theory [5], the correlated multi-band metal is easily driven to the Mott insulator by a very weak splitting of the orbital degeneracy. Our results seem to be successfully consistent with this model.

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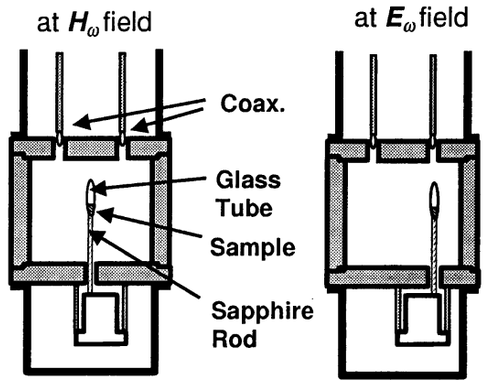

Fig. 1 Schematic view of the microwave cavity resonator.

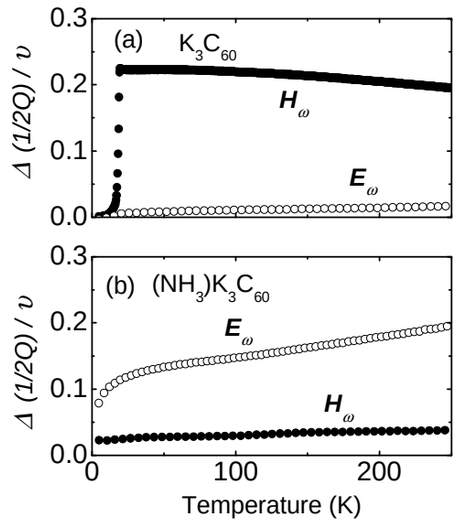

Fig.2 The temperature dependence of $\Delta(1/2Q)$ at H_ω- and E_ω fields for (a) K_3C_{60} and (b) $(NH_3)K_3C_{60}$.

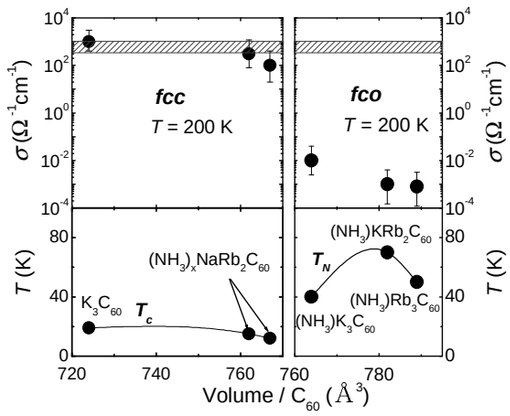

Fig. 3 The microwave conductivity at 200 K and the phase diagram of the ammonia intercalated fullerides as functions of the effective volume per C_{60} molecule.