Electric field control of spin-resolved edge states in graphene quantum nanorings

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The electric-field effect on the electronic and magnetic properties of triangular and hexagonal graphene quantum rings with zigzag edge termination is investigated by means of the single-band tight-binding Hamiltonian and the mean-field Hubbard model. It is shown how the electron and spin states in the nanoring structures can be manipulated by applying an electric field. We find different spin-depolarization behaviors with variation of electric field strength due to the dependence of spin densities on the shapes and edges of this kind of nanorings.

In the case of triangular quantum rings, the magnetization on the inner and outer edges can be selectively tuned and the spin states depolarize gradually as the field strength is increased, while in the case of hexagonal nanorings, the transverse electric field reduces the magnetic moments on both inner and outer edges symmetrically and rapidly.

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I. INTRODUCTION

The enormous interest in graphene research, both in theory and experiments is driven by its unusual electronic and magnetic properties and great potential applications in modern photovoltaic and nanoelectronic devices\textsuperscript{4,5}. The fabrication and manipulation of the graphene quantum dots are of the most desirable developments in this field\textsuperscript{6,7}. Moreover, a carbon-based structure is a good candidate as the fundamental logic gates for designing ultra-fast spintronic devices\textsuperscript{8} and field effect transistors\textsuperscript{9,10}.

In graphene quantum dots, magnetic properties may arise from their topological structure and edge effects. Due to the repulsive electron-electron interaction in zigzag-edge structures, $\pi$ electrons cannot be paired simultaneously. Therefore, zigzag-edge graphene nanostructures such as triangular or hexagonal quantum dots and rings can exhibit magnetic ordering\textsuperscript{9,10} because of an imbalance between the number of atoms belonging to the two sublattices in graphene bipartite nanostructures, which produces a net magnetization at the ground state. In recent years, ring-type nanostructures made of carbon-based materials like graphene nanoribbons with different shapes and edges have been studied theoretically\textsuperscript{11}. Among them, zigzag-edge graphene quantum rings (GQRs) have a larger magnetic moment compared to graphene quantum dots due to more zigzag edges per unit area\textsuperscript{12}. In this regard, magnetic properties, energy levels, and electronic correlations in graphene rings have also been investigated\textsuperscript{13,14,15}.

On the other hand, the ability to control electronic and magnetic properties of material by external electric field is at the heart of the modern electronic and spintronic devices\textsuperscript{16,17}. In monolayer structures, such as graphene, the screening of electric field is extremely reduced, and hence, less energy is consumed. Moreover, graphene quantum rings and dots with controllable band-gaps, smaller size compared to the coherence length, and individual electronic and magnetic properties are more interesting for nanoelectronic applications\textsuperscript{18-22}. Therefore, it is essential to study the effect of electric field in such structures and several studies by focusing on this effect in graphene quantum dots with zigzag edges have been reported accordingly\textsuperscript{23-25}. Moreover, half-metallicity in graphene nanoribbons induced by electric field has been predicted\textsuperscript{26}. In these studies, the electronic and magnetic properties of graphene nanoribbons and quantum dots by variation of electric field strength have been demonstrated; however, to our knowledge, the magnetic behavior of GQRs under an external electric field has not yet been explored.

In this paper, by combining the single-band tight-binding approximation and the mean-field Hubbard model, we numerically analyze the influence of transverse electrical-field on electronic and magnetic properties of narrow triangular and hexagonal GQRs with zigzag edges. The spin-resolved electronic structure and the sensitivity of energy gap to the field strength in these nanorings are investigated. We show that the distribution of local magnetic moments on the inner and outer edges can be tuned differently by adjusting the strength of electric field. The evolution of magnetic moments strongly depends on the type of nanorings, and in the case of triangular structures, such evolution is even size-dependent. Therefore, after a short description about our theory in Section II, we will present our numerical results in Section III, to see how by applying an electric field rather than a magnetic field the spin states can be manipulated in such zigzag-edge nanorings.

II. MODEL AND METHOD

We simulate the triangular and hexagonal GQRs [shown in Fig.1], using the $\pi$-orbital tight-binding model and the Hubbard repulsion treated in the mean-field approximation to include the effect of electron-electron interaction in the graphene nanoring calculations, which may induce magnetic localized moments on the zigzag-shaped edge\textsuperscript{9,10}. The Hubbard Hamiltonian for an $N$-electron ring with $N$ lattice sites in the presence of a transverse electric field can be written as\textsuperscript{25,26}.
The first term in Eq. (1) which is defined as

Here, the operator \( c_i \) \(^\dagger\) (\( c_i \)) creates (annihilates) an electron with spin \( \sigma \) at site \( i \) and \( n_i,\sigma = c_i,\sigma c_i,\sigma^\dagger \) is a number operator. The first term in Eq. (1) which is defined as \( \epsilon_i,\sigma = -e x_i E \) describes the effect of electric field along the \( x \)-axis, where \( e \) is the electron charge, \( E \) denotes the electric field strength, and \( x_i \) is the position of the \( i \)th carbon atom along the \( x \)-direction. The second term corresponds to the single \( \pi \)-orbital tight-binding Hamiltonian, while the third term accounts for the on-site Coulomb interaction \( U \). Accordingly, the magnetic moment (spin) at each atomic site can be expressed as:

\[
m_i = \langle S_i \rangle = (\langle n_i,\uparrow \rangle - \langle n_i,\downarrow \rangle)/2.
\]

(2)

Note that, in our calculations the transfer integral between all nearest-neighbor sites and the Hubbard parameter are set to, \( t = -2.66 \) and \( U = 2.82 \) eV, respectively.

III. RESULTS AND DISCUSSION

In order to study the electronic states and magnetic properties of the nanorings in the presence of electric field, we start from an anti-ferromagnetic spin configuration as an initial condition and solve the mean-field Hubbard Hamiltonian self-consistently. As shown in Fig. 1, we consider the electric field distribution along the \( x \)-axis for triangular and hexagonal nanorings. The electric field is generated by two gate electrodes (not shown in the figure) with opposite electrostatic potentials at the outside of each ring and parallel to the \( y \)-axis. These ring structures have well-defined zigzag shape in both the inner and outer edges. We describe the number of benzene rings which forms the thickness of each ribbon by \( W \), and the number of carbon atoms in the inner edge of each ribbon by \( L \). The triangular graphene ring with \( W=3 \) and \( L=7 \) consists of 285 carbon atoms. This ring structure has different number of \( A \)- and \( B \)-type atoms on each edge. Moreover, the inner and the outer edges, each have only one type of atom. Therefore, the total spin, \( S = \sum m_i \), at zero electric field, according to the Lieb’s theorem\(^{13,27} \), reaches \( S = 4.5 \) with a maximum value of \( S = 0.13 \). In addition, the magnetic configurations of the inner and outer edge atoms in the triangular graphene ring are in opposite directions with different distribution of magnetic moment values, as shown in Fig. 1(a).

On the other hand, the hexagonal graphene ring consists of 198 carbon atoms, which has the same number of \( A \)- and \( B \)-type atoms. In this structure, the inner and outer edges, each consist of both types of atoms. Therefore, the total spin value of the ring is zero and the localized magnetic moments on both edges form an antiferromagnetic spin configuration, as shown in Fig. 1(b).

Now we study the single particle energy and the edge magnetic moment for the case of triangular ring in the presence of electric field. The strength of the electric field in these calculations is taken in the range of several volts per nanometer, which is experimentally achievable if small electrodes are made. In Fig. 2, the energy spectra for spin-up and spin-down electrons are plotted near the Fermi energy that is set to zero [Figs. 2(a)- 2(d)]. Without considering the effect of on-site Coulomb repulsion, the energy spectrum for the edge states at the Fermi energy collapses to a shell of degenerate states.

Fig. 2(a) shows that in the absence of electric field, the electron-electron interaction opens a gap in the degenerate shell at the Fermi level. This interaction has a great influence on the degree of spin polarization of the triangular GQRs and
induces a total spin equal to 4.5. By switching the electric field on, the quasi degeneracy of edge states is lifted due to the Stark effect and the energy dispersion increases as shown in Fig. 2(b). In this electric field strength, the net magnetization remains unchanged. The spin depolarization, however, occurs when the electrostatic field is beyond a critical value \( E_c \). In the case of a triangular ring (shown in Fig. 1(a)) the critical field is 2.5 V/nm. As the electric field increases to 3.5 V/nm, the energy levels of spin-up (spin-down) states, right above (below) the Fermi energy, shift below (above) the Fermi level and, consequently, the spin gap between the two spin branches with the same eigenstate index decreases, and the total spin of the ring reaches \( S = 2.5 \) [Fig. 2(c)]. A further increase in the strength of the electric field causes a dramatic reduction in the spin gap of the edge states with the same index which results in a remarkable decrease in the total spin value. As it can be seen in Fig. 2(d), an external electric field with the strength of 5 V/nm can strongly reduce the spin gap, and hence, the total spin of the ring reaches 0.5. Our numerical results show that the total spin of graphene rings is harder to depolarize than the total spin of graphene quantum dots, indicating more sensitivity to the electric field in the graphene quantum dots compared to the nanorings.

In order to have a better understanding of the magnetic evolution of edge states in triangular GQRs as the electric field changes, we have shown in Fig. 3(a) and 3(b) the distribution of magnetic moments under the influence of two different values of field strength. The distribution of magnetic edge states under the electric field in the range of 0-2.5 V/nm remains unchanged. By increasing the field strength beyond \( E_c = 2.5 \) V/nm, the magnetic ordering of edge atoms decreases. The spin-up densities on the outer edges and far away from the \( y \)-axis vanish, while the spin-down densities on the inner edges and the spin-up densities near the origin on the outer edge of the ring do not change significantly [Fig. 3(a)]. By further increasing the electric field strength, the local magnetic moments are restricted to the center region along the \( y \)-axis of the ring, as shown in Fig. 3(b).

This magnetic evolution can be understood from Eq. 1. The electrostatic potential has low values at atomic sites near the center line along the \( y \)-axis in the ring, and the corresponding on-site energies have small values compared to the on-site energy of atoms far away from the center region. Therefore, the influence of electric potential on the atoms located far away from the center is much stronger than the effect of electron-electron interaction. However, as the strength of the electric field is increased above 8 V/nm, the local magnetic moments due to the on-site Coulomb interaction are dramatically reduced, even for the inner edge atoms. Accordingly, only the carbon atoms on the outer edges and near the \( y \)-axis have non-magnetic moment with a total spin equal to 1/2. Note that the net magnetization vanishes only for the triangular graphene structures with an even number of atoms. The total spins as a function of electric field strength for two triangular rings with different sizes are shown in Fig. 3(c). It is clear that the spin depolarization can happen when the field strength is beyond a critical value. In other words, the spin depolarization begins to happen when the total spin decreases by one quantum \( \hbar \) as the electric field is increased above a critical value. This spin reduction manifests itself as a steplike function of the electric field strength. The electric field cannot completely depolarize the total spin of a triangular ring with \( W = 3 \) and \( L = 7 \), and such a structure has the minimal value \( S = 1/2 \) due to the odd number of electrons. On the other hand, the complete spin depolarization occurs for a triangular carbon ring with \( W = 2 \) and \( L = 7 \). This triangular structure consists of 186 carbon atoms which indicates an even number of electrons at half filling, and hence, the total spin completely depolarizes, i.e., \( S = 0 \). In our calculations, the critical value of the electric field for starting the spin depolarization in the larger (smaller) triangular ring is \( E = 2.5 \) V/nm (3.5 V/nm) and the minimum spin can reach \( E = 5 \) V/nm (6 V/nm). This means that the larger GQR, which is affected by a larger electrostatic potential, depolarizes easier than the smaller ring. Accordingly, the spin depolarization of such triangular rings depends on the number of electrons at half filling and also the field strength. To investigate the field effect on the electronic structure of the nanorings, we have shown in Fig. 3(d) the energy gap as a function of field strength for the two different triangular rings.

It is clear that the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) gap reduces as the field strength increases. For instance, in the case of thinner ring, the energy gap decreases to the small value of 20 meV at \( E \geq 5 \) V/nm. However, in contrast with the rectangular graphene nanodots, in which the nanodots switch from antiferromagnetic to diamagnetic ground state at low electric fields, and the HOMO-LUMO gap becomes spin-dependent.
the ground state of the triangular nanorings remains antiferromagnetic even at high electric fields such as $E = 5$ V/nm. This indicates the strong influence of the geometry, quantum size, and edges on the induced magnetic moments in this type of nanoring compared to those in the square nanodot.

Now we consider the field effect on the electronic and magnetic properties of the hexagonal nanoring with zigzag edge termination, shown in Fig. 1(b). In Fig. 4, we have depicted the spin-resolved energy spectra for spin-up and spin-down electrons as the strength of electric field is increased. This kind of nanorings does not show any spin gaps in its electronic spectra and the net magnetization remains zero in both the presence and absence of the applied field. On the contrary, the local magnetic moment on each side of the hexagonal ring is nonzero, even under the influence of electric field. To see this behavior, we have shown in Fig. 5(a) and 5(b) the distribution of induced magnetic moments in the presence of two field strengths $E = 1.7$ V/nm and $E = 1.8$ V/nm, respectively. It is clear that, at $E = 1.7$ V/nm, the spin states show the same evolution on each side (edge) of the six inner or outer edges of the hexagonal ring. Moreover, the local magnetic moments on each side of the ring vanish rapidly when the electric field strength varies from 1.7 to 1.8 V/nm. Comparing the Figs. 5(a) and 5(b), one can conclude that the electronic structure of the hexagonal ring is more sensitive to the field strength, compared to the triangular structures, and its spin depolarization occurs at lower values of electric field (see Figs. 3(a) and 3(b) and 5(a) and 5(b) for comparison). We find that, applying a transverse electric field, even on a small part of the hexagonal ring, reduces the spin states symmetrically on the zigzag edge atoms [not shown here].

In Fig. 5(c), the maximum value of the local magnetic moments ($M = \mu_B S$) in the hexagonal ring is shown as the electric field is varied. It can be seen that the magnetic moment is relatively small, compared to the triangular rings. Since the value of $S$ is less than 1, the spin depolarization does not behave like what was observed in the triangular rings, in which the depolarization occurs when the electric field strength is beyond some critical values. Consequently, the local magnetization in the hexagonal systems is easier to depolarize than the local magnetic moment in the triangular structures. We have also shown the behavior of the energy gap as a function of field strength in Fig. 5(d). Our results reveal that, although the HOMO-LUMO gap of the hexagonal rings responds to the variation of field strength very similar to that of the triangular rings, it drops and vanishes much faster than those in Fig. 3(d).

IV. CONCLUSION

In summary, the influence of electric field and electron-electron interaction on the edge states in the zigzag-edge GQRs was studied by means of the single-band tight-binding Hamiltonian and the mean-field Hubbard model. There is a competition between the electron-electron interaction and the external electric field to depolarize the local magnetic moments on the inner and outer edges of the nanorings. We showed that the strength of electrostatic potential strongly affects the magnetization in our systems. The presence of electrostatic field mainly affects the electronic spectrum in the vicinity of the Fermi energy, while the single particle states which are located far away from this energy, remain unchanged and therefore the magnetic properties of nanorings are determined by the $\pi$-electrons around the Fermi level.

In the case of triangular rings, the inner and the outer edge states show different evolutions under the external electric field, and the net spin-polarization of the nanorings might be enhanced by choosing appropriate field strengths such as the non-uniform electrostatic fields proposed in ref. 22. On the con-
trary, the evolution of the magnetic moments on all of the edges (whether they be the inner edges or the outer edges) of the hexagonal nanorings is the same by variation of field strength, which clearly indicates that the size and the shape of the nanorings are crucial factors in the electric-field-induced depolarization in this type of nanorings. Although by increasing the size of the nanorings many particle interactions are expected to be more important, these findings can still be useful to qualitatively understand the variation of energy spectra and spin depolarization under the influence of electric field in larger graphene nanorings.

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1 Novelosov K S, Geim A K, Morozov S V, Jiang D, Zhang Y, Dubono S V, Grigorieva I V and Firso A A 2004 Science 306 666
2 Gülü A D G, Potasz P, Vozny O, Korkusinski M and Hawrylak P 2009 Phys. Rev. Lett. 103 246805
3 Novelosov K S, Geim A K, Morozov S V, Jiang D, Katsnelson M I, Grigorieva I V, Dubono S V and Firso A A 2005 Nature 438 197
4 Sanvito S 2007 Nature nanotechnology 2 204
5 Silvestrov P G and Efetov K B 2007 Phys. Rev. Lett. 98 016802
6 Wang W L, Yazyev O V, Meng S and Kaxiras E 2009 Phys. Rev. Lett. 102 157201
7 Schwierz F 2010 Nature nanotechnology 5 487
8 Diez-Perez I, Li Z, Hihath J, Li J, Zhang C, Yang X, Zang L, Dai Y, Feng X, Muellen K and Tao N J 2010 Nature communication 1 31
9 Farghadan R, Saffarzadeh A and Semiromi E H 2013 J. Appl. Phys. 114 214314
10 Fujita M Wakabayashi K Nakada K and Kusakabe K 1996 J. Phys. Soc. Jap. 65 1920
11 Russo S Oostinga J B Wehenkel D Heersche H B Sobhani S S Vanderven L M K and Morpurgo A F 2008 Phys. Rev. B 77 085413
12 Grujic M, Tadic M and Peeters F M 2013 Phys. Rev. B 87 085434
13 Potasz P Güclü A D and Hawrylak P 2010 Phys. Rev. B 82 075425
14 Bahamon D A, Pereira A L C and Schulz P A 2009 Phys. Rev. B 79 125414
15 Agapito L A, Kioussis N and Kaxiras E 2010 Phys. Rev. B 82 201411
16 Potasz P Güclü A D Vozny O Folk J A and Hawrylak P 2011 Phys. Rev. B 83, 174441
17 Saffarzadeh A and Farghadan R 2011 Appl. Phys. Lett. 98 023106
18 Fernández-Rossier J and Palacios J J 2007 Phys. Rev. Lett. 99 177204
19 Zheng X H, Song L L, Wang R N, Hao H, Guo L J and Zeng Z 2010 Appl. Phys. Lett. 97 153129
20 Dong Q R 2013 J. Appl. Phys. 113 234304
21 Ma W L and Li S S 2012 Phys. Rev. B 86 045449
22 Zheng H and Duley W 2008 Phys. Rev. B 78 155118
23 Güclü A D, Potasz P and Hawrylak P 2011 Phys. Rev. B 84 155118
24 Son Y W, Cohen M L and Louie S G 2007 Nature 444 347
25 Farghadan R and Saievar E 2012 J. Appl. Phys. 111 014304
26 Guo J, Gunlycke D and White C T 2008 Appl. Phys. Lett. 92 163109
27 Lieb E H 1989 Phys. Rev. Lett. 62 1201

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