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Novel Molecular System C₆₀: Fullerites and Fullerides

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NOVEL MOLECULAR SYSTEM C60: FULLERITES AND FULLERIDES

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ABSTRACT Absorption spectra and TEM and STM images are observed on C₆₀ thin films prepared by MBE technique. Superconductivity behaviors of alkali metal-doped single crystals and thin films are also reported.

INTRODUCTION

Since the first preparation of C_{60} molecules, 1 a lot of novel characters of the molecule have been revealed and versatilities for investigation not only on the molecule (Fullerene) but also on the solid state (Fullerite) have been much more increased after the invention of the preparation method for large amount of C_{60} material. 2 Moreover, the discovery of superconductivity of potassium-doped C_{60} has greatly accelerated the study on the solid state C_{60} . In the first report by Hebard et al., thin film of K-doped C_{60} as well as powders were proved to show superconducting transition around $18K.^3$ Elaborated studies on metal-doped C_{60} polycrystalline powders have shown the superconducting transitions for K_3C_{60} , Rb_3C_{60} , Cs_2RbC_{60} and $Rb_{3-x}Tl_xC_{60}$ at 19.3, 30, 33 and 42.5K, respectively. 4,5,6

In this report we show structural features and some optical properties of fullerite films prepared by MBE technique.⁷ We also present electrical transport behaviors of alkali metal-doped fullerides single crystals as well as films.

RESULTS AND DISCUSSION

TEM images of a fullerite film showed epitaxial growth of C_{60} crystal on an LiF single crystal of which structure is fcc. The optical absorption spectrum of the fullerite film manifested the appearance of forbidden bands in the molecular state around 450 and 650 nm besides the similar absorption bands in the 200-400 nm region of molecular

absorption.⁷ These bands may be assigned to the transition associated with the HOMO-LUMO gap transition. STM image of a monomolecular film of C₆₀ on an HOPG showed one-dimensional chain structure of molecules. Details of the observation will appear elsewhere.

Furthermore, a deposited film C₆₀(~60 nm in thickness) on quartz substrate showed distinct nonlinear optical effect, SHG and THG. There should be some asymmetry in the vertical direction in the film to afford the SHG. A rather high $\chi^{(3)}$ was also estimated to be 2×10⁻¹⁰ esu for the THG.8

Alkali metal doping to single crystals or thin films was carried out under various conditions. A single crystal obtained from CS₂ solution was doped with potassium at 200-220°C for 28 hrs and annealed at 230°C for 12 hrs. The resistivity of the crystal before doping was higher than $10^8 \Omega cm$ at room temperature, and it was reduced to ~10 $m\Omega$ cm after doping and annealing. The temperature dependence of the resistivity of this crystal was "metallic" from room temperature to 20 K, and superconductivity transition took place at 19-20 K very sharply. This is the first observation of the metallic behavior on the resistivity measurement so far.

Superconductivity of the K-doped films was also observed at 2-15 K with a little diffuse transition, although there were more difficulties in doping conditions compared with single crystal case. Doping experiments with other alkali metals or other metals are on the way.

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