Anatomy of the magnetic anisotropy energy mediated by tight-binding Rashba electrons

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Abstract

The magnetic anisotropy is a fundamental quantity, defining the orientational stability of the magnetic state. Due to its importance, many different approaches have been put forth to explain its properties and behavior, ranging from simple models to fully first-principles calculations based on the electronic structure of a chosen material. In this work, we focus on a simple tight-binding model of spin-orbit-coupled electrons exchange-coupled to a background ferromagnetic order, meant to abstract the essential physics at the interface between a ferromagnetic layer and a heavy-metal layer. We propose a new method to calculate the magnetic anisotropy energy, based on the spin susceptibility of the electrons. This is compared to other approaches, such as energy differences between different orientations of the ferromagnet, or the anisotropy of the spin-orbit energy. We investigate not only their compatibility but also what physical insights can be gained from each of them. In particular, we establish under what conditions perpendicular magnetic anisotropy is favored, and how it can be maximized in the present model. The computation of the magnetic anisotropy energy from the spin susceptibility is general, and can be readily extended to realistic electronic structure calculations.
I. INTRODUCTION

Spintronics aims to utilize the electron spin as the active degree of freedom for information storage and processing. To uncover the fundamental physical mechanisms enabling such functionality, bilayers consisting of a magnetic layer and a heavy-metal layer have been studied extensively. Besides the properties intrinsic to each layer, the interface between the two plays a crucial role. At the interface, the strong spin-orbit coupling (SOC) derived from the heavy-metal layer meets the strong exchange interaction of the magnetic layer, breaking both time-reversal and inversion symmetry. This leads to many interesting physical phenomena: perpendicular magnetic anisotropy (PMA), the Dzyaloshinskii-Moriya interaction, spin-orbit torques, Rashba-Edelstein effect, etc. These properties can be engineered by tuning the chemical composition and interface structure, or by applying a gate voltage, as demonstrated for the magnetic anisotropy.

The magnetic anisotropy energy (MAE) is the dependence of the energy of a magnetic system on the real-space orientation of its magnetization. The MAE is responsible for the orientational stability of magnetic domains, an observation that lies at the heart of both magnetic hard disk drives and magnetic random access memories. There are two main contributions to the MAE: the magnetocrystalline anisotropy (of electronic origin) and shape anisotropy (arising from the magnetostatic dipolar interaction). For a thin ferromagnetic film, the magnetostatic energy is minimized when the magnetization is in the plane of the film, leading to in-plane magnetic anisotropy (IMA). To stabilize PMA, the magnetocrystalline anisotropy energy must overcome the shape anisotropy. From the technological point of view, PMA is very important, as it allows to increase the bit storage density, by shrinking the size of the magnetic domains that store each bit of information. This spurred a lot of experimental and theoretical efforts to design, grow and understand the properties of magnetic materials displaying PMA.

It was pointed out by Bloch and van Vleck that the MAE comes about through the SOC, and Brooks outlined its description in terms of the electronic structure. Bruno made the appealing connection between the MAE and the anisotropy of the orbital magnetic moment, which was generalized in Ref. 14. As an example, density functional theory (DFT) calculations for transition-metal systems show that Bruno’s connection holds for 3d transition metals and their compounds, and also for thin films. However there are several
counter-examples, e.g. as found experimentally and theoretically in Ref. 17. Total energy differences from self-consistent DFT calculations are the obvious but challenging way of computing the MAE for a target system\textsuperscript{18}. A simplification based on the magnetic force theorem\textsuperscript{19} replaces the former with the difference of band energies\textsuperscript{15}, which are easier to converge. A different approach is to evaluate directly the derivative of the energy with respect to the ferromagnetic orientation, which led to the so-called torque method\textsuperscript{20}. Recently it has been proposed by Antropov \textit{et al.}\textsuperscript{21} that a numerically stable way of computing the MAE is to evaluate half of the anisotropy in the SOC energy term in the hamiltonian, adapting to electronic structure calculations an idea already advanced by van der Laan\textsuperscript{22}.

The Rashba model\textsuperscript{23} is the prototype for the effects of SOC on the band structure and materials properties of surfaces and interfaces. In combination with the exchange interaction generated by the magnetic layer, it has been extensively employed to interpret the properties of magnetic/heavy-metal bilayers, among other systems\textsuperscript{6}. Ref. 24 presented a simple theoretical description of the MAE using the free-electron Rashba model. However, the same work also argued that the finite bandwidth must be taken into account in order to correctly account for the PMA, and this was confirmed in Ref. 25. A gate voltage was experimentally demonstrated to control the Rashba coupling strength\textsuperscript{26}, which might provide a route to the electrical control of the MAE\textsuperscript{7}.

In this work we study the magnetocrystalline anisotropy of a ferromagnet/heavy-metal bilayer, driven by an interfacial Rashba SOC, highlighting different physical regimes and considering different ways of interpreting the results. We develop the theory for the finite bandwidth case, employing the tight-binding approximation. The behavior of the MAE is analyzed with respect to the three competing energy scales: the non-relativistic kinetic energy $t'$, the Rashba SOC strength $t''$, and the exchange coupling $J_{sd}$ to the ferromagnetic order parameter. We contrast the global definition of the MAE (energy difference between different ferromagnetic directions of the system) with its local definition (curvature of the energy for a given ferromagnetic direction). This curvature of the energy is evaluated from the electronic spin susceptibility, providing a new way to compute the MAE. The role of the Fermi surface is identified from the behavior of the intraband and interband contributions to the susceptibility. The analytic treatment of the half-filled case provides a figure of merit for PMA in this model, and numerical calculations recover the $\text{IMA} \rightarrow \text{PMA} \rightarrow \text{IMA}$ behavior of the MAE when the filling is increased from zero to two electrons\textsuperscript{25}. The recent
proposal that the MAE can be estimated as half of the anisotropy in the SOC energy term in the Hamiltonian is also explored. We consider three qualitatively different parameter regimes for a detailed study: (i) strong exchange \((J_{sd} \gg t' \gg t'')\), (ii) intermediate exchange \((J_{sd} \sim t' \gg t'')\), and (iii) weak exchange \((t' \gg J_{sd} \sim t'')\).

The paper is organized as follows. In Sec. II we present the tight-binding model and the theoretical and numerical methods, and illustrate the main features of the electronic structure. Different ways of computing the MAE and an overview of the results are discussed in Sec. III, connecting to existing work. The half-filled case is analyzed using perturbation theory in Sec. IV, proving that it always leads to PMA and providing a useful figure of merit for the MAE. Then the MAE is studied in detail in Sec. V, focusing on three physically distinct cases via suitable combinations of model parameters. Our conclusions are gathered in Sec. VI, and some derivations and analytical calculations are presented in three appendices.

II. MODEL AND METHODS

To describe the itinerant electrons with SOC, we consider a square lattice with one orbital per site and nearest-neighbor hopping only, and a Rashba-like spin-momentum locking:

\[
\mathcal{H}_e = -t \sum_{<i,j>} \sum_{s,s'} c_{is}^\dagger \left( \cos \phi_R \sigma^0_{ss'} - i \sin \phi_R \left( \hat{z} \times \hat{R}_{ij} \right) \cdot \sigma_{ss'} \right) c_{js'}.
\]

Here \(c_{is}^\dagger\) and \(c_{is}\) are the creation and annihilation operators for an electron with spin \(s\) at a lattice site \(\mathbf{R}_i\), the hopping strength is given by \(t\), \(\sigma^0\) is the unit \(2 \times 2\) spin matrix, and \(\sigma = (\sigma^x, \sigma^y, \sigma^z)\) is the vector of Pauli matrices. The vector connecting site \(i\) to site \(j\) is \(\mathbf{R}_{ij} = \mathbf{R}_j - \mathbf{R}_i\), and the cross product makes the spin-momentum locking perpendicular to the bond direction, \(\hat{R}_{ij} = \mathbf{R}_{ij}/|\mathbf{R}_{ij}|\) and to the normal to the lattice plane, \(\hat{z}\). The angle \(\phi_R\) sets the balance between the conventional spin-independent part of the hopping and the chiral Rashba part.

We impose Born-von Karman periodic boundary conditions and introduce the lattice Fourier transforms of the operators,

\[
c_{is} = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} e^{i\mathbf{k} \cdot \mathbf{R}_i} c_{s}(\mathbf{k}) , \quad \frac{1}{N} \sum_{\mathbf{k}} e^{i\mathbf{k} \cdot (\mathbf{R}_i - \mathbf{R}_j)} = \delta_{ij} , \quad \frac{1}{N} \sum_{i} e^{i(\mathbf{k} - \mathbf{k}') \cdot \mathbf{R}_i} = \delta_{\mathbf{k}\mathbf{k}'} ,
\]

where \(N\) is the number of lattice sites. This leads to the \(k\)-space representation of the
Hamiltonian matrix elements,

\[ \mathcal{H}_e(k) = -t' (\cos k_x + \cos k_y) \sigma^0 - t'' (\sin k_x \sigma^y - \sin k_y \sigma^x) = \mathcal{H}_0(k) + \mathcal{H}_R(k) \quad . \]  

(3)

The lattice constant is taken as the unit of length, \( a = 1 \), and we define \( t' = 2t \cos \phi_R \) and \( t'' = 2t \sin \phi_R \). These parameters set the relative importance of the conventional and Rashba hopping terms. For small \( k \)-vectors (setting \( \hbar = 1 \) and ignoring the leading constant term),

\[ \mathcal{H}_e(k) \approx \frac{t'(k_x^2 + k_y^2)}{2} \sigma^0 - t'' (k_x \sigma^y - k_y \sigma^x) \]

(4)

\[ = \frac{k_x^2 + k_y^2}{2m^*} \sigma^0 + \alpha (k \times \hat{z}) \cdot \sigma \quad , \]

(5)

which is the form of the Hamiltonian for a Rashba electron gas, with \( m^* \) the effective mass of the parabolic dispersion, and \( \alpha \) the Rashba parameter.

We next consider that at each lattice site there is a ferromagnetic exchange coupling \( J_{sd} > 0 \) between the itinerant electrons and localized magnetic moments \( \mathbf{S}_i \). The latter are treated as classical spins of unit length, which define a background magnetic field acting on the spin of the itinerant electrons. For a ferromagnetic background (\( \mathbf{B} = J_{sd} \mathbf{S} \)), the Hamiltonian is translationally invariant (cf. Eq. (3)),

\[ \mathcal{H}(k) = \mathcal{H}_e(k) - \mathbf{B} \cdot \sigma = \mathcal{H}_0(k) + \mathcal{H}_R(k) + \mathcal{H}_B \quad , \]

(6)

and its matrix elements are

\[ \mathcal{H}(k) = E_0(k) \sigma^0 - \mathbf{b}(k) \cdot \sigma \quad , \]

(7)

where

\[ E_0(k) = -t' (\cos k_x + \cos k_y) \quad , \]

(8)

and

\[ \mathbf{b}(k) = \mathbf{b}_R(k) + \mathbf{B} = (B_x + t'' \sin k_y) \hat{x} + (B_y - t'' \sin k_x) \hat{y} + B_z \hat{z} \quad . \]

(9)

Here \( \mathbf{b}_R(k) \) is the Rashba spin-orbit field, and the coupling to the ferromagnetic background is given by \( \mathbf{B} = J_{sd} (\cos \varphi \sin \theta \hat{x} + \sin \varphi \sin \theta \hat{y} + \cos \theta \hat{z}) \), with the spherical angles \( \theta \) and \( \varphi \) specifying its orientation.

We can then immediately diagonalize the Hamiltonian,

\[ \mathcal{H}(k) = E_+(k) P_+(k) + E_-(k) P_-(k) \quad , \]

(10)
with the band dispersions

$$E_{\pm}(k) = E_0(k) \mp |b(k)|$$

and corresponding eigenspace projectors

$$P_{\pm}(k) = \frac{1}{2} \left( \sigma^0 \pm \hat{b}(k) \cdot \sigma \right) , \quad \hat{b}(k) = \frac{b(k)}{|b(k)|} .$$

The plus sign corresponds to the majority band and the minus sign to the minority band. The band dispersions are plotted in Fig. 1 for some representative cases.

In the following expressions the explicit dependence on the model parameters and the orientation of the ferromagnetic background is omitted for simplicity. The electronic density of states (DOS) is given by

$$\rho(E) = \sum_{n=\pm} \int \frac{dk}{(2\pi)^2} \delta(E - E_n(k)) ,$$

which leads to the number of electrons per lattice site,

$$N_e = \int \frac{dk}{(2\pi)^2} \left( f_+(k) + f_-(k) \right) = \int_{-\infty}^{E_F} dE \rho(E) .$$

The integral is over the first Brillouin zone, and $f_n(k) = \Theta(E_F - E_n(k))$ is the occupation of the corresponding eigenstate $E_n(k)$. The coupling to the ferromagnetic background induces a net spin moment on the itinerant electrons, given by

$$M = \int \frac{dk}{(2\pi)^2} \left( f_+(k) - f_-(k) \right) \hat{b}(k) = \int_{-\infty}^{E_F} dE \ m(E) ,$$

which defines the spin-polarized DOS (the net vector spin polarization at a given energy).

The energetics of the itinerant electrons can be obtained from the internal energy, that we consider at zero temperature,

$$U = \sum_{n=\pm} \int \frac{dk}{(2\pi)^2} f_n(k) E_n(k) = \int_{-\infty}^{E_F} dE \rho(E) E .$$

Some properties of the internal energy are summarized in Appendix A. Further insight can be gained by separating the different contributions to the internal energy (cf. Eq. (6)),

$$U = \sum_{n=\pm} \int \frac{dk}{(2\pi)^2} f_n(k) \text{Tr} P_n(k) \left( \mathcal{H}_0(k) + \mathcal{H}_R(k) + \mathcal{H}_B \right) = U_0 + U_R + U_B .$$

The first term is the usual tight-binding kinetic energy, the second term is the Rashba or SOC energy, and the third is the spin polarization energy due to the ferromagnetic background.
FIG. 1. Band dispersions given by Eq. (11) for representative cases, and their contributions to the DOS. (a) No Rashba splitting and finite exchange coupling to the background magnetization leads to a constant vertical splitting of the bands. Parameters: $\phi_\text{R} = 0$ ($t' = 2t$, $t'' = 0$), $J_\text{sd} = t$. (b) Finite Rashba splitting and no background magnetization leads to a k-dependent horizontal splitting of the bands. Parameters: $\phi_\text{R} = \pi/6$ ($t' = \sqrt{3}t$, $t'' = t$), $J_\text{sd} = 0$. (c,d) When the Rashba splitting and the background magnetization are both finite, the dispersion depends on the orientation of the magnetization with respect to the lattice. Parameters: $\phi_\text{R} = \pi/6$ ($t' = \sqrt{3}t$, $t'' = t$), $J_\text{sd} = t$. (c) When the magnetization is normal to the plane of the surface ($\mathbf{B} \parallel \mathbf{z}$) the system has fourfold rotational symmetry. (d) When the magnetization is along a nearest-neighbor direction ($\mathbf{B} \parallel \mathbf{x}$) the bands have a unidirectional shift in the perpendicular direction ($\mathbf{y}$). For this choice of parameters there are two degeneracy points, at $\mathbf{k} = (0, -\pi/2)$ and $\mathbf{k} = (\pm \pi, -\pi/2)$, one being visible in the figure. These points of degeneracy do not lead to any features in the DOS.

For derivations and some calculations, it is more convenient to employ the Green function

$$G(\mathbf{k}, E) = (E - \mathcal{H}(\mathbf{k}))^{-1} = \sum_{n=\pm} \frac{P_n(\mathbf{k})}{E - E_n(\mathbf{k})} .$$

(18)
Its basic properties and the connection to the internal energy and its derivatives are presented in Appendix B. For instance, the DOS is given by

$$\rho(E) = -\frac{1}{\pi} \text{Im} \text{Tr} \int \frac{dk}{(2\pi)^2} G(k, E),$$  \hspace{1cm} (19)$$

and the spin-polarized DOS by

$$m(E) = -\frac{1}{\pi} \text{Im} \text{Tr} \int \frac{dk}{(2\pi)^2} \sigma G(k, E),$$  \hspace{1cm} (20)$$

where the traces are over the spin components. The symbol Im stands for the procedure described in Eq. (B4).

All ground state properties can be expressed in terms of the Green function, including correlation functions. In particular, the static uniform spin susceptibility for a fixed number of electrons is given by (using Eqs. (7), (15) and (20), and the property (B3))

$$\chi^{\alpha\beta} = \frac{\partial M^{\alpha}}{\partial B^{\beta}} \bigg|_{N_e} = \frac{1}{\pi} \text{Im} \text{Tr} \int_{-\infty}^{E_F} dE \int \frac{dk}{(2\pi)^2} \sigma^{\alpha} G(k, E) \sigma^{\beta} G(k, E) - \frac{m^{\alpha}(E_F) m^{\beta}(E_F)}{\rho(E_F)}. \hspace{1cm} (21)$$

The last term is a Fermi energy correction, and comes from ensuring \(\partial N_e/\partial B^\beta = 0\), as done in the derivation of Eq. (B9). Its role is illustrated in Appendix C1 for a ferromagnetic system without SOC. The susceptibility can also be expressed directly in terms of the eigenvalues and eigenvectors of the Hamiltonian, as derived in Appendix C.

Some comments on the numerical evaluation of the different quantities are in order. Every quantity is to be calculated at constant filling \(N_e\), which requires an accurate determination of the Fermi energy \(E_F\). Keeping all other parameters fixed (magnetization orientation, etc.), \(E_F\) is a monotonic function of \(N_e\), so it can be efficiently determined using the bisection algorithm with high accuracy. For \(N_e = 1\), \(E_F = 0\). This ensures the particle-hole symmetry of the numerical results. \(E_F\) is iteratively refined using the bisection algorithm until the computed \(N_e\) is within a \(\pm 10^{-8}\) range of the desired input value. The integrals over the Brillouin zone are done with a k-mesh of 1000 \(\times\) 1000 equidistant points. To compute the DOS, the \(\delta\)-functions in Eq. (13) are approximated by Lorentzian functions with a broadening \(\eta = 10^{-3} t\). All other quantities can be computed by direct numerical summation of the contributions from each k-point, using either the analytical expressions or by contour integration of the energy dependence of the Green function expressions. Both approaches lead to very similar results.
III. COMPUTING THE MAGNETIC ANISOTROPY ENERGY

In our model, the MAE is due to the variation of the internal energy of the itinerant electrons due to a rotation of the ferromagnetic background. Following the arguments of Bloch and van Vleck\textsuperscript{10,11}, it vanishes if there is no Rashba coupling (\(\phi_R = 0\) so \(t'' = 0\)). Phenomenologically, the MAE is expanded in angular functions that respect the symmetry of the system\textsuperscript{9}. For the square lattice (tetragonal symmetry),

\[ U_{\text{MAE}}(\theta, \varphi) = K_2 \sin^2 \theta + (K_4 + K'_4 \cos 4 \varphi) \sin^4 \theta + \ldots, \tag{22} \]

with \(\theta\) and \(\varphi\) the spherical angles describing the orientation of the ferromagnetic background. Perturbation theory yields \(K_{2n} \propto t'' \left( \frac{t''}{t''_{\text{sd}}} \right)^{2n-1}\) with \(n \geq 1\) for the present model, as derived in Section IV. Higher anisotropy constants are expected to quickly decrease in magnitude, as they depend on higher powers of the ratio between the spin-orbit interaction and the spin splitting, which is typically assumed to be small. Besides perturbation theory, there are other approaches to the MAE, which we outline next.

The first and most obvious approach is to compute the internal energy of the system for several orientations of the ferromagnetic background. The anisotropy constants can then be defined either by mapping the angular dependence of the internal energy, or by assuming a model form for the angular dependence and computing only the internal energy of a few different ferromagnetic orientations. Keeping all other parameters fixed, the internal energy given by Eq. (16) is an explicit function of the angles describing the ferromagnetic orientation, \(U(\theta, \varphi)\). Assuming that the model form in Eq. (22) holds, evaluating the internal energy for three orientations is sufficient to establish whether we have PMA or IMA:

\[ \begin{align*}
U(\pi/2, 0) - U(0, 0) &= K_2 + K_4 + K'_4 \quad \left\{ \begin{array}{l}
U(\pi/2, \pi/4) - U(0, 0) = K_2 + K_4 - K'_4 \end{array} \right. > 0 \quad \text{for PMA}. \tag{23}
\end{align*} \]

This approach gives a global definition of the MAE, as it employs different magnetic states for its characterization. In Fig. 2 we show how the internal energy differences alternate between IMA \(\rightarrow\) PMA \(\rightarrow\) IMA as a function of the filling of the bands, as already found in Ref. 25. Making use of Eq. (17), we can also split the internal energy differences into three terms, and in particular explore whether the MAE can be estimated as half of the SOC energy term\textsuperscript{21,22}. We undertake this analysis in Sec. V.
FIG. 2. MAE from internal energy differences, $\Delta U = U(\pi/2, 0) - U(0, 0)$, as a function of electron filling. (a) For increasing Rashba strength and fixed coupling strength to the ferromagnetic background. Parameters: $J_{sd} = t$. (b) For fixed Rashba strength and increasing coupling strength to the ferromagnetic background. Parameters: $\phi_R = \pi/6$ ($t' = \sqrt{3}t$, $t'' = t$).

Alternatively, we can establish a local characterization of the MAE in two ways. For a chosen orientation of the ferromagnetic background, we may compute either the magnetic torque (first derivatives of the internal energy) or the curvature of the internal energy (second derivatives). The Hellmann-Feynman theorem\textsuperscript{27,28} yields the first derivatives of the internal energy (the detailed derivation is presented in Appendix B). Using Eq. (B7) we have

\begin{equation}
\frac{\partial U}{\partial \theta} = -\mathbf{M} \cdot \frac{\partial \mathbf{B}}{\partial \theta} = (K_2 + 2 (K_4 + K'_4 \cos 4\varphi \sin^2 \theta) \sin 2\theta) \sin 2\theta ,
\end{equation}

\begin{equation}
\frac{\partial U}{\partial \varphi} = -\mathbf{M} \cdot \frac{\partial \mathbf{B}}{\partial \varphi} = -4 K'_4 \sin 4\varphi \sin^4 \theta ,
\end{equation}

The corresponding derivatives of Eq. (22) were included for comparison. As indicated by the phenomenological expression for $U_{\text{MAE}}(\theta, \varphi)$, the magnetic torque $\mathbf{M} \times \mathbf{B}$ must vanish for the high-symmetry directions. These are the nearest- and next-nearest-neighbor directions ($\theta = \pi/2$ and $\varphi = n\pi/4$, with $n \in \{0, 1, \ldots, 7\}$), and the normal to the lattice plane ($\theta = 0, \pi$).

On the other hand, the second derivatives of the internal energy are particularly simple for these high-symmetry directions. Only the cartesian components of the spin susceptibility tensor for the plane perpendicular to a chosen direction are required (transverse susceptibility). For the high-symmetry directions the net spin moment of the itinerant electrons is
aligned with the ferromagnetic background, \( \mathbf{M} \parallel \mathbf{S} \), and so the Fermi energy corrections in Eq. (21) vanish for the transverse susceptibilities. For \( \mathbf{M} \parallel \hat{x} \) (one of the nearest-neighbor directions), Eqs. (21) and (B9) lead to the non-zero derivatives

\[
\frac{1}{2} \left. \frac{\partial^2 U}{\partial \theta^2} \right|_{\mathbf{M} \parallel \hat{x}} = \left( \frac{J_{sd}}{2} \right)^2 \frac{M}{J_{sd}} - \chi^{zz},
\]

(26)

\[
\frac{1}{2} \left. \frac{\partial^2 U}{\partial \varphi^2} \right|_{\mathbf{M} \parallel \hat{x}} = \left( \frac{J_{sd}}{2} \right)^2 \frac{M}{J_{sd}} - \chi^{yy},
\]

(27)

Here \( M = |\mathbf{M}| \) is the magnitude of the net spin moment of the itinerant electrons, which depends on the orientation of the ferromagnetic background. Although the spherical coordinate system is singular at the poles, the second derivative can be defined there as

\[
\frac{1}{2} \left. \frac{\partial^2 U}{\partial \theta^2} \right|_{\mathbf{M} \parallel \hat{z}} = \left( \frac{J_{sd}}{2} \right)^2 \frac{M}{J_{sd}} - \chi^{xx}.
\]

(28)

It is independent of the azimuthal angle due to the high symmetry of the square lattice (\( \chi^{xx} = \chi^{yy} \) and \( \chi^{xy} = \chi^{yx} = 0 \)). The model form of Eq. (22) was again used for comparison.

We can gain further insight into the MAE by separating the transverse spin susceptibility into intraband and interband contributions, as explained in Appendix C. The intra and interband contributions to the spin susceptibility become

\[
\chi^{\alpha \alpha}_{\text{intra}} = \int \frac{d\mathbf{k}}{(2\pi)^2} \left( \mathbf{\hat{b}}_\alpha(\mathbf{k}) \right)^2 \sum_n \delta(E_F - E_n(\mathbf{k}))
\]

(29)

\[
\chi^{\alpha \alpha}_{\text{inter}} = -\int \frac{d\mathbf{k}}{(2\pi)^2} \left( \mathbf{\hat{b}}_\alpha(\mathbf{k}) \right)^2 \frac{f_+(\mathbf{k}) - f_-(\mathbf{k})}{|\mathbf{b}(\mathbf{k})|}
\]

(30)

with \( \alpha = x, y, z \). Here \( \mathbf{\hat{b}}_\alpha(\mathbf{k}) \) are the cartesian components of the unit vector defining the spin quantization axis for each \( \mathbf{k} \), see Eq. (9). \( \chi^{\alpha \alpha}_{\text{intra}} \) arises from the Fermi surface and is positive, while \( \chi^{\alpha \alpha}_{\text{inter}} \) arises from the Fermi sea and is negative. We define the anisotropic volume susceptibility starting from Eq. (15) and subtracting the third contribution to the spin susceptibility, Eq. (C9),

\[
\chi^0 = \frac{M}{J_{sd}} - \bar{\chi} = \int \frac{d\mathbf{k}}{(2\pi)^2} \frac{\mathbf{S} \cdot \mathbf{b}_R(\mathbf{k})}{J_{sd}} \frac{f_+(\mathbf{k}) - f_-(\mathbf{k})}{|\mathbf{b}(\mathbf{k})|}.
\]

(31)

Its sign can be positive or negative, and vanishes when SOC is absent.

Whether we have PMA or IMA can then be established in two ways. Setting \( \mathbf{M} \parallel \hat{z} \) we find that \( \chi^0 = 0 \), so the sign of the MAE is decided by the competition between the intraband and interband contributions to the spin susceptibility \( \chi^{xx} \). On the other hand,
when \( \mathbf{M} \parallel \hat{x} \) we have \( \chi^{zz} = 0 \), and sign of the MAE is determined by \( \chi^0 \). The detailed analysis in Sec. V shows that both results are consistent, and can be given a meaningful interpretation.

IV. PERTURBATION THEORY FOR THE GAPPED HALF-FILLED CASE

Let us study in detail the gapped half-filled case, when the majority band is full, \( f_+ (k) = 1 \), and the minority band is empty, \( f_- (k) = 0 \). This case was found numerically to have PMA, both in our calculations and in Ref. 25, and now we shall prove this analytically. Starting from Eqs. (9), (11) and (16), the internal energy for this case is simply

\[
U = \int \frac{dk}{(2\pi)^2} \left( E_0(k) - |b(k)| \right)
\]  

(32)

We see that only the second term in the integrand contains information about the orientation of the ferromagnetic background, given by the angles \( \theta \) and \( \varphi \).

We next expand the spin splitting \( |b(k)| \) in order to extract the \( \theta \) and \( \varphi \) dependence:

\[
|b(k)| = (|b_R(k)|^2 + (J_{sd})^2 + 2J_{sd} S \cdot b_R(k))^\frac{1}{2}
\]

\[
= b_0(k) \left( 1 + \cos \gamma(k) \right)^\frac{1}{2} = b_0(k) \sum_{n=0}^{\infty} \left( \frac{1}{2} \right)_n \left( \cos \gamma(k) \right)^n
\]  

(33)

defining

\[
b_0(k) = (|b_R(k)|^2 + (J_{sd})^2)^\frac{1}{2}, \quad \cos \gamma(k) = \frac{2 J_{sd} S \cdot b_R(k)}{|b_R(k)|^2 + (J_{sd})^2}
\]  

(34)

The binomial series was used. We next rearrange the expansion,

\[
|b(k)| = \sum_{n=0}^{\infty} \left( \frac{1}{2} \right)_n \frac{(2J_{sd} t''(k))}{(b_0(k))^{2n-1}} (\sin \theta)^n (\sin k_y \cos \varphi - \sin k_x \sin \varphi)^n
\]

\[
= \sum_{n=0}^{\infty} \left( \frac{1}{2} \right)_n \frac{(2J_{sd} t''(k))}{(b_0(k))^{2n-1}} (\sin \theta)^n \sum_{p=0}^{n} \binom{n}{p} (-1)^p (\sin k_y \cos \varphi)^p (\sin k_x \sin \varphi)^{n-p}
\]

\[
= \sum_{n=0}^{\infty} \sum_{p=0}^{n} B_n^p (k) (\sin \theta)^n (\cos \varphi)^p (\sin \varphi)^{n-p},
\]  

(35)

with the expansion coefficients

\[
B_n^p (k) = (-1)^p 2^n \left( \frac{1}{2} \right)_n \binom{n}{p} \frac{(J_{sd} t''(k))^n (\sin k_y)^p (\sin k_x)^{n-p}}{|b_R(k)|^2 + (J_{sd})^2}^{n- \frac{1}{2}}
\]  

(36)
The internal energy has then the expansion

\[ U(\theta, \varphi) = \sum_{n=0}^{\infty} \sum_{p=0}^{n} U_n^p (\sin \theta)^n (\cos \varphi)^p (\sin \varphi)^{n-p}, \quad (37) \]

with the coefficients

\[ U_n^p = -\int \frac{d\mathbf{k}}{(2\pi)^2} B_n^p(\mathbf{k}). \quad (38) \]

The behavior of the integrand under either \( k_x \to -k_x \) or \( k_y \to -k_y \) dictates that \( U_{2n+1}^{2p+1} = 0 \) and \( U_{2n}^{2p} = 0 \), so that only terms even in both \( p \) and \( n \) survive. In combination with the symmetry of the binomial coefficients, we also have

\[ U_{2n}^{2p} - 2U_{2n}^{2p+2} = 0. \]

The first terms in the expansion are then

\[ U(\theta, \varphi) \approx U_0^0 + U_0^2 \sin^2 \theta + \left( \frac{6U_0^0 + U_4^2}{8} + \frac{2U_4^0 - U_4^2}{8} \cos 4\varphi \right) \sin^4 \theta, \quad (39) \]

in agreement with the phenomenological form given in Eq. (22).

As we are considering the gapped half-filled case, we may take \(|t''| \ll J_{\text{sd}}\) and expand the integrands

\[ B_0^0(\mathbf{k}) \approx J_{\text{sd}} + \frac{|b_R(\mathbf{k})|^2}{2J_{\text{sd}}} \left( 1 - \frac{|b_R(\mathbf{k})|^2}{4(J_{\text{sd}})^2} \right), \quad (40a) \]

\[ B_0^2(\mathbf{k}) \approx -\frac{(t'')^2}{2J_{\text{sd}}} \sin^2 k_x \left( 1 - \frac{3|b_R(\mathbf{k})|^2}{2(J_{\text{sd}})^2} \right), \quad (40b) \]

\[ B_4^0(\mathbf{k}) \approx -\frac{5(t'')^4}{8(J_{\text{sd}})^3} \sin^4 k_x, \quad (40c) \]

\[ B_4^2(\mathbf{k}) \approx -\frac{15(t'')^4}{4(J_{\text{sd}})^3} \sin^2 k_x \sin^2 k_y. \quad (40d) \]

The following integral can then be used to generate all \( U_n^p \) coefficients:

\[ I_\ell(x, y) = \frac{1}{(2\pi)^2} \int_{-\pi}^{\pi} \int_{-\pi}^{\pi} \, dk_x \, dk_y \, (x \sin^2 k_x + y \sin^2 k_y)^\ell \]

\[ \approx \sum_{k=0}^{\ell} \binom{\ell}{k} x^k y^{\ell-k} \frac{4}{\pi^2} \int_0^{\pi} \, dk_x \, (\sin k_x)^{2k} \int_0^{\pi} \, dk_y \, (\sin k_y)^{2(\ell-k)} \]

\[ = \sum_{k=0}^{\ell} \binom{\ell}{k} \frac{(2k-1)!!}{(2k)!!} \frac{(2(\ell-k)-1)!!}{(2(\ell-k))!!} x^k y^{\ell-k}. \quad (41) \]

For the general case of the integrand we derive

\[ \frac{(\ell-m-n)!}{\ell!} \frac{\partial^{\ell-m-n} I_\ell}{\partial x^m \partial y^n}(1, 1) = \int \frac{d\mathbf{k}}{(2\pi)^2} \left( \sin^2 k_x \right)^m \left( \sin^2 k_y \right)^n \left( \sin^2 k_x + \sin^2 k_y \right)^{\ell-m-n}. \quad (42) \]
The polynomials that will be needed in the following are

\[ I_0(x, y) = 1, \quad I_1(x, y) = \frac{x + y}{2}, \quad I_2(x, y) = \frac{3}{8} (x^2 + y^2) + \frac{1}{2} xy. \] (43)

The coefficients in the expansion of the internal energy are (skipping the constant shift of the energy)

\[ U_2^0 = \frac{1}{2} \frac{(t'')^2}{J_{sd}} \frac{\partial I_1}{\partial x}(1, 1) - \frac{3}{8} \frac{(t'')^4}{(J_{sd})^3} \frac{\partial I_2}{\partial x}(1, 1) = \frac{1}{4} \frac{(t'')^2}{J_{sd}} - \frac{15}{64} \frac{(t'')^4}{(J_{sd})^3}, \] (44a)

\[ U_4^0 = \frac{5}{16} \frac{(t'')^4}{(J_{sd})^3} \frac{\partial^2 I_2}{\partial x^2}(1, 1) = \frac{15}{64} \frac{(t'')^4}{(J_{sd})^3}, \] (44b)

\[ U_4^2 = \frac{15}{8} \frac{(t'')^4}{(J_{sd})^3} \frac{\partial^2 I_2}{\partial x \partial y}(1, 1) = \frac{15}{16} \frac{(t'')^4}{(J_{sd})^3}. \] (44c)

From Eqs. (22) and (39), the anisotropy coefficients are then

\[ K_2 = \frac{1}{4} \frac{(t'')^2}{J_{sd}} - \frac{15}{32} \frac{(t'')^4}{(J_{sd})^3}, \quad K_4 = \frac{75}{256} \frac{(t'')^4}{(J_{sd})^3}, \quad K_4' = -\frac{K_4}{5}. \] (45)

This proves that the gapped half-filled case always displays PMA (when perturbation theory is valid). The fourth order correction softens \( K_2 \), but \( K_4 \) reinforces the easy-axis character. The in-plane anisotropy is weak when compared to the uniaxial one, and favors alignment along the nearest-neighbor directions. For comparison, Appendix C.2 derives the same results starting from the transverse spin susceptibility.

A useful figure of merit against which the MAE can be compared is thus the leading contribution to the PMA,

\[ K_{ref} = \frac{1}{4} \frac{(t'')^2}{J_{sd}}. \] (46)

Fig. 3 shows the region of validity and the breakdown of perturbation theory for this case. The maximum value of the PMA is obtained when the gap between the bands is about to close (e.g. when \( J_{sd} \approx 4t \) for small \( \phi_R \) or \( t'' \ll t' \) ), which sets a limit on how much the PMA can be enhanced by reducing the magnitude of \( J_{sd} \).

V. THREE CASE STUDIES

We now present a detailed analysis of the MAE for three different choices of model parameters, meant to illustrate different physical regimes at the ferromagnet/heavy-metal interface: (i) strong exchange \( (J_{sd} \gg t' \gg t'') \), (ii) intermediate exchange \( (J_{sd} \sim t' \gg t'') \),
FIG. 3. MAE from perturbation theory for the half-filled case, $N_e = 1$, as a function of the coupling strength to the ferromagnetic background and of the Rashba coupling strength. Solid lines are the numerically calculated internal energy differences $\Delta U = U(\pi/2, 0) - U(0, 0)$. Dashed lines are the corresponding combination of anisotropy coefficients in Eq. (23), using the analytical forms of Eq. (44). The vertical dotted lines mark the closing of the gap in the weak SOC limit. We plot the results in two ways. (a) The energy axis is scaled by $t''/16$, to factor out the expected dependence on the SOC strength. This shows that the maximum possible anisotropy for a given Rashba strength occurs when the band gap closes, as marked by the horizontal dotted line for the weak SOC limit. (b) The energy axis is scaled by the figure of merit $K_{\text{ref}}$ given in Eq. (46). The horizontal dotted line is the estimate $\Delta U = K_{\text{ref}}$. The vertical dotted line can be used to classify the deviations from this estimate: higher SOC contributions become important (to the right of this line), or the perturbation theory breaks down (to the left of this line).

and (iii) weak exchange ($t' \gg J_{\text{sd}} \sim t''$). We fix the SOC strength to be smaller than the non-relativistic bandwidth, by setting $\phi_R = \pi/20$ ($t' = 2.0t$ and $t'' = 0.3t$). The three case studies are then defined by how the exchange energy due to the ferromagnetic coupling compares to these two energy scales. We shall compare the local characterization of the MAE via the susceptibility with the global one using band energy differences. For the present model, the contribution to the MAE from the volume susceptibility (Eq. (31)) vanishes when $\mathbf{M} \parallel \hat{z}$, while it is the only non-vanishing contribution for $\mathbf{M} \parallel \hat{x}$.

First we consider the case where the exchange energy dominates, by setting $J_{\text{sd}} = 10t$. 


FIG. 4. MAE for the strong exchange case, $J_{sd} \gg t' \gg t''$. (a) Total DOS and number of electrons as a function of energy, for $M \parallel \hat{z}$. (b) MAE from the second derivatives of the band energy. For $M \parallel \hat{x}$ (Eq. 26), only $\chi^0$ contributes (Eq. (31)). For $M \parallel \hat{z}$ (Eq. 28), $\chi^0$ does not contribute, and we plot the intraband (Eq. (29)) and interband (Eq. (30)) contributions from the uniform spin susceptibility, as well as the net result. (c) Internal energy differences $U(M \parallel \hat{x}) - U(M \parallel \hat{z})$, decomposed using Eq. 17. The curve showing half of the difference in the SOC energy overlaps almost perfectly with the net internal energy differences, which in turn agrees very well with the results obtained from the susceptibility calculations, c.f. panel (b). Parameters: $J_{sd} = 10t$ and $\phi_R = \pi/20$ ($t' = 2.0t$, $t'' = 0.3t$).

This leads to two well-separated bands, as shown in Fig. 4(a). Fig. 4(b) estimates the MAE from the spin susceptibility, for two stable orientations of the ferromagnetic background. We see that for most values of $N_e$ we find IMA, with PMA in a narrow range around $N_e = 1$. When $M \parallel \hat{z}$ (Eq. 28), the interband contribution to the susceptibility (Eq. (30)) favors PMA, while the intraband contribution (Eq. (29)) favors IMA. The amplitude of the intraband contribution is larger than the interband one. PMA arises due to the different behavior when $N_e = 1$: the intraband contribution must vanish (the system is gapped), while the interband term remains finite. The sum of the two terms gives the net MAE for this configuration. When $M \parallel \hat{x}$ (Eq. 26), the volume susceptibility (Eq. (31)) is the only contribution present, and reproduces essentially the same MAE as found for $M \parallel \hat{z}$. This agreement shows that the higher-order anisotropy constants ($K_4$ and $K'_4$) are very small when compared with $K_2$, as anticipated from perturbation theory. Fig. 4(c) plots the MAE from the band energy difference between $M \parallel \hat{x}$ and $M \parallel \hat{z}$. The MAE from this approach is in perfect agreement with the one extracted from the susceptibility. Decomposing the band
energy into its constituents, see Eq. (17), we see that: (i) the anisotropy of the non-relativistic kinetic energy $\Delta U_0$ matches the intraband contribution to the susceptibility $\chi^{\text{intra}}(\mathbf{M} \parallel \hat{z})$, (ii) the anisotropy of the spin polarization energy $\Delta U_\text{p}$ matches the interband contribution to the susceptibility $\chi^{\text{inter}}(\mathbf{M} \parallel \hat{z})$, and (iii) half of the anisotropy of the Rashba energy $\Delta U_\text{R}/2$ matches the contribution from the volume susceptibility $\chi^0(\mathbf{M} \parallel \hat{x})$. We also verify that the MAE is given by half of the anisotropy in the SOC (Rashba) energy, as proposed in Refs. 21 and 22.

Next we consider the case where the exchange energy is comparable to the non-relativistic bandwidth, by setting $J_{sd} = t'$. Now the two bands overlap, as shown in Fig. 5(a), with the higher band beginning to fill for $N_e > 0.5$ ($E_F > -2t$), and the lower band being completely full for $N_e > 1.5$ ($E_F > 2t$). Fig. 5(b) estimates the MAE from the spin susceptibility, and shows that PMA is found in a much wider range of $N_e$ than in the previous case. Contrasting Eq. (29) and Eq. (30), we see that the Fermi sea term can be enhanced by reducing the $k$-dependent spin splitting $|b(k)|$, which we achieved by weakening $J_{sd}$, so that

FIG. 5. MAE for the intermediate exchange case, $J_{sd} \sim t' \gg t''$. (a) Total DOS and number of electrons as a function of energy, for $\mathbf{M} \parallel \hat{z}$. (b) MAE from the second derivatives of the band energy. For $\mathbf{M} \parallel \hat{x}$ (Eq. 26), only $\chi^0$ contributes (Eq. (31)). For $\mathbf{M} \parallel \hat{z}$ (Eq. 28), $\chi^0$ does not contribute, and we plot the intraband (Eq. (29)) and interband (Eq. (30)) contributions from the uniform spin susceptibility, as well as the net result. (c) Internal energy differences $U(\mathbf{M} \parallel \hat{x}) - U(\mathbf{M} \parallel \hat{z})$, decomposed using Eq. 17. The curve showing half of the difference in the SOC energy overlaps almost perfectly with the net internal energy differences, which in turn agrees very well with the results obtained from the susceptibility calculations, c.f. panel (b). Parameters: $J_{sd} = t'$ and $\phi_R = \pi/20$ ($t' = 2.0t$, $t'' = 0.3t$).
FIG. 6. MAE for the weak exchange case, $t' \gg J_{sd} \sim t''$. (a) Total DOS and number of electrons as a function of energy, for $M \parallel \hat{z}$. (b) MAE from the second derivatives of the band energy. For $M \parallel \hat{x}$ (Eq. 26), only $\chi^0$ contributes (Eq. (31)). For $M \parallel \hat{z}$ (Eq. 28), $\chi^0$ does not contribute, and we plot the intraband (Eq. (29)) and interband (Eq. (30)) contributions from the uniform spin susceptibility, as well as the net result. (c) Internal energy differences $U(M \parallel \hat{x}) - U(M \parallel \hat{z})$, decomposed using Eq. 17. The curve showing half of the difference in the SOC energy overlaps almost perfectly with the net internal energy differences, which in turn agrees very well with the results obtained from the susceptibility calculations, c.f. panel (b). Parameters: $J_{sd} = t''$ and $\phi_R = \pi/20$ ($t' = 2.0t$, $t'' = 0.3t$).

now the interband contribution dominates over the intraband one. However, near $N_e = 0$ (likewise near $N_e = 2$), the intraband contribution is linear in $N_e$ while the interband one is quadratic, so that the former can overtake the latter, and thus favors IMA. As already shown in Fig. 3, the MAE reaches only 20% of $K_{ref}$ at $N_e = 1$ (gapless system), in line with the discussion of Sec. IV. Fig. 5(c) plots the MAE from the band energy difference between $M \parallel \hat{x}$ and $M \parallel \hat{z}$ and its decomposition. Once again the band energy difference agrees very well with the results obtained from the susceptibility calculations, and with the estimate of $\Delta U_R/2$. The previous identifications between the intraband and interband contributions to the susceptibility and the anisotropies of the non-relativistic kinetic energy and of the spin polarization energy, respectively, are seen to hold only while one of the bands is either completely empty ($N_e < 0.5$) or completely full ($N_e > 1.5$). Although those two contributions to the energy exhibit a discontinuous behavior when both bands are partially filled, their sum is continuous, as can be concluded from $\Delta U_{total}$.

Lastly we consider the case where the exchange energy is comparable to the SOC strength,
by setting $J_{sd} = t''$. In this regime the splitting between the two bands is small, as seen in Fig. 6(a), as the bandwidth is mostly set by $t'$, and $t' \gg t'' \sim J_{sd}$. The intraband and interband contributions to the susceptibility are almost identical, Fig. 6(b), leading to a small net value of the MAE. Now we find PMA for almost all values of $N_e$, except at the band edges ($N_e \approx 0$ or 2) where IMA is recovered. For these limiting values of the filling the band dispersions can be approximated by the free-electron Rashba model, for which IMA is the expected result$^{24,25}$. Fig. 6(c) provides a better view of the behavior of the MAE, using the band energy difference between $\mathbf{M} \parallel \hat{x}$ and $\mathbf{M} \parallel \hat{z}$ and its decomposition. As found for the previous case, when both bands are partially filled there is no direct correspondence between the contributions to the band energy difference and the contributions to the susceptibility. Estimating the band energy difference by half of the anisotropy of the Rashba energy remains an excellent approximation, also in very good agreement with the results from the volume susceptibility.

VI. DISCUSSION AND CONCLUSIONS

In this work we explored a simple tight-binding model of spin-orbit-coupled electrons exchange-coupled to a background ferromagnetic order parameter, meant to abstract the essential electronic structure properties of the interface between a ferromagnetic layer and a heavy-metal layer. The simplicity of the model made it attractive to consider different approaches to the calculation of the magnetic anisotropy energy: a global approach, based on band energy differences, and a local approach, based on the curvature of the energy for an equilibrium ferromagnetic orientation. Besides reproducing the results of previous works$^{24,25}$, we provided a detailed view on how the competition between in-plane and perpendicular magnetic anisotropies is settled, by decomposing the spin susceptibility into intra and interband contributions, and connecting them to the anisotropy of different energy terms in the hamiltonian. Reassuringly, the global and local approaches to the magnetic anisotropy are found to be compatible, due to the weak higher-order anisotropy contributions. Perturbation theory was used to prove analytically that when the system is gapped perpendicular magnetic anisotropy always ensues.

We found that the perpendicular magnetic anisotropy can be enhanced by tuning the splitting of the energy bands to the point where the gap between them is about to close.
(besides the obvious path of increasing the magnitude of the spin-orbit coupling). This has the added advantage of increasing the range of filling values for which perpendicular magnetic anisotropy is present in the model. The impact of tuning the effective splitting of the energy bands at the interface between a ferromagnet and a heavy-metal can be explored, both with density functional theory calculations and experimentally, by inserting dopants or a decoupling layer at the interface. These studies would also uncover which are the generic features of the interface-driven magnetic anisotropy and which are the model-specific ones.

On the theoretical side, the calculation of the magnetic anisotropy energy from realistic band structures remains a challenging problem. The magnetic force theorem has been employed to replace the total energy difference between two self-consistent calculations for orthogonal directions of the magnetization by the corresponding difference in band energies, requiring only one self-consistent calculation\textsuperscript{15}. In a similar vein, the first derivative of the energy with respect to the orientation of the magnetization (the so-called magnetic torque) has also been effectively deployed\textsuperscript{20}. Here we proposed to utilize the static uniform spin susceptibility to obtain the curvature of the energy for an equilibrium orientation of the magnetization, which would require also only one self-consistent calculation. We also validated the proposal of van der Laan\textsuperscript{22} and Antropov\textsuperscript{21} to consider the anisotropy of the spin-orbit coupling energy term in the hamiltonian as an accurate approach to compute the magnetic anisotropy energy. These last two methods deserve further exploration within the context of realistic electronic structure calculations.

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Appendix A: Internal energy vs. grand potential

The properties of a system with a fixed number of electrons held at zero temperature can be derived from the internal energy, Eq. (16). Suppose the hamiltonian depends on a set of parameters \( X \), and we wish to find how the internal energy changes upon small changes in those parameters. The first derivative is

\[
\frac{\partial U}{\partial X_i} \bigg|_{N_e} = \frac{\partial U}{\partial X_i} \bigg|_{E_F} + \frac{\partial U}{\partial E_F} \frac{\partial E_F}{\partial X_i} = \int_{-\infty}^{E_F} dE \frac{\partial \rho(E, X)}{\partial X_i} E + \rho(E_F, X) E_F \frac{\partial E_F}{\partial X_i} . \tag{A1}
\]

The vertical bars indicate which variables are kept fixed. Using Eq. (14) and the requirement of fixed number of electrons, its derivative must be zero,

\[
0 = \frac{\partial N_e}{\partial X_i} = \frac{\partial N_e}{\partial X_i} \bigg|_{E_F} + \frac{\partial N_e}{\partial E_F} \frac{\partial E_F}{\partial X_i} = \int_{-\infty}^{E_F} dE \frac{\partial \rho(E, X)}{\partial X_i} + \rho(E_F, X) \frac{\partial E_F}{\partial X_i} , \tag{A2}
\]

so the first derivative of the internal energy can be rewritten as

\[
\frac{\partial U}{\partial X_i} \bigg|_{N_e} = \int_{-\infty}^{E_F} dE \frac{\partial \rho(E, X)}{\partial X_i} (E - E_F) . \tag{A3}
\]

This coincides with the first derivative of the grand potential,

\[
\Phi = \frac{1}{T=0} U - E_F N_e , \quad \frac{\partial \Phi}{\partial X_i} \bigg|_{E_F} = \frac{\partial U}{\partial X_i} \bigg|_{N_e} , \tag{A4}
\]

which is the expected thermodynamic result. The grand canonical ensemble is often used instead of the canonical one, as calculations tend to be simpler.

Starting from Eq. (A3), the second derivative of the internal energy is

\[
\frac{\partial^2 U}{\partial X_i \partial X_j} \bigg|_{N_e} = \int_{-\infty}^{E_F} dE \frac{\partial^2 \rho(E, X)}{\partial X_i \partial X_j} (E - E_F) - \frac{\partial E_F}{\partial X_i} \int_{-\infty}^{E_F} dE \frac{\partial \rho(E, X)}{\partial X_j} + \rho(E_F, X) \frac{\partial E_F}{\partial X_i} \frac{\partial E_F}{\partial X_j} . \tag{A5}
\]

We see that the second derivatives are related by a factor which is related to how the number of electrons changes upon variation of the parameters in the hamiltonian. This correction clearly vanishes for a gapped system (\( \rho(E_F, X) = 0 \)) or when varying the parameters leaves the Fermi energy unchanged.

Appendix B: Green functions and the Hellmann-Feynman theorem

For our purposes, the Green function is the resolvent of the hamiltonian,

\[
(E - \mathcal{H}(X)) G(E, X) = \mathcal{I} , \tag{B1}
\]
where the hamiltonian is assumed to depend on some parameters \( X \), and \( \mathcal{I} \) is the identity matrix for a chosen representation. Taking the derivate with respect to the energy parameter we find

\[
\frac{\partial G(E, X)}{\partial E} = -G(E, X) G(E, X),
\]

and with respect to a hamiltonian parameter we get

\[
\frac{\partial G(E, X)}{\partial X_i} = G(E, X) \frac{\partial H(X)}{\partial X_i} G(E, X).
\]

Using the Dirac identity we obtain the spectral density matrix from the discontinuity of the Green function across the real energy axis,

\[
\delta (E - H(X)) = -\frac{1}{2\pi i} \lim_{\eta \to 0^+} (G(E + i\eta, X) - G(E - i\eta, X)) \equiv -\frac{1}{\pi} \text{Im} G(E, X).
\]

The density of states of the system is then given by

\[
\rho(E, X) = -\frac{1}{\pi} \text{Im} \text{Tr} G(E, X),
\]

and its derivative with respect to a hamiltonian parameter by

\[
\frac{\partial \rho(E, X)}{\partial X_i} = -\frac{1}{\pi} \text{Im} \text{Tr} G(E, X) \frac{\partial H(X)}{\partial X_i} G(E, X) = \frac{1}{\pi} \text{Im} \text{Tr} \frac{\partial G(E, X)}{\partial E} \frac{\partial H(X)}{\partial X_i},
\]

using the cyclic property of the trace.

We can now replace these results in the first derivative of the internal energy, Eq. (A3),

\[
\left. \frac{\partial U}{\partial X_i} \right|_{N_e} = \frac{1}{\pi} \text{Im} \text{Tr} \int_{-\infty}^{E_F} dE \frac{\partial G(E, X)}{\partial E} \frac{\partial H(X)}{\partial X_i} (E - E_F)
\]

\[
= -\frac{1}{\pi} \text{Im} \text{Tr} \int_{-\infty}^{E_F} dE G(E, X) \frac{\partial H(X)}{\partial X_i} \equiv \left\langle \frac{\partial H(X)}{\partial X_i} \right\rangle_{E_F},
\]

after integration by parts. This is the Hellmann-Feynman theorem\textsuperscript{27,28}: the derivative of the energy with respect to a parameter is given by the ground state expectation value of the derivative of the hamiltonian with respect to the same parameter.

In Eq. (A2) we find

\[
\left. \frac{\partial N_e}{\partial X_i} \right|_{E_F} = \int_{-\infty}^{E_F} dE \frac{\partial \rho(E, X)}{\partial X_i} = \frac{1}{\pi} \text{Im} \text{Tr} G(E_F, X) \frac{\partial H(X)}{\partial X_i} \equiv -\left\langle \frac{\partial H(X)}{\partial X_i} \right\rangle_{E_F},
\]

and using this and Eq. (B3) we can express the second derivative of the internal energy, Eq. (A5), as

\[
\left. \frac{\partial^2 U}{\partial X_i \partial X_j} \right|_{N_e} = \left\langle \frac{\partial^2 H(X)}{\partial X_i \partial X_j} \right\rangle - \frac{1}{\pi} \text{Im} \text{Tr} \int_{-\infty}^{E_F} dE G(E, X) \frac{\partial H(X)}{\partial X_i} G(E, X) \frac{\partial H(X)}{\partial X_j} + \frac{1}{\rho(E_F, X)} \left\langle \frac{\partial H(X)}{\partial X_i} \right\rangle_{E_F} \left\langle \frac{\partial H(X)}{\partial X_j} \right\rangle_{E_F}.
\]
The last term must be omitted for a gapped system (no Fermi surface).

Appendix C: Anatomy of the static uniform susceptibility

In this appendix the expression for the susceptibility using Green functions, Eq. (21) (see also Eq. (B9)), is recast in the more familiar form from perturbation theory. We recall the spectral representation of the Green function, Eq. (18):

\[ G(k, E) = \sum_n \frac{P_n(k)}{E - E_n(k)} \quad , \quad P_n(k) = \frac{1}{2} \left( \sigma^0 + n \hat{b}(k) \cdot \sigma \right) , \quad n = \pm . \tag{C1} \]

We only have to rewrite the term involving the product of Green functions,

\[ \chi^{\alpha\beta} \big|_{E_F} = \frac{1}{\pi} \text{Im Tr} \int_{-\infty}^{E_F} \frac{dE}{(2\pi)^2} \sigma^\alpha G(k, E) \sigma^\beta G(k, E) \]

\[ = \sum_{n'n} \frac{1}{\pi} \text{Im Tr} \int_{-\infty}^{E_F} \frac{dE}{(2\pi)^2} \sigma^\alpha \frac{P_{n'}(k)}{E - E_{n'}(k)} \sigma^\beta \frac{P_n(k)}{E - E_n(k)} \] . \tag{C2} \]

To evaluate the energy integral we require the partial fraction decomposition of

\[ \frac{1}{E - E_{n'}(k)} \frac{1}{E - E_n(k)} = \frac{1}{E_{n'}(k) - E_n(k)} \left( \frac{1}{E - E_{n'}(k)} - \frac{1}{E - E_n(k)} \right) , \tag{C3} \]

which holds only if \( n \neq n' \) (interband contribution), and contributes simple poles to the energy integral:

\[ -\frac{1}{\pi} \text{Im Tr} \int_{-\infty}^{E_F} \frac{dE}{E - E_n(k)} = \Theta(E_F - E_n(k)) \equiv f_n(k) \] . \tag{C4} \]

When \( n' = n \) we have degeneracies (intraband term), which contribute a second-order pole and so have to be treated separately:

\[ -\frac{1}{\pi} \text{Im Tr} \int_{-\infty}^{E_F} \frac{dE}{(E - E_n(k))^2} = \frac{\partial f_n(k)}{\partial E_n(k)} = -\delta(E_F - E_n(k)) \] . \tag{C5} \]

The matrix elements are given by

\[ \mathcal{M}^{\alpha\beta}_{n'n}(k) = \text{Tr} \sigma^\alpha P_{n'}(k) \sigma^\beta P_n(k) \]

\[ = \frac{1 - n'n}{2} \delta_{\alpha\beta} + n'n \hat{b}_\alpha(k) \hat{b}_\beta(k) - i \frac{n}{2} \sum_{\gamma} \varepsilon_{\alpha\beta\gamma} \hat{b}_\gamma(k) \] , \tag{C6} \]

with \( \varepsilon_{\alpha\beta\gamma} \) the Levi-Civita symbol. We can then split the susceptibility as \( \chi^{\alpha\beta} = \chi^{\alpha\beta}_{\text{intra}} + \chi^{\alpha\beta}_{\text{inter}} + \bar{\chi} \delta_{\alpha\beta} \), with

\[ \chi^{\alpha\beta}_{\text{intra}} = \int \frac{d^2k}{(2\pi)^2} \hat{b}_\alpha(k) \hat{b}_\beta(k) \sum_{n = \pm} \delta(E_F - E_n(k)) - \frac{m^\alpha(E_F) m^\beta(E_F)}{\rho(E_F)} \] , \tag{C7} \]

\[ 23 \]
\[\chi_{\alpha\beta}^{\text{inter}} = 2 \int \frac{dk}{(2\pi)^2} \hat{b}_\alpha(k) \hat{b}_\beta(k) \frac{f_-(k) - f_+(k)}{E_-(k) - E_+(k)} , \quad (C8)\]

\[\bar{\chi} = -2 \int \frac{dk}{(2\pi)^2} \frac{f_-(k) - f_+(k)}{E_-(k) - E_+(k)} . \quad (C9)\]

The intraband term collects the contributions from the Fermi energy, while the interband term collects those from the Fermi sea. \(\bar{\chi}\) is also an interband contribution, but splitting it off simplifies the equations in the main text and makes the interpretation of the results more straightforward. The contribution of the antisymmetric part of the matrix element to the interband term cancels out.

1. **Ferromagnetic system without spin-orbit coupling**

For this example we can take \(\hat{b}(k) = S = \hat{z}\) without loss of generality, as without SOC the system is invariant under spin rotations. The energy dispersion of Eq. (11) becomes

\[E_n(k) = E_0(k) - nJ_{sd} \quad , \quad n = \pm , \quad (C10)\]

and the matrix elements simplify to

\[M_{\alpha\beta}^{n' n}(k) = \delta_{\alpha\beta} \frac{1 - n'n}{2} + n'n \delta_{\alpha z} \delta_{\beta z} - i \varepsilon_{\alpha\beta z} \frac{n' - n}{2} . \quad (C11)\]

The longitudinal susceptibility \((\alpha = \beta = z)\) arises from the intraband contributions \((n' = n)\), while the transverse susceptibility \((\alpha, \beta = x, y)\) arises from the interband contributions \((n' \neq n)\). From Eq. (C7), the longitudinal susceptibility is thus

\[\chi^{zz} = \rho_+(E_F) + \rho_-(E_F) + \frac{(\rho_+(E_F) - \rho_-(E_F))^2}{\rho(E_F)} = \frac{4\rho_+(E_F)\rho_-(E_F)}{\rho(E_F)} , \quad (C12)\]

with \(\rho_n(E_F)\) the density of states at the Fermi energy of the \(n\)-band (check Eq. (13)). Here the correction term is crucial: if one band is partially occupied, \(\rho_+(E_F) \neq 0\), and the other band is empty, \(\rho_-(E_F) = 0\), then \(\chi^{zz} = 0\), as the increase in the spin moment (the \(\rho_+(E_F)\) contribution from the first term) is cancelled by the requirement of fixed number of electrons (enforced by the correction term). From Eq. (C8), the transverse susceptibility is (check Eq. (15))

\[\chi^{xx} = \chi^{yy} = -\int \frac{dk}{(2\pi)^2} \frac{f_-(k) - f_+(k)}{J_{sd}} = \frac{M}{J_{sd}} . \quad (C13)\]
This cancels precisely the volume susceptibility, $\chi^0$, and makes the derivatives of the internal energy with respect to the angles defining the ferromagnetic direction vanish, Eqs. (26), (27) and (28). As discussed in Sec. III, this term is also present in the general case both in the transverse susceptibilities and in the volume susceptibility, and so those quantities are defined in the main text by analytically subtracting this term from both of them.

2. Gapped system at half-filling with $S = \hat{z}$

Now the Fermi energy lies in the gap, so one of the bands is fully occupied, $f_+(k) = 1$, and the other is empty, $f_-(k) = 0$. Thus there are no intraband contributions to the susceptibility and the Fermi surface corrections vanish. From Eq. (C8) and inserting the band dispersions of Eq. (11), the susceptibility is then

$$\chi^{\alpha\beta} = \int \frac{dk}{(2\pi)^2} \frac{\delta_{\alpha\beta} - \hat{b}_\alpha(k) \hat{b}_\beta(k)}{|b(k)|}. \quad (C14)$$

The longitudinal susceptibility is

$$\chi^{zz} = \int \frac{dk}{(2\pi)^2} \frac{1 - (\hat{b}_z(k))^2}{|b(k)|} = \int \frac{dk}{(2\pi)^2} \frac{(\hat{b}_x(k))^2 + (\hat{b}_y(k))^2}{|b(k)|} \neq 0, \quad (C15)$$

which shows that the net spin moment is not saturated, due to SOC. Using Eq. (9) we find

$$\chi^{zz} = \int \frac{dk}{(2\pi)^2} \frac{(t'')^2 (\sin^2 k_x + \sin^2 k_y)}{\left(\left(t''\right)^2 (\sin^2 k_x + \sin^2 k_y) + (J_{sd})^2\right)^{\frac{3}{2}}}$$

$$= -t'' \frac{\partial}{\partial t''} \int \frac{dk}{(2\pi)^2} \left(\left(t''\right)^2 (\sin^2 k_x + \sin^2 k_y) + (J_{sd})^2\right)^{-\frac{3}{2}}$$

$$\approx \frac{t''}{J_{sd}} \frac{\partial}{\partial t''} \int \frac{dk}{(2\pi)^2} \left(\frac{(t'')^2}{2(J_{sd})^2} (\sin^2 k_x + \sin^2 k_y) - \frac{3(t'')^4}{8(J_{sd})^4} (\sin^2 k_x + \sin^2 k_y)^2\right)$$

$$= \frac{(t'')^2}{(J_{sd})^3} \left(\frac{\sin^2 k_x + \sin^2 k_y}{2(J_{sd})^2} - \frac{3(t'')^2}{2(J_{sd})^2} (\sin^2 k_x + \sin^2 k_y)^2\right)$$

$$= \frac{(t'')^2}{(J_{sd})^3} \left(1 - 15(t'')^2\right) \quad (C16)$$

The generating polynomial of Eq. (41) was used to systematically evaluate the integrals.
The transverse susceptibility is

\[
\chi^{xx} = \int \frac{dk}{(2\pi)^2} \frac{1 - (\hat{b}_x(k))^2}{|b(k)|} = \int \frac{dk}{(2\pi)^2} \frac{(\hat{b}_y(k))^2 + (\hat{b}_z(k))^2}{|b(k)|}
\]

\[
= \int \frac{dk}{(2\pi)^2} \frac{(t'')^2 \sin^2 k_x + (J_{sd})^2}{(t'')^2 \sin^2 k_x + (J_{sd})^2 + (J_{sd})^2} \]

\[
= \int \frac{dk}{(2\pi)^2} \frac{(t'')^2 \sin^2 k_x + (J_{sd})^2}{(t'')^2 \sin^2 k_x + (J_{sd})^2 + (J_{sd})^2} \]

\[
= - \left( \frac{t''}{2} \frac{\partial}{\partial t''} + J_{sd} \frac{\partial}{\partial J_{sd}} \right) \int \frac{dk}{(2\pi)^2} \left( (t'')^2 (\sin^2 k_x + \sin^2 k_y) + (J_{sd})^2 \right)^{-\frac{1}{2}} . \quad (C17)
\]

The symmetry of the integrand allows the replacement shown on the third line, and in turn shows that \( \chi^{yy} = \chi^{xx} \). Substituting the results from the longitudinal susceptibility,

\[
\chi^{xx} = \frac{1}{2} \chi^{zz} - J_{sd} \frac{\partial \chi^0}{\partial J_{sd}} = \frac{1}{2} \chi^{zz} - \frac{\partial M}{\partial J_{sd}} + \chi^0 = -\frac{1}{2} \chi^{zz} + \chi^0 . \quad (C18)
\]

These identifications follow from the expression for the longitudinal uniform susceptibility and from the definition of the spin moment, Eq. (15), which under the present assumptions leads to the volume susceptibility

\[
\chi^0 \equiv \frac{M}{J_{sd}} = \int \frac{dk}{(2\pi)^2} \frac{\hat{b}_z(k)}{J_{sd}} = \int \frac{dk}{(2\pi)^2} \frac{1}{|b(k)|} . \quad (C19)
\]

Using the expansion found for the longitudinal susceptibility,

\[
\chi^0 \approx \int \frac{dk}{(2\pi)^2} \left( \frac{(t'')^2}{2(J_{sd})^2} (\sin^2 k_x + \sin^2 k_y) - \frac{3(t'')^4}{8(J_{sd})^4} (\sin^2 k_x + \sin^2 k_y)^2 \right)
\]

\[
= \frac{(t'')^2}{2(J_{sd})^2} \left( 1 - \frac{15(t'')^2}{16(J_{sd})^2} \right) , \quad (C20)
\]

and from Eq. (28) we obtain the uniaxial magnetic anisotropy coefficient,

\[
\frac{\partial^2 U}{\partial \theta^2} \bigg|_{M/\hat{z}} = \frac{1}{2} (J_{sd})^2 \chi^{zz} = \frac{1}{2} \frac{(t'')^2}{J_{sd}} - \frac{15}{16} (J_{sd})^4 = 2K_2 , \quad (C21)
\]

in perfect agreement with the direct calculation of Sec. IV.
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