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Publisher's version / Version de l'éditeur:

<https://doi.org/10.1103/PhysRevB.83.174441>

Physical Review. B, Condensed Matter and Materials Physics, 83, 17, pp. 174441-1-174441-6, 2011-05-31

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Electronic and magnetic properties of triangular graphene quantum rings

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(Received 9 February 2011; revised manuscript received 6 April 2011; published 31 May 2011)

Electronic and magnetic properties of triangular graphene rings potentially fabricated using carbon nanotubes as masks are described as a function of their size and width. The electronic properties of the charge neutral system are calculated using tight-binding method and interactions are treated using the mean-field Hubbard model. We show that for triangular quantum dots with a triangular hole, the magnetic properties are determined by the width of the ring, leading to ferromagnetic corners and antiferromagnetic sides. The electronic properties of gated graphene quantum rings as a function of additional number of electrons or holes are described by a combination of tight-binding, Hartree-Fock, and configuration interaction methods. The outer edge is found to be maximally spin polarized for almost all filling factors while the evolution of the excitation gap as a function of shell filling shows oscillations as a result of electronic correlations.

DOI: 10.1103/PhysRevB.83.174441

PACS number(s): 75.75.-c, 73.22.Pr, 81.05.ue

I. INTRODUCTION

There is currently significant interest in electronic properties^{1–5} and potential technological applications of graphene.^{5–8} While ideal carbon graphene sheet is in itself nonmagnetic, theoretical research suggests occurrence of local magnetic moments in the vicinity of defects^{9,10} or zigzag-type boundaries of graphene sheet.^{11–15} The zigzag edges lead to the existence of degenerate states near the Fermi level, predicted by tight-binding model and confirmed by density functional theory (DFT) calculations.^{11,16–21} Experimentally, these states were observed using scanning tunneling microscope (STM) near monoatomic steps on a graphite surface.^{22,23}

For graphene nanostructures with zigzag edges, electron exchange interactions lead to antiferromagnetic order in graphene ribbons¹¹ but ferromagnetic order in triangular graphene quantum dots (TGQD).^{13,14,20,24} Apart from edge magnetism, interactions in partially filled edge states result in other correlated ground states.^{24,25} In particular, magnetism in TGQD can be completely destroyed by adding extra electron to the charge neutral system.²⁴

The fabrication of triangular graphene quantum dots with well-defined shape and zigzag edges is potentially challenging as it requires control of the edges at the atomic level. In this work we explore the use of carbon nanotubes (CNTs) as masks with atomically precise shape and size for producing graphene triangular nanostructures. As shown in Fig. 1 three carbon nanotubes allow the fabrication of a triangular graphene quantum dot but with a hollow center, triangular graphene quantum ring (TGQR). In this paper, we describe electronic and magnetic properties of TGQR as a function of size, width and charge controlled by the gate. We show that many of the properties of triangular graphene quantum dots survive and that new properties related to ring formation appear.

The paper is organized as follows. In Sec. II, we describe our triangular graphene ring model and propose an experimental method for designing graphene nanostructures with well-defined edges. In Sec. III, we study the single particle properties using tight-binding (TB) Hamiltonian in the nearest neighbor approximation. In Sec. IV, we investigate

magnetic properties of the charge neutral rings using mean-field Hubbard model and density functional theory (DFT). In Sec. V, we study magnetic properties and the role of correlations in gated charged ring using a combination of tight-binding, Hartree-Fock, and configuration interaction methods (TB + HF + CI). We analyze electronic and magnetic properties of graphene rings with given width as a function of the number of electrons occupying degenerate shell. Section VI contains the summary.

II. TRIANGULAR GRAPHENE QUANTUM RING MODEL

As shown in Fig. 1, TGQR can be fabricated using carbon nanotubes (CNT) as a mask in the etching process.^{26,27} One can place CNT over the graphene sheet along a given crystallographic direction and cover atoms lying below, e.g., along a zigzag direction. Three carbon nanotubes can be arranged in a triangular shape, along three zigzag edges. As a result one expects to obtain a triangular structure with well defined zigzag edges and a hole in the center, as shown on the right in Fig. 1. We also note that this method can be used to obtain, e.g., bow-tie structures with potential application as quantum information logic devices.¹⁴ TGQD with a zigzag edge shown in Fig. 1 consists of $N_{\text{out}}^2 + 4N_{\text{out}} + 1$ atoms,²⁸ where N_{out} is the number of edge atoms on one outer edge and edge atoms are defined as those having only two neighbors. In order to create a triangular ring, we remove a small triangle from the center (Fig. 1). The small triangle consists of $N_{\text{inn}}^2 + 4N_{\text{inn}} + 1$ atoms, where N_{inn} is the number of edge atoms on one inner edge. The resulting TGQR has $N = N_{\text{out}}^2 - N_{\text{inn}}^2 + 4(N_{\text{out}} - N_{\text{inn}})$ atoms. Its width satisfies $N_{\text{out}} - N_{\text{inn}} = 3(N_{\text{width}} + 1)$, where N_{width} is the width counted in the number of benzene rings. For instance, the structure in Fig. 1 has $N_{\text{width}} = 2$. In the full triangle, the imbalance between number of A type (N_A) and B type (N_B) of atoms in bipartite honeycomb graphene lattice, proportional to N_{out} , leads to appearance of zero-energy states in the TB model in the nearest-neighbors approximation. The number of zero-energy states can be defined as $N_{\text{zero}} = |N_A - N_B|$.²⁹ Removing a small triangle from the center lowers the imbalance between

FIG. 1. (Color online) Proposed experimental method for designing a TGQR. Three CNTs arranged in equilateral triangle along zigzag edges play the role of a mask. By using etching methods one can obtain TGQR with well-defined edges. The circumference of CNT determines the width of TGQR. Red (light gray) and blue (dark gray) colors distinguish between two sublattices in the honeycomb graphene lattice.

two types of atoms in the structure, leading to a decreased number of zero-energy states $N_{\text{zero}} = 3(N_{\text{width}} + 1)$. Thus, interestingly, the number of zero energy states in TGQR's only depends on the width of the ring, and not the size.

III. SINGLE PARTICLE PROPERTIES OF TRIANGULAR GRAPHENE QUANTUM RINGS

We calculate the single particle energy spectra using the tight-binding model, which has been successfully applied to other carbon nanostructures.^{16–18,20,24,30} The TB Hamiltonian is given by

$$H_{TB} = \sum_{i,j,\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma}, \quad (1)$$

where t_{ij} are the hopping integrals, $c_{i\sigma}^\dagger$ and $c_{i\sigma}$ are creation and annihilation operators of electrons on π_z orbitals on site i with spin σ . All in-plane dangling bonds at the edges are assumed passivated with hydrogen. We distinguish between two types of atoms in the unit cell of the honeycomb lattice of graphene sheet, labeled as A type and B type, and indicated by red (light gray) and blue (dark gray) colors, respectively, in Fig. 1. We note that while outer edges are built of A-type atoms, inner edges are built of B-type atoms (Fig. 1).

In Fig. 2, we show the single particle spectra obtained by diagonalizing the TB Hamiltonian Eq. (1) in the nearest-neighbors approximation. Figure 2(a) shows the energy spectrum for TGQR with $N_{\text{width}} = 2$ consisting of 171 atoms and shown in Fig. 1. It has $N_{\text{out}} = 11$ and $N_{\text{inn}} = 2$ and the number of zero-energy states is $N_{\text{zero}} = 9$. Similarly, in Fig. 2(b) we show the TB spectrum for TGQR with $N_{\text{width}} = 5$ and consisting of 504 atoms. It has $N_{\text{out}} = 21$ and $N_{\text{inn}} = 3$, giving $N_{\text{zero}} = 18$, consistent with our formula $N_{\text{zero}} = 3(N_{\text{width}} + 1)$. We note that the states of the zero-energy shell consist of orbitals belonging to one type of atoms,³¹ indicated by red (light gray) color in Fig. 1, and lie mostly on the outer edge.¹⁹ On the other hand, the other states close to the Fermi level consist of orbitals belonging to both sublattices but lie mostly on inner edge (not shown here). This fact has implications for

FIG. 2. Single particle TB levels for TGQR with (a) $N_{\text{width}} = 2$, consisting of 171 atoms and (b) $N_{\text{width}} = 5$, consisting of 504 atoms. The degeneracy at the Fermi level (dashed line) is a function of the width $N_{\text{zero}} = 3(N_{\text{width}} + 1)$, for (a) $N_{\text{zero}} = 9$ and for (b) $N_{\text{zero}} = 18$.

the magnetic properties of the system, described in the next section.

IV. MAGNETISATION OF CHARGE NEUTRAL TRIANGULAR GRAPHENE QUANTUM RINGS

The presence of the degeneracy in the vicinity of the Fermi level can lead to the appearance of finite magnetic moment due to exchange interaction that favors parallel spin configuration. Charge neutral system corresponds to half-filled degenerate TB zero-energy shell. Therefore a finite magnetic moment proportional to the number of degenerate states N_{zero} is expected. In order to study magnetic properties of the system we use Hubbard model in the mean-field approximation:

$$H = t \sum_{\langle i,j \rangle, \sigma} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_i (n_{i\uparrow} \langle n_{i\downarrow} \rangle + n_{i\downarrow} \langle n_{i\uparrow} \rangle), \quad (2)$$

where the first term is the TB Hamiltonian given by Eq. (1) in the nearest-neighbor approximation and $n_{i\uparrow} = c_{i\uparrow}^\dagger c_{i\uparrow}$ is the spin-up particle number operator. We choose $t = -2.5$ eV and on-site interaction $U \sim 2.75$ eV calculated using Slater orbitals, for effective dielectric constant $\kappa = 6$.³² The ratio $|U/t| = 1.1$ resides in commonly used range (1.1–1.3)³³ and is close to DFT results.³⁴ A self-consistent solution of the Hubbard Hamiltonian is characterized by single-particle spectrum and the spin density $M_i = (n_{i\uparrow} - n_{i\downarrow})/2$ on each atom i .

In Fig. 3 we show spectra obtained (a) from Hubbard model in the mean-field approximation and (b) using DFT implemented in the SIESTA package³⁵ for TGQR with $N_{\text{width}} = 2$, consisting of 171 atoms, $N_{\text{out}} = 11$ and $N_{\text{inn}} = 2$. This corresponds to nine zero-energy TB levels, shown in Fig. 2(a). Interactions open a spin dependent gap in the single-particle zero-energy shell, resulting in maximum spin polarization of those states. The total spin of the system is $S_{\text{tot}} = 9/2$, in accordance with Lieb's theorem.²⁹ In Figs. 3(c) and 3(d) we show corresponding spin density. The net total spin is mostly localized on the outer edge and vanishes as one moves to the center, similar to full triangle results.²⁰ Good agreement between results obtained from the mean-field Hubbard and DFT calculations (Fig. 3) validates the applicability of the mean-field Hubbard model and allows us to study efficiently structures consisting of larger number of atoms.

FIG. 3. (Color online) Energy spectra from (a) self-consistent mean-field Hubbard model and (b) DFT calculations for TGQR with the width $N_{\text{width}} = 2$ and 171 atoms. States up to the Fermi level (dashed line) are occupied. (c) and (d) are corresponding spin densities. The radius of circles is proportional to the value of spin density on a given atom. Proportions between size of circles in (c) and (d) are not retained.

In Fig. 4 we show the results of the Hubbard model for a larger structure with 315 atoms, $N_{\text{out}} = 20$ and $N_{\text{inn}} = 11$, with the same width $N_{\text{width}} = 2$. The energy spectrum, Fig. 4(a), looks similar to that from Fig. 3(a) and the total spin is again $S_{\text{tot}} = 9/2$. On the other hand, spin density in Fig. 4(b) is different than in Fig. 3(c). Here, the outer edge is still spin polarized, but the inner edge reveals opposite polarization. This fact can be understood in the following way. Electrons with majority spin (spin up) occupy degenerate levels of the zero-energy shell which are built exclusively of orbitals localized on atoms belonging to the sublattice labeled as A . These states are localized on the outer edge. Due to repulsive on-site interaction, spin-up electrons repel minority spin electrons (spin-down) to the sublattice labeled as B . After self-consistent calculations, spin-up and spin-down densities are spatially separated occupying mostly sublattices A and B , respectively. Local imbalance between the two sublattices occurs near edges, resulting in local magnetic moments, seen in Fig. 4(b). As a result, we observe that the outer and inner edges

FIG. 4. (Color online) (a) Self-consistent energy spectra and (b) corresponding spin densities from the mean-field Hubbard model for TGQR with the width $N_{\text{width}} = 2$ and 315 atoms. The radius of circles is proportional to the value of spin density on a given atom.

FIG. 5. (Color online) (a) Average magnetic moment as a function of size (N is the number of atoms) in corner and edge regions. Structures reveal stable ferromagnetic order in corners, but a change from ferromagnetic to antiferromagnetic on edges with increasing size. (b) Total spin in corner region as a function of width. Linear dependence is due to increased number of zero-energy states.

are oppositely spin polarized, similar to graphene nanoribbons. However, the magnetic moments are not equal resulting in local antiferromagnetic state in contrast to the antiferromagnetic state in graphene nanoribbons.

Magnetic moment of the inner edge is the highest close to the middle of the edge and decreases toward the corners. This allows us to distinguish between two types of regions in the structure: corners and edges. Due to triangular symmetry of the system, in further analysis we can focus on only one corner and one edge. We define average magnetization in a given region as $\langle M \rangle = \sum_i M_i / N'$, where summation is over sites in a given region and N' is corresponding total number of atoms. In Fig. 5(a) we show average magnetization in one corner and one edge as a function of the size of TGQR for a given width, $N_{\text{width}} = 2$. Small structures ($N < 200$ atoms) reveal finite and comparable magnetic moments in both regions, consistent with Fig. 3(c), where most of the spin density is distributed on outer edges. There are two effects related to increasing size: the length of the internal edge increases increasing spin polarization opposite to the outer edge spin polarization [see Fig. 4(b)] and increase of the overall number of atoms in the edge region. The first effect leads to antiferromagnetic coupling between opposite edges and second one to vanishing average magnetization, seen in Fig. 5(a). We note here that although the average magnetization rapidly decreases with size, it never approaches zero. On the other hand, average magnetization at the corner is stable and nearly independent of the size. This fact is related to fixed number of atoms in corner region.

According to Lieb's theorem, the total spin of the system must be $S = 3(N_{\text{width}} + 1)/2$. Moreover, the spin density for smaller structures is equally distributed along the outer edge [see Fig. 3(c)]. Partitioning structure into six approximately equal regions, three corners and three edges [see inset in Fig. 5(a)], gives an approximately equal total spin in each domain. In further analysis we show that this is true for arbitrary size triangular rings. In Fig. 5(b), we present the total spin in one corner $S_c = \sum_i M_i$ as a function of the width of the ring. Summation is over all sites in one corner. We obtain linear dependence $S_c \sim N_{\text{width}}$, which, for the best choice of cuts, should be described by relation $S_c = (N_{\text{width}} + 1)/4$, which is one-sixth of the total spin S of the entire structure. In this ideal case, all six regions reveal equal total spin S_c , independently

of the size of the structure. We relate this fact to the behavior in edge and corner regions. For sufficiently large structures, magnetic moments on the edge region are distributed on a large number of atoms, giving vanishing average magnetic moment but always finite total spin equal to $S_c = (N_{\text{width}} + 1)/4$. With increasing size, the length of the inner edge increases. In order to satisfy the relation $S_c = (N_{\text{width}} + 1)/4$, the magnetic moment on the outer edge increases proportionally to oppositely polarized magnetic moment on the inner edge, resulting in antiferromagnetic coupling between opposite edges. On the other hand, in corners, there is always a fixed number of atoms independent of size, giving a constant average magnetic moment and the total spin equal to $S_c = (N_{\text{width}} + 1)/4$. We note that above conclusions were confirmed by investigation of TGQR with a width in the range $2 \leq N_{\text{width}} \leq 9$ for structures up to 1500 atoms. Thus, we can treat large TGQR as consisting of three ferromagnetic corners connected by antiferromagnetic ribbons, with ribbons exhibiting finite total spin. This result can be useful in designing spintronic devices. Choosing CNT with proper circumference, one can obtain TGQR with desired magnetic moment localized in the corners.

At the end of this section, we investigate a triangular ring with a hexagonal hole in the center. A removed hexagon consists of an equal number of A- and B-type atoms. Thus, despite a smaller number of atoms in the structure as compared to a full triangle, the imbalance between A- and B-type atoms is maintained. This fact allows us to conclude that the number of degenerate states in the system does not depend on the size of the hexagonal hole. Figure 6(a) shows self-consistent energy levels for the structure consisting of 681 atoms, $N_{\text{zero}} = 27$. Similar to TGQR, interactions open a spin gap and the structure reveals finite magnetic moment. In Fig. 6(b) we show the corresponding spin density. The outer edge is spin-up polarized, which is related to a local majority of sublattice A atoms. On the other hand, consecutive inner edges consist of a different type of atoms and consequently are oppositely polarized. Performing partitioning for corners and edges, similar to TGQR, the system can be treated as consisting of three ferromagnetic triangles connected by antiferromagnetic ribbons.

FIG. 6. (Color online) (a) Self-consistent energy spectra and (b) corresponding spin density for triangular structure with 681 atoms and hexagonal hole in the center. Structure can be treated as consisting of three ferromagnetic triangles connected by antiferromagnetic ribbons. The radius of circles in (b) is proportional to the value of spin density of a given atom.

V. SPIN AND CORRELATIONS IN GATED GRAPHENE QUANTUM RINGS

Previous work^{24,25} shows that the Hubbard model and DFT calculations describe well properties of the charge neutral graphene quantum dots. On the other hand, gated triangular graphene quantum dots reveal effects related to electronic correlations in the partially filled zero-energy shell.²⁴ This requires approaches beyond the mean-field approximation. In order to study charged systems, we use a combination of TB, HF, and configuration interaction (CI) methods. The calculation method is as follows. First we solve the mean-field problem using a combination of TB and self-consistent HF method for the system with an empty degenerate shell. A large gap between the degenerate shell and the valence band provides a reference state with fully occupied valence band that can be described by a single Slater determinant of HF self-consistent orbitals. The mean-field Hamiltonian for this reference state is given by²⁴

$$H_{MF} = \sum_{i,l,\sigma} t_{il} c_{i\sigma}^\dagger c_{l\sigma} + \sum_{i,l,\sigma} \sum_{j,k,\sigma'} (\rho_{jk\sigma'} - \rho_{jk\sigma'}^{\text{bulk}}) \langle ij|V|kl \rangle - \langle ij|V|lk \rangle \delta_{\sigma,\sigma'} c_{i\sigma}^\dagger c_{l\sigma}, \quad (3)$$

where the first term is the TB Hamiltonian given by Eq. (1) and second term corresponds to interactions in mean-field approximation. TB parameters are $t = -2.5$ eV for the nearest neighbors, and $t = -0.1$ eV for the next-nearest neighbors.³⁶ $\rho_{jk\sigma'}$ and $\rho_{jk\sigma'}^{\text{bulk}}$ are density matrices for quantum dot and bulk graphene, respectively. After iteratively diagonalizing the Hamiltonian given by Eq. (3) we obtain new quasiparticle levels describing unoccupied states, including the zero-energy shell. In Figs. 7(a) and 7(b) we show Hartree-Fock energy levels for TGQR with $N_{\text{width}} = 2$ and 171 atoms. The degeneracy of the zero-energy shell is slightly removed, with a group of three states separated from the rest by a small gap, similar to full triangle results.²⁴ These states correspond to the wave function localized in the three corners of the triangle. We then start filling these degenerate states by adding extra electrons one by one and solve the many-body Hamiltonian corresponding to the added electrons, given by

$$H_{MB} = \sum_{s,\sigma} \epsilon_s a_{s\sigma}^\dagger a_{s\sigma} + \frac{1}{2} \sum_{\substack{s,p,d,f, \\ \sigma,\sigma'}} \langle sp|V|df \rangle a_{s\sigma}^\dagger a_{p\sigma'}^\dagger a_{d\sigma} a_{f\sigma}, \quad (4)$$

where the first term describes Hartree-Fock energies and the second term describes interactions between quasiparticles occupying degenerate states denoted by s, p, d, f indices. We calculated the two-body quasiparticle scattering matrix elements $\langle sp|V|df \rangle$ from the two-body localized on-site Coulomb matrix elements $\langle ij|V|kl \rangle$ by using Slater π_z orbitals,³⁷ where i, j, k, l are the site indices. In numerical calculations, on-site, scattering, and exchange terms up to the next-nearest neighbors, as well as all long-range direct terms are included. The few largest Coulomb matrix elements are given in Ref. 32. We build our basis from all possible many-body configurations within degenerate shell for a given number of electrons N_{el} . Finally, in this many-body basis we

FIG. 7. (a) and (b) Hartree-Fock energy levels for TGQR with $N_{\text{width}} = 2$ consisting of 171 atoms and filled by $N_{\text{el}} = 10$ electrons. The configuration represented by arrows in (a) corresponds to all occupied spin-down orbitals and one occupied spin-up orbital. The configuration represented by arrows in (b) is the configuration from (a) with one spin-down flipped. (c) The low-energy spectra for the different total spin S for $N_{\text{el}} = 10$ electrons. The ground state has $S = 4$, indicated by *a*, with one of the configuration shown in (a). The lowest energy excited state, indicated by *b*, is ~ 4 meV higher in energy, corresponds to spin-flip configurations with one of the configuration shown in (b).

diagonalize the Hamiltonian given by Eq. (4) in each subspace with a projection of total spin onto the z axis S_z .

In Fig. 7(a) we show an example of a configuration related to $N_{\text{el}} = 10$ electrons. This corresponds to a half-filled degenerate shell with all spin-down states of the shell filled and one additional spin-up electron. For maximal total spin $S = 4$ there are nine possible configurations corresponding to the nine possible states of the spin-up electron. An energy spectrum obtained by diagonalizing full Hamiltonian, Eq. (4), for total spin $S = 4$ is shown in Fig. 7(c). We see that, by comparison with total spin states with $S = 0, 1, \dots, 4$, the ground state corresponding to configurations of the type *a* [one of which is shown in Fig. 7(a)] is maximally spin polarized, with the excitation gap in the $S = 4$ subspace of ~ 40 meV. However, the lowest energy excitations correspond to spin-flip configurations with total spin $S = 3$, one of which is shown Fig. 7(b). These configurations involve spin-flip excitations from the fully spin polarized electronic shell in the presence of one additional spin-up electron.

The energy $E_{\text{gap}} = 4$ meV for $N_{\text{el}} = 10$, indicated in Fig. 7(c), is shown in Fig. 8(a) together with the energy gap for all electron numbers $1 < N_{\text{el}} < 18$ and hence all filling factors. In Fig. 8(b), we show the total spin S of the ground and the first excited state as a function of the number of electrons occupying the degenerate shell. For arbitrary filling, except

FIG. 8. (Color online) (a) Energy spin gap between ground and first excited states. Black long arrow corresponds to half-filled shell with $E_{\text{gap}} \sim 28$ meV. Significant reduction in the spin-flip energy gap for one additional electron, $E_{\text{gap}} \sim 4$ meV, indicated by small black arrow, is the signature of correlation effects. (b) Total spin of the ground and first excited states as a function of the number of electrons N_{el} . The small black arrow indicates excited state for $N_{\text{el}} = 10$ electrons with one of the configurations shown schematically with arrows in Fig. 7(b).

for $N_{\text{el}} = 2$, the ground state is maximally spin polarized. Moreover, the first excited state has total spin consistent with spin-flip excitation from the maximally spin polarized ground state as discussed in detail for $N_{\text{el}} = 10$. The signature of correlation effects is seen in the dependence of the excitation gap on the shell filling, shown in Fig. 8(a). For half-filling at $N_{\text{el}} = 9$, indicated by an arrow, the excitations are spin-flip excitations from the spin polarized zero-energy shell. This energy gap, ~ 28 meV, is significantly larger in comparison with the energy gap of ~ 4 meV for spin flips in the presence of an additional spin-up electron. The correlations induced by an additional spin-up electron lead to a much smaller spin-flip excitation gap. This is to be compared with full graphene triangular dots where spin-flip excitations have a lower energy leading to full depolarization of the ground state.²⁴

VI. CONCLUSIONS

To conclude, we described here the electronic and magnetic properties and the role of correlations in triangular graphene rings potentially fabricated using carbon nanotubes as masks. The TGQR structures exhibit a magnetic moment due to

the presence of a degenerate band of states at the Fermi level. We showed that the degeneracy of the zero-energy band, and thus the total magnetization of the system is determined by the width of the structure. For TGQR with a small inner hole, only the outer edges are spin polarized, similarly to full triangle results. However, as the size of the hole is increased, the inner edges become spin polarized as well, showing antiferromagnetic configurations. We show that TGQR with a large hole can be treated as a system consisting of three spin polarized corners connected by antiferromagnetic ribbons. Designing structures with a given width enables us to obtain systems with arbitrary magnetic moment, opening the possibility of using TGQR in designing spintronic devices. The robustness of the total spin formation to charging is assessed.

In charged graphene rings, correlation effects are found to play a role, affecting the energy required to flip a spin for different filling factors.

ACKNOWLEDGMENTS

The authors thank NRC-CNRS CRP, Canadian Institute for Advanced Research, Institute for Microstructural Sciences, Quantum Works for support. P.P. acknowledges financial support from the fellowship co-financed by European Union within European Social Fund. This work was financed from the sources granted by the Department of Scientific Research for science development in the years 2010–2012, as a research project, Grant No. NN202 48839.

- ¹K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, Y. Zhang, S. V. Dubonos, I. V. Grigorieva, and A. A. Firsov, *Science* **306**, 666 (2004).
- ²K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, M. I. Katsnelson, I. V. Grigorieva, S. V. Dubonos, and A. A. Firsov, *Nature (London)* **438**, 197 (2005).
- ³Y. B. Zhang, Y. W. Tan, H. L. Stormer, and P. Kim, *Nature (London)* **438**, 201 (2005).
- ⁴A. H. C. Neto, F. Guinea, N. M. R. Peres, K. S. Novoselov, and A. K. Geim, *Rev. Mod. Phys.* **81**, 109 (2009).
- ⁵A. K. Geim and K. S. Novoselov, *Nat. Mater.* **6**, 183 (2007).
- ⁶A. Rycerz, J. Tworzydło, and C. W. Beenakker, *Nature Phys.* **3**, 172 (2007).
- ⁷F. Xia, T. Mueller, Y.-M. Lin, A. Valdes-Garcia, and P. Avouris, *Nature Nanotechnol.* **4**, 839 (2009).
- ⁸T. Mueller, F. Xia, and P. Avouris, *Nature Photon.* **4**, 297 (2010).
- ⁹O. V. Yazyev and L. Helm, *Phys. Rev. B* **75**, 125408 (2007).
- ¹⁰M. W. C. Dharma-Wardana and M. Z. Zgierski, *J. Phys. E* **41**, 80 (2008).
- ¹¹Y. Son, M. L. Cohen, and S. G. Louie, *Nature (London)* **444**, 347 (2006).
- ¹²O. V. Yazyev and M. I. Katsnelson, *Phys. Rev. Lett.* **100**, 047209 (2008).
- ¹³W. L. Wang, S. Meng, and E. Kaxiras, *Nano Lett.* **8**, 241 (2008).
- ¹⁴W. L. Wang, O. V. Yazyev, S. Meng, and E. Kaxiras, *Phys. Rev. Lett.* **102**, 157201 (2009).
- ¹⁵H. Lee, Y.-W. Son, N. Park, S. Han, and J. Yu, *Phys. Rev. B* **72**, 174431 (2005).
- ¹⁶K. Nakada, M. Fujita, G. Dresselhaus, and M. S. Dresselhaus, *Phys. Rev. B* **54**, 17954 (1996).
- ¹⁷M. Ezawa, *Phys. Rev. B* **73**, 045432 (2006).
- ¹⁸T. Yamamoto, T. Noguchi, and K. Watanabe, *Phys. Rev. B* **74**, 121409 (2006).
- ¹⁹M. Ezawa, *Phys. Rev. B* **76**, 245415 (2007).
- ²⁰J. Fernandez-Rossier and J. J. Palacios, *Phys. Rev. Lett.* **99**, 177204 (2007).
- ²¹J. Akola, H. P. Heiskanen, and M. Manninen, *Phys. Rev. B* **77**, 193410 (2008).
- ²²Y. Niimi, T. Matsui, H. Kambara, K. Tagami, M. Tsukada, and H. Fukuyama, *Appl. Surf. Sci.* **241**, 43 (2005).
- ²³Y. Kobayashi, K. I. Fukui, T. Enoki, K. Kusakabe, and Y. Kaburagi, *Phys. Rev. B* **71**, 193406 (2005).
- ²⁴A. D. Güçlü, P. Potasz, O. Voznyy, M. Korkusinski, and P. Hawrylak, *Phys. Rev. Lett.* **103**, 246805 (2009).
- ²⁵B. Wunsch, T. Stauber, F. Sols, and F. Guinea, *Phys. Rev. Lett.* **101**, 036803 (2008).
- ²⁶F. Molitor, J. Güttinger, C. Stampfer, D. Graf, T. Ihn, and K. Ensslin, *Phys. Rev. B* **76**, 245426 (2007).
- ²⁷C. Stampfer, J. Güttinger, F. Molitor, D. Graf, T. Ihn, and K. Ensslin, *Appl. Phys. Lett.* **92**, 012102 (2008).
- ²⁸D. A. Bahamon, A. L. C. Pereira, and P. A. Schulz, *Phys. Rev. B* **79**, 125414 (2009).
- ²⁹E. H. Lieb, *Phys. Rev. Lett.* **62**, 1201 (1989).
- ³⁰P. R. Wallace, *Phys. Rev.* **71**, 622 (1947).
- ³¹P. Potasz, A. D. Güçlü, and P. Hawrylak, *Phys. Rev. B* **81**, 033403 (2010).
- ³²P. Potasz, A. D. Güçlü, and P. Hawrylak, *Phys. Rev. B* **82**, 075425 (2010).
- ³³O. V. Yazyev, *Phys. Rev. Lett.* **101**, 037203 (2008).
- ³⁴L. Pisani, J. A. Chan, B. Montanari, and N. M. Harrison, *Phys. Rev. B* **75**, 064418 (2007).
- ³⁵J. M. Soler, E. Artacho, J. D. Gale, A. Garcia, J. Junquera, P. Ordejon, and D. Sanchez-Portal, *J. Phys. Condens. Matter* **14**, 2745 (2002).
- ³⁶R. S. Deacon, K.-C. Chuang, R. J. Nicholas, K. S. Novoselov, and A. K. Geim, *Phys. Rev. B* **76**, 081406(R) (2007).
- ³⁷B. J. Ransil, *Rev. Mod. Phys.* **32**, 245 (1960).