Origin of the Dzyaloshinskii-Moriya Interactions in the Intermetallic RMn_2Si_2 (R= La, Ce, Yb and Y) Materials: A DFT Study

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Origin of the Dzyaloshinskii-Moriya interactions in the intermetallic RMn_2Si_2 (R= La, Ce, Yb and Y) materials: A DFT study

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Abstract

The Dzyaloshinskii-Moriya interactions (D_M) are investigated using first-principles calculations by means of the WIENNCM code, an implementation of the FP-LAPW method. The intermetallic RMn_2Si_2 (R= La, Ce, Yb, and Y) materials exhibit a large spin-orbit effect after the density of states; they found a strong hybridization between Mn-Si and Mn-R atoms. Also, show a large noncollinear magnetic configuration depending on the R atoms. By using ab-initio calculations, the RKKY effect is observed in the RMn_2Si_2 materials, which shows explicitly the existence of the giant magnetoresistance (GMR) in these materials. Explicitly, the mechanisms responsible for the magnetoelectric coupling are due to relatively the effect of the presence of the Dzyaloshinskii-Moriya term.

Keywords: RMn_2Si_2 materials; DFT study; Dzyaloshinskii-Moriya interactions; Magnetoelectric coupling; RKKY interactions.

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1-Introduction

Recently, the ternary intermetallic RT_2X_2 compounds (R is rare earth, T is transition metal and X is Si or Ge) have attracted significant attention because of their remarkable properties and potential application as high-temperature thermoelectric [1]. On the other hand, these materials have been reexamined by neutron diffraction experiments as they show a positive giant magnetoresistance [2]. Several studies indicate that in the majority of these compounds, the Mn metal (except Mn) carries no magnetic moment. R atoms usually order antiferromagnetically at low temperatures. Magnetic studies found that RMn_2Si_2 compounds are either ferromagnetic or antiferromagnetic [3,4]. Nevertheless, the various studies devoted to RMn_2Si_2 (R = La, Ce, Yb, Y, Pr, Nd, Sm, Gd) [2, 5-8] found some discrepancies. Moreover, these materials show a rich variety of magnetic behaviors depending on R (Ba, Ca, La, Rare Earths) [9]. This magnetic diversity should give rise to different transport properties and magnetoresistance (GMR) coefficients. In addition, the noncollinear magnetism in $LaMn_2Si_2$ compound has been studied in detail in Ref. [2]. Furthermore, these materials crystallize in the $ThCr_2Si_2$ structure, whereas, the structural and magnetic properties of $BaMn_2P_2$ and $BaMn_2As_2$ [10,11] are similar to those of the archetypal magnetoelectric material chromium sesquioxide Cr_2O_3 [12, 13]. Magnetoelectric has a wide interest in several applications as multifunctional devices such as magnetic-electric transducers, actuators, sensors, and spintronic devices [14,15]. There are two principal types of magnetoelectric materials: magnetostrictive-piezoelectric composites and single-phase crystals and polycrystals [16]. In the next year, another two interesting multiferroic materials were discovered: orthorhombic $TbMn_2O_5$ [17] and hexagonal $HoMnO_3$ [18] is similar to the $TbMnO_3$ compound, this demonstrated a strong magnetoelectric coupling, giving a switchable polarization upon magnetic field, although its ferroelectricity is poor. Moreover, the origin of magnetoelectric materials based on the spin-orbit coupling can be explained by two possible

mechanisms, Dzyaloshinskii-Moriya (DM) interaction [19, 20] and spin-dependent metal-ligand hybridization [19].

The present paper is to study the physical properties of RMn_2Si_2 ($\text{R} = \text{La}, \text{Ce}, \text{Yb}, \text{and Y}$) materials by using a non-collinear first-principles calculation. Firstly, we found an important RKKY coupling in the intermetallic RMn_2Si_2 materials; this effect is related to the noncollinear magnetism, which also is the sign of GMR in this compound. Secondly, we calculated the Dzyaloshinskii-Moriya interaction (DM) in these types of materials; we found this effect is related to the spin-orbit coupling, also the relationship between magnetoelectric coupling and DM interaction was presented. For this reason, the intermetallic RMn_2Si_2 materials could be suggested as a good candidate for magnetoelectric applications.

2-Computational Methods

The intermetallic RMn_2Si_2 ($\text{R} = \text{La}, \text{Ce}, \text{Yb}, \text{and Y}$) compounds is studied using the full potential linearized augmented plane wave FP-LAPW method implemented in the WIENNCM package [21] with the noncollinear magnetism. While the exchange and correlation potentials have been treated by the Generalized Gradient Approximation parameterized by Perdew, Burke, and Erzehof (GGA-PBE) [22]. All the calculations take into account spin-orbit coupling with a 300 k-point in the irreducible wedge of the Brillouin zone. The RMn_2Si_2 compounds crystallize with the tetragonal ThCr_2Si_2 -type crystal structure in the following space group ($I4/mmm$) with the lattice parameter (see **Table 1**). The atoms R, Mn and Si are located in $[0,0,0]$, $[0,0.5,0.25]$ and $[0,0,z]$, respectively.

Table 1: Structural parameters of RMn_2Si_2 ($\text{R} = \text{La}, \text{Ce}, \text{Yb}$ and Y) materials.

	La [23]	Y [24]	Ce [25]	Yb [26]
$a(\text{\AA})$	4.1103	3.9200	4.0417	3.8864
$c(\text{\AA})$	10.5969	10.4750	10.5546	10.4040
$z(\text{Si})$	0.3805	0.3870	0.3820	0.3838

Fig. 1: Unit cell structure of RMn_2Si_2 ($\text{R} = \text{La, Ce, Yb and Y}$) materials.

The Hamiltonian of the system (see, **Fig. 1**) is described by Heisenberg model:

$$H = -J_{\text{Mn-Mn}} \sum_{\langle i,j \rangle} \vec{S}_i \vec{S}_j - J_{\text{RKKY}} \sum_{\langle i,k \rangle} \vec{S}_i \vec{S}_k - H_{\text{DM}} \quad (1)$$

where, $J_{\text{Mn-Mn}}$ is the exchange coupling interactions between the first nearest neighbor. J_{RKKY} is the RKKY interaction between the first nearest neighbor's intra-plans. H_{DM} is the Dzyaloshinskii-Moriya interaction given by:

$$\vec{D}_M * (\vec{S}_i \times \vec{S}_j) \quad (2)$$

where $\vec{S} = (S_x, S_y, S_z)$ and $\vec{D}_M = (D_x, D_y, D_z)$

3-Results and discussion

3.1-Collinear calculation

3.1.1-Density of states and hybridization

The total and partial densities of states of intermetallic RMn_2Si_2 ($\text{R} = \text{La, Y, Ce and Yb}$) compounds are plotted in **Figs. 2**. From this figure, it is found that these compounds exhibit metallic behavior. The maximum contribution in the total DOS is due to the Mn atom in both up and down spins states, whereas, the R ($\text{R} = \text{La, Yb, Y and Ce}$) and Si atoms are lowly

contributing in the DOS states. The densities of states in RMn_2Si_2 changed with changing the R atoms. For $\text{R}=\text{La}$, Y, Ce, and Yb the maximum of dos reached the value 24.56 (states/eV), 6.17 (states/eV), 25.02 (states/eV), and 79.40 (states/eV), respectively. However, the difference in hybridization between Mn-Si and Mn-R states is observed. For all compounds, there are number peaks in the valence band partial DOS of the Mn d states. The Mn (d) and Si (p) states are hybridized mostly in the energy range from -5 eV to -2.0 eV relative to the Fermi level (see, **Figs. 2 (a-d)**).

Fig. 2: Total and partial density of states for RMn_2Si_2 materials: a) LaMn_2Si_2 b) YMn_2Si_2 , c) CeMn_2Si_2 , and d) YbMn_2Si_2 .

3.2-Non-collinear calculation

The magnetic moment value of Mn in each compound for the different angles is illustrated in **Table 2**. From this table, it is found that the magnetic moment of Mn atom in all materials is in good agreement with the experimental results [23-26].

Table 2: Magnetic moment RMn_2Si_2 (R= La, Ce, Yb and Y) materials.

Compounds	Present work ($\mu_B(\text{Mn})$)					Experimental values ($\mu_B(\text{Mn})$)
	30°	60°	90°	120°	150°	
LaMn_2Si_2	2.22	2.23	2.16	2.17	2.21	2.49 [23]
YMn_2Si_2	2.12	2.07	2.05	2.10	2.11	1.98 [24]
CeMn_2Si_2	2.16	2.13	2.08	2.12	2.17	2.30 [25]
YbMn_2Si_2	2.05	2.04	2.06	2.05	2.05	2.00 [26]

3.2.1-The Ruderman–Kittel–Kasuya–Yosida (RKKY) interactions

The RMn_2Si_2 compounds (R=La, Y, Yb, and Ce) form a natural multilayer system where the planes of the rare earth ions are separated from the transition metal layers. The Mn-Mn atoms are formed by two magnetic plans separated by a nonmagnetic plane of Si. By varying the distance between Mn-Mn in different planes, we can calculate the exchange coupling interactions (J_{RKKY}). **Fig. 3** shows the exchange coupling interaction as a function of the distance between Mn-Mn atoms in different planes. This figure found that the long-range damping oscillatory behavior can be clearly seen; this oscillatory depended on the spin-orbit interaction, the atomic number of the R atom, and structural parameters. We are found that all family of RMn_2Si_2 compounds exhibits RKKY interactions (see, **Figs. 3(a-d)**). These results are in good agreement with the experimental studies and show the contribution of the non-collinearity is due to the GMR effect of these materials [2].

Fig. 3: Exchange coupling interactions J_{RKKY} as a function of the distance between Mn-Mn atoms: (a) LaMn_2Si_2 (b) YMn_2Si_2 (c) CeMn_2Si_2 and (d) YbMn_2Si_2 .

3.2.2-Dzyaloshinskii-Moriya interaction

The antisymmetric Hamiltonian is expressed by $H_{DM} = D_M * (\vec{S}_i \times \vec{S}_j)$, in Heisenberg model $\vec{S} = (S_x, S_y, S_z)$. [28]:

$$H_{DM} = D_x(S_y S_z - S_z S_y) - D_y(S_x S_z - S_z S_x) + D_z(S_x S_y - S_y S_x) \quad (3)$$

where D_x , D_y and D_z are calculated coefficients of D_M by choosing the configuration that take into consideration one direction and cancel the two other components.

The D_M has been calculated for the RMn_2Si_2 (R=La, Y, Ce, and Yb) compounds using ab initio calculations. The results of D_M are presented in **Table 3**. From these results the RMn_2Si_2 (R=La, Y, Ce and Yb) compound exhibit a strong D_M except YMn_2Si_2 give a small D_M ($D_x=0.36\text{meV}$, $D_y=0.0\text{meV}$ and $D_z=0.65\text{meV}$). This difference was related to the spin-orbit effect in each compound, it's normal to find a D_M in this type of compounds due to the noncollinear inter-layers coupling existing between Mn-Mn atoms.

Table 3: Dzyaloshinskii-Moriya interactions values in RMn_2Si_2 materials.

Materials	(θ_1, θ_2) and (φ_1, φ_2)	D_M (meV)
LaMn ₂ Si ₂	(60,120)-(90,270)	$D_x=0.09$
	(60,120)-(0,180)	$D_y=1.75$
	(0,0)-(60,120)	$D_z=5.73$
YMn ₂ Si ₂	(60,120)-(90,270)	$D_x=0.36$
	(60,120)- (0,180)	$D_y=0.0$
	(0,0)- (60,120)	$D_z=0.65$
CeMn ₂ Si ₂	(45,135)- (90,270)	$D_x=10.90$
	(60,120)- (0,180)	$D_y=1.25$
	(0,0)- (90,180)	$D_z=5.16$
YbMn ₂ Si ₂	(60,120)- (90,270)	$D_x=108.29$
	(60,120)- (0,180)	$D_y=1.95$
	(0,0)- (30,150)	$D_z=1.55$

Moreover, the LaMn_2Si_2 compound presents a large D_M along in the z-axis directions and a little effect in the y-axis directions. The YbMn_2Si_2 illustrates a maximum (108.29meV) of D_M along the x-axis directions and a weak value in the y-axis and z-axis directions. The maximum of D_M for the CeMn_2Si_2 compound is along the x-axis direction, also an important value in the z-axis and a weak value in the y-axis direction. This difference is due to the hybridization between orbital d for Mn and p for Si. However, it's clear that when we change the tilt angle the value of the D_M changes too, we can conclude the D_M depended strongly on the angle. We found a strong D_M in the intermetallic RMn_2Si_2 (R=La, Ce, and Yb) compounds, this interaction allows coupling between magnetism and ferroelectricity [15, 16]. This interaction resembles the form of antisymmetric superexchange interaction due to relativistic spin-orbit coupling, being proportional to the vector product of spins, whereas, the D_M interaction favors non-collinear spin ordering. These types of materials are an excellent model system for studying magnetoelectric coupling.

4-Conclusions

In summary, the electronic and magnetic properties of the ternary intermetallic RMn_2Si_2 (R= La, Ce, Yb, and Y) compounds are calculated. Using the first-principles calculations, we demonstrate that these materials exhibit noncollinear magnetism in agreement with experimental results. By varying the distance between Mn-Mn atoms, we indicate that the presence of RKKY interactions. Also, the Dzyaloshinskii-Moriya interactions in these materials are discussed. These materials could be suggested as a good candidate for magnetoelectric applications.

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Authors' contributions:

-Conception and design of study: Najim Tahiri, Widad Bazine,

-Acquisition of data: Widad Bazine

-Analysis and/or interpretation of data: Najim Tahiri, Widad Bazine, Omar El Bounagui and Hamid Ez-Zahraouy.

-Drafting the manuscript: Najim Tahiri, Widad Bazine

-Revising the manuscript critically for important intellectual content: Najim Tahiri, Widad Bazine and Omar El Bounagui

-Approval of the version of the manuscript to be published: Najim Tahiri, Widad Bazine, Omar El Bounagui and Hamid Ez-Zahraouy.

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Figures

Figure 1

Unit cell structure of RMn_2Si_2 (R= La, Ce, Yb and Y) materials.

Figure 2

Total and partial density of states for RMn₂Si₂ materials: a) LaMn₂Si₂ b) YMn₂Si₂, c) CeMn₂Si₂, and d) YbMn₂Si₂.

Figure 3

Exchange coupling interactions JRKKY as a function of the distance between Mn-Mn atoms: (a) LaMn₂Si₂ (b) YMn₂Si₂ (c) CeMn₂Si₂ and (d) YbMn₂Si₂.