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First-principles study of exchange interactions and Curie temperatures of half-metallic ferrimagnetic full Heusler alloys Mn_2VZ (Z = Al, Ge)

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Abstract

We report the parameter-free, density functional theory calculations of interatomic exchange interactions and Curie temperatures of half-metallic ferrimagnetic full Heusler alloys Mn_2VZ (Z = Al, Ge). To calculate the interatomic exchange interactions we employ the frozen-magnon approach. The Curie temperatures are calculated within the mean-field approximation to the classical Heisenberg Hamiltonian by solving a matrix equation for a multi-sublattice system. Our calculations show that, although a large magnetic moment is carried by Mn atoms, competing ferromagnetic (inter-sublattice) and antiferromagnetic (intra-sublattice) Mn–Mn interactions in Mn_2VAl almost cancel each other in the mean-field experienced by the Mn atoms. In Mn_2VGe the leading Mn–Mn exchange interaction is antiferromagnetic. In both compounds the ferromagnetism of the Mn subsystem is favoured by strong antiferromagnetic Mn–V interactions. The obtained value of the Curie temperature of Mn_2VAl is in good agreement with experiment. For Mn_2VGe there is no experimental information available and our calculation is a prediction.

1. Introduction

The increased interest in the field of spin electronics attracts strong attention to the magnetic systems suitable for the realization of spin injection into a semiconductor [1]. The Heusler alloys are considered as a promising class of materials that can possess the necessary combination of properties. Among the features useful for applications are high Curie temperature, high electron spin polarization at the Fermi level and very small lattice mismatch with widely employed semiconductors [2]. Some of the Heusler compounds were found to

have a half-metallic ground state [3] characterized by a 100% spin-polarization of the charge carriers. An interesting combination of physical properties makes Heusler alloys the subject of intensive experimental and theoretical investigations [4–9].

In full Heusler compounds characterized by the formula X_2YZ the Mn atoms usually enter as the Y element. The compounds where Mn assumes the X positions are very rare. To our knowledge there have only been two systems of this type studied experimentally: Mn_2VAl [10] and Mn_2VGa [11].

Mn_2VAl has received much experimental and theoretical attention. The neutron diffraction experiment by Itoh *et al* [10] gave the ferrimagnetic state of compound with Mn magnetic moment of $1.5\pm0.3\ \mu_B$ and V moment $-0.9\ \mu_B$. Jiang *et al* examined the magnetic structure of Mn_2VAl by x-ray diffraction and magnetization measurements [12]. They found that Mn_2VAl was nearly half-metallic with a total magnetic moment of $1.94\ \mu_B$ at 5 K. The Curie temperature of the sample was found to be about 760 K and the loss of half-metallic character was attributed to the small amount of disorder. The electron structure calculation by Ishida *et al* performed within the local-density approximation (LDA) to the density functional theory resulted in a ground state of Mn_2VAl close to half-metallicity [13]. Recently a detailed theoretical study of the magnetism of Mn_2VAl was reported by Weht and Pickett [14], who used the generalized gradient approximation (GGA) for the exchange correlation potential and showed that Mn_2VAl is a half-metallic ferrimagnet, with the atomic moments of $1.5\ \mu_B$ and $-0.9\ \mu_B$ on Mn and V in very good agreement with experiment. The Fermi level was found to lie in the minority spin band.

The main purpose of the present work is a detailed study of the exchange interactions in two half-metallic Mn_2VZ compounds: Mn_2VAl and Mn_2VGe. (Mn_2VGe has not yet been synthesized. Its half-metallicity has been predicted theoretically [4].) Both intra-sublattice and inter-sublattice exchange interactions are calculated. We show that the pattern of exchange interactions in these systems deviates from the physical picture that can be expected on the basis of the experimental information available. Indeed, the Mn–Mn distance of 2.96 Å (3.04 Å) in Mn_2VAl (Mn_2VGe) is substantially smaller than the Mn–Mn distance of about 4 Å in the X_2MnZ-type Heusler alloys [15]. (For Mn_2VGe we use the interatomic distance of Mn_2VGa [11].) On the other hand, it is comparable with the Mn–Mn distance in the antiferromagnetic fcc Mn (2.73 Å) [16]. According to the Bethe–Slater curve [17], there are physical reasons to expect that smaller distances between the 3d atoms stimulate the formation of the antiferromagnetic structure whereas larger distances make the ferromagnetic structure energetically preferable. Among the Heusler alloys, a smaller distance between pairs of the Mn atoms is obtained in the case of random occupation by Mn and Z atoms of the Y and Z sublattices (see, e.g., the system with the B2-type crystal structure in [15]). Indeed, the experiment gives for such systems the antiferromagnetic ordering [15]. Therefore in the case of Mn_2VAl an antiferromagnetism of the two Mn sublattices can be expected.

Our study shows, however, that the situation is more complex. The nearest-neighbour (nn) Mn–Mn exchange interaction is found to be ferromagnetic whereas the next nn Mn–Mn interaction is antiferromagnetic. As a result the contributions of different Mn atoms into the exchange field experienced by a given Mn atom compensate strongly, not giving substantial contribution into the Curie temperature. The main role in the formation of the magnetic structure and the magnetic transition temperature is played by the strong Mn–V exchange interaction. We show that in Mn_2VGe the ferromagnetic ordering of the Mn subsystem is also governed by the Mn–V exchange interaction.

The paper is organized as follows. In section 2 we present the calculational approach. Section 3 contains the results of the calculations and discussion. Section 4 gives the conclusions.

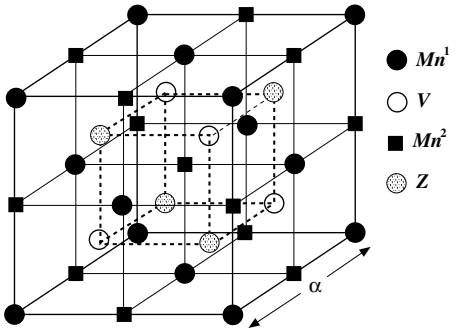

Figure 1. Schematic representation of the $L2_1$ structure. The lattice consists of four interpenetrating fcc sublattices with the positions (0, 0, 0) and $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ for the Mn and $(\frac{1}{4}, \frac{1}{4}, \frac{1}{4})$ and $(\frac{3}{4}, \frac{3}{4}, \frac{3}{4})$ for the V and Z, respectively.

2. Calculational method

The calculations are carried out with the augmented spherical waves (ASW) method [18] within the generalized gradient approximation (GGA) [19] for the exchange–correlation potential. We use the experimental lattice parameter of Mn_2VAl [15][1]. The radii of all atomic spheres are chosen equal.

We describe the interatomic exchange interactions in terms of the classical Heisenberg Hamiltonian:

$$H_{\text{eff}} = -\sum_{\mu,\nu} \sum_{\substack{\mathbf{R},\mathbf{R}' \\ (\mu\mathbf{R}\neq\nu\mathbf{R}')}} J^{\mu\nu}_{\mathbf{R}\mathbf{R}'} \mathbf{s}^{\mu}_{\mathbf{R}} \mathbf{s}^{\nu}_{\mathbf{R}'}. \tag{1}$$

In equation (1), the indices μ and ν label different sublattices, $\mathbf{R}$ and $\mathbf{R}'$ are the lattice vectors specifying the atoms within sublattices, and $\mathbf{s}^{\mu}_{\mathbf{R}}$ is the unit vector pointing in the direction of the magnetic moment at site $(\mu, \mathbf{R})$. The systems considered contain three 3d atoms in the unit cell (see figure 1).

We employ the frozen-magnon approach [20, 21] to calculate the interatomic Heisenberg exchange parameters. The calculations involve few steps. In the first step, the exchange parameters between the atoms of a given sublattice μ are computed. The calculation is based on the evaluation of the energy of the frozen-magnon configurations defined by the following atomic polar and azimuthal angles:

$$\theta^{\mu}_{\mathbf{R}} = \theta, \qquad \phi^{\mu}_{\mathbf{R}} = \mathbf{q}\cdot\mathbf{R} + \phi^{\mu}. \tag{2}$$

The constant phase ϕ^{μ} is always chosen equal to zero. The magnetic moments of all other sublattices are kept parallel to the z axis. Within the Heisenberg model (1) the energy of such configuration takes the form

$$E^{\mu\mu}(\theta, \mathbf{q}) = E^{\mu\mu}_0(\theta) + \sin^2\theta J^{\mu\mu}(\mathbf{q}) \tag{3}$$

where $E^{\mu\mu}_0$ does not depend on $\mathbf{q}$ and the Fourier transform $J^{\mu\nu}(\mathbf{q})$ is defined by

$$J^{\mu\nu}(\mathbf{q}) = \sum_{\mathbf{R}} J^{\mu\nu}_{0\mathbf{R}} \exp(\mathrm{i}\mathbf{q}\cdot\mathbf{R}). \tag{4}$$

[1] To our knowledge, Mn_2VGe has not been studied experimentally. We perform calculations of Mn_2VGe using the lattice constants of Mn_2VAl and Mn_2VGa. In the lattice constant of Mn_2VGa we found it half-metallic, while it was nearly half-metallic with the lattice constant of Mn_2VAl. We will present the results of calculation for the former case.

Table 1. Magnetic moments (in μ_B) of Mn_2VZ (Z = Al, Ge). a is the lattice parameter (in Å).

		a (Å)	Mn	V	Z	Cell
Mn_2VAl	Present work	5.932[a]	1.525	−1.022	−0.031	2.00
	Weht[b]	5.875	1.500	−0.900	−0.100	2.00
	Galanakis[c]	5.932[a]	1.413	−0.786	−0.020	2.02
	Jiang (expt)[d]	5.920	—	—	—	**1.94**
Mn_2VGe	Present work	6.095[e]	1.003	−0.969	−0.037	1.00
	Galanakis[c]	5.932[a]	0.750	−0.476	−0.020	1.00

[a] Reference [15].
[b] Reference [14].
[c] Reference [4].
[d] Reference [12].
[e] See footnote 1.

In the case of $\nu = \mu$ the sum in equation (4) does not include $\mathbf{R} = 0$. Calculating $E^{\mu\mu}(\theta, \mathbf{q})$ for a regular $\mathbf{q}$-mesh in the Brillouin zone of the crystal, and performing back Fourier transformation, one gets the exchange parameters $J_{0\mathbf{R}}^{\mu\mu}$ for sublattice μ.

The determination of the exchange interactions between the atoms of two different sublattices μ and ν is discussed in [22].

The Curie temperature is estimated within the mean-field approximation for a multi-sublattice material by solving the system of coupled equations [22, 23]

$$\langle s^\mu \rangle = \frac{2}{3k_B T} \sum_\nu J_0^{\mu\nu} \langle s^\nu \rangle \tag{5}$$

where $\langle s^\nu \rangle$ is the average z component of $\mathbf{s}_\mathbf{R}^\nu$ and $J_0^{\mu\nu} \equiv \sum_\mathbf{R} J_{0\mathbf{R}}^{\mu\nu}$. Equation (5) can be represented in form of an eigenvalue matrix problem:

$$(\boldsymbol{\Theta} - T\mathbf{I})\mathbf{S} = 0 \tag{6}$$

where $\Theta_{\mu\nu} = \frac{2}{3k_B} J_0^{\mu\nu}$, $\mathbf{I}$ is a unit matrix, and $\mathbf{S}$ is the vector of $\langle s^\nu \rangle$. The largest eigenvalue of matrix Θ gives the value of the Curie temperature [23].

3. Results and discussion

The crystal structure is presented in figure 1. The two Mn sublattices are equivalent. The nearest Mn atoms belong to two different sublattices.

In table 1 we present calculated magnetic moments. For comparison, the available experimental values of the moments and the results of previous calculations are given. The net magnetic moment per unit cell is 2 μ_B for Mn_2VAl and 1 μ_B for Mn_2VGe. The magnetic alignment is ferrimagnetic in both systems. The Mn moments are parallel and assume the values close to 1.5 μ_B in Mn_2VAl and to 1 μ_B in Mn_2VGe. The moment of V is close to -1 μ_B in both systems. The values of the moments are in agreement with the results of previous calculations.

The calculated Heisenberg exchange parameters are presented in figures 2 and 3. We obtained a strong dependence of the pattern of the Mn–Mn and V–V exchange interactions on the type of the Z atom. A similar result was obtained earlier for Ni-based full Heusler alloys [22]. The nearest Mn–Mn distance is half of the lattice constant a. The exchange interaction between the nearest Mn atoms is ferromagnetic (figure 3). Simultaneous examination of both intra- and inter-sublattice Mn–Mn exchange interactions (figures 2, 3) indicates the RKKY-type oscillations.

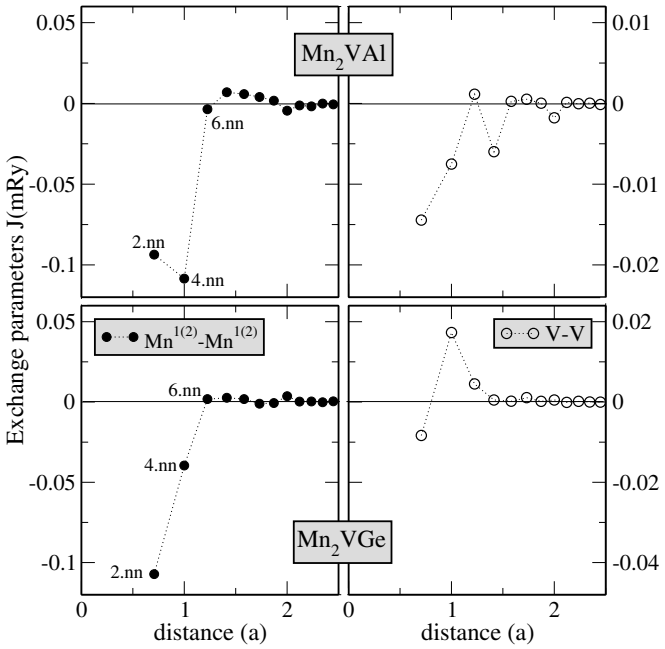

Figure 2. Intra-sublattice Mn–Mn (left-hand part) and V–V (right-hand part) exchange interactions in Mn_2VZ (Z = Al and Ge) as a function of the distance given in units of the lattice constant.

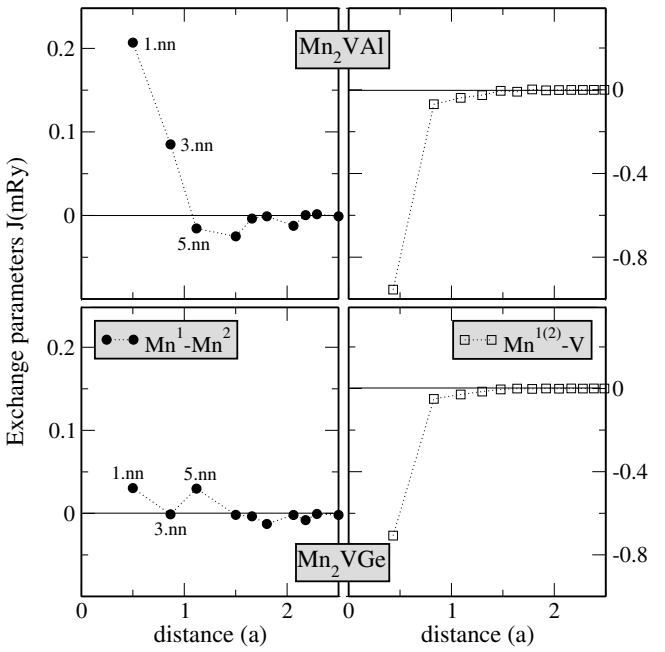

Figure 3. Inter-sublattice Mn^1–Mn^2 (left-hand side) and Mn–V (right-hand side) exchange interactions in Mn_2VZ (Z = Al and Ge) as a function of the distance given in units of the lattice constant.

Table 2. Mean-field estimation of the Curie temperatures for Mn_2VZ (Z = Al, Ge). The second (third) column gives the Curie temperature calculated with account for Mn–Mn (Mn–V) interactions only. In the fourth column both types of interactions are taken into account. The experimental value of the Curie temperature for Mn_2VAl is taken from [15].

	$T_{c,N}^{Mn-Mn[MFA]}$ (K)	$T_c^{Mn-V[MFA]}$ (K)	$T_c^{[MFA]}$ (K)	$T_c^{[Exp]}$ (K)
Mn_2VAl	30	623	638	760
Mn_2VGe	170	488	413	—

If only Mn–Mn exchange interactions are considered, in Mn_2VAl ferromagnetic interactions prevail, while in Mn_2VGe the Mn–Mn dominate antiferromagnetic interactions. The corresponding Curie and Neél temperatures are given in the second column of table 2.

The interactions between V atoms are very small and can be neglected. The formation of the ferrimagnetic structure with all Mn moments being parallel to each other and the V moments directed oppositely is determined by the strong antiferromagnetic exchange interactions between the nearest Mn and V moments. In Mn_2VAl this interaction is five times larger than the nearest-neighbour Mn–Mn interaction. In Mn_2VGe this factor increases to 20. The strong Mn–V antiferromagnetic coupling makes a parallel direction of the Mn moments surrounding a V atom energetically preferable, leading to the ferromagnetism of the Mn sublattices. In Mn_2VGe this trend overcomes the direct antiferromagnetic Mn–Mn interaction.

In the third column of table 2 we present the Curie temperature calculated with the Mn–Mn exchange interaction being neglected. The Curie temperature given in the fourth column takes into account both Mn–V and Mn–Mn interactions. It is clearly seen that the main contribution for both systems comes from the Mn–V interaction. In Mn_2VAl, the correction of T_c due to the Mn–Mn exchange interaction is positive and very small. In Mn_2VGe, it is negative and amounts to 15%.

In Mn_2VAl, where the experimental estimation of the Curie temperature is available, both the theoretical and experimental values are in good agreement. The theoretical Curie temperature of Mn_2VGe should be considered as a prediction.

4. Conclusion

In conclusion, we have systematically studied exchange interactions and Curie temperatures in two half-metallic ferrimagnetic full Heusler alloys Mn_2VZ (Z = Al, Ge). The calculations are performed within the parameter-free density functional theory. We show that various Mn–Mn exchange interactions compensate each other and the ferromagnetism of the Mn subsystem is favoured by very strong antiferromagnetic Mn–V interactions. Good agreement with experiment is obtained for the Curie temperature of Mn_2VAl. We give a prediction for the Curie temperature of Mn_2VGe.

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