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Introducing Nonuniform Strain to Graphene: Toward Strain Engineering

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1. Introduction

Due to the linear gapless energy spectrum, graphene exhibits lots of unusual phenomena, which are fascinating from the viewpoint of fundamental sciences but also are important for applications. For its application to electronic devices, the formation of a band gap is indispensable, because the inherent conduction in graphene is metallic. Several methods have been proposed for forming band gaps in graphene, including the electron confinement in graphene nanoribbons[1], the application of perpendicular electric fields to bilayer graphene[2], and the chemical modification of graphene[3]. However, the Dirac fermionic nature of electrons prevents the confinement by electrostatic potentials (called Klein tunneling), and high sensitivity of the electron transport to atomic-scale disorder makes the realization of insulating states inside the gap challenging [4].

Recently, another method to induce a band gap was proposed, which was based on the generation of a pseudomagnetic field by applying a nonuniform strain to graphene[5,6]. By using pseudomagnetic fields, one can confine electrons, form nanoribbons, and produce a Landau-level-like energy spectrum with multiple energy gaps at high energies. Thus, the strain engineering is a possible breakthrough for graphene-based electronics.

Experimentally, strain-induced Landau levels were observed in a scanning tunneling microscopy study on the top of nanobubbles which were naturally formed in chemical-vapor-deposition (CVD)-grown graphene[7]. The corresponding pseudo-magnetic field exceeded 300 T. On the other hand, designed pseudo-magnetic fields or strain-induced band gaps in graphene have not been attained yet, mainly because a practical method for introducing a designed nonuniform strain is lacking.

In this paper, we describe a technique to introduce designed nonuniform strain to graphene. For this purpose, we place nanopillars with any shape made of a dielectric material (electron beam resist) between a graphene film and the Si substrate. The strength and spatial pattern of the strain can be designed by adjusting the shape, position and size of the pillars.

2. Experiments

For the fabrication of nanopillars on the surface of Si substrates, we take advantage of an uncommon property of e-beam resists; excess exposure to e-beam makes resists such as lift-off resist (LOR) and polymethyl methacrylate (PMMA) insoluble even in their developers and removers. An example is shown in Fig. 1, in which rectangular nanopillars with side of 300 nm form a triangular lattice between graphene and the substrate. The nanopillars are fabricated with an e-beam dose of 60,000 $\mu C/cm^2$, which is more than 100 times larger than the normal exposure.

When the substrate is dipped into distilled water at the final stage in removal of the resist without excess exposure, the graphene portions between nanopillars are attached to the substrate, as shown in Fig. 2(a), indicating that the graphene film is nonuniformly stretched. In this sample, to estimate the amount of stretch in graphene films, we place additional PMMA disks on top of the LOR pillars. In Fig. 2(a), each PMMA disk is exactly on top of the corresponding LOR pillar, indicating that the graphene did not shift laterally when stretching. The average stretch is defined as the increase of the length of a graphene section between a pair of pillars after the stretch divided by the original graphene length (separation of the pillars). From Fig. 2(a), one can estimate the average stretch of graphene between pillars, which ranges from 6 to 20%, depending on the direction, as shown in Fig. 2(b), in which pillars A to D correspond to those in Fig. 2(a). The absence of the triangular symmetry in the average stretch is presumably because of a minimal lateral shift of graphene, which is undetectable by the present method. This shift also causes wrinkles in Fig. 2(a). Figure 2(c) shows an another example, in which the top PMMA disks are shifted in the direction perpendicular to the graphene edge (indicated by the dashed line). The amount of the shift becomes larger as pillars approach the graphene edge. Even in this case, though, the graphene film stretches along the shift direction with an average stretch of 7%, as estimated from the SEM image.

To confirm the nonuniform strain in graphene, we performed spatially resolved micro-Raman spectroscopy. The Raman spectra were acquired using a laser excitation of 532 nm (2.33 eV) with an incident power of 100 μW and a spot size of ~ 0.5 μm. The result is shown in Fig. 3. Figure 3(a) is a schematic view of the sample, in which a triangular lattice of LOR pillars (indicated by open circles)

130 nm high and 200 nm wide is placed underneath graphene. The separation of pillars is 1.5 μm. The average stretch of graphene between adjacent pillars is estimated to be ~ 6 - 9% from SEM images (not shown), depending on the direction. Figure 3(b) to 3(d) is the Raman spectra around the 2D band for several points, A_1, A_2, B_1, …, E_2, indicated in Fig. 3(a). Here, A_i (i = 1, 2) is on a pillar and B_i to D_i are located near the midpoint of adjacent pillars. Points X_1 and X_2 (X = A, ..., D) are equivalent in the triangular lattice. E_1 and E_2 are near the graphene edge. On the pillars (Fig. 3(b)), the 2D peak is located at (2663 ± 1) cm^{-1}, which downshifts in comparison with the value of ~ 2680 cm^{-1} for graphene placed on a Si substrate (without pillars), as shown in Fig. 3(e). Note that the LOR pillars (without graphene) do not show any Raman peak around the 2D band. On the other hand, near the midpoint between adjacent pillars (Fig. 3(c)), the spectrum is almost independent of the direction within the accuracy of the measurement, and exhibits a maximum at (2670 ± 2) cm^{-1}. The difference of the 2D peak positions in Figs. 3(b), 3(c), and 3(e) presumably originates from the variation of strain. Actually, near a graphene edge, where one can expect a relaxation of strain due to the existence of the edge, a smaller downshift is observed as shown in Fig. 3(d), in which the 2D peak is situated at (2677 ± 1) cm^{-1}.

3. Summary

We have developed a technique to introduce nonuniform strain in graphene by using pillars made of an electron beam resist. The amount of stretch, estimated from SEM images, reached ~ 20%. Raman spectroscopy indicated the application of nonuniform strain. This technique has a possibility to be applied to the formation of band gaps in graphene.

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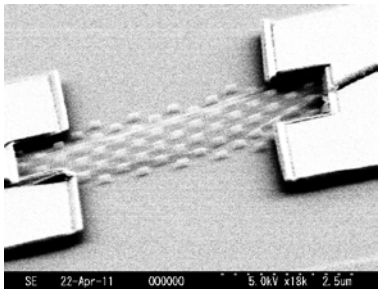

Fig. 1. SEM image of nanopillars sandwiched between graphene and the substrate.

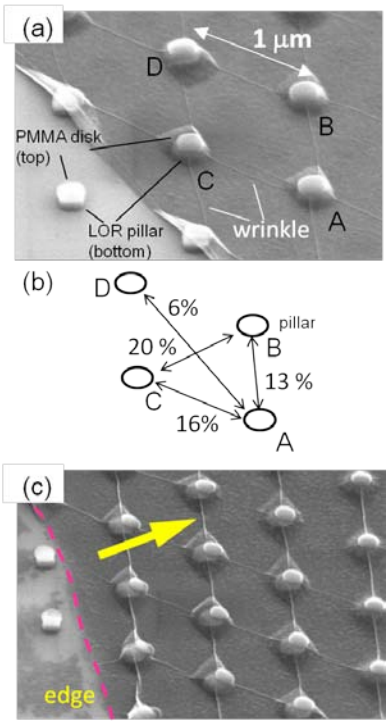

Fig. 2. (a) SEM image of graphene attached to the substrate. (b) The average stretch between nanopillars estimated from (a). (c) SEM image of graphene with noticeable lateral shift.

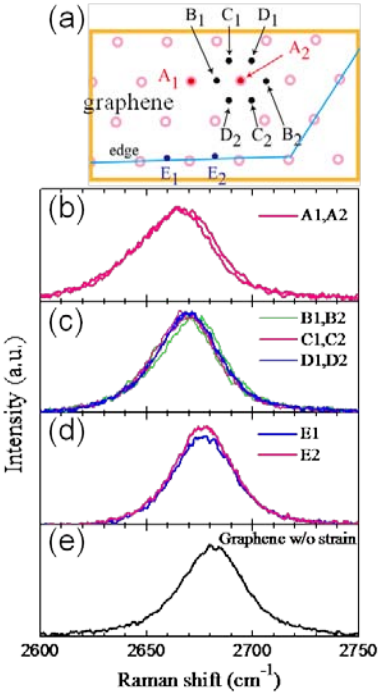

Fig. 3. (a) Schematic top view of strained graphene. (b)-(d) Raman spectra around 2D band for graphene on pillars (b), for graphene near the midpoint of adjacent pillars (c), and for graphene near an edge (d). Positions for each spectrum is indicated in (a). (e) Typical Raman spectrum for graphene without strain.