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Electrostatic gating enables key functionality in modern electronic devices by altering the properties of materials. While classical electrostatics is usually sufficient to understand the effects of gating in extended systems, the inherent quantum properties of gating in nanostructures offer unexplored opportunities for materials and devices. Using first-principles calculations for Co/bilayer graphene, Co/BN/graphene, and Co/BN/benzene, as well as a simple physical model, we show that heterostructures with two-dimensional materials yield tunable magnetic proximity effects. Van der Waals bonding is identified as a requirement for large electronic structure changes by gating, enabling both the magnitude and sign change of spin polarization in physisorbed graphene. The ability to electrically reverse the spin polarization of an electrode provides an alternative to using the applied magnetic field or spin transfer torque in spintronic devices, thus transforming a spin-valve into a spin transistor.

Electrostatic modulation of carrier density is not only central to electronics, as in field-effect transistors, but also controls electronic phase transitions, changing an insulator into a superconductor,[1,2] or suppressing a metal-insulator transition.[3] An upper range of the imposed electric field, $\mathbf{E}_{ext}$, to influence electronic structures can be understood from the breaking of a chemical bond on the order of 1 V/Å. Gate-induced fields of this magnitude can be attained experimentally.[4] For comparison, a dielectric breakdown in air occurs at $\sim 10^{-4}$ V/Å.

While electrostatic models[5,6] show important modifications from large field-induced changes of carrier density, the role of gating can be more complex with an inherent role of quantum tunneling effects.[7] Electrostatic gating can profoundly alter magnetic materials by inducing ferromagnetism[8–10] and controlling Skyrmions.[11] Field-induced redistribution of carriers can yield striking changes in the Curie temperature.[12]

Here we examine tunable magnetic proximity effects[13] and reveal electric control of spin-dependent properties of nonmagnetic materials. An importance of magnetic proximity effects has recently emerged in the search for Majorana states[14] and realizing exotic properties of topological insulators.[15] We look for materials systems in which gating could enable large changes in the density of states (DOS) and its spin polarization.
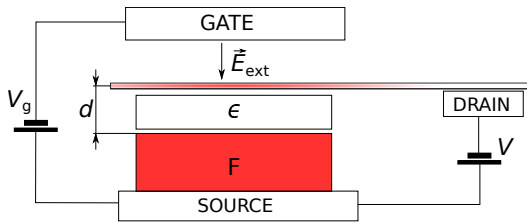

FIG. 1. Schematic of a lateral geometry. A ferromagnet, F, is separated from a graphene by a dielectric, D. The gate voltage V_g and $\mathbf{E}_{ext}$-field are marked. The color (red) depicts a proximity-effect induced spin polarization in graphene.

We focus on graphene (Gr)-based layered heterostructures deposited on a metallic ferromagnet (F) as shown in Fig. 1. The significance of this choice is two-fold: (i) such systems include van der Waals (vdW) heterostructures with atomically sharp interfaces[16] which simplify the implementation of electrostatic gating,[2,17] (ii) these are key building blocks for graphene spintronics[18] with a prospect of gate-tunable magnetic proximity effects — an important precursor for lateral spin injection needed in many applications.[8,19–27] This path to tunable magnetic proximity effects contradicts common expectations that a magnetic insulator is required to avoid a short-circuit effect of a metallic F.[28–30]

In bulk materials the proximity-induced exchange splitting and magnetization decay over a much shorter distance compared to the spin-diffusion length, which is important for spin injection.[8] Consequently, the description of the spin injection usually completely neglects equilibrium proximity effects in the nonmagnetic materials. However, in atomically-thin vdW materials the magnetic proximity length exceeds their thickness. Thus, describing spin injection should include magnetic proximity effects.[26] It is helpful to distinguish two mechanisms for magnetic proximity effects: (a) The wave functions from Gr penetrate into the insulating F (as evanescent states, since there are no states at the Fermi level, E_F, there) and acquire exchange splitting from its native ferromagnetism. (b) The wave functions from the metallic F penetrate into Gr, directly polarizing its electronic structure at the E_F.

Even the simple effect of electrostatic screening in a metal is intrinsically quantum mechanical, as it is apparent from the spin-dependent screening in F,[31,32] shown in Fig. 2 for a Co slab. The electrostatic field is screened from the interior of a metal by induced surface charges of both spin projections. However, in F the induced surface charge has inequivalent majority and minority spin contributions, leading to the changes of surface magnetic properties. The relative amount of these charge contributions[31] have a quantum mechanical origin: they depend on the spin polarization of DOS, $P = (N_\uparrow - N_\downarrow)/(N_\uparrow + N_\downarrow)$, at E_F. For Co, in which $P < 0$, most of the screening charge comes from the minority-spin channel.[33]

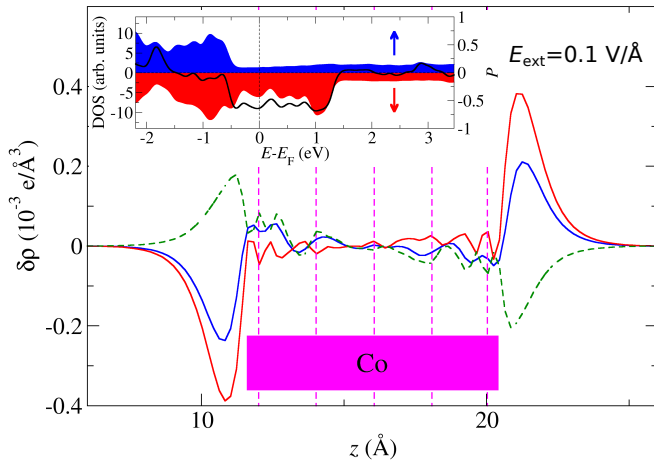For spintronics applications it is important to understand

FIG. 2. Field-induced charge rearrangement ($x - y$ plane averaged) for a 5 layer Co(0001) slab. Majority (minority) spin: red (blue) solid lines; normalized magnetization: green dashed line. Vertical lines: Co layers. Inset: majority (minority) spin DOS: red (blue) shading; DOS spin polarization: black line.
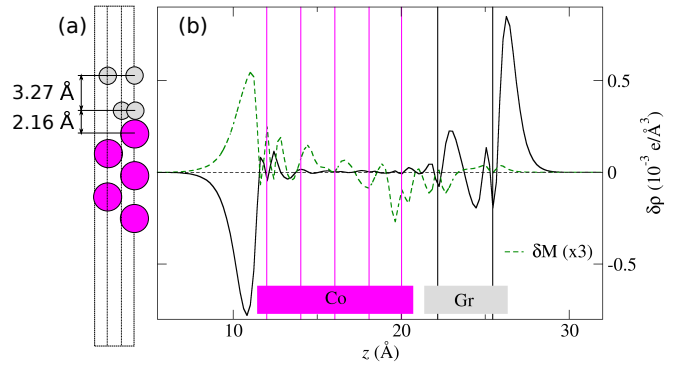

FIG. 3. (a) Computational geometry: 5 layer Co(0001) slab and a bilayer Gr (two C atoms/layer, AB stacking). (b) Field-induced charge rearrangement ($x - y$ plane averaged): sum for both spins (black), 3 × normalized magnetization (green). Vertical lines: Co and Gr layers. $E_{ext} = 0.1$ V/Å.

what happens when a nonmagnetic dielectric is attached to F, and if the magnetic properties of this system could be altered through gating. Unfortunately, there is a disconnect between many gating experiments and the lack of predictive and materials-specific methods to accurately describe them. Consequently, it is unclear what materials systems are needed to ensure that the gating will work and enable large changes in their electronic structure. For many novel materials the corresponding screening lengths are one or two lattice constants, requiring an accurate description of interfaces.

While first-principles methods provide atomically-resolved information about heterointerfaces of interest for gating experiments, standard approaches have important limitations. The use of periodic boundary conditions introduces spurious interactions with periodically repeated images of the system which needs to be corrected.[34,35] To avoid these difficulties, we implement gating[36] in a real-space density-functional code.[37] For an accurate description of bonding it is crucial to include vdW interaction, which is missing in the commonly used (semi)local functionals.[38] This is achieved seamlessly by using vdW-DF,[39] a nonlocal correlation functional.[37,40]

To explore the feasibility of gating in F/Gr heterostructures, we consider a Co (0001) slab attached to a bilayer Gr. The bottom Gr layer (depicted as a dielectric in Fig. 1) is chemisorbed (chemical bonding) to Co, but the top Gr layer is only weakly bound through vdW interactions. Correspondingly, in the bottom Gr layer there is a strong hybridization with Co and the Dirac cone is destroyed, similar to other metallic F/Gr junctions.[26] In contrast, with a weak vdW bonding Dirac cone is largely preserved in the top Gr layer. Figure 3 shows a striking difference in the response of the two Gr layers to gating. The field-induced charge rearrangement in the top Gr layer is three times larger compared to the bottom Gr layer. Essentially, the latter is electrically grounded through strong bonding with the Co metal. Unlike the pure Co slab (Fig. 2),

spin-dependent screening in both Gr layers is negligible. This seems to imply that gating can not modify the spin polarization in a nonmagnetic region, but the actual situation is more complicated.

To investigate the viability of gating, we examine the influence of the electric field on the layer-resolved DOS. As seen in Fig. 4, the adsorption of Gr changes the spin-dependent DOS of the top Co layer.[41] This is consistent with the expected magnetic softening[42,43] of a surface Co layer from chemisorbed Gr. However, for both the pure Co slab and Gr-covered Co there are no field-induced changes in DOS. In contrast to negligible DOS changes of the bottom Gr layer [Fig. 4(b)], the changes with gating in the top layer are considerable [Fig. 4(c)]. This points to a likely trend that strongly bonded heterostructures are unsuitable for gating: the chemical bonds ground the attached dielectric to the metallic F (Fig. 1), precluding charge transfer and control of P. However, the top Gr layer exhibits large field-induced changes in DOS and P.

Intuitively, large bonding distance could provide a large voltage drop, while small DOS suppresses screening of the external field E_{ext}. The resulting charge transfer for the region (top Gr layer) with a small DOS at the Fermi level $N(E_F)$ will induce appreciable changes in its electronic structure. Thus, to facilitate the tunability of P, one should seek an energy window with a small DOS in both spin channels. In Fig. 4(c) this is observed at $E_{ext} \sim -0.4$ V/Å for the vdW-bound top Gr layer, where the Dirac cone is largely preserved.

For practical applications, it is important to ascertain that this tunability is not unique to a specific system, and that such control of DOS and P can be realized at the Fermi level. To this end, we repeated the calculations with the bottom Gr layer replaced by hexagonal boron nitride (BN), an insulator commonly used in vdW heterostructures to improve their charge and spin properties, as well as to implement topological states.[16,44–48]

The comparison of Co/Gr/Gr and Co/BN/Gr (see Ref. 35 for computational geometry and bonding distances) in Figs. 5
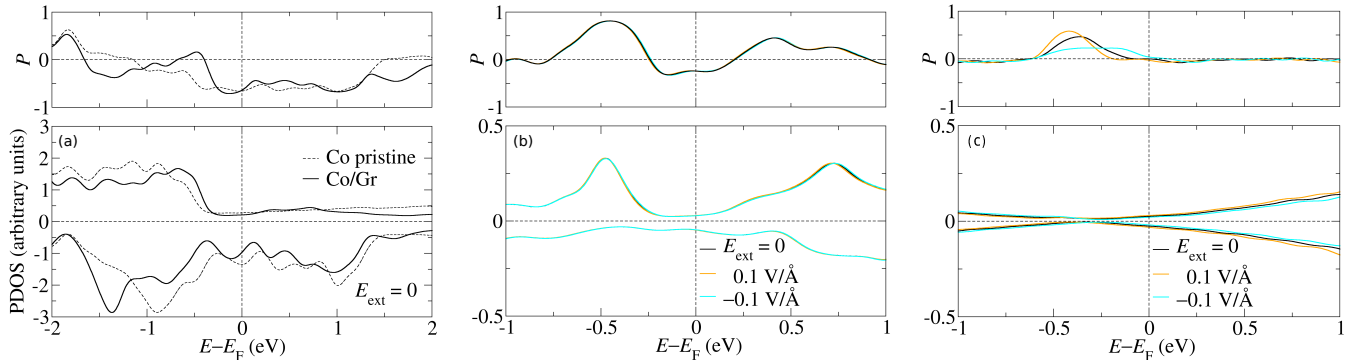
FIG. 4. (a) Projected DOS (majority, minority) and the spin polarization, P, on the surface Co atom of the pristine slab (dashed) and Co slab with attached bilayer Gr (full). Field-dependent PDOS and P on the C atoms of (b) the bottom Gr layer (attached to Co surface) and (c) top Gr layer (vdW bonded to the bottom Gr layer, chemisorbed on Co).
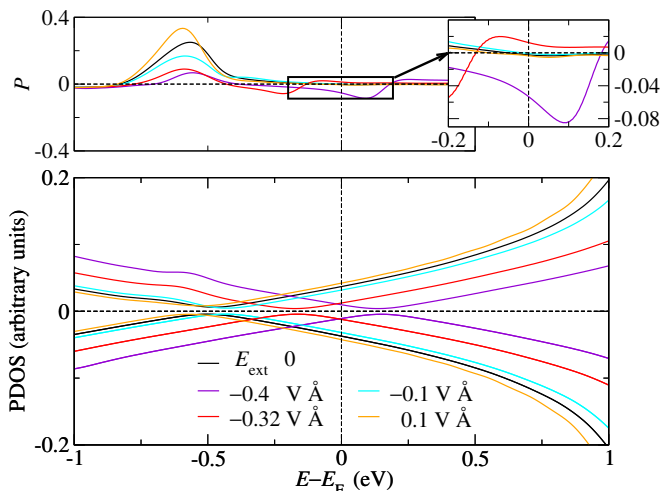

FIG. 5. (a) Field-dependent projected DOS and (b) the spin polarization on the C atoms of Co (slab)/ BN/Gr system. (c) Zoom of the spin polarization in (b).

and 4(c) shows that the replacement of the bottom Gr with BN preserves the overall DOS shape of the top Gr while slightly shifting its Dirac point. These results suggest that vdW bonding of the top Gr layer facilitates gating for two reasons: (i) It preserves the Dirac cone, maintaining a region of small DOS; (ii) This region of small DOS can be shifted in energy by E_{ext}, in contrast to chemical bonding which prevents DOS changes.

Although field-induced DOS changes seen in Fig. 5 resemble a (nonlinear in the field strength) rigid shift in energy, the behavior of P is quite complicated. Similar to the small DOS region in Fig. 4(c) at $E \approx -0.4$ eV, in Fig. 5(b) we see large changes in P with E_{ext} near the Dirac point at $E \approx -0.5$ eV. More importantly, large changes in P are also observed [Fig. 5(c)] in the range of field magnitudes that bring the Dirac point close to E_F. Surprisingly, the electric field changes both the magnitude and the sign of $P(E_F)$. This is a result of a

complicated hybridization between Co, BN, and the two inequivalent C atoms (see Ref. 35).

While moderate E_{ext} changes [from -0.32 to -0.4 V/Å in Fig. 5(c)] lead to the changes in P on the order 0.1 near E_F, we expect that even larger changes in spin polarization could be achieved at finite bias in experiments on lateral transport. Similar trends of enhanced bias-dependent spin polarization have been studied self-consistently in magnetic p-n junctions.[49]

This path towards tunable magnetic proximity effect is in contrast to the common expectations that having a magnetic insulator is required to avoid a short-circuit effect of a metallic F.[28–30] In our approach the first attached layer is indeed grounded to the metallic F, but the second layer offers unexplored opportunities for spintronic devices. Even common metallic F combined with vdW bonding could provide considerable room-temperature changes in P. With the gate-controlled reversal of spin polarization, a simple spin-valve would effectively be transformed into a spin transistor.

To predict the gating effects in systems similar to the one shown in Fig. 1, but not limited to Gr as the top layer, we can adapt a simple electrostatic model.[50] We would like to estimate the shift of the DOS for Gr relative to the F (the "ground") when an external field E_{ext} is applied by the gate. The simple calculation assumes energy-independent DOS, which is correct when the relative shift is small. We assume that charge can freely transfer between F and Gr over the typical time scales of the experiment (e.g. through tunneling) to establish thermodynamic equilibrium. Thus, the electrochemical potential is the same in F and Gr. When the gate voltage is applied, we then have $\delta E_F^1 - e\delta V_1 = \delta E_F^2 - e\delta V_2$, where δE_F^j are Fermi level shifts in the region j (1: F; 2: Gr), and δV_j are the electrostatic potential shifts under gating. The charge transferred from region 1 to region 2 per unit area is $|\sigma| = eN_j\delta E_F^j$, where N_j is the DOS per unit area in region j.[51] The shifts δV_j correspond to an induced electric field between F and Gr: $\delta E_{in} = (\delta V_2 - \delta V_1)/d$. Finally, the electrostatic boundary condition for Gr gives $\epsilon E_{in} - \epsilon_0 E_{ext} = \sigma$, where ϵ_0 is the vacuum permittivity. Combining these equations, we find,
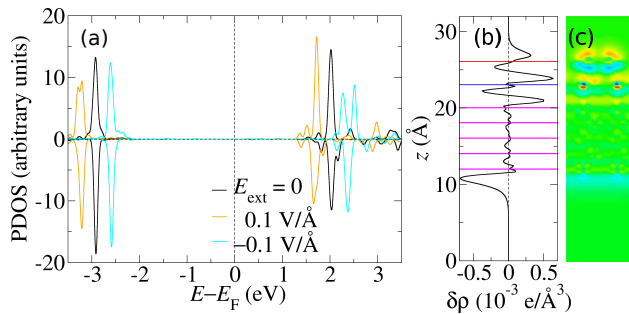
FIG. 6. (a) Field-dependent projected DOS for benzene molecule, attached to Co slab covered with BN. (b) Surface charge transfer along the z direction normal to the slab. (c) Charge transfer at $E_{ext} = 0.1$ V/Å. The color scale represent the charge accumulation and varies from 0.002 $e/Å^3$ (red) to -0.002 $e/Å^3$ (blue), shown for a cross section through a plane.

using $N_1 \gg N_2$,

$$\delta V = \epsilon_0 E_{ext} d/(\epsilon + e^2 N_2 d), \tag{1}$$

where $\delta V = \delta V_2 - \delta V_1$ (and $\delta V_1 \ll \delta V_2$). This corresponds to an effective dielectric constant $\epsilon_{eff} = \epsilon + e^2 N_2 d$.

We see an interesting interplay between classic electrostatics and quantum mechanics through the DOS of the gated material (N_2). Small N_2 is required to achieve effective gating, while large d is desirable.

The results of the model agree well with our calculations for Co/Gr, Co/Gr/Gr, and Co/BN/Gr structures. The DOS is an inherent property of the adsorbed top layer and a large d is characteristic of dominant vdW bonding. These findings suggests the importance of vdW bonding for effective gating. While our model is limited to layered structures, there are also non-layered vdW-bonded structures. For example, organic molecules are very promising building block for spintronics.[52]

To examine the importance of the periodicity of the top vdW-bonded layer, we replace Gr by a non-periodic structure. Benzene, a dominantly vdW-bonded organic molecule, is such an example. Despite its lack of in-plane periodicity, benzene attached to Co/BN shows in Fig. 6 a very similar behavior to all the previous cases where a periodic monolayer (Gr) was attached. DOS changes with E_{ext} confirm effective gating, consistent with the prediction of the layered model [Eq. (1)] requiring a small DOS at E_F of the adsorbed benzene, combined with a large vdW bonding distance. Moreover, the same vdW-bonding requirement for gating applies to adsorbed single atoms, as calculated for Xe.[35]

A weak vdW binding (physisorption) may seem incompatible with charge transfer. However, it is impossible to have "pure physisorption" with no charge rearrangement.[5,35,53] A pure vdW attraction would bring two parts of the system together which is always prevented by the counterbalancing force due to the Pauli repulsion, known as the "push back" or "pillow" effect.[5,35] Such a charge rearrangement is indeed visible in Figs. 6(b), (c), indicating that the vdW-bound part is inevitably slightly doped.

A straightforward method to detect the gate-controlled P is the lateral spin transport. The changes of P in Gr produced by the spin injector are detected by a laterally-separated F.[8,19–24] It may also be possible to directly measure proximity-induced P in Gr using the magneto-optical Kerr effect (MOKE) with the single F region in the geometry of Fig. 1. MOKE could detect very small spin polarizations, $P \sim 10^{-5}$, and has been shown to be very sensitive in determining the structural and spin-dependent properties of Gr.[54] A large background P from F itself could be removed using double modulation scheme[55] and lock-in amplifiers. One would only detect the changes of P in Gr corresponding to the chosen driven frequency of the gate voltage (say $\sim$ 1 kHz).

While we have focused on the control of the magnetic proximity effects, the underlying principles of effective electrostatic gating have broader implications. Many systems are vdW-bonded, which, according to our results, facilitates the gating of the physisorbed part of the heterostructure. This can be already seen for gating-induced superconductivity in vdW-bonded ZrNCl[2] and we expect it to be relevant for many vdW heterostructures, including those based on transition-metal dichalcogenides[56,57] and emerging Dirac materials,[58] sharing with graphene an underlying electronic structure of a Dirac cone. In these systems the requirement of a small density of states at the Fermi level for effective gating is readily achieved. With dual gating structures, similar to the schematic from Fig. 1, carrier density and the electric field can be independently changed opening opportunities to realize unexplored material properties.

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