Magnetism in Manganese-based Heuser Compounds

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Heusler compounds are well-known and intensively studied due to their diverse properties, that is caused by the large variety of underlying interacting physical mechanisms. In this context, theoretical studies based on density functional theory have been performed treating experimentally known in parallel as yet not investigated stoichiometries. The systematically conducted theoretical studies thus were capable of modelling the experimental findings, and to elucidate on the so far unclear aspects of the cubic to tetragonal distortion of the lattice that has been observed in Mn_3Ga and further shape memory alloys. Thus, in the present work it was intended to consequently model the alloys of the Mn_2YGa-family (Y representing transition metals) by means of stepwise relaxing the crystallographic and magnetic ground states from the cubic to the tetragonal lattices. Along with the structural relaxation the inherent modification of the magnetic properties, Heisenberg exchange interactions and anisotropies, have been studied. We aim to establish a model to adjust the properties as function of the number of valence electrons, that were shown to be a critical ingredient in the design scheme. Transitions from in-plane to out-of-plane anisotropy can be achieved on the basis of the presented work.

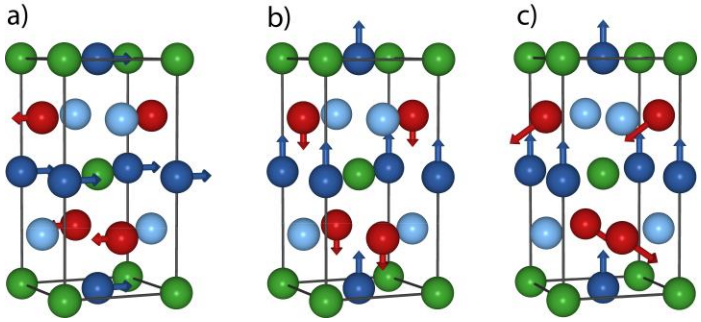

Fig. 1 A set of generalized tetragonal Heusler structures (spacegroup 119) is shown depicting the four Wyckoff-Positions, that are to be occupied according to Mn (red, blue), Y (light blue), Ga (green, Z) to achieve inverse-type order; a) in-plane ($K_u < 0$) and b) out-of-plane ($K_u > 0$) orientation of the magnetization. c) Exemplary non-collinear order for competing in-plane and out-of-plane anisotropy contributions.

Cubic Mn_2YGa compounds.

A deliberately chosen set of compounds of the type Mn_2YGa has been studied in order to obtain a fundamental understanding of the dependencies between structure and magnetic order, to deeply elucidate the microscopic properties in terms of Heisenberg exchange coupling constants, consequently the Curie temperature (T_C), and the respective magneto-crystalline anisotropy that arises according to the inset of the tetragonal distortion.

In the restriction to cubic crystal structures, due to which the so-called *regular*- and *inverse*- Heusler structures are the only ordering possibilites for ternary compositions, it was shown that for systems, involving early transition metals (ETM) on the *Y*-position ($Y = Sc, Ti, V, Cr$), the *regular*-type ordering is favored, where as the *inverste* order is preferred for the late transition metal (LTM, $Y = Mn, Fe, Co, Ni, Cu$) containing alloys. Inherently connected with the crystal structure is the magnetic ordering on the one hand, and the nature of the local moments on the other hand. Thus, the magnetic ordering of the ETM systems was found to be ferrimagnetic (FiM1), with mangenese site moments being aligned anti-parallel to the ETM, and exhibiting a rather localized character of the local moment. In contrast, the LTM compounds exhibit two different manganese sites, which obviously determine the magnetic order that is ferrimagentic (FiM2), but substantially different from FiM1 in terms of the relative orientation of the manganese moments, that occupy different sites (Mn(4*d*) and Mn(4*b*)) and show anti-parallel alignment of a smaller itinerant moment on Mn(4*d*) ($m_{4d} = -1.5\mu_B$) and larger moment on Mn(4*b*) ($m_{4b} = 3\mu_B$), that stays unchanged over the LTM-part of the series. A strong stabilization is observed that is evaluated by means of the Heisenberg exchange constants, which result in Curie temperatures larger than $T_C > 700\,K$ in the mean-field approximation. The T_C increases with increasing valence electron count (N_V) for the LTM compounds, where as it is found to be rather constant for the ETM

materials. Summarizing we found a dependence of the magnetic ordering in along with changes in the crystallographic order, accompagnied with Slater-Pauling behavior for the cubic compounds across the compensation point ($N_V = 24$) at which the structural preference changes from *regular* to *inverse*-type order. Along with the structral preference the T_C starts increasing with N_V, due to the strengthening of $J_{Mn4d-Mn4b}$ interaction that dominates the formation of the magnetic order.

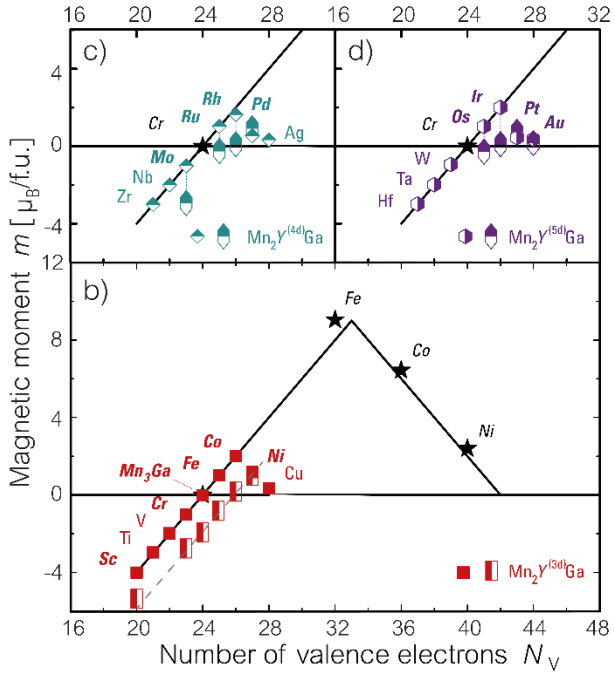

Fig. 2 Slater-Pauling plot of the magnetic moments of Mn_2YGa compounds, visualizing the constant shift to lower values for the tetragonally distorted systems.

Tetragonally distorted Mn_2YGa compounds.

Mn_3Ga and Mn_2NiGa are well known to exhibit a tetragonal distortion of the lattice, thus naturally it has been asked for the mechanisms behind the distortion, and to explore the physics of familiar compounds thus establishing trends that lead to a general understanding. The lattices of compounds where subject to non-volume conserving distortions, and it has been observed that with the inset of preferred *inverse*-order (LTM), most the lattices are sensitive towards a elongation of the unit cell. Compressed variants have not been observed. The emergence of dips in the energy-landscape, did already set in the range of ETM alloys, though. It could be shown, that the Mn(4*d*) and *Y*(4*c*) states at the Fermi edge are primary responsible for the instabilty of the cubic lattices.

The tetragonal distortion has a direct impact on the magnetic properties as compared to the cubic parent compounds, that is manifested in increased Mn(4*d*) local moments, where as the Mn(4*b*) moments are not affected; highlighting once again the peculiar role of Mn(4*b*). Elevated T_C are computed for the tetragonally elongated *inverse* compounds, that are not due to strengthened Mn(4*d*)-Mn(4*b*) exchange coupling, but due to the removal of competing interactions, that are weakened/degraded in response to the distortion. As a consequence of the arising uniaxial symmetry of the tetragonally modfied lattices, the emergence of magnetocrystalline anisotropy (MCA) is observed and quantitatively evaluated. Summarizing the numerical work, we observed a change of sign, thus reorientation of the preferred magnetization direction based on the optimized lattice data. The magnitude of the MCA energy strongly decreases within the tetragonal *inverse* systems and changes sign for compounds with Mn_2YGa ($Y = \mathrm{Ni, Pd, Pt}$) providing 10 valence *d*-electrons on the *Y*-position. The reorientation is thus qualitatively related to the experimentally already realized compound Mn_2RhSn exhibiting the same number of valence electrons, and thus being chemically similar to the above mentioned systems.

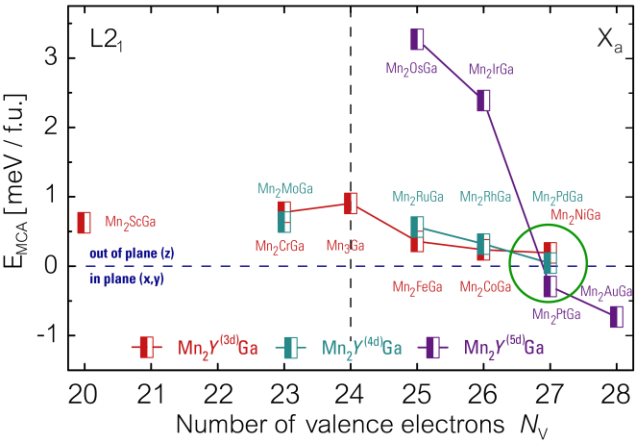

Fig. 3 Magneticrystalline anisotropy energy of the studied systems

References

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