

Tuning of Structural and Electronic properties of Epitaxial Graphene by Substrate Microfabrication

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1. Introduction

Graphene is a promising material owing to its novel physical properties [1], such as band structure [2] and diamagnetism [3], which are often thickness-dependent.

To fulfill these properties toward device applications, thickness-controlled epitaxial graphene should be therefore produced. The recent advance in epitaxy of graphene on SiC makes it possible to produce high-quality graphene on a wafer scale [4]. An unintentional variation in thickness of the epitaxial graphene is introduced because of random nucleation near steps on the SiC substrate. It is therefore important to reduce the density of surface step on SiC substrate, in order to acquire the homogeneous epitaxial graphene film.

In this paper, we demonstrate self-organization of the epitaxial graphene with the aid of SiC substrate microfabrication (μ -EG). Spectromicroscopies confirm an improvement in the thickness-variation of μ -EG by adjusting microfabrication patterns. The improvement is due to a suppression of the random nucleation by decreasing surface step density on the microfabricated SiC. Furthermore electronic properties of μ -EG vary with the microfabrication pattern.

2. Experimental

We used 6H-SiC(0001) nitrogen-doped with a resistivity of $\sim 0.02 \Omega \text{ cm}$ as the substrates. The substrate microfabrication was performed by electron-beam lithography, followed by the etching using fast atom beam of SF₆. The etching depth was typically $\sim 100 \text{ nm}$. The samples were then etched by in hydrogen before graphene epitaxy. Graphene growth was carried out in a vertical cold-wall reactor comprising a double-walled, water-cooled quartz tube and a graphite susceptor in a slow flow of argon (purity 5.0). LEEM and PEEM measurements were carried out by LEEM III (Elmitec GmbH) installed at BL17SU, SPring-8.

3. Results & Discussions

Microscopic thickness variation of μ -EG

Figures 1a-b show LEEM images of μ -EG on differently-oriented rectangular pattern microfabricated on 6H-SiC(0001). From the LEEM image of μ -EG on non-fabricated region, the miscut of the substrate is in the $[11\bar{2}0]$ direction. Consequently the steps run parallel along the $[1100]$ direction. The shortening of the side in the $[11\bar{2}0]$ direction remarkably decrease the thickness variation (Fig. 1a), while that in the $[1100]$ direction cannot decrease the thickness variation (Fig. 1b).

The effect of the shortening of the side is remarkable in the rectangle pattern whose side in the $[11\bar{2}0]$ direction is $< 2 \mu\text{m}$. By AFM observations, it was found that no bunched step exists in the smaller patterns where no thickness variation is observed in Fig. 1a. It is thus concluded that thickness variation can be suppressed by decreasing the density of surface step where random nucleation of graphene occurs [5].

Electronic properties of μ -EG

Pattern-dependent electronic properties of μ -EG were probed by electron-reflectivity from the LEEM images because, in previous papers [6, 7], it is reported that the dip position is closely related to work function.

Figure 1c shows typical electron-reflectivity spectra of μ -EG (single layer) from the LEEM image (Figure 1a). The dip position varies with the pattern size. Figure 1d shows the shift of the dip position by the pattern size and shape. The dip position redshifts by decreasing the length of side in the $[11\bar{2}0]$ direction, while the dip position is independent of the size of the pattern whose short side is in the $[1100]$ direction. By comparison to the LEEM images (Figs. 1 a and b), these results indicate that the dip position is sensitive to the thickness-variation of μ -EG. On the same SiC substrate, the redshift of dip position indicates the change in the work function of graphene [7]. Our result therefore suggests that the degree of the doping of μ -EG varies with shape and size of the microfabrication pattern, maybe due

to the change of the density of impurities near the bunched steps where the thickness of the epitaxial graphene change.

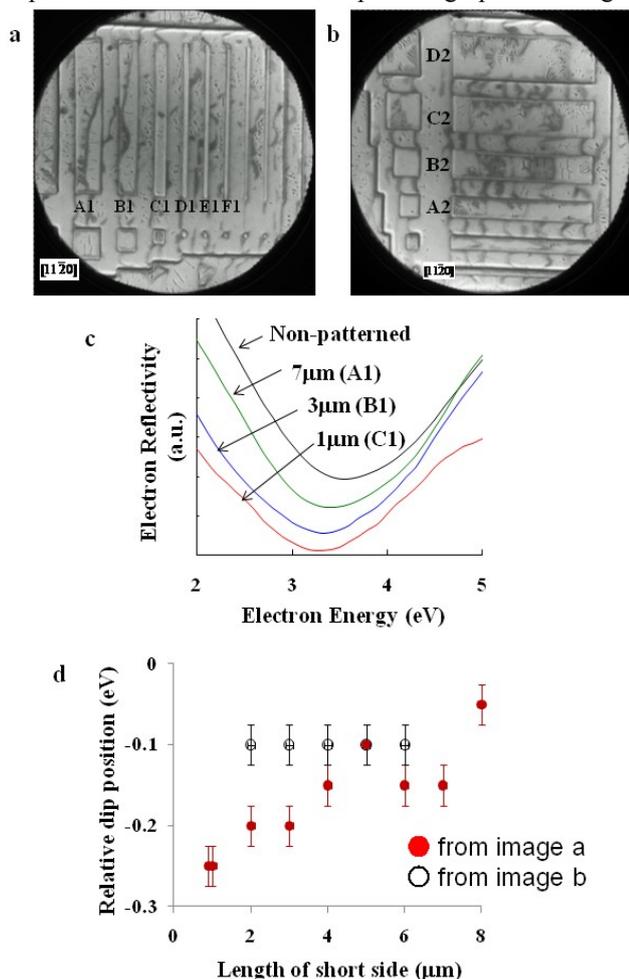


Fig. 1 (a, b) LEEM images of μ -EG on different rectangular patterns on the microfabricated 6H-SiC(0001) substrate. (c) typical electron reflectivity spectra of single layer of μ -EG on the rectangular pattern from image a. (d) Dependence of the dip position in the electron reflectivity spectra of single layer graphene on the rectangular pattern and size, relative to the dip position of EG. The filled and white circles are the dip positions of the spectra of the rectangle patterns whose shorter sides are in the directions of $[11\bar{2}0]$ (image a) and $[1100]$ (image b), respectively.

3. Conclusions

In summary, we have demonstrated that the epitaxial growth on microfabricated SiC(0001) provides uniform graphene layers, without thickness-variation, by adjusting size and shape of the microfabrication pattern. The uniform single graphene layer can be obtained in smaller pattern owing to the reduction of the density of surface step which originates the random nucleation. All necessary steps (substrate microfabrication, hydrogen etching, and graphene synthesis) can be fit with current semiconductor manufacturing. In combination with advanced microdevice production technologies, highly reliable graphene-based devices can be fabricated on a wafer scale.

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References

- [1] K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, Y. Zhang, S. V. Dubonos, I. V. Grigorieva, A. A. Firsov, *Science* **306** (2004) 666.
- [2] T. Ohta, A. Bostwick, Th. Seyller, K. Horn, E. Rotenberg, *Science* **313** (2006) 951.
- [3] M. Koshino and T. Ando, *Phys. Rev. B* **76** (2007) 085425.
- [4] V. Borovikov and A. Zangwill, *Phys. Rev. B* **80** (2009) 121406.
- [5] K. V. Emstev, A. Bostwick, K. Horn, J. Jobst, G. L. Kellogg, L. Ley, J. L. McChesney, T. Ohta, S. A. Reshanov, J. Röhl, E. Rotenberg, A. K. Schmid, D. Waldmann, H. B. Weber, and Th. Seyller, *Nature Mater.* **8** (2009) 203
- [6] H. Hibino, H. Kageshima, F. Maeda, M. Nagase, Y. Kobayashi, and H. Yamaguchi, *Phys. Rev. B* **77** (2008) 075413 (2008).
- [7] H. Hibino, H. Kageshima, F. Maeda, M. Nagase, Y. Kobayashi, and H. Yamaguchi, *e-J. Surf. Sci. Nanotech.* **6** (2008) 107.