Ab-intio compositional tuning of the functionalities in Heusler materials

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A diversity of the magnetic and transport properties in Heusler compounds suggests them as a suitable source of materials used in various technological fields. Our group engineers their functional characteristics based on the first-principles electronic structure methods. The recent focus was given to the Mn-rich Heusler compositions, which provide unusual magnetic and transport phenomena, discussed below.

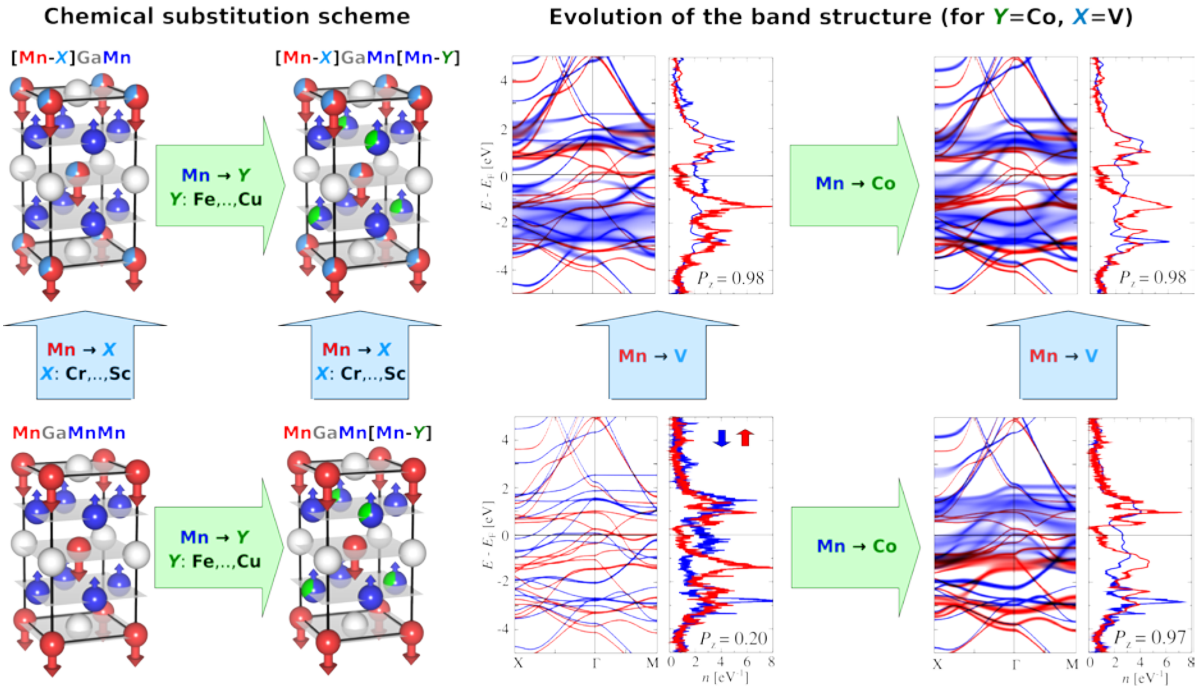

*Fig. 1. Random substitution of Mn in tetragonal Mn_3Ga Heusler compound (**MnGaMnMn**: **Mn** in 2a (red), **Ga** in 2b (gray), **Mn** in 2c/2d=4c (blue) Wyckoff sites; arrows indicate atomic magnetic moments) either by small amount of **Y**=Fe,..,Cu (**MnGa[Mn-Y]Mn**) or by **X**=Cr,..,Sc (**[Mn-X]GaMnMn**) elements, or in combination (**[Mn-X]Ga[Mn-Y]Mn**), produces the electron localization (broadened regions in corresponding band structure diagrams) within the energy windows, which appear to be different in different spin-channels (blue – spin-down, red – spin-up). One of these windows (spin-down) is located at the E_F whereas those for spin-up – below or above. This leads to a strong suppression of the DC conductivity in the spin-down channel compared to the spin-up one.*

Half-metals. For several decades the half-metallicity permanently remains in the focus of the intensive research in the field of spintronics. Being a peculiar electronic state, with a semiconducting band gap in one spin-channel, so that the conducting electrons belong to a another single spin-channel only, it provides an excellent base for the modern spintronic elements in which electric current can be efficiently controlled by the magnetic field. Our recent first-principles studies of the random chemical occupation which typically occurs in polyatomic systems such as Heusler alloys, have shown a principal possibility to manipulate almost independently by different components of the linear-response conductivity tensor. These effects might provide an alternative resource of materials with high spin-polarization as it is often required in spintronics. Their principal difference from the conventionally exploited half-metals is the presence of the so-called spin-selective localization [1-3] which implies a strong disorder scattering of the conducting electrons in one spin-channel and extremely weak scattering in another one. Being based on the different mechanism, their high spin-polarization can then be combined with additional properties which normally are incompatible with in the ordered systems, such as conventional half-metals – high spin-polarization and broad variability of their magnetization combined with a strong magnetocrystalline anisotropy. Fig.1 shows how different chemical substitutions of Mn in Mn_3Ga by other 3*d* transition elements lead to the localization of the spin-down electronic states in the vicinity of the Fermi energy. By the first-principles calculations of the spin-resolved residual conductivities based on the Kubo-Greenwood formalism, we have shown that the spin-polarization P_z (along the *c*-axis of a tetragonal unit cell) might be dramatically increased by using already a very small substitution rates. In the metallic regime P_z depends much stronger on the ratio of the spin-resolved electron mobilities rather than on spin-resolved concentrations of conducting electrons. Our latest studies on other Heusler systems, for example $Mn_{3-x}Pt_xGa$ series, indicate that the spin-selective localization is rather widely spread phenomenon, which occurs in a large number of tetragonal and cubic Mn-based Heusler alloys.

Noncollinear magnets. Noncollinear distribution of the magnetization gains nowadays a special focus initiated by novel principles of the MRAM technologies. Especially interesting are the so-called magnetic Skyrmion structures: long-range vortices formed by the interplay of several factors, among which the spin-orbit coupling and the absence of the inversion symmetry are the most necessary ingredients. Recently, we have computed and analyzed these ingredients for the group of tetragonal Mn-based alloys [4], which suggests the Skyrmion order in Mn_2RhSn Heusler material (Fig.2):

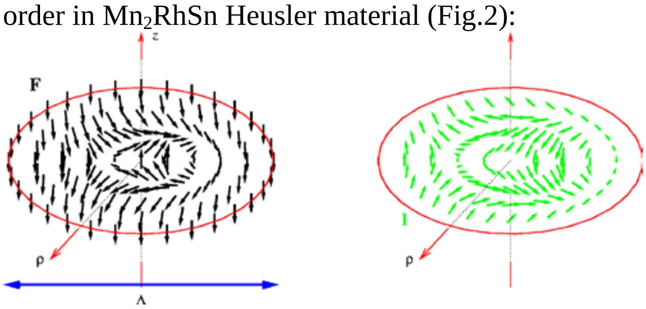

Fig. 2: Schematic view of the magnetic skyrmion phase in Mn_2RhSn. The z-axis is oriented along the tetragonal c-axis. Right: ferromagnetic mode; left: antiferromagnetic mode. The chiral twisting length estimated by micromagnetic model is Λ~130 nm.

Antiparallel alignment of the magnetic moments in two different Mn sublattices is typically encountered in most of the Mn_2YZ compounds, but not all of them exhibit noncollinearity. The most significant exchange coupling between the nearest Mn_I and Mn_{II} atoms in different Wyckoff sites is characterized by a large exchange constant (J_I~−20 meV) that leads to a typical collinear ferrimagnetic state. However, we have identified several systematic cases (tetragonal Mn_2RhSn, Mn_2PtIn and Mn_2IrIn), in which the collinear ferrimagnetic order cannot explain the measured saturated magnetization. In this systems we figured out that their collinear order can be perturbed by the next important interaction *j* between the next-nearest planes, e.g., between Mn_{II}-*Y* planes (Fig.3).

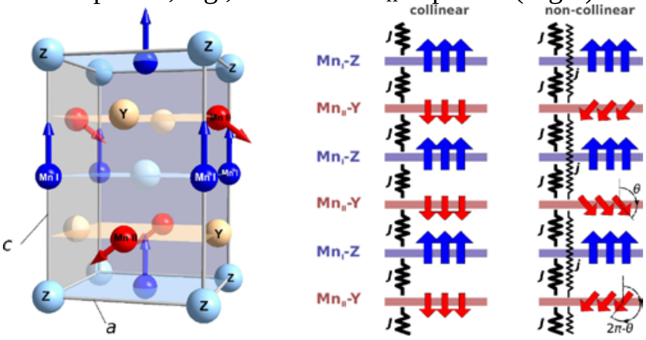

Fig 3. Mn_2RhSn unit cell: two types of Mn atoms (red and blue); arrows indicate their magnetic moments. Classical exchange picture explains the short-range magnetic canting: additional antiparallel coupling (j) competes with the strongest nearest neighbor antiparallel coupling (J).

This interaction is antiparallel due to its indirect origin realized through the main-group element *Z* (super-exchange). Since *j* tends to rotate the moments of the nearest Mn_{II}-*Y* planes antiparallel to each other, it competes with the strong antiparallel exchange *J* and may then result in a nontrivial canting angle ($\theta \neq 0°;180°$). The relevant θ-dependent part of the Heisenberg Hamiltonian contains only two types of antiparallel interactions $H(\theta)=-J\cdot\cos\theta-½\cdot j\cdot\cos2(\pi-\theta)$, the first term being the coupling of the nearest planes (Mn_I-*Z* with Mn_{II}-*Y*) and the second – of the next-nearest (Mn_{II}-*Y*) ones. The extrema of the $H(\theta)$ function, $\theta_{1,2}=180°\pm\arccos(J/2j)$, give a noncollinear solution for $j > ½\cdot J$, i.e. the canting occurs only if the next-nearest exchange *j* is sufficiently strong. Parameterized by the *ab-intio* *J* and *j* exchange constants, this model reasonably describes the θ-dependence of the total energy exhibiting two minima at $\theta_{1,2}=180°\pm55°$, which was also confirmed by neutron diffraction ($\theta_{1,2}=180° \pm 58.9°$).

References

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