Electronic and Magnetic Properties of $\text{Mn}_2\text{Sb}_{1-x}\text{As}_x$ ($x = 0, 0.5, 1$)

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The electronic and magnetic properties of $\text{Mn}_2\text{Sb}_{1-x}\text{As}_x$ (where $x = 0, 0.5, 1$) are investigated from first-principles total-energy calculations. The predicted magnetic ground states (ferrimagnetism for $x = 0$ and antiferromagnetism for $x = 1$) are in agreement with experimental observations; however, this is not the case for $x = 0.5$. Here, it is found that the artificial change of the surrounding atoms of Mn stabilizes the antiferromagnetism. A similar change stabilizes ferrimagnetism (antiferromagnetism) for $x = 1$ ($x = 0$). These results indicate that the environment around the Mn atoms plays a very important role in the stabilization of antiferromagnetism in the $\text{Mn}_2\text{Sb}_{1-x}\text{As}_x$ system. In addition, an analysis of the sign of an effective exchange interaction as a function of the distance between the Mn atoms is performed.
[\[doi:10.2320/matertrans.M2014251\]](https://doi.org/10.2320/matertrans.M2014251)

(Received July 3, 2014; Accepted September 12, 2014; Published November 25, 2014)

Keywords: Mn_2Sb - Mn_2As system, antiferromagnetism, ferrimagnetism

1. Introduction

Mn_2Sb has a tetragonal crystal structure of the Cu_2Sb type with a 129th space group ($P4/nmm$).¹⁾ In the unit cell, there are two different crystallographic sites for the Mn atoms (Mn(I) and Mn(II)), where the Mn(I) (Mn(II)) atom is tetrahedrally (octahedrally) surrounded by Sb atoms. The ground state of Mn_2Sb is ferrimagnetic, and the magnetic moments of Mn(I) and Mn(II) are aligned antiparallel to one another.^{2–5)} Compounds of the forms $\text{Mn}_{2-x}\text{T}_x\text{Sb}$ and $\text{Mn}_2\text{Sb}_{1-x}\text{Z}_x$ (where $T = \text{V, Cr, Co, Cu, Zn}$ and $Z = \text{As, Ge, Sn}$) show a first-order phase transition from ferrimagnetic (FR) to antiferromagnetic (AF) states as temperature decreases.^{6–12)} The FR to AF transition is accompanied by a large change in the volume, resistivity, magnetization, and so on.^{13,14)} The application of a magnetic field induces the first-order AF to FR transition, which is accompanied by large magnetoresistance, magnetostriction, and so on.^{13,14)} Therefore, these compounds have attracted attention as magnetic-field-controlled materials.

We are interested in the origin of the stabilization of the AF state in the above-mentioned compounds. We begin by examining $\text{Mn}_2\text{Sb}_{0.5}\text{As}_{0.5}$, in addition to the stoichiometric compounds Mn_2Sb and Mn_2As for the following reasons. The ground state of Mn_2Sb (Mn_2As) is FR (AF), and the antiferromagnetic structure appearing in $\text{Mn}_{2-x}\text{T}_x\text{Sb}$ and $\text{Mn}_2\text{Sb}_{1-x}\text{Z}_x$ is the same as that appearing in $\text{Mn}_2\text{Sb}_{1-x}\text{As}_x$. We will discuss the origin of the stabilization of the AF state from the results of several first-principles total-energy calculations.

2. Approach

The crystal structure of Mn_2Sb is shown in Fig. 1, where the Mn atoms occupy two different crystallographic positions: $(0, 0, 0)$ and $(1/2, 1/2, 0)$ for Mn(I) and $(0, 1/2, u)$ and $(1/2, 0, -u)$ for Mn(II). The Sb atoms occupy the $(0, 1/2, v)$ and $(1/2, 0, -v)$ positions. Here, u and v are the atomic position parameters.¹⁾

To simulate a compound, such as $\text{Mn}_2\text{Sb}_{0.5}\text{As}_{0.5}$, we use a simple model with a 25th space group ($Pmm2$)¹⁾ as follows.

Fig. 1 Crystal structure of Mn_2Sb and schematic representations of several magnetic structures.

The unit cell is kept the same as that of Mn_2Sb , and one of the two Sb atoms is replaced with an As atom. In the later section, other models are considered.

For magnetic phases, the following five structures are considered: paramagnetic or non-magnetic (P), ferromagnetic (F), ferrimagnetic (FR), and two antiferromagnetic (AF1 and AF2) states (see Fig. 1). These magnetic phases (or spin configurations) were investigated about 40 years ago for the Mn_2Sb - Mn_2As system by molecular field theory.¹⁵⁾ The FR and AF1 structures are equal to those reported as the magnetic ground states for Mn_2Sb ³⁾ and Mn_2As ,¹⁶⁾ respectively.

First-principles total-energy calculations are performed using the full-potential linearized augmented plane wave (FLAPW) method.¹⁷⁾ The generalized gradient approximation of Perdew *et al.*¹⁸⁾ is used for the exchange-correlation potential. The plane wave cutoff is $RK_{\text{max}} = 7.0$, where R is the smallest atomic sphere radius and K_{max} is the magnitude of the largest K vector. For the atomic sphere radius, we used the values 2.24 a.u. for Mn and 2.13 a.u. for Sb and As. In the calculation of equilibrium positions of the atoms in a cell, we used a reverse-communication trust-region quasi-Newton method.¹⁷⁾

3. Results and Discussions

3.1 Mn_2Sb and Mn_2As

For nonmagnetic Mn_2Sb , we calculated the total energy,

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Table 1 Lattice constants a and c and atomic positions u and v of Mn_2Sb and Mn_2As . The atomic position is expressed in units of c . See text for detail.

	Mn_2Sb				Mn_2As			
	a (Å)	c (Å)	u	v	a (Å)	c (Å)	u	v
exp.*	4.080	6.560	0.710	0.279	3.761	6.265	0.735	0.330
P	3.818	6.139			3.590	5.980		
F	3.964	6.373			3.594	5.987		
FR	3.965	6.375			3.696	6.157		
AF1	3.974	6.390			3.692	6.150		
AF2	3.957	6.362			3.594	5.987		
P	3.818	6.139	0.778	0.331	3.590	5.980	0.685	0.281
FR	3.965	6.375	0.723	0.293	3.696	6.157	0.682	0.274
AF1	3.974	6.390	0.723	0.294	3.694	6.150	0.668	0.270

*Ref. 5) for Mn_2Sb and ref. 19) for Mn_2As .

Table 2 Total energies relative to a nonmagnetic state (P) for several magnetic states. The values in parentheses show the results after optimization of atomic positions for Mn(II), Sb, and As.

	Total energy per formula unit (eV)				
	P	F	FR	AF1	AF2
Mn_2Sb	0.00 (−0.41)	−0.74	−0.95 (−1.00)	−0.90 (−0.92)	−0.72
Mn_2As	0.00 (−0.28)	−0.03	−0.43 (−0.95)	−0.43 (−0.97)	−0.001

E_{tot} , as a function of c/a by fixing the cell volume, V , at the experimental value ($V = 109.2 \text{ Å}^3$). The result showed that E_{tot} is minimal at $c/a = 1.610$. This value is very close to the experimental one, $c/a = 1.608$.⁵⁾ Therefore, we did not optimize the value of c/a for the other magnetic states, and instead used the experimental values $c/a = 1.608$ ⁵⁾ and 1.666 ¹⁹⁾ for Mn_2Sb and Mn_2As , respectively.

The optimal values for the lattice constant, a , and atomic positions are listed Table 1. These values were determined by the following steps. First, we calculated E_{tot} as a function of a and estimated the value, a_{min} , that gives the minimum total energy. Here, the atomic positions were fixed at the experimental values.^{5,19)} Second, we fixed the lattice constant, a , at a_{min} and optimized the atomic positions of Mn(II), Sb, and As by minimizing the forces acting on those atoms. Third, we optimized the value of a using the atomic positions determined in the second step. Concerning the lattice constant, the discrepancy between our results and the experimental ones is below 3% for both Mn_2Sb and Mn_2As .

The E_{tot} values for several magnetic states are listed in Table 2. Before optimizing the atomic positions, the values of E_{tot} for two magnetic states (F and AF2) were higher than those for FR and AF1 in both Mn_2Sb and Mn_2As . Therefore, we did not optimize the atomic positions for F and AF2. After the optimization of the atomic positions, the differences, $\Delta E_{\text{tot}} = E_{\text{tot}}(\text{FR}) - E_{\text{tot}}(\text{AF1})$ were found to be -0.08 eV and $+0.02 \text{ eV}$ for Mn_2Sb and Mn_2As , respectively. Here, $E_{\text{tot}}(\text{FR})$ and $E_{\text{tot}}(\text{AF1})$ mean the total energy per formula unit for the FR and AF1 states, respectively. The value of ΔE_{tot} for Mn_2As is very close to that of the early all-electron FLAPW calculation ($+0.03 \text{ eV}$).²⁰⁾ Although ΔE_{tot} is small, the predicted magnetic ground states from ΔE_{tot} are

Table 3 Magnetic moment per formula unit (M_{tot}) and those on constituent atoms (units μ_B).

		M_{tot}	Mn(I)	Mn(II)	Sb/As	
Mn_2Sb	theory	1.33	−2.17	3.39	0.003	this work
		1.40	−2.31	3.58	0.04	Ref. 10)
		1.76	−2.11	3.65	0.01	Ref. 21)
	experiment	1.80	−2.10	3.90		Ref. 4)
		1.78	−1.77	3.55		Ref. 5)
		1.74	−2.13	3.87		Ref. 3)
Mn_2As	theory	1.10	−1.80	2.90		Ref. 16)
			−1.89	3.24	−0.05	this work
			−1.98	3.44		Ref. 20)
	experiment		−1.87	3.43		Ref. 22)
			−3.70	3.50		Ref. 16)

consistent with experimental reports^{3,16)} because the experimental observations showed that Mn_2Sb (Mn_2As) has the FR (AF1) state for its magnetic ground state.

Our results for the magnetic moments of FR Mn_2Sb and AF1 Mn_2As are summarized in Table 3, together with previous theoretical^{20–22)} and experimental results.^{3–5,10,16)} This table shows that our results are in agreement with other theoretical calculations in both Mn_2Sb and Mn_2As . The discrepancy between theoretical and experimental results is small in Mn_2Sb . In Mn_2As , there is a good agreement between theoretical and experimental results for the magnetic moment of Mn(II). However, the experimental value of Mn(I) is twice as much as its theoretical value. The cause of this discrepancy is unknown.

3.2 $\text{Mn}_2\text{Sb}_{0.5}\text{As}_{0.5}$

3.2.1 Chemical ordered structure

For this compound, two magnetic states, FR and AF1, are considered. The lattice constants and atomic positions are determined by the following four steps. First, we calculated E_{tot} as a function of a by fixing c/a and the atomic positions at their experimental values for Mn_2As .¹⁹⁾ Then, we estimated the value, a_{min} (or V_{min}), that gives a minimum total energy. Second, we calculated E_{tot} as a function of c/a using V_{min} and the experimental atomic positions, and then we estimated the value, $(c/a)_{\text{min}}$ that gives a minimum total energy. Third, we optimized the atomic positions using V_{min} and $(c/a)_{\text{min}}$. In the fourth step, we re-estimated $(c/a)_{\text{min}}$ using V_{min} and the atomic positions determined in the third step. The result of total energy vs. c/a is shown in Fig. 2, and the obtained data are summarized in Table 4. The obtained magnetic data are listed in Table 5.

Figure 2 shows that the energy difference between FR and AF1 is negligible before the optimization of the atomic positions. However, the FR state is more stable than the AF1 state after optimization. This result indicates that the optimization of the atomic positions is crucial to the determination of the relative stability of the two magnetic phases, FR and AF1.

From Tables 1 and 4, we noticed that both a and c decrease with increasing As composition. This finding is in good agreement with previous experimental observations.¹⁵⁾ Our

Fig. 2 Total energy per formula unit vs. c/a for $\text{Mn}_2\text{Sb}_{0.5}\text{As}_{0.5}$. Upper (lower) two lines show the results for before (after) the optimization of the atomic positions. The point A denotes $a_{\min}$. The points B and C denote $(c/a)_{\min}$. See text for details.

Table 4 Lattice constants (a and c) and atomic positions (z , u , and v) of Mn(I), Mn(II), Sb, and As in $\text{Mn}_2\text{Sb}_{0.5}\text{As}_{0.5}$. The atomic positions are expressed in units of lattice constant c . The words “A”, “B”, and “C” mean the points shown in Fig. 2.

		FR			AF1		
		A	B	C	A	B	C
	a (Å)	3.797	3.784	3.802	3.798	3.750	3.800
	c (Å)	6.325	6.368	6.308	6.326	6.490	6.319
	c/a	1.666	1.683	1.659	1.666	1.731	1.663
Mn(I)	(0, 0, z_1)	0	0	0.011	0	0	0.010
	(1/2, 1/2, z_2)	0	0	0.011	0	0	0.010
Mn(II)	(0, 1/2, u_1)	0.735	0.735	0.706	0.735	0.735	0.702
	(1/2, 0, u_2)	0.265	0.265	0.312	0.265	0.265	0.312
Sb	(0, 1/2, v_1)	0.330	0.330	0.286	0.330	0.330	0.290
As	(1/2, 0, v_2)	0.670	0.670	0.721	0.670	0.670	0.720

Table 5 Magnetic moments (units μ_B) on constituent atoms and per formula unit (M_{tot}) in $\text{Mn}_2\text{Sb}_{0.5}\text{As}_{0.5}$. The words “B” and “C” mean the points shown in Fig. 2.

$\text{Mn}_2\text{Sb}_{0.5}\text{As}_{0.5}$	M_{tot}	B			C		
		Mn(I)	Mn(II)	M_{tot}	Mn(I)	Mn(II)	
FR	1.09	−2.10	3.09	1.32	−2.08	3.29	
AF1		−2.21	3.01		−1.96	3.25

Fig. 3 Surrounding atoms of constituent atoms in the FR Mn_2Sb (upper), AF1 Mn_2As (lower) states. The symbols “I” and “II” refer to the Mn(I) and Mn(II) atoms, respectively. For example, the upper panel shows that the Mn(I) atom has four Mn(II), four Sb, and four Mn(I) atoms in the 1st, 2nd, and 3rd neighbor positions, respectively.

result for the magnetic ground state does not agree with the result from the literature¹⁵⁾ because the FR state is more stable than the AF1 state. To clarify the cause of this disagreement, we consider the following two aspects: the chemical or compositional disorder between the Sb and As atoms and the atoms surrounding the constituent atoms. The second aspect is especially expected to be important because the neighboring atoms of each of the constituent in the FR Mn_2Sb case are much different from those in the AF1 Mn_2As case, as shown from Fig. 3, where the FR and AF1 states are the magnetic ground states for Mn_2Sb and Mn_2As , respectively.

3.2.2 Chemical disorder between Sb and As

To investigate the effect that chemical or compositional disorder between the Sb and As atoms has on the magnetic phases of $\text{Mn}_2\text{Sb}_{0.5}\text{As}_{0.5}$ (FR and AF1), we considered three other models (types 2, 3, and 4) shown in Fig. 4, which have different configurations of Sb and As from the type 1 model (already considered in the previous section). In the type 1 and 2 models, either Sb or As exists on the c plane. In the type 3 (type 4) model, Sb and As are alternated in either (both) the a and b directions on the c plane. For the AF1 state, we used the same cells (see Fig. 4) for the type 2 and 4 models.

The calculations for types 3 and 4 converged to the state that corresponded with the initial magnetic arrangement, FR or AF1, but those for type 2 converged only to the FR state irrespective of the initial magnetic arrangement. As shown in Fig. 5, the energy of the FR state is lower than that of the AF1 state in three of the model types. These results indicate that the relative stability of two magnetic phases may not be sensitive to chemical or compositional disorder between the Sb and As atoms.

3.2.3 Environment of constituent atom

By fixing the lattice constants (a and c) at their optimal values for the FR Mn_2Sb case (which has the lowest total energy among several magnetic phases) and decreasing the value of the atomic position z of Mn(II), we calculated the total energy, E_{tot} , for each of the FR and AF1 states. In this case, the Mn(II) atom was moved closer to the Sb atom. We performed the same calculations for Mn_2As ($\text{Mn}_2\text{Sb}_{0.5}\text{As}_{0.5}$) as those performed Mn_2Sb , fixing a and c at their optimal values for the AF1 Mn_2As (FR $\text{Mn}_2\text{Sb}_{0.5}\text{As}_{0.5}$) case. In the Mn_2As case, the Mn(II) atom was separated farther from the As atom in the z direction. Although we could have independently moved the two Mn(II) atoms in the cell of

Fig. 4 Several models for $\text{Mn}_2\text{Sb}_{0.5}\text{As}_{0.5}$. Fig. 5 Total energies (per formula unit) of the type 1, 3, and 4 models for $\text{Mn}_2\text{Sb}_{0.5}\text{As}_{0.5}$.

$\text{Mn}_2\text{Sb}_{0.5}\text{As}_{0.5}$, for simplicity, we moved one Mn(II) atom closer to the Sb atom and the other closer to the As atom by the same amount.

In all cases, the movement of Mn(II) resulted in an increase of E_{tot} , but changed the sign of ΔE_{tot} , as shown in Fig. 6, where $\Delta E_{\text{tot}} = E_{\text{tot}}(\text{AF1}) - E_{\text{tot}}(\text{FR})$. For Mn_2Sb and $\text{Mn}_2\text{Sb}_{0.5}\text{As}_{0.5}$, the sign was changed from positive to negative. This means that the AF1 state was relatively stabilized by a decrease in the distance between the Mn(II) and Sb (or As) atoms in the z direction. On the other hand, for Mn_2As , the sign was changed from negative to positive. This means that the FR state was relatively stabilized by the increase in the distance between the Mn(II) and As atoms in the z direction.

To clarify the environment of a constituent atom in the cell, we show its 1st, 2nd, and 3rd nearest neighbor atoms in Fig. 7. For example, the Mn(I) atom in the upper panel of Fig. 7(a) has four Mn(II), four Sb, and four Mn(I) atoms as its 1st, 2nd, and 3rd nearest neighbors, respectively. The most remarkable features observed are as follows. Each of the Mn(II) and Sb (or As) atoms becomes the 1st nearest neighbor when the AF1 state is relatively stable. On the other hand, each of Mn(I) and Mn(II) atoms becomes the 1st nearest neighbor when the FR state is relatively stable.

Fig. 6 Total energy per formula unit vs. atomic position. See text for details.

3.3 Effective exchange interaction

In Fig. 8, we show the partial density of states (*DOS*) of Mn(I) and Mn(II) in the FR Mn_2Sb and AF1 Mn_2As cases, which have been optimized with respect to a , c , and atomic positions (see Sec. 3.1). Note that our *DOS* are quite similar to those obtained by other researchers,^{10,20–22} and that the *DOS* for both the FR and AF1 states are very much alike except for a few details. The most notable feature is that the magnetic moments of Mn(I) and Mn(II) may be regarded as localized (as explained by Küber *et al.* for Mn atoms in some Heusler alloys²³). Thus, in this section, we reconsider the results of Sec. 3.2.2 in terms of an effective exchange interaction, $J(r)$, between Mn atoms with interatomic distance r , which is described schematically in Fig. 9.

First, we focus on the Mn(II) atom because its magnetic moment is larger than that of the Mn(I) atom (see Table 3). In Table 6, we show the interatomic distances between the Mn(II) atoms and their 1st to 3rd nearest neighbors in Mn_2Sb , Mn_2As , and $\text{Mn}_2\text{Sb}_{0.5}\text{As}_{0.5}$. The 1st, 2nd, and 3rd neighbor's distances are denoted as r_1 , r_2 , and r_3 , respectively. As seen in Fig. 10, the magnetic moments between the 2nd neighbors must be parallel (ferromagnetically-coupled) in both the FR and AF1 states, and those between the 3rd neighbors must also be parallel. These facts indicate that $A < r_2$ and $r_3 < B$. In the FR state, $A < r_1$, because the magnetic moments between the 1st neighbors must be parallel. Whereas, in the AF1 state, $A > r_1$, because the magnetic moments between the 1st neighbors must be antiparallel (antiferromagnetically-

Fig. 7 Surrounding atoms of constituent atoms in (a) Mn_2Sb , (b) Mn_2As , and (c) $\text{Mn}_2\text{Sb}_{0.5}\text{As}_{0.5}$. For example, the Mn(I) atom in the upper panel of Fig. (a) has four Mn(II), four Sb, and four Mn(I) atoms in the 1st, 2nd, and 3rd neighbor positions, respectively. In each panel, the upper part shows the initial state and the lower part shows the state in which relative stabilization between FR and AF1 is changed. The changes of the atomic position, u , of Mn(II) are as follows: -0.055 , 0.044 , and 0.040 for (a), (b), and (c), respectively.

coupled). From Table 6 and the above relationship among A , B , r_1 , r_2 , and r_3 , we conclude that $A \sim 3.5 \text{ \AA}$ and $B > 5.1 \text{ \AA}$.

Note that, Endoh and Ishikawa²⁴⁾ estimated $J(r)$ from the magnetic structures of Pt_3Mn , PtMn , PtMn_3 , and other compounds and reported that $A = 3 \text{ \AA}$ and $B = 4 \text{ \AA}$. Concerning $J(r)$ for Ni_2MnSn , $A = 3.2 \text{ \AA}$ and $B = 7.7 \text{ \AA}$.²⁵⁾ Our estimation for the value of A does not differ much from theirs and that for B is an intermediate value between two cases.

Here, we focus on the Mn(I) atom. In Table 7, we show the interatomic distances between Mn(I) and Mn (Mn(I) or Mn(II)) and the 1st to 4th nearest neighbors in Mn_2Sb , Mn_2As , and $\text{Mn}_2\text{Sb}_{0.5}\text{As}_{0.5}$. The positions of the neighboring Mn atoms are depicted in Fig. 11, where the definition of $r_{\text{I-I}}$, $r'_{\text{I-I}}$, $r_{\text{I-II}}$, and $r'_{\text{I-II}}$ is given. The table shows that $2.61 \text{ \AA} \leq r_{\text{I-I}} \leq 2.80 \text{ \AA}$, $3.69 \text{ \AA} \leq r'_{\text{I-I}} \leq 3.96 \text{ \AA}$, $2.56 \text{ \AA} \leq r_{\text{I-II}} \leq 2.90 \text{ \AA}$, and $2.78 \text{ \AA} \leq r'_{\text{I-II}} \leq 2.97 \text{ \AA}$. As seen in Fig. 11, the magnetic moments for the distances $r_{\text{I-I}}$ and $r'_{\text{I-I}}$ must be parallel (ferromagnetically-coupled) in both the FR and AF1 states. Whereas, the magnetic moments for the distances of $r_{\text{I-II}}$ and $r'_{\text{I-II}}$ must be antiparallel (antiferromag-

netically-coupled) in both the FR and AF1 states. Therefore, $J_{\text{I-I}}(r) > 0$ for $2.61 \text{ \AA} \leq r \leq 2.80 \text{ \AA}$ and $3.69 \text{ \AA} \leq r \leq 3.96 \text{ \AA}$, $J_{\text{I-II}}(r) < 0$ for $2.56 \text{ \AA} \leq r \leq 2.97 \text{ \AA}$. Here, $J_{\text{I-I}}$ ($J_{\text{I-II}}$) denotes an effective exchange interaction between the Mn(I) and Mn(I) (Mn(I) and Mn(II)) atoms.

We estimated only the sign of an effective exchange interaction, $J(r)$, between Mn atoms from the data obtained by our calculation. Using theoretically estimated $J_{\text{I-I}}(r)$, $J_{\text{I-II}}(r)$, and $J_{\text{II-II}}(r)$, we can explain the magnetic transition from FR to AF1 by the Heisenberg Hamiltonian or molecular field theory, where $J_{\text{II-II}}(r)$ denotes an effective exchange interaction between Mn(II) atoms. However, further studies are needed to qualitatively estimate $J(r)$.

4. Summary

We investigated several magnetic phases for $\text{Mn}_2\text{Sb}_{1-x}\text{As}_x$ ($x = 0, 0.5, 1$) by first-principles total-energy calculations. The predicted magnetic ground states for $x = 0$ (ferrimagnetic) and $x = 1$ (antiferromagnetic) are consistent with previous experimental observations, but that for $x = 0.5$

Fig. 8 Partial density of states of Mn(I) and Mn(II) in FR Mn_2Sb and AF1 Mn_2As . The Fermi energy is at $E = 0$.

Fig. 9 Effective exchange interaction $J(r)$.

Table 6 Interatomic distances ($\text{\AA}$) between Mn(II) from the 1st to 3rd nearest neighbors. The number of neighbors is shown in parentheses. For each material, the upper (lower) row shows the data of the FR (AF1) state (see Fig. 6).

	dz	1st	2nd	3rd
Mn_2Sb	0	3.96 (4)	3.99 (4)	4.51 (4)
	-0.055	3.53 (4)	3.96 (4)	5.08 (4)
Mn_2As	0.044	3.69 (4)	3.69 (4)	4.40 (4)
	0	3.33 (4)	3.69 (4)	4.85 (4)
$\text{Mn}_2\text{Sb}_{0.5}\text{As}_{0.5}$	0	3.66 (4)	3.80 (4)	4.67 (4)
	0.040	3.34 (4)	3.80 (4)	5.09 (4)

(antiferromagnetic) does not agree with the literature. However, we gained further insight into the stabilization of the antiferromagnetic state for not only $x = 0.5$ but also $x = 0$

Fig. 10 Neighbors of Mn(II) and distances between Mn(II) atoms.

Table 7 Neighboring atoms of Mn(I) and their distances ($\text{\AA}$). For each material, the upper (lower) row shows the data of the FR (AF1) state (see Fig. 6). The notation (I, 4) and (I', 4), (II, 2), and (II', 2) denote that four Mn(I), four Mn(II), two Mn(II), and two Mn(II) atoms exist as neighbors of Mn(I) for the distances $r_{\text{I-I}}$, $r'_{\text{I-I}}$, $r_{\text{I-II}}$, and $r'_{\text{I-II}}$, respectively (see Fig. 11).

	dz	1st	2nd	3rd	4th
Mn_2Sb	0	2.65 (II, 4)	2.80 (I, 4)	3.96 (I', 4)	
	-0.055	2.80 (I, 4)	2.90 (II, 4)	3.96 (I', 4)	
Mn_2As	0.044	2.56 (II, 4)	2.61 (I, 4)	3.69 (I', 4)	
	0	2.61 (I, 4)	2.75 (II, 4)	3.69 (I', 4)	
$\text{Mn}_2\text{Sb}_{0.5}\text{As}_{0.5}$	0	2.61 (II, 2)	2.68 (I, 4)	2.78 (II', 2)	3.80 (I', 4)
	0.040	2.69 (I, 4)	2.79 (II, 2)	2.97 (II', 2)	3.80 (I', 4)

Fig. 11 Neighbors of Mn(I) and distances between Mn atoms. For Mn_2Sb and Mn_2Sb , $r_{\text{I-II}} = r'_{\text{I-II}}$.

from additional total-energy calculations. The magnetic phase is not sensitive to the chemical or compositional disorder between the Sb and As atoms. If the Sb and/or As atoms become nearest to one of the two nonequivalent Mn atoms in the cell (that is Mn(II)), then the antiferromagnetic state stabilizes. On the other hand, if the two non-equivalent Mn atoms in the cell become nearest to each other, then the ferrimagnetic state stabilizes. In addition, another interpretation of these results is given in terms of an effective exchange interaction between Mn atoms.

Acknowledgments

This work was supported by JSPS KAKENHI Grant Number 24560855.

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