Opposite Changes in Gap Width of Opposite Spin States Induced by Rashba Effect in Anti-ferromagnetic Graphene on Ni(111)

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Graphene is a promising candidate for applications in spintronics. In this paper, Density Functional Theory method is used to calculate the band structure and magnetic properties of graphene on Ni(111). Our results show that once there is anti-ferromagnetic order in graphene, an external electric field at the order of $10^9$ V/m can induce a gap width difference of tens of meV for opposite spin states near the Fermi surface.

I. INTRODUCTION

Graphene is a two-dimensional material with many interesting properties. Behavior of Dirac Fermions in graphene has been extensively studied. Particularly, magnetic properties have attracted lots of interests.

Graphene is considered a promising candidates for spintronics due to its long spin relaxation time and large spin relaxation length. Spin splitting is usually a desirable effect in potential applications. Although the intrinsic spin orbit coupling (SOC) is quite weak, considerable extrinsic SOC effects have been proposed and studied, among which the Rashba SOC resulted from broken inversion symmetry received lots of attention. The Rashba Hamiltonian can be expressed as

$$ H = \alpha (p \times \sigma) \cdot E $$ (1)

$\alpha$, $p = \hbar k$, $\sigma$ and $E$ respectively stands for Rashba strength parameter, electron momentum, Pauli spin operator and the electric filed.

Graphene/Ni system is studied here for two reasons: One is that graphene films can be conveniently prepared on Ni substrate; the other is that Ni is a common candidate for spin injection electrode and spin detection electrode. Published work suggests that the Rashba-induced spin gap can be as large as tens to hundreds of meV.

However, systematic study of the influences of Rashba effect on the Dirac fermions around the K point near the Fermi surface which greatly affect the transport properties is still lacking.

In this paper, we study few layer graphene(FLG)/Ni(111) system using density-functional theory (DFT) by considering various factors including substrate interaction, layer number and SOC.

II. METHODS

For First Principle Calculations LDA with PAW in VASP has been used to optimize the Few Layer Graphene (FLG)/Ni system, which according to previous works is effective and sufficient. The energy cut-off of optimization is 500eV and the convergence criteria is that the Hellmann-Feynman forces be less than 0.03 eV/Å for the geometry optimization. The graphene layers and upper three layers of the total six layers Ni(111) are allowed to relax. A vacuum slab more than 15 Å is applied to avoid interaction between supercells.

The original $1 \times 1$ unit cell of Ni substrate is used, onto which the $1 \times 1$ unit cell of graphene is mapped. In monolayer case the graphene is of the well studied top-fcc site, as shown in FIG. 1(b); for bilayer the bottom layer remains the same while the layer above is of hcp-fcc site, in case of more than two layers of graphene, an
AB stacking model is used. This configuration is shown in FIG.1(a).

Then SOC is included into calculation to determine the influence of Rashba effect on the properties of monolayer case.

III. RESULTS AND DISCUSSIONS

The calculated band structures along \( K - \Gamma - K \) for 1-4 layers of graphene on Ni(111) are demonstrated in FIG. 2. We perform projected band structure calculation, in which the radius of red/blue spots demonstrates the weight of \( p_z \) orbit of top layer graphene. FIG. 2(a)/2(b) display band structure near Fermi surface for majority/minority spin of monolayer case, while that of bilayer, trilayer and quadrilayer are respectively shown in FIG. 2(c)/2(d), 2(e)/2(f) and 2(g)/2(h).

As is shown in FIG. 2(a) and 2(b), the existence of Ni substrate seriously changes the band structure of graphene compared with freestanding situation with gapless Dirac cone and linear energy dispersion. Due to the broken of the equivalence of the two sublattices of graphene caused by the substrate interaction, the Dirac Cone must be destroyed and a gap about 0.25eV opens up. The dense gray bands below the gap is those of Ni 3d orbit. The ferromagnetism of Ni leads to a 0.4eV spin gap, indicating the spin-polarized characteristics of graphene layer. The detail magnetization is displayed in FIG. 3, in which red and blue circle represents magnetism of C atoms parallel and anti-parallel to that of Ni atoms while the radius stands for the magnitude respectively. For convenience, the top layer Ni substrate is drawn as large gray circles. The graphene has an antiferromagnetic order, with one sublattice, namely A, under which is a first layer Ni atom, of magnetic momentum anti-parallel (0.01\( \mu_B \)) to that of Ni, and the other sublattice, namely B, of a parallel magnetism (0.03\( \mu_B \)).

For bilayer graphene, the parabolic energy dispersion is recovered since interation between graphene layers is weak. The Dirac point is lowered by about 0.4eV, suggesting strong charge transfer and vertical electric field, which can explain the small energy gap. And the spin gap is almost immeasurable, displaying the non spin-polarized character of top layer. Our calculation of the magnetization is also one magnitude lower than monolayer case. In Fig. 2(e) to 2(f), the energy of Dirac cone is at 0.2eV. In quadrilayer case, the Dirac Cone is almost at the Fermi Level, as shown in FIG. 2(g) and 2(h). This can be understood since less charge will be transferred to upper layer. And we expectedly find no difference between spins and negligible magnet moment.

Thus from the discussion above, we conclude that the Ni substrate will result in a magnetization of graphene atoms and a vertical electric field. Both effects are weaken when layer number of graphene increase. Then Non-collinear Calculation is done for monolayer case since it has observable magnetization. We are inter-

FIG. 2: (Color online) Band structure near Fermi surface for majority/minority spin of monolayer (a) and (b), bilayer (c) and (d), trilayer (e) and (f), quadrilayer (g) and (h). The radius of red spots demonstrate the weight of \( p_z \) orbit of top layer graphene. The insets show the detail near -K point and Fermi Level.
FIG. 3: (Color online) Calculated magnet moment of graphene atoms. Red/blue circle represents C atoms with magnetism parallel/anti-parallel to that of Ni atoms and the radius stands for the magnitude respectively.

FIG. 4: (Color online) (a) Calculated band structure of monolayer graphene /Ni(111) system near Fermi level after introducing SOC. Red/blue circles stand for majority/minority spin bands, and their radius represents weight of graphene atoms’ π orbits. (b) Rashba splitting of upper (grey square) and lower (green circle) band. (c)/(d) the contribution of π orbit of sublattice A/B to the bands, the radius indicates the weight. The insets show the detail near -K point and Fermi Level.
FIG. 5: (Color online) Schematic diagram to apply our method, excluding exchange splitting. (a) with zero electric field in Rashba Hamiltonian, black lines stands for bands of both spin up and spin down. (b) with the electric field in Ni(111) case, blue/red line respectively represents spin up/down. (c) with external electric field or strengthened magnetization of graphene, the Rashba effect is enlarged and the system is converted from semiconductor to half metal.

which is approximately $0.4\text{eV}/\text{Å}^{22}\) Such effect can be tuned through extrinsic electric field perpendicular to the interface, and an electric field strong enough may completely close the gap for one spin and remarkably enlarge the gap for the other spin, which is the ideal situation in spintronics. FIG. 5 is a schematic diagram in which we exclude exchange splitting and only consider Rashba effect. FIG. 5(a) is the case with zero electric field and no Rashba effect, (b) is the Ni(111) case. It may be converted from semiconductor to half metal if we further apply external electric field or strengthen the magnetization of graphene, as shown in FIG. 5(c). Another critical implication is its application in valleytronics. Due to the linear dependence of Rashba splitting on wavevector, the degeneracy of the valley degree of freedom is broken and the shrink/enlarge effect is opposite at K/-K. This indicates that under proper configuration one spin may only transport at K and the other only at -K. Thus the Rashba effect we discuss here can also be an effective valley filter.

IV. CONCLUSIONS

In summary, graphene/Ni (111) are studied considering several factors including magnetism induced by substrate and Rashba effect. At last, the Rashba effect is found to produce a 30 meV/-60 meV extra spin gap on upper/lower band at K near Fermi surface, which can further adjusts the spin gap with Rashba effect. In addition, the possibility to make discrepancy between energy gaps of different spin is discovered and interpreted, which provides a completely new method to efficiently optimize graphene/Ni system for the application in spintronics. And the opposite effect at K/K makes it also a promising valley filter. This method is tunable and can be extended to other systems with similar characters. Actually, we expect such effect in all graphene systems with different magnetization of two sublattices. Graphene on semiconductor substrate is desirable for application and SiC is a promising candidate. Another perhaps better system is carbon nanostructures without any substrate, which can display antiferromagnetic behavior due to topological frustration\textsuperscript{23}. Graphene nanoribbon is an particularly expected choice, which is under way.

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