



Noncollinear Magnetism in Ultrathin Films with Strong Spin-Orbit Coupling from *Ab Initio*

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Fe and Pt are paradigms for ferromagnetism and strong spin-orbit coupling, respectively. Their combination—in an ultrathin Fe film on a Pt(111) substrate—is thus expected to modify the magnetic structures. We report on a theoretical investigation of a monolayer of Fe on Pt(111), using a generalized Heisenberg model that includes the complete spin interaction matrices I_{ij} computed from first principles. We find a noncollinear periodic configuration that is strongly determined by the Dzyaloshinskii-Moriya interaction. Taking into account a magnetic field to mimic recent experiments, this noncollinear structure solves the disagreement between the experimental magnetization and the average magnetization for a ferromagnetic system. The critical temperature decreases from 670 K to 590 K due to spin-orbit coupling.

Keywords: Dzyaloshinskii-Moriya Interaction, Ultrathin Films, Magnetic Ground State Properties.

1. MOTIVATION

The properties of ultrathin magnetic films depend strongly on the substrate,¹ as has been shown experimentally by spin-polarized scanning tunneling microscopy and magneto-optical Kerr spectroscopy. In particular spin-orbit coupling (SOC) induces a symmetry breaking in magnetic systems, which e.g., shows up as magnetocrystalline anisotropy. Together with the exchange interaction, the magnetic anisotropy manifests itself in the formation of domain walls. In non-centrosymmetric systems, the Dzyaloshinskii-Moriya (DM) interaction results in non-collinear magnetic structures, preferably at surfaces and in ultrathin films.² The magnetic properties of a monolayer Fe on Pt(111) are still not understood completely. On one hand, Moulas et al.³ showed that by applying an external magnetic field of 5 T the magnetization is barely $1.2 \mu_B$; the saturation field is roughly estimated to 10 T. On the other hand, first-principles electronic-structure calculations using a Korringa–Kohn–Rostoker Green’s function method³ or the Vienna *Ab-Initio* Simulation Package⁴ support ferromagnetic order, an Fe magnetic moment of about $3.0 \mu_B$ and an induced Pt moment of $0.25 \mu_B$. We regard these contradictory results as an indication for a noncollinear magnetic structure in Fe/Pt(111) which may be driven by the strong SOC in Pt. Noncollinear magnetism due to SOC has been shown for FePt-alloy clusters

deposited on Pt(111).^{5,6} In this paper we report on a first-principles investigation of a monolayer Fe on Pt(111) that includes SOC. Using a generalized Heisenberg model, we focus on the question on how spin-orbit coupling manifests itself in the magnetic ground-state properties and the magnetic structure of Fe/Pt(111).

2. OUTLINE OF THE THEORETICAL APPROACH

2.1. First-Principles Calculations

To investigate the magnetic properties of a monolayer Fe on Pt(111), we performed first-principles electronic-structure calculations using a relativistic Korringa–Kohn–Rostoker Green’s function method.^{7,8} Solving the Dirac equation for a spin-polarized system, spin-orbit coupling and magnetism are treated on equal footing. Wavefunctions and scattering matrices have been calculated up to an orbital momentum of $l_{\max} = 3$. The site-dependent potentials are described in the atomic sphere approximation. The film-substrate system is taken as translationally invariant parallel to the layers but semi-infinite perpendicular to the layers. We adopt the interlayer spacing derived by Hardrat et al.¹ who found an inward relaxation of the monolayer fcc Fe by 12.7% of the Pt bulk interlayer distance (lattice constant $a = 2.81 \text{ \AA}$; Fig. 1).

From the *ab initio* calculations, we derived the complete spin interaction matrix of a generalized Heisenberg

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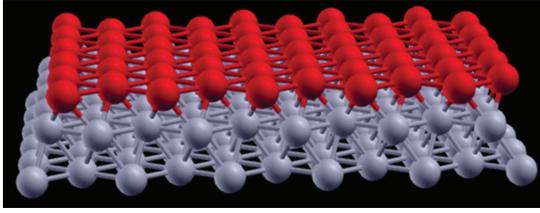


Fig. 1. Schematic view of a monolayer Fe on Pt(111). Only the first two layers of Pt (gray spheres) are important for the spin structure in the Fe film (red spheres).

model,⁷ within the framework of the magnetic force theorem.^{9–14}

2.2. Spin Hamiltonian and Interaction Matrix

The Heisenberg model

$$H = - \sum_{i \neq j} J_{ij} \mathbf{m}_i \cdot \mathbf{m}_j \quad (1)$$

where \mathbf{m}_i and \mathbf{m}_j are classical normalized magnetic moments and J_{ij} is their exchange constant, is a standard model for magnetic materials. Since the J_{ij} are isotropic and symmetric, it does not account for spin-orbit coupling effects. A straightforward generalization is given by

$$H = - \sum_{ij} \mathbf{m}_i I_{ij} \mathbf{m}_j \quad (2)$$

where I_{ij} is the full 3×3 interaction matrix ($I_{ij} = I_{ij}^T$). Decomposing I_{ij} as

$$I_{ij} \equiv J_{ij} \mathbf{1} + S_{ij} + A_{ij} \quad (3)$$

one recovers the Heisenberg exchange $J_{ij} = (1/3) \text{tr} I_{ij}$. The anisotropic and traceless parts S_{ij} and A_{ij} split into a symmetric and an antisymmetric contribution, $S_{ij} = (1/2) \cdot (I_{ij} + I_{ij}^T) - J_{ij} \mathbf{1}$ and $A_{ij} = (1/2)(I_{ij} - I_{ij}^T)$. This implies that only the J_{ij} force the system into a collinear state. The anisotropic antisymmetric components include the Dzyaloshinskii-Moriya vectors

$$D_{ij}^\alpha \equiv \frac{1}{2} \sum_{\beta\gamma} \varepsilon_{\alpha\beta\gamma} I_{ij}^{\beta\gamma}, \quad \alpha, \beta, \gamma = x, y, z \quad (4)$$

that describe the noncollinearity between two magnetic moments;¹⁵ they are closely linked to the symmetry of the system.

The complete Hamiltonian then reads

$$H = - \sum_{i \neq j} [J_{ij} \mathbf{m}_i \cdot \mathbf{m}_j + \vec{D}_{ij} \cdot (\mathbf{m}_i \times \mathbf{m}_j) + \mathbf{m}_i S_{ij} \mathbf{m}_j] - \sum_i \mathbf{m}_i I_{ii} \mathbf{m}_i - \sum_{ij} \mathbf{m}_i Q_{ij} \mathbf{m}_j + H_{\text{ext}} \quad (5)$$

The fourth term correlates to the uniaxial anisotropy of the system, whereas the fifth term accounts for the

dipole–dipole interaction (shape anisotropy). The dipolar matrix Q_{ij} is given by

$$Q_{ij}^{\mu\nu} = \frac{\mu_0}{8\pi} \frac{3r_{ij}^\mu r_{ij}^\nu - r_{ij}^2 \delta^{\mu\nu}}{r_{ij}^5}, \quad r_{ij} = r_i - r_j; \quad \mu, \nu = x, y, z \quad (6)$$

where r_i is the location of the atom i . Eventually, an external magnetic field is described by the Zeeman term in H_{ext} . The magnetic properties of the systems in thermal equilibrium are obtained by a standard Monte-Carlo method^{16,17} using the Metropolis algorithm.

3. RESULTS AND DISCUSSION

3.1. Spin-Resolved Electronic Properties

The electronic-structure calculations yield that the majority bands in the Fe layer are almost completely filled, unlike the minority bands that show a sharp maximum at the Fermi level (Fig. 2). As a consequence of the reduced dimensionality of the film, the Fe magnetic moment ($3.03 \mu_B$) is increased with respect to Fe bulk ($2.26 \mu_B$) and agrees with that given by Hardrat et al.¹ ($3.10 \mu_B$). Hybridization of Fe and Pt electronic states results in induced Pt magnetic moments that decrease rapidly toward the bulk (first Pt-layer $0.26 \mu_B$; second Pt-layer $0.01 \mu_B$; Fig. 2). This is a first hint on that the Pt substrate, with its large spin-orbit coupling, can indeed have a profound influence on the magnetic structure in the Fe adlayer.

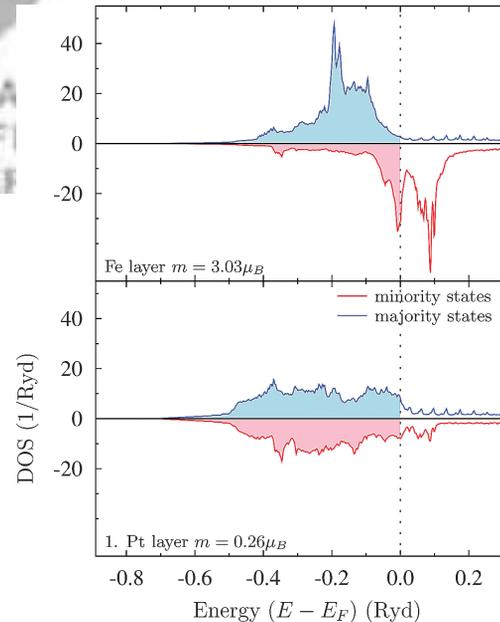


Fig. 2. Spin-resolved electronic structure of a monolayer Fe on Pt(111). The density of states (DOS) is given for the Fe layer (top) and the first Pt layer (bottom). The strong exchange splitting in Fe ($3.03 \mu_B$) and the hybridization of Fe with Pt electronic states leads to a sizable induced moment in the first Pt layer ($0.26 \mu_B$).

3.2. Magnetic Configuration of a Monolayer Fe on Pt(111)

In the Monte Carlo simulations for the magnetic ground state we consider the first three layers (Fe layer and the two subsequent Pt layers) because only these show relevant magnetic moments and spin interactions.

First, we discuss the effects of the individual contributions in the spin Hamiltonian on the magnetic configurations. The nearest-neighbor Heisenberg exchange constant exhibits ferromagnetic behavior ($J_{\text{INN}} > 0$); the magnetocrystalline anisotropy is in-plane (parallel to the surface). Hence, without the DM contribution, we obtain an in-plane collinear ground state with an average atomic moment of $1.59 \mu_{\text{B}}$, in clear contrast to experiment ($1.0 \mu_{\text{B}}$ from Ref. [3]).

Inclusion of the symmetric anisotropic part S_{ij} in the Hamiltonian maintains this spin configuration in general. In contrast to the DM part, this term tends to tilt pairs of magnetic moments in the same direction. Hence it can be interpreted as an additional contribution to the magnetocrystalline anisotropy that slightly changes the easy axis for each pair of moments. As a consequence, the local magnetic moments deviate more strongly from the general easy axis than without S_{ij} , even at very low temperatures. These fluctuations of the easy axes can be viewed as a broadening of the global energy minimum that also gives rise to fluctuations of the transition region width w .

The Dzyaloshinskii-Moriya contribution turns out to be significant only for nearest neighbor sites, as was deduced by successively reducing the interaction range in the MC calculations; it tends to tilt magnetic moments mutually and is strongly linked to the symmetry of the system (Fig. 3). It produces as an outcome of our calculations a periodic noncollinear configuration which shows up in both the Fe layer and the subsurface Pt layers (Fig. 4; not shown for Pt); this corroborates the significant coupling of Fe and Pt found in the density of states (Fig. 2). Since the magnetic structure contains in-plane and perpendicular components, it reminds at a combination of (very narrow) Bloch and Néel walls.

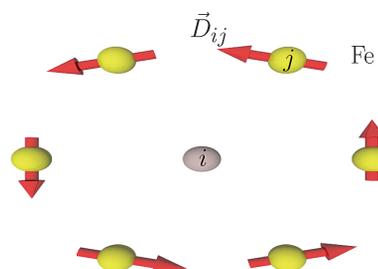


Fig. 3. Dzyaloshinskii-Moriya interaction in a monolayer Fe on Pt(111). Arrows depict the DM vectors D_{ij} between two magnetic moments i (central site, fixed) and j in the Fe adlayer. Because of the three-fold symmetry, DM contributions from an in-plane ring structure, with antisymmetric interactions with respect to site i .

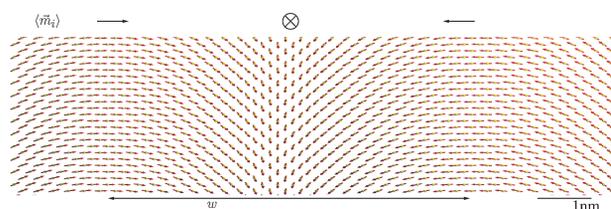


Fig. 4. Noncollinear magnetism in a monolayer Fe on Pt(111). Because of strong spin-orbit coupling effects, the local magnetic moments (arrows) form a periodic noncollinear magnetic structure. The moment averaged over one magnetic unit cell vanishes, but the moment averaged over the length w of the transition range is about $1.0 \mu_{\text{B}}$.

An impression of the interplay of the individual contributions to the Hamiltonian is provided by ratios $J:K:S:D$. We obtain for the Fe–Fe interaction 75:1:9:16, in contrast to the Fe–Pt interaction 8:1:1:2. While in both cases the Heisenberg exchange dominates, the ratio $J:D$ is almost identical for Fe–Fe and Fe–Pt (about 8:1), giving further support to the importance of the SOC and hybridization. The magnetic lattice constant can be estimated from domain-wall theory within a continuum model. Without DM interaction, the width w of the transition region is given by

$$w = \sqrt{\frac{A}{K}} \quad (7)$$

where A and K are the exchange density and the average anisotropy, respectively. With DM contribution, an analytical solution for w has been achieved only for DM vectors aligned along the z -direction, enlarging the transition region w . From the calculated parameters we obtain $w = 3.8 \text{ nm}$, which is slightly smaller than half of the magnetic lattice constant (Fig. 4). Taking into account the DM contribution, w is increased to 6.7 nm . A closer analysis of the MC simulations yields that this increase can indeed be attributed to the DM contributions.

Averaging over the magnetic unit cell gives a vanishing net magnetization. Restricting the average to the transition region, as indicated by the double arrow in Figure 4, produces a net moment of about $1.0 \mu_{\text{B}}$.

The perpendicular components of the local moments are very well described by an arithmetic function. Further, the symmetric anisotropic interaction leads to tiny fluctuations of the magnetic lattice constant; these are attributed to the anisotropic symmetric contributions to the exchange matrices.

We now focus on the magnetic phase transition. Since a noncollinear configuration is not well characterized by its average magnetization, we deduce the critical temperature from the nearest-neighbor spin correlation function

$$S = \frac{1}{N} \sum_i \frac{1}{N_i} \sum_j |\mathbf{m}_i \cdot \mathbf{m}_j| \quad (8)$$

Here, N is the number of sites in the sample and N_i the number of nearest neighbor atoms of site i . An alternative

measure is the spin–spin correlation function (SSCF)

$$s(dr) = \langle \mathbf{m}_r \cdot \mathbf{m}_{r+dr} \rangle_r \quad (9)$$

of magnetic moments at a distance dr . The SSCF of Fe/Pt(111) behaves like $\cos(\pi dr/w)$ at very low temperatures. For increasing thermal fluctuations, its amplitude decreases and eventually vanishes at the Curie temperature T_C . From this general behavior we derive Curie temperatures of 670 K without DM interaction. With DM interaction, the Curie temperature is reduced to 590 K which is in good agreement with 0.6 of T_C for bulk Fe (1043 K), as found by Rausch and Nolting.¹⁸

Eventually, we consider the dependency of the average magnetization on an external magnetic field perpendicular to the surface. We recall that a magnetization of $1.2 \mu_B$ has been found experimentally at 5 T; even at 10 T the sample was not driven into saturation.³ To mimic these experiments we performed MC simulations with the magnetic-field term (up to 10 T). As consequence of the strong SOC, the local magnetic moments maintain sizable in-plane components (cf. Fig. 4 for zero field) but are tilted out-of-plane with increasing field strength. For a field of 5 T we obtain a net magnetization of $1.3 \mu_B$. Also at 10 T, the local magnetic moments are not completely rotated out-of-plane, indicating incomplete saturation. Since these findings agree nicely with experiment,³ we conclude that the theoretically predicted noncollinear structure solves the aforementioned puzzle.

4. CONCLUSION

Ultrathin films with strong spin-orbit coupling show noncollinear spin structures, as is demonstrated for a monolayer Fe on Pt(111). Using Monte-Carlo calculations for a generalized Heisenberg model, in which spin-orbit contributions are taken into account and whose parameters are obtained from first-principles calculations, we find a periodic arrangement of toroidal structures. This noncollinear structure solves the discrepancy of the experimental magnetization and the theoretical magnetization for a ferromagnetic configuration. Our findings call for new experiments in order to verify the predicted magnetic structure.

We expect a strong impact of the spin-orbit interaction on time-dependent phenomena, as is currently investigated within an atomistic approach based on the stochastic Landau–Lifshitz–Gilbert equation. First calculations for systems perturbed by a magnetic field pulse show long-term excitations which may excite magnons.

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