

Origin of Charge Puddle of Graphene on α - SiO₂: First Principles Study.

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Ripples and charge impurity effect of graphene are considered as the origin of charge puddles in graphene sheet on SiO₂. However, this topic is very controversial among researchers in graphene community. In this study, by using density functional theory, we calculate the band structure of the rippled graphene model and charged impurity model that is located close to the (0001) α -quartz surface. We expect that this study will provide great insight on this matter.

INTRODUCTION

Graphene has attracted great attention because of exceptional electronic properties, including high carrier mobility. However, this ultrahigh mobility has been achieved only in suspended graphene or on exfoliated flakes of hexagonal boron nitride (h-BN). [1-2]

One way to solve this problem is that ripple shape of graphene, due to surface roughness of substrate, can substantially affect charge carrier mobility. [3] The other idea is that common large-surface area substrates such as oxides can make charged impurities. [4]

Controlling and eliminating the carrier scattering from these charged puddles are fundamentally important and needed for practical electronic applications.

In this study, we investigated the electronic structure of rippled graphene and SiO₂-supported graphene model. We will be able to clearly

identify the origin of carrier mobility degradation of graphene on SiO₂

MODELS

First, to make the ripple graphene model, we used 10 ×10 supercell from the unit cell of pristine graphene (Fig.1(a)). If the lattice constant of pristine graphene is defined as 'b', that of rippled graphene model is defined as 'a' (a<b) and the graphene is induced with the strain defined as ' $\text{Strain} = (1 - \frac{a}{b}) \times 100(\%)$ ' where b is constant, 24.81 Å (Fig.1 (b)). We can get the rippled graphene models through the strain-induced graphene was calculated by geometry optimization (Fig.1 (c)). The table 1 shows the lattice constant, diameter, height and height/diameter ratio with respect to the induced strain. It shows that the larger the strain, higher the height/diameter ratio and result in a larger curvature.

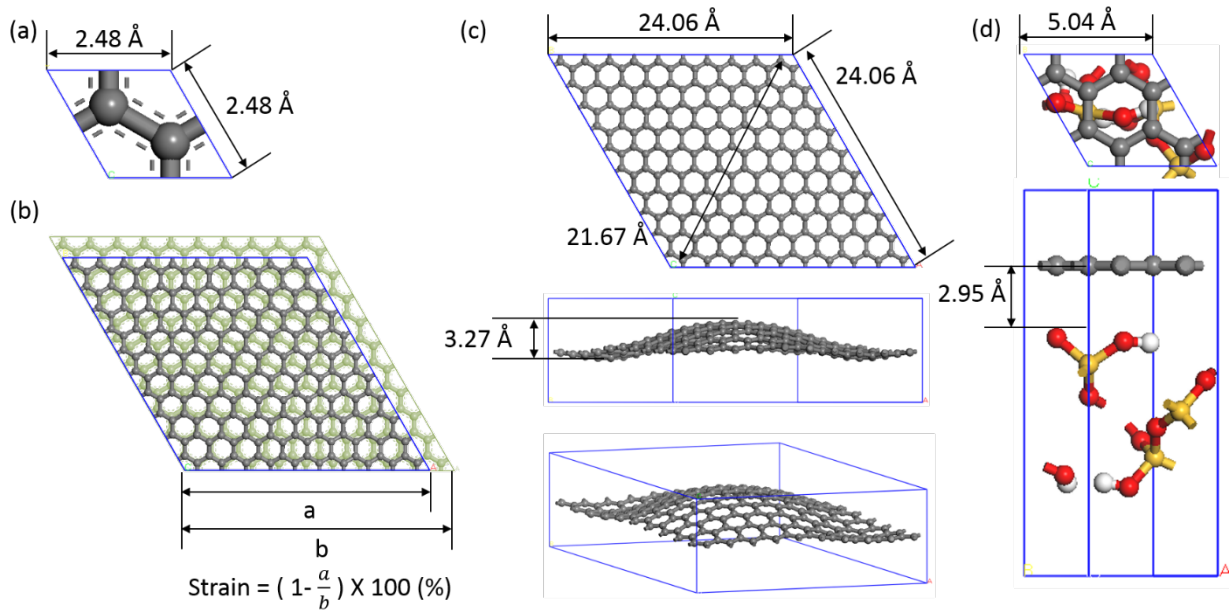


Fig. 15. (a) Unit cell of pristine graphene within hexagonal cell and lattice constant, $a=2.48 \text{ \AA}$. (b) Size of pristine (strain=0%) graphene and strained graphene model, strain is defined by $(1-a/b) \times 100(\%)$, where b is constant ($b = 24.81 \text{ \AA}$). (c) 3% strained ripple model, They foamed ripple shape with lattice constant($a=24.06 \text{ \AA}$), diameter (about 21 \AA) and height (3.27 \AA). (d) Graphene on (0001) α -quartz surface model. The lattice constant is 5.04 \AA and distance between graphene and α -quartz surface is 2.95 \AA .

Strain [%]	1.0	1.5	2.0	2.5	3.0	3.5	4.0	4.5
Lattice constant [Å]	24.557	24.433	24.309	24.185	24.061	23.937	23.813	23.689
Diameter [Å]	22.132	22.016	21.901	21.786	21.665	21.549	21.434	21.320
Height [Å]	1.719	2.156	2.533	2.912	3.268	3.540	3.789	4.017
H/D ratio	0.078	0.098	0.116	0.134	0.151	0.164	0.177	0.188

Table 13. Lattice constant, diameter height and height/diameter ratio (H/D ratio) of ripple models depending on induced strain.

Here, instead of amorphous SiO_2 , we considered crystalline structures to reduce the calculation cost. Here, instead of amorphous SiO_2 , we considered crystalline structures to reduce the calculation cost. Both crystalline and amorphous SiO_2 show similar electrical properties. We considered six types of model to describe various types of amor-

phous surface, hydrogen, hydroxyl, oxygen dangling bond and silicon dangling bond.

The α -quartz crystal model with three layers of oxide pair is used to construct the slab of SiO_2 with (0001) orientation which is normal to surface. In order to avoid vertical coupling effect, the vacuum is set with more than 20 \AA and bottom

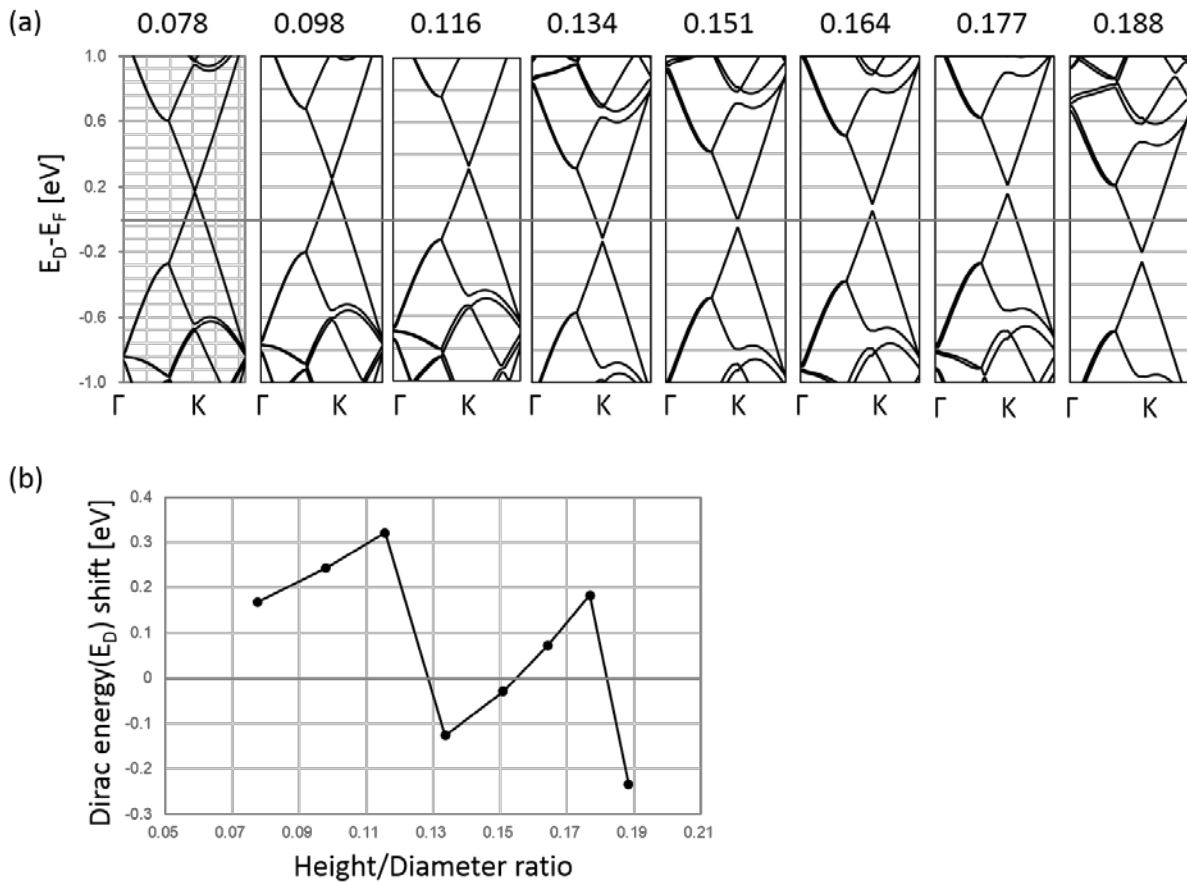


Fig. 2. (a) Band structure of ripple graphene model with respect to height/diameter ratio from 0.078 to 0.188 near Fermi energy. (b) Dirac energy (E_D) shift vs. Height/Diameter ratio with respect to ripple models

atoms are passivated with hydrogen.

The SiO_2 -graphene interface, containing a graphene layer with 8 carbon atoms, is simulated using a hexagonal unit cell with lattice parameters $a = b = 5.043 \text{ \AA}$. The 1×1 cell of α -quartz crystal can be properly matched for the 2×2 cell of graphene with small lattice mismatch of 1.45%. The equilibrium distance between the carbon of graphene and the closest silicon atom from graphene is 4.146 \AA (Fig.1(d)).

CALCULATION METHODS

We first study rippled and charged impurity models as shown in Fig. 1, using generalized gradient approximation (GGA - PBE) in LCAODFTLab of the EDISON Nano-physics that can calculate density functional theory (DFT). The Brillouin zone was done using a Monkhorst-Pack grid of $30 \times 30 \times 1$ for energy convergence and $150 \times 150 \times 1$ for electronic structure calculations.

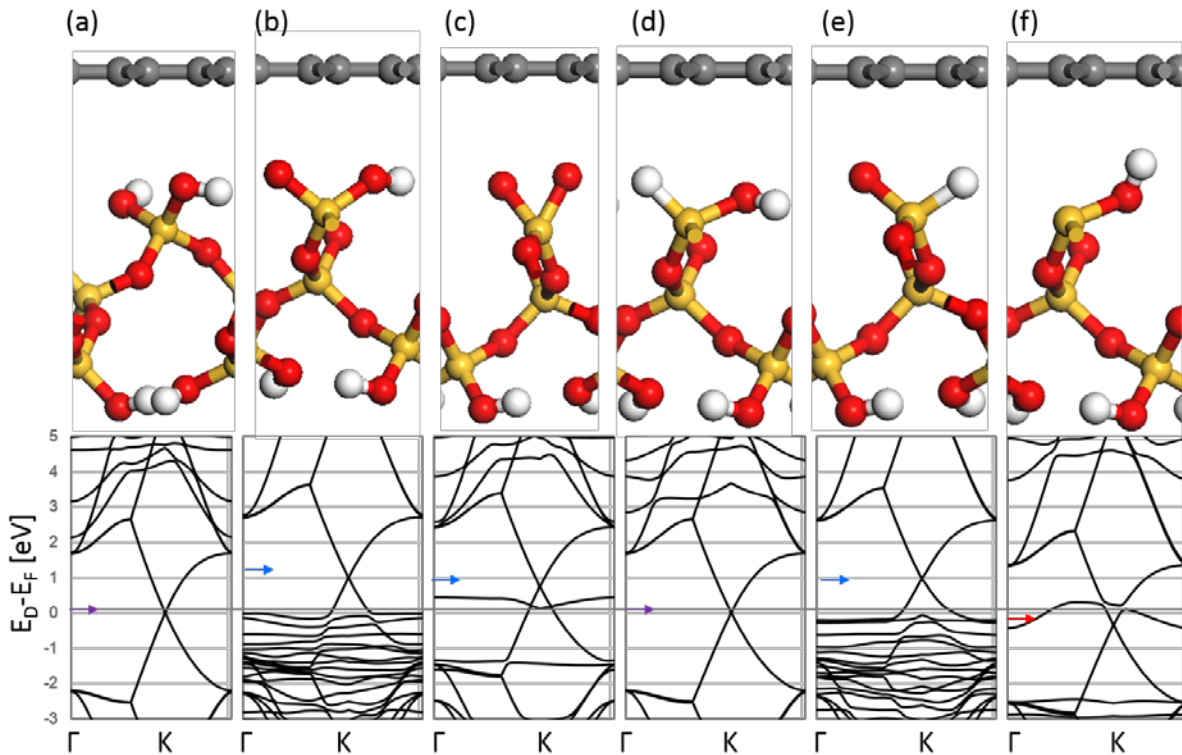


Fig. 3. Atomic configuration and band structure of graphene on α -quartz substrate. (a) hydroxyl ($-OH$) and hydroxyl, (b) hydroxyl and oxygen dangling bond, (c) oxygen dangling bond and oxygen dangling bond, (d) hydroxyl and hydrogen passivation, (e) oxygen dangling bond and hydrogen passivation, (f) hydroxyl and silicon dangling bond.

RESULTS AND DISCUSSION

1. Ripple graphene model

As shown in Fig. 2, we calculated the band structure of rippled graphene models. The Dirac energy (E_D) was shifted and it shows p-type or n-type behavior with respect to height/diameter ratio of rippled graphene models. It means that the curvature of ripples in graphene affects the formation of charge puddles with various size and types. Electron scattering from these charge puddles degrades charge carrier mobility.

2. SiO_2 /Graphene interface model

and structure of α -quartz/Graphene interface models with six types of surface.

In Fig 3(a), (d), the α -quartz surface has two of hydroxyl ($-OH$) (Fig.3(a)) and hydroxyl and hydrogen passivation (Fig.3(d)) within the unit cell. They have intrinsic graphene property where there is no shift in Dirac energy, due to the lack of dangling bonds.

In Fig.3 (b), (c), (e), the α -quartz surface have a hydroxyl and an oxygen dangling bond, two oxygen dangling bonds and an oxygen dangling bond and a hydrogen passivation, respectively. They have p-type behavior with Dirac energy shift due to dangling bonds from oxygen atoms.

In Fig.3 (f), the α -quartz surface has a hydroxyl and a silicon dangling bond. It unusually shows n-type behavior with dirac energy shift due to dangling bonds from silicon atoms.

It means that hydrogen passivated surface do not affect the charge puddle, on the other hand, the dangling bonds from oxygen and silicon atoms can affect the charge puddles to be p-type or n-type, respectively.

CONCLUSION

In order to find out the charge carrier mobility degradation effect of graphene on SiO_2 substrate, we calculated the electronic structure of rippled graphene model with respect to the various curvatures and SiO_2 /graphene interface model with respect to various substrate surface. The curvature of ripple in graphene affects formation of charge puddles with various size and types. In SiO_2 /graphene interface, the dangling bonds from oxygen and silicon atoms can affect the charge puddles to be p-type or n-type, respectively. We can expect that both ripple shape of graphene and SiO_2 substrate degrade charge carrier mobility due to the scattering effect from the charge puddles.

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