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Research articles

Electric-field control of magnetism in graphene on chromia

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ABSTRACT

First-principle calculations are used to investigate how an external electric field controls the spin polarization in graphene on chromia, a system of interest in the area of spin field-effect transistors. Both free-standing chromia thin films and graphene-bilayers are considered. The effect of the electric field depends on the thickness of the chromia and ranges from moderately strong and linear effects to very strong nonlinear magnetoelectricity. The graphene modifies and generally enhances the nonlinear magnetoelectric effect. We also find that the external electric field drastically changes the energy-dependent spin polarization in the graphene layers, which is predicted to reach values of up to about 80%.

1. Introduction

The spin field-effect transistor (spin-FET) is a potential application in spintronics where two ferromagnet layers (source and drain) are connected by a two-dimensional semiconductor channel [1–3], in contrast to conventional FETs, where the source and drain are p- or n-type semiconductors. The concept of spin-FET was introduced by Datta and Das in 1989, who proposed that the spin can be controlled by Rashba spin-orbit interaction [1,4,5]. The spin-FET may improve the conventional FET by needing small voltages to operate and exhibiting low power consumption. Among the materials in consideration and of specific interest in this paper is graphene on magnetic substrates, where one attempts to use the high electron mobility of graphene [6] for narrow-channel conduction [6–8].

The injection and transmission of spins involve source-channel and channel-drain interfaces, and the transmission of electrons through the channel-drain interface depends on the orientation of the spins in the drain. However, the underlying spin manipulation is not an easy task. The device requires a dielectric gate, for example, a magnetic insulator, that induces some spin polarization in the graphene [9]. Chromia (Cr_2O_3) is a room temperature magnetoelectric material suitable for use as a voltage-controlled gate in spin-FETs [7,8]. Antiferromagnetic Cr_2O_3 exhibits a linear magnetoelectric (ME) effect and the linear ME coupling constant of bulk Cr_2O_3 is $\alpha = 4$ ps/m [10,11]. The thin Cr_2O_3 film has been suggested for data storage in magnetoelectric random access memories (MERAM) [12]. Recent experiments by Borisov et al. [13] have shown that the ME effect in a Cr_2O_3 film of thickness 500 nm is very similar to the bulk, but the situation in thin films may or may not be very different from that in thick films.

Stuart et al. [14] experimentally investigated the graphene-chromia system and were able to demonstrate magnetoelectric (ME) coupling in a single domain of chromia for Gr/Cr_2O_3 film. On the theoretical side, there exist a few models [6] for spin polarization in graphene using a magnetic gate, and we have recently performed *ab initio* simulations of the spin polarization in the chromia layers [15] that indicate a very complex magnetic structure, with a layer-thickness-dependent competition between ferromagnetic and antiferromagnetic couplings.

In the present paper, we use first-principle calculations to investigate the effect of an external electric field on the magnetism of free-standing Cr_2O_3 layers and of graphene on Cr_2O_3.

2. Computational details

In our density-functional calculations, we have employed the local spin-density approximation (LSDA) for the exchange and correlation functional and used the projector augmented wave (PAW) method [16], as implemented in the Vienna Ab-initio Simulation Package (VASP) [17]. We have taken the Cr_2O_3 bulk lattice parameters, $a = b = 4.9607$ Å, which yields an interface lattice mismatch of 0.03% for Gr/Cr_2O_3 [0001]. The films are constructed using the supercell approach, and the vacuum along the z-direction is taken as 18 Å. We have also investigated bulk and thin-film Cr_2O_3 using an on-site Coulomb interaction on Cr atom, using the LSDA

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+ U approach [18], but little change in magnetic properties is affected by the parameter U. This is in accordance with studies by Niranjal et al. [19], who investigated the interface magnetoelectric effect in $Fe_3O_3/BaTiO_3$. For this reason, the parameter U is not explicitly discussed in this paper.

The convergence criterion for the self-consistent calculations is 10^{-7} eV for the total energy per supercell and an energy cutoff of 520 eV is used for the electronic wave functions. The interlayer distance at the graphene-chromia interface is obtained after relaxation, performed until the Hellmann-Feynman forces on the atoms are less than 20 meV/Å. In all calculations, the electric field is perpendicular to the film plane. In present work, electric field direction from down to up (↑) represents the positive field while from up to down (↓) represents the negative field. Strength of the electric field is $\sim 10^{-9}$ V/m which is one order higher than the typical breakdown electric field of bulk chromia. The electric breakdown limit or dielectric strength depends on various real-structure parameters, such as defect density, thickness and cannot, therefore, be compared to bulk and thin films. It increases with decreasing film thickness and reaches high values in thin films in many cases [20].

3. Magnetoelectric effect in chromia thin films

Fig. 1 shows the free-standing chromia layers whose magnetoelectric behavior is investigated in this paper. The notation is the same as in Ref. [15], where we investigated the spin structure of freestanding chromia thin films in the absence of an electric field. The thinnest film (I), which contains 6 atomic layers, is ferromagnetic. The other two films (II and III), which contain 9 and 15 atomic layers, respectively, exhibit antiferromagnetic Cr-Cr interactions that lead to a ferrimagnetic spin structure.

The thinnest FM chromia film (I) shows a linear surface magnetoelectric effect described by the linear coupling constant $\alpha_s = 3.79$ ps/m, which is close to the coupling constant of bulk Cr_2O_3. Fig. 2 shows the electric-field dependence of the corresponding moment per surface Cr atom, which varies at a rate of about 0.2 μ_B per 0.1 V/Å.

By contrast, the magnetoelectric effect in the thicker films (II and III) is strongly nonlinear. The moment of film II, Fig. 3(a), remains nearly zero in a positive electric field, increasing to about 0.045 μ_B per supercell in a field of 0.5 eV/Å. In a negative field, the magnetic moment per supercell rapidly jumps to 2.7 μ_B per supercell and then remains nearly constant. Fig. 3(b) shows the magnetic moment per supercell for film III, whose magnetoelectricity is also highly nonlinear.

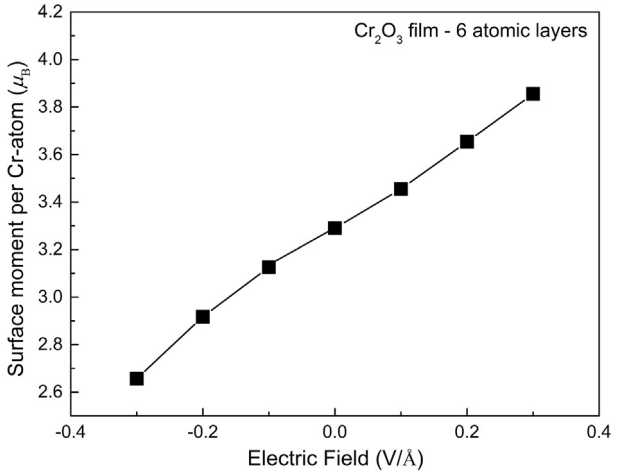

Fig. 2. The magnetic moment of the surface Cr atoms in the film I as a function of the applied electric field.

To understand the origin of the nonlinear magnetoelectric coupling, we have investigated the electron charge distribution in the electric field. Fig. 4 visualizes the charge distribution in the thickest film (III) for $E = \pm 0.3$ eV/Å. The distribution shown in the figure is $\Delta\rho = \rho(E) - \rho(0)$, that is, the respective red and green colours indicate enhanced and reduced charge densities relative to $E = 0$. Compared to metallic surfaces [21], the charge transfer rather uneven: the charge is not able to move smoothly with the electric field and the charge transfer contribution comes from all over the film. A major aspect of this discontinuity is the poor screening of the electric field in the insulating films.

4. Spin polarization and charge density in Gr/Cr_2O_3

Fig. 5 shows the schematic atomic and spin structure of Gr/Cr_2O_3. The graphene is on top of a film of type II, that is, on 9 atomic layers of Cr_2O_3. Fig. 6 compares the electric-field-dependence of the magnetic moments per supercell with and without graphene. In zero and positive electric fields, the graphene

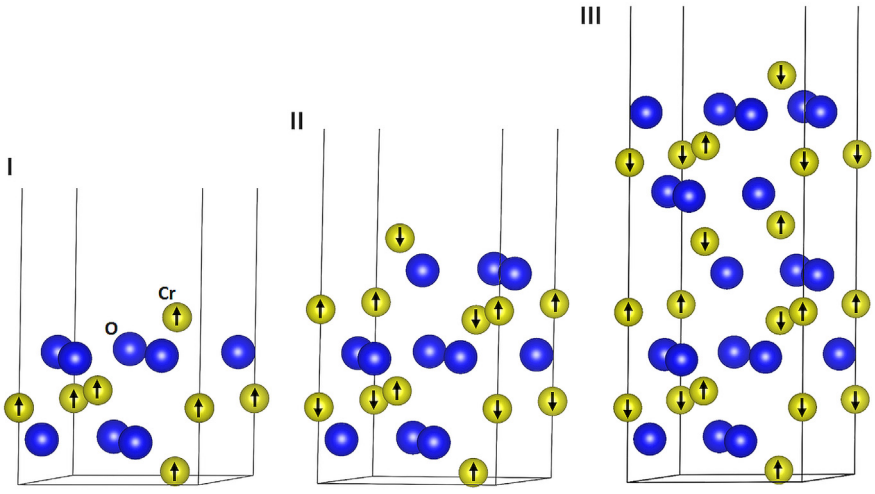

Fig. 1. Freestanding Cr_2O_3 thin films having 6 (I), 9 (II), and 15 (III) atomic layers. Cr and O atoms are yellow and blue, respectively. The total film thickness ranges from 4 Å (I) to 11 Å (III).