https://doi.org/10.22226/2410-3535-2020-4-491-495 PACS: 81.05.ue; 61.48.+c; 07.05.Tp

The effect of the occurrence of a magnetic field of a current loop in hybrid graphene/C_{60} carbon systems

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In this article, using *in silico* methods we investigate the simultaneous effect of temperature, external electric field, and structural defects of graphene atomic network on the patterns of positioning of the C_{60} molecule on a graphene sheet. The conditions for the appearance of a circular current on graphene during the ordered motion of a charged C_{60} fullerene controlled by regularly adsorbed hydrogen atoms and external electric field are found. Numerical experiments are carried out using the molecular dynamics and the self-consistent charge density functional tight-binding (SCC-DFTB) method. It is found that at a temperature of 175 K and an electric field strength of $1 \cdot 10^6$ V/m, the circular current value is ~10.4 nA. The magnetic field induced by the circular current is 6.2 μT. It is shown that the appearance of a circular current is caused by two most important factors: the arrangement of hydrogen atoms on graphene and the temperature. A defect in the form of chemically adsorbed hydrogen atoms should form a bowl-shaped well in the energy surface of interaction of C_{60} with grapheme, along the edges of which the fullerene will make a circular motion. As a way to amplify the circular current and induced magnetic field, it is proposed to vary the size of the local hydrogenation region of graphene. The external electric field directs the charged fullerene C_{60} to the defect region. It is predicted that the detected physical effect can be the basis for operation of new miniature magnetic field sources for various nanodevices.

Keywords: graphene/C_{60} hybrid system, magnetic field of a circular current, local hydrogenation, external electric field, density-functional tight-binding method.

УДК: 537.9; 004.942

Эффект возникновения магнитного поля кругового тока в гибридных углеродных системах графен/C_{60}

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В данной работе методами *in silico* исследуется одновременное влияние температуры, внешнего электрического поля и структурных дефектов атомной сетки графена на закономерности позиционирования молекулы C_{60} на листе графена. Обнаружены условия появления кругового тока на графене при упорядоченном движении заряженного фуллерена C_{60}, контролируемом с помощью регулярно адсорбированных атомов водорода и внешнего электрического поля. Численные эксперименты проводились с использованием метода молекулярной динамики и метода функционала плотности в приближении сильной связи с самосогласованным определением атомных зарядов (SCC-DFTB). Установлено, что при температуре 175 К и напряженности внешнего электрического поля $1 \cdot 10^6$ В/м величина кругового тока составляет ~10.4 нА. Индукция магнитного поля, индуцированного круговым током, составляет 6.2 мкТ. Показано, что двумя наиболее важными факторами в появлении кругового тока являются расположение атомов водорода на графене и температура. Дефект в виде химически адсорбирующихся атомов водорода должен сформировать в энергической поверхности взаимодействия молекулы C_{60} с графеном яму чашеобразной формы, по краям которой фуллерен будет совершать круговое движение. В качестве способа усиления кругового тока и индуцированного им магнитного поля предлагается варьировать размерами области локального наводораживания графеновой атомной сетки. Внешнее электрическое поле позволяет доставить заряженный фуллерен C_{60} к области расположения дефекта. Прогнозируется, что обнаруженный физический эффект может быть положен в основу физического принципа работы новых миниатюрных источников магнитного поля для различных наноустройств.

Ключевые слова: гибридная система графен/C_{60}, магнитное поле кругового тока, локальное наводораживание, внешнее электрическое поле, метод функционала плотности в приближении сильной связи.

1. Introduction

Currently, hybrid nanostructures, which are a combination of graphene with other nanoscale materials, including semiconductor nanoparticles, attracted great attention of researchers [1]. In particular, hybrid carbon nanosystems consisting of C_{60} fullerene films deposited on monolayer graphene have wide application prospect in many fields [1–6]. Various devices of molecular electronics and nanoelectronics have been proposed based on graphene/C_{60} films, including mid-infrared photodetector [2], ultra-thin, flexible solar cells [3], vertical field-effect transistors [4, 5], supercapacitors [6]. In a real experiment, the deposition of C_{60} films on a graphene substrate is carried out using the well-proven thermal evaporation method [7, 8] or spray coating method [5, 6]. Investigations of the properties of this hybrid material are intensively developing at the present. Much attention is paid to studying the electronic structure of graphene/C_{60} system [9–11], as well as the charge transfer between C_{60} atoms and graphene atoms during the interaction of two objects [12–15]. It was found that in the graphene/C_{60} system, the C_{60} molecule functioned as an electron acceptor affording hole doping of graphene [13]. External gating can change the C_{60} from an electron acceptor to an electron donor [15]. Another promising research topic is the study of the regularities in the positioning and behavior of the C_{60} molecule on graphene from the position of the discovery of new physical effects. In particular, some papers are devoted to the study of the behavior of the C_{60} fullerene on graphene at various temperatures [16–20]. These papers have shown that at low temperatures the chaotic movement of C_{60} is replaced by ordered motion, whose trajectory is determined by the graphene topology and the magnitude of temperature.

In this work, the simultaneous effect of temperature, external electric field, and structural defects of the graphene atomic network on the regularities in the positioning of the C_{60} molecule on graphene is first studied.

2. Computational details

The optimum ground state geometry of graphene/C_{60} system was calculated using the SCC-DFTB method [21]. The dynamic behavior of the C_{60} molecule on graphene was simulated using the classical molecular dynamics [22] in combination with the SCC-DFTB method. To take into account the external electric field imparted additional energy to the charged C_{60} molecule, the force generated by the electric field was added to the equation of motion. The SCC-DFTB method allows us to take into account the charge transfer between the atoms of C_{60} molecule and graphene during the simulation. The Verlet algorithm was used to integrate the equations of motion [23]. Step of numerical integration was equal to 1 fs. The temperature was kept constant at 300 K in simulation using the Berendsen thermostat [24]. All simulations were performed using the Kvazar-Mizar package [25].

3. Results and discussion

The object of this study was a graphene sheet with a length of 30 Å along the zigzag edge and 33 Å along the armchair edge. As a way to manipulate fullerene on a graphene substrate, we propose to modify the atomic structure of graphene in such a way as to make the movement of the C_{60} molecule controllable. In search of the optimal structural modification of the graphene sheet providing the desired effect, we considered the case of local hydrogenation of graphene by a group of 19 hydrogen atoms chemically adsorbed in the central region of the graphene atomic network. The creation of such defect led to a deflection of the graphene atomic network in the region of the location of hydrogen atoms, as shown in Fig. 1. The energy profile of the interaction between a C_{60} molecule and a graphene sheet for this case is also shown in Fig. 1. As can be seen, a convex region is observed in the energy profile in the center of the graphene sheet. The sizes of this region correspond to the sizes of the local hydrogenation region of graphene.

In order to identify the regularities in the dynamical behavior of a C_{60} molecule on a graphene substrate of this topology, we carried out a series of numerical experiments at temperatures ranging from 50 to 300 K. As a result of the MD calculations, it was found that achieving the local hydrogenation region, the fullerene starts to move just within this region due to the action of the repulsive energy barrier. At a certain temperature, the movement of fullerene within the specified region acquires a cyclical character. According to our calculations this effect appears at 160 K. Fig. 2 a shows the trajectory of the center of mass of a C_{60} molecule in this case. As can be seen, the motion of C_{60} has irregular character accompanied by multiple attempts to describe a loop form of trajectory. In order to stabilize the circular motion of fullerene within the local hydrogenation region, an external electric field with a strength of $1 \cdot 10^6$ V/m was applied. The charge $+1e$ was previously added to the C_{60} molecule by encapsulating the ion. The analysis of the calculated C_{60} trajectory showed that the motion of fullerene within the local hydrogenation region acquired a more ordered character due to the action of an electric field. Starting from the time point of 80 ps, the C_{60} makes a cyclic motion with some periodicity. It is important to note the role of the electric field direction. By setting the electric field vector along the Y axis we force C_{60} molecule to make cyclic rotation clockwise. The fragment of the loop-shaped trajectory of the C_{60} molecule within the local hydrogenation region of graphene atomic network (bowl-shaped region with a diameter of 9 Å) is shown in Fig. 2 b. The non-ideal shape of the loop can be caused by the

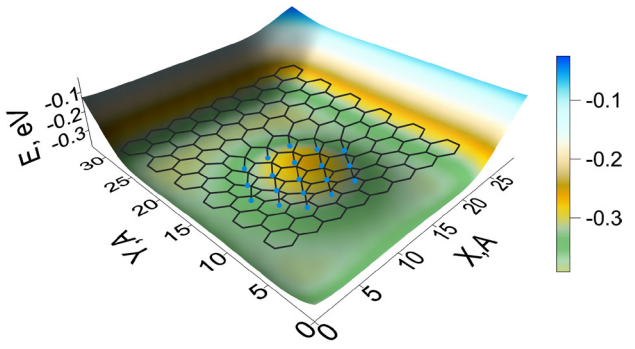
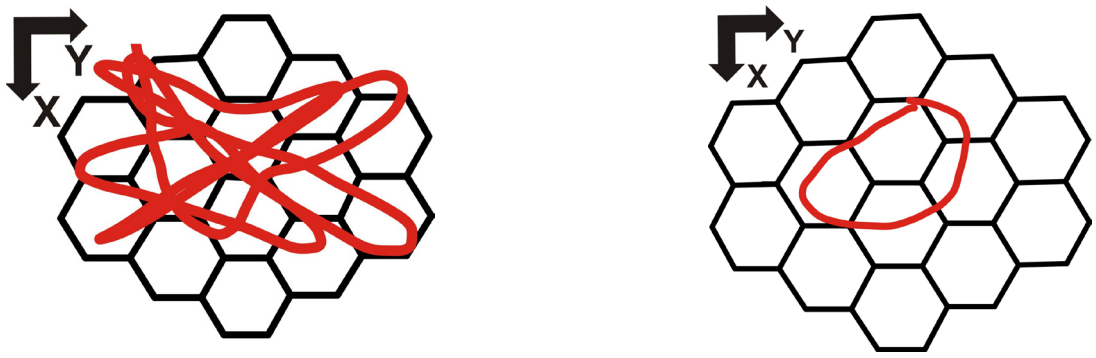


Fig. 1. (Color online) Atomic structure of the graphene/C_{60} hybrid system with a defect in the form of local hydrogenation of graphene by a group of 19 atoms and the energy profile of the interaction of C_{60} with graphene.



a b

Fig. 2. (Color online) A top view of the fragments of trajectory of the center of mass of a molecule C_{60} within the local hydrogenation region of graphene atomic network (bowl-shaped region with a diameter of 9 Å) at 160 K without electric field (a) and in the presence of electric field with a strength of $1 \cdot 10^6$ V/m (b).

topological features of the graphene atomic network within in this region, primarily, by the pronounced curvature.

Taking into account the cyclic character of the charged fullerene motion, one can draw a direct analogy with the motion of electron within an atom in circle orbits [26]. Such motion is equivalent to a closed current loop. Thus, it can be assumed that the phenomenon of current flow can be occurred during the C_{60} cyclic motion within the local hydrogenation region. Let us numerically estimate of the cyclic current induced in the graphene/C_{60} complex. The C_{60} fullerene moves along a closed trajectory which is an ellipse with major and minor semi-axes equal to $a = 3.75$ Å and $b = 2$ Å, respectively. The length of the ellipse is 18.48 Å; the area of the ellipse is 23.56 Å^2. The current I was calculated according to the known formula:

$$I = e\upsilon = e\nu / 2\pi r, \qquad (1)$$

where e is the electron charge, υ is the number of turns per second, ν is the average velocity of fullerene's center of mass, r is the radius of the circular orbit. Since the average velocity of the C_{60} molecule is ~65 m/s in considered case, the circular current is ~6 nA.

We have estimated the value of the magnetic induction B of a magnetic field generated by a closed current loop. This loop represented the closed elliptical trajectory for a charged C_{60} fullerene. The calculation of the magnetic induction was carried out using the well-known formula for the circular current:

$$B = \mu_0 IpL / 4\pi(p^2 + x^2)^{3/2}, \qquad (2)$$

where the focal parameter p of the ellipse is used instead of the radius of the circular radius, I is the circular current, L is the length of the ellipse, μ_0 is the vacuum permeability. The maximum value of the magnetic induction B is achieved in the center of an ellipse. The magnetic induction decreases when moving away from the current loop as shown in Fig. 3. At point O the magnetic induction B reaches a maximum value of 0.95 μT; further, when moving along the straight line OA, the magnetic induction weakens. The magnetic induction becomes zero at the distance of 6 Å away from an ellipse.

After determining the conditions for the occurrence of a circular current in the graphene/C_{60} system, it was necessary to find a way to increase its value. The results of the performed simulations have shown that an increase in the external field strength forced the C_{60} molecule to leave the circular orbit and, thereby, to stop the current flow. Therefore, there an idea occurred to solve the noted problem by varying the geometrical sizes of the bowl-shaped region of graphene atomic network by using a larger amount of chemically adsorbed hydrogen atoms on graphene. To test this idea, a series of numerical experiments was carried out with a local defect in the chemisorption of a group of 44 hydrogen atoms on graphene. This defect led to the formation of a bowl-shaped graphene region with a larger diameter of 14.6 Å. The energy profile of the interaction of C_{60} molecule and graphene for this case is shown in Fig. 4. This figure shows that the energy profile has undergone significant changes. A well with a depth of 1.8 eV is observed in the center of the energy profile, in contrast to the previous case with 19 hydrogen atoms, where a convex region was observed. Consequently, at a certain concentration of hydrogen atoms chemically adsorbed on graphene, the convex energy profile transforms into a profile with a potential well.

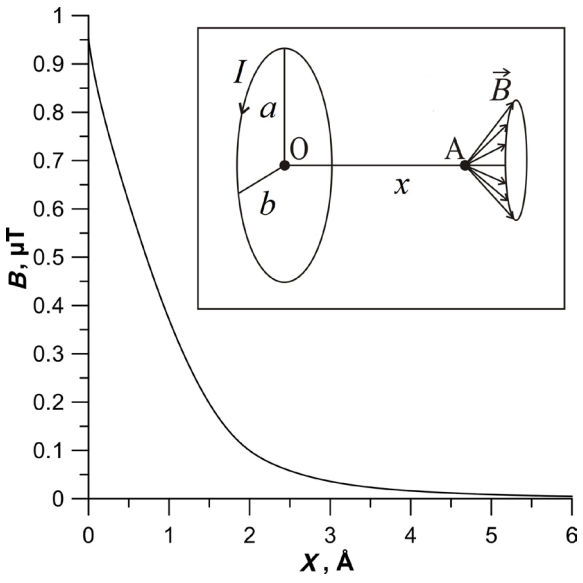


Fig. 3. Change in the magnetic induction B of a magnetic field generated by the current loop with distance X from the loop center in the case of a bowl-shaped graphene region with a diameter of 9 Å.

For the new case, we determined the temperature at which the trajectory of the C_{60} molecule close to the loop shape will be observed. According to our calculations, this temperature is 175 K. Further, an external electric field with a strength of $1 \cdot 10^6$ V/m was applied to the graphene/C_{60} system along the Y axis of the graphene plane. After the application of an external electric field, the trajectory of the C_{60} motion gradually takes the form of a loop, which repeats approximately every 20 ps. The fragments of the loop trajectory of the C_{60} molecule at consecutive times are presented in Fig. 5.

Fig. 5 shows that, over time, the C_{60} molecule revolves in the elliptical trajectories in the center of the bowl-shaped graphene region. Herewith the C_{60} molecule rotates clockwise according to the direction of the applied field. The circular current was calculated for one of the loop trajectories shown in Fig. 5. This trajectory is an ellipse with major and minor semi-axes equal to $a = 3.32$ Å and $b = 1.28$ Å, respectively. The area of the ellipse is 13.35 Å^2. The calculated circular current is ~10.4 nA. Fig. 6 shows the change in the magnetic induction B of a magnetic field generated by the current loop as the distance away from the center of loop. As can be seen, the magnetic induction B decreases with distance from the current loop, reaching a maximum value of $B = 6.2$ μT at the center of the ellipse.

Thus, an increase in the diameter of the bowl-shaped graphene region by only 5.6 Å made it possible to achieve an almost twofold increase in the circular current flowing in a closed loop. The magnetic field generated by the current loop has also increased. Consequently, the proposed way for enhancing the circular current and the generated magnetic field is effective and can be implemented in practice. It can be assumed that by choosing the appropriate dimensions of the local hydrogenation region of graphene atomic network, it is possible to achieve an increase in the circular current and magnetic induction of the generated magnetic field by tens to hundreds of times.

4. Conclusions

Based on the results of numerical simulation of the behavior of the C_{60} molecule on a graphene substrate containing defects in the form of local hydrogenation, we have found a new effective way to manipulate the fullerene molecules on graphene. It was shown that the chemisorption of a group of hydrogen atoms on a graphene sheet lead to the creation of a bowl-shaped region in the graphene atomic network. The finite motion of the C_{60} molecule was observed within this region. It was found that at a certain temperature and strength of the external electric field, the C_{60} molecule starts to move within the bowl-shaped region in elliptic trajectories. In the course of studying the C_{60} circular motion, we have found a physical effect of appearance of the loop current. The value of this current was equal to 10.4 nA at temperature of 175 K and electric field strength of $1 \cdot 10^6$ V/m. As the effective way for the enhancing current we proposed to vary the sizes of the bowl-shaped graphene region. It can be assumed that the models of new miniature magnetic field sources for a variety of nanodevices will be developed on the base of the obtained results.

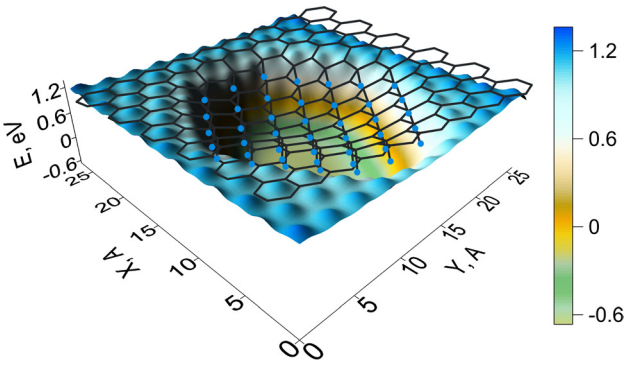


Fig. 4. (Color online) Atomic structure of the graphene/C_{60} hybrid system with a defect in the form of local hydrogenation of graphene by a group of 44 atoms and the energy profile of the interaction of C_{60} with graphene.

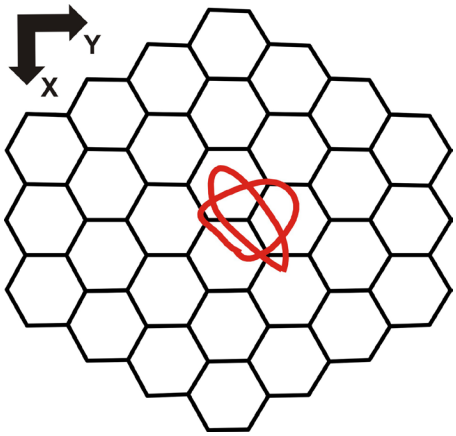


Fig. 5. (Color online) Top view of fragments of the loop trajectory of the center of mass of a molecule C_{60} within the local hydrogenation region of the graphene atomic network (bowl-shaped region with a diameter of 14.6 Å) at 175 K in the presence of an electric field with a strength of $1 \cdot 10^6$ V/m.

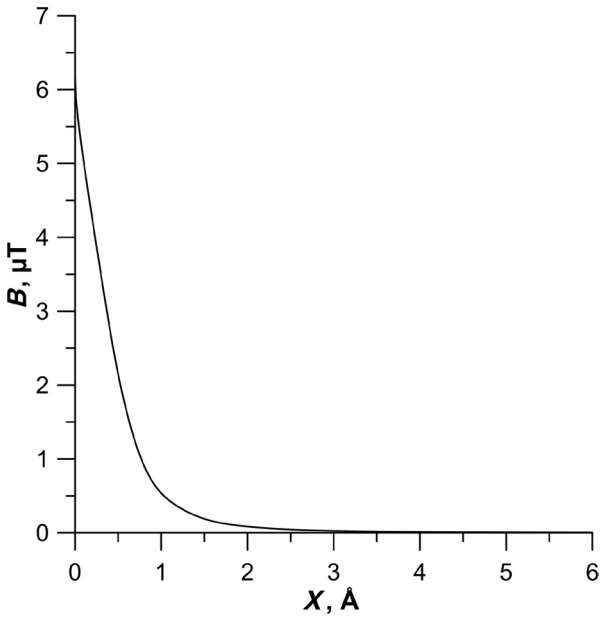


Fig. 6. The change in the magnetic induction B a magnetic field generated by the current loop as the distance away from the center of loop for the case of bowl-shaped graphene region with a diameter of 14.6 Å.

Acknowledgements. This study was supported by the Ministry of Science and Higher Education of the Russian Federation in the framework of the government task (project No. FSRR-2020-0004).

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