Exchange Bias Driven by Dzyaloshinskii-Moriya Interactions

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The exchange bias effect in a compensated $IrMn_3/Co(111)$ system is studied using multiscale modeling from *ab initio* to atomistic spin model calculations. We evaluate numerically the out-of-plane hysteresis loops of the bilayer for different thicknesses of the ferromagnetic layer. The results show the existence of a perpendicular exchange bias and an enhancement of the coercivity of the system. To identify the origin of the exchange bias, we analyze the hysteresis loops of a selected bilayer by tuning the different contributions to the exchange interaction across the interface. Our results indicate that the exchange bias is primarily induced by Dzyaloshinskii-Moriya interactions, while the coercivity is increased mainly due to a spin-flop mechanism.

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In magnetic heterostructures where a ferromagnet (FM) is in contact with an antiferromagnet (AFM), the exchange interaction between the FM and the AFM may induce a unidirectional anisotropy, which is reflected in the hysteresis loops by a shift along the magnetic field axis. This effect is called exchange bias [1]. Most of the theories which have been developed to explain the exchange bias (EB) assume uncompensated spins at the interface of the antiferromagnet to pin the ferromagnet and, therefore, fail to explain the origin of the EB in a system with a compensated interface [2]. To cure this problem, EB models based on a domain state in diluted AFMs due to an imbalance of the number of impurities [3,4], spin-flop coupling [5], biquadratic exchange interaction [6], formation of domain walls [7], or anisotropic exchange interactions across the interface [8] were developed. Recently, based on symmetry properties, Dzyaloshinskii-Moriya (DM) interactions have been proposed as possible mechanisms responsible to EB in compensated systems [9,10].

The $L1_2$-type $IrMn_3$ is a triangular AFM with a noncollinear spin ground state, called a $T1$ Néel state. It exhibits a large second-order magnetic anisotropy due to anisotropic exchange interactions. This high effective anisotropy entails an easy plane (111), to which the ground state is confined [11]. When $IrMn_3$ is capped by fcc Co, the magnetic properties of both the AFM and FM are modified close to the interface [12]. In particular, sizable DM interactions arise between the Co and Mn atoms, owing to the breaking of inversion symmetry at the interface. The (111) interface is perfectly compensated, with an equal number of atoms belonging to the three magnetic sublattices of the AFM.

In this Letter, we focus on addressing the origin of the EB in compensated $IrMn_3/Co(111)$ bilayers by performing numerical calculations of the hysteresis loops and identifying the roles played by different types of exchange interactions between Mn and Co atoms. We find a strong perpendicular EB effect. The main mechanism responsible for the perpendicular EB is the DM interaction, nevertheless, with other minor contributions due to the anisotropy in the exchange interactions through the interface.

This Letter is organized as follows. First, we introduce a spin model which is based on *ab initio* calculations. Then, we analyze the DM interactions across the interface and formulate a model for the exchange bias field. In the next section, we present spin-dynamics simulations and compare them with our theoretical model. We finish with a discussion of the FM switching process and the origin of the EB.

We study the magnetic properties of our system in the spirit of a hierarchical multiscale model linking *ab initio* calculations with dynamical spin model simulations. In terms of the fully relativistic screened Korringa-Kohn-Rostoker method [13,14], we perform self-consistent calculations of an $IrMn_3/Co(111)$ bilayer. Based on a spin-cluster expansion technique [12], we define a classical Hamiltonian, which will be used later in our spin-dynamics simulations, and derive the exchange interactions in the system. The Hamiltonian is a generalized Heisenberg model

$$\mathcal{H} = -\frac{1}{2}\sum_{i,j}\vec{s}_i \boldsymbol{J}_{ij}\vec{s}_j - \sum_i \vec{s}_i \boldsymbol{K}_i \vec{s}_i - \sum_i \mu_i \vec{H}_A \vec{s}_i, \quad (1)$$

where $\vec{s}_i$ represent classical spins, i.e., unit vectors along the direction of each magnetic moment at sites i, occupied by either cobalt or manganese atoms. The first term stands for the exchange contribution to the energy, with $\boldsymbol{J}_{ij}$ denoting the tensorial exchange interaction. The second term comprises the on-site anisotropy and the magnetostatic energy, and $\boldsymbol{K}_i$ is called the anisotropy matrix. In the presence of an external magnetic field $\vec{H}_A$, the last term adds a Zeeman contribution to the Hamiltonian, where μ_i is the magnetic moment of the atom i.

The exchange interactions can further be decomposed into three terms $\boldsymbol{J}_{ij} = J_{ij}^{\mathrm{iso}}\boldsymbol{I} + \boldsymbol{J}_{ij}^{S} + \boldsymbol{J}_{ij}^{A}$ [15], with $J_{ij}^{\mathrm{iso}} = (1/3)\mathrm{Tr}[\boldsymbol{J}_{ij}]$ the isotropic exchange interaction, $\boldsymbol{J}_{ij}^{S} = (1/2)(\boldsymbol{J}_{ij} + \boldsymbol{J}_{ij}^{T}) - J_{ij}^{\mathrm{iso}}\boldsymbol{I}$ the traceless symmetric (anisotropic) part, and $\boldsymbol{J}_{ij}^{A} = (1/2)(\boldsymbol{J}_{ij} - \boldsymbol{J}_{ij}^{T})$ the antisymmetric part of the exchange tensor. The latter one is clearly related to the DM interaction $\vec{s}_i \boldsymbol{J}_{ij}^{A} \vec{s}_j = \vec{D}_{ij} \cdot (\vec{s}_i \times \vec{s}_j)$, with $\vec{D}_{ij}$ as the DM vector.

The DM interaction arises due to the spin-orbit coupling and favors a perpendicular alignment of the spins [16,17]. In a metallic system, the DM interaction can be understood as an anisotropic exchange (or RKKY) interaction [18]. It vanishes if the system is centrosymmetric, but for solids with complex lattices or at interfaces and surfaces, where the inversion symmetry is broken, the DM interaction might play an important role [19].

The stacking model of the bilayer is depicted in Fig. 1(a). For simplicity, we assume that the fcc Co and the $L1_2$ $IrMn_3$ lattices match perfectly without structural relaxation. We used the 2D lattice parameter of $IrMn_3$, $a = 0.3785$ nm. Because of symmetry, the three Mn sublattices in $L1_2$ $IrMn_3$ present uniaxial on-site anisotropy with orthogonal easy axes but an identical anisotropy constant $K_{\mathrm{Mn}} = 0.54$ meV [11]. The fcc Co has an on-site anisotropy smaller than 1 μeV; this contribution will be neglected in the following.

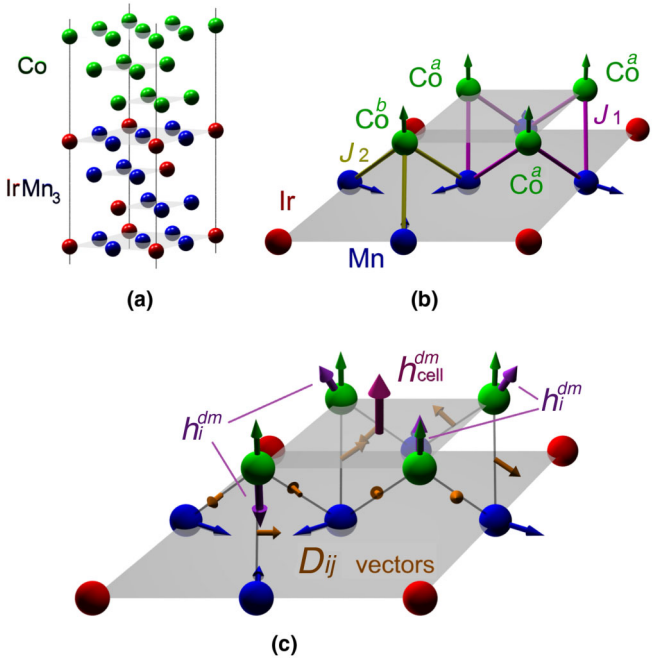

FIG. 1 (color online). (a) Stacking order for the $L1_2$ $IrMn_3$ fcc Co bilayer near the interface. Red, blue, and green spheres represent Ir, Mn, and Co atoms. (b) Sketch of the magnetic order at the (111) interface. The Mn moments (blue arrows) display a perfect $T1$ state. J_1 and J_2 label the two kinds of isotropic exchange interactions between Co-Mn NNs. (c) DM vectors $\vec{D}_{ij}$ between Co-Mn nearest neighbors (orange arrows), a DM interface field $\vec{h}_i^{dm}$, and a DM field acting on the Co atoms per unit cell $\vec{h}_{\mathrm{cell}}^{dm}$ are displayed by violet and magenta arrows.

EB is intimately related to the exchange interaction across the interface between the FM and the AFM. Unfortunately, it is in practice impossible to explore these interactions experimentally. First-principles calculations present the only tool to determine interface interactions between magnetic atoms.

In Fig. 1(b), the 2D unit cell is sketched at the interface, comprising four Co atoms in the upper plane, while there are three Mn and one Ir atoms in the layer below. We can distinguish two kinds of Co atoms at the interface with either two or three nearest neighbor (NN) Mn atoms Co^a and Co^b, respectively. The Co^a atoms and their Mn NNs interact ferromagnetically with an isotropic exchange constant $J_1 = 1.24$ meV, while the Co^b atoms interact antiferromagnetically with their Mn NNs with exchange constant $J_2 = -6.37$ meV. Considering even further interactions, the effective isotropic exchange across the interface has an antiferromagnetic character.

From Eq. (1), the contribution of the DM interactions to the interfacial energy can be expressed as

$$\mathcal{H}_{\mathrm{int}}^{dm} = -\sum_{i,j} \vec{s}_i \cdot (\vec{s}_j \times \vec{D}_{ij}) = -\sum_{i} \vec{s}_i \cdot \vec{h}_i^{dm}, \qquad (2)$$

where now the index i labels only Co atoms and j labels their Mn nearest neighbors. Correspondingly, $\vec{h}_i^{dm}$ is the DM field experienced by a Co spin $\vec{s}_i$ due to its Mn neighbors. As shown in Fig. 1(c), the DM vectors between Co-Mn NNs lie practically in the (111) plane, with the magnitudes $|\vec{D}_{ij}| = 0.58(0.42)$ meV for $\mathrm{Co}^{a(b)}$-Mn nearest neighbors. The DM interactions between Co-Co and Mn-Mn pairs at the interface are non-negligible but less relevant to EB [20].

To simplify the discussion of the EB effect, let us consider only NN interactions and suppose that the Co spins are in a ferromagnetic state while the Mn atoms form a perfect $T1$ state. Then, the DM interactions across the interface induce an effective magnetic field acting on the Co atoms per unit cell $|\vec{h}_{\mathrm{cell}}^{dm}| = |\sum_{i=1}^{4} \vec{h}_i^{dm}| \approx 1.05$ meV that points normal to the interface. Concomitantly, the DM interactions across the interface favor a perpendicular alignment of the Co and Mn moments. The direction of $\vec{h}_{\mathrm{cell}}^{dm}$, pointing either towards the Co or the $IrMn_3$ part of the interface, depends on the chirality of the $T1$ state. Supposing no distortion of the $T1$ state during the FM magnetization reversal, an exchange bias arises due to the DM interactions with the EB field

$$H_{\mathrm{EB}}^{dm} = (|\vec{h}_{\mathrm{cell}}^{dm}| \cos\theta)/(4\mu_{\mathrm{Co}} t_{\mathrm{Co}}), \qquad (3)$$

where θ is the angle of FM magnetization with respect to the effective DM field, and t_{Co} is the thickness of the Co layer.

The DM interactions can easily be modified by a mismatch or relaxation of the lattices. In order to check this, we also studied the case of an $IrMn_3/Co$ bilayer with a relaxation of 17% of the Co lattice parameter normal to the interface. In this case, the magnitude and the direction of the DM vectors are indeed modified. However, $\vec{h}^{dm}_{\text{cell}}$ still points normal to the interface and its magnitude is reduced by only 15% with respect to the unrelaxed case.

In the spin-dynamics simulations, the antiferromagnet was modeled by three intercalated Mn sublattices, forming in total $20 \times 20 \times 6$ unit cells, and the ferromagnet by $20 \times 20 \times t_{\text{Co}}$ unit cells, t_{Co} denoting the number of Co atomic monolayers (ML), in the following labeled $[IrMn_3]_6/[Co]t_{\text{Co}}$. As an insignificant simplification, we supposed the Ir atoms were nonmagnetic, and the magnetic moments of Mn and Co atoms were taken uniformly $\mu_{\text{Mn}} = 2.2\mu_B$ and $\mu_{\text{Co}} = 1.6\mu_B$. We considered magnetostatic interaction in the FM layer approximated by a uniaxial shape anisotropy $K_{\text{Co}} = -0.084$ meV.

To study the possible existence of EB, we evaluate numerically the out-of-plane hysteresis loops of several $[IrMn_3]_6/[Co]t_{\text{Co}}$ bilayers. The hysteresis loops are calculated as a succession of equilibrium states determined by solving the stochastic Landau-Lifshitz-Gilbert equation in the context of the generalized Heisenberg model described in Eq. (1), where the exchange interactions are considered up to sixth NNs; see Refs. [21,22] for more details.

Prior to calculating the hysteresis loops, we prepared the system similarly to experiments by simulating a field-cooling (FC) process. The FC process starts from a random spin configuration in the AFM part, at an initial temperature above the Néel temperature of the AFM and below the Curie temperature of the FM, and proceeds to a final temperature $T_f = 0$ K under the influence of an external applied (cooling) field $H_{\text{cf}} = 1.5$ T. After the FC process, the magnetic moments in the FM are oriented along the direction of the cooling field and perpendicular to the AFM easy plane. The AFM presents a quasi-$T1$ state, slightly distorted at the interface due to the effective antiferromagnetic interaction between the Co and Mn atoms. This distortion gives rise to a small net magnetization in the AFM that is antiparallel to the FM magnetization. Therefore, the spin configuration after the simulated FC process is similar to a *spin-flop* state. This state is quite robust against variation of the speed of the cooling process.

Using this spin configuration as the initial magnetic state, the simulated hysteresis loops display a quasisquare shape, a negative exchange bias (H_{EB}), and a high coercivity. We present the values of the coercive (H_C) and EB fields, determined from the hysteresis loops, as a function of t_{Co} in Figs. 2(a) and 2(b), respectively. The coercivity shows a $1/t_{\text{Co}}$ dependence. This result clearly indicates that the large H_C can be attributed to the interface. As expected, the EB field decreases as the Co thickness increases, but the fit to the theoretical expression [Eq. (3)] is not perfect for small

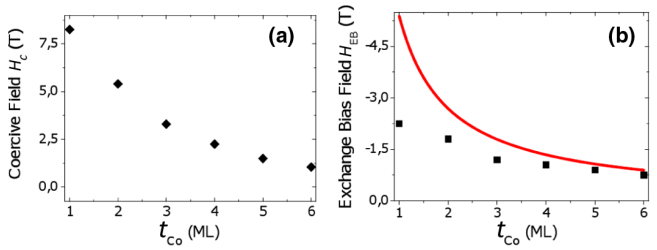

FIG. 2 (color online). Dependence of (a) the coercive field and (b) the EB field on the thickness of Co capping t_{Co}. Solid symbols represent the numerical results, and the line corresponds to Eq. (3).

thicknesses. However, this deviation from the theoretical $1/t_{\text{Co}}$ dependence can be understood by noting that the influence of the interface is not restricted to the adjacent FM and AFM atomic layers, but it extends to at least two or three atomic layers from the interface on both sides.

Based on the spin-dynamics simulations, we can affirm that the FM magnetization switches through a quasicoherent rotation, and during the FM switching, the AFM also switches between two spin-flop states. As it was shown by Schulthess and Butler in Ref. [23], if only isotropic exchange interactions are considered, this kind of magnetization inversion process leads to a uniaxial rather than unidirectional anisotropy. Therefore, the high value of the coercive field might be explained with a spin-flop mechanism, but it is not related to the exchange bias effect.

Inspecting the effect of the DM interactions during the magnetization reversal, we note that the out-of-plane component of the interfacial DM field only depends on the in-plane components of the AFM magnetization which practically remain unchanged during the switching process. This implies that in the descending branch of the hysteresis loop, $\vec{h}^{dm}_{\text{cell}}$ opposes the switching of the FM, while in the ascending branch, it favors the inversion of the FM magnetization. This is the simple picture of how the interfacial DM field generates the perpendicular exchange bias in the $IrMn_3/Co$ bilayer.

To corroborate our hypothesis for the origin of the EB, we investigated the changes in the hysteresis loop of the $[IrMn_3]_6/[Co]_1$ bilayer when artificially switching on or off the different contributions to the exchange interactions across the interface. In all cases, the initial magnetic state of the simulation was the same, namely, the spin configuration obtained after a FC process with all the interactions between FM and AFM layers switched on.

If all parts of the exchange interactions across the interface are removed, the hysteresis loop is perfectly square shaped and, as expected, does not display any EB; see Fig. 3(a). In this case, the coercive field ($H_C = 2.65$ T) is larger than expected considering only the shape anisotropy K_{Co}. The shape of the hysteresis loop indicates a perpendicular anisotropy. These two effects may be explained by

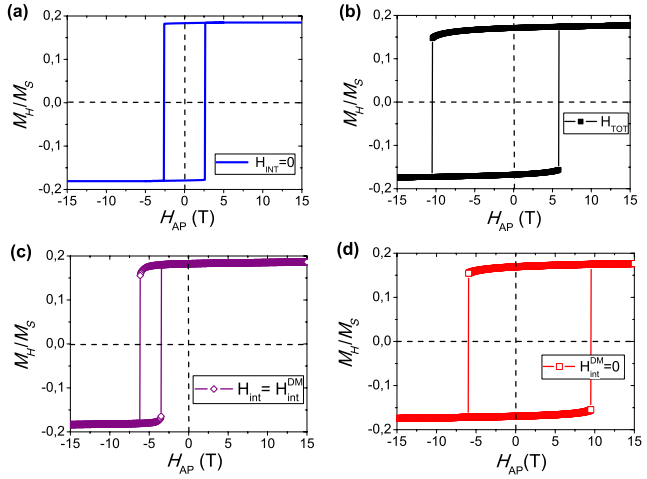

FIG. 3 (color online). Out-of-plane hysteresis loops of $[IrMn_3]_6/[Co]_1$: (a) Removing all the interactions across the interface ($\mathcal{H}_{\rm int} = 0$). (b) Considering all the interactions across the interface. (c) Considering only the DM interactions between the Co and Mn atoms ($\mathcal{H}_{\rm int} = \mathcal{H}_{\rm int}^{dm}$). (d) Removing only the DM interaction between the Co and Mn atoms ($\mathcal{H}_{\rm int}^{dm} = 0$).

an increase of the symmetric anisotropic part of the Co-Co exchange interactions close to the interface [12].

Figure 3(b) shows the hysteresis loop when all the interactions across the interface are considered. Apparently, the system exhibits a highly enhanced coercivity ($H_C = 8.2$ T) and high negative exchange bias ($H_{\rm EB} = -2.3$ T). In comparison to the previous case, it is obvious that both features are related to the exchange interactions across the FM-AFM interface.

Considering only the asymmetric DM contributions to the exchange interactions across the interface, the hysteresis loop shows a drastic reduction of the coercivity and an increase of the EB with respect to the case when all the interactions are included; see Fig. 3(c). Since in this case the isotropic exchange interactions between the Co-Mn pairs are switched off, there is no distortion of the AFM $T1$ state near the interface. Consequently, the net AFM magnetization becomes zero near the interface, thus preventing the increment of the coercivity.

It is interesting to compare the value of the exchange bias obtained in the simulation $H_{\rm EB} = -4.07$ T, with the corresponding value given by Eq. (3) $H_{\rm EB}^{dm} \approx -5.38$ T. These values are of the same order and sign, but the theoretical value is, however, considerably larger than $H_{\rm EB}$. This difference may be a consequence of neglecting the DM interactions between Co-Co and Mn-Mn neighbors in the EB model.

On the other hand, if only the DM interactions between the Mn-Co pairs are removed, the coercivity of the system is almost not affected ($H_C \approx 7.8$ T). Nevertheless, a strong reduction in the magnitude of the EB is observed and even its sign is changed ($H_{\rm EB} \approx 1.8$ T); see Fig. 3(d). This suggests that beyond the DM interactions between the Co-Mn neighbors, there are other, less significant, sources of the EB. In brief, the DM interactions between the Co-Co and Mn-Mn pairs make the distortion of the two spin-flop states asymmetric. Since in the reversed spin-flop state the net magnetization of the AFM layer is somewhat enlarged, the effective antiferromagnetic exchange coupling between the Co and Mn atoms induces a positive EB in the hysteresis loop.

In comparison to experimental values of perpendicular EB fields (e.g., in IrMn/[Co/Pt] [24,25] and IrMn/Co/[Co/Pt] [26] multilayers), our results are 20–100 times higher. Note that these experimental data are evaluated at room temperature, while ours are calculated at 0 K. Although we expect that the exchange integrals as determined by our first-principles calculations are only weakly temperature dependent [27,28], the EB fields will be reduced by thermal spin fluctuations, as it was shown in Ref. [3]. Also, it is possible that Mn diffusion deteriorates the first Co atomic layer [25], and the chemical disorder decreases the DM interactions between Mn and Co atoms at the interface, leading to a reduction of the EB. On the other hand, values have been reported for the in-plane EB field in Mn-Ir/$Co_{100-x}Fe_x$ bilayers [29,30] of the same order of magnitude as our prediction for a Co thickness of 4 nm.

In the light of our results in terms of combined *ab initio* and spin-dynamics simulations, we conclude that the principal source of perpendicular EB in $IrMn_3$/Co(111) is the DM interactions across the interface that favor a perpendicular orientation between the Mn and Co moments and a unidirectional anisotropy perpendicular to the interface. The high coercivity obtained from our simulations is due to a combination of at least two factors, an enhanced Co-Co two-site anisotropy (the symmetric anisotropic part of the exchange) close to the interface and, more importantly, the isotropic exchange between Mn-Co neighbors which results in a distortion of the $T1$ state close to the interface, which leads to a net magnetization in the AFM interface layer. During the switching of the FM, the AFM also switches between two spin-flop states, thus resulting in a considerable enhancement of the coercivity.

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