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Interface-induced spin polarization in graphene on chromia

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Abstract — The induced spin polarization of graphene on Cr₂O₃ (001) is investigated using density-functional theory (DFT) and model calculations. The magnetic moment in graphene is a proximity effect and can be regarded as a second-order Stoner scenario, and similar mechanisms are likely realized for all graphene systems with an insulating magnetic substrate. In the absence of charge transfer, the magnetic moment would be quadratic in the exchange field, as contrasted to the usually encountered approximately linear dependence. The net magnetization of the graphene is small, of the order of 0.01 μ_B per atom, but the energy-dependent spin polarization exhibits pronounced peaks that have a disproportionately strong effect on the spin-polarized electron transport and are therefore important for spin-electronics applications.

Index Terms — Cr₂O₃ thin films, graphene, proximity magnetization, Stoner criterion, spin electronics

Induced spin polarization due to the proximity of magnetic atoms or a magnetically ordered substrate continues to be a fascinating topic. The effect has been reported for a variety of molecules on ferromagnetic substrates as well as for graphene [Dedkov 2008, Dedkov 2010, Dzemiantsova 2011, Mandal 2014, Marchenko 2015, Weser 2010, Weser 2011, Usachov 2015], also on ferromagnetic substrates. This trend towards an induced polarization in the adlayer may be rationalized in terms of the Stoner criterion [Ma'Mari 2015], and induced polarization from proximity effects is well described by mean field arguments [Dowben 1991, Mathon 1986, Schwenk 1998].

Induced polarization in graphene and other two-dimensional materials has some potential for narrow-channel conduction in a spin field effect transistor (spin-FET). For such a spin FET to actually work, based on the induced polarization from the gate, the device requires a dielectric gate with interface polarization in proximity to the graphene. Most promising would be a nonvolatile spin-FET, with a multiferroic or magnetoelectric gate dielectric, having high interface polarization like chromia [Binek 2015, Dowben 2015], where the interface polarization can be controlled by voltage [Cao 2014, He 2010, Street 2014]. There have already been several efforts to investigate the induced magnetism or spin polarization in graphene supported on magnetic insulators. For example, based on transport measurements, Wang *et al.* [2015] concluded that graphene on yttrium-iron garnet becomes magnetic, without being able to determine or even estimate the moment. The induced magnetism in graphene on magnetic insulators such as MnO and EuO substrates was investigated using density-functional theory calculations and spin-polarization was inferred in graphene in both the substrates [Ilyasov 2011, Yang 2013]. Similar studies on graphene on Fe₂O₃ indicate a significant charge transfer from graphene to surface of Fe₂O₃, which results in a significant shift of the Fermi energy away from the Dirac point.

Putting graphene onto a magnetic substrate might be expected to yield a large magnetization in the graphene, but experiment [Dedkov 2010] indicates that this is not the case. Here we show that, in spite of the small net magnetization, the energy-dependent spin polarization assumes very high values at some energies (\uparrow dominates 0 to +1 eV; \downarrow dominates 0 to -1 eV), as discussed herein. These large spin polarizations, induced in graphene, may be exploited in a magnetoelectric field-effect transistor. The extent to which magnetic atoms can polarize an adjacent paramagnetic layers is related to an interatomic exchange J^* , which translates into an interatomic exchange field $H^* \sim J^*$. The value of J^* is typically of the order of 0.1 eV, corresponding to an exchange field of 1000 T [Mohn 2003]. The size of the induced moment, $m = H^* \chi$, depends on the electronic structure of the material to be spin-polarized, described by its magnetic susceptibility [Janak 1977, Mohn 2003]

$$\chi = \frac{\chi_p}{1 - I D(E_F)} \quad (1)$$

Here $\chi_p \sim D(E_F)$ is the noninteracting or Pauli susceptibility, $D(E_F)$ is the density of states at the Fermi level, and the Stoner parameter I , of the order of 1 eV for a broad range of materials [Janak 1977], describes the Coulomb interaction of the electrons. The density of states at the Fermi level has a two-fold effect in this mechanism. First, the Pauli susceptibility is proportional to $D(E_F)$. Second, the Stoner-type interaction yields an exchange enhancement of the susceptibility that also increases with $D(E_F)$. When $I D(E_F) = 1$, then the susceptibility of Eq. (1) diverges and the system becomes ferromagnetic, the threshold being known as the *Stoner criterion*. Simple metals, such as Al, have a rather low density of states at the Fermi level, so that χ_p is small and only slightly enhanced by the interaction term $\chi/\chi_p \approx 1.36$ [Janak 1977]. For this reason, Al is only weakly spin-polarized by neighboring magnetic atoms, as exemplified by Al in MnAl, where the moment $m = 0.061 \mu_B$ per Al atom [Manchanda 2014]. Platinum is a well-known

exchange-enhanced Pauli paramagnet ($\chi/\chi_p \approx 3.8$), closer to satisfying the Stoner criterion and, in the vicinity of Fe or Co, develops a magnetic moment m of the order of $0.5 \mu_B$ per atom [Kashyap 1999].

When *graphene* is deposited onto a magnetic substrate, then the substrate also exerts an exchange field J^* . However, the electronic structure of graphene is very different from simple metals and exchange-enhanced Pauli paramagnets. In fair approximation, graphene is described by a single hopping parameter t , which describes the interatomic hybridization between the p_z orbitals and is of the order of 1 eV, somewhat dependent on strain [Wallace 1947, Pereira 2009, Kumar 2014]. The corresponding density of states at the chemical potential (the Fermi level) is low and, for charge-neutral graphene, $D(E_F) = 0$, meaning that $\chi_p = 0$ and that there is no Stoner enhancement. In other words, graphene represents an extreme case, where the Stoner criterion is maximally violated. This finding seems to be at odds with experiments that indirectly show a magnetic effect in graphene on magnetic substrate, as evidenced by transport measurements [Wang 2015]. In fact, spin polarization, not magnetic moment, is the key to spin FET and spin transport, as this is what influences the spin current.

Figure 1 shows the effect of a small interatomic exchange J^* on the density of states of graphene. The magnetic substrate exerts an exchange field on the graphene, which yields opposite shifts of the \uparrow and \downarrow subbands. The strength of the shift, and therefore the magnitude of the induced magnetic moment m , is essentially given by Eq. (1). In the most simple approximation for graphene, $D(E_F) = 0$ and the moment $m = 0$ in graphene. Yet there are two effects that contribute to a proximity magnetic moment in graphene, both explained in Fig. 1. First, in the case of surfaces terminated by metallic magnetic atoms, there is some charge transfer from the substrate to the graphene (n -doping), which means that $D(E_F) > 0$ and magnitude of the moment in the graphene layer is nonzero, i.e. $|m| > 0$. Second, even in the absence of charge transfer, which may be the case in substrates with mixed metal-oxygen termination, there is a second-order magnetic moment, because the J^* shift makes the DOS at the Fermi level nonzero.

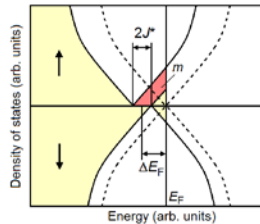


Fig. 1. Perturbative origin of the p_z graphene moment (red area). ΔE_F is the chemical-potential difference responsible for n -doping and J^* describes the exchange field caused by the Cr_2O_3

The graphene magnetic moment m is determined by evaluating the \uparrow and \downarrow areas in Fig. 1. This can be done analytically, but it is necessary to consider two cases, depending on the exchange shift J^* and Fermi-

energy shift ΔE_F due to the n -doping. In the first case, which we will see to be the more typical one, $\Delta E_F > J^*$ and the graphene spin moment, measured in μ_B ,

$$m = 2 \gamma J^* \Delta E_F \quad (2)$$

where γ is the magnitude of the first derivative of the density of states at the Fermi level, $\gamma = |\partial D(E_F)/\partial E|$. Unlike $D(E_F)$, γ is nonzero and continuous near the "Dirac" point, determined by the interatomic hopping t of the graphene $2p_z$ electrons and scaling as $\gamma \sim 1/t^2$. The magnetic moment is accompanied by a charge transfer $\Delta n = \gamma(\Delta E_F^2 + J^{*2})$.

In the opposite case of $\Delta E_F < J^*$, $m = \gamma(\Delta E_F^2 + J^{*2})$ and $\Delta n = 2 \gamma J^* \Delta E_F$. In particular, $m = \gamma J^{*2}$ for $\Delta E_F = 0$ and $m = 2 \gamma J^{*2}$ for $\Delta E_F = J^*$. The latter two equations show that the graphene proximity moment m is essentially quadratic in the exchange field exerted by the substrate. An order-of-magnitude estimate, based on $t \sim 1$ eV and $J^* \sim 0.1$ eV, is $m = 0.01 \mu_B$. This is a small but finite moment, in spite of the naive Stoner prediction $m = 0$ for graphene.

The findings of the previous sections are limited to weak exchange fields, but *ab-initio* calculations using density-functional theory (DFT) confirm these findings. Figure 2 shows the structure of the considered graphene- Cr_2O_3 thin films. The graphene grows on a Cr-terminated surface, as experimentally established [Lübbe 2009], and one Cr atom is "sticking out" somewhat and located close to the graphene.

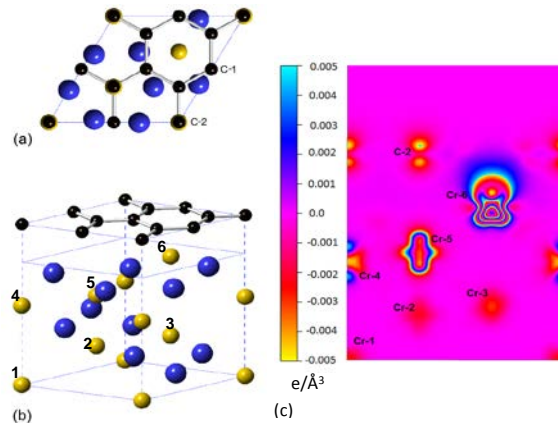


Fig. 2. Atomic structure and charge density of graphene- Cr_2O_3 : (a) top and (b) side view of used supercell, and (c) charge density difference with isovalues from -0.005 to $0.005 \text{ e}/\text{\AA}^3$. Cr, O and C atoms are shown in yellow, blue and black colors. C-1 and C-2 are the carbon atoms near to and away from the top-Cr atom, respectively. The films are separated by a supercell vacuum of 18 \AA .

Experimental lattice parameters of bulk Cr_2O_3 [Khamlich 2011] are used for the graphene/ Cr_2O_3 system, and a graphene- Cr_2O_3 distance of 2.00 \AA was obtained after relaxation. The DFT calculations were performed using the Vienna *ab-initio* simulation package (VASP) [Kresse 1993, Kresse 1996], employing the local spin density approximation (LSDA). We have used a $11 \times 11 \times 1$ Γ -centered k -point grid for the self-consistent

calculations and taken an energy cutoff of 520 eV for the plane-wave basis set. As in bulk Cr_2O_3 , the Cr atoms form antiferromagnetically coupled sublattices located on alternating (111) planes [Fiebig 2005], as contrasted to the somewhat more complicated situation in Cr_2O_3 thin films with free surfaces [Choudhary 2015].

Figure 3 shows total and local densities of states for isolated and bilayer thin films. The charge transfer of electrons from the substrate chromia to graphene shifts the chemical potential well into the chromia valence band, Fig. 3(c-d). Note that the otherwise insulating antiferromagnetic chromia might well be metallic at the graphene interface due to band bending.

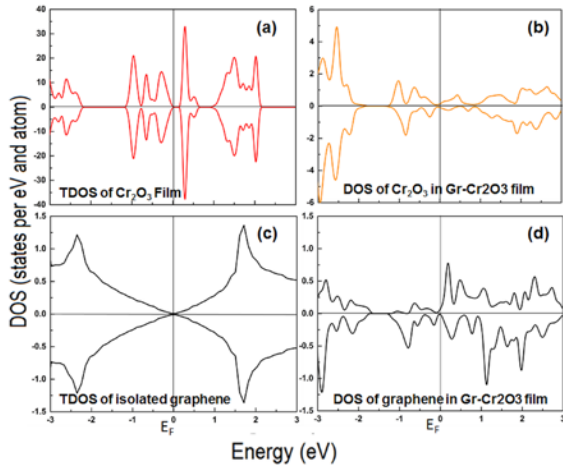


Fig. 3. Densities of states (DOS): (a) Cr_2O_3 thin film, (b) Cr_2O_3 in the bilayer, (c) isolated graphene, and (d) graphene in the bilayer (C-1 and C-2 combined). The Cr_2O_3 DOS (a-b) contain all Cr and O atoms.

The calculated graphene moments per atom are $0.0015 \mu_B$ for the C-1 sites that form the hexagon centered over the terminating Cr atom and $0.0205 \mu_B$ for the C-2 sites away from the hexagon. The estimated error for the two moments is about $0.002 \mu_B$, so that the magnitude of the C-1 moment may not be significant. These moments are small, especially compared to the Cr moment of about $2.5 \mu_B$. However, the charge transfer is significant and the spin polarizations at selected energies are large (especially near -0.8 eV). Figure 2(c) shows the charge density difference for the graphene on Cr_2O_3 system, defined as $\Delta\rho = \rho_{\text{Gr/Cr}_2\text{O}_3} - \rho_{\text{Cr}_2\text{O}_3} - \rho_{\text{Gr}}$, where $\rho_{\text{Gr/Cr}_2\text{O}_3}$, $\rho_{\text{Cr}_2\text{O}_3}$ and ρ_{Gr} are the respective charge densities of graphene/ Cr_2O_3 , Cr_2O_3 and isolated graphene at the same position in the system as for graphene- Cr_2O_3 . Charge density is transferred from the Cr_2O_3 to the graphene, predominantly to the C-2 atoms.

We see that the material is *n*-doped, by about 0.1 electrons per C atom, and exhibits a small but finite exchange energy shift of about 0.1 eV. The vicinity of Cr (and O atoms) strongly distorts the DOS of the graphene layers, creating resonance peaks in a broad energy range. When the distance between graphene and Cr_2O_3 is increased from 2 Å to 4 Å, the graphene moment drops from about $0.01 \mu_B$ to effectively (or nearly) zero.

Figure 4 shows the spin asymmetries in the Cr_2O_3 substrate (a) and in the graphene layer on top of the Cr_2O_3 (b). The spin asymmetry in isolated graphene is zero, but the Cr spins create a spin polarization in the graphene layer. Isolated Cr_2O_3 has a high spin polarization but no conduction electrons, so that both the Cr_2O_3 substrate and graphene layer are necessary for spin-polarized transport.

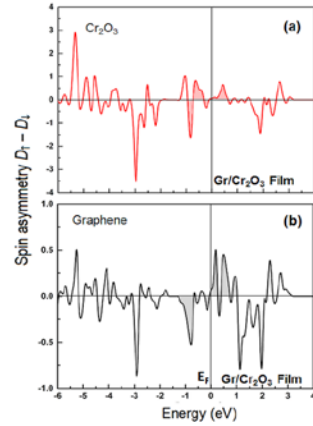


Fig. 4. Spin asymmetries in the graphene-chromia system: (a) substrate Cr_2O_3 and (b) graphene layer.

It is important to note that while the induced magnetic moment is extremely low in our calculations, there are regions where the spin polarization is high (particular in the bias region of -1 to $+1$ eV) and thus there remains the potential for nonvolatile gated spin transport at select bias voltages, mediated by a magnetoelectric. If not chromia, then some other dielectric multiferroic with high boundary polarization will have significant hybridization with graphene. Aside from Cr-terminated $\text{Cr}_2\text{O}_3(001)$ and $\text{NiO}(111)$. In these two cases, the net magnetic moment is very low, comparable to the accuracy of the DFT calculations. Since the mechanism considered in this paper is very general, the finding of nearly zero net moments also applies to other magnetic substrates. We have focused on Cr_2O_3 , because this material is a voltage-switchable antiferromagnet. Furthermore, both the charge and the moment of Cr^{3+} are larger than those of Ni^{2+} , which enhances net magnetization and spin polarization in the graphene.

Finite-temperature spin excitations and interface morphology, as discussed for Pd on yttrium-iron-garnet [Lin 2014], quantitatively modify the picture outlined in the present paper but leave the qualitative picture unchanged. Finite-temperature effects moderately reduce the spin polarization due to spin mixing [Skomski 2002], and interface features such as oxygen termination are likely reduce the spin polarization. However, as outlined in the introduction, the behavior of exchange-enhanced Pauli paramagnets such as Pd and Pt in contact with magnetic phases reflects a *high* rather than low DOS at the Fermi level. The magnetization of paramagnets like Pd is therefore Landau-Ginzburg-like, similar to the

exponential decay due to Curie-Weiss paramagnetism near surfaces [Dowben 1991, Skomski 2008].

It is instructive to compare the DOS of isolated graphene (red curves in Fig. 5) with those of graphene on Cr₂O₃ (black curves in Fig. 5) and with the analytical model predictions of Fig. 1. The chemical-potential difference is clearly visible: the Fermi level is no longer at the Dirac point but shifted to the right. The exchange splitting (J^*) is much less pronounced, corresponding to the small net moment of only 0.01 μ_B per carbon atom.

In conclusion, we have investigated how one anti-ferromagnetic dielectric oxide with high boundary polarization, used as a substrate, affects the physical behavior of graphene. The exchange field from the substrate induces a small net magnetization, but since it qualitatively changes the behavior of graphene, it has a disproportionately strong effect on electronic transport. This explains why the efforts towards developing an interface gated spin FET should consider the induced spin polarization in graphene even when the actual induced moment might be low.

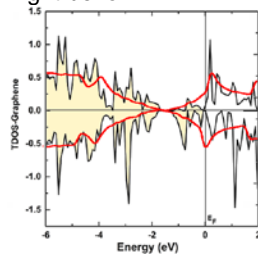


Fig. 5. Origin of graphene moment. Compared to Fig. 1, the Cr₂O₃ introduces a peak structure (black) into the graphene DOS, modifying the original graphene DOS (red). Both the n -doping ($\Delta E_F > 0$) and the spin asymmetry ($0 < J^* \ll \Delta E_F$) are clearly visible. Approximately, $J^* = 0.05$ eV and $\Delta E_F = 1.5$ eV.

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