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Formation process of graphene on SiC (0 0 0 1)

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ABSTRACT

High-resolution transmission electron microscopy has revealed the formation process of graphene layers on SiC (0 0 0 1). Initially, nucleation occurs at SiC steps, covering them with a few layers of graphene. These curved graphene layers stand almost perpendicularly on the lower terrace. Graphene subsequently grows over the terrace region. The growth is often pinned by lattice defects of the SiC substrate.

1. Introduction

Graphene is a two-dimensional material consisting of carbon atoms. Extensive studies have been conducted concerning its peculiar electronic state [1–4]. It is known that the electronic state of graphene can be controlled by the number of layers, the stacking sequence [5], an applied electric field [6], and structural modification due to the substrate [7]. Silicon carbide is an attractive substrate to grow high quality graphene. Annealing the SiC crystal in a vacuum furnace leads to the sublimation of silicon atoms from the surface and the subsequent formation of graphene layers by the remaining carbon atoms [8–14]. We refer to this approach as the "SiC surface decomposition method" [8]. Using this method, wafer-scale graphene layers can be spontaneously formed on the insulating substrate. For the formation of high quality graphene, details of the formation process need to be clarified [15–21]. Previous studies to investigate the formation process of graphene on SiC have been carried out using atomic force microscopy and scanning tunneling microscopy [6,8,9]. However, the atomic-scale structure in the vicinity of a SiC step cannot be examined using these microscopies. In this study, we report the formation process of graphene on SiC (0 0 0 1) from the results of high-resolution transmission electron microscope (HRTEM) cross-sectional observations.

2. Experimental Procedure

Graphene on SiC (0 0 0 1) samples were prepared by annealing on-axis 6H-SiC (0 0 0 1) and 4°off-axis (the averaged terrace width before graphitization is about 5 nm) 4H-SiC (0 0 0 1) single crystals at 1350–1450 °C. Details of the preparation procedure were described in our previous paper [13]. Thin specimens for observation were obtained by Ar-ion thinning. Observations were carried out at room temperature using JEM-2010- and JEM-2010F-type transmission electron microscopes (TEM) at an accelerating voltage of 200 kV.

3. Experimental results

In this study, our main purpose was to investigate the formation process of graphene layers on SiC. Accordingly, we focused on the region around a step on the SiC where graphene nucleates.

Fig. 1 shows some typical HRTEM images of growth on stepped SiC. Figures (a) and (e) are the images from 4H-SiC with the electron incidence of [1 0 $\bar{1}$ 0] direction, while (b), (c), (d), and (f) are from 6H-SiC with the electron incidence of [1 1 $\bar{2}$ 0] direction. We carefully checked the observation conditions by comparison with simulated images using about 40 nm underfocus and sample thicknesses varying from 1.2 to 6.0 nm, as described in our previous paper [13]. In all the images, graphene layers appear as a dark line contrast on the SiC substrate. The SiC substrate appears as a bright dotted contrast, and has higher resolution in the lateral direction in (a) and (e) than in (c), (d), and (f). This difference comes from the electron incidence difference. Early in the growth

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process, about four curved layers of graphene covering the SiC steps are observed, as indicated by the arrows in (a). These line contrasts are present around the step positions, but are not visible clearly in other regions of the image. This indicates that the graphene was initially formed around the location of steps. Another interesting feature is that the edges of the graphene layers meet the lower terrace almost perpendicularly.

Fig. 1(b) shows an image of a wide area, indicating that growth proceeds from the steps to the terrace regions. Fig. 1(c) and (d) are enlargements of parts of (b). In (d), it is clear that curved graphene layers on a SiC step spread over the terrace region to the left. Here again, graphene layers intercept the lower terrace vertically. In another step-like region shown in (c), there are five layers on the right side of the step and three layers on the left side. In this case, graphene layers seemed to grow from the right to the left accompanying the decomposition of SiC. In other words, growth stopped in that region. It should be noted that areas of darker contrasts of SiC are observed as indicated by the arrows in (c). They indicate the presence of strain due to some kind of lattice defects. It appears that graphene growth was pinned by lattice defects of SiC in this region.

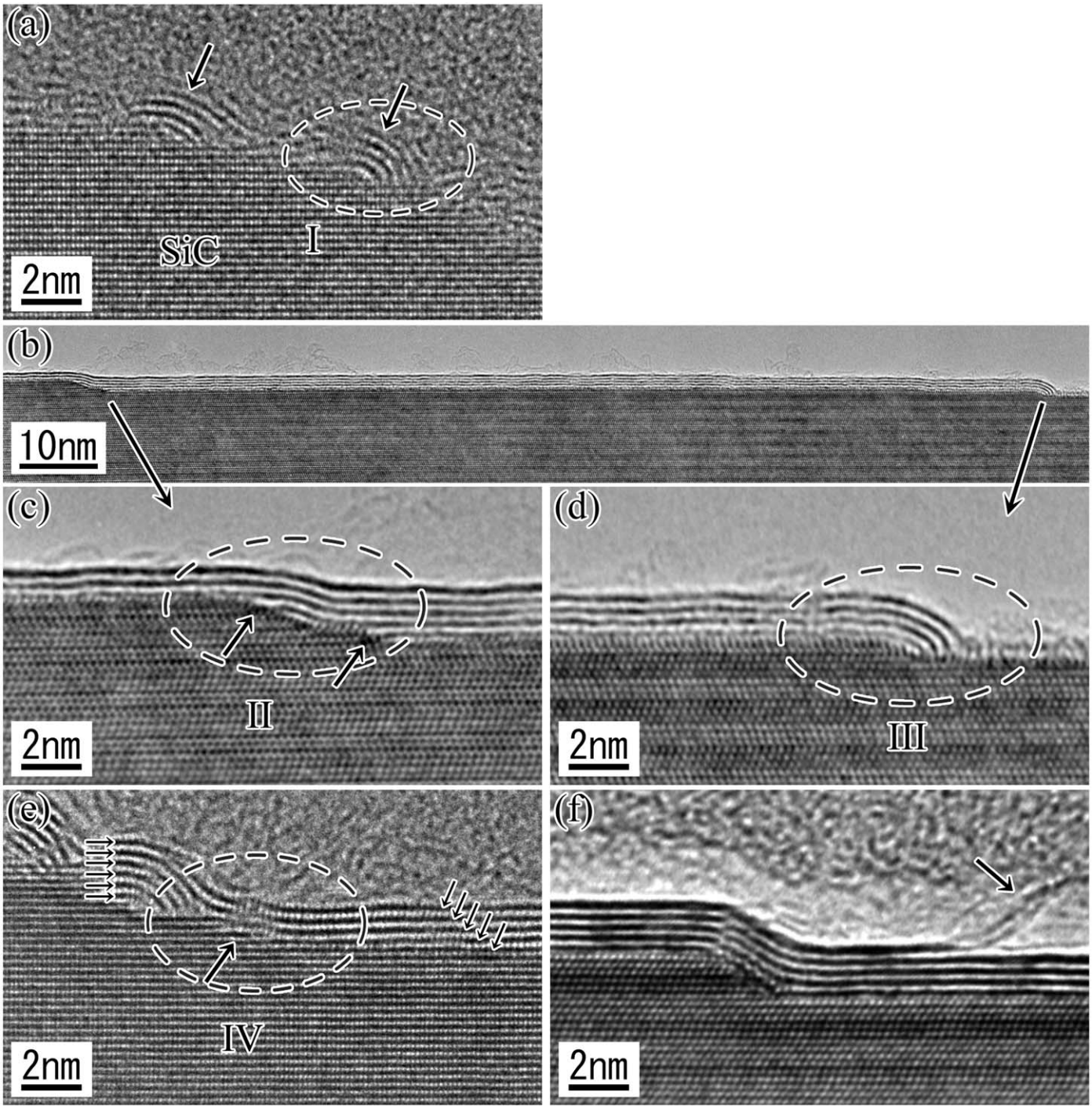

Fig. 1. HRTEM images of typical graphene layers formed on stepped SiC. Graphene layers are observed as dark line contrasts. (a) Initial growth occurs on steps. (b) Wide-area scan at a later stage of growth. The images in (c) and (d) are enlargements of part of (b). Images (e) and (f) show graphene around the step.