

# The formation of atomic nanoclusters on graphene sheets

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## Abstract

The formation of atomic nanoclusters on suspended graphene sheets has been investigated by employing a molecular dynamics simulation at finite temperature. Our systematic study is based on temperature-dependent molecular dynamics simulations of some transition and alkali atoms on suspended graphene sheets. We find that the transition atoms aggregate and make various size nanoclusters distributed randomly on graphene surfaces. We also report that most alkali atoms make one atomic layer on graphene sheets. Interestingly, the potassium atoms almost deposit regularly on the surface at low temperature. We expect from this behavior that the electrical conductivity of a suspended graphene doped by potassium atoms would be much higher than in the case doped by the other atoms at low temperature.

(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

Graphene is a newly realized two-dimensional electron system [1, 2] which has produced a great deal of interest because of the new physics which it exhibits and because of its potential as a new material for electronic technology. The agent responsible for many of the interesting electronic properties of graphene sheets is the non-Bravais honeycomb-lattice arrangement of carbon atoms, which leads to a gapless semiconductor with valence and conduction  $\pi$ -bands. States near the Fermi energy of a graphene sheet are described by a massless Dirac equation which has chiral band states in which the honeycomb-sublattice pseudospin is aligned either parallel to or opposite to the envelope function momentum [3]. The linear energy–momentum dispersion has been confirmed by recent observations [4, 5]. There are significant efforts to grow graphene epitaxially [6] by thermal decomposition of silicon carbide (SiC), deposition of graphene sheets on solid [7] or by vapor deposition of hydrocarbons on catalytic metallic surfaces which could later be etched away leaving graphene on an insulating substrate.

An unusual feature of the single-atom-thick layer of carbon atoms is the absence of strong localization [8, 9] when charge impurities, short range ripples and surface roughness exist on graphene sheets. The issue of localization in graphene has recently attracted some attention and the chiral nature of the electron behavior has been discussed in the literature [10, 11]. Suzuura and Ando [10] claimed that the quantum correction to the conductivity in graphene can differ from what is observed in a normal two-dimensional electron gas due to the nature of elastic scattering in graphene. This is possibly because of changing the sign of the localization correction and turning weak localization into weak antilocalization for the region when the intervalley scattering time is much larger than the phase coherence time. Further consideration of the behavior of the quantum correction to the conductivity in graphene [11] concluded that this behavior is entirely suppressed due to time-reversal symmetry breaking of electronic states around each degenerate valley.

Another special feature of graphene is the capacity for using as a gas sensor because of the special characteristics of graphene [12–16]. Graphene-based gas sensors enable sensitivity to detect individual events when a gas molecule is absorbed by a graphene sheet. The absorbed molecules change

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the local carrier concentration in graphene. Constant mobility of charge carriers in graphene with increasing chemical doping of NO<sub>2</sub> has been observed in experiments [12]. Note that doping molecules add some charge carriers but also induce charged impurities. The latter effect results in a decreasing of the mobility. Different possible scenarios have been discussed to prove the constant mobility of charge carriers [12].

Chen *et al* [17] have reported a systematic theoretical and experimental study of the charged impurities mechanism by monitoring a reduction of the charge carrier mobility. The density of the charged impurities is induced by adding potassium atoms onto a graphene surface placed on an SiO<sub>2</sub>/Si substrate in ultrahigh vacuum at low temperature. They have reported that the addition of charged impurities produces a more linear behavior of conductance, reduces the mobility and moreover indicates that the minimum conductance depends on the density of charged impurities.

In addition, the enhancement of electrical conductivity in single-wall carbon nanotubes which are doped by alkali atoms has been studied both experimentally and theoretically [18–20]. Gao *et al* [20] carried out a molecular dynamics simulation to find the optimum structure of the doped potassium atoms for different numbers of potassium atoms in two different packing schemes.

A series of measurements on suspended graphene have been performed by experimental groups [21, 22] and showed that graphene has an extraordinary stiffness which can support an additional weight of many crystalline copper nanoparticles. The results from an STEM micrograph of graphene sheets incorporating copper atoms showed an aggregation shape that nanoparticles with different sizes distributed on graphene randomly. On the other hand, the pinning of size-selected gold and nickel atoms on graphite has been investigated [23] by using the MD simulation and it is shown that the atoms aggregated on the surface and gold clusters are shown to be flatter and more spread out than the nickel cluster, which is more compact. Moreover, they calculated the pinning energy thresholds and showed that there is a good agreement with those measured in experiments [24].

Particle aggregation in materials science is a direct consequence of mutual attraction between particles, atoms or molecules via van der Waals forces or chemical bonding [25]. When there are collisions between particles, there are chances that particles will attach to each other and become a larger particle. There are three major physical mechanisms to form aggregates: Brownian motion, fluid shear or motion, and differential settling. Because of interparticle forces, not all collisions may be successful in producing aggregates. If there is strong repulsion between particles then practically no collision gives an aggregate. It is known that collisions brought about by Brownian motion do not generally lead to the rapid formation of very large aggregates. In addition, differential settling becomes more important when the particles are large and dense and this mechanism can promoting aggregation.

In general, there are two main methods for investigating the physical behaviors of the absorbed particles on a surface: a Monte Carlo method, which is a sophisticated approach, while the other one is a molecular dynamics simulation.

The purpose of this paper is the study of deposition and aggregation of atoms on graphene sheets by using molecular dynamics simulations. Eventually, we generally report that the potassium atoms arrange regularly and deposit on a suspended graphene sheet at low temperature. This is evidence that the electrical conductivity of the doped potassium atoms on a suspended graphene sheet should be higher than the electrical conductivity of other examined atoms.

It is worthwhile to note that, since adding atoms are placed in the setback distance from the graphene sheet, the effect of charged impurities can be negligible for the suspended graphene due to large screening effects. There are at least two reasons to explain why the effect of charge impurities should be important in graphene placed on a substrate. The first effect comes from the fact that the averaged background dielectric constant value of graphene placed on a substrate is more or less three times larger than the vacuum dielectric constant. The second effect is related to the image that the charged impurity [26] induces in another side of the dielectric substrate when there is a charged impurity above graphene. In the latter case, an induced electric dipole in the system increases the effect of electron scattering. We consequently expect that the difference between the carrier mobility for a suspended graphene and a graphene placed on a substrate incorporating both the potassium atoms at low temperature.

The contents of this paper are described briefly as follows. In section 2, we introduce the models and theory. Section 3 contains our numerical calculations. Finally, we conclude in section 4 with a brief summary.

## 2. Model and theory

We have used the empirical inter-atomic interaction potential, carbon–carbon interaction in graphite [27], which contains a three-body interaction for molecular dynamics simulation of a suspended graphene sheet at finite temperature. The two-body potential gives a description of the formation of a chemical bond between two atoms. Moreover, the three-body potential favors structures in which the angle between two bonds is made by the same atom. Many-body effects of the electron system, on average, are considered in the Brenner potential [27] through the bond order and, furthermore, the potential depends on the local environment.

We have considered a graphene sheet including  $280 \times 160$  atoms with periodic boundary condition. The area of a graphene sheet is  $A_0 = 1173.5 \text{ nm}^2$ . Considering the canonical ensemble ( $NVT$ ), we have employed a Nosé–Hoover thermostat to control temperature. Our simulation time step is 1 fs in all cases and the thermostat's parameter is 5 fs. Therefore, we have found a stable two-dimensional graphene sheet in our simulation.

First of all, we have simulated the stable graphene sheet at finite temperature, and then extra atoms have been distributed randomly on the graphene. The number of adatoms is equal to 2500 and distributed in a portion of area,  $A = \frac{4}{9}A_0$  in order to avoid boundary effects. We let the system achieve its equilibrium condition where the atoms above the surface are mostly fluctuating around their equilibrium positions. At

**Table 1.** Physical parameters and Lennard-Jones 12 – 6 parameters for some atoms and molecules.

Element	$\sigma$ (Å)	$\varepsilon$ (eV)	Atomic mass (g mol <sup>-1</sup> )
Cu	2.338	0.409 33	63.546
Ag	2.644	0.3447	107.8682
Au	2.637	0.441 47	196.96
Li	2.839	0.2053	6.941
Na	3.475	0.1378	22.989 77
K	4.285	0.114 44	39.09
C	3.369	0.002 63	12.01
Cu–C	2.853	0.0328	—
Ag–C	3.006	0.0301	—
Au–C	3.003	0.0341	—
Li–C	3.104	0.0232	—
Na–C	3.422	0.0190	—
K–C	3.827	0.0173	—

the beginning of the simulation, the atoms are initially located on a height equal to 3.65 Å above the graphene sheet. To further proceed, we do need to implement the interparticle interactions. We have used van der Waals potentials between atom–atom and carbon–atom denoted by X–X and C–X, respectively. The parameters of two-component interactions between two types of atoms can be estimated by simple average expressions proposed by Steel *et al* [28]:

$$\sigma_{C-X} = \frac{\sigma_{X-X} + \sigma_{C-C}}{2} \quad (1)$$

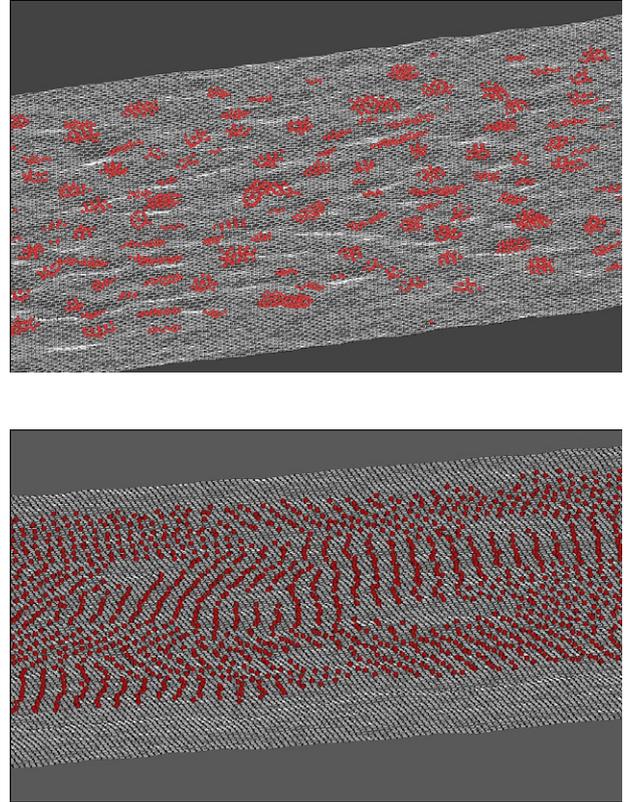
$$\varepsilon_{C-X} = \sqrt{\varepsilon_{X-X} \cdot \varepsilon_{C-C}},$$

where  $\sigma_{i-j}$  is the collision diameter and  $\varepsilon_{i-j}$  is the depth of the primary energy well between  $i$ th and  $j$ th atoms. The parameters which have been used in our simulations are given in table 1, adopted from [29]. It is worthwhile to note that our final results are independent of the explicit values of parameters appearing in van der Waals potentials. We have used a new set of parameters [30–32] and our final results did not change.

### 3. Numerical results

In this section we present our numerical results based on the method described above. To this purpose, we have considered a system incorporating the transition atoms like copper, silver and gold atoms or the alkali atoms like lithium, sodium and potassium atoms on graphene sheets. Note that the gravitational force is very small in comparison to interparticle forces and tends to go unnoticed. Our main results have been summarized in figure 1. The copper atoms (figure 1(a)) are aggregated and the potassium atoms are deposited (figure 1(b)) on graphene sheets at  $T = 50$  K.

Here, a graphene sheet plays the role of a substrate for external absorbed atoms. Figure 2 shows three snapshots of distributed nanoclusters on graphene at  $T = 50$  K. The transition atoms show aggregation configurations producing different nanocluster sizes. Apparently each nanocluster trapped around the height is out-of-plane because of graphene roughness and preferably made a nanocluster. Furthermore,

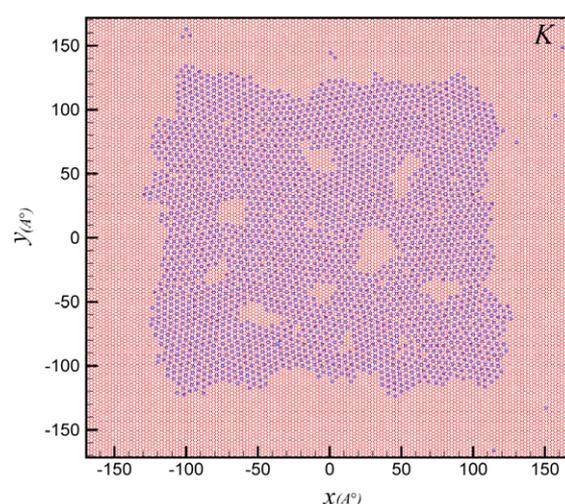
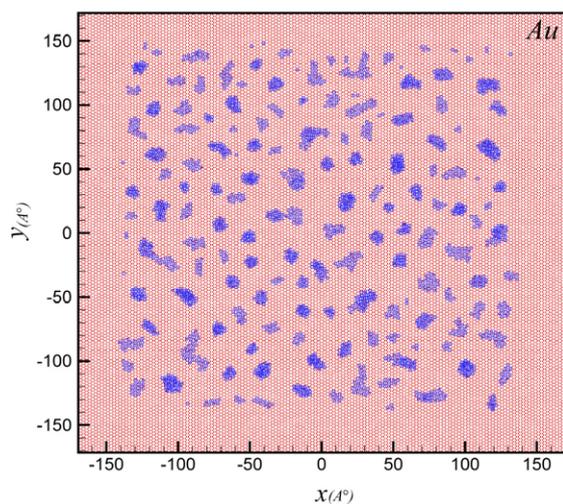
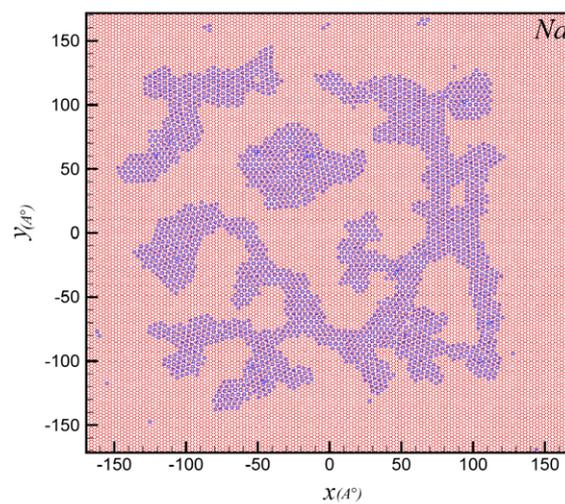
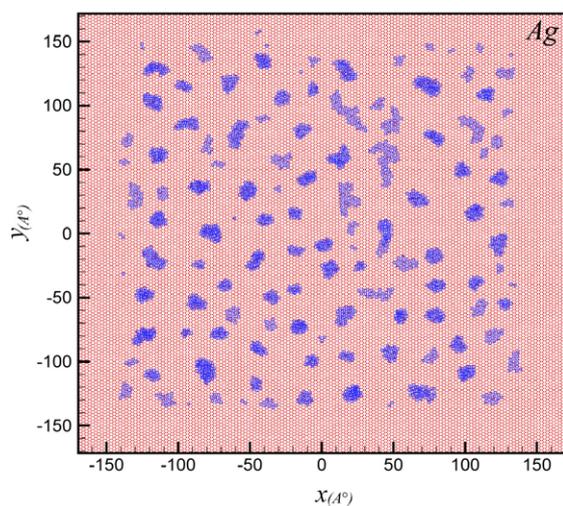
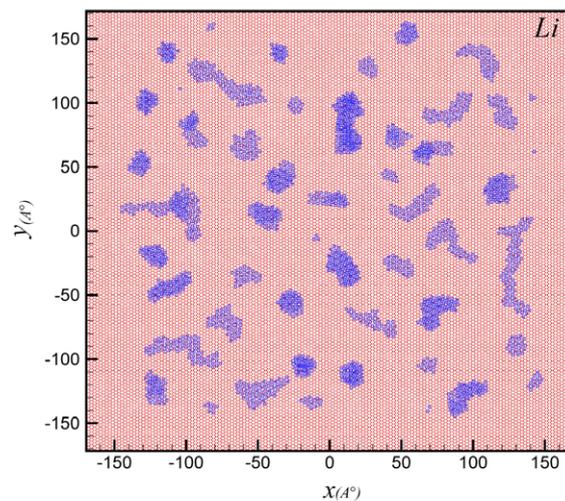
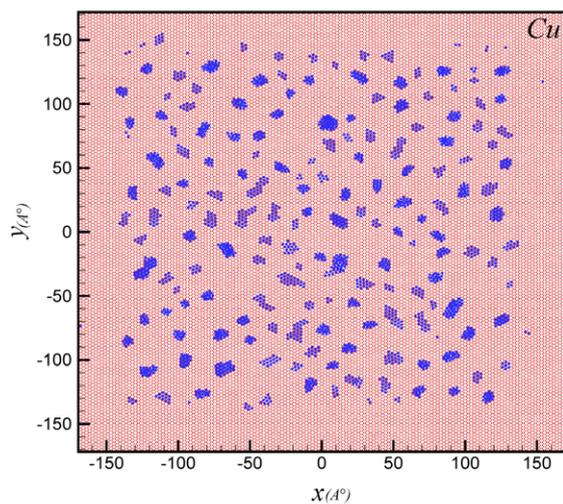


**Figure 1.** Distribution of copper atoms (top) and potassium atoms (bottom) on graphene sheets at  $T = 50$  K. The copper and potassium atoms are denoted by red spheres and graphene sheets are denoted by white spheres as a background. The number of atoms added on graphene is 2500 for both cases.

since  $\varepsilon_{Cu-C}/\varepsilon_{Cu-Cu}$  is smaller than  $\varepsilon_{Au-C}/\varepsilon_{Au-Au}$ , the copper atoms thus distribute laterally on the surface much more than the gold atoms. In this case, the gold atoms prefer to grow more vertically in comparison to the copper atoms. Our numerical simulations confirm such behavior. Furthermore, our finding for the copper atoms is quite similar to that observed from STEM micrographs of graphene sheets incorporating copper atoms by an experimental group [21]. To be sure about the independence of interparticle interaction potentials, we have examined the Morse potential [29] and we get entirely the same results.

We have seen, on the other hand, new physics by adding the alkali atoms on graphene sheets at  $T = 50$  K where the results are depicted in figure 3. The lithium atoms form a few layers of nanoclusters on graphene apparently having regular shapes. Importantly, by using sodium atoms, a layer an atom thick is arranged above the surface. This layer shows a sort of percolation configuration. Surprisingly, the potassium atoms arranged regularly and noticeably wet the graphene's surface. There are some small vacant islands where the potassium atoms could not occupy because they are bounded locally by bumps on the graphene surface. For graphene the doping is usually realized by surface transfer doping.

The electronic properties that result from absorption depend on the ionic and covalent character of the bonds formed between carbon and the metal atoms. As alkali atoms easily



**Figure 2.** Aggregation of transition atoms on graphene sheets at  $T = 50$  K. Nanoclusters are distributed randomly above the surface.

**Figure 3.** Distribution of alkali atoms on graphene sheets at  $T = 50$  K. Potassium atoms deposited on graphene.

release their valence electron, they may effectively induce n-type doping [5]. Since the alkali atoms play the role as a cation, they can give electrons to the system easily. The amount of charge transfer to graphene by potassium atoms is

a challenging problem and has been studied for many years in potassium-deposited graphite surfaces [33]. The charge transfer between a paramagnetic molecule  $\text{NO}_2$  and a graphene layer has been recently calculated [34] and a charge transfer

of  $1e$  per molecule is found. Furthermore, the effect of potassium doping on the electrical properties of graphene by using density-functional theory (DFT +  $U$ ) has been recently studied [35]. They predicted a charge transfer of  $0.51e$  per potassium atom at  $U = 0$ . As  $U$  increases, the charge transfer is linearly suppressed.

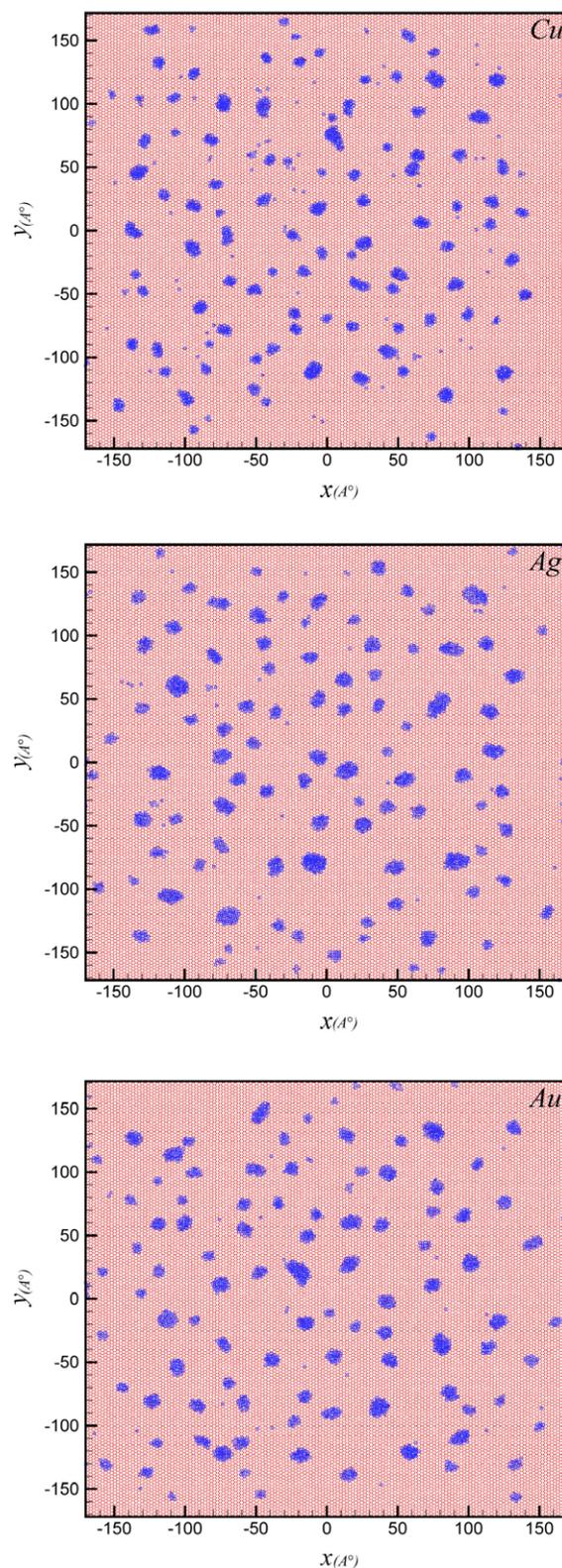
The regular arrangement distributions of potassium (K) atoms on top of the surface indicate that the value of the opening gap due to breaking symmetry would be much smaller than those values induced by the other metallic adatoms. Moreover, it is shown [36] that the conductance of charge carriers decreases by increasing the value of the gap. Therefore, the K atoms give rise to an increase in electrical conductivity: however, the transition atoms evidently play a role as the central scatterers and result in decreasing the electrical conductivity of the system. Note that the effect of charged impurities from chemical doping are negligible for suspended graphene [13]. Our results regarding the regular arrangement of K atoms on graphene are similar to the alkali atoms doped on the single-wall carbon nanotube [20].

It is well known [25] that, for the alkali metal cations, the critical coagulation concentration values which are estimated by assuming that no energy barrier exists, decreases in the order  $\text{Li}^+ > \text{Na}^+ > \text{K}^+$ , showing that the most hydrated ion ( $\text{Li}^+$ ) is the least effective in reducing the repulsion term in particle–particle interactions. Consequently, the K atoms deposit easily on the surface with respect to Li and Na atoms.

We expect physically that the atoms escape from graphene sheets when the temperature increases because of increasing the kinetic energy of atoms. The number and sizes of nanoclusters decrease by increasing temperature up to  $T = 300$  K and at higher temperature atoms escape from the surface. In figure 4, we have shown the aggregation behavior of transition atoms at room temperature. The number of nanoclusters decreases by increasing temperature. Similar to the results shown in figure 2, the nanoclusters have no regular crystalline configuration at higher temperature and moreover the distribution of potassium atoms changes and they show a percolation on top of graphene sheets as shown in figure 5.

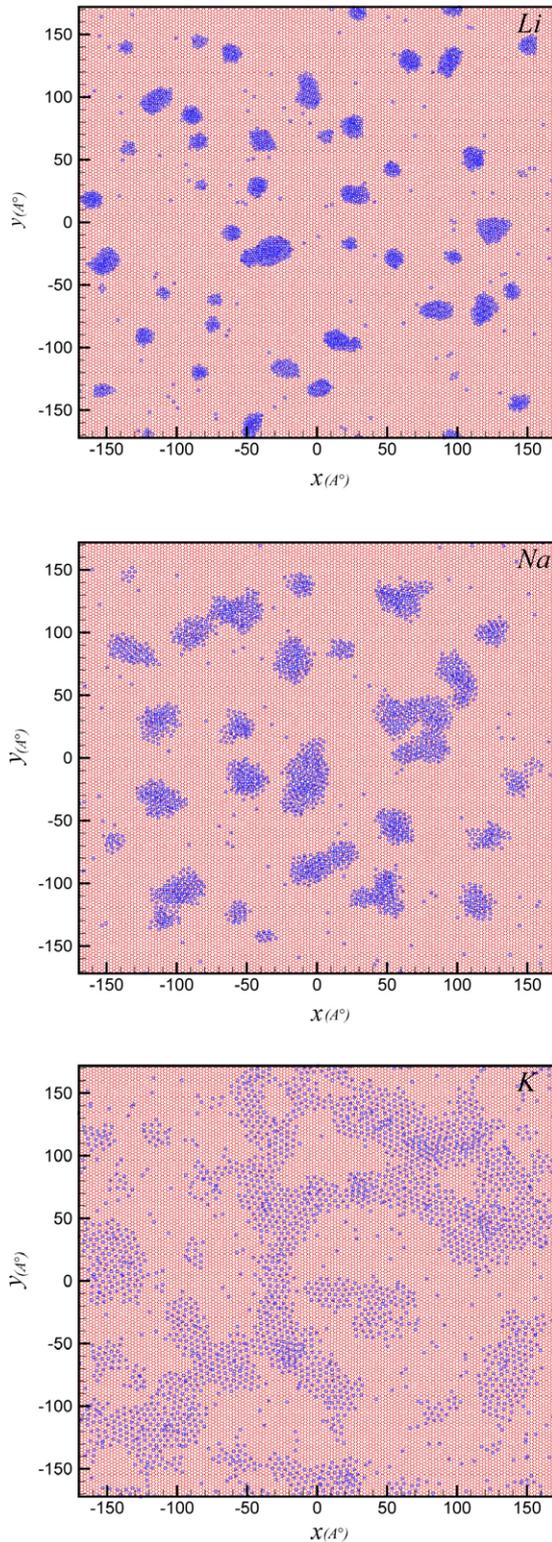
For estimating the typical size of nanoclusters at different temperatures, we have calculated the density number of copper atoms,  $P(N)$ , as a function of  $N$ , where  $N$  is the number of copper atoms in a nanocluster as depicted in figure 6. The number of copper atoms in each nanocluster decreases by increasing temperature and some of the atoms escape from the surface.

One of the interesting subjects in studying alkali metal absorption on graphite is the phase transition in the structural shapes of the overlayer [33]. The most studied alkali metal on graphite is potassium. A common convention for overlayer structure is a  $2 \times 2$  phase in which eight carbon atoms are surrounded by one K atom [33], which is the most condensed phase of alkali atom absorption on the graphite sheet at a temperature lower than room temperature. There are other less condensed structure such as a  $7 \times 7$  phase where 98 carbon atoms are surrounded by one K atom. Two relevant parameters for change in the coverage type in the graphite sheet are temperature and the density of adatoms. Increasing



**Figure 4.** Aggregation of transition atoms on graphene sheets at  $T = 300$  K.

the number of K atoms on the graphite at low temperatures ( $\sim 90$  K), close-packed islands ( $2 \times 2$ ) cover the entire surfaces but also it may be the existence of the other low condensed phase. After saturation of K atoms over the graphene it starts to



**Figure 5.** Distribution of alkali atoms on graphene sheets at  $T = 300$  K.

make other layers over the first one [33]. In the case of Li atoms most observations indicate the intercalation into the graphite even at 100 K [37]. For Na atoms growing some multilayered non-ordered islands over each other have both experimentally and computationally been observed [38].

In our calculation, quite interestingly, the atomic pattern ( $2 \times 2$ ), one K atom per eight carbons, for an overlayer in high density for K atoms and a snapshot of the structure is shown in figure 7. This pattern is similar to that predicted for K atoms on graphite. In the low density, on the other hand, we did not observe low density phases ( $7 \times 7$ ) whereas small islands of the  $2 \times 2$  structure have been observed. The absence of low density structures might be understood from the dominance of thermal vibration which is larger in a strictly two-dimensional system with respect to three-dimensional structure in graphite.

Furthermore, the type of ordering of sodium atoms on the  $C_{60}$  molecules has been investigated by Roques *et al* [39]. Their results showed that the ordering of deposited atoms is temperature- and concentration-dependent. They found that for up to eight atoms on the surface of a  $C_{60}$  molecule there is no homogeneous deposition and they begin to form nanoclusters on the surface. This is because of saturation of charge transfer from alkali atoms to the substrate when the substrate has not enough surface (limiting by curvature) for accepting other charges. In comparison with our calculations, crudely deducing, a graphene sheet with the very large radius of a carbon cage has enough available surface for adding more alkali atoms and those alkali atoms enable us to wet the surface more and more.

We are also interested in calculating the out-of-plane carbon atoms in the presence of external atoms above graphene. The common procedure for measuring the roughness exponents of a rough surface is to use a surface structure function [40, 41]:

$$S_x(\delta) = \langle |h(x + \delta, y) - h(x, y)|^2 \rangle, \quad (2)$$

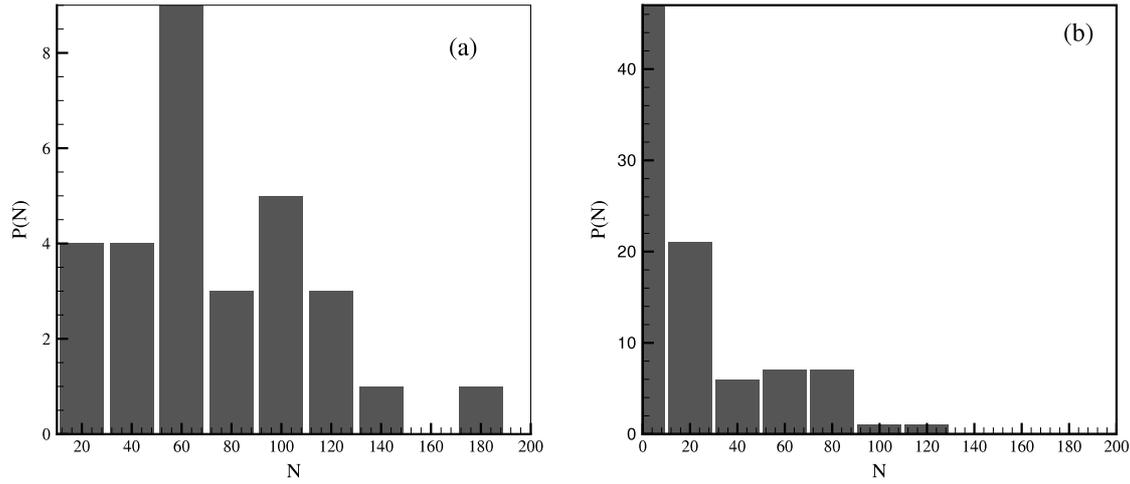
where the average is taken over some different  $y$  values. The variation of  $S_x(\delta)$  for different systems at  $T = 50$  K is shown in figure 8. The characteristic length is defined by the position that curves are bent. From our finding, the characteristic length in all cases is less than the case of graphene without absorbed atoms. Moreover, the characteristic length of the system by using the transition atoms are smaller than when the alkali atoms are used. The reason is as follows: the nanoclusters made by transition atoms distributed randomly over the graphene sheets increase the surface randomness. The  $S_x(\delta)$  referring to the situation where the potassium atoms are added on graphene is very similar to the one in which graphene is itself alone because correlation between the potassium atoms and the carbon atoms is strong at low temperature. Consequently, this sort of atom added on graphene does not change the morphology of the surface at low temperature.

Another interesting quantity is normal-normal correlation for a surface. The normal-normal correlation in the  $x$  direction is defined as

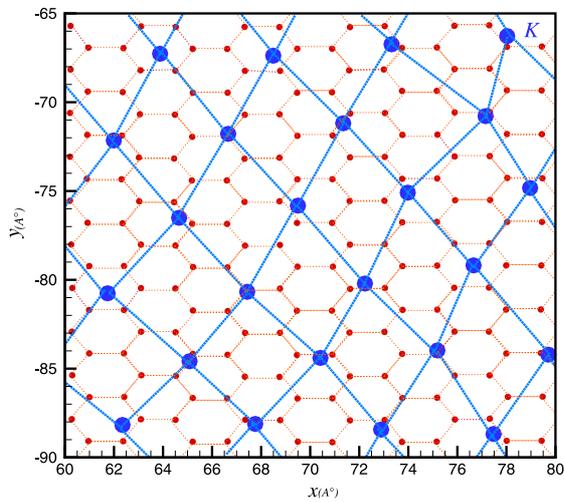
$$c(\delta, T) = \langle \hat{n}(x, y) \cdot \hat{n}(x + \delta, y) \rangle. \quad (3)$$

The persistence length,  $l_p(T)$ , which is a criterion of surface stiffness, is expressed as follows [42]:

$$l_p(T) = \frac{\delta}{\ln c(0, T) - \ln c(\delta, T)}. \quad (4)$$



**Figure 6.** Density number of copper atoms as a function of the number of atoms at  $T = 50$  K (a) and  $T = 300$  K (b).



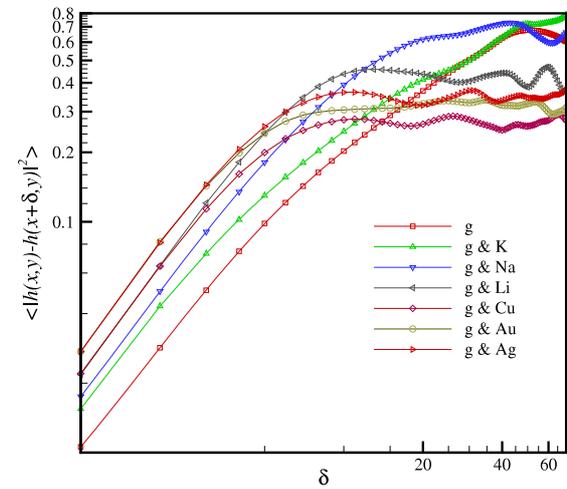
**Figure 7.** Pattern of potassium atoms distribution ( $2 \times 2$ ) on a graphene sheet at  $T = 50$  K. The large circles represent the potassium atoms and the small dots represent the C atoms.

A surface is rigid in a length smaller than the persistence length and it behaves as a soft membrane for longer length. To calculate the adatom dependence of persistence length of graphene sheets, we have calculated this parameter for different atoms used at three temperatures which are listed in table 2. Moreover, by using the Morse potential for the transition atoms, the same persistence length values have been obtained.

As is clear from table 2, the persistence length decreases by incorporating atoms and importantly the persistence length of potassium atoms is close to the  $l_p$  of graphene itself. In all cases,  $l_p$  decreases by increasing temperature.

#### 4. Conclusion

We have studied the formation of atomic nanoclusters on suspended graphene sheets by using a molecular dynamics simulation at finite temperature. We have used the model



**Figure 8.** Surface structure function,  $S_x(\delta)$ , as a function of  $\delta$  at  $T = 50$  K. Graphene is denoted by g.

**Table 2.** Persistence length of graphene (denoted by g) and the presence of adatoms on graphene at different temperatures.

Element	$l_p$ ( $\text{\AA}$ ) at $T = 50$ K	$l_p$ ( $\text{\AA}$ ) at $T = 200$ K	$l_p$ ( $\text{\AA}$ ) at $T = 300$ K
g and Cu	88.1	60.0	52.0
g and Ag	82.5	58.0	52.0
g and Au	87.2	58.5	52.1
g and Li	95.3	60.0	54.3
g and Na	94.5	60.5	53.5
g and K	102.1	67.2	54.0
g	112.0	69.0	55.5

of interparticle potentials: the Brenner potential for carbon-carbon interactions and van der Waals model potentials for the interactions between atoms and the carbon atoms. We have shown that the transition atoms aggregated on the surface with different nanocluster sizes. However, the sodium and the potassium atoms produced one atomic layer on graphene. Interestingly, the potassium atoms were arranged regularly on

graphene sheets at low temperature and it indicates that the value of the opening gap due to breaking symmetry would be much smaller than those values induced by the other metallic adatoms.

As a consequence, the charge carriers' electrical conductivity of a suspended graphene doped by potassium atoms would be higher than a suspended graphene doped by other atoms. We qualitatively expect that the electrical conductivity of the system would increase by adding potassium atoms on suspended graphene sheets at low temperature. Note that, since the induced charged impurities are set far from the suspended graphene sheet, the effect of charged impurities can be ignored due to large screening effects. Our finding at low temperature would be verified by experiments.

We remark that a model going beyond the molecular dynamics simulation is necessary to account quantitatively for the effect of potassium atoms on electrical conductivity for a suspended graphene sheet at low temperature. One approach would be the density-functional theory together with the molecular dynamics simulation, where the transport properties of charge carriers in the presence of extra atoms are considered.

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