Exchange Coupling and Exchange Bias in FM/AFM Bilayers for a Fully Compensated AFM Interface

B. Skubic, J. Hellsvik, L. Nordström and O. Eriksson

Department of Physics and Materials Science, Uppsala University

Box 530, SE-75121, Uppsala, Sweden

We address the interlayer coupling in a ferromagnet/antiferromagnet bilayer where the interface of the antiferromagnet is fully compensated. We discuss the role of different types of exchange interaction for the interlayer coupling and exchange bias. We propose two types of corrections to the ideal Heisenberg Hamiltonian which may explain exchange bias. The first is a correction for the angular dependence of the exchange interactions and the second a correction due to magnetostriction and interface imperfections. The first correction contributes to an anisotropy at the interface and favors either parallel or perpendicular coupling across the interface. The second correction contributes to the exchange bias. Our analysis is based on atomic spin dynamics simulations, and our results show that small corrections to the ideal Heisenberg Hamiltonian may have macroscopic consequences in systems with frustrated interatomic interactions.

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1. Introduction

The coupling between ferromagnetic (FM) and antiferromagnetic (AFM) thin films is still not well understood, despite several decades of research [1, 2]. Two common features of this coupling is (1) an exchange bias or a shift of the magnetization curve of the FM layer away from the zero field axis and (2) an increased coercivity or uniaxial anisotropy of the FM layer due to contact with the AFM. These two properties of the FM/AFM coupling are utilized in modern data storage technology for pinning FM layers. One reason for the lack of theoretical understanding of the coupling is the sensitivity to interface morphology and preparation techniques. A full understanding of the coupling requires consideration of the true interface structure including interface mixing, roughness and structural relaxation and how these properties affect the exchange and magnetostatic interactions across the interface. Even though details of the interface structure play an important role for the coupling in FM/AFM systems it is still of interest to examine how different interactions across an ideal interface contribute to the overall coupling.

The scope of this article is to discuss the role of the exchange interaction across the interface. Our starting point is the atomic moment approximation (AMA), a description of the magnetic system in terms of an atomic moment picture, where a rigid atomic moment is associated with each atom [3]. The interatomic exchange interaction, which governs the interaction between the atomic moments, relies on the electronic structure of the system and is often accurately calculated within density functional theory (DFT). The Heisenberg Hamiltonian provides a simple but often accurate model for the in-

teratomic exchange interactions. A DFT ground state structure can be mapped onto a Heisenberg Hamiltonian. The Liechtenstein–Katsnelson–Gubanov method (LKGM) [4], which is the most widely used technique for obtaining exchange interactions for a Heisenberg Hamiltonian, results in exchange parameters valid in the small angle limit. The generalized-perturbation-method (GPM) [5] provides an alternative method of producing parameters strictly valid only for the paramagnetic state. Our discussion in this article is based on the LKGM mapping. We introduce a small deviation to the ideal Heisenberg Hamiltonian which we denote exchange asymmetry and which accounts for a correction to the angular dependence of the exchange parameters. We do not attempt to calculate the magnitude of this correction. Instead, we treat the correction as a parameter and we discuss how it affects the coupling across an interface of a FM/AFM bilayer where the AFM interface is fully compensated. Secondly, we introduce a correction to the exchange parameters due to magnetostrictive effects. Within the Heisenberg model the atomic positions are usually regarded as fixed and the exchange parameters are therefore independent of the magnetostrictive motion of the atoms. Here we do not attempt to calculate the magnetostrictive effect. Instead we identify interface sites where magnetostrictive forces differ. An asymmetry between these different sites is introduced in the exchange parameters. The asymmetry is described by a parameter and we investigate how this asymmetry affects the coupling across the interface of the bilayer.

It has previously been shown that a theoretical model of an ideal FM/AFM structure based on the Heisenberg Hamiltonian results in a strong perpendicular coupling

between the moments of the FM and AFM layers [6, 7]. Nevertheless, experiments sometimes report parallel coupling and in other cases a perpendicular coupling across the interface. We find within our model that even for a very small value of the exchange asymmetry, discussed above, we obtain a rather strong energy lowering mechanism which may favor either parallel or perpendicular coupling across the interface. Hence the exchange asymmetry, or the first order correction of the angular dependence of the exchange parameters, has profound consequences for the coupling in the AFM/FM system and results in a uniaxial anisotropy of the FM layer. Moreover, it has previously been shown that the Heisenberg model does not result in an exchange bias of the FM layer. By introducing a symmetry breaking mechanism between different sublattices of the AFM at the interface Lederman et al. [8] were able to explain an exchange bias. We identify magnetostriction as a possible source for the asymmetry and by including this asymmetry we are able to explain the presence of an exchange bias.

2. Description of model system

As a model system we choose the Fe/NiO (001) system (see Fig. 1). The coupling of Fe on top of NiO has been shown to be collinear in experimental studies [9], i.e. the Fe moments are either parallel or antiparallel to the Ni moments. The reverse system, NiO on top of
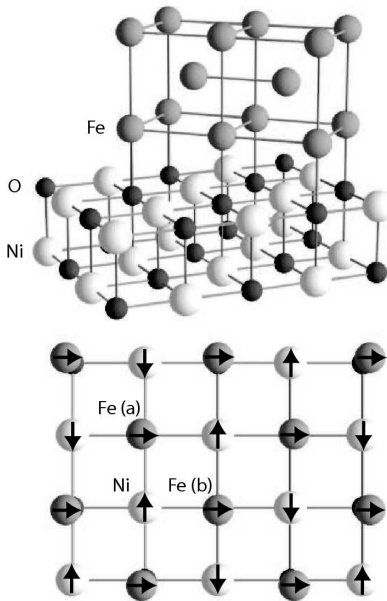

Fig. 1. Structure of the simulated Fe/NiO interface. The top figure shows the crystallographic structure of the simulated system. Black balls represent O atoms, white balls Ni atoms and gray balls Fe atoms. The bottom figure shows a top view of 1 ML Fe on top of NiO. The atomic moments are illustrated by arrows and we have chosen to plot the case of perpendicular coupling across the interface. In the lower part we mark out sites a and b which are inequivalent in model 2 (see Sect. 5).

TABLE

Exchange parameters J_x^i for three models used in the simulations where i is the number of the coordination shell and x is the atomic pairs for which the interaction acts. The index a or b refers to site index (see Fig. 1). Each chemical unit cell of NiO contains 2 Ni sites at the (001) interface. Values are given in mRy. The parameters β and γ are unitless parameters for the exchange asymmetry and magnetostrictive asymmetry respectively, defined in Sects. 4 and 5.

Parameter	Heisenberg model	Model 1	Model 2
J_{FeFe}^1	1.3	1.3	1.3
J_{FeFe}^2	0.8	0.8	0.8
J_{NiNi}^1	0.1	0.1	0.1
J_{NiNi}^2	−1.4	−1.4	−1.4
$J_{\text{FeNi},a}^1(< 90°)$	0.1	0.1	0.1
$J_{\text{FeNi},a}^1(> 90°)$	0.1	0.1β	0.1β
$J_{\text{FeNi},b}^1(< 90°)$	0.1	0.1	0.1γ
$J_{\text{FeNi},b}^1(> 90°)$	0.1	0.1β	$0.1\gamma\beta$

Fe has shown perpendicular coupling [10]. This difference can be a result of different interface structures. NiO forms in the NaCl structure and has the type-II AF order, i.e. with an ordering wave vector along the [111] direction. We consider here a bilayer of 5 ML of bcc Fe on top of 32 ML of NiO. The crystalline and magnetic structure is shown in Fig. 1. Let us note that in this figure the Ni moments point in the y-direction (up or down) and throughout our simulations the y-direction is defined to be parallel to the orientation of the Ni moments. In Fig. 1 the Fe moment is drawn to lie along the x-axis, but in the actual simulations, described below, the Fe moment can point in a general direction. In our model we use the exchange parameters obtained from Ref. [11] for bulk NiO. In lateral dimensions we use a 2×2 repetition of the chemical unit cell in order to fit the type-II AF ordering of NiO. The simulation cell contains 296 atoms and periodic boundary conditions were used in the in-plane directions. For Fe we use typical exchange parameters reported in Ref. [5]. The interaction between Fe and Ni in this system is unknown and we assume only nearest neighbor ferromagnetic interactions of the order 0.1 mRy, a typical strength of nearest neighbor transition metal atoms. Experiments reveal the existence of an FeO layer at the interface [12]. For simplicity we have chosen to neglect the oxide layer in our simulations. The exchange parameters used in the simulations are listed in Table. Furthermore, we have used the atomic spin dynamics (ASD) [13] simulation package to perform relaxation simulations of the magnetic structures. The default damping parameter of $\alpha = 0.1$ is used. First, we reproduce the perpendicular bilayer coupling seen within the ideal Heisenberg model. We then introduce the parameterized corrections to the Heisenberg Hamiltonian and demonstrate how this affects the FM/AFM coupling.

3. Theoretical considerations

Let us first examine the bilayer coupling within the Heisenberg model. Within the Heisenberg model, $H = -\sum_{ij} J_{ij} e_i e_j$, the interaction parameters $J_{ij}(\theta, \phi)$ are independent of the angle between the interacting moments, i.e. $J_{ij}(\theta, \phi) = J_{ij}$. Let us consider now the Fe/NiO system with the Heisenberg parameters given in Table. In Fig. 2 we present a simulation of a relaxation of the bilayer system starting from completely random spin orientations of all the atomic moments. The figure shows the time evolution of the magnetization of the Fe layer. The relaxation pattern of the Fe layer corresponds to the
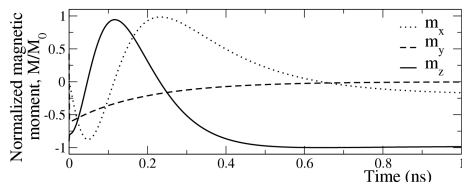
Fig. 2. Simulation of magnetic relaxation starting from a completely random distribution of the orientations of the atomic moments in a Fe/NiO bilayer system. The plots show the evolution of the x, y and z-components of the magnetization of the Fe layer for the Heisenberg model. Initially on a timescale of fs (not shown) there is a quick relaxation to FM order of the Fe layer. On a longer timescale (shown here) the orientation of the magnetization of the FM layer relaxes in the presence of the exchange of the AFM layer. The relaxation pattern is equivalent to an easy-plane anisotropy resulting in perpendicular coupling.

relaxation pattern of a magnetization in an easy-plane anisotropy where the easy plane is perpendicular to the magnetization axis of the NiO layer. Figure 2 shows that the y-component of the Fe-magnetization is zero, which with our definition of the Ni moments always lying along the y-direction means that Fe and Ni moments couple perpendicularly. Hence the simulations show that the Heisenberg model results in a perpendicular coupling between the FM and the AFM layers, reproducing the results from Ref. [7]. Here we have neglected magnetostatic interactions which result in a strong shape anisotropy which would lead to an in-plane confinement of the magnetization.

The mechanism behind the perpendicular coupling can be understood in terms of a simple model of the system. First, we assume that all the NiO moments are fixed in their bulk orientations and that all of the Fe moments are parallel with each other. The only degree of freedom within the system is the relative orientation of the moments of the FM (Fe) layer and the moments of the AFM (NiO) layer. With the assumption that the Ni atoms stay in a perfect antiferromagnetic coupling there is no energetically preferred direction for the magnetization of the FM layer. To illustrate this fact, let us note that if the FM layer is perpendicular to the orientation of the magnetization of the AFM layer, all the Fe atoms and all

the Ni moments are perpendicular, resulting in a zero energy contribution to the coupling between the AFM and FM layers from the Heisenberg term. For a parallel arrangement the Heisenberg term gives alternatively positive and negative terms depending on if the Fe–Ni coupling is parallel or antiparallel. Hence, the sum of all the terms in the Heisenberg model is zero in this case.

With the assumption of rigid Ni moments we have neglected magnetic relaxation at the interface, i.e. the possibility of some atoms to deviate from collinear coupling. Let us now go beyond this assumption and allow for relaxation of the interface moments. For the case of parallel alignment between the FM and AFM layer there is no relaxation path within the Heisenberg model towards lower energy of the parallel alignment. For perpendicular alignment however, the interface Ni moments may relax towards the direction of the Fe layer resulting in a lowering of the energy. Hence interface relaxation of the AFM (Ni) moments results in a favoring of perpendicular coupling. The total energy of the system has an energy minimum at a slight tilt angle of the Ni moments towards the Fe moments. By taking interface relaxation into account the Heisenberg model produces a negative energy contribution which explains the perpendicular coupling across an FM/AFM interface with fully compensated AFM moments. If we assume that interface relaxation occurs only at the interface layer of NiO, the coupling energy is given by a simple expression

$$E(\theta) = 2J_{\text{NiO},2}[\cos(\theta) - 1]$$

$$+ 4J_{\text{NiO},2}[\cos(2\theta) - 1] - 8J_{\text{FeNi}}\sin(\theta), \qquad (1)$$

where θ is the angle with which the Ni moments deviate from antiferromagnetic order. The coupling curve for different magnitudes of the exchange coupling is shown in Fig. 3. It may be seen that for increasing strength of the Fe–Ni exchange coupling, the angle between Ni moments deviate from aniferromagnetic coupling to a larger and larger degree. In reality, the relaxation process at the interface is more complex involving more layers than just the interface layer of NiO and the relaxation is also af-
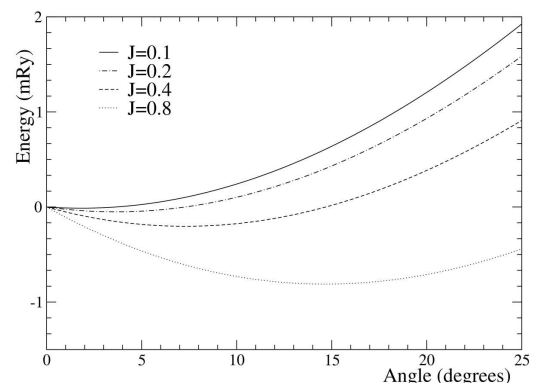
Fig. 3. The energy lowering by the tilting mechanism for different values of the Fe–Ni exchange parameter, J, within the Heisenberg model.

fected by the magneto-crystalline anisotropy in both NiO and Fe which tends to counteract the tilting mechanism.

4. A model beyond the Heisenberg: asymmetric exchange — model 1

The validity of using a Heisenberg model for describing an FM/AFM interface can be questioned. For many systems the validity of the Heisenberg model is limited to small angle rotations from the ground state. At an interface between an FM and an AFM material, frustration occurs to such a degree that we might expect large deviations from the Heisenberg behavior. As a first order correction we include a biquadratic term in the exchange interaction

$$J(\theta, \phi) = J_{\mathrm{FM}} + (J_{\mathrm{AFM}} - J_{\mathrm{FM}})[1 - \cos(\theta)]/2, \qquad (2)$$

where J_{FM} is the coupling for a ferromagnetic alignment and J_{AFM} is the coupling for an antiferromagnetic alignment of the magnetic moments. The fact that these two parameters in general differ is seen in electronic structure calculations [14] where both the FM and AFM structure of a material (e.g. Fe) can be stabilized and exchange parameters for both structures can be extracted. In the actual simulations presented below we use an approximation of this expression by utilizing $J(\theta, \phi) = J_{\mathrm{FM}}$ for $\theta < 90°$ and $J(\theta, \phi) = J_{\mathrm{AFM}}$ for $\theta > 90°$. Instead of calculating J_{AFM} we define the exchange asymmetry $\beta = J_{\mathrm{AFM}}/J_{\mathrm{FM}}$ and we study the coupling with respect to β.

With this small correction of the asymmetry between ferromagnetic and antiferromagnetic atomic couplings across the interface, we find an energy lowering mechanism which, depending on β, may favor parallel or perpendicular coupling between the AFM and FM thin films. With this form of the exchange interaction the Heisenberg term will no longer vanish for parallel coupling. Instead, based on this mechanism one should expect either a large preference or penalty for parallel coupling between the interfaces, depending on whether $J_{\mathrm{AFM}} > J_{\mathrm{FM}}$ or $J_{\mathrm{AFM}} < J_{\mathrm{FM}}$, respectively. The coupling energy per atom due to this mechanism is given by $J_{\mathrm{AFM}} - J_{\mathrm{FM}}$. In Fig. 4 we present relaxation simulations starting from a completely random spin configuration, for values of $\beta = 0.9$ and $\beta = 1.1$. For $\beta = 0.9$ we see a relaxation pattern similar to the relaxation pattern of a magnetization in an easy-axis anisotropy. The y-component of the Fe magnetization is saturated, showing a parallel coupling between the Fe and Ni moments. For $\beta = 1.1$ we see a pattern similar to that in Fig. 4, which indicates an easy-plane anisotropy. Here, the y-component of the Fe moment becomes zero indicating a perpendicular coupling between Fe and Ni moments. Hence the exchange asymmetry results in a distinct uniaxial anisotropy and the size determines whether a parallel or perpendicular coupling is favored.

Exchange asymmetry should appear in most systems since most systems are poorly described by the Heisenberg model for large angle rotations between the
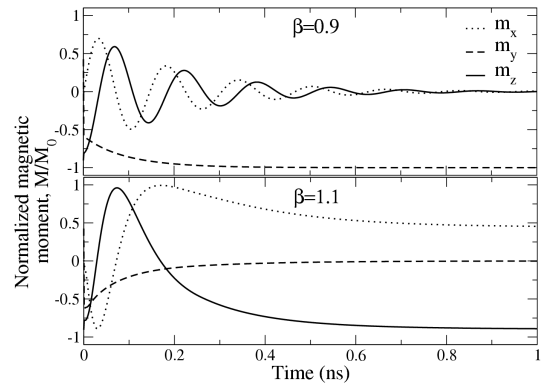

Fig. 4. Simulation of magnetic relaxation starting from a completely random distribution of the orientations of the atomic moments in a Fe/NiO bilayer system. The upper part shows the evolution of the top Fe monolayer for model 1 with $\beta = 0.9$ and the lower part shows the evolution of model 1 with $\beta = 1.1$. The relaxation pattern of the upper part resembles relaxation in an easy-axis anisotropy resulting in parallel coupling. The relaxation pattern of the lower part resembles relaxation in an easy-plane anisotropy resulting in perpendicular coupling.
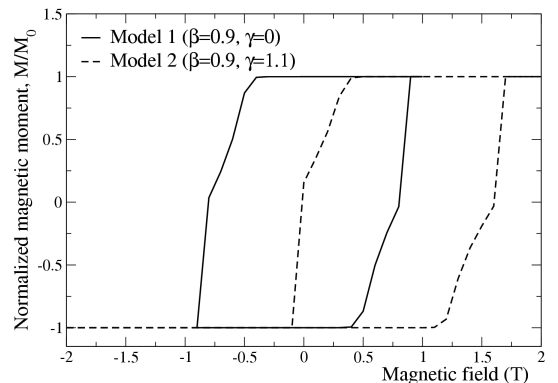

Fig. 5. Magnetization loops of the Fe layer for model 1 and model 2. Model 1 results in a coercivity. Model 2 in addition results in an exchange bias. With current exchange parameters and values for β and γ a rather large uniaxial anisotropy applied to the NiO layer was necessary in order to avoid switching from occurring far away from the interface within the AFM with a rearrangement of the magnetic structure during the switching process.

atomic moments. The final exchange contribution to the coupling should depend on a competition between the strength of the exchange asymmetry and the tilting mechanism discussed in Sect. 3 which favors perpendicular coupling.

5. A model beyond Heisenberg: magnetostriction — model 2

Magnetostriction, i.e. a force induced by a change of the magnetic order of a system may lead to an altering of

the lattice structure at the interface. At the bilayer interface there are two types of Fe sites (see Fig. 1). Each Fe has four nearest neighbor Ni atoms and one second nearest neighbor Ni atom (not shown in Fig. 1). In the case of parallel coupling, which we will focus on, two nearest neighbor couplings are FM and two are AFM. The second nearest neighbor coupling is either FM or AFM depending on the Fe site. The two interface Fe sites differ in the way in which the nearest neighbor FM and AFM couplings are arranged and in the type of the second nearest neighbor coupling. If a magnetostriction effect was present the two Fe sites would be submitted to differently directed forces favoring a slight buckling of the interface. The nearest neighbor interactions would lead to an in-plane buckling of the Fe layer and the second nearest neighbor interactions would lead to an out-of-plane buckling. This buckling would in turn affect the exchange interactions for the two sites differently. Hence the asymmetry which is manifested in the magnetization has due to magnetostriction led to a manifestation in the structural properties at the interface. As we will show, this effect may lead to an exchange bias. An essential requirement is that the structural deformation remains also during a magnetization reversal of the FM layer. Whether such a permanent structural deformation can take place spontaneously or if it can take place in a quench in an external field is an open question.

A buckling of the FeO interface layer at the interface in the Fe/NiO system has been shown in an *ab initio* calculation [12]. This is still different from what is required to produce an exchange bias. We now extend model 1 to also account for the possibility of a site dependent coupling. This is modeled in something which we refer to as model 2, where we introduce site dependent (a, b) exchange parameters, where atomic sites a and b refer to Fe–Ni pairs that are buckled into or away from each other, respectively. We define a buckling parameter by $\gamma = J_{\mathrm{AFM},a}/J_{\mathrm{AFM},b} = J_{\mathrm{FM},a}/J_{\mathrm{FM},b}$. In Fig. 5 we present ASD simulations of a field magnetization loop for a system with $\beta = 0.9$ and $\gamma = 1.1$, which is clearly seen to exhibit an exchange bias. The loop is compared to a simulation with $\beta = 0.9$ and $\gamma = 0$. For just a small asymmetry between the sites we find a strong exchange bias, which on a qualitative level is in accordance with observations.

6. Conclusions

In conclusion, we have shown that small corrections to the ideal Heisenberg model has profound consequences for the exchange coupling across an FM/AFM interface, for systems where the AFM interface is compensated. The reason for this is the large degree of frustration that occurs at the interface of this type of system. We show that corrections for the angular dependence of the exchange interaction gives rise to a uniaxial anisotropy of the FM layer. Moreover, we propose a buckling/magnetostrictive mechanism where interface frustration results in a structural rearrangement of the interface which contributes to the exchange bias. Our study stresses the importance of accurate *ab initio* spin-dynamic methods in combination with first principles calculations for understanding systems with a high degree of internal frustration and complexity.

Acknowledgments

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