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**First-principles exchange interactions between ferro and
antiferromagnetic films:
Co on NiMn, a case study.**

T. C. Schulthess and W. H. Butler

Metals and Ceramics division, Oak Ridge National Laboratory

Oak Ridge, TN 37831

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Abstract

Heisenberg exchange parameters at the interface of antiferromagnetic NiMn with ferromagnetic Co are calculated from first-principles. We use a layer version of the Korringa-Kohn-Rostocker multiple-scattering approach and an expression, which is based on the expansion of the band energy, to calculate the exchange parameters from the underlying electronic structure. For bulk systems, the parameter sets yield Curie temperatures that are in good agreement with experimental values. In the interface region, the inter-layer interactions in NiMn change significantly compared to the bulk while the intra-layer interactions are almost unchanged.

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I. INTRODUCTION

Exchange interactions between ferro and antiferromagnetic films are of considerable interest since their understanding is closely related to the effect known as exchange anisotropy (or exchange bias). Even though this effect has been known since the late 1950s¹ and has very interesting technological application, its understanding from a microscopic point of view is not established. A simple model that postulates exchange interactions between the antiferromagnet (AF) and the ferromagnet (FM) was introduced by Mauri *et al.*² and explains the effect by a domain wall which is pushed into the AF when the magnetization in the FM is reversed. Malozenoff³ explained the exchange interaction with a random field that is attributed to the interface roughness. However experiments by Nogues *et al.*⁴ showed that the exchange anisotropy is present with perfectly smooth interfaces and that the roughness actually reduces the bias field. Recently Koon⁵ showed, that a generic Heisenberg model with nearest neighbor interactions may lead to the FM to orienting perpendicular to the easy axis of the AF. This non-trivial result is due to relaxation of the moment directions near the AF-FM interface and leads to bias fields that are of reasonable magnitude. But the generic Heisenberg model introduced by Koon yields exchange anisotropy only when the spins are confined to the plane parallel to the interface⁶. Since there is no immediate physical justification for this confinement more thorough investigations of realistic materials are needed. The major unknowns for any application of a Heisenberg type model to realistic materials are the exchange parameters, and it is the purpose of this work to provide such a set of parameters for a system that may be of significant relevance in technological applications.

In the present work we follow an approach proposed by Liechtenstein *et al.*⁷ in which Heisenberg exchange parameters can be determined directly from first principles electronic structure calculations. The system that we investigate is chosen to give the best compromise between relevance to real applications and the possibilities of state of the art electronic structure methods. Usually Ni-Fe is used as a FM on AF NiMn substrates⁸. In order to

simplify the calculations we replace the random Ni-Fe alloy by a pure FM. Since no FM fits naturally onto NiMn we chose Co because it has magnetic properties which are fairly robust with respect to structural distortions and can thus be viewed as a good representative of a FM material to interface with NiMn.

II. METHOD

Our aim is to map the rather complex spin dependent part of the energy functional of density functional theory within the local density approximation onto a simple Heisenberg Hamiltonian,

$$E = - \sum_{ij, i \neq j} J_{ij} \hat{\mathbf{s}}_i \cdot \hat{\mathbf{s}}_j, \quad (1)$$

which is the sum over all exchange interaction, J_{ij} , between moments of fixed magnitude, located on sites labeled by i and represented by a vector of unit length $\hat{\mathbf{s}}_i$. For the description of the electronic structure, multiple scattering theory (MST) provides a natural starting point, since it expands the Green function in a localized basis around every site. The key operator used in this context is the scattering-path operator (SPO), $[\tau_{ij}(\varepsilon)]_{Ls, L's'}$, from which the one-particle Green function and hence the charge and moment density may be calculated. It gives, at energy ε , the scattered spherical wave of angular momentum $L' = (l', m')$ and spin s' from site j resulting from an incident wave of angular momentum L and spin s at site i . An expression for the exchange parameters in terms of these SPOs, which is based on an expansion of the band energy, was first given for the ferromagnetic case by Liechtenstein *et al.*⁷, and later extended to the case of arbitrary orientation of the atomic moments by Antropov *et al.*⁹ who wrote the pair exchange parameter as.

$$[J_{ij}]_{\alpha\beta} = -\frac{1}{4\pi} \text{Im} \int_{-\infty}^{\varepsilon_F} \text{Tr}_L \left(\Delta_i \tau_{ij}^\alpha \Delta_j \tau_{ji}^\beta - \Delta_i \tau_{ij}^0 \Delta_j \tau_{ji}^0 \right) d\varepsilon. \quad (2)$$

In this expression $\Delta_i = 1/2 \sum_{\nu=1,3} \text{Tr}_s (t_i^{-1} \sigma^\nu)$ and $\tau_{ij}^\nu = 1/2 \text{Tr}_s (\tau_{ij} \sigma^\nu)$ where t_i is the single site scattering matrix, σ^0 is the 2×2 unit matrix, and $\sigma^{\nu=1,3}$ are the three Pauli matrices. In

the present case we will be concerned with collinear ferro and anti-ferromagnetically aligned atomic moments which we take to be parallel to the z-axis¹⁰. Thus the exchange interactions reduce to

$$J_{ij} = -\frac{1}{4\pi} \text{Im} \int_{-\infty}^{\epsilon_F} \text{Tr}_L \left(\tilde{\Delta}_i \tau_{ij}^{\uparrow} \tilde{\Delta}_j \tau_{ji}^{\downarrow} \right) d\epsilon, \quad (3)$$

with $\tilde{\Delta}_i = (t_{i\uparrow}^{-1} - t_{i\downarrow}^{-1}) \text{sgn}(\hat{\mathbf{z}} \cdot \hat{\mathbf{s}}_i)$, and (\uparrow, \downarrow) denote the actual spin channel. There exists an important sum rule^{7,9}, which for collinear spins can be written as,

$$\begin{aligned} J_i &= \sum_{j \neq i} J_{ij} \text{sgn}(\hat{\mathbf{s}}_i \cdot \hat{\mathbf{s}}_j) \\ &= -\frac{1}{4\pi} \text{Im} \int_{-\infty}^{\epsilon_F} \text{Tr}_L \left(\tilde{\Delta}_i \left[\tau_{ii}^{\uparrow} - \tau_{ii}^{\downarrow} \right] + \tilde{\Delta}_i \tau_{ii}^{\uparrow} \tilde{\Delta}_i \tau_{ii}^{\downarrow} \right) d\epsilon. \end{aligned} \quad (4)$$

This last expression will be used to check the convergence of the calculated exchange interactions.

In order to comply with the symmetry requirement in thin films and planar interfaces, we consider the solid as a superposition of 2D-periodic atomic layers. We use the layer version of the Korring-Kohn-Rostocker (LKKR) method¹¹ to calculate the electronic structure and the approach outlined in reference¹² to determine the SPOs that enter the application of equation (3) to the layered geometry. The potentials are treated in the atomic sphere approximation (ASA).

III. ELECTRONIC STRUCTURE AND EXCHANGE INTERACTIONS IN NIMN AND CO

NiMn crystallizes in a face centered tetragonal structure in which Ni and Mn-layers alternate in the (001) direction. Our calculations¹³ are performed at experimental lattice constant for thin films⁸, $a = 3.697 \text{ \AA}$ with a c/a ratio of 0.9573. In accordance with the experimental magnetic structure¹⁴, we find Ni to be non-magnetic and the Mn layers to consist of two square sub-lattices that have opposite magnetic moments. The magnitude of the Mn moment is $3.2 \mu_{Bohr}$, which is slightly smaller than the experimental value of $3.8 \pm 0.3 \mu_{Bohr}$.

The crystal structure of Co is assumed to match that of NiMn, i.e., it is fct with the same basal lattice constant as NiMn but with a c/a ratio chosen to conserve the experimental atomic volume of hexagonal Co. This approximation is imposed by the requirement of the LKKR-method that there has to be an overall 2D-periodicity of the system under consideration. However, the robustness of the magnetic structure of Co with respect to changes in the crystal structure allows us to assume that this structural approximation will not significantly alter the magnetic properties of the Co film and the exchange parameters between the NiMn and Co. The magnetic moment of Co in this artificial fct structure is $1.60\mu_{Bohr}$ which is similar to $1.63\mu_{Bohr}$, the value calculated for hcp Co.

In table I the exchange interactions of bulk Co and NiMn are compiled. The cutoff is chosen in order that the agreement between the sum over calculated J_{ij} and the value determined directly from Eq.(4) is better than one percent. The results clearly indicate that in Co not only the magnitude but also the coupling between the magnetic moments on different sites are robust with respect to the change of the structure. The Curie temperature that corresponds to the set of exchange interactions can be estimated within a mean field consideration from⁷ $T_c = 2/3J_i$ and is found to be 1375K for fcc Co, which is in good agreement with the experimental value of 1388K. Similarly, the Neel temperature of NiMn is estimated to 1187K which is slightly higher than the experimental value of 1070 ± 40 K given by Pál *et al.*¹⁴.

IV. RESULTS FOR THE CO-NIMN INTERFACES

Since not very much is known about the details of the interface between NiMn and a ferromagnetic material, we consider the two possibilities of perfect interfaces, i.e., the cases where Co is next to Ni and Mn, respectively. In the calculations the electronic charge was relaxed self-consistently in 12 atomic layers consisting of 6 Co layers and 3 repeat units of NiMn. On the AF side of the interface the potentials were fixed to the bulk values and on the other side we used potentials that were converged for Co with the Fermi energy fixed to

the value of NiMn. This assures that the electro-chemical potential is the same on both side of the interface. We have explicitly checked the effect of this shift in the Fermi energy on the magnetic moments and the exchange interaction in Co and found that it is negligible. The relative size of the atomic spheres of Ni and Mn was chosen such that in the bulk every site is charge neutral.

Even though the method applied here is not limited to the case of collinear atomic moments, the magnetization in Co is chosen to be aligned with the moments of one of the two Mn sub-lattices. This particular (but arbitrary) choice of the relative magnetization directions provides an important check of the transferability of the exchange parameters. Breaking the symmetry by introducing the NiMn-Co interface with collinear magnetic moments introduces two types of Co-Mn interactions which are equivalent from a geometrical point of view but inequivalent with respect to the relative directions of the moments. The difference between the two gives an estimate of the error introduced when one assumes that the exchange interactions are scalars instead of tensors as actually required by Eq. (2). The results for the exchange interactions of the two types of interfaces are given in Figure (1). Only values that differ from the corresponding bulk values are shown. Whenever the values between the two subsets mentioned previously differ, they are both shown. As can be seen, there is no qualitative difference between the two subsets. Whether the quantitative difference is relevant will have to be determined in an actual micromagnetics calculations which uses these Heisenberg parameters.

While all interactions converge rapidly to their bulk values inside Co, the convergence is much slower on the NiMn side. Despite the effect of the interface in NiMn, the intra-layer interactions in the interface region are qualitatively the same as in the bulk. The effect of the interface is stronger on the inter-layer coupling. The most significant effect is between next nearest out of plane Mn neighbors, where the interactions reverse sign and further enforce the antiferromagnetic alignment of the moments. The values for nearest out of plane neighbors are reduced by about 30% but remain strong. In the case where Co layers border with Mn, the Co-Mn coupling between next nearest neighbors is almost the same as

between corresponding Mn sites and consequently one order of magnitude larger than the equivalent interactions in Co. The interactions between nearest Co-Mn neighbors however roughly correspond to the average value between the interactions in Co and NiMn. For the other interface, where Ni is next to Co, the coupling between Co and Mn is much smaller, but the Ni layer at the interface has a non-vanishing moment of $0.30\mu_{Bohr}$ which leads to finite interactions between Ni and its nearest Co and Mn neighbors.

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REFERENCES

- ¹ W. H. Meiklejohn and C. P. Bean, Phys. Rev. **105**, 904 (1957).
- ² D. Mauri, H. C. Siegmann, P. S. Bagus, and E. Kay, J. Appl. Phys **62**, 3047 (1987).
- ³ A. P. Malozemoff, J. Appl. Phys. **63**, 3874 (1988).
- ⁴ J. Nogues, D. Lederman, T. J. Moran, Ivan K. Schuller, and K. V. Rao, App. Phys. Lett. **68**, 3186 (1996).
- ⁵ N. C. Koon, Phys. Rev. Lett. **78**, 4865 (1997).
- ⁶ T. C. Schulthess and W. H. Butler, unpublished.
- ⁷ A. I. Liechtenstein, M. I. Kastnelson, V. P. Antropov, and V. A. Gubanov, J. Mag. Magn. Mater. **67**, 65 (1987).
- ⁸ Tsann Lin, Daniele Mauri, Norbert Staud, Cherngye Hwang, and Grace L. Gorman, Appl. Phys. Lett. **65**, 1183 (1994).
- ⁹ V. P. Antropov, M. I. Kastnelson, B. N. Harmon, M. van Shilfgaarde, A. I. Liechtenstein, unpublished.
- ¹⁰ Since spin-orbit coupling is not included in the present calculations the spin direction can be chosen independently of the lattice.
- ¹¹ J. M. MacLaren, S. Crampin, D. D. Vvedensky, and J. B. Pendry, Phys. Rev. B **40**, 12164 (1989)
- ¹² T. C. Schulthess, R. Monnier, and S. Crampin, Phys. Rev. B **50**, 18564 (1994).
- ¹³ We have used *s*, *p*, and *d* partial waves, 16 energy points on a semi circular contour in the complex energy plane, 36 special k-points in the 1/8th section of the 2D Brillouin zone and 37 plane waves for the interlayer scattering.
- ¹⁴ L. Pál, E. Krén, G. Kádár, P. Szabó, and T. Tarnóczy, J. Appl. Phys. **39**, 538 (1968).

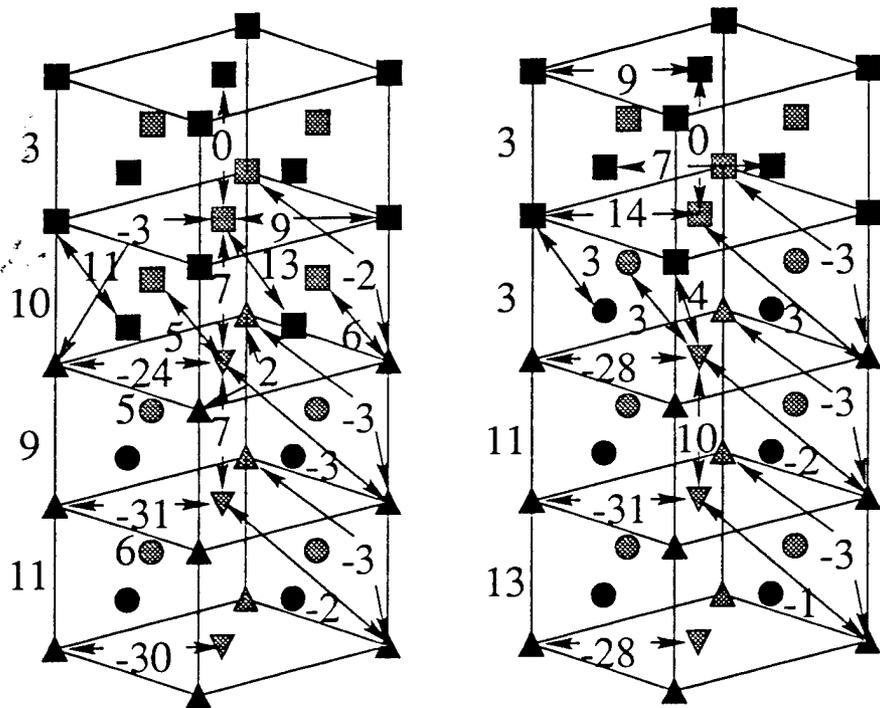
TABLES

TABLE I. Calculated values for J_i (first row) and J_{ij} (remaining rows) in meV for bulk Co and MnNi. Values for Co are given for the fct and the fcc structure.

Co	fct	fcc	Mn	fct
J_i	179.6	177.8	J_i	153.4
(.5,0.,.5)	14.1	12.4	(.5,.5,0.)	-27.1
(.5,.5,0.)	8.1	12.4	(0.,0.,1.)	16.1
(0.,0.,1.)	1.2	0.4	(1.,0.,0.)	6.7
(0.,1.,0.)	0.7	0.4	(.5,.5,1.)	1.8
(.5,.5,1.)	1.2	1.5	(0.,1.,1.)	-2.3
(1.,.5,.5)	2.0	1.5		

FIGURES

FIG. 1. Exchange interaction in meV for Co-MnNi (left panel) and Co-NiMn (right panel). Squares, circles, up and down triangles represent respectively, Co, Ni, up and down Mn.



Schulthess and Butler, Figure 1

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