Magnetic order at surfaces beyond the Heisenberg model

Spin structures at surfaces driven by higher-order exchange interactions

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In Short

- Higher-order exchange interactions beyond pair wise Heisenberg exchange can play an important role for magnetism at surfaces.
- Novel spin structures can occur which are interesting with respect to hybrid structures with superconductors or as the origin of topological orbital magnetization.
- We use density functional theory in combination with atomistic spin models to explore the effect of higher-order exchange interactions for ultrathin films studied experimentally by our collaborators via spin-polarized scanning tunneling microscopy.

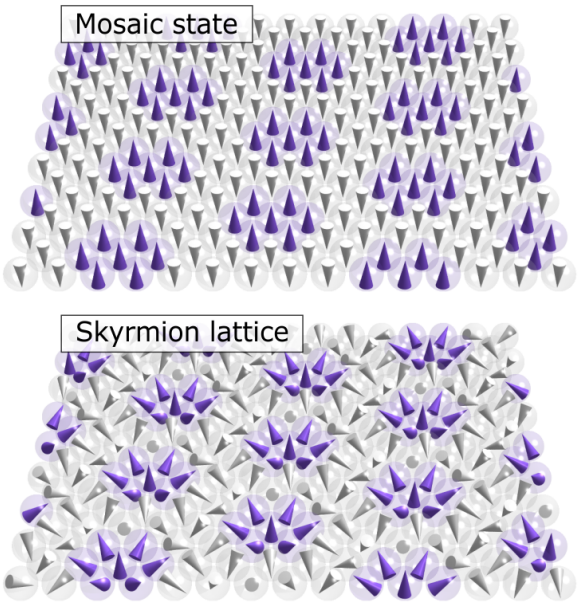

***Figure 1:** The image shows the different orientation of the magnetic moments in an Fe monolayer: In a magnetic mosaic state (above), they are oriented in groups either upwards (purple) or downwards (white). In the skyrmion lattice (below), on the other hand, they point in all directions. The mosaic state is stabilized by four spin exchange in Fe/Rh films on the Ir(111) surface [18].*

The magnetic ground state of a material is governed by the magnetic interactions between atomic magnetic moments. In addition to the Heisenberg pairwise exchange interactions also higher-order exchange interactions can play a role, where exchange between more than two sites is involved [1]. In complex three-dimensional spin structures on the nanometer scale such interactions can be the decisive factor for the magnetic ground state. While several spin structures at surfaces stabilized by higher-order terms are known [2–6], the understanding of these interactions is still very limited. Recently, an additional higher-order exchange interaction was proposed theoretically [7] in order to understand discrepancies between results from density functional theory (DFT) and previously used atomistic spin models [8]. A realistic spin model, that describes the magnetic interactions accurately, is indispensable for further investigations regarding predictions for materials with tailored properties, such as magnetic ground state, phase transitions, thermodynamical stability, and spin dynamics.

In this project we aim to gain deeper insight into the role of higher-order interactions at transition-metal interfaces. We study thin magnetic films on single crystal surfaces which serve as model systems to understand the microscopic origin of complex magnetic order and the involved spin interactions. In our approach we combine DFT calculations and atomistic spin dynamics simulations. We collaborate with the experimental group of Prof. R. Wiesendanger, in particular, with Dr. Kirsten von Bergmann who is an expert in spin-polarized scanning tunneling microscopy. Our goal is to discover novel spin structures driven by higher-order exchange interactions, to identify their origin and to establish a suitable atomistic spin model. We anticipate that the fundamental understanding of the role and occurrence of higher-order interactions obtained here will also be relevant beyond complex magnetic ground states in monolayers, e.g. in the context of hybrid systems where non-collinear magnetic order is in contact with superconductors or is the source of topological orbital magnetization [9–11].

We use DFT calculations to obtain total energies for a large number of collinear and non-collinear spin structures including spin-orbit coupling. From these first-principles calculations we can parametrize an atomistic spin model including pair-wise Heisenberg exchange, the Dzyaloshinskii-Moriya interaction as well as higher-order terms such as the biquadratic interaction and the four-spin interactions [12]. Atomistic spin dynamics simulations using our in-house developed Kiel code (as e.g. in [13,14]) are applied to find new magnetic ground states based on this DFT parametrized atomistic spin model.

We have shown with this approach that the stability of topological spin structures such as skyrmions

or antiskyrmions can be drastically enhanced due to higher-order exchange interactions [14]. This allows the stabilization of such spin structures even in the absence of DMI. In particular, the four-site four spin interaction plays a crucial role and depending on its sign it can lead to an increase or decrease of the energy barrier protecting a skyrmion or antiskyrmion against the collapse to the ferromagnetic state.

We have also explained the recent discovery of the triple-Q state in a Mn monolayer on the Re(0001) surface [15]. We have shown that the triple-Q state is actually distorted due to topological-chiral magnetic interactions [16] which were only recently proposed [17]. The reduced symmetry of the distorted 3Q state facilitates an effective coupling of the spin state to the atomic lattice, e.g. via the easy in-plane magnetic anisotropy of Mn/Re(0001), which can explain the experimental observations [15].

Recently, we have demonstrated that – in contrast to previous experimental findings and theoretical considerations – higher-order exchange interactions can induce spontaneous nano-scale two-dimensional multi-Q states with collinear spin structure (Fig. 1). Using spin-polarized scanning tunneling microscopy our experimental collaborators have revealed the magnetic ground states of hexagonal Fe/Rh films with different stacking sequence and thickness on the Ir(111) surface [18]. By the combination of DFT and an atomistic spin model we show that these complex spin structures originate from the delicate interplay of frustrated pair-wise and higher-order exchange while the Dzyaloshinskii-Moriya interaction (DMI) is weak. In particular, we find that the only recently proposed three-site four spin interaction plays a key role for the formation of collinear multi-Q states [18].

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More Information

[1] M. Takahashi, *J. Phys. C Solid State Phys.* **10**, 1289 (1977).

[2] P. Kurz, G. Bihlmayer, K. Hirai, and S. Blügel, *Phys. Rev. Lett.* **86**, 1106 (2001).

[3] S. Heinze, K. von Bergmann, M. Menzel, J. Brede, A. Kubetzka, R. Wiesendanger, G. Bihlmayer, and S. Blügel, *Nat. Phys.* **7**, 718 (2011).

[4] Y. Yoshida, S. Schröder, P. Ferriani, D. Serrate, A. Kubetzka, K. von Bergmann, S. Heinze, and R. Wiesendanger, *Phys. Rev. Lett.* **108**, 087205 (2012).

[5] A. Krönlein, M. Schmitt, M. Hoffmann, J. Kemmer, N. Seubert, M. Vogt, J. Küspert, M. Böhme, B. Alonazi, J. Kügel, H.A. Albrithen, M. Bode, G. Bihlmayer, and S. Blügel, *Phys. Rev. Lett.* **120**, 207102 (2018).

[6] N. Romming, H. Pralow, A. Kubetzka, M. Hoffmann, S. von Malottki, S. Meyer, B. Dupé, R. Wiesendanger, K. von Bergmann, and S. Heinze, *Phys. Rev. Lett.* **120**, 207101 (2018).

[7] M. Hoffmann and S. Blügel, *Phys. Rev. B* **101** 024418 (2020).

[8] A. Al-Zubi, G. Bihlmayer, and S. Blügel, *Phys. Status Solidi B* **248**, 2242 (2011).

[9] M. Hoffmann, J. Weischenberg, B. Dupé, F. Freimuth, P. Ferriani, Y. Mokrousov, and S. Heinze, *Phys. Rev. B* **92** 020401 (R) (2015).

[10] J.-P. Hanke, F. Freimuth, A. K. Nandy, H. Zhang, S. Blügel, and Y. Mokrousov, *Phys. Rev. B* **94**, 121114 (R) (2016).

[11] S. Nakosai, Y. Tanaka, and N. Nagaosa, *Phys. Rev. B* **88**, 180503 (R) (2013).

[12] M. Gutzeit, S. Haldar, S. Meyer, and S. Heinze, *Phys. Rev. B* **104**, 024420 (2021).

[13] S. Haldar, S. von Malottki, S. Meyer, P. F. Bessarab, and S. Heinze, *Phys. Rev. B* **98** 060413 (R) (2018).

[14] S. Paul, S. Haldar, S. von Malottki, and S. Heinze, *Nat. Commun.* **11**, 4756 (2020).

[15] J. Spethmann, S. Meyer, K. von Bergmann, R. Wiesendanger, S. Heinze, and A. Kubetzka, *Phys. Rev. Lett.* **124**, 227203 (2020).

[16] S. Haldar, S. Meyer, A. Kubetzka, and S. Heinze, *Phys. Rev. B* **104**, L180404 (2021).

[17] S. Grytsiuk, J.-P. Hanke, M. Hoffmann, J. Bouaziz, O. Gomonay, G. Bihlmayer, S. Lounis, Y. Mokrousov, and S. Blügel, *Nat. Commun.* **11**, 511 (2020).

[18] M. Gutzeit, A. Kubetzka, S. Haldar, H. Pralow, M. A. Goerzen, R. Wiesendanger, S. Heinze, and K. von Bergmann, *Nat. Commun.* **13**, 5764 (2022).

Project Partners

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