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SURFACE
AND THIN FILMS

Spontaneous Growth of Single Crystals of Various Shapes in Tin–Fullerite Films

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Abstract—Changes in the structure and elemental and phase compositions of tin–fullerite films after their storage in air are investigated by X-ray diffraction, scanning electronic microscopy, atomic force microscopy, X-ray spectroscopic microanalysis, and Auger electron spectroscopy. Formation of the new phase Sn_xC_{60}, whisker tin and Sn_xC_{60} crystals, and petal and flowerlike fullerite crystals under the action of internal stress is established.

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INTRODUCTION

Fullerite is a new allotropic form of carbon, which attracts the attention of researchers owing to its unusual properties. It is a molecular crystal, which can be crystallized into either cubic or hexagonal phases at temperatures above 260 K. Molecules rotating with a frequency of 1×10^{10} s^{-1} occupy fullerite lattice sites. This high rotational mobility is related to the fact that a C_{60} molecule in the lattice is in a spherically symmetric potential [1]. At $T < 260$ K, transition to the low-temperature phase (a simple cubic lattice with partially frozen rotation of molecules) occurs.

The diameter of C_{60} molecules is 0.711 nm and, therefore, the size of octahedral and tetrahedral voids in the fullerite lattice is fairly large. As a result, doping of fullerite by atoms of some metals leads to the formation of new chemical compounds. C_{60} molecules in the lattice are bound by van der Waals forces. The sublimation temperature of fullerite is 580 K. Therefore, diffusion processes may be significant in some metal–fullerite systems at room temperature.

The purpose of this study is to investigate the interactions between Sn atoms and C_{60} molecules and the structural and phase changes in thin tin–fullerite films after their storage in air.

EXPERIMENTAL

Films were prepared by successive chemical vapor deposition in a VUP-5M system. Oxidized single-crystal Si(111) wafers were used as substrates. First, a 150-nm-thick fullerite film was deposited on a substrate and then a 130-nm-thick tin film was deposited. Fullerenes were sublimated from a tantalum crucible, whose temperature was maintained at $T = 770$ K. This value ensured a high deposition rate of films (2 nm/s). The purity of starting C_{60} and Sn was no worse than 99.98 and 99.999%, respectively.

The phase composition of the films was measured on a DRON-3.0 diffractometer in copper K_α radiation. The film structure was investigated under an LEO-1455 VP scanning electron microscope at accelerating voltages of 10 and 20 kV. The surface morphology was analyzed under a Solver PRO-P47 atomic force microscope. To provide operation in the semicontact mode, the atomic force microscope was equipped with noncontact cantilevers having a hardness coefficient of 2.5–10 N/m. Auger depth profiling of the films was performed on a PHI-660 scanning electron spectrometer. Films were sputtered by Ar^+ ions ($E = 3.5$ keV) at a rate of 15 nm/min. Detailed recording of Auger peaks was performed at the accelerating voltage $U = 3$ kV and electron irradiation doses no higher than 10^{16} cm^{-2}. Analysis of the elemental composition of new formations was performed by X-ray electron-probe microanalysis via the use of a Roentec energy-dispersive microanalyzer. The concentrations of light elements (C, O) and tin were found from the intensities of K and L lines, respectively.

EXPERIMENTAL RESULTS AND DISCUSSION

As-prepared fullerite films had a fine-grained structure with an average grain size of 50–100 nm (Fig. 1a). The surface roughness, measured by atomic force microscopy, was (10 ± 3) nm. The structure of a tin film