



# Finite-temperature magnetism from first principles

PhD Thesis Booklet

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# Background

Since the end of the last century nanomagnetism has obtained a central position in technological applications. After the discovery of giant magnetoresistance (GMR) and its subsequent rapid application in hard disk drive technology—essentially marking the birth of spintronics—ever more experimental and theoretical effort has been made to harness magnetism, and more generally, the electron spin, in electronic applications. Following the employment of GMR and tunnelling magnetoresistance in spin valves, magnetic alternatives to semiconductor-based memories have also been sought, leading to the invention of the magnetoresistive random access memory (MRAM) and the racetrack memory. Second-generation MRAM techniques are already being developed, which are based on thermal-assisted switching and the spin-transfer torque. The latter effect is also a primary candidate for the manipulation of more exotic spintronic-logic devices implemented through the use of magnetic nanostructures, including nanoskyrmion lattices, paving the way for ‘skyrmionics’. Magnetic effects are being exploited also outside electronics, with ongoing research in areas such as magnetic shape-memory alloys and magnetic refrigeration, in close relation to the magnetocaloric effect.

One aim of theoretical investigations is to broaden the spectrum of available materials for industrial applications. Other aspects concern the basic understanding of the physics behind the relevant effects, such as the superparamagnetism of nanoclusters and how that could be tailored in order to increase bit density in magnetic data storage applications. Due to obvious practical reasons such theoretical efforts must involve finite-temperature calculations, as in the great majority of applications the system is at or near room temperature. While potential candidates for magnetic recording can be filtered through zero-temperature *ab initio* calculations by studying the magnetocrystalline anisotropy of materials, a complete description from a technical point of view must include the high-temperature (or at least room-temperature) behaviour of the candidates. For investigations regarding the magnetocaloric effect the impact of finite temperature need not be explained.

While there is a multitude of techniques allowing for the accurate description of real materials in their ground states from first principles, finite-temperature effects pose a much less tractable problem, especially for magnetic materials. Practical finite-temperature descriptions either introduce some kind of mean field approximation or step away from the *ab initio* approach by introducing spin models. The former approach provides a much more fundamental description, however it involves huge computational effort and is rarely feasible for materials showing non-collinear magnetism. Spin models, on the other hand, can give detailed information about crucial finite-temperature behaviour such as

susceptibilities, specific heats and temperature-induced phase transitions, at the cost of moving away from the first-principles approach.

The relativistic disordered local moment method (relativistic DLM, RDLM) implemented within the screened Korringa–Kohn–Rostoker (screened KKR, SKKR) multiple scattering theory provides a mean field *ab initio* description of temperature-induced spin fluctuations in terms of the coherent potential approximation. The theory allows to include the impact of spin disorder on the electronic structure self-consistently. For systems with reduced dimensions and complex magnetic structure it is practical to extract spin model parameters using either the so-called spin-cluster expansion (SCE) or the relativistic torque method (RTM), compatible with a paramagnetic (PM) or an ordered ferromagnetic (FM) reference state, respectively. The corresponding spin model can then be used in a multi-scale approach by performing Monte Carlo or atomistic spin dynamics simulations to obtain the ground state spin configuration or finite-temperature excitations.

## Objectives

The aim of the thesis is to investigate the finite-temperature magnetism of metallic magnets in terms of the self-consistent RDLM-SKKR method and to further develop the available techniques in order to broaden the scope of our research.

In the first part of the thesis I study bulk magnets important from the point of view of industrial applications. I assess the stability of the local moment of bcc-Fe, followed by calculations concerning the magnetocrystalline anisotropy of L1<sub>0</sub>-FePt and the metamagnetic phase transition of CsCl-ordered FeRh. Finally, I investigate the effects of transversal spin fluctuations and surface termination on the local moment of hcp-Gd in terms of the LSDA+U approach.

In the second part of the thesis I turn to the thin film system Fe<sub>1</sub>/Rh(001), and extract spin model parameters from the electronic structure in order to determine the ground-state spin configuration. I also develop the generalization of our methods used for the computation of *ab initio* exchange interactions, namely the spin-cluster expansion and the relativistic torque method, in order to extract spin model parameters directly from the finite-temperature electronic structure.

## Novel scientific findings

My research detailed in the thesis and the corresponding novel findings are summarized in the following thesis statements:

1. I made essential contributions to implement the self-consistent relativistic disordered local moment scheme into the screened Korringa–Kohn–Rostoker method. I used the new code to explore the variation of the magnetization and the local spin moment of bcc-Fe with temperature. I found a 15% reduction of the Fe moment from the ferromagnetic ground state to the high-temperature paramagnetic state, suggesting limited rigidity of the Fe moment in the bulk. The calculations overestimated the Curie temperature due to a shortcoming of mean field theory inherent in RDLM theory. Therefore, I suggested that the results obtained from the RDLM method should be interpreted on the basis of the average magnetization rather than the temperature.

Related publication: [I]

2. By using the RDLM-SKKR code I performed systematic calculations of the magnetocrystalline anisotropy energy (MAE) of FePt alloys subject to spin and chemical disorder due to finite temperature and intermixing between layers nominally populated by Fe and Pt. The calculations demonstrated that the MAE decays rapidly with increasing chemical disorder at any temperature and for fixed intermixing it shows approximate power-law dependence on the total magnetization of the system. I found that the corresponding exponent increases towards increasing temperature, suggesting that the value of 2.1 of the exponent quoted in the literature only applies at very low temperatures. The magnetization-dependence of the local spin moments suggests that Fe is quite rigid in FePt, while the induced moment of Pt is likely to be described by a simple Weiss-field picture.

Related publication: [I]

3. I used the self-consistent RDLM-SKKR method to map the phase diagram of FeRh as a function of lattice parameter and mean field temperature. Zero-temperature total energy calculations suggest that the ground state is antiferromagnetic (AFM) below  $a = 3.11 \text{ \AA}$  and FM above. From the calculated Curie and Néel temperatures I compiled the theoretical FM-PM and AFM-PM phase boundaries as a function of the lattice parameter. The triple point marked by the crossing of these two curves occurs at  $a = 2.99 \text{ \AA}$ . In between these two lattice parameters I concluded the presence of a temperature-induced metamagnetic transition. I constructed the phase boundary between the AFM and FM phases by searching for crossovers in

the free energy curves corresponding to these two states.

Related publication: [I]

4. I implemented the LSDA+U method within the RDLM-SKKR program package. Using this code I compared the magnetization of Gd in the bulk and on the (0001) surface, in the zero-temperature FM and the high-temperature PM phases. In the bulk I found a considerable spin splitting in the conduction band, owing to a local moment of  $0.77 \mu_B$ . In the PM phase this splitting almost vanishes, yet the conduction band still possesses a spin moment of  $0.41 \mu_B$  due to the asymmetry of spectral weights in the two spin channels. The results suggest non-Stoner behaviour of the Gd conduction band moment, but also reveal that the polarization effect due to the Weiss field generated by the  $4f$  states is also significant.

Related publication: [III]

5. I explored the magnetic interactions in the  $\text{Fe}_1/\text{Rh}(001)$  thin film using the relativistic torque method and the spin-cluster expansion combined with RDLM independently and found considerable differences between the two models. The results reveal that the tensorial Heisenberg model is insufficient for the description of the magnetism in this system, indicating that higher-order two-spin interactions and multi-spin interactions are most likely needed to grasp the complex magnetic structure found experimentally.

Related publication: [II]

6. I developed and implemented the SCE-RDLM method below the Curie temperature in order to obtain the effect of thermal spin fluctuations on the exchange couplings from first principles. Calculations for bcc-Fe show the correct limit towards the PM state, however in the  $T \rightarrow 0$  K limit the interactions are inconsistent with the RTM. I then developed and implemented the RTM-RDLM method and found that the resulting exchange couplings produce the correct limits. In anticipation of applications in ultrafast demagnetization simulations I explored the dependence of the exchange couplings on magnetic order parameter and electronic temperature up to the Stoner–Curie point.

## Publications related to the thesis statements

- [I] A. Deák, E. Simon, L. Balogh, L. Szunyogh, M. dos Santos Dias, and J. B. Staunton, *Metallic magnetism at finite temperatures studied by relativistic disordered moment description: Theory and applications*, Physical Review B **89**, 224401 (2014)

- [II] A. Deák, K. Palotás, L. Szunyogh, and I. A. Szabó, *Magnetic correlations beyond the Heisenberg model in an Fe monolayer on Rh(001)*, Journal of Physics: Condensed Matter **27**, 146003 (2015)
- [III] L. Oroszlány, A. Deák, S. Khmelevskiy, and L. Szunyogh, *Magnetism of gadolinium: a first-principles perspective*, arXiv:1503.08940 (2015), submitted to Physical Review Letters

## Other publications

- [IV] E. Simon, K. Palotás, B. Ujfalussy, A. Deák, G. M. Stocks, and L. Szunyogh, *Spin-correlations and magnetic structure in an Fe monolayer on 5d transition metal surfaces*, Journal of Physics: Condensed Matter **26**, 186001 (2014)
- [V] J. B. Staunton, R. Banerjee, M. dos Santos Dias, A. Deak, and L. Szunyogh, *Fluctuating local moments, itinerant electrons, and the magnetocaloric effect: Compositional hypersensitivity of FeRh*, Physical Review B **89**, 054427 (2014)
- [VI] A. Deák, L. Szunyogh, and B. Ujfalussy, *Thickness-dependent magnetic structure of ultrathin Fe/Ir(001) films: From spin-spiral states toward ferromagnetic order*, Physical Review B **84**, 224413 (2011)
- [VII] M. dos Santos Dias, J. B. Staunton, A. Deak, and L. Szunyogh, *Anisotropic spin-spin correlations in  $Mn_1/X(111)$  ( $X = Pd, Pt, Ag, \text{ and } Au$ )*, Physical Review B **83**, 054435 (2011)