Physical origin of the incommensurate spin spiral structure in Mn_3Si

M. Hortamani,[1,a)] L. Sandratskii,[1] P. Zahn,[2] and I. Mertig[1,2]
[1]*Max-Planck-Institut für Mikrostrukturphysik, D-06120 Halle, Germany*
[2]*Martin-Luther-Universität Halle-Wittenberg Institut für Physik, D–06099 Halle, Germany*

(Presented 12 November 2008; received 23 September 2008; accepted 4 November 2008; published online 23 February 2009)

We report first-principles study of the magnetic structure of Mn_3Si. The Mn atoms form two sublattices. One of them is (Mn_I) characterized by large atomic magnetic moments and the other one (Mn_{II}) possesses induced moments. It is shown that in agreement with experiment the magnetic ground state is a spin spiral. We found that the spiral magnetic structure is formed as a result of the competition between direct antiferromagnetic interaction of the Mn_I magnetic moments and indirect ferromagnetic interaction of the same moments through the magnetic moment of the Mn_{II} sublattice. We demonstrate that this competition is strongly volume dependent and leads to volume dependence of the wave vector of the spin spiral. These properties are related to the volume dependence of the induced moments of Mn_{II} atoms. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3068422]

I. INTRODUCTION

In the previous work we discussed the possibility of formation of a Mn_3Si film on the Si(111) surface. This film has features that are useful for the realization of spin injection through the metal-semiconductor interface.[1,2] Mn_3Si belongs to the class of Heusler compounds of the type X_2YZ.[3] The crystal structure can be described in terms of four interpenetrating fcc Bravais lattices. The Si and Mn_I atoms are located at (0,0,0) and $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$, respectively, while the Mn_{II} atoms occupy $(\frac{1}{4},\frac{1}{4},\frac{1}{4})$ and $(\frac{3}{4},\frac{3}{4},\frac{3}{4})$ sites. All atomic positions are given in units of lattice parameter a. The Mn_I sites are surrounded by eight Mn_{II} nearest neighbors and the nearest neighbors of Mn_{II} are four Mn_I and four Si atoms. The Mn_3Si was found experimentally to have incommensurate magnetic spiral structure with zero net magnetization[4,5] and a spiral vector of $0.425(2\pi/a)(111)$.[6,7] The magnetic transition temperature is 23 K. The experimental values of the atomic moments are $\mu_I=1.7\mu_B$ and $\mu_{II}=-0.19\mu_B$.[4]

II. CALCULATIONAL DETAILS

The calculations are performed within the framework of the density functional theory using the augmented spherical wave method.[8,9] The generalized gradient approximation to the exchange-correlation potential is employed. The spin spiral is characterized by the wave vector $\mathbf{q}$ defining the relative directions of the magnetic moments. For an atom at the position $\mathbf{r_i}$ the polar angle ϕ is given by $\phi_i=\mathbf{q}\cdot\mathbf{r_i}$. The azimuthal angle θ was chosen to be 90° for all atomic moments. We perform calculations for the vectors $\mathbf{q}$ parallel to the (111) axis. The wave vector is presented in the form $\mathbf{q}=q(2\pi/a)(111)$. Therefore q gives the length of vector $\mathbf{q}$ in units of $2\pi\sqrt{3}/a$.

III. RESULTS AND DISCUSSION

In Fig. 1 we present the total energy as a function of lattice parameter for the nonmagnetic state and the magnetic state with $q=q_{exp}=0.425$. The curves are fitted using the Murnaghan equation. The minimum energy of the magnetic structure is lower than the minimum energy of the nonmagnetic state by 0.35 eV per formula unit. At the lattice parameter of $a=10.84$ Å, which corresponds to the theoretical equilibrium volume, the magnetic moments of the Mn sublattices are 2.6 and $-0.8\mu_B$ for Mn_I and Mn_{II}, respectively. These values of the magnetic moments are substantially larger than the values extracted from the neutron scattering experiment. On the other hand, for a lattice parameter of $a=10.25$ Å the calculated atomic moments 1.78 and $-0.18\mu_B$ are very close to the experimental values. At present we cannot suggest an explanation why the values of the magnetic moments at the equilibrium volume deviate from the experimental ones. The experiment[4] shows that the magnetism of the system is highly unusual and strong magnetic fluctuations can influence the values of the observed moments.

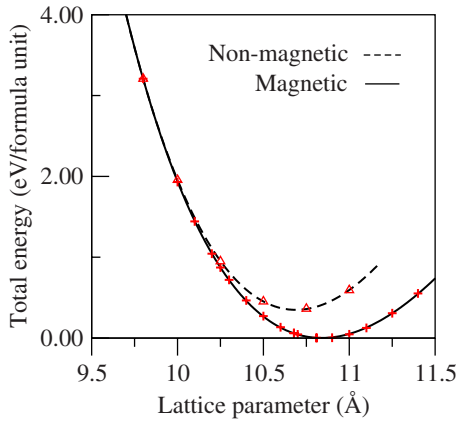

FIG. 1. (Color online) The total energy as a function of lattice parameter for the nonmagnetic state and the magnetic state with $q=q_{exp}=0.425$. The energy origin is taken at the minimal energy of the magnetic state.

a)Electronic mail: hortamani@mpi-halle.mpg.de.

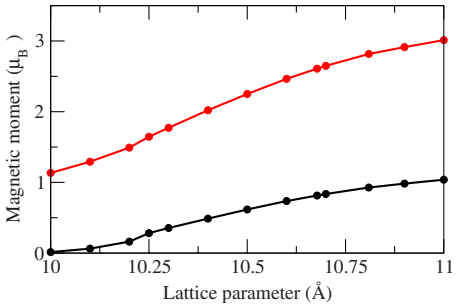

FIG. 2. (Color online) Magnetic moments of the two sublattices for the spiral structure with $q=q_{exp}$ as a function of lattice parameter. The upper curve shows Mn_I moment and the lower curve shows Mn_{II} moment.

To get a deeper insight into the origin of the spiral magnetic structure in Mn_3Si, we study in this paper the magnetic properties of the system at different volumes varying lattice parameter from 10 to 11 Å. At the beginning of the lattice parameter interval, the value of the moment of Mn_{II} sublattice is very small (Fig. 2). The magnetic moments of both sublattices increase monotonously with volume expansion (Fig. 2). This is an expected behavior since increasing volume leads to narrower energy bands.

Above we discussed calculations for the spiral structures with experimental wave vector. Now we turn to the study of the dependence of the electronic properties on the value of the wave vector. Figure 3 shows the q-dependence of the magnetic moments of both sublattices for two values of the lattice parameter. The q-dependence of the moment of the Mn_I sublattice is relatively weak. It varies between 1.64 and $1.50\mu_B$ at $a=10.25$ Å and between 3.01 and $2.55\mu_B$ at $a=11$ Å. The induced magnetic moment of the second sublattice has a very different behavior. At $a=10.25$ Å, the moment is small for all q. It remains almost unchanged up to $q=0.3$ and decreases monotonously for larger q, goes to zero at $q=1$. At $a=11$ Å, this induced magnetic moment of Mn_{II} sublattice has a value close to $1\mu_B$ in the q interval from 0 to about 0.6. For larger q values the moment decreases monotonously, vanishing at $q=1$.

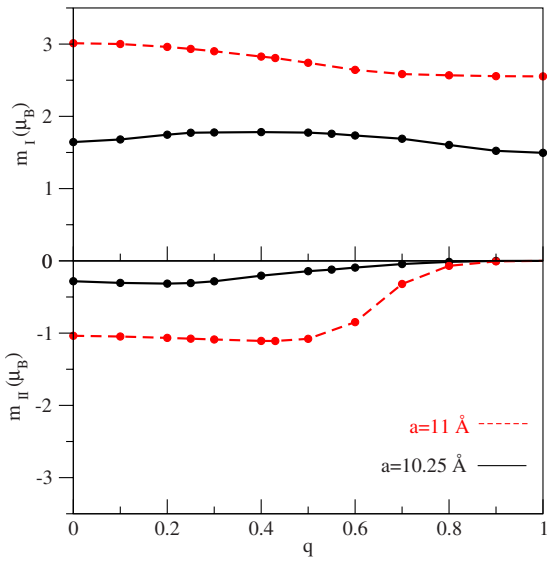

FIG. 3. (Color online) Mn magnetic moments as a function of q for $a=10.25$ and $a=11.00$ Å. The upper panel presents the moment of Mn_I atoms and the lower panel shows the moment of Mn_{II} sublattice.

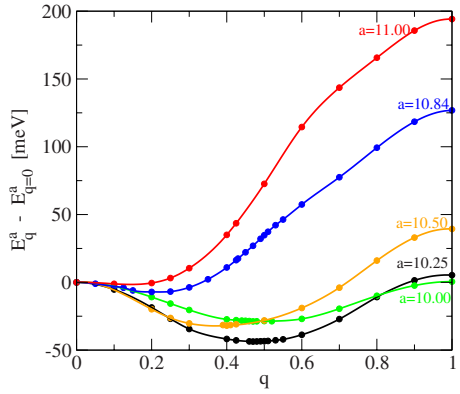

FIG. 4. (Color online) Total energy vs q for a number of lattice parameters. For each volume the energy origin is selected at the energy of the $q=0$ structure.

It is important to notice that the magnetic structure of the Mn_I sublattice is collinear ferromagnetic for both $q=0$ and $q=1$ and is collinear antiferromagnetic for $q=0.5$. On the other hand the relative orientation of the moments of the Mn_I and Mn_{II} sublattices changes continuously with the variation in q in the whole interval from 0 to 1. For $q=0$ the moments of two sublattices are collinear whereas for $q=1$ they are orthogonal to each other. This orthogonality according to the q value explains why the moment of the second sublattice tends to zero. Since the moments of the Mn_{II} atoms are induced by the moments of the Mn_I sublattice, they vanish if the directions of the moments become orthogonal.

A closer inspection of the $m_I(q)$ shows that it is almost symmetric with respect to $q=0.5$ at $a=10.25$ Å, while it decreases monotonously in the whole q interval at $a=11$ Å. This difference in the $m_I(q)$ behavior is related to the value of induced moments of the Mn_{II} sublattice.

Next we consider the q dependence of the total energy calculated for various values of the lattice parameter (c.f. Figure 4). For $a=10$ Å and $a=10.25$ Å the energy curves are almost symmetric with respect to $q=0.5$. The form of the curves can be well described by one cosine function with periodicity of 1. The position of the minimum of the curves is close to the experimental wave vector of the spiral structure. The closeness of the energy curve to a symmetric form is in correlation with the same property of $m_I(q)$ and reveals that the induced moment of Mn_{II} sublattice has weak influence on the equilibrium magnetic structure. Therefore one can conclude that a direct exchange interaction between Mn_I moments stimulates the formation of a collinear antiferromagnetic structure. In terms of the Heisenberg model this one-harmonic behavior corresponds to the nearest-neighbor antiferromagnetic interaction between Mn_I moments.

With expansion of the crystal the behavior of the $E(q)$ curve changes strongly. It deviates from the simple cosine-type, which means that other interatomic interactions become important. At a lattice parameter of $a=11$ Å the $E(q)$ curve becomes rather close to a cosine-type with a period twice as large as at $a=10.25$ Å. A larger period in q space

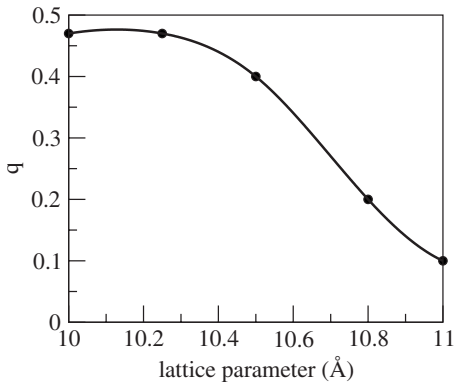

FIG. 5. The dependence of the spin spiral wave vector q on the lattice parameter.

corresponds to the magnetic interaction between atoms that are closer in the real space. In our case this is the result of the increasing value of the induced moments of the Mn_{II} sublattice. Indeed for $a=11$ Å the induced moment assumes a large value of about $1\mu_B$ in a broad q interval (Fig. 3). The form of the energy curve shows that an indirect exchange interaction between Mn_I moments mediated by the Mn_{II} moments is ferromagnetic. This conclusion is additionally supported by the fact that, from the point of view of the Mn_I sublattice, both the $q=0$ and $q=1$ cases correspond to the same collinear ferromagnetic structure. A very large energy difference between $q=0$ and $q=1$ for $a=11$ Å results from the vanishing of the moments of Mn_{II} atoms for $q=1$.

Now we can give the following interpretation of the formation of the magnetic structure. The direct exchange interaction between nearest atoms of the Mn_I sublattice (Mn_I-Mn_I) is antiferromagnetic stimulating the formation of a collinear antiferromagnetic configuration of the moments of this sublattice. With increasing lattice volume, the value of the magnetic moments of the Mn_{II} sublattice increases strongly. The exchange interactions involving Mn_{II} moments stimulate parallel orientation of the Mn_I moments. As a result there is a competition between effective ferromagnetic and antiferromagnetic interactions in the system leading to the formation of the noncollinear spiral configuration in a broad interval of the lattice volumes.

In Fig. 5 we show that the lattice parameter dependence of the spiral wave vector decreases monotonously from the value close to $q=0.5$ at lattice parameter $a=10$ Å to $q=0.1$ at $a=11$ Å. The stronger the intersublattice exchange interaction the smaller the equilibrium q value. On the other hand, the weaker the intersublattice interaction, the closer the magnetic structure to the commensurate antiferromagnetic configuration with $q=0.5$.

IV. CONCLUSION

We found that the spiral magnetic structure is formed as a result of the competition between direct antiferromagnetic interaction of the Mn_I magnetic moments of the main magnetic sublattice and indirect ferromagnetic interaction of the same moments through the Mn_{II} sublattices with induced moments. We demonstrate that this competition is strongly volume dependent and leads to a strong variation in the spin spiral wave vector.

[1] M. Hortamani, P. Kratzer, and M. Scheffler, Phys. Rev. B **76**, 235426 (2007).
[2] M. Hortamani, Ph.D. thesis, Freie Universität, Berlin, 2006.
[3] R. de Groot and F. Mueller, Phys. Rev. Lett. **50**, 2024 (1983).
[4] C. Pfleiderer, J. Beouf, and H. Löhneysen, Phys. Rev. B **65**, 172404 (2002).
[5] S. Tomiyoshi, Y. Yamaguchi, and M. Ohashi, Phys. Rev. B **36**, 2181 (1987).
[6] S. Tomiyoshi, E. Cowley, and H. Onodera, Phys. Rev. B **73**, 024416 (2006).
[7] S. Tomiyoshi and H. Watanabe, J. Phys. Soc. Jpn. **39**, 295 (1975).
[8] A. R. Williams, J. Kübler, and C. D. Gelatt, Phys. Rev. B **19**, 6094 (1979).
[9] L. M. Sandratskii, Adv. Phys. **47**, 91 (1998).