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Electronic Structure of Graphene on Co_2FeSi Heusler Alloy

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Abstract

Due to high carrier mobility and long spin diffusion length graphene is a promising material for spintronics applications. In order to achieve effective spin transport and increase spin injection efficiency, graphene interfaces with highly spin-polarised materials, such as Heusler alloys, are needed. In this work, first-principles calculations of graphene/Co_2FeSi electronic structure are done in the frame of density functional theory. It is shown that the high percent of spin polarization in this system is combined with the linear dispersion of the π-states of graphene. The results suggest that the Co_2FeSi Heusler alloy is a promising candidate for graphene-based spintronic devices.

Keywords: Graphene; Heusler alloys; Spintronics

1. INTRODUCTION

Thanks to its exceptional physical properties, such as high carrier mobility and long spin diffusion length, graphene is a prospective material for use in spintronics [1]. For its practical applications, a graphene/ferromagnet heterostructure with effective spin transport is needed. One approach for increasing spin injection efficiency is the use of highly spin-polarized materials, such as Heusler alloys. Heusler alloys have a unique electronic structure that is different for the majority and minority spin bands. While the majority band shows metallic behavior with non-zero density of states, the minority band has a gap at the Fermi level, causing nearly 100% spin polarization.

The synthesis of the graphene/Heusler alloy interface has been demonstrated in several works [2–5]. In Ref. [2], graphene was grown using CVD onto a magnetron-sputtered single crystalline $Co_2Fe(Ge_{0.5}Ga_{0.5})$ substrate. The spin-dependent properties of the heterostructure were confirmed using depth-resolved X-ray magnetic circular dichroism measurements. Density functional theory (DFT) calculations showed that the intrinsic electronic properties of graphene and $Co_2Fe(Ge_{0.5}Ga_{0.5})$ are preserved in the vicinity of the interface.

Another way of implementing graphene/Heusler alloy to spintronics consists of utilizing graphene as an interlayer between a semiconducting channel and a Heusler alloy electrode [3,4]. A graphene tunnel barrier between the Heusler alloy and the channel modulates the Schottky barrier height and decreases the resistance—area product of the spin diode. Graphene/Heusler alloy interfaces are also interesting from the fundamental point of view: graphene interlayer between the substrate and Heusler alloy films allows investigation of the interplay between different mechanisms of Heusler alloy epitaxy [5].

One of the potential approaches to the direct synthesis of the graphene/Heusler alloy interface consists in intercalation of graphene with atoms of ferromagnetic metals and silicon. It has been shown that the use of an intercalation approach allows synthesis of ferromagnetic cobalt and iron silicides under CVD graphene [6] and graphene grown on silicon carbide [7]. Preliminary information about electronic structure and magnetic properties of such systems can be obtained from first principles. Here, we report *ab initio*

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calculation of the electronic structure of graphene on the surface of the Co_2FeSi Heusler alloy.

2. CALCULATION DETAILS

The calculations were carried out using the spin-polarized DFT as implemented in the Quantum ESPRESSO open-source package [8]. The energy cutoff for the plane waves was set to 100 Ry. The electron-core interaction was described using the projected augmented wave (PAW) [9] pseudopotentials within the general gradient approximation (GGA) [10,11]. The Brillouin zone was sampled using an 8×8×1 mesh of the Monkhorst-Pack k-points [12]. Before the electronic structure calculations, the geometry optimization was done using the Broyden–Fletcher–Goldfarb–Shanno (BFGS) algorithm. The dispersion interaction was taken into account with the use of the semi-empirical dispersion correlation DFT-D2 method of Grimme [13]. For the convenience of data interpretation, wave functions were expanded in the basis of atomic orbitals.

3. RESULTS AND DISCUSSION

In the first stage, the electronic structure of bulk Co_2FeSi Heusler alloy (space group $Fm\bar{3}m$) was calculated. The lattice constant was set to 5.64 Å following the experimental data [14]. The resulting electronic structure has shown good agreement with previous calculations [15]. To simulate the graphene/Co_2FeSi system, we put graphene on top of the Co_2FeSi(111) face to match the symmetry of graphene and the substrate. The supercell consisted of 3×3 graphene unit cells (18 C atoms) and a Co_2FeSi(111) slab (18 Co atoms, 9 Fe atoms, and 9 Si atoms). The size of the vacuum gap was 15 Å. We used Co- and Si-terminated layers of Co_2FeSi to investigate the effect of different surface structures on graphene electronic states. Then we carried out geometry relaxation for the case of graphene placed on top of Co-terminated Co_2FeSi. The geometry optimization has led to an out-of-plane corrugation of the graphene lattice. The difference between the C atoms positions lies in the range of 0.2 Å (Fig. 1a). The distance between C and Co atoms is 2.15 Å, which corresponds to the typical distance between graphene and metal surface [16]. Then, calculations for the case of graphene placed on Si-terminated Co_2FeSi were done. Geometry optimization has shown that graphene is flat and the distance between graphene and the substrate is equal to 3.15 Å (Fig. 1b), which is close to the case of graphene on cobalt silicide [17].

In the second stage, we carried out the self-consistent calculations of the band structure of the optimized supercells. The fragments of the electronic structure near the K-point of the graphene Brillouin zone are shown in Figure 2. For convenience, graphene states are highlighted. In the

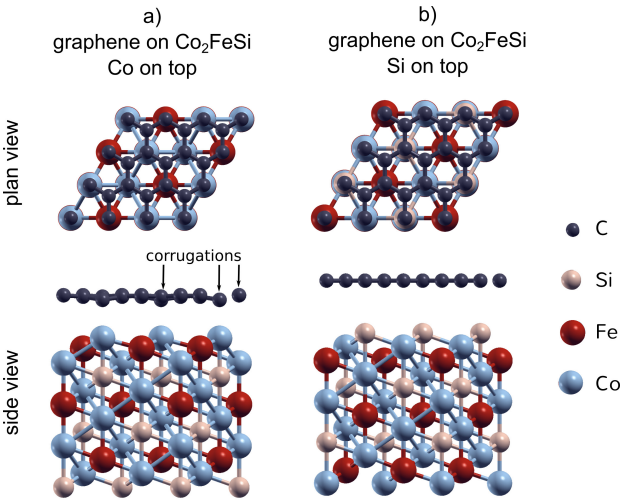

Fig. 1. The supercells used for calculation of the graphene/Co_2FeSi electronic structure with graphene on top of Co- (a) and Si-terminated (b) surfaces.

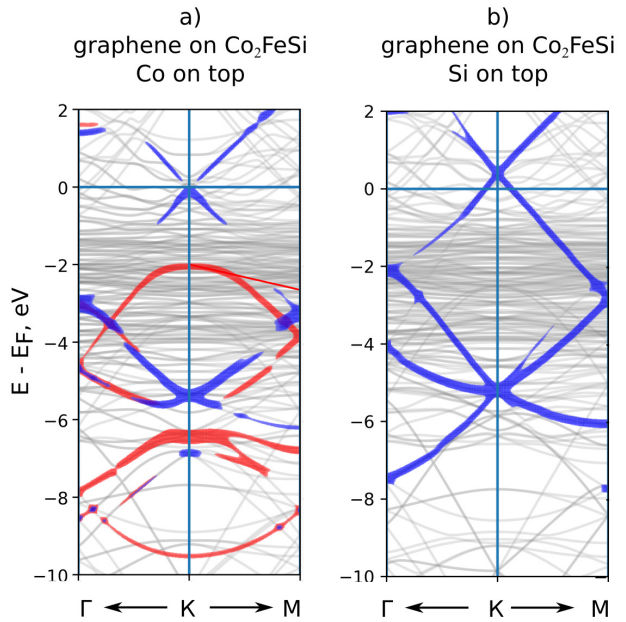

Fig. 2. Comparison of electronic structure for graphene on Co- (a) and Si-terminated (b) faces of Co_2FeSi.

case of graphene on the Co-terminated layer of Co_2FeSi, the interaction of graphene with the substrate is determined by the hybridization of the p_z states of C and d states of Co atoms (Fig. 2a). The hybridized states are shown in red color. These states lie 2 eV below the Fermi level, as in the graphene/Co(0001) system [17]. However, for those C atoms, which are not placed directly above Co atoms, linear dispersion of p_z states around the K point is preserved and the Dirac cone is seen. These states are shown in Fig. 2a in blue color. Dirac point is located in the vicinity of Fermi level. A different situation is realized when graphene is located on top of a Si-terminated surface. The electronic structure for this case is shown in Fig. 2b. Graphene states are shown in blue color. Only states with linear dispersion

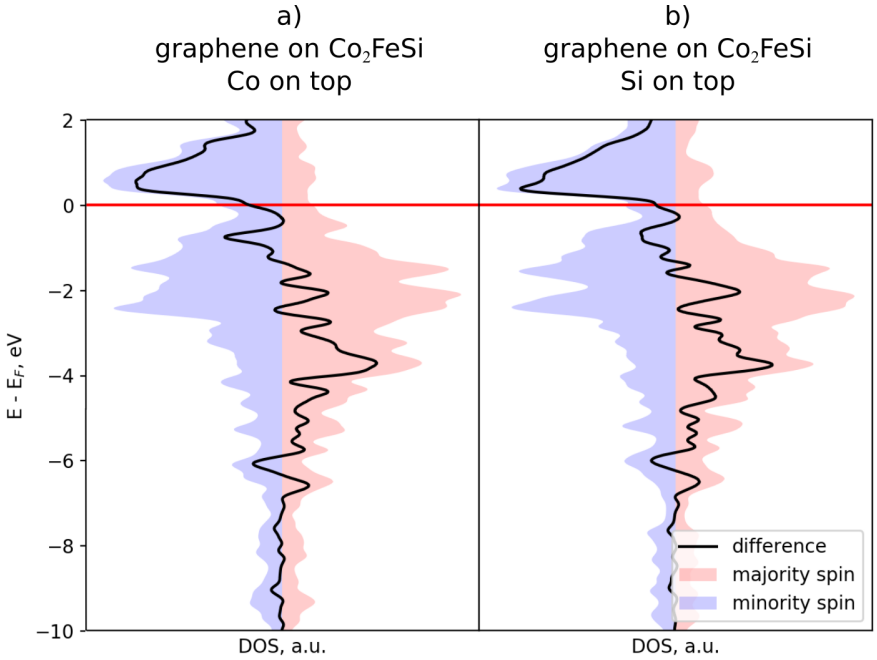

Fig. 3. Comparison of densities of states (DOS) for graphene on Co- (a) and Si-terminated (b) faces of Co_2FeSi.

exist which indicates the quasi-freestanding nature of graphene. Graphene is slightly p-doped with the Dirac point located 0.5 eV above the Fermi level.

Spin-resolved densities of states are shown in Figure 3. Spin polarization at the Fermi level is 75% for both types of surface terminations. The values of the magnetic moments in the bulk crystal and at the boundary differ insignificantly.

4. CONCLUSIONS

In this work, *ab initio* calculations of the graphene/Co_2FeSi system electronic structure are done. It is shown that different graphene states can be observed depending on the surface termination. The electronic structure of graphene on Co-terminated Co_2FeSi demonstrates both strong hybridization for the C atoms located above metal and linear dispersion for the rest C atoms. This duality may find its application in spin injection into graphene. Therefore, the combination of the high percent of spin polarization with the linear dispersion of the π-states of graphene at the vicinity of the Fermi level makes the Co_2FeSi a promising material for graphene-based spintronic devices. For the further evaluation of spin-injection efficiency, nonequilibrium spin transport calculations are to be done.

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Электронная структура графена на поверхности сплава Гейслера Co_2FeSi

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Аннотация. Благодаря высокой подвижности носителей заряда и большой длине спиновой диффузии графен является перспективным материалом для применения в спинтронике. Для достижения эффективного спинового транспорта и повышения эффективности спиновой инжекции необходимы интерфейсы графена с материалами с высокой спиновой поляризацией, такими как сплавы Гейслера. В данной работе проведены первопринципные расчеты электронной структуры графена/Co_2FeSi в рамках метода функционала плотности. Показано, что высокий процент спиновой поляризации в этой системе сочетается с линейной дисперсией π-состояний графена. Результаты показывают, что сплав Гейслера Co_2FeSi является многообещающим кандидатом для устройств спинтроники на основе графена.

Ключевые слова: графен; сплавы Гейслера; спинтроника