

Disorder dependence of the magnetic moment of the half-metallic ferromagnet NiMnSb from first principles

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Using half-metallic ferromagnets in spin-dependent devices, like spin valves and ferromagnetic tunnel junctions, is expected to increase the device performance. However, using the half-metallic ferromagnet NiMnSb in such devices led to much less than ideal results. One of the possible sources for this behavior is atomic disorder. First-principles calculations of the influence of atomic disorder on the electronic structure of NiMnSb underline the sensitivity of half-metallic properties in NiMnSb to atomic disorder. In this article, we report on the disorder dependence of the total magnetic moment calculated by applying the layer Korringa–Kohn–Rostoker method in conjunction with the coherent potential approximation. We consider the following types of disorder: (1) intermixing of Ni and Mn, (2) partial occupancy of a normally vacant lattice site by Ni and Mn, and (3) partial occupancy of this site by Mn and Sb. In all cases the composition is kept stoichiometric. All three types of disorder decrease the moment monotonically with increasing disorder levels. For the experimentally seen disorder of 5% Mn and 5% Sb on the normally vacant lattice site, the total moment is decreased by 4.1%. The results suggest that precise measurement of the saturation magnetization of NiMnSb thin films can give information on the disorder. © 2000 American Institute of Physics. [S0021-8979(00)50008-5]

INTRODUCTION

Spin-dependent devices increasingly gain importance in many applications. Spin valves are being used in hard-disk read heads and in many sensors. Both spin valves and ferromagnetic tunnel junctions are considered as building blocks for nonvolatile memory devices. Spin-dependent devices gain efficiency if the spin polarization in their ferromagnetic thin films is increased. An ideal 100% spin polarization at the Fermi energy is found in half-metallic ferromagnets. The semi-Heusler alloy NiMnSb is such a ferromagnet^{1,2} and it has been attempted to use NiMnSb in spin-dependent devices.^{3–6} However, the results are not consistent with a high spin polarization. It has been pointed out that atomic disorder in the related half-metallic ferromagnet PtMnSb can significantly influence the half-metallic property of this material.⁷ First-principles electronic structure calculations for NiMnSb show indeed that a disorder level of only a few percent is sufficient to create minority-spin states at the Fermi energy and significantly reduce the spin polarization.⁸ Such disorder levels are realistic in thin films⁹ and thus indeed could be the explanations for the present failure to produce half-metallic NiMnSb films in spin-dependent devices. In this article we will show that a second consequence of atomic disorder is the reduction of the magnetic moment in NiMnSb.

CALCULATION

NiMnSb crystallizes in the $C1_b$ structure. It consists of four interpenetrating fcc lattices labeled A, B, C, and D. Sites A, B, and D are occupied by Ni, Mn, and Sb, respectively, while site C is unoccupied. We apply the layer Korringa–Kohn–Rostoker method in conjunction with the coherent potential approximation (LKRR–CPA)¹⁰ to calculate the total magnetic moment of NiMnSb with atomic disorder. The types of disorder we consider are summarized in Table I. Type A–B has intermixing of Ni and Mn. In the case of disorder type C (C') the normally empty site C is occupied by Ni and Mn (Mn and Sb). In all cases the composition is kept stoichiometric. X-ray diffraction measurements by Kautzky *et al.*¹¹ are consistent with disorder type C' and a disorder level of 5%. The experimental lattice constant¹² of 5.927 Å is used throughout. We use the atomic sphere approximation with an empty sphere for unoccupied sites. In our calculations we use the layer doubling algorithm,¹⁰ repeating (111) layers with four sites in the layer

TABLE I. Types of disorder considered in this article. The second column shows the sites with interchange of atoms (or vacancies). Occupancies of the four lattice sites are shown for the three types of disorder.

Disorder	Occupancies					
	Type	Scheme	Site A	Site B	Site C	Site D
A–B	A↔B		Ni _{1-x} Mn _x	Ni _x Mn _{1-x}	...	Sb
C	AB↔C		Ni _{1-x}	Mn _{1-x}	Ni _x Mn _x	Sb
C'	BD↔C		Ni	Mn _{1-x}	Mn _x Sb _x	Sb _{1-x}

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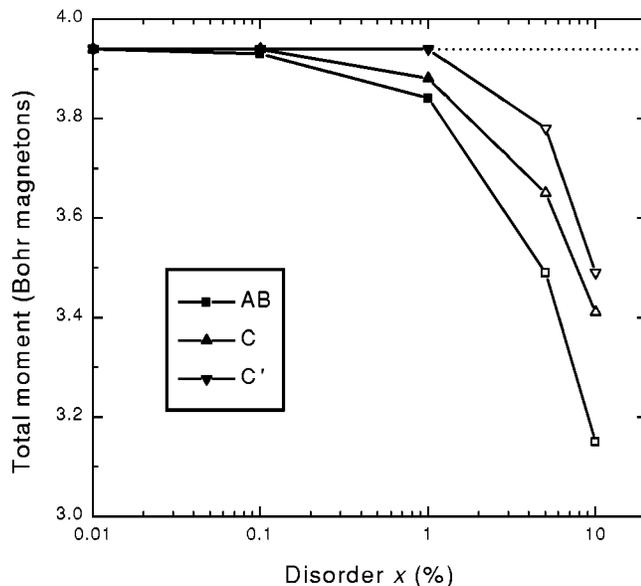


FIG. 1. Calculated total magnetic moment as a function of the disorder level for A-B-type disorder (squares), C-type disorder (up triangles), and C'-type disorder (down triangles). The dotted line shows the moment calculated for ordered NiMnSb. Open symbols indicate a spin polarization below 100%. The lines between the data points are to guide the eyes.

unit cell. All numerical results are based on fully self-consistent calculations performed with a basis set that includes s , p , d , and f partial waves on each site and 75 two-dimensional plane waves. The Brillouin zone sums are performed over 64 special \vec{k}_{\parallel} -points in the Brillouin zone. For the energy integrals we use 16 points on a semicircle in the complex plane.

RESULTS

We find a total magnetic moment of $3.94 \mu_B$ for ordered NiMnSb. The small deviation from the expected (integer) value of $4.00 \mu_B$ is related to approximations made in determining charge densities and moments in the calculations.¹³ For all three types of disorder the total moment decreases monotonically with an increasing level of disorder. This disorder dependence is shown in Fig. 1. Also indicated are cases with minority-spin states at the Fermi energy, i.e., cases with a spin polarization smaller than 100%, as reported in Ref. 8. In these cases, the moment is always smaller than $3.8 \mu_B$ (a reduction by 3.6%), while the half-metallic cases have a moment above this value. For the experimentally observed¹¹ disorder of type C' at a disorder level of 5% the calculated moment is $3.78 \mu_B$ or a reduction by 4.1%.

DISCUSSION AND CONCLUSION

The reduction of the moment is caused by disorder-induced minority-spin states which also cause the reduction of the spin polarization for disorder levels above 1%. If we scale our calculated moments by the accepted moment of $4.00 \mu_B$ for ordered NiMnSb we obtain a moment of $3.86 \mu_B$ below which the spin-polarization is smaller than 100%.

Experimental data for bulk NiMnSb is very well in agreement with a moment of $4.00 \mu_B$ or only slightly higher. Hordequin *et al.*¹⁴ measure 4.03 ± 0.02 and $4.025 \pm 0.02 \mu_B$ in different directions of single-crystal samples and 4.01

$\pm 0.02 \mu_B$ in a polycrystalline sample. Kabani *et al.*³ report a moment of $3.97 \pm 0.08 \mu_B$ for single-crystal material. The same group reports a moment of $3.9 \pm 0.2 \mu_B$ for thin-film samples obtained by coevaporation of Ni, Mn, and Sb. In this case, the samples were annealed at 500°C for 2 h. The apparent reduction of the moment should be more pronounced for NiMnSb thin films used in spin valves and tunnel junctions which require lower annealing temperatures and thus higher levels of disorder can be expected.

In order to better understand the effect of disorder on the half-metallic property of NiMnSb and the implication for spin-dependent devices it will be helpful to obtain precise structural and magnetic measurements on thin-film samples. A precise measurement of the magnetization in thin films for which both the atomic disorder and the spin-polarizations are known would in our opinion lead to an independent check of predicted changes of the electronic structure due to atomic disorder and thus help validate the argument that failure to produce half-metallic NiMnSb films is due to atomic disorder.

In conclusion, we reported on the dependence of the total magnetic moment of NiMnSb on atomic disorder. The moment decreases with increasing disorder. A calculated moment below $3.8 \mu_B$ was seen in those cases with minority-spin states at the Fermi energy. While more data on atomic disorder in NiMnSb thin films is needed it will also be interesting to precisely measure the low-temperature saturation magnetization.

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- In the LKKR-CPA method the charge density is determined from the Green function and the total charge and moments are determined from numerical integration over the atomic sphere. Together with the truncation of the basis sets this may lead, in the present case, to small deviation of the moment from integer values.
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