New Mn- and Ni-based Heusler Compounds

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Heusler and related compounds are the main focus of the synthesis group. The goal is the design of Heusler compounds with desired functionalities. Examples are wide and low band gap semiconductors, Li-based compounds for electrode materials, exchange bias materials, thermoelectric materials, hard magnetic materials, compensated ferrimagnets, antiferromagnets , and magnetocaloric materials. As an example, the manganese-rich Heusler compounds are discussed.

Heusler compounds are an interesting class of compounds with wide-ranging and tunable properties. They are intermetallic compounds with the general composition X_2YZ, where X and Y are transition or rare-earth metals and Z is a main-group element. Heusler compounds include half-metallic ferri- and ferromagnetic materials, shape memory alloys, topological insulators, high potential magnetocalorics, and materials for spintronic applications.

New Mn2-based Heusler Compounds

Mn2-based Heusler compounds have attracted a great deal of interest as half-metallic ferri- and ferromagnets and as materials for spintronic applications. We have synthesized and studied in [1] the crystal structure and disorder type of the new Heusler compounds Mn_2RuGe and Mn_2RhGa and redetermined the crystal structure of Mn_2RuSn. They crystallize cubic with $L2_{1b}$ structure in *Fm*-3*m*, which is an inverse Heusler structure with a transition metal disorder of the type $(Mn_{0.5},Y_{0.5})_2MnZ$, where Y = Ru or Rh and Z = Ge, Ga, or Sn.

An analysis of the different types of substitutional disorder (Fig. 1) showed that an inverse Heusler structure can generally gain stability through the configurational part of the entropy of mixing at elevated temperatures without losing too much enthalpy in the configurational part of the enthalpy of mixing owing to a special bond arrangement in the inverse type structure.

Furthermore, the enthalpy of formation of transition-metal-based Heusler compounds and Al, Ga, and Ge as the main group metals obtained from DFT calculations and experimental data were used to confirm Burch's rule, which predicts the stability of transition-metal-based inverse Heusler compounds. It was found that alloying tendencies as manifested in binary phase diagrams and the enthalpies obtained from the Miedema model are correlated with the stability of Heusler compounds. Burch's rule was found to be in excellent agreement with the currently available experimental data. The DFT data and general alloying tendencies show that deviations are expected according to this rule. Miedema's model allows the estimation of the enthalpy of formation for the transition- and main group metal-based Heusler compounds, except for those having period 6 elements and Pd.

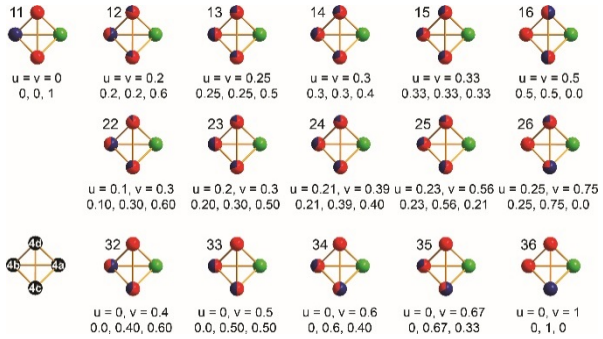

Fig. 1: Different types of substitutional disorder (X,Y) for the cubic Heusler structure of composition X_2YZ. The Z component fully occupies Wyckoff position 4a. For each type of disorder, the bcc tetrahedron is shown. The general formula is $(X_{1-u},Y_u)^{4d}(X_{1-v},Y_v)^{4c}(X_{u+v},Y_{1-u-v})^{4b}Z^{4a}$. Parameters in second row at the bottom of each bcc tetrahedron are occupancy factors of Y for 4d, 4c, and 4b sites in F-43m.

Magnetic and transport properties in the Heusler series $Ni_{2-x}Mn_{1+x}Sn$ affected by chemical disorder

The crystal structure and magnetic and transport characteristics of $Ni_{2-x}Mn_{1+x}Sn$ Heusler series have been studied [2] with the emphasis on chemical disorder effects. The structure and disorder character in this series can be predicted by using simple rules as outlined in [1]. Ni_2MnSn is a ferromagnetic, congruent melting phase, which crystallizes cubic in the $L2_1$ structure type. By increasing x, Ni and Mn atoms randomly mix occupy the heterocubic sites of the regular Heusler structure, and the magnetic structure becomes ferrimagnetic.

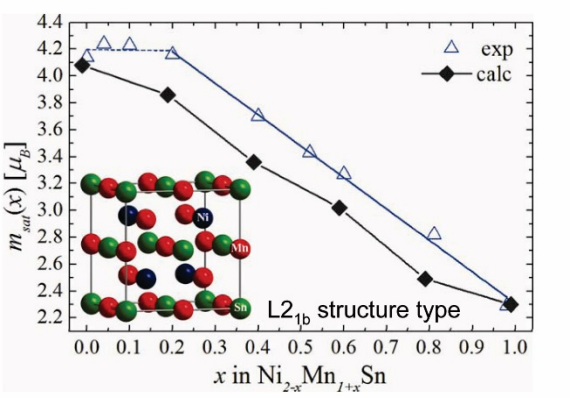

Fig. 2: The saturation magnetization $m_{sat.}$ at 1.8 K, from magnetometry and theoretically calculated as a function of the Mn content. The inset shows the inverse Heusler structure of Mn_2NiSn, which becomes the $L2_{1b}$ structure type in case of random mix occupation of the heterocubic sites by (Ni,Mn).

The total magnetic moment, $m_{sat.}$, decreases linearly in the range $0.2 \leq x \leq 1$, while the Curie temperature T_C increases. At low Mn content ($x < 0.2$), the unit cell volume shows anomalous behavior, characterized by a constant $m_{sat.}$ and T_C. It was found that the electrical resistivity, Seebeck coefficient, and thermal conductivity strongly depend on the amount of disorder, which increases with the Mn content.

Results of first-principle calculations based on the coherent potential approximation (CPA) alloy theory for the magnetic and electrical properties are in reasonable agreement with the simple rules and all experimental data. Data for the saturation magnetization m_{sat} at 1.8 K, from magnetometry and theoretically calculated for the example are shown in Fig. 2.

Increasing Curie temperature in the tetragonal Mn_2RhSn Heusler compound through substitution of Rh by Co and Mn by Rh

The tetragonal Mn_2RhSn Heusler compound shows better lattice mismatch with MgO than do $Mn_{3-x}Ga$ and tetragonal $Mn_{3-x}Co_xGa$ Heusler compounds and hence is better suited for spin transfer torque applications. Because of its tetragonal structure, Mn_2RhSn does not obey the Slater-Pauling rule, and its low saturation magnetic moment and its hard magnetic properties make it suitable for STT applications.

We found [3] when increasing the Rh content of $Mn_{2-x}Rh_{1+x}Sn$ that the Curie temperature increased; however, the tetragonal structure was not stable and changed to the cubic structure. When the amount of Mn increased in the $Mn_{2+x}Rh_{1-x}Sn$ series, mixtures of hexagonal Mn_3Sn and tetragonal Mn_2RhSn were observed. Substitution of Rh by Co in the series $Mn_2Rh_{1-x}Co_xSn$ series led to the tetragonal structure for $x = 0.1 – 0.6$ and the cubic structure in the Co-rich samples of $x = 0.7 – 1$. All cubic samples showed Slater-Pauling type behavior and soft hysteresis loops. In comparison to $Mn_{3-x}Ga$ and $Mn_{3-x}Co_xGa$, the $Mn_2Rh_{1-x}Co_xSn$ system showed better lattice match with MgO, similar to Mn_2RhSn. The Curie temperature in this series increased with an increase in the amount of Co. Thus, introducing Co in Mn_2RhSn increased the Curie temperature and preserved the tetragonal phase with a low magnetic moment up to $x = 0.6$.

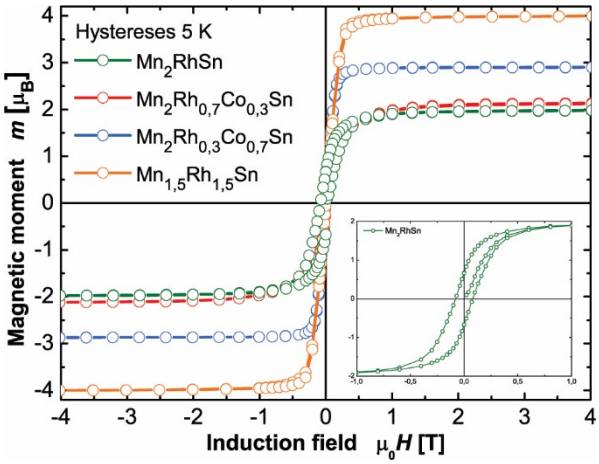

Fig. 3: Magnetic properties of $Mn_{2-x}Rh_{1+x}Sn$ and $Mn_2Rh_{1-x}Co_xSn$. The MH-curves were measured at T = 5 K. The inset shows the zoomed-in view of the hysteresis curves at T = 5 K for Mn_2RhSn.

References

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