MŰEGYETEM 1782

Theoretical study of the magnetic ordering of thin films and nanostructures

Thesis Booklet

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Background

Magnetic thin films play important role in technological applications of the last decades. In magnetic data storage technology the information is stored in the direction of the magnetic domains, and the magnetic anisotropy plays a crucial role in it. Materials with magnetic domains perpendicular to the surface increase considerably storage density. The first hard disk in the market using perpendicular magnetic anisotropy (PMA) was implemented by Toshiba in 2005 [1]. For the further increase of the storage density the size of the magnetic grains needs to be further decreased, which involves the use of materials with high uniaxial anisotropy [2–4].

The magnetic anisotropy energy (MAE) has different contributions. A prominent role is played by magnetocrystalline anisotropy (MCA) and shape anisotropy (SA), which two often prefer opposite magnetic arrangements. These contributions behave in different ways to the change in the thickness or temperature of the system, leading to the change of the sign of MAE. In such cases, a spin reorientation transition can take place in the system, where the magnetic moments rotate in a new, more favourable direction [5, 6]. In recent years, it has been revealed that the Dzyaloshinsky–Moriya interaction (DMI) has a dynamic finite temperature contribution to the anisotropy, which can influence this process [7].

Topological spin structures, such as skyrmions have been a popular research topic in solid-state physics for the past decade [8]. Modern technology now allows us to create, move and annihilate skyrmions individually [9], treating them as quasi-particles. Their nanoscale size, exceptional stability and mobility make them perfect candidates for use in high-density data storage systems [10] or even quantum computers [11].

The development of experimental tools made it possible to individually move atoms on a substrate with the tip of a scanning tunnelling microscope [12], and thus opened the way for the intensive investigation of spin chains. The investigation of magnetic nanostructures placed on non-magnetic substrates gained a lot of momentum thanks to the discovery of Majorana bound states hosted in nanochains placed on the surface of superconductors [13, 14], which are considered as a possible way to implement quantum computers.

Objectives and summary of research

During my work, I dealt with the phenomena presented above.

One of the main directions of the research was the temperature-dependent investigation of magnetic thin films using the well-tempered metadynamics method. I modelled these systems as classical Heisenberg spins. First, I reproduced the expected temperature dependence of the magnetic anisotropy energy [15] on model systems, then I investigated the spin reorientation transitions on mono- and bilayers with special attention to the order of the phase transformation. After that, I investigated two experimentally motivated systems, Fe bi- and trilayers on Au(001) surface and Fe bilayer on W(110) surface. I described these systems with coupling parameters obtained from first principle calculations [16]. While in the case of the former system, the goal was to investigate the thickness-dependent reorientation transition between the bi- and trilayers, in the case of Fe_2/W(110), the focus was on the temperature-dependent behaviour, with particular regard to the importance of the anisotropy term induced by the DMI.

An important part of my PhD work was the further development of the metadynamics code, so that I could also examine the topological structures hosted in magnetic thin layers. I investigated the appearing skyrmions and antiskyrmions in ($Pt_{0.95}Ir_{0.05}$) /Fe/Pd(111) and Pd/Fe/Ir(111) bilayers. The topological charge that characterises the grid itself not enough to identify different skyrmions. However, by carefully examining the configuration, the various objects can be identified. This allowed me to identify, depending on the temperature and the external magnetic field, a region where the skyrmions behave as particles on the lattice, and thus I was able to calculate the chemical their potential.

Another main direction of the research was the investigation of the magnetic structure of Fe chains placed on non-magnetic substrates by means of *ab initio* optimization. The significance of the investigation of the chains placed on the Re(0001), Rh(111) and Nb(110) surfaces was that the ground state magnetic ordering can be determined directly from the electronic structure without the assumption of a spin model. My studies extended to the investigation of the effect of the ℓ_{max} angular momentum cutoff used in the electronic structure calculation, and to the comparison of the resulting configurations with the results based on classical quadratic spin-model calculations.

Methods

The electronic structure of the various systems I investigated has been determined by using the Korringa–Kohn–Rostoker method (KKR) [16].

I described the magnetic thin films using classical Heisenberg-spin models [17, 18] and investigated their temperature-dependent properties using the well-tempered metadynamics method [19–21], which made it possible to map the free energy surface as a function of a collective variable depending on the physical problem. Thus, I was able to determine both the temperature-dependent magnetic anisotropy energy, which plays a key role in explaining spin reorientation transitions, and the temperature-dependent chemical potential of skyrmionic particles.

The electronic structure of the atomic chains was calculated using the embedded cluster technique implemented in the KKR method [22]. During the *ab initio* optimization of the magnetic ground state, I worked without spin models, directly connecting the electronic structure and the minimization of the torque acting on each spin.

Novel scientific findings

The results of my PhD research and corresponding novel findings are summarized in the following thesis statements:

I. First I used the well-tempered metadynamics method on model spin Hamiltonians describing magnetic thin films. I investigated the temperature dependence of the magnetic anisotropy energy in the case of purely on-site and purely two-site anisotropies and found a good agreement on the magnetic anisotropy energy with the Callen–Callen theory. I found that these two terms have a different temperature dependence, which can lead to a spin reorientation transition (SRT) due to the competition of the two. This phenomenon has been demonstrated first in a single monolayer. In the case of a bilayer system the richer behaviour of the phase transitions have been found, namely first and second order phase transitions were discovered depending on the distribution of the anisotropy of the system.

   These results are published in Pub. II.

II. I studied the magnetic anisotropy of $\mathrm{Fe}_n/\mathrm{Au}(001)$ ($n = 2, 3$) and $\mathrm{Fe}_2/\mathrm{W}(110)$ using well-tempered metadynamics method.

   For the first system, I calculated the magnetic exchange parameters for different Fe-Au layer-to-layer distances, then I used a selected one in the simulations to calculate the temperature dependent magnetic anisotropy energy. I showed, that this system fulfills the Callen–Callen theory. The investigation of free energy surfaces showed that a thickness driven spin reorientation transition happens between $n = 2$ and $n = 3$.

   The Dzyaloshynsky–Moriya interaction (DMI) plays a crucial role in the spin reorientation transition taking place in the Fe bilayer on W(110). I showed that at finite temperature an additional anisotropy term appears, which can be explained by the arising finite temperature fluctuations in the system. I proved that without the DMI, the spin-reorientation transition would not happen in the system.

These results are published in Pub. I., II. and III.

III. I further developed the well-tempered metadynamics code to use the topological charge as a collective variable. With this modified version I investigated the temperature dependence of creation and annihilation of skyrmionic objects in heavy metal/iron bilayers, namely $(\mathrm{Pt}_{0.95}\mathrm{Ir}_{0.05})$/Fe/Pd(111) and Pd/Fe/Ir(111). I found that at the low temperatures in a low density the skyrmions and antiskyrmions act as particles, thus their respective chemical potential can be defined. Finite temperature chemical potential curves showed that for the investigated $(\mathrm{Pt}_{0.95}\mathrm{Ir}_{0.05})$/Fe/Pd(111) bilayer both skyrmion and antiskyrmion creation is energetically unfavourable, but for Pd/Fe/Ir(111) in a magnetic field range, where the ground state is field polarized I found negative chemical potential for skyrmions, showing that their formation is a favourable process, i. e. with the given $B-T$ parameters their creation from the field polarized background is a favourable process.

    The corresponding results are published in Pub. V.

IV. I used *ab initio* optimization to obtain the ground state magnetic configuration of Fe chains on different heavy metal substrates. The calculations on Re substrate highlighted how the choice on the angular momentum cutoff in the electronic structure calculation can influence the magnetic structure. I reproduced the experimentally measured spin–spiral states in the Fe chains on Re(000) and Nb(110), with a good agreement on the wavelengths. The extensive study on Fe atomic chains on Rh(111) showed how the stacking can influence the magnetic configuration, and the comparison with spinmodel simulations pointed out how each term of the exchange coupling contributes to the formation of the magnetic configuration.

    The results are published in Pub. IV.

Publications related to the thesis statements:

My PhD dissertation contains the results of the following pulications:

I. B. Nagyfalusi L. Udvardi and L. Szunyogh, *First principles and metadynamics study of the spin-reorientation transition in Fe/Au(001) films*, IOP Conf. Ser.: J. Phys. Conf. Ser. **903**, 012016 (2017)

II. B. Nagyfalusi, L. Udvardi and L. Szunyogh, *Metadynamics study of the temperature dependence of magnetic anisotropy and spin-reorientation transitions in ultrathin films*, Phys. Rev. B **100**, 174429 (2019)

III. B. Nagyfalusi, L. Udvardi L. Szunyogh, and L. Rózsa, *Spin reorientation in an ultrahin Fe film on W(110) induced by Dzyaloshinsky-Moriya interactions*, Phys. Rev. B **102**, 134413 (2020)

IV. B. Nagyfalusi, L. Udvardi and L. Szunyogh, *Magnetic ground state of supported monatomic Fe chains from first principles*, J. Phys. Cond. Mat. **34** 395803 (2022)

V. B. Nagyfalusi, L. Udvardi L. Szunyogh, and L. Rózsa, *Chemical potential of magnetic skyrmion quasiparticles in heavy metal/iron bilayers*, sent to Phys. Rev. B., under review, arxiv: 2312.05535 (2023)

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