Finite-temperature magnetism of ultrathin films and nanoclusters

PhD Thesis Booklet

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Background of the research

Magnetic materials continue to play an ever more important role in modern information technology. Their conventional applications in data storage have expanded due to the widespread use of the giant magnetoresistance and exchange bias effects in hard drives and magnetic memory devices. Furthermore, recent developments in the field have concentrated on taking advantage of the spin information in logic circuits, coupled to the charge degree of freedom of electrons in spintronics, in the form of spin waves in magnonics, or by concentrating on the topological properties as in skyrmionics. They also continue to attract the interest of scientists working in fundamental research; as a few examples, we mention the recently observed spin, topological and skyrmion Hall effects, as well as noncollinear magnetic phases such as spin spirals and skyrmion lattices.

Similarly to semiconductor-based technologies, the drive for an increased data density has motivated the miniaturization of devices and the decrease in dimensionality of the applied systems. Ultrathin magnetic films with the thickness of a few atoms and the breadth of a few micrometres or hundred nanometres represent a step in this direction, before the system size reaches clusters consisting of several dozen atoms, such as short one-dimensional chains. The aim of the theoretical description of low-dimensional magnetic systems is to construct a model with a few input parameters which can account for different phenomena observed in experiments, and is also capable of opening new directions for progress by making predictions. Since eventually the proposed new devices should operate in room-temperature environments, the range of applicability of the theoretical model should include finite-temperature effects beyond ground state properties.

Ab initio electronic structure calculation methods are capable of describing the band structure of solids and several connected experimental quantities, if the elements constituting the system and their geometrical positions are known in advance. Out of these techniques, the advantage of the Korringa–Kohn–Rostoker method[Korringa, 1947; Kohn, 1954] is the capability to treat systems which lack translational invariance in three dimensions in real space, either by using the screening method for two-dimensional surfaces and interfaces, or by relying on the embedding technique for small clusters of atoms. Based on the adiabatic decoupling of magnetic and electronic degrees of freedom, it is possible to understand magnetism in these systems by treating the localized magnetic moments in the lattice as classical unit vectors.

The stochastic Landau–Lifshitz–Gilbert equation[Brown, 1963] provides an empirical description of the time evolution of classical magnetic systems at finite temperature. While it is supposed in the model that the length of the spin vectors does not change during the calculations, the damping mechanism and the coupling to the heat bath provides a way for describing thermal equilibrium properties and relaxation effects. The spin dynamics simulation method corresponds to the numerical solution of this equation of motion and the extraction of thermodynamic quantities. Although Monte Carlo simulations based on the Metropolis algorithm do not follow the time evolution directly, they are constructed for describing systems at finite temperature in equilibrium. Combining the Korringa–Kohn–Rostoker method with these classical simulation methods enables the successful description of several magnetic phenomena from first principles.

Objectives

The aim of my thesis was to investigate low-dimensional magnetic systems at finite temperature, and describe the low-energy excitations, the possible phase transitions and metastable states. During my investigations I chose different descriptions for ultrathin films and nanoclusters.

I have described ultrathin film systems with a model Hamiltonian, where the coefficients were determined from *ab initio* calculations. The first objective was to reproduce and explain different phenomena observed in experiments. The spin wave spectrum of ferromagnetic Fe mono- and double-layers on W(110) surface has been measured using spin-polarized electron energy loss spectroscopy. It was found that the spin wave frequencies decrease as the temperature is increased[Zakeri, 2014]; I intended to find a quantitative explanation for this phenomenon. At low temperature, Fe double-layer on W(110) orders in a noncollinear spin spiral state, while it becomes ferromagnetic at high temperature as mentioned above. My aim was the theoretical modelling of this spin reorientation transition observed in spin-polarized scanning tunnelling microscopy experiments[von Bergmann, 2006]. I also planned to investigate the spin spiral, skyrmion lattice and field-polarized phases for different values of the external magnetic field in PdFe bilayer on Ir(111), similarly motivated by previous spin-polarized scanning tunnelling microscopy experiments[Romming, 2013].

The second objective was predicting new phenomena. I intended to examine the magnetic ordering and phase transitions in an Fe monolayer on the (110) surface of Ta instead of W. Regarding the PdFe bilayer on Ir(111), I wanted to investigate how the different ordered states transform into the paramagnetic state at elevated temperature, and how the lifetime of skyrmions is affected by thermal fluctuations.

In the case of nanoclusters I treated the interactions between the localized moments directly on *ab initio* grounds. The objective was to implement temperature effects in the calculations and to investigate the differences between the direct *ab initio* calculations and the spin models. I intended to construct numerical solvers to the stochastic Landau–Lifshitz–Gilbert equation which could be applied directly within the *ab initio* formalism, and test their speed and stability. Finally, I planned to examine a simple system, a chain of ten Co atoms on Au(001) surface.

Methods

I used a program based on the screened Korringa–Kohn–Rostoker method[Szunyogh, 1994] for the *ab initio* calculations. I have developed and implemented numerical solvers to the stochastic Landau–Lifshitz–Gilbert equation in the code. Due to the specifics of the program, I had to construct stochastic numerical integration methods which use the quantities determined from first principles, and are able to describe the thermal equilibrium correctly.

I used a program based on the Metropolis Monte Carlo algorithm for performing simulations using a spin Hamiltonian. I modified the program for spin dynamics simulations by implementing a solver to the stochastic Landau–Lifshitz–Gilbert equation. I expanded the code by calculating different quantities, such as the static and dynamic structure factors and the topological charge.

I also performed theoretical calculations based on spin wave excitations and mean-field theory. I have created simple programs capable of executing the necessary numerical

calculations, since I treated noncollinear states such as spin spirals and skyrmion lattices, where the number of variables in the equations is high.

New scientific results

The main results of my thesis are summarized in the thesis statements below.

1. I have determined the linear response function of a magnetic monolayer to an external time-dependent magnetic field using spin dynamics simulations. I have demonstrated that thermal fluctuations decrease the spin wave frequencies, and that this phenomenon can be quantitatively explained by perturbation theory based on spin waves. The results are published in paper [I].

2. I have determined the interaction coefficients and ground states in Fe mono- and double-layers on W(110). The in-plane ferromagnetic phase of the monolayer is in agreement with the experiments. Although the *ab initio* calculations could not reproduce the spin spiral ground state of the double-layer, Monte Carlo simulations confirmed the observed reorientation of the easy axis from out-of-plane to in-plane as the temperature is increased. I have demonstrated possible phase transitions between a spin spiral phase and the ferromagnetic phase magnetized perpendicularly to the spiral plane as a function of temperature using mean-field theory and Monte Carlo simulations. I have predicted the presence of an elliptic conical spin spiral state in Fe double-layer on W(110), which has not been observed experimentally so far. The results are partially published in papers [III] and [IV].

3. I have determined the interaction coefficients and ground states in an Fe monolayer on Ta(110). Depending on the distance between the monolayer and the substrate, I have identified four possible ground states in the system, three of them being noncollinear spin spiral states. I have predicted a transition from the ferromagnetic to the SS I, as well as from the SS II to the SS I state as the temperature is increased, by relying on spin wave expansion and Monte Carlo simulations. I have used mean-field theory to explain the transition from the SS III to the SS II state observed in the simulations. The results are published in paper [IV].

4. I have determined the $B-T$ phase diagram of PdFe bilayer on Ir(111) using Monte Carlo simulations. Besides the long-range ordered cycloidal spin spiral, skyrmion lattice and field-polarized states, I have found an intermediate state displaying short-range order, where the topological charge was fluctuating. Based on the similarity of the calculated static susceptibility and static structure factor to the experimental results for the bulk skyrmionic system MnSi, I have identified this intermediate regime as the fluctuation-disordered state. I have emphasized the importance of finite skyrmion lifetime in this regime at finite external magnetic field, and calculated said lifetime from spin dynamics simulations. The results are published in paper [V].

5. I have constructed three numerical solvers to the stochastic Landau–Lifshitz–Gilbert equation written in the local coordinate system, which I have implemented in the screened Korringa–Kohn–Rostoker code. Based on the stability analysis performed on a simple Heisenberg chain, I have determined that the one-step scheme is the

most suitable one for the application in question. Using *ab initio* spin dynamics simulations, I have examined a chain of ten Co atoms on Au(001) surface. Despite a slight canting of the ground state due to the Dzyaloshinsky–Moriya interactions, I have demonstrated that the thermal equilibrium properties may be sufficiently described by a nearest-neighbour Heisenberg model, while including an on-site anisotropy term can explain the observed switching times. The results are published in paper [II].

Publications connected to thesis statements:

[I] L. Rózsa, L. Udvardi and L. Szunyogh, *Relativistic and thermal effects on the magnon spectrum of a ferromagnetic monolayer*, J. Phys.: Condens. Matter **25**, 506002 (2013).

[II] L. Rózsa, L. Udvardi and L. Szunyogh, *Langevin spin dynamics based on ab initio calculations: numerical schemes and applications*, J. Phys.: Condens. Matter **26**, 216003 (2014).

[III] L. Rózsa, L. Szunyogh and L. Udvardi, *Non-Collinear Magnetic Configurations at Finite Temperature in Thin Films*, IEEE Trans. Magn. **50**(11), 1300704 (2014).

[IV] L. Rózsa, L. Udvardi, L. Szunyogh and I. A. Szabó, *Magnetic phase diagram of an Fe monolayer on W(110) and Ta(110) surfaces based on ab initio calculations*, Phys. Rev. B **91**, 144424 (2015).

[V] L. Rózsa, E. Simon, K. Palotás, L. Udvardi and L. Szunyogh, *Complex magnetic phase diagram and skyrmion lifetime in an ultrathin film from atomistic simulations*, Phys. Rev. B **93**, 024417 (2016).

Other publications:

[VI] E. Simon, K. Palotás, L. Rózsa, L. Udvardi and L. Szunyogh, *Formation of magnetic skyrmions with tunable properties in PdFe bilayer deposited on Ir(111)*, Phys. Rev. B **90**, 094410 (2014).

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