Quantum Monte Carlo Study of Semiconductor Artificial Graphene Nanostructures

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Semiconductor artificial graphene nanostructures where Hubbard model parameter $U/t$ can be of the order of 100, provide a highly controllable platform to study strongly correlated quantum many-particle phases. We use accurate variational and diffusion Monte Carlo methods to demonstrate a transition from antiferromagnetic to metallic phases for experimentally accessible lattice constant $a = 50 \text{ nm}$ in terms of lattice site radius $\rho$, for finite sized artificial honeycomb structures nanopatterned on GaAs quantum wells containing up to 114 electrons. By analysing spin-spin correlation functions for hexagonal flakes with armchair edges and triangular flakes with zigzag edges, we show that edge type, geometry and charge nonuniformity affect the steepness and the crossover $\rho$ value of the phase transition. For triangular structures, the metal-insulator transition is accompanied with a smoother edge polarization transition.

Keywords: artificial graphene, graphene quantum dots, quantum simulators, variational Monte Carlo, diffusion Monte Carlo

In recent years, technological advances in photonic and condensed matter based artificial superlattices gives us opportunities to develop practical quantum simulators [1][14]. These quantum simulators allow us to replicate complex systems that are hard to fabricate and provide us with a playground to verify theoretical predictions. In this respect, artificial graphene (AG) nanostructures, designed by imitating the 2D honeycomb pattern of graphene, have been proven to be good candidates for being reliable and controllable sources for both fabrication and investigation of many physical phenomena related to Dirac fermions [13][19]. In particular, AG nanostructures can be formed using semiconductor materials. While earlier reports on nanopatterned artificial graphene on GaAs quantum well (QW) structures found no evidence of massless Dirac fermions (MDFs), presumably because of the relatively large lattice periods [20][22], in recent experimental works using modulation-doped AlGaAs/GaAs quantum wells [13][14], shrinking down of the lattice constant of the honeycomb array to approximately $50 \text{ nm}$ allowed the observation of the predicted graphene-like behavior [21][23][25].

Observation of Dirac fermions in AG also opens up a fresh way of studying graphene quantum dots [20] where geometry, size and edge type is expected to give rise to several physical properties such as bandgap opening [27], edge magnetization [28][30] and optical control [31]. However, the fabrication and reliability issues such as edge reconstruction or presence of impurities, make it harder to observe interesting phenomena predicted to occur in nanostructured graphene. Semiconductor AG nanostructures, on the other hand, offer several advantages such as tunability of system parameters including lattice constant, site radius and potential depth, which, in turn, allow to control electron-electron interactions and tunneling strength between sites, in particular Hubbard parameter $U/t$. In experimental structures with lattice constant $a = 50 \text{ nm}$ [13], $U/t$ can be as high as 350 (as we will argue below) i.e. two orders of magnitude larger than the critical value for antiferromagnetic Mott transition predicted by calculations based on Hubbard model for honeycomb lattice [32][35]. Moreover, unlike in real graphene, long range electron repulsion does not cancel the attraction of the artificial confining potential in AG even near charge neutrality. For such large and long-ranged electron interactions, a non-perturbative many-body approach is desirable for careful treatment of correlation effects.

Earlier theoretical work on electron interaction effects in semiconductor AG nanostructures based on density functional theory (DFT) showed that Dirac bands were stable against interactions [23][24], which was also confirmed using path integral Monte Carlo calculations [25]. However, recent calculations using Hartree-Fock and exact diagonalization approaches for a triangular zigzag geometry with $a = 12.5 - 15 \text{ nm}$ show that a transition from antiferromagnetic (AF) insulator to metallic phases occurs, pointing to the importance of electron interactions [36].

In this work, we use continuum variational Monte Carlo (VMC) and diffusion Monte Carlo (DMC) methods for non-perturbative and accurate treatment of many-body correlations within the fixed-node approximation to study GaAs based AG nanostructures. First, we consider an hexagonal armchair geometry which serves as a bridge between the finite-size samples and bulk graphene [27], with lattice constant $a = 50 \text{ nm}$ following recent experimental work [14]. We show that, a transition from AF to metallic phase occurs, but is affected by a nonuniform charge distribution in the sample due to finite size effects. This charge nonuniformity which is not present in real graphene quantum dots, causes the phase transition to be steeper and to occur at a smaller value of $\rho$. We also investigate AG quantum dots with triangular zigzag geometry and show that edge magnetization survives the phase transition, in agreement with previous theoretical
prediction for smaller lattice constants [36].

Our model of nanostructured semiconductor AG consists of \(N\) interacting electrons in a honeycomb array of \(N\) confining potentials, described by the many-body Hamiltonian

\[
H = -\frac{1}{2} \sum_{i}^{N} \nabla_{i}^{2} + \sum_{i}^{N} V(\mathbf{r}_{i}) + \sum_{i}^{N} k|\mathbf{r}_{i}|^{2} + \sum_{i<j}^{N} \frac{1}{r_{ij}} \tag{1}
\]

in effective atomic units (electronic charge \(e\), dielectric constant \(\varepsilon\), effective mass \(m^*\), and \(\hbar\) are set to 1), where \(1/r_{ij}\) is the Coulomb interaction between the electrons, \(V(\mathbf{r}_{i})\) is the total potential energy of the confining potentials, and \(k\) is the spring constant of quadratic gate potential located at the center of the system which controls the finite size effects. Typical material properties for GaAs, effective electron mass \(m^* = 0.067m_{0}\) and dielectric constant \(\varepsilon = 12.4\), are used. Corresponding effective Bohr radius is \(a_{B} = 9.794\) nm, and the effective Hartree energy is 11.857 meV. The honeycomb array of potential wells is modelled using gaussian-like functions [25],

\[
V(\mathbf{r}) = V_{0} \sum_{\mathbf{R}_{0}} \exp[-(|\mathbf{r} - \mathbf{R}_{0}|/\rho)^{s}] \tag{2}
\]

where \(s \geq 1\), \(V_{0}\) is the potential depth, \(\rho\) is the radius and \(s\) is the sharpness of the potential wells. \(\mathbf{R}_{0}\) is the location of the potential wells. In our numerical calculations, dot-to-dot distance (lattice constant) was fixed to \(a = 50\) nm, while several radius values from \(\rho = 10\) nm to 35 nm were covered. Three different sharpness values were used; \(s = 1\) for a gaussian potential, \(s = 2.8\) for a sharp, muffin-tin like potential, and \(s = 1.4\) in between. \(V_{0}\) values vary depending on dot radius (e.g. increasing monotonically with dot radius from \(-38\) meV to \(-15\) meV for \(s = 1.4\) and \(N = 42\)), tuned to keep the total energy of the system close to zero, since our aim is to imitate the charge neutral behavior of finite sized graphene quantum dot. For too high values of \(V_{0}\), electrons tend to escape the system during VMC or DMC simulations, while for too low values over localization occurs [24].

Accuracy of the numerical calculations depends on trial wave functions in both VMC and DMC methods. One starts with a set of single-particle orbitals (e.g. localized gaussians or from self-consistent calculations) to build a Slater-Jastrow trial wave function \(\Psi_{T}(\mathbf{R})\) which is a linear combination of products of up- and down-spin Slater determinants of these orbitals multiplied by a Jastrow factor (The details of our Jastrow factor is given in Ref. [37]). After the VMC calculations where Jastrow parameters as well as the gaussian functions width are optimized using energy minimization technique [38], we use fixed-node DMC [39, 40] to project the optimized many-body wave function onto a better approximation of the true ground state, an approximation that has the same nodes as \(\Psi_{T}(\mathbf{R})\). The resulting fixed-node DMC energy is an upper bound to the true energy and depends only on the nodal structure of the Slater part of the trial wave function \(\Psi_{T}(\mathbf{R})\) [39].

In order to form Slater determinants, we prepare three different types of orbitals aiming to capture metallic or AF insulator phases, depending on the potential well radius \(\rho\): (i) Localized gaussian functions that are proven to be one of the most suitable functions for 2D systems of quantum dots [37] [41, 42] and are expected to provide a better description of strongly localized states. (ii) Tight-binding (TB) orbitals, on the other extreme, may be used to describe metallic phases in which electrons move more freely. (iii) Mean-field Hubbard (MFH) orbitals can describe both localized and liquid-like states depending on the ratio \(U/t\). Corresponding variational and fixed-node energies of those three types of orbitals are expected to hint us at a possible transition from metallic state to an AF order as a function of \(\rho\). In this work, all quantities that do not commute with the Hamiltonian were calculated using an extrapolated estimator,

\[\langle \hat{O} \rangle = 2\langle \hat{O} \rangle_{\text{DMC}} - \langle \hat{O} \rangle_{\text{VMC}} \tag{39}\]

FIG. 1. Armchair hexagonal flake results, \(s = 1.4, k = 0\) and \(N = 42\) plotted for several trial wave functions. (a) VMC total energy vs \(\rho\), (b) DMC total energy vs \(\rho\), (c) Extrapolated pair spin density results for \(\rho = 10\) nm, using tight-binding trial wave function. (d) Extrapolated pair spin density results for \(\rho = 35\) nm, using tight-binding trial wave function. The reference electron is located at the X marked point for c and d.

Figure 1a shows the VMC energies obtained from TB, MFH and gaussian orbitals, as a function of \(\rho\), for an armchair hexagonal geometry with 42 sites and 42 electrons. The parabolic gate potential parameter is turned off (\(k = 0\)) and its effect will be discussed in a later section. At low \(\rho\), gaussian and MFH \(U = 20t\) orbitals provides better variational energies. As \(\rho\) is increased to \(\approx 27\) nm, a clear crossover (from insulator to metallic
phase) occurs above which TB and MFH $U = 2t$ orbitals take the lead. Fixed-node DMC energies, however, reveal a somewhat different picture, shown in Fig. 1b. From $\rho = 10$ nm to $\rho \approx 18$ nm, all trial wave functions give similar energies within the statistical error bars, and split near $\rho \approx 18$ nm. After the split, the ground state of the system is represented by TB and MFH $U = 2t$ trial wave functions. These results show that, surprisingly, TB trial wave function has equally good nodal structure as the gaussian orbitals at low $\rho$ values, raising questions about the true nature of the ground state. To reveal the underlying electronic and magnetic structure, we consider the pair densities $p_{\sigma\sigma_0}(r, r_0)$, probability of finding an electron with spin $\sigma$ at location $r$ when an electron with spin $\sigma_0$ is fixed at location $r_0$, and the pair spin densities, $p_{\uparrow\uparrow}(r, r_0) - p_{\downarrow\downarrow}(r, r_0)$. Figures 2-a and b show the pair spin densities for a reference spin down electron fixed on top of a site (chosen to break the system symmetry and away from the edges) shown with a cross. At $\rho = 10$ nm, electrons are well localized at the sites, leading to an AF insulator. On the other hand, at large values of $\rho$, spin-spin correlations are weak and short-ranged up to nearest neighbors. While these results confirm that a transition from AF insulator to a metallic phase does occur as observed in previous work [30], there are several issues left to address: (i) Inconsistent signature regarding the crossover $\rho$ value obtained from VMC and DMC energies and the underlying dynamics of the transition. (ii) Finite size and edge effects. (iii) Relationship to bulk properties.

![FIG. 2. DFT band structure of bulk artificial graphene for (a) $\rho = 15$ nm and (b) $\rho = 25$ nm. Inset figure shows that $U/t$ ratio plotted against dot radius $\rho$, predicted by DFT and single particle calculations.](image)

Now, we turn our attention to the 2D bulk properties for similar system parameters used in our calculations. As mentioned earlier, $V_0$ values are tuned to keep the total energy of the system close to zero. The question is whether such set of parameters leave the Dirac cone structure intact, which may otherwise have repercussions on electronic and magnetic properties. In Fig. 2 we show our DFT calculations results in the local density approximation (LDA). The Dirac cone structure is preserved for the range $\rho = [12.5, 30]$ nm; outside this range the Dirac fermion picture becomes distorted. Moreover, we can estimate the TB hopping parameter $t$ from the slope of the Dirac cones as $t = \frac{2}{\pi} a^2 \frac{E}{dk}$ where $a$ is the lattice constant. As we see from Fig. 2 $t$ increases with increasing $\rho$ but decreasing $s$, as expected. In addition to $t$, to build a Hubbard model, we estimated the $U$ value for a single well using $U = 2\pi \int r n(r)V(r) dr$ after solving single particle Schrödinger equation. In the inset of Fig. 2, we plot $U/t$ as a function of $\rho$ which shows a fast decay from $\approx 300$ to 50 between $\rho = 15$ and 20. Surprisingly, these results indicate that the critical $U/t$ value for metal-insulator transition in AG is much higher than the critical value of $\sim 3.8$ predicted by Hubbard calculations [32][33], presumably due to the importance of long-range interactions and deviation from the nearest neighbor TB approximation as $\rho$ increases. On the other hand, according to our quantum Monte Carlo (QMC) calculations, MFH trial wave functions based on LDA estimation of $U/t$ do not provide the most suitable fixed-node energies and the nodal structure of the simplest TB trial wave functions works best for the whole range of system parameters regardless of the underlying electronic or magnetic state.

![FIG. 3. Effect of $k$ on total electron densities for armchair hexagonal flake. $s = 1.4$, $\rho = 25$ nm and $N = 42$ using tight-binding trial wave function. (a) $k = 0$. (b) $k = 3.56 \times 10^{-4}$ meV/nm$^2$.](image)

Next, we focus on finite size effects. While $V_0$ is tuned to imitate overall charge neutrality, long-range electron interactions still affects the charge distribution inside the system, pushing the electrons towards the edges and/or corners, as can be seen in Fig. 3 for the hexagonal armchair system with $N = 42$, $s = 1.4$ and $\rho = 25$ nm. While, in principle, $V_0$ can be decreased further to achieve charge uniformity, this would localize the electrons too strongly to their sites and make the system negatively charged unlike in real graphene systems. An alternative solution is to apply a quadratic gate potential controlled by the parameter $k$ in Eq. 1. When $k > 0$, quadratic gate potential attracts the electrons towards the center of the system so that the charge uniformity is preserved, and finite size effects are minimized, as shown in Fig. 3 for $k = 3.56 \times 10^{-4}$ meV/nm$^2$.

In order to understand more in detail the dynamics of the transition from AF insulator to metallic phase...
including effects of charge nonuniformity and quantum well potential sharpness, we consider a real space spin-spin correlation function $g$ normalized by the density-density correlations, $g = \langle s_i s_j \rangle / \langle n_i n_j \rangle$ where $s_i$ and $n_i$ are the average total spin and total electron densities on site $i$ within a radius $r$, and $i,j$ are the nearest neighbor sites. We used $r = a/2$ in all spin-spin correlation calculations, where $a$ is the lattice constant. Output values of the function remain in [-1,1] range, with $g = -1$ corresponding to AF and $g = 0$ corresponding to metallic configuration ($g = 1$ means that all spins are in the same direction, which does not happen in the subspace $S_z = 0$ considered here). In Fig. 4a, $g$ is plotted against $\rho$ obtained from different trial wave functions, for $s = 1.4$ and $k = 3.56 \times 10^{-4}$ meV/nm$^2$ to obtain charge uniformity. We have also added a weighted average of all trial wave functions using Boltzmann distribution at $T = 4$ K, representing the ground state, to ensure that no effects are missed when various trial wave functions lead to same ground state energies within statistical noise. The emerging picture is that the system remains strongly AF between $\rho = 10 - 18$ nm, then starts fading smoothly, finally reaching fully metallic regime around $\rho = 30$ nm. We note that the DMC energies for $k > 0$ split around $\rho = 26$ nm (see the inset of Fig. 4b), in contrast with the $k = 0$ results in Fig. 4a where the split occurs near $\rho = 18$ nm. Figure 4b summarizes all weight averaged $g$ values obtained for different $k$ and $s$ values, and for both flake sizes. Interestingly, for $k = 0$ the transition from AF to metallic regime is sharper than for charge uniform systems ($k > 0$), albeit at lower $\rho$ values. Sharpness $s$ and system size $N$, on the other hand, does not have a significant effect on the transitions.

For triangular zigzag structures, spin polarized edges even in the metallic phase is expected due to the imbalance between the two sublattices $2S = 30$ 30 43, with a non-zero ground state total spin $S_z = (N_A - N_B)/2$ where.

$N_A$ and $N_B$ are number of $A$ and $B$ sublattice atoms, according to Lieb’s theorem 44. In our AG flake, the total spin is $S_z = 2$ for $N = 46$ and $S_z = 5/2$ for $N = 61$. The analysis of the spin-spin correlation functions for $s = 1.4$ and $k = 2.7 \times 10^{-4}$ meV/nm$^2$ leads to a similar picture as before; the system with $N = 46$ sites is perfectly AF between $\rho = 10 - 20$ nm, then smoothly vanishes between $\rho = 20 - 35$ nm, before becoming completely metallic. For $N = 61$ sites, although the transition is not as smooth presumably due to larger statistical fluctuations at large system size, a similar picture emerges (Fig. 5c). In order to understand the edge magnetization during the transition, we consider an edge polarization ratio defined as

$$p_e = \frac{\langle |s|_{\text{edge}} \rangle - \langle |s|_{\text{bulk}} \rangle}{\langle |s|_{\text{edge}} \rangle + \langle |s|_{\text{bulk}} \rangle},$$

which gives one if only edge sites are polarized, zero if edge and bulk sites are equally (un)polarized. In Fig. 5c, edge polarization ratio increases as $\rho$ increases, indicating that spins are polarized more at the edges as the system goes into metallic phase similar to real triangular zigzag graphene quantum dots, and consistent with the spin density results in Fig. 5b and insets, demonstrating a metallic phase with edge-polarized spins at $\rho = 35$ nm for 46 sites and at $\rho = 30$ nm for 61 sites. Additionally, in Fig. 5b weighted average analysis also confirms that the triangular zigzag AG flake undergoes a metal-insulator transition between $\sim 18$ and 30 nm dot radius.
However, the edge polarization transition occurs slower compared to the metal insulator transition.

In summary, we have shown that a metal - antiferromagnetic insulator transition occurs in nanopatterned GaAs based artificial graphene structures including up to $N = 114$ electrons with lattice constant $a = 50$ nm as a function of site radius, using accurate variational and diffusion Monte Carlo methods, within the Dirac regime as confirmed by our density functional calculations. Our approach where a simple tight-binding trial wave function combined with a Jastrow factor is found to be sufficient to account for electron correlation effects in both metallic and antiferromagnetic regimes, allows direct modelling of system parameters, making it possible to systematically investigate the effects of edge type, geometry, size, quantum well shape and gate potentials. We have shown that the steepness and the crossover $\rho$ value of the phase transition is affected by charge nonuniformity due to finite size effects. For triangular structures exhibiting magnetized edge states, the edge polarization transition is shown to occur more slowly compared to the metal insulator transition.

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