Half-metallic behavior of Co$_2$MnSi/Co$_2$MnAl/MgO interface and its coherent tunneling conductance

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Abstract. We study electronic and transport properties of Co$_2$MnSi/Co$_2$MnAl/MgO(001) junctions on the basis of the first-principles density functional calculations. We found that an insertion of thin Co$_2$MnAl layers into a Co$_2$MnSi/MgO junction can eliminate interface states in the minority-spin gap and provide a 100% spin-polarization at the interface. A transfer of electrons from the minority- to majority-spin states of interfacial Mn atoms plays an important role to recover the half-metallic gap in the MnAl-terminated interface. Furthermore, the magnetic tunnel junction with Co$_2$MnSi/(Co$_2$MnAl)$_n$/MgO(001) ($n=1$∼$3$) retains coherent tunneling of the $\Delta_1$-band electrons in parallel magnetization, which is in contrast to that with Co$_2$MnSi/(Co$_2$CrAl)$_n$/MgO(001) where the tunneling of the $\Delta_1$-band electrons is reduced significantly at the Co$_2$CrAl layer.

1. Introduction
Metal-insulator heterojunctions have received much attention in spintronics owing to their possibility to derive a spin-polarized current from ferromagnetic metals. This feature is of great importance in spin-dependent devices, such as a tunneling magnetoresistance (TMR) device[1], which is essential for creation of ultrahigh density magnetic random access memory. The half-metallic ferromagnets (HMFs) [2] are one of key materials in obtaining huge TMR. Among many theoretical predictions of the HMFs, Co-based full Heusler alloys are promising materials to be applied as a source of the spin-polarized current because of their high spin-polarization and high Curie temperature[3]. In particular, Co$_2$MnSi is experimentally proved to be half-metallic. A large TMR ratio of 570% has been observed at 2K for the magnetic tunnel junction (MTJ) of Co$_2$MnSi/AlO$_x$/Co$_2$MnSi[4].

So far, the MTJs composed of the half-metallic Co-based full Heusler alloys were fabricated, and the TMR ratio has been improved steadily[5, 6, 7]. However, experimentally observed TMR ratios at room temperature are still smaller than that expected for MTJs with ideal HMFs. Recently, Chioncel, et al. investigated density of states of HMFs by using the dynamical mean-field theory (DMFT) and suggested that formation of non-quasiparticle states in the minority-spin gap at finite temperature reduces the spin-polarization and causes tunneling conductance for MTJs in anti-parallel magnetization[8]. However, recent experiments on x-ray photoemission spectroscopies of Co$_2$MnSi [9] show no significant temperature dependence in the valence-band photoemission spectra, which is in contrast to the theoretical prediction by the DMFT calculations[8]. On the other hand, Mavropoulos, et al. suggested that interface states appeared in the minority-spin gap at the interface of MTJs can also contribute to the tunneling
conductance in the anti-parallel magnetization through spin mixing processes such as magnon excitations and inelastic scattering[10]. We consider that the effect of interface states can be excluded if completely half-metallic HMFs/insulator heterojunctions is fabricated.

In our previous study[11], we found that the CrAl-terminated interface of Co$_2$CrAl/MgO(001) junction is half-metallic without any interfacial states in the minority-spin gap in both sides of the junctions, while the MnSi termination of Co$_2$MnSi/MgO(001) junction looses the half-metallicity of bulk Co$_2$MnSi. Furthermore, we have proposed that the junction inserted one-monolayer (one Co-layer and one CrAl-layer) of Co$_2$CrAl between Co$_2$MnSi electrode and MgO barrier can retain both the half-metallic character and the tunneling conductance through the $\Delta_1$ channel. However, tunneling conductance of a MTJ with Co$_2$MnSi/Co$_2$CrAl/MgO(001) junction decreases compared to that of a MTJ without Co$_2$CrAl layer, because Co$_2$CrAl has no $\Delta_1$ band around the Fermi level. The $\Delta_1$ band in the majority-spin state is also essential to obtain a large TMR ratio for MgO-based MTJs[12]. In this paper, we propose that an insertion of a Co$_2$MnAl layer instead of the Co$_2$CrAl into the Co$_2$MnSi/MgO(001) junction is effective to form the half-metallic interface without any reduction of the tunneling conductance owing to the additional layer.

2. Computational Details
Calculations of supercells consisting of Co$_2$YZ (YZ = MnSi, MnAl or CrAl) and MgO have been done using the Quantum-ESPRESSO package[13] with the ultra-soft pseudopotentials and a plane-wave basis set. We adopt the generalized gradient approximation [14] for the exchange and correlation term. The cutoff energy for the wave function and charge density is set to 30Ry and 300Ry, respectively. The number of $k$ points is taken to be 10×10×1 for all cases, and Gaussian smearing method with a broadening parameter of 0.01Ry is used. For conductance calculations, we consider an open quantum system consisting of a scattering region corresponding to MgO and a junction with Co$_2$YZ attached to left and right semi-infinite electrodes corresponding to bulk Co$_2$YZ[11]. We solve scattering equations for some fixed in-plane wave vector $k_{//} = (k_x, k_y)$ and spin index by an approach of Choi and Ihm[15].

The in-plane lattice parameter of the tetragonal supercell for Co$_2$YZ/MgO(001) junction (YZ = MnSi, MnAl) is fixed at 3.99Å and 4.03Å for YZ=MnSi and MnAl, respectively. These values correspond to $a_0/\sqrt{2}$, where $a_0$ is the lattice constant of the bulk Co$_2$MnSi(5.65Å) and Co$_2$MnAl(5.69Å). The Co$_2$YZ/MgO(001) interface has two types of termination on Co$_2$YZ, namely, the Co termination and the YZ termination. Furthermore, each of them has three possible structures, in which atoms terminating Co$_2$YZ are positioned on top of O atoms (O-top), Mg atoms (Mg-top) or hollow sites of MgO(001) surface. We adopted the YZ-terminated O-top configuration as a Co$_2$YZ/MgO interface structure, because the YZ terminations of the Co$_2$YZ/MgO(001) junctions are thermodynamically stable as compared with the Co terminations[11]. We prepare the supercell of multilayer containing 9 atomic layers of MgO and 15 atomic layers of Co$_2$YZ, respectively.

3. Results and Discussions
Figure 1(a) shows the spatial distribution of the spin-polarization at the Fermi level of the Co$_2$MnSi/(Co$_2$MnAl)$_1$/MgO(001), Co$_2$MnAl/MgO(001) and Co$_2$MnSi/MgO(001) junctions evaluated from the local density of states (LDOSs) projected on to each atomic sphere. In the case of the Co$_2$MnSi/MgO(001), the spin-polarization degrades significantly at the interfacial region, and the half-metallic character does not recover over 6 atomic layers of Co$_2$MnSi from the interface with MgO because of non-bonding Mn 3d states (especially $d_{yz}$ and $d_{zx}$) appearing around the Fermi level[11]. The spin-polarization in the MgO slab region ($z \geq 0.0$) is due to the metal induced gap states appeared in the region close to the interface. In contrast, in the case of Co$_2$MnSi/(Co$_2$MnAl)$_1$/MgO(001) the half-metallic behavior is observed throughout the
The spin-polarization at the Fermi level projected onto each atomic sphere as a function of the distance from interface for Co$_2$MnSi/(Co$_2$MnAl)$_1$/MgO(001), Co$_2$MnAl/MgO(001) and Co$_2$MnAl/MgO(001), respectively. Schematic figures of the supercell of the Co$_2$MnSi/(Co$_2$MnAl)$_1$/MgO(001) junction are shown above the spin-polarization. Local density of states of Mn 3$d$ as a function of energy relative to the Fermi energy. The sign of LDOS indicates the majority-spin (positive) and minority-spin (negative) states. Logarithmic plot of the averaged majority-spin conductance over $k// = (k_x, k_y)$ at the Fermi level in $e^2/h$ unit as a function of the number of Co$_2$Y$Z$ layer $n$ for the Co$_2$MnSi/(Co$_2$Y$Z$)$_n$/(MgO)$_n$/Co$_2$MnSi MTJ ($YZ$=MnAl,CrAl) in the parallel magnetization.

interfacial region, indicating that insertion of the Co$_2$MnAl layer between Co$_2$MnSi electrode and MgO barrier can eliminate interface states in the minority-spin gap at the Fermi level. In the case of the Co$_2$MnAl/MgO(001), the spin-polarization is not unity even far from the interface because the bulk Co$_2$MnAl is not half-metallic with the spin-polarization of 70–80%. This can be confirmed by the LDOSs of Mn 3$d$ in bulk Co$_2$MnAl in Fig.1(b) (dotted broken lines), where the Fermi level crosses the valence band edge in the minority-spin state. On the other hand, the spin-polarization at the Co$_2$MnAl/MgO(001) interface is relatively high compared to the bulk one. We consider that the high spin-polarization at the Co$_2$MnAl/MgO(001) interface can be explained by a similar mechanism to that of the Co$_2$CrAl/MgO(001) interface[11], i.e., electron transfer from the minority- to majority-spin states at the interface.

In general, formation of non-bonding states at interfaces causes a transfer of electrons. In this case, minority-spin electrons of interfacial Mn 3$d$ non-bonding states transfer to the majority-spin Mn 3$d$ states, because the bulk Co$_2$MnAl has unoccupied majority-spin Mn 3$d$ states and can accommodate additional electrons. The electron transfer push up the non-bonding minority-spin Mn 3$d$ states, and reconstruct the energy gap for the interfacial Mn. In fact, the local spin moment of Mn in the MnAl-terminated interface (3.75$\mu_B$) increases from the bulk one (2.95$\mu_B$) owing to the electron transfer. However, the Fermi level of interfacial Mn 3$d$ still cross the valence band edge in the minority-spin states(see broken lines in Fig1(b)), leading a ~90% spin-polarization at the Co$_2$MnAl/MgO(001) interface. On the other hand, in the case of the Co$_2$MnSi/(Co$_2$MnAl)$_1$/MgO(001), an additional valence electron of Co$_2$MnSi compared to Co$_2$MnAl lifts the Fermi level of the interfacial Mn 3$d$, and positions it at the middle of the energy gap(see solid lines in Fig1(b)). Thus, the Co$_2$MnSi/(Co$_2$MnAl)$_1$/MgO(001) junction has a complete spin-polarization throughout the interfacial region.

Then, we discuss transport properties of the Co$_2$MnSi/(Co$_2$Y$Z$)$_n$/MgO/(Co$_2$Y$Z$)$_n$/Co$_2$MnSi (001) MTJs (Y$Z$=MnAl,CrAl) (n=0~3). Figure 1(c) shows the averaged majority-spin
conductance over in-plane wave vector \( k_{//} = (k_x, k_y) \) at the Fermi level for the MTJ in parallel magnetization. The averaged conductance for \( YZ=\text{CrAl} \) decreases rapidly with increasing number of adlayer \( n \), while for \( YZ=\text{MnAl} \) the averaged conductance keeps same order of magnitude against \( n \). Since \( \text{Co}_2\text{CrAl} \) has no \( \Delta_1 \) band at the Fermi level in the majority-spin state, the reduction of the tunneling conductance for the \( \Delta_1 \) channel of \( \text{Co}_2\text{MnSi} \) electrode is significant. On the other hand, \( \text{Co}_2\text{MnAl} \) has an additional valence electrons compared to \( \text{Co}_2\text{CrAl} \), and the Fermi level of \( \text{Co}_2\text{MnAl} \) lies in the bottom of the \( \Delta_1 \) band. Thus, electrons with the \( \Delta_1 \) symmetry can get through the \( \text{Co}_2\text{MnAl} \) layer without reflection, and the MTJ having \( \text{Co}_2\text{MnSi}/(\text{Co}_2\text{MnAl})_n/\text{MgO} \) interface can retain relatively large tunneling conductance in the parallel magnetization. The large tunneling conductance in majority-spin state is crucial in obtaining huge TMR ratio in MTJs using half-metal.

4. Conclusions
In this paper, we have studied electronic and transport properties of the \( \text{Co}_2\text{MnSi}/(\text{Co}_2\text{MnAl})_n/\text{MgO}(001) \) junction \((n=0\sim3)\) on the basis of the first-principles density functional calculations. We have obtained a 100% spin-polarization at the MnAl-terminated interface of the \( \text{Co}_2\text{MnSi}/(\text{Co}_2\text{MnAl})_1/\text{MgO}(001) \) junction. The transfer of electrons from minority- to majority-spin states in Mn 3d plays an important role to make the half-metallic gap in the MnAl-terminated \( \text{Co}_2\text{MnAl}/\text{MgO} \) junction. Furthermore, we have shown that the junction inserted thin \( \text{Co}_2\text{MnAl} \) layer between \( \text{Co}_2\text{MnSi} \) electrode and MgO barrier can preserve large tunneling conductance as compared with that with \( \text{Co}_2\text{CrAl} \) adlayer. Fabrication of half-metallic \( \text{Co}_2\text{MnSi}/\text{Co}_2\text{MnAl}/\text{MgO}(001) \) junctions will be possible experimentally by depositing a few layers of \( \text{Co}_2\text{MnAl} \) on \( \text{Co}_2\text{MnSi} \) electrode or MgO barrier, and is worth further investigations.

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