First principles simulations of magnetic nanostructures

PhD thesis booklet

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Background of the research

Development of experimetal tools, in particular spin-polarized scanning tunnelling microscopy, (SP-STM) made it possible to explore non-collinear magnetic structures in atomic resolution. [Bode et al., 2007]

Magnetic properties of transition metal systems are often described by effective spin-models. [Nowak, 2007] In nanoclusters the symmetry of the system is lower compared to higher dimensionality structures and there are many different coupling parameters and higher order spin-interactions might also be present. [Antal et al., 2008] In the case of a magnetic overlayer the inversion symmetry is broken which results in a non-vanishing Dzyaloshinsky-Moriya (DM) interaction. The presence of the DM interaction may lead to formation of exotic magnetic patterns such as magnetic skyrmions. [Heinze et al., 2011] *Ab initio* calculations on magnetic nano-structures can help a lot to interpret the experimental results more clearly and to attain better understanding of the underlying physical phenomena.

Objectives

In this work, magnetic nanostructures deposited onto magnetic or non-magnetic substrate are investigated theoretically based on first principles calculations. In the studied systems spin-orbit coupling (SOC) plays an important role. Fully relativistic electronic structure calculations are used, therefore, the SOC is taken into account in a non-perturbative way. It is demonstrated how the relativistic effects lift the degeneracy of the non-relativistic ground state.

In Thesis Statement 1, a demonstration is provided that a fully *ab initio* based Monte Carlo (MC) simulation of the magnetic structure of a nanocluster can be performed. An effort is made to compare the first principles description to an appropriate spin-model description.

The structure of a domain wall through a ferromagnetic nanocontact shows interesting behaviour. The measurements of Calvo *et al.* [Calvo et al., 2009] and Autès *et al.* [Autès et al., 2008] on ferromagnetic 3d metal nanocontacts motivated the investigations of the magnetic ground state and the magnetic anisotropy of a Co point contact between oppositely magnetised ferromagnetic Co leads in Thesis Statement 2. It has been found that the magnetocrystalline anisotropy energy (MAE) of the central atom distinguishes between the helical and the cycloidal domain walls. Strong correlation between the MAE and the anisotropy of the orbital moment is expected since their common origin is the SOC. [Bruno, 1989] This correlation is also confirmed by the numerical investigation of the MAE and the orbital moment of the central atom of the cobalt point contact.

The classic example of frustration is the antiferromagnetically coupled symmetric trimer where it is impossible to set all the three spin-pairs in their lowest energy state simultaneously. The measurements of Gao *et al.* [Gao et al., 2008] and the calculations of Stocks *et al.* [Stocks et al., 2007] and Antal *et al.* [Antal et al., 2008] motivated the study of the chirality and anisotropy of Cr trimers on the Au(111) surface. The energy of two magnetic configurations with opposite chirality turned out to be different as it has also been confirmed by Antal *et al.* [Antal et al., 2008]. However, compact Cr trimer on the Au(111) surface is a well-studied system, [Antal et al., 2008, Bergman et al., 2007, Gotsis et al., 2006, Stocks et al., 2007]

some new aspects have been considered in the thesis. There is consensus in the theoretical literature that the ground state is an in-plane 120° Néel state. In a non-relativistic description the Néel configurations with different chirality or different in-plane orientation have the same energy. This degeneracy is lifted by the Dzyaloshinsky-Moriya and the pseudo-dipolar interaction. The results of this topic are presented in Thesis Statement 3.

Methods and techniques

There are several theoretical tools available for determining the electronic structure of magnetic systems. In my thesis the Korringa–Kohn–Rostoker [Kohn and Rostoker, 1954, Korringa, 1947] method is applied. The magnetic properties of nanoclusters (cobalt, chromium) deposited onto a magnetic or onto a non-magnetic substrate (cobalt, copper, gold) have been calculated by using an embedded cluster Green's function technique [Lazarovits et al., 2002, Lazarovits, 2003] as combined with the KKR method.

Classical spin models for the clusters are constructed according to the symmetry of the systems. From the *ab initio* band energy, some of the spin model parameters are determined by the torque [Liechtenstein et al., 1987, Udvardi et al., 2003] and the rotational energy [Szunyogh et al., 2009, 2011] methods.

New results.
Thesis Statements

I formulate the new results of my thesis in three Thesis Statements.

1. I implemented a fully *ab initio* Monte Carlo simulation by extending the KKR code and calculated the temperature dependent magnetization of a 4×4 Co cluster deposited on the Cu(001) surface. The isotropic exchange parameters and an estimation of the uniaxial on-site anisotropy of the Heisenberg model of this system were determined. The spin model MC simulation was compared to the fully *ab initio* MC simulation. I found that the magnetization curves from the two simulations practically agree. The validity of the application of the spin model is confirmed. The advantage of the *ab initio* MC approach is that it does not rely on an *a priori* spin model and its disadvantage is the extreme computational demand.

   Publication II belongs to this thesis statement.

2. The magnetic structure of a model of a Co point contact between two oppositely magnetized Co leads has been investigated by means of *ab initio* calculations. The strong ferromagnetic coupling and the symmetry enable two distinct domain walls: helical wall (HW) and cycloidal wall (CW), the CW being 30 meV lower in energy. The width of the domain walls followed the length of the point contact under a deformation of $-15\% \ldots +15\%$. Strong uniaxial anisotropy of the central atom was experienced with an easy axis perpendicular to the

leads which was the main reason of the lower energy of the CW. Anisotropy of the orbital moment of the central atom was also revealed in strong correlation with the magnetic anisotropy energy.

Publication III belongs to this thesis statement.

3. Equilateral compact Cr trimers deposited onto fcc hollow or hcp hollow positions of the Au(111) surface were investigated. Antiferromagnetic coupling between the Cr atoms results in a ground state of an in-plane 120° Néel state with two possible chirality. The ground state out of the two Néel states was the result of an interplay between the Dzyaloshinsky–Moriya (DM) interaction and the symmetric part of the two-site anisotropy. The DM interaction depended intriguingly on the geometry. In the case of a Cr monolayer on the Au(111) surface the non-vanishing z component of the DM coupling results in an energy difference between the Néel structures with opposite chirality.

   Publications I and **IV** belong to this thesis statement.

Publications connected to the Thesis Statements

The following papers are covered by my thesis. The publications are listed in chronological order.

[I] Antal, A., Lazarovits, B., Balogh, L., Udvardi, L. & Szunyogh, L. Multiscale studies of complex magnetism of nanostructures based on first principles. *Philosophical Magazine* **88**, 2715–2724 (2008). DOI: 10.1080/14786430802389213.

[II] Balogh, L., Lebecki, K. M., Lazarovits, B., Udvardi, L., Szunyogh, L., & Nowak, U. Monte Carlo study on magnetic nanoparticles from first principle. *Journal of Physics: Conference Series* **200**, 072103 (2010). DOI: 10.1088/1742-6596/200/7/072103.

[III] Balogh, L., Palotás, K., Udvardi, L., Szunyogh, L. & Nowak, U. Theoretical study of magnetic domain walls through a cobalt nanocontact. *Phys. Rev. B* **86**,024406 (2012). DOI: 10.1103/PhysRevB.86.024406.

[IV] Balogh, L., Udvardi, L. & Szunyogh, L. Magnetic anisotropy and chirality of frustrated Cr nanostructures on Au(111). *Journal of Physics: Condensed Matter* **26**, 436001 (2014). DOI: 10.1088/0953-8984/26/43/436001.

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