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Epitaxial C_{60} thin films on Bi(0001)

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Abstract

We have used the Bi(0001)/Si(111) template to grow highly ordered C_{60} epitaxial thin films and analyzed them using scanning tunneling microscopy and low-energy electron microscopy. The in situ low-energy electron microscope investigations show that the initial nucleation of the C_{60} islands on the surface takes place at surface defects, such as domain boundaries and multiple steps. The in-plane lattice parameters of this C_{60} film turns out to be the same as that of the bulk fcc(111) C_{60}. The line-on-line epitaxial structure is realized in spite of a weak interaction between the C_{60} molecules and Bi(0001) surface, while scanning tunneling spectroscopy indicates that there is a negligible charge transfer between the molecules and the surface.
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Organic materials are quickly becoming popular and important ingredients of the modern electronic devices. Among others, the buckminsterfullerenes [1] are worth to be noted as very promising in fabrication of electro-active elements in solar cells and active layers in organic thin film transistors [2,3]. The n-type conductivity of the C_{60} also induces significant interest in its application to electronic devices [4]. However, in majority of practical applications based on thin molecular films the key problem is the formation of homogeneously ordered structures, since the defects can critically affect the performance of device. Therefore, optimization of the growth parameters of organic structures, which would allow them to be successfully applied to organic-based devices, is a very relevant topic.

A search for the solution to this problem sparked a great deal of investigations related to C_{60} structures on various semiconductors [5–8]. The crystallographic ordering of the fullerene film is generally governed by a balance of two interactions: an intermolecular interaction and the interaction between C_{60} molecule and a substrate. Usually, the interaction between a semiconductor surface and an adsorbed fullerene molecule is rather strong due to high concentration of dangling bonds at the surface, and may have a significant influence on the morphology of thin solid films of van der Waals-bonded C_{60}. In case of metallic substrates, the electronic interaction leads to the mixing of substrate states with the LUMO orbitals, affecting strongly the electronic structure of the C_{60} monolayer [9].

In our previous works we have shown that semimetallic, chemically inert, well-ordered Bi films on the Si(111)-7×7 can be utilized as templates for growth of pentacene (Pn), where the weak interaction between Bi(0001) surface and Pn molecules results in the formation of highly crystalline molecular films [10,11]. In this letter we show that the Bi(0001)/Si(111) template is successfully applied to the growth of highly crystalline, epitaxial C_{60} films.

All experiments have been carried out in two separate ultra-high vacuum (UHV) systems – low energy electron

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microscope (LEEM) and a scanning tunneling microscope (STM), with base pressures in the low 10^{-10} Torr range. The preparation of Bi(0001)/Si(111) template is described elsewhere [12]. The depositions of C_{60} films have been performed from Ta-pocket evaporators on substrates kept at ~400 K. The C_{60} deposition rate was in the range of 0.05–0.1 ML/min, where 1 ML corresponds to the molecular density of the C_{60} bulk fcc(111) plane. The nucleation and morphology of the films have been followed in situ in LEEM experiments, while the STM data have been obtained after cooling the samples down to room temperature (RT).

STM images shown in Fig. 1, obtained from a monolayer of C_{60} grown on Bi(0001)/Si(111) show a highly crystalline layer with a perfect molecular ordering within a hundreds of nanometers. A single monolayer of C_{60} completely covers Bi(0001) surface, with the single-bilayer high Bi steps visible underneath, as shown in Fig. 1a. A high-resolution STM image in Fig. 1b shows an array of hexagonally arranged protrusions, each corresponding to a single C_{60} molecule. An additional modulation visible in this STM image is attributed to a Moiré pattern originating from the overlapping Bi(0001) and C_{60} lattices, and it is an immediate indication of the epitaxial relation existing between Bi substrate and C_{60} monolayer. Similar Moiré patterns were observed previously in the Pn/Bi(0001) system [13]. In the present case the modulation periodicity corresponds almost perfectly to a specific relation between C_{60} and Bi(0001) in-plane lattice parameters, defined by: $5|\mathbf{a}_{C_{60}}| \equiv 11|\mathbf{a}_{Bi}|$[1]. This fact and the distribution of the periodic pattern indicate line-on-line type of the epitaxial matching, characterized by C_{60} in-plane lattice vector aligning to each of three equivalent directions of the trigonal Bi substrate surface, $\langle 10\rangle$, $\langle 01\rangle$ and $\langle \bar{1}\bar{1}\rangle$, respectively.

Real-time LEEM observation gives more insight into the initial C_{60} nucleation and a further development of the C_{60} domains. There has been determined that the nucleation density of the C_{60} islands is rather high – typically exciding 10 μm^{-2} at substrate temperature of 400 K. Moreover, the chain-like alignment of the monolayer-high C_{60} islands (darker features) visible in the bright-field LEEM image recorded during C_{60} deposition and shown in Fig. 2a suggests that the initial nucleation of the C_{60} domains occurs preferentially on certain defects, which have been further identified as the Bi twin domain boundaries [14]. Additional STM data of the sub-monolayer C_{60} film, example of which (processed digitally [15] in order to enhance certain features in the image), shown in Fig. 2b, confirm that indeed the initial nucleation of the C_{60} domains occurs preferentially on the Bi domain boundaries (marked by dashed line in Fig. 2a) as well as on other defects, such as multiple-bilayer Bi steps.

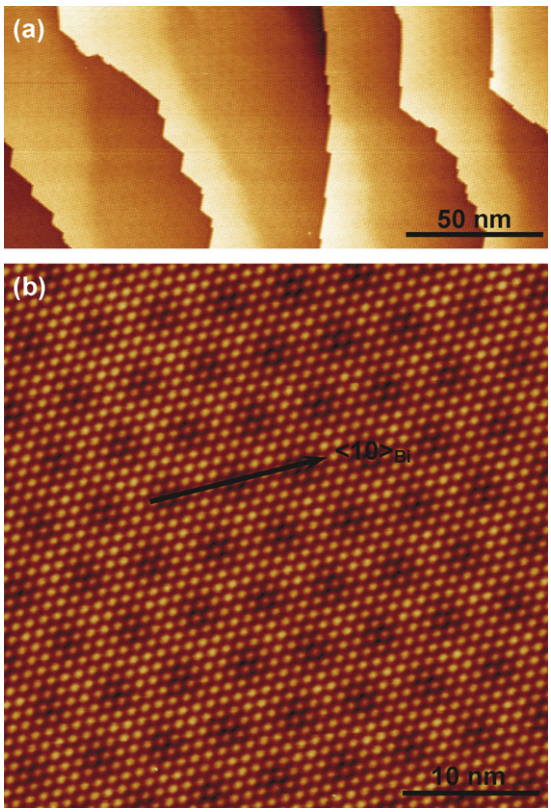

Fig. 1. (a) A large area STM image ($V_S = +2.3$ V, $I = 20$ pA) taken from the monolayer of C_{60} grown on Bi(0001) surface at ~400 K – the bilayer-high Bi steps are visible underneath the C_{60} layer; (b) A high-resolution STM image ($V_S = +2.3$ V, $I = 20$ pA) taken from the C_{60} film shown in (a); each protrusion corresponds to a single C_{60} molecule; additional modulation is attributed to a Moiré pattern originating from the overlapping C_{60} and Bi(0001) lattices.

In order to establish the exact relation between the C_{60} and Bi(0001) crystal lattices, the low-energy electron diffraction (LEED) patterns were acquired in the LEEM experiments. A series of such LEED patterns is shown in Fig. 3. The LEED pattern taken from a clean Bi(0001) surface prior to C_{60} deposition (Fig. 3a) shows hexagonal distribution of the diffraction spots, corresponding to the trigonal symmetry of the Bi(0001) surface [12]. Upon the deposition of 0.4 ML of C_{60}, the diffraction spots from the C_{60} layer become visible (Fig. 3b), while the Bi-related spots (an example is marked by an arrow in Fig. 3b) are still present. From the LEED pattern taken from sub-monolayer C_{60} film it is apparent that the directions of the inverse lattice vectors for both C_{60} and Bi(0001) lattices are same, which confirms that the C_{60} monolayer has a line-on-line epitaxial relation with the Bi(0001) surface. Moreover, having both the C_{60} and Bi-related spots visible simultaneously, we were able to accurately determine the C_{60} in-plane lattice parameter to be 10.04 ± 0.02 Å, which is same, within an experimental error, as that for the C_{60} bulk fcc(111) plane (10.02 Å)

[1] A natural cleaved (111) plane of rhombohedral Bi is called in the present paper the (0001) plane for hexagonal crystal indexing, with the simplest trigonal in-plane coordinates, $|a| = |b| = 4.546$ Å, and out-of-plane trigonal axis $|c| = 11.7967$ Å; the C_{60} bulk fcc(111) in-plane lattice parameter is 10.02 Å.

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