New spin injection scheme based on spin gapless semiconductors: A first-principles study

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Abstract – Spin injection efficiency based on a conventional and/or half-metallic ferromagnet/semiconductor is greatly limited by the Schmidt obstacle due to conductivity mismatch; here we proposed that by replacing the metallic injectors with spin gapless semiconductors can significantly reduce the conductive mismatch to enhance spin injection efficiency. By performing first-principles calculations based on superlattice structure, we have studied a representative system of Mn2CoAl/semiconductor spin injector scheme. The results showed that a high spin polarization was maintained at the interface in systems of Mn2CoAl/Fe2VAl constructed with (100) interface and Mn2CoAl/GaAs with (110) interface, and the latter is expected to possess long spin diffusion length. Inherited from the spin gapless feature of Mn2CoAl, a pronounced dip was observed around the Fermi level in the majority spin density of states in both systems, suggesting fast transport of the low-density carriers.

Introduction. – Rapid development of spintronics requires large sources of spin-polarized charge carriers, turning spin injection into a field of growing interest in recent decades. Conventionally, spin injection utilized the ferromagnet/semiconductor (SC) interface [1], for which the injection efficiency was greatly limited due to the conductivity mismatch (theoretically modeled by Schmidt et al. [2]) and low spin polarization degree of the magnetic source. Subsequently, with the emerging of half-metallic ferromagnets (HMF) that possess nearly 100% spin polarization, the HMF/SC heterostructures were proposed for enhancing the spin injection efficiency [3,4]. Nevertheless, the conductivity mismatch between the metal and semiconductor still exists. Tunnel contacts were raised as one way to circumvent this obstacle [5,6]. In some cases, the metal and semiconductor interface can form a natural Schottky barrier [7,8], but more commonly a thin oxide layer was inserted to form tunneling barriers, such as FM/MgO/SC heterostructures [9,10], where the spin injection efficiency has been greatly enhanced. On the other hand, magnetic semiconductors were also investigated to realize highly polarized spin current [11,12], but they are restricted to low temperatures and sometimes need large field bias.

Here in this letter we proposed another spin injector scheme that not only keep a high spin polarization of the injection source like HMF, but can also effectively overcome the conductivity mismatch. The scheme we considered is based on the spin gapless semiconductors (SGS), a kind of gapless semiconductor accompanying with fully spin-polarized charge carriers [13]. As seen in fig. 1 for the DOS sketch of a SGS, a zero gap was realized in one spin direction (↑) like in gapless semiconductors, but in the other spin (↓) a finite gap was opened resulting from spin splitting, thus the transport carriers were theoretically fully spin polarized like in HMF. Owing to its expected semi-conductivity, it could be more compatible with the semiconducting substrate from the impedance.

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Among the SGS candidates, the Heusler alloy Mn$_2$CoAl has been predicted to be a spin gapless semiconductor both theoretically and experimentally [14]. The reported data for the conductivity of Mn$_2$CoAl is in the order of $10^3$ S cm$^{-1}$, about two orders lower than the traditional HMF (for example, Co$_2$MnSi is $\sim 10^5$ S cm$^{-1}$ [15]). Considering that the electronic states of SGS are extremely sensitive to the atomic order, we evaluated the conductivity of fully ordered Mn$_2$CoAl by employing the BoltzTraP code [16] based on the semiclassical Boltzmann transport theory. The calculated room temperature conductivity (with respect to a constant relaxation time) for the three typical systems Co$_2$MnSi, Mn$_2$CoAl and GaAs was presented in the right panel of fig. 1. It can be seen that the conductivity of perfect Mn$_2$CoAl is much lower (nearly ten times) than that of Co$_2$MnSi, while it is very close to that of GaAs. The small conductivity mismatch between SGS and SC is promising to enhance the spin polarization degree in the SC region according to the Schmidt model [2]. In addition, the Heusler-type SGS we dealt with in the present study possess the advantage of high Curie temperature and compatibility to the industrial semiconductor from both structure and lattice constant.

The interfacial spin polarization can be evaluated by building a heterostructure model using the first-principles method, as done in most systems of HMF/SC [3,17]. In the present letter we investigated the SGS/SC (SGS = Mn$_2$CoAl, SC = Fe$_2$VAl, GaAs) heterostructures in terms of their layer-by-layer properties. We found that high spin polarization can be preserved for certain interface configurations.

**Calculation details.** – The first-principle calculations were performed within the framework of density functional theory by the pseudopotential method implemented in the CASTEP package [18,19]. The exchange correlation energy was treated under the generalized gradient approximation (GGA) [20]. Theoretically, the spin-injected system can be simulated with a superlattice structure as long as the constituting layers are thick enough to restore the bulk properties in the center part of the slabs [4]. In the present study, we investigated the heterostructure properties by constructing supercells containing several unit cells of the constituent material. Two typical kinds of semiconductor substrates were chosen: one is the nonmagnetic Heusler alloy Fe$_2$VAl, which has been experimentally proved to exhibit semiconductor-like properties [21]; the other is the stereotype semiconductor of GaAs. In fig. 2, the Mn$_2$CoAl/Fe$_2$VAl constructed with (100) interface and Mn$_2$CoAl/GaAs with (110) interface were presented separately. The in-plane lattice parameters ($a$, $b$) of the supercells were determined to be in accordance with the bulk substrate, and the parameter $c$ along the stacking direction was manually optimized. Based on the experimental value of $a_{Fe_2VAl}$ = 5.76 Å and $a_{GaAs}$ = 5.65 Å, the lattice constant set for the supercell is $a = b = a_{Fe_2VAl}/\sqrt{2}$ for the (100) geometry, and $a = a_{GaAs}$, $b = a_{GaAs}/\sqrt{2}$ for the (110) geometry. The lattice mismatch between Mn$_2$CoAl ($a = 5.8$ Å) and these two substrates is 0.7% and 2.7%, respectively. For all cases we apply a plane-wave basis set cut-off energy of 500 eV to ensure good convergence and a mesh of $12 \times 12 \times 4$ $k$-points for the (100) interface and $12 \times 8 \times 4$ for the (110) interface. All DOS curves were plotted with a smearing width of 0.05 eV.

**Mn$_2$CoAl/Fe$_2$VAl heterostructures.** – We first determine the effect of the interface on the structural and electronic properties of all-Heusler Mn$_2$CoAl/Fe$_2$VAl heterostructures. As seen in fig. 2, Mn$_2$CoAl/Fe$_2$VAl heterostructures constructed along the [001] direction consist of alternate Co-Mn, Mn-Al and Fe-Fe, V-Al atomic layers. The interfacial layers that combine the two compounds
can be either Co-Mn/V-Al (Mn-Al/Fe-Fe, the other side) or Co-Mn/Fe-Fe (Mn-Al/V-Al, the other side). We calculated both of them and found that the superlattice with Co-Mn/Fe-Fe (Mn-Al/V-Al) interface exhibits no spin polarization at all. In all-Heusler situation, as also discussed in ref. [4], the final spin polarization of the heterostructures can be simply anticipated by examining the junction components. The so-called constructive junction layers are usually those being semiconducting or ferromagnetic that bridge their bulk neighbors, like Co-Mn/V-Al (Mn-Al/Fe-Fe) here. Therefore, we studied in detail for the superlattices of [Mn$_2$CoAl/Fe$_2$VAl]$_4$ with Co-Mn/V-Al (Mn-Al/Fe-Fe) interface. As shown in fig. 3 for the [100] projected superlattice, 32 non-equivalent atoms are distributed in 16 atomic planes. To obtain the layer-resolved magnetic moment, we added the moment of each atom in one layer, and the local spin polarization was defined as 

$$P = \frac{N(\varepsilon_F)^{\uparrow} - N(\varepsilon_F)^{\downarrow}}{N(\varepsilon_F)^{\uparrow} + N(\varepsilon_F)^{\downarrow}},$$

where $N(\varepsilon_F)$ is the density of states at the Fermi level. It should be pointed out that in experiments the definition of spin polarization can vary depending on the measuring methods [22], the form here is simplified but has been widely recognized as a theoretical estimate [23]. It can be seen in fig. 3 that in the middle of each slab, the bulk moment was well reproduced, with the total moment to be 8.09$\mu_B$, deviating little from the bulk values of Mn$_2$CoAl ($2\mu_B \times 4$). When approaching the interface (in the middle and at the boundary of the figure), the moment of the magnetic layer decreased and, on the other hand, a small moment was induced in the semiconductor layer. Correspondingly, the degree of spin polarization on the Mn$_2$CoAl side remained high, but dropped quickly on the semiconductor side, implying a short spin diffusion length in this heterostructure.

The right panel of fig. 3 shows the detailed evolution of the layer-resolved DOS for four layers around the interface. In the minority spin, a large gap was observed for all layers. The overall DOS pattern showed a small difference with respect to the layer change, indicating weak interface scattering resulting from high structure similarities of this system. Still, comparing the same composition layer, for example the 6th and the 8th ones (both a Co-Mn layer), the DOS of the 8th layer revealed a less pronounced exchange splitting due to interface bonding states. The spin splitting of the semiconducting layer (from the 9th to the 12th) decreased with increasing distance from the interface, as indicated by the symmetry change of the spin-resolved DOS. Notably, the Fermi level located near to a valley in the majority spin, inherited from the gapless feature of SGS (DOS scheme in fig. 1), making it advantageous with lower carrier densities comparing to the traditional metallic injection source.

**Mn$_2$CoAl/GaAs heterostructures.** To guide future experimental efforts, here results for the Mn$_2$CoAl/GaAs heterostructures are also presented. For the GaAs substrate, the heterostructure constructed in the [100] direction lost its polarization at the interface according to our calculations. Studies on the Mn$_2$CoAl
unchanged with the bulk value of 2
Mn polarization. Right panel: the layer-resolved DOS for the corresponding layer-resolved magnetic moment and spin spectrum to different [Mn
2
succeeding discussion.

In the interface, we focus on the former condition in the atomic magnetic moments of the latter decreased much or Co-As (Al-Ga) considering their bonding ways. As the (110) interface, which can be denoted as Co-Ga (Al-As)

There are also two kinds of atom-connected ways for the connected full Heusler alloy and GaAs systems [17,25].

high spin polarization has been reported in other (110) properties of the bulk form. Consistently with our result, of the cubic phase, so they are expected to restore the properties of the bulk form. Consistently with our result, high spin polarization has been reported in other (110) connected full Heusler alloy and GaAs systems [17,25].

There are also two kinds of atom-connected ways for the (110) interface, which can be denoted as Co-Ga (Al-As) or Co-As (Al-Ga) considering their bonding ways. As the atomic magnetic moments of the latter decreased much in the interface, we focus on the former condition in the succeeding discussion.

The magnetic moment and spin polarization with respect to different [Mn
2
CoAl/GaAs] planes were given in fig. 4 in the same way as Mn
2
CoAl/Fe
2
VAl. On the Mn
2
CoAl side, the moment of each layer remained almost unchanged with the bulk value of 2μB, while the corresponding spin polarization was largely reduced compared with the bulk of 100%. From the DOS of the 3rd and 4th layers, small states emerged in the minority spin when getting near to the boundary, since the majority DOS is also very small, the spin polarization can be easily destroyed due to the compensation of the states at the Fermi level. Prominently, on the other hand, high spin polarization was observed for all layers on the semiconducting side, which is most desirable in designing a spin injector system.

The DOS of the 5th and 6th layers still present strong exchange splitting, indicating probable long spin diffusion length in this system. Like in the case of Fe
2
VAl, a pronounced dip was also found in the majority spin DOS, suggesting low carrier concentration that may facilitate fast transport of electrons.

Prospect. – To realize the application of the above scheme, the first step is to fabricate well-ordered SGS films on SC substrates. Recently, Mn
2
CoAl films oriented in the (100) direction was deposited on both silicon and GaAs substrates, exhibiting ferromagnetism and semiconducting-like transport properties [26,27]. Future work will concentrate on enhancing the structure ordering and further controlling the film growth direction and terminated layers, which greatly affects the injected spin polarization degree according to above discussions.

Summary. – Using first-principles density functional calculations, we have investigated the spin injection in two representative systems of Mn
2
CoAl/SC (SC = Fe
2
VAl, GaAs), based on the assumption that SGS/SC can reasonably enhance the spin injection efficiency by reducing the conductivity mismatch. The computed results showed that systems of Mn
2
CoAl/Fe
2
VAl constructed with (100) interface and Mn
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CoAl/GaAs with (110) interface were favored for maintaining high spin polarization. Particularly, in the Mn
2
CoAl/GaAs system, a high degree of spin polarization was achieved in the semiconducting region, implying a long spin diffusion length. Remarkably, in both systems, the layered DOS reveal the spin gapless feature with a dip in the majority spin, which means that the transport carriers should be relatively low. This may give rise to higher mobility of the carriers compared to the traditional metallic injection system.

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