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Multilayer heterostructures of magnetic Heusler and binary compounds from first principles

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ABSTRACT

Employing first-principles state-of-the-art electronic structure calculations, we study a series of multilayer heterostructures composed of ferro/ferrimagnetic half-metallic Heusler compounds and binary compounds presenting perpendicular magnetic anisotropy. We relax these heterostructures and study both their electronic and magnetic properties. In most studied cases the Heusler spacer keeps a large value of spin-polarization at the Fermi level even for ultrathin films which attends the maximum value of 100% in the case of the Mn_2VSi/MnSi multilayer. Our results pave the way both experimentally and theoretically towards the growth of such multilayer heterostructures and their incorporation in spintronic/magnetoelectronic devices.

1. Introduction

Devices based on half-metallic magnetic compounds are particularly attractive for applications in spintronics and magnetoelectronics [1]. Such compounds are hybrids between usual metals and semiconductors presenting an energy gap at the Fermi level for the one spin-directions while their electronic band structure in the other spin-direction is that of a usual metal [2]. This unique feature enables the creation of a 100% spin-polarized current optimizing the performance of devices leading to versatile applications [3]. Heusler compounds consist a huge family of ternary and quaternary compounds encompassing a variety of electronic properties [4–7]. Among them several have been predicted and confirmed experimentally to be half-metallic magnets with particularly high values of Curie temperature making them ideal candidates for realistic applications [8–10].

In a recent publication (Ref. [11]) we have studied the magnetic and electronic properties of unrelaxed Co_2MnSi/CoPd multilayer heterostructures. Such junctions combine the half-metallic properties of the ferromagnetic Co_2MnSi full Heusler compound to the perpendicular magnetic anisotropy (PMA) of CoPd, a typical binary ferromagnetic compound [12]. More precisely, the full-potential nonorthogonal local-orbital minimum-basis band structure scheme (FPLO) [13] within the localized spin density approximation [14] had been employed where the Dirac-derived Kohn–Sham-like equations [15] are solved self-consistently. Our first-principles calculations on ultrathin Co_2MnSi/CoPd multilayers in Ref. [11] suggest that the spacers show behavior close to the one of the perfect bulk compounds. Co_2MnSi, although lost its half-metallicity, still exhibited a high value of spin-polarization at the Fermi level which could be further enhanced for thicker spacers. Thus the studied ultrathin Co_2MnSi/CoPd multilayers combined high values of spin-polarization with the perpendicular magnetic anisotropy of binary compounds and could find application in spintronic/magnetoelectronic devices.

2. Computational details

In the present study we expand our previous study in Ref. [11] to cover a variety of multilayer heterostructures made up of ferro/ferri-magnetic half-metallic Heusler compounds and binary magnetic compounds. Heusler compounds crystallize in the cubic $L2_1$ structure and binary compounds in the so-called tetragonal $L1_0$ structure. But as explained in detail in Ref. [11] (see Fig. 1 in the reference), if the unit cell of the $L1_0$ structure is doubled the two lattices coincide and thus if growth along the [001] direction is

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assumed there would be a common interface layer made of a 3*d* transition metal atom. We have studied the cases of Co_2MnSi/CoPd, Co_2MnSi/CoPt, Fe_2MnSi/FePd, Fe_2MnSi/FePt, Mn_2VSi/MnPd, Mn_2VSi/MnPt and Mn_2VSi/MnSi multilayer heterostructures. Co_2MnSi and Fe_2MnSi are well known to be ferromagnetic half-metallic full-Heusler compounds [8], while Mn_2VSi is a ferrimagnetic half-metallic full-Heusler compound with V and Mn atoms having antiparallel spin magnetic moments [16]. The Heusler spacers have been combined with binary compounds containing Pd/Pt atoms since the latter have a large value of spin-orbit coupling leading to large values of perpendicular magnetic anisotropy energy in the corresponding binary magnetic compounds [12]. MnSi is not expected to exhibit a strong PMA but we have included it also in our study since in that case, the Mn_2VSi/MnSi multilayer shows a smoother change along the interface since interface Mn atoms have as nearest-neighbors V/Si atoms from one side and Si atoms from the other side.

To perform our first-principles calculations, we have employed both (a) the Quantum-ESPRESSO program [17,18] and the (b) the ab initio total-energy and molecular-dynamics program VASP (Vienna ab initio simulation program) developed at the Institut für Materialphysik of the Universität Wien [19, 20]. In all calculations we have used the Generalized-Gradient Approximation (GGA) to the exchange–correlation potential in the Perdew–Burke–Erzenhof formulation [21]. Both Quantum-ESPRESSO and VASP are based on pseudopotentials (in the latter also the possibility to used projector-augmented-waves (PAW) basis is implemented) and have been extensively used in the study of materials. Both codes have intrinsic algorithms allowing the relaxations of both the dimension of the unit cell and the position of the atoms (due to symmetry reasons in the systems under study the atoms do not change their relative positions within each atomic layer and only the distance between consecutive layers changes). Both Quantum-ESPRESSO and VASP (using both pseudopotential or PAW basis) produced very similar results for all multilayers under study and thus we will present results obtained by the former.

All structures within Quantum-ESPRESSO were relaxed with a variable cell optimization with the convergence thresholds set to 10^{-4} a.u. for the energy, 10^{-3} Ry/a.u. for the forces and 0.5 kbar for the pressure. The sampling of the Brillouin zone was performed with a $4\times4\times2$ Monkhorst–Pack grid [22], since benchmark calculations with denser grids revealed no significant change in the results. In all calculations we used scalar relativistic ultrasoft pseudopotentials from the Quantum-ESPRESSO repository, while the kinetic energy cutoff for the wavefunctions and the charge density was set to 40 Ry and 400 Ry, respectively. Finally, we should note that we were not able to calculate PMA for the structures under study, since it requires convergence of the difference of the total energies for two different orientations of the magnetization [23]. PMA converges much slower with the number of **k**-points than the structural, electronic and magnetic properties presented below and our computer resources were not sufficient to achieve convergence.

3. Results and discussion

Prior to presenting our results, we should note that we have performed calculations both for ultrathin spacers (each spacer has a thickness of one unit cell as presented in Figure 1 in Ref. [11]) and much thicker multilayers (we have doubled the thickness of each spacer separately and then of both spacers simultaneously and even made tests where we have quadrupled the thickness of the spacers). In all cases the properties at the interface did not change with respect to the ultrathin multilayer heterostructures and, as will be also discussed later on, already in the case of the ultrathin films, the atoms within the spacers present bulk-like behavior highlighting the importance of the local environment in these systems. Thus in the following we will concentrate ourselves on the thin multilayers where each spacer has the thickness of one unit cell to make discussion easier to follow.

Table 1
For the multilayers under study we compare the calculated thickness of the two spacers (Heusler and binary compound) to the bulk values derived also from our calculations. All presented results are for the equilibrium ground state. For the bulk binary compounds we have imposed a cubic symmetry although real compounds are tetragonal crystallizing in the $L1_0$ structure (as shown in Fig. 1 of Reference [11] the cube is two times the primitive cell of the $L1_0$ lattice). In the last column we present the in-plane lattice constant of the multilayer shown in Fig. 1.

Lattice	Heusler		Binary		In-plane
constants (Å)	Bulk	Spacer	Bulk	Spacer	Multilayer
Co_2MnSi/CoPt	5.588	5.647	6.039	7.232	5.508
Co_2MnSi/CoPd	5.588	5.646	6.005	7.194	5.483
Fe_2MnSi/FePt	5.553	6.173	6.143	7.499	5.448
Fe_2MnSi/FePd	5.553	6.249	6.120	7.529	5.399
Mn_2VSi/MnPt	5.593	5.745	6.172	7.280	5.589
Mn_2VSi/MnPd	5.593	5.744	6.138	7.119	5.604
Mn_2VSi/MnSi	5.593	5.747	5.511	5.542	5.537

3.1. Relaxations

We will start the discussion of our results from the relaxations. First, as a reference we relaxed the lattice constants of all three Heusler compounds Co_2MnSi, Fe_2MnSi and Mn_2VSi and the results are presented in Table 1. The calculated equilibrium lattice constants are 5.588 Å, 5.533 Å and 5.593 Å. These values are close to each other and also slightly smaller than the experimental values (e.g. for Co_2MnSi the experimental value is 5.65 Å [4] but obtained values show variations depending on the sample [24]). For the binary compounds it is well known that they crystallize in tetragonal structures. To make more clear the effect of relaxations we have calculated their equilibrium lattice constants obliging them to keep a cubic symmetry (as discussed in Ref. [11] in the case of no tetragonalization $L1_0$ structure coincides with the $L2_1$ structure of Heuslers). In Table 1 we present the calculated values assuming a unit cell similar to the Heusler compounds to make values comparable. We see that in their case the equilibrium lattice constants are considerable larger that the Heusler compounds exceeding the 6 Å, with the single exception of MnSi which has a lattice constant of 5.511 Å since Si atoms have a much smaller atomic radius than Pd/Pt atoms. These values give a first hint that in the case of multilayers large relaxations of the distance of the atomic layers are expected.

Our last assumption is confirmed by our calculations on the multilayers. First, we should focus on the in-plane lattice parameter which is presented in the last column of Table 1. In the case of Co_2MnSi/CoPd(CoPt) multilayers the in-plane lattice parameter is about 0.1 Å smaller than of Co_2MnSi itself. In the case of Fe_2MnSi/FePd the difference increases to 0.15 Å, while in the case of Mn_2VSi/MnPt(MnPd,MnSi) compounds the difference between the in-plane lattice constant and the Heusler Mn_2VSi compound is even smaller. Thus it seems that in all cases under study it is the Heusler compound which imposes the in-plane lattice constant and not the binary compound. The latter compounds, with the exception of MnSi, are thus expected to show a large expansion along the growth axis to account for their in-plane contraction when used as spacers in the heterostructures under study.

In Table 1 we have included the thickness of the two spacers in the various heterostructures under study. To make these values easier to understand in Fig. 1 we present the percentage change of the thickness of the two spacers with respect to the corresponding