

III. CONCLUSIONS

Since it is technologically possible to control the grain size by varying doping concentration and deposition ratio of Ba and Si atomic fluxes, we suggest the grain size at room temperatures does not significantly affect values of electron mobility. The size of grains should be carefully considered only at low temperature. From the other side, the potential barrier at grain boundaries crucially decreases the maximum mobility values at the whole temperature range. We suppose the same tendencies to be observed in the case of hole as main carriers.

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AB INITIO SIMULATION OF GRAPHENE INTERACTION WITH SiO₂ SUBSTRATE FOR NANOELECTRONICS APPLICATION

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I. INTRODUCTION

Graphene is a promising material with high charge mobility [1-3]. The substrate material has a significant negative influence on the charge carriers mobility. Quartz well established as substrate in graphene technology. However, the mobility of charge carriers in such systems is worse than the theoretical calculations of a pure graphene sheet [2, 3]. The space charge inhomogeneity (so-called charge puddle) leads to degradation of the graphene electronic properties [4-6]. Physical characteristics leading to the occurrence of electron-hole puddles not identified uniquely [1, 4, 7]. Possible reasons for the formation of a charge puddle: open dangling bonds [8], point defects in the graphene layer [9-11], and charged impurities [12, 13].

The results of experimental work [14] prove that the presence of dangling bonds on the surface of the SiO₂ quartz substrate during the fabrication of the device may be the main reason for the occurrence of an electron-hole puddle in graphene.

Amorphous SiO₂ has various types of surface species. Therefore, impact of amorphous SiO₂ surface variety on the structure and electronic properties of graphene has been study in present theoretical investigation.

The α -quartz is the most stable polymorphic modification of SiO₂, and its most stable surface is (001) [15, 16]. Two typical quartz substrates were chosen for simulation. Ones have been with open oxygen (O_{unsat}) or silicon (Si_{unsat}) dangling bonds.

It is possible to passivate the broken bonds by substrate surface adsorbed hydrogen atoms for each of these types of bonds (Si_{sat} and O_{sat}). In theoretical work [17] have been determined that the most stable is surface α -SiO₂ with the atomic configuration (0001) (O_{sat}). Calculations were performed by quantum-

mechanical simulation without taking into account of Van der Waals forces which are significant in layered structures.

Thus, the quantum-mechanical study of a graphene monolayer deposited on all four types of surfaces: O_{unsat} , O_{sat} , Si_{unsat} and Si_{sat} using methods that take into account Van der Waals forces are of interest.

II. COMPUTATIONAL DETAILS

Calculations were performed based on the density functional theory (DFT). All of calculations have been carried out using VASP (Vienna Ab initio Simulation Package) [18-20]. The projector-augmented wave (PAW) potentials and Perdew–Burke–Ernzerhof (PBE) functional have been used. A cutoff energy of 520 eV. The valence electron configurations for Si, O, C, and H were $[Ne] 3s^2 3p^2$, $[He] 2s^2 2p^4$, $[He] 2s^2 2p^2$ and $1s1$, respectively. The atomic structures were relaxed until the forces on all unconstrained atoms were smaller than 0.01 eV/Å. A vacuum layer of 20 Å along z direction was constructed to eliminate the interaction with spurious replica images. The calculations were carried out without spin polarization. Integration in reverse energy space was carried on the $7 \times 7 \times 7$ and $5 \times 5 \times 5$ k-points grids determined by a fine grid of gamma-centered method.

The exchange-correlation functional was chosen based on preliminary calculations of the structural parameters of the graphite unit cells and α -SiO₂ (Table 1).

The DFT-D3 method of Grimme demonstrated demonstrated the smallest difference between the calculated and experimental magnitude of lattice constants and have been used for further calculation.

Table 1 – Exchange-correlation functional for graphite and α -SiO₂ structure calculation

Structure	Functional	Lattice constant			Difference with experiment, %
		a, Å	c, Å	c/a	
Graphite-2H	LDA[21]	2.4479	6.4596	2.6388	3.1109
	PBE[22]	2.4692	7.5047	3.0393	11.5938
	PBE-D2[23]	2.4647	6.3218	2.5649	5.8242
	PBE-D3(zero)[24]	2.4682	6.7947	2.7529	1.0771
	PBE-D3(BJ)[25]	2.4674	6.6772	2.7061	0.6386
	Experiment[26]	2.4642	6.7114	2.7235	-
α -SiO ₂ (quartz)	LDA[21]	5.0433	5.5341	1.0973	0.2563
	PBE[22]	5.0038	5.5869	1.1165	1.4903
	PBE-D2[23]	5.0886	5.5742	1.0954	0.4279
	PBE-D3(zero)[24]	5.0867	5.5745	1.0959	0.3853
	PBE-D3(BJ)[25]	5.0801	5.5704	1.0965	0.3293
	Experiment[27]	4.9133	5.4053	1.1001	-

Static self-consistent calculations were performed using the tetrahedron method and Bloch corrections. Integration in reverse energy space was carried on the $11 \times 11 \times 1$ k-points grids. Structural figures and charge density drawings were produced by VESTA package [28].

III. RESULTS AND DISCUSSION

Figure 1 shows the positions of the graphene sheet with respect to the position of the unit cell of quartz which was study in our work. Ground states were found for each type of the surface and position of graphene sheets. The adsorption energy was calculated. The binding energy of carbon atoms in graphene with a substrate was determined on the basis of relation

$$E_{\text{ads}} = \frac{E_{\text{total}} - E_{\text{gr}} - E_{\text{surf}}}{n},$$

where n – number of graphene unit cells used in supercell, E_{total} – total energy of graphene-SiO₂ (0001) supercell, E_{gr} – total energy of the graphene, E_{surf} – total energy of quartz substrate.

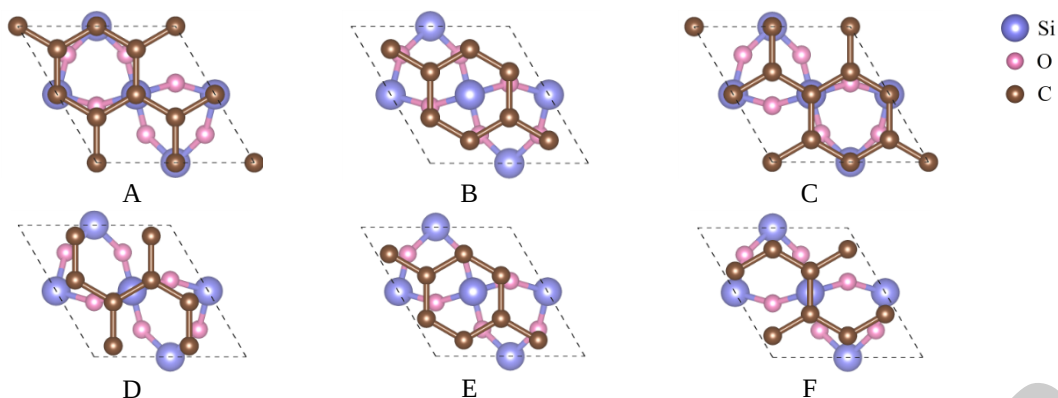


Figure 1 – Position of carbon atoms on quartz (0001) substrate

Interlayer distances and adsorption energy were calculated. The absorption of graphene on the surface of quartz with oxygen dangling bonds is most energetically favorable ($E_{ads} = -281$ meV). Binding energy between graphene and SiO₂ (0001) substrate is in range (-85 to -72.5 meV) for the other types of surface. Optimized structures of O_{unsat}, O_{sat}, Si_{unsat} and O_{sat} with adsorbed graphene sheet are shown on Figure 2. The distances between the graphene layer and the surface of the substrate range from 3.31 to 4.32 Å, which corresponds to physical adsorption with a strong influence of the Van der Waals forces.

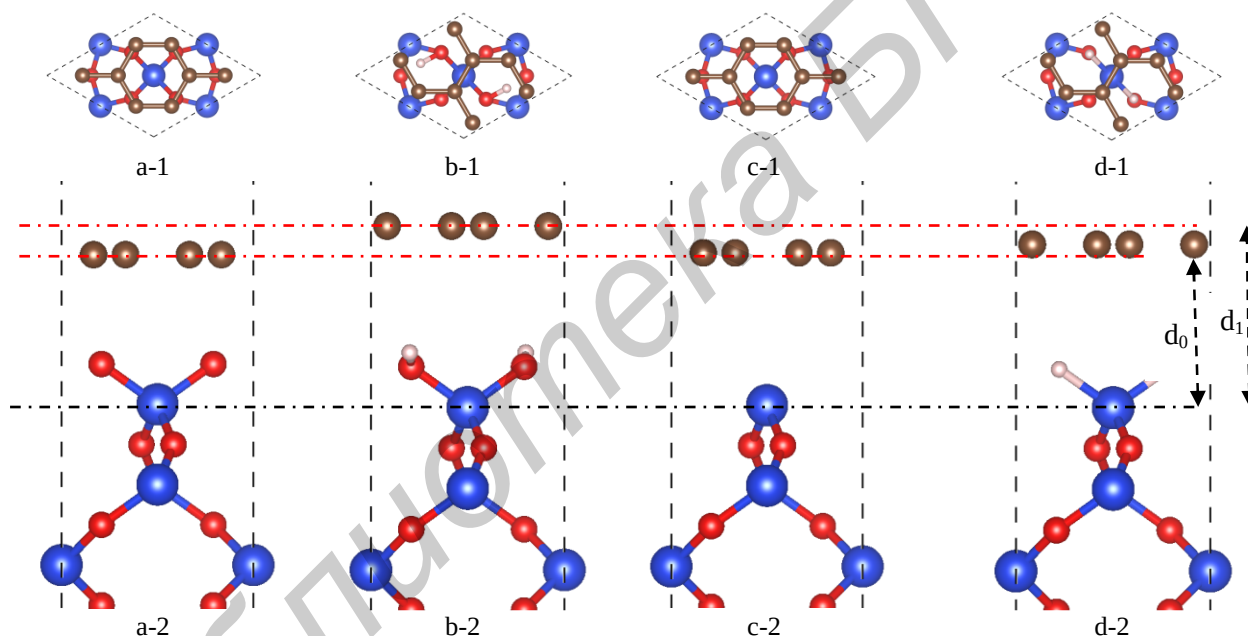


Figure 2 – Optimized structures of O_{unsat}, O_{sat}, Si_{unsat} and Si_{sat} with adsorbed graphene sheet

The band structures were calculated for the quartz substrate O_{unsat}, graphene and graphene on quartz. Changes in the states of graphene under the action of a substrate are observed in the immediate vicinity of the Fermi level. Energy gap arises width from 20 to 75 meV between the bonding and antibonding π -zones of graphene (Figure 3).

IV. CONCLUSIONS

Ground state of graphene position on the obtained SiO₂ surfaces has been determined. Interlayer distances between graphene and different types of quartz substrate have been calculated and are from 3.31 to 4.32 Å.

The energy spectrum of graphene on substrate near the Fermi level is linear. The Fermi level shifts toward to the valence band, which indicates a small redistribution of charge on the substrate. Overflow of charge occurs on near-surface oxygen atoms (0.07 e/atom). The binding energy from 20 to 80 meV for the physisorption of graphene sheet on a quartz substrate. Electronic structure of adsorbed graphene has no changes.

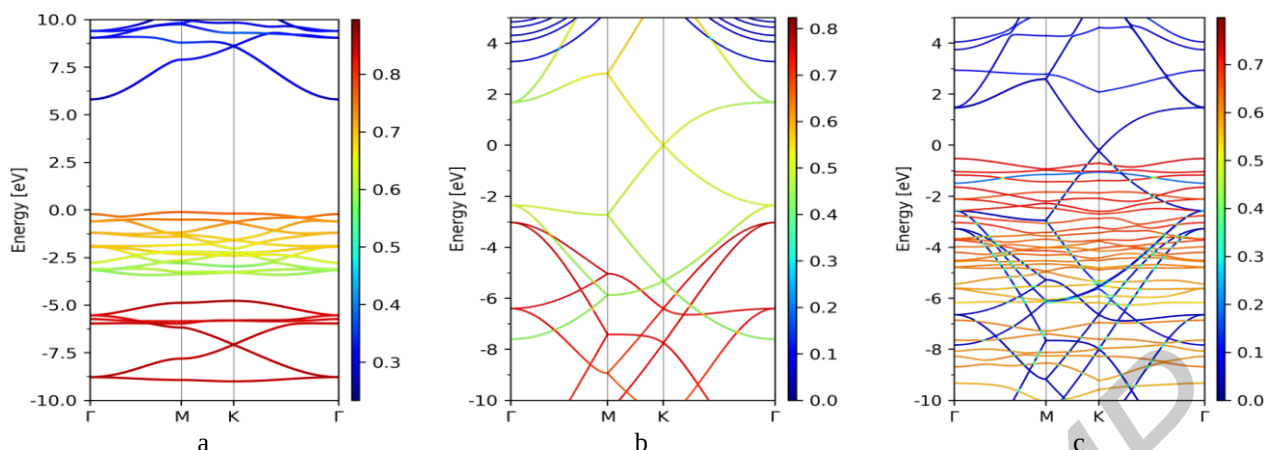


Figure 3 – Band structure of quartz (a), graphene (b) and graphene on quartz substrate (c)

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