Proximity Effects Induced in Graphene by Magnetic Insulators: First-Principles Calculations on Spin Filtering and Exchange-Splitting Gaps

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Graphene is very attractive for spintronics [1] since long spin lifetimes are expected within this material due to its intrinsic weak spin-orbit coupling and hyperfine interaction [2]. However, inducing magnetism in graphene is still jeopardizing for its applications. One way to induce magnetic states in graphene is using magnetic substrates, e.g. transition metals Co and Ni [3]. The properties of these epitaxial films have been extensively studied, however they are grown on conducting substrates which limit graphene applications for electronic devices. Alternative possibility is to use magnetic insulating material EuO as a substrate [4]. Here we addressed this problem from first principles and report promising potential for producing high spin polarization and exchange splitting band-gap values. Our calculations were performed using density functional theory based VASP [5] and SIESTA [6] packages. Considering that Eu is a heavy element and its outer shell ($4f^7 6s^2$) contains f electrons, GGA approach fails to describe strongly correlated localized 4f electrons giving the metallic ground state of EuO, while a clear band gap is observed in experiment [7]. Thus, we introduced Hubbard-U parameter to describe the strong intra-atomic interaction in a screened Hartree-Fock like manner, which produces correct ground state of EuO.

Using the optimized structure of graphene on EuO [Fig. 1], we found that the interaction with the magnetic substrate remarkably affects the magnetic properties of graphene [8]. First, a spin polarization defined as a difference between minority and majority states normalized by the total density of states at the Fermi level may reach 24% [8]. Next, the traditional linear dispersion of the graphene band structure is modified yielding a band gap opening at the Dirac point [Fig. 2]. Interestingly, the degeneracy lifting at the Dirac point is spin dependent as can be demonstrated with a simple fit of the band structure with spin-dependent Dirac dispersion relation $E_\sigma(q) = \pm\sqrt{(\hbar v_\sigma q)^2 + (\Delta_\sigma/2)^2}$ which gives gap widths of $\Delta_\downarrow$=98meV and $\Delta_\uparrow$=134meV for minority and majority states, respectively, while the Fermi velocities $v_\downarrow$=1.40x10^6 m/s and $v_\uparrow$=1.15x10^6 m/s are also polarized [inset of Fig. 2]. The corresponding polarization, around 20% for both gaps and velocities, is very significant [8]. We also simulated the situation of internal pressure and strain by calculating the electronic properties as a function the interlayer spacing Δ_z as described in Figs. 1(a) and 1(c). It is found [8] that the impact on the band dispersion of graphene is markedly strong [Fig. 3]. When compressing the bilayer by 0.5Å, more electrons (and spins) are transferred to the graphene layer due to enhanced overlap between C p_z and Eu 4*f* orbitals. Accordingly, the Dirac point is moved deeper inside the valence bands compared to the equilibrium situation [cf. Figs. 2 and 3(a)]. In contrast, for larger layer separation, the Dirac cone is clearly seen to be shifted out from the valence band of EuO, approaching the Fermi level of the system [Figs. 3(b) and 3(c)]. Simultaneously, with the shifting of the Dirac point out of the EuO valence band, the gap between spin-up and spin-down bands is continuously reduced. Finally, for Δ_z=5Å, the spin-up and spin-down branches become almost degenerated and the Dirac point crosses the Fermi level, i.e., approaching a typical band structure characteristics of isolated graphene [Fig. 3(d)].

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References

[1] A. Fert et al, *Mat. Sci. Eng. B,* **84** (2001) 1; S. A. Wolf et al, Science, **294** (2001) 1488.
[2] D. Huertas-Hernando et al, Eur. Phys. J. Special Topics, **148** (2007) 177.
[3] A. Varykhalov et al, Phys. Rev. Lett., **101** (2008) 157601; O. Rader et al, Phys. Rev. Lett., **102** (2009) 057602.
[4] H. Haugen et al, Phys. Rev. B, **77** (2008) 115406.
[5] G. Kresse and J. Hafner, Phys. Rev. B, **47** (1993) 558; P. E. Blöchl, Phys. Rev. B **50** (1994) 17953; G. Kresse and J. Joubert, *ibid.* **59** (1999) 1758.
[6]J. M. Soler, E. Artacho, J. D. Gale, A. Garcı`a, J. Junquera, P. Ordejo´n and Daniel Sa´nchez-Portal, J. Phys. Condens. Matter **14**, 2745 (2002)
[7] N. J. C. Ingle et al, Phys. Rev. B., **77** (2008) 121202; A Mauger et al, Phys. Reports, **141** (1986) 51.
[8] H. X. Yang, A. Hallal, D. Terrade, X. Waintal, S. Roche and M. Chshiev, Phys. Rev. Lett. **110**, 046603 (2013).

Figures

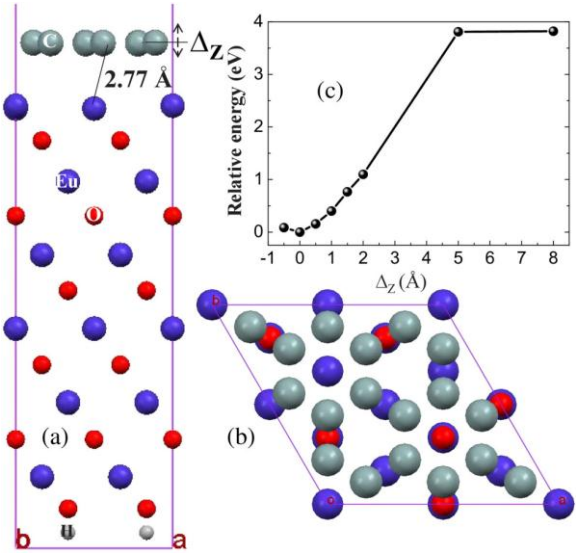

Figure 1 *(a) side view and (b) top view of the calculated crystalline structures for graphene on top of a six bilayer EuO film. The bottom of EuO is terminated with hydrogen atoms. (c) relative energy (to the optimized structure) of graphene-EuO as a function of shifting distance (_z) between the graphene and the substrate.*

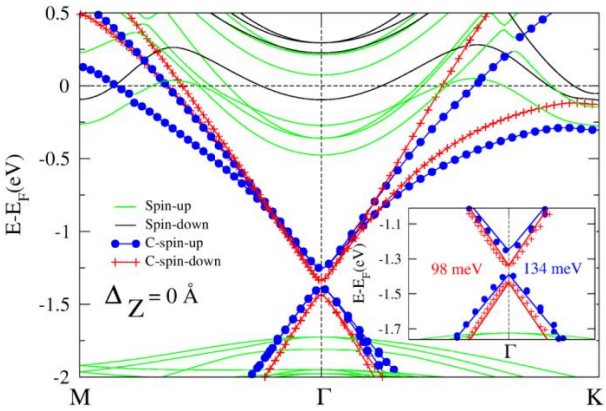

Figure 2*. Band structure of graphene on EuO. Green (blue) and black (red) represent spin up and spin down bands of EuO (graphene), respectively. Inset: zoom around the Dirac cone, the symbols correspond to DFT data while the lines correspond to the fit according to Eq. (2).*

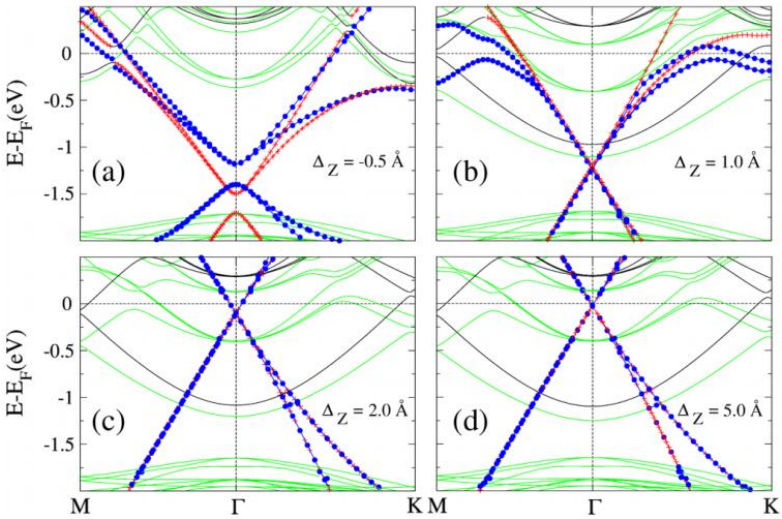

Figure 3*. Band structures for graphene on EuO with graphene shifted (a) inward (compared to optimized structure) 0.5A, (b) outward 1.0A, (c) outward 2.0 A, and (d) outward 5.0 A, respectively.*