Study of ferromagnetic materials under pressure from a theoretical approach

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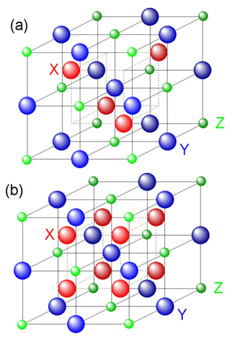
Abstract

The crystalline systems presented in this work belong to a group known as Heusler alloys. These materials are characterized by a strong coupling between their structure and magnetic properties. Usually, a change in one of them can alter the other, which implies changes in other electronic or structural properties. The properties and its dependence with external fields make these materials interesting, both from a fundamental point of view, as well as on their different possible applications. We present a theoretical investigation of the dependence of the structural, electronic and magnetic properties of some Heusler alloys as function of applied external pressure. Basically, we can analyze our results from two points of view. On one side, structural, electronic and magnetic properties at room pressure, such as, unit cell volume, bulk modulus, compressibility factor, magnetization, density of electronic states, spin polarization, magnetic exchange constants and Curie temperature. On the other hand, the pressure dependence of the Curie temperature and spin polarization in these particular ferromagnetic systems can be studied.

Introduction

Heusler alloys are magnetic multi-component intermetallic alloys with XYZ (half Heusler) or X_2YZ (full Heusler) stoichiometries.



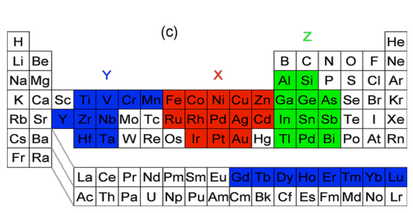


Figure 1. Atomic structure of a) Full Heusler alloys b) Half Heusler alloys and c) Major combinations of Heusler alloy formation [1].

Spatial group: 225
Simmetry: Fm3m

Wychoff Positions:
(0,0,0)
(1/2,1/2,1/2)
(1/4,1/4,1/4)
(3/4,3/4,3/4)

These alloys are based on the face-centered cubic structure with half Heusler alloys having the $C1_b$ structure and full Heusler alloys having the $L2_1$ structure.

Properties and Applications

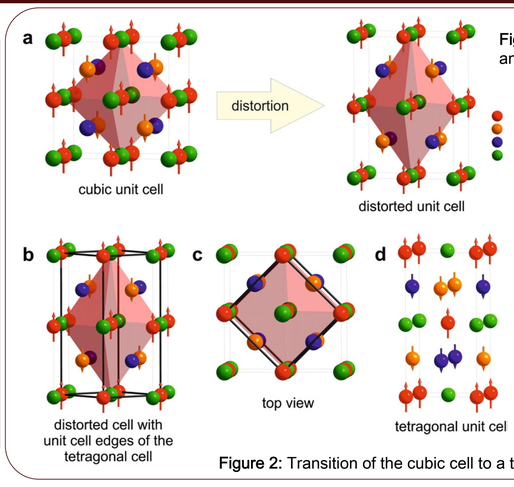


Figure 2: Transition of the cubic cell to a tetragonal distorted cell (Left).

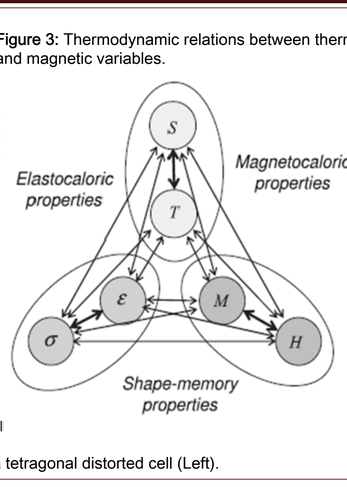
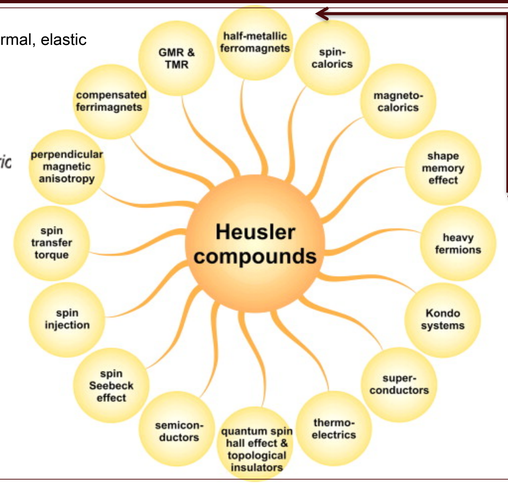


Figure 3: Thermodynamic relations between thermal, elastic and magnetic variables.



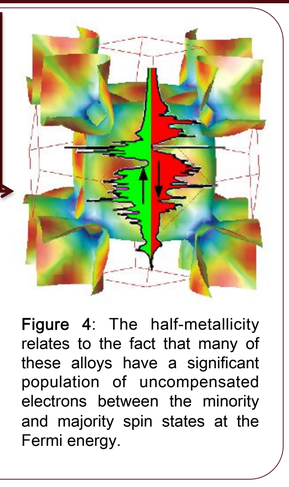
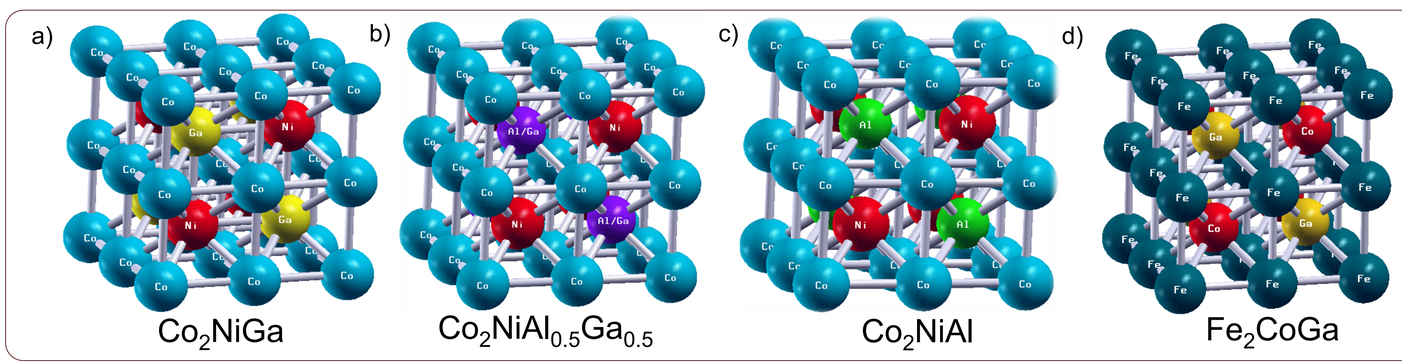


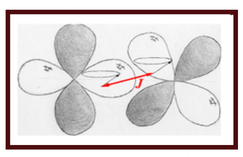
Figure 4: The half-metallicity relates to the fact that many of these alloys have a significant population of uncompensated electrons between the minority and majority spin states at the Fermi energy.

Systems

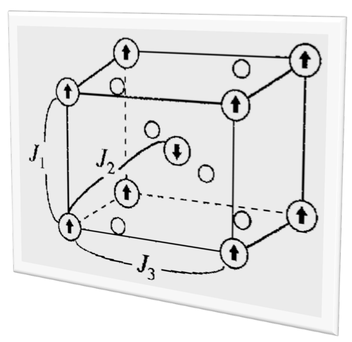


Theory

The magnetic exchange interaction constants J_{ij} have been determined by mapping the problem onto classical Heisenberg model:



$$\mathbf{H} = -\frac{1}{2}\sum_{i \neq j} J_{ij}\mathbf{m}_i \cdot \mathbf{m}_j$$



where i and j are atomic indices, J_{ij} represents the magnetic exchange coupling and **m** are the unitary local magnetic moments. J_{ij} are calculated using ab initio calculation based on the KKR method with the formulation proposed by Lichtenstein [6].

$$J_{ij} = \frac{1}{4\pi}\int^{\epsilon_F} ImTr_L\{\Delta_i \hat{T}^{ij}_{\uparrow}\hat{T}^{ij}_{\downarrow}\}\,d\epsilon$$

Comp. Details & Structural Results at ambient pressure

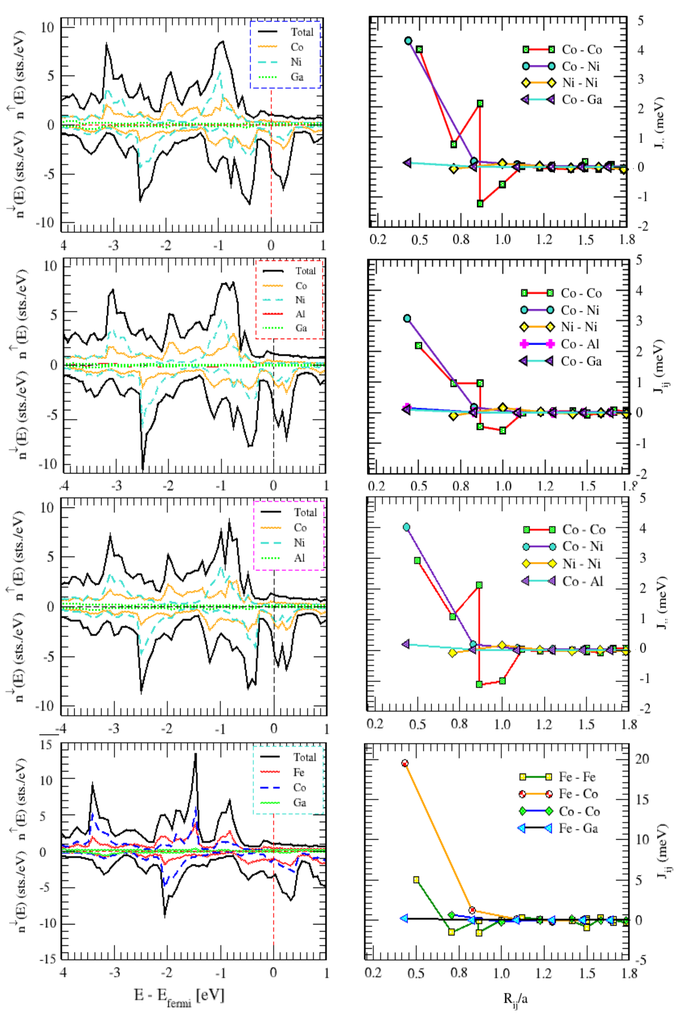


Figure 5: Electronic density of states and magnetic exchange interaction parameters J_{ij} between an atom i and its neighbor j located at a distance of R_{ij} for a) Co_2NiGa, b) $Co_2NiAl_{0.5}Ga_{0.5}$, c) Co_2NiAl and d) Fe_2CoGa at ambient pressure where the distances are normalized with respect to the lattice parameter, a. Notice the variation in the values of the vertical scale . Energy is reported with respect to the Fermi energy in DOS plots.

The structural properties were obtained from a density functional theory (DFT) calculation as implemented in the software VASP [2]. The electron–ionic core interaction has been accounted for by applying the PAW method [3] and the exchange correlation energy has been expressed using the GGA exchange–correlation potential of Perdew, Burke, and Ernzerhoff [4]. All calculations have been performed under the Local Spin Density Approximation to account for the spin density distribution. For the Brillouin-zone sampling, we use a 13x13x13 Monkhorst-Pack k-point mesh. The energy cut-off was chosen to be 470 eV for Co_2NiGa, Co_2NiAl and $Co_2NiAl_{0.5}Ga_{0.5}$ and 405 eV for Fe_2CoGa. With these fixed variables a very good energy convergence has been observed. The Density of states (DOS) and magnetic exchange coupling constants were obtained using the SPRKKR package [5] where Atomic sphere approximation (ASA) geometry is considered for the charge density and potential. The effect of pressure has been taken into account by changing the volume using the initial cell parameter optimized from the VASP code.

Structure	Lattice parameter [Å]	Bulk Modulus [GPa]	Compressibility Factor [GPa^{-1}]	Magnetization [μB]	Curie Temp. [K]
Fe_2CoGa	5.765	155.90	6.093×10^{-3}	6.065	1208.88
Co_2NiGa	5.693	180.15	5.280×10^{-3}	2.751	380.02
$Co_2NiAl_{0.5}Ga_{0.5}$	5.682	179.93	5.327×10^{-3}	2.724	411.20
Co_2NiAl	5.670	178.27	5.462×10^{-3}	2.731	390.73

Table 1: Calculated data on lattice parameters, bulk modulus, compressibility factor, magnetic moment and Curie temperature at ambient pressure for Co_2NiGa, Co_2NiAl, $Co_2NiAl_{0.5}Ga_{0.5}$ and Fe_2CoGa. The Curie temperature have been derived by mean field theory (MFT).

Results under pressure

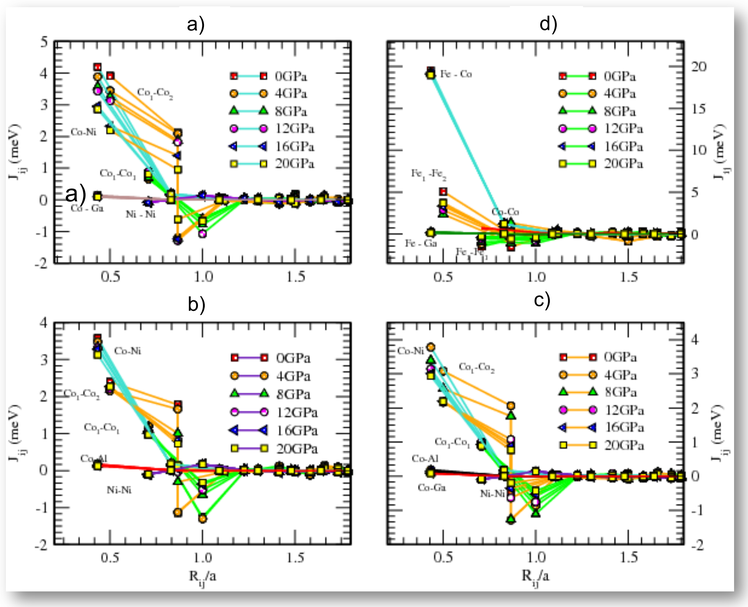


Figure 6: Magnetic exchange couplings J_{ij} as function of pressure for a) Co_2NiGa, b) $Co_2NiAl_{0.5}Ga_{0.5}$, c) Co_2NiAl and d) Fe_2CoGa.

Structure	Spin polarization as function of pressure					
	0 GPa	4 GPa	8 GPa	12 GPa	16 GPa	20 GPa
Fe_2CoGa	-0.5254	-0.4067	-0.5965	-0.7231	-0.7205	-0.7221
Co_2NiGa	-0.5297	-0.5446	-0.5471	-0.4931	-0.6026	-0.6797
$Co_2NiAl_{0.5}Ga_{0.5}$	-0.5796	-0.6295	-0.7322	-0.7558	-0.7572	-0.7724
Co_2NiAl	-0.5555	-0.5425	-0.5414	-0.6215	-0.7376	-0.7376

Figure 7: Magnetization as function of pressure for a) Based Co alloys and b) for Fe_2CoGa, c) Curie temperature for based Co alloys and d) for Fe_2CoGa. e) Spin polarization as function of pressure for Fe_2CoGa.

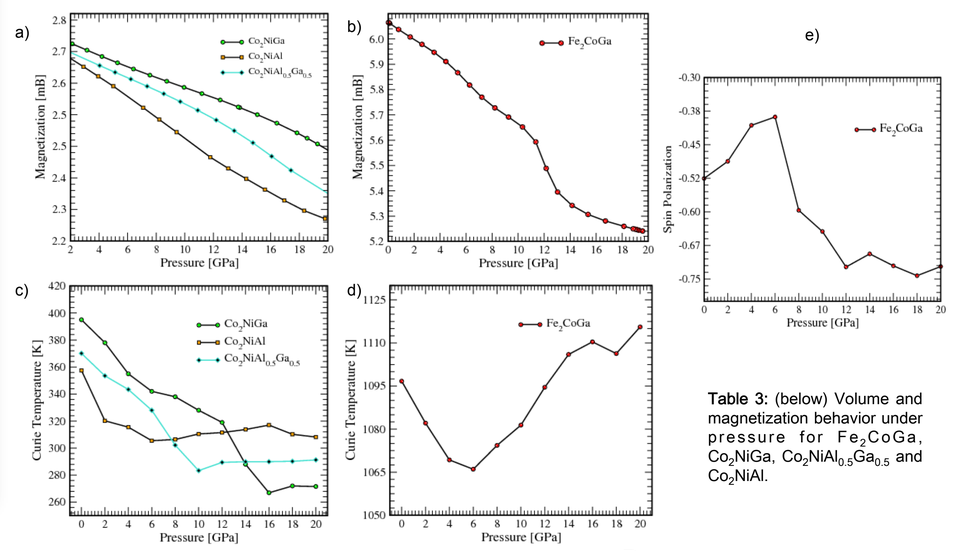


Table 2. Spin polarization for Fe_2CoGa, Co_2NiGa, $Co_2NiAl_{0.5}Ga_{0.5}$ and Co_2NiAl as function of pressure (Left).

Table 3: (below) Volume and magnetization behavior under pressure for Fe_2CoGa, Co_2NiGa, $Co_2NiAl_{0.5}Ga_{0.5}$ and Co_2NiAl.

	Pressure	
	0 GPa →	20 GPa
System	Volume	Magnetization
Fe_2CoGa	3.28%	5.04%
Co_2NiGa	3.01%	10.96%
$Co_2NiAl_{0.5}Ga_{0.5}$	2.99%	13.70%
Co_2NiAl	2.98%	15.46%

Conclusions

1. A systematic study of the structural, electronic and magnetic properties for various Heusler alloys with the $L2_1$ crystal structure as function of pressure are presented.
2. The electronic DOS at the Fermi level is dominated by d-states, which in turn are contributed mostly by the cobalt atoms for Co based alloys and Fe for Fe_2CoGa.
3. The compressibility dependence of the volume is quadratic with the pressure and the coefficients were determined.
4. Based on the obtained equation of states, we are able to study the changes of the magnetic properties as function of pressure.
5. As pressure is increased, interatomic distances shorten. This is accompanied by depletion of the d-band, increased overlapping of d-states and an increased metallicity and delocalization, this result in a decrease in magnetization with pressure .
6. We find agreement of all structural and elastic properties with reported experimental and/or theoretical values.
7. The large dependence observed for the properties on the Heusler alloys with pressure, indicates that pressure can be used in experiments to trigger particular magnetic properties of interest.

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Acknowledgements

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