Electric-field controlled spin in bilayer triangular graphene quantum dots

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We present theoretical results based on mean-field and exact many-body approaches showing that in bilayer triangular graphene quantum dots with zigzag edges the magnetism can be controlled by an external vertical electric-field. We demonstrate that without electric field the spins of the two layers of the quantum dot interact ferromagnetically. At a critical value of the electric-field, the total spin of the bilayer structure can be turned off or reduced to a single localized spin, a qubit isolated from contacts and free from interaction with nuclear spins.

Graphene exhibits unusual electronic properties including relativistic nature of quasi-particles, sublattice pseudospin, and zero energy bandgap. The application of graphene for logic devices requires opening of the bandgap which can be achieved by either size quantization, or chemical modification, or bringing in a second layer and applying an external electric-field, or size quantization in bilayer graphene nanostructures. In particular, when graphene is reduced to a triangular quantum dot with zigzag edges, sublattice symmetry is broken, size dependent energy gap is open, and a band of degenerate states at the Fermi level is created leading to finite macroscopic spin polarization. This allows simultaneous size, shape and edge engineering of magnetic, electrical, and optical properties in a single material-graphene.

In this work, we investigate electronic and magnetic properties of bilayer triangular graphene quantum dots with zigzag edges under external vertical electric-field. We show that the magnetic moment of bilayer triangular graphene quantum dots can be controlled by the vertical electric-field. Without the electric-field, the magnetic moments of the two layers are shown to be coupled ferromagnetically. Using configuration interaction and mean-field calculations based on tight-binding model, we demonstrate that the ferromagnetism can be either turned off or reduced to a single electron/hole spin. The single electron spin is hence isolated in a charge neutral structure by the application of an electric-field, independent of the size of the quantum dot and without decoherence due to contacts. The electric-field control of the ferromagnetism and isolation of a single spin opens new applications in spintronics and quantum information processing.

However, not all the A2 atoms have a B1 partner as required by Bernal stacking. A more natural configuration choice is shown on the right hand side of Fig.1a. The top layer triangle has its floating atoms removed, making it smaller than the bottom layer triangle. Such a bilayer construction has the interesting property of having an odd number of degenerate states at the Fermi level (zero-energy shell of states) independent of its size, allowing to isolate a single spin in a charge neutral structure and hence isolated from the contacts. This is illustrated in Fig.1b, where the spin density isosurfaces are shown for zero electric-field (left hand side) and finite electric-field (right hand side), as obtained from our tight-binding based configuration interaction calculations explained in detail below. When the electric-field is off, both layers have a finite magnetic moment, as in single layer triangles, differing by one spin due to the size difference of the two triangles. The magnetic moments of the two layers are coupled ferromagnetically. When
a sufficiently high electric field is applied, electrons from the lower layer reduce their energy by flipping their spin and transferring to the top layer filling up all the available spin up and down zero-energy states, leaving behind one single spin.

We have confirmed the above picture through calculations with varying levels of accuracy. As a first step, we diagonalize the tight-binding Hamiltonian given by

\[ H_{TB} = \sum_{ij\sigma} \tau_{ij} c^\dagger_{i\sigma} c_{j\sigma} + \sum_{i\sigma} V_i c^\dagger_{i\sigma} c_{i\sigma} \]  

where the operator \( c^\dagger_{i\sigma} \) creates an electron on a \( p_z \) orbital on site \( i \) with spin \( \sigma \). The tight-binding parameters \( \tau_{ij} \) are fixed to their bulk values \( t = -2.8 \) eV for in-plane nearest neighbors \( i \) and \( j \) and \( t_\perp = 0.4 \) eV for inter-layer hopping. The effect of the potential difference induced by an external perpendicular electric-field \( E \) is taken into account through \( V_i = -\Delta V/2 \) for the bottom layer atoms and \( V_i = \Delta V/2 \) for the top layer atoms. Figure 2a shows the energy spectrum near the Fermi level for \( \Delta V = 0 \) eV here. First, we study a small BQD of 107 atoms and 9 zero-energy states. The magnetic properties of BQD structures as a function of applied electric field can be studied by solving the Hubbard model within the self-consistent mean-field approach:

\[ H_{MF} = \sum_{ij\sigma} \tau_{ij} c^\dagger_{i\sigma} c_{j\sigma} + \sum_{i\sigma} V_i n_{i\sigma} + U \sum_{i\sigma} (n_{i\sigma} - \frac{1}{2}) n_{i\sigma} \]  

where \( U \) is the on-site Hubbard term taken to be 2.75 eV here. First, we study a small BQD of 107 atoms and 9 zero-energy states. Figure 3 shows the energies of lowest energy states with different total spin \( S \) as a function of potential difference \( \Delta V \) between the layers, with respect to the ferromagnetic configuration \( S_{max} = 4.5 \) for a system of 107 atoms and 9 zero-energy states.

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or hole spin in a neutral BQD by applying an external electric field.

The procedure of isolating single electron or hole should occur regardless of the size of the system since the top layer has always one less zero-energy state than the bottom one. In order to investigate the size dependence, in Fig.4a we show the energy difference between the ferromagnetic and antiferromagnetic (FM-AFM) states calculated in the mean-field Hubbard approximation as a function of applied voltage for several sizes up to 1507 atoms. We note that, due to the unusually high degeneracy of the states, self-consistent iterations occasionally get trapped in a local energy minima. We have thus repeated the calculations several times using different initial conditions and/or convergence schemes to assure that the correct ground state was reached. As expected, at $\Delta V = 0$, the FM-AFM gap increases with the size of the system $N$. In fact, the FM-AFM gap energy per $N$ approaches a constant value of 14.3 meV (top panel). Critical value $\Delta V_c$ where the transition occurs as a function of number of atoms $N$ (bottom panel).

We now test the predictions of the mean-field Hubbard model by including long range interaction and correlation effects, which is computationally feasible for the system size studied in Fig.3. We first solve a Hartree-Fock generalization of the Eq.2, but with empty zero-energy states.

$H_{HF} = \sum_{ij\sigma} \tau_{ij\sigma} c_i^\dagger c_j + \sum_{i\sigma} V_i n_i\sigma + \sum_{i\sigma} \sum_{j\sigma'} (\rho_{j\sigma'} - \rho_{j\sigma'}) \times (\langle ij|V_{ee}|kl\rangle - \langle ij|V_{ee}|kl\rangle \delta_{\sigma,\sigma'}) c_i^\dagger c_k$  

(3)

where the tight-binding term now includes the intra-layer next-nearest neighbour hopping $t = -0.1$ eV, inter-layer next-nearest hoppings $\gamma_3 = 0.3$ eV and $\gamma_4 = 0.15$ eV $[2]$. The terms $\rho$ and $\rho_{\text{bulk}}$ are quantum dot and bulk density matrices, respectively $[13]$. The two-body Coulomb matrix elements $\langle ij|V_{ee}|kl\rangle$ computed using Slater $p_z$ orbitals include on-site interactions, all scattering and exchange terms within next-nearest neighbors, and all long range direct interactions. We have previously tested the validity of our approach by comparing to density functional calculations and obtained good agreement$[13]$. After diagonalizing the Hartree-Fock Hamiltonian, we obtain Hartree-Fock quasi-particles denoted by the creation operator $b_{p\sigma}^\dagger$, with eigenvalues $\epsilon_p$ and eigenfunctions $|p\rangle$. We can now fill the new quasi-particle zero-energy states with electrons and solve the many-body Hamiltonian given by

$H = \sum_{p\sigma} \epsilon_p b_{p\sigma}^\dagger b_{p\sigma} + \frac{1}{2} \sum_{pqrs\sigma\sigma'} \langle pq|V_{ee}|rs\rangle b_{pq\sigma}^\dagger b_{rs\sigma'} b_{r\sigma'}^\dagger b_{s\sigma}$  

(4)

Figure 5a shows the evolution of spin states for the same system studied in Fig.3, but now obtained by diagonalizing the many-body Hamiltonian. We observe two main differences from the mean-field Hubbard results: (i) At low $\Delta V$, the antiferromagnetic configuration $S = 1/2$ is no longer the first excited state. Although the AFM-FM energy gap is still comparable to the Hubbard result, other spin excitations are now closer to the ground state due to correlation effects. (ii) The ground state spin transitions do not occur as abruptly as in a Hubbard model.
As shown in Fig.5b, the total spin of the charge neutral system (red color online, solid line with circles) evolves towards the minimum spin state $S = 1/2$ gradually between $\Delta V = 0.5 - 1.0$ eV. This is mainly due to long range interactions. As the electrons are transferred one by one from the top layer into the bottom layer, they leave a positively charged hole behind which makes it harder to transfer more electrons. We note that this behavior of gradual spin transition is also obtained within a mean-field Hubbard model with long range interactions included (not shown). Finally, in Fig.5b, we also study the effect of charging the BQD system. The asymmetry between magnetic moment of $+e$ and $-e$ charged systems reflects the correlation induced spin depolarization process that occurs in single layer TGQDs as discussed in our previous works [13].

In summary, we demonstrate that the graphene bilayer triangular quantum dots exhibit a shell of degenerate states at the Fermi level. At half filling, the shell is maximally spin polarized. By the application of a vertical electric field the total spin of the bilayer structure can be turned off or reduced to a single localized spin, a qubit isolated from contacts and free from interaction with nuclear spins. This opens new possibilities in the area of spintronics and quantum information processing.

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