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Theory of the Interfacial Dzyaloshinskii-Moriya Interaction in Rashba Antiferromagnets

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(Dated: May 14, 2018)

In antiferromagnetic (AFM) thin films, broken inversion symmetry or coupling to adjacent heavy metals can induce Dzyaloshinskii-Moriya (DM) interactions. Knowledge of the DM parameters is essential for understanding and designing exotic spin structures, such as hedgehog Skyrmions and chiral Néel walls, which are attractive for use in novel information storage technologies. We introduce a framework for computing the DM interaction in two-dimensional Rashba antiferromagnets. Unlike in Rashba ferromagnets, the DM interaction is not suppressed even at low temperatures. The material parameters control both the strength and sign of the interfacial DM interaction. Our results suggest a route toward controlling the DM interaction in AFM materials by means of doping and electric fields.

Relativistic spin-orbit coupling (SOC) is the foundation of spin orbittronics, a rapidly developing branch of spintronics [1, 2]. Anisotropic magnetoresistance [3] and the anomalous Hall effect [4] are established SOC transport phenomena. More recent discoveries include the spin Hall and inverse spin Hall effects [5], topological surface states [6, 7], spin-orbit torques [1, 8–11], and chiral domain walls and Skyrmions [12–18]. These phenomena are essential to enable novel ultrafast, nonvolatile, nanoscale spin-based storage and computation devices.

The Dzyaloshinskii-Moriya (DM) interaction between localized spins in a magnetic material is induced by SOC [19–21]. The DM interaction is important for stabilizing noncollinear magnetic structures in ferromagnets. The original proposal by Dzyaloshinskii and Moriya, however, concerned antiferromagnetic (AFM) systems with weak magnetization, i.e., weak ferromagnets; this weak magnetism can be explained in terms of antisymmetric exchange interaction, which is also referred to as the DM interaction.

The energy associated with the DM interaction between two neighboring spins in a lattice, $S_1$ and $S_2$, can be written in the form of the mixed product $H_{DM} = -D_{12} \cdot S_1 \times S_2$, where $D_{12}$ is called the DM vector. The DM interaction, therefore, favors a perpendicular orientation of neighboring spins. By contrast, the Heisenberg exchange interaction $H_{ex} = J_{ex} S_1 \cdot S_2$ favors a collinear magnetic order, which can be either ferromagnetic (FM) for $J_{ex} < 0$ or AFM for $J_{ex} > 0$. The competition between the Heisenberg exchange interaction and the DM interaction leads to the formation of exotic structures such as chiral domain walls, helices, and Skyrmions [2].

In the bulk of a noncentrosymmetric magnetic crystal, the DM vector points along one of the lattice vectors and gives rise to so-called Bloch-like structures [19–21].

FIG. 1: (a) The Rashba-AFM model is a model for systems with inversion asymmetry: an AFM/HM bilayer (left) and an AFM thin film asymmetrically embedded between two different insulator layers (right). (b), (c) Electronic dispersion relations in the limit of strong exchange coupling (b) and in the limit of high SOC (c). The superscript $\eta = \pm$ refers to the conduction (blue) and valence (red) bands, whereas the superscript $s = \pm$ specifies chiral bands. In these two extreme limits, the band structure is almost isotropic for an AFM system with in-plane anisotropy.

In an ultrathin magnetic film with impurity- or interface-induced SOC, there is also an interfacial DM interaction that corresponds to a DM vector pointing out along the interface. This gives rise to so-called Néel-type textures [14–21]. Both the sign and the amplitude of the DM vector are of critical importance for observing and engineering different chiral structures [27–40, 43–45].

AFM materials have recently attracted considerable attention as active spintronics elements [46–47]. The absence of stray fields, the possibility of operating at terahertz frequencies, and the existence of spin waves with opposite helicities make antiferromagnets promising can-
didates for use in the next generation of spin-based memory and processing devices [46, 47] with a nanoscale element base. The coexistence of strong SOC and spin ordering at the interfaces of AFM/heavy-metal (HM) bilayers makes such heterostructures particularly promising for low-dimensional spin orbitronics applications [48]. An AFM thin film sandwiched between insulators is also a common functional geometry for spin-orbitronics due to interfacial SOC. The interfacial SOC in such a system is effectively described by Rashba SOC [39]. For magnetic films, the two-dimensional (2D) Rashba model captures the main physics and trends of SOC with a broken inversion symmetry [40], such as fieldlike and dampinglike spin-orbit torques [11–13], intrinsic spin Hall effects [14], intrinsic anomalous Hall effects [50], inverse Faraday effects [51], and magnetic anisotropy [52].

In this Letter, we develop a framework for computing the interfacial DM interaction in AFM layers with inversion asymmetry. The typical system illustrated in Fig. 1 is described on the basis of an effective 2D AFM-Rashba Hamiltonian. We find that both the sign and the magnitude of the DM interaction can be tuned by modifying the ratio relating three parameters, which can represent either the total magnetization asymmetry. The typical system illustrated in Fig. 1, which also includes the s-d coupling, and $S_B[n]$ is the bosonic action describing the dynamics of the localized spins (magnons) in the absence of itinerant electrons. In our model, it is the coupling between the itinerant electrons and the local moments that determines the DM interaction, which is also directly linked to the SOC of the itinerant electrons. We will not specify the bosonic part of the action $S_B[n]$ since it is irrelevant for the subsequent discussion.

The bosonic action reads

$$S_B = \int d\tau d^3r \Phi^\dagger \left[ -i g \frac{\partial \Phi}{\partial \tau} \right] \Phi,$$

where $\beta = 1/k_B T$ is the inverse temperature and $\tau$ is the imaginary time. The inverse Green’s function operator is $g \frac{\partial}{\partial \tau}$ in terms of the Hamiltonian of Eq. [1]. We compute the effective theory for the vector field $n(r)$ by integrating out the fermionic degrees of freedom. This standard procedure results in an additional, effective contribution to the bosonic action of the form

$$\Delta S_B^{\text{eff}}[n] = \int_0^\beta d\tau \int d^3r \left( -h \text{Tr} \ln(-G^{-1}) \right).$$

Below, we analyze $\Delta S_B^{\text{eff}}[n]$ and its influence on the magnet in the AFM-Rashba model.

In our system, the symmetry-breaking direction ($z$ direction) is perpendicular to the plane $xx$. Without the loss of generality, we choose the $x$ axis to be in the direction of the in-plane vector field $n$. A small deviation of the unit vector from its equilibrium direction is then, parameterized by $n = \hat{x} + \delta n$, where $\delta n = -(\delta n_x^2 + \delta n_z^2)/2, \delta n_x, \delta n_z$. The effective action is obtained from a perturbation with respect to $\delta n$, [53] that holds irrespective of the value of the exchange parameter. The action $S$ defines the system partition function $Z = \int d[\Phi^*] d[\Phi] d[n] e^{-S[\Phi^*, \Phi, n]/\hbar}$, where $\Phi$ is the Grassmannian coherent-state spinor and $\hbar$ is Planck’s constant. In the s-d approach to magnetic systems, the action is decomposed into the sum $S = S_F + S_B$, where $S_F[\Phi^*, \Phi, n]$ is the fermionic action corresponding to the Hamiltonian of Eq. [1], which also includes the s-d coupling, and $S_B[n]$ is the bosonic action describing the dynamics of the localized spins (magnons) in the absence of itinerant electrons. In our model, it is the coupling between the itinerant electrons and the local moments that determines the DM interaction, which is also directly linked to the SOC of the itinerant electrons. We will not specify the bosonic part of the action $S_B[n]$ since it is irrelevant for the subsequent discussion.

The bosonic action reads

$$S_B = \int d\tau d^3r \int d^3r' \Phi^\dagger \left[ -h G^{-1}_{\tau \tau'} \right] \Phi_{\tau' \tau},$$

where $\beta = 1/k_B T$ is the inverse temperature and $\tau$ is the imaginary time. The inverse Green’s function operator is $h G^{-1}_{\tau \tau'} = -\left( \delta \tau + \hbar \right) \delta(\tau - \tau')$ in terms of the Hamiltonian of Eq. [1]. We compute the effective theory for the vector field $n(r)$ by integrating out the fermionic degrees of freedom. This standard procedure results in an additional, effective contribution to the bosonic action of the form

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strength $J_{sd}$. The effective action is conveniently established from the Dyson equation, $G^{-1} = G^0^{-1} - \Sigma$, where the unperturbed Green’s function refers to $\delta n = 0$ and the self-energy $h\Sigma = J_{sd} \Omega \delta(\mathbf{r} - \mathbf{r'})/\delta(\tau - \tau')$ is introduced.

Computing the self-energy up to the second order in $\delta n$ yields

$$\Delta \Sigma_{\text{eff}}[n] = \hbar \beta \sum_{k \neq 0} \delta n^a_{k,l} \Pi_{k,l}^b \delta n^b_{-k,-l}, \quad (3)$$

where the indices $a, b = (y, z)$ denote the transverse vector components with respect to the equilibrium $\hat{x}$ direction, $\nu_l = 2n\pi/\beta$ denotes the bosonic Matsubara frequencies, and

$$\Pi_{k,l}^b = \frac{J_{sd}^2}{2\hbar \beta} \sum_{q,n} \text{Tr}[\Gamma_a G_{q,n}^0 \Gamma_b G_{q+k,n+n+i\nu_l}] \quad (4)$$

is the dynamical susceptibility tensor, pictured schematically in Fig. 2a. Here, $G_{q,n}^0 = (i\hbar \omega_n - \mathcal{H})^{-1}$ is the equilibrium Green’s function, and the $\omega_n = (2n + 1)\pi/\beta$ are the fermionic Matsubara frequencies. We compute the sum over the fermionic Matsubara frequencies by using the identity $\sum_n (i\hbar \omega_n - E)^{-1}/\beta = f(E)$, where $f(E)$ is the Fermi distribution.

By expanding the static limit, $\nu_l = 0$, on the spin susceptibility to the second order in the wave vector $k$, we find both the electronic contribution to the symmetric Heisenberg exchange stiffness, which is determined by the symmetric terms in the diagonal elements of the susceptibility tensor, and the antisymmetric exchange interaction (DM interaction), which is determined by the antisymmetric terms in the off-diagonal elements. From the partition function of the canonical ensemble, $Z = \int d\mathbf{n} e^{-\mathcal{S}_{\text{eff}}[\mathbf{n}]/\hbar} = \int d\mathbf{n} e^{-\beta F[\mathbf{n}]}$, we obtain $\delta n \mathcal{S}_{\text{eff}}/\hbar = \beta \delta n F[\mathbf{n}]$, where the micromagnetic free energy, including the stiffness and the DM interaction, is $F[\mathbf{n}] = \int d^2 \mathbf{r} (A(\nabla \mathbf{n})^2 - D \mathbf{n} \cdot (\hat{x} \times \nabla) \times \mathbf{n})$. By comparing the microscopic free energy with the expression for the effective action, Eq. (3), we define the micromagnetic parameters $A$ and $D$, which characterize the free carrier contributions to the exchange stiffness and the DM interaction, respectively.

Upon expanding the off-diagonal elements of the tensor $\Pi$ to the first order in the wave vector $k$ [see Fig. 2b)], we obtain the relation

$$D = -\frac{\partial^2 \Pi_{zy}^y}{\partial k_y^2} \bigg|_{k=0} = -\frac{\partial^2 \Pi_{xy}^y}{\partial k_y^2} \bigg|_{k=0} \quad (5a)$$

$$= \frac{J_{sd}^2}{2\beta} \sum_{q,n} \text{Tr}[\Gamma_y G_{q,n}^0 \Gamma_z G_{q,n}^0 \nu_y G_{q,n}^0] \quad (5b)$$

where $v_y = \hbar^{-1} \partial \mathcal{H}_q/\partial q_y$ is the $y$ component of the velocity operator. From the second-order terms, we obtain the electron contribution to the exchange stiffness:

$$A = -\frac{\partial^2 \Pi_{zy}^y}{\partial k_x^2} \bigg|_{k=0} = -\frac{\partial^2 \Pi_{xz}^z}{\partial k_x^2} \bigg|_{k=0}. \quad (6)$$

In our model, $A$ describes the contribution to the AFM exchange interaction from a superexchange-type interaction between the localized spins in the AFM layer via the itinerant spins.

We should emphasize here that in this approach, we have ignored the spin fluctuations of the localized AFM spins, which is a valid omission as long as the system temperature is much less than the critical Néel temperature.

To model an AFM system with interfacial SOC, we use the 2D AFM-Rashba Hamiltonian on a square lattice:

$$\mathcal{H} = \gamma_k \tau_z \mathbf{s}_0 + J_{sd} \tau_z \mathbf{s} \cdot \mathbf{n} - \alpha R \tau_z (\mathbf{s} \times \mathbf{k}) \cdot \hat{z}, \quad (7)$$

where $\mathbf{s}$ and $\tau$ are the vectors of Pauli matrices representing the spin and AFM sublattice degree of freedom, respectively; $\sigma_0$ is the identity matrix; $\mathbf{n}$ is the staggered order parameter (the normalized Néel vector); and $\alpha R$ is the strength of the Rashba SOC. The kinetic energy of the itinerant electrons is $\gamma_k = a^2 t (k^2 - k_0^2)$, where $t$ is the nearest-neighbor hopping energy and $k_0 = 2/a$, with $a$ being the lattice constant.

The band structure of the AFM-Rashba Hamiltonian of Eq. (7) is, in general, anisotropic. It is convenient to parameterize the four spectral branches as follows:

$$E_{k}^{s,0} = \eta \sqrt{\alpha^2 + J_{sd}^2 + \alpha^2 k_x^2 + 2s\alpha R k_x^2}, \quad (8)$$

where $s, \eta = \pm 1$ are the spin chirality and electron/hole band indices, respectively. We also introduce $\xi_k = (\gamma_k^2 + J_{sd}^2 \cos^2 \phi)^{1/2}$ with the in-plane wave vector $k$ parameterized by the angle $\phi$, such that $k = k(\cos \phi, \sin \phi, 0)$.

Let us now analyze the expressions of Eqs. (5) and (6) for the AFM-Rashba Hamiltonian of Eq. (7) in the limit of weak spin-orbit interaction, $\{k_0 \alpha R, J_{sd}\} \ll \min\{t, \epsilon F\}$, and at zero temperature. Using the relation $\Gamma_a = \tau_z \sigma_a$, we obtain the relation

$$D = -\frac{A k_0^2}{4t} \alpha R, \quad (9)$$

where the stiffness parameter is

$$A = \frac{t J_{sd}^2}{2\pi \epsilon F} \begin{cases} 2 - \epsilon F^2/16t^2, & \epsilon F < 4t, \\ 1, & \epsilon F > 4t, \end{cases} \quad (10)$$

which is manifestly independent of the SOC strength in the limit of weak spin-orbit interaction. Here, the Fermi energy $\epsilon F$ is measured with respect to the center of the energy gap [see Figs. 4(b) and (c)]. In this regime, the
Hamiltonian of the Rashba SOC symmetry, the DM interaction vanishes inside the gap but remains finite outside the gap, even at zero temperature. Thus, we conclude that the DM interaction exhibits a qualitatively different behavior in antiferromagnets compared with that in ferromagnets.

In Fig. 3, we illustrate the behavior of the DM interaction on the basis of a numerical analysis of Eq. (5), beyond the weak SOC regime of Eqs. (9-11), where typical material parameters are assumed: \( t = 3 \) eV, \( \epsilon_F = 2 \) eV, \( J_{sd} = 1 \) eV, \( a = 0.4 \) nm and \( k_0 \alpha_R = 0.1 \) eV. The results indicate a rather large interfacial DM interaction, with \( D \approx -1.6 \) pJ/m. Moreover, when the Fermi energy lies within the gap, i.e., \( |\epsilon_F| < J_{sd} \), we find an order-of-magnitude enhancement of the DM interaction with the opposite sign, \( D \approx 12.7 \) pJ/m. Importantly, the estimated strength of the DM interaction is at least an order of magnitude larger than that for a Rashba-FM layer with the same parameters.

For small Fermi energies, the DM interaction is positive and almost independent of the Fermi energy (see the top panel in Fig. 3), as might be expected from Eq. (11). In the metallic regime (i.e., for Fermi energies well above the gap), the DM interaction is negative, and its strength decreases in proportion to \( \epsilon_F^{-2} \), in good agreement with Eqs. (9) and (10). The sign inversion of the DM interaction is rather sharp and occurs at \( \epsilon_F = \sqrt{J_{sd}^2 + \alpha_R^2 t^2} \). The bottom panel of Fig. 3 also confirms that the strength of the DM interaction is linearly proportional to \( \alpha_R \) in the weak SOC regime, in agreement with Eq. (9). Thus, we conclude that the DM interaction in an AFM material may vary by orders of magnitude depending on the material parameters, as illustrated in Fig. 3.

Our calculations also show that, unlike in FM-Rashba systems, the temperature dependence of the DM interaction in the AFM-Rashba model of Eq. (7) is weak due to a large contribution to the DM interaction from the valence bands (not shown). As we have already discussed, this is correct if the system temperature is much less than the Néel temperature, i.e., in a regime in which the spin fluctuations of the AFM layer are suppressed.

Controlling the DM interaction is essential for engineering chiral magnetic structures. If the DM interaction parameter exceeds a certain critical value, which is determined by the Heisenberg exchange interaction and the uniaxial anisotropy, then the ground state changes from a collinear configuration to either a helimagnetic state or a Skyrmion lattice. A weaker DM interaction enables the stabilization of isolated Skyrmions in a metastable state. In chiral magnets, the sign of the DM interaction determines the direction, or handedness, of spin rotation.

The asymmetry of the spin-wave dispersion in spin-polarized electron energy-loss spectroscopy and Brillouin light scattering is a measure of the DM interaction in FM...
Although the interfacial DM interactions in a few FM/HM bilayers have been experimentally studied in recent years, we are not aware of similar measurements in AFM heterostructures. However, the first observation of a bulk DM interaction in an AFM system, namely, the noncentrosymmetric $\alpha - \text{Cu}_2\text{V}_2\text{O}_7$, has recently been reported in Ref. [43] based on inelastic neutron scattering. Very recently, a large DM interaction in Fe/Ir bilayers on Rh(001) has been predicted on the basis of ab initio calculations [61]. We hope that our work will stimulate new experiments and ab initio works on such AFM heterostructures.

In summary, we have computed the DM interaction, both analytically and numerically, using an effective model for AFM layers with interfacial Rashba SOC. In both analytically and numerically, using an effective model for AFM layers with interfacial Rashba SOC. In summary, we have computed the DM interaction, both analytically and numerically, using an effective model for AFM layers with interfacial Rashba SOC. In summary, we have computed the DM interaction, both analytically and numerically, using an effective model for AFM layers with interfacial Rashba SOC. In summary, we have computed the DM interaction, both analytically and numerically, using an effective model for AFM layers with interfacial Rashba SOC. In summary, we have computed the DM interaction, both analytically and numerically, using an effective model for AFM layers with interfacial Rashba SOC. In summary, we have computed the DM interaction, both analytically and numerically, using an effective model for AFM layers with interfacial Rashba SOC.
In general, the effective action has two extra terms related to the spin susceptibilities with a long wavelength nature, i.e., $k = 0$, which have no contributions in the exchange stiffness and DM interaction: \[
\Delta S_{\text{eff}}[\mathbf{n}] = \hbar \beta \left( \delta n_\nu < \mathbf{\hat{a}} > + \sum_{l} \delta n_\nu \left( \hbar \beta \sum_{i} \mathbf{\Gamma}_{\nu l}(\mathbf{i}) + \mathbf{\Gamma}_{\nu l}(\mathbf{i})^{T} \right) \right).
\] The static spin susceptibility related to the first-order term in $\delta n$ is $\chi_{ab}^\nu = \frac{\mathbf{J}_{ab}}{\hbar \beta} \sum_{\mathbf{q},\mathbf{\omega}_0} \text{Tr}[\mathbf{\Gamma}_{\nu} G_{\mathbf{q},\mathbf{\omega}_0}^{0}], \text{ and the dynamic spin susceptibility} \text{ related to the second-order term in } \delta n_a$ is $\Pi_{ab}^{\nu \nu} = \sum_{\mathbf{q},\mathbf{\omega}_0} \left\{ - \left( \mathbf{J}_{ab}/2\hbar \beta \right) \text{Tr}[\mathbf{\Gamma}_{\nu} G_{\mathbf{q},\mathbf{\omega}_0}^{0} \delta_{ab} + (J_{ab}^{2}/2\hbar \beta) \text{Tr}[\mathbf{\Gamma}_{\nu} G_{\mathbf{q},\mathbf{\omega}_0}^{0} \mathbf{\Gamma}_{\nu} G_{\mathbf{q},\mathbf{\omega}_0}^{0}]], \right\}$, where $\delta_{ab}$ is the Kronecker delta function.