Chapter 6

Summary and Outlook

This thesis presents a theoretical investigation of magnetic properties of laterally nanostructured films during growth. A phenomenological model for MBE growth is developed, obtaining realistic film morphologies. We propose an appropriate micromagnetic model for the description of the magnetic behavior whose basic physical idea is to describe nanostructured films as systems of interacting magnetic islands with varying island positions, sizes and shapes. Inter-island dipole and exchange interactions and a uniaxial single-island anisotropy are taken into account on equal footing. As an approximation, we apply a two-state model $S_i = \pm 1$ for the stable island magnetization directions along the uniaxial easy axis, taking into account an anisotropy energy barrier during magnetization reversal for the dynamics. Using this model, we studied nanostructures, ranging from separated dipole-coupled uniaxial islands for film coverages much below the percolation threshold Θ_{P} to connected ferromagnetic films for coverages much larger than Θ_{P}, and the transition between these extremal cases. We investigated the long-range magnetic ordering of such ultrathin film systems by using equilibrium MC simulations. Furthermore, kinetic MC simulations were performed in order to examine the magnetic relaxation processes into thermal equilibrium and to treat metastable states far from equilibrium. For this, a new cluster MC method was developed, taking into account correlated island magnetization rotations. The application of this method leads to a much more efficient and more realistic relaxation into equilibrium of irregularly connected island systems and allows for the calculation of the magnetic properties in the entire coverage range within the same model. Most of our results cannot be obtained by conventional methods.

The general aim of this study is the investigation of magnetic properties in thermal equilibrium and nonequilibrium and their dependence on the irregular nanostructure, the film coverage, the temperature, the time, and the magnetic interactions. Coming back to the leading questions of this thesis, which were formulated in the Introduction, p. 17 and p. 20, our investigations lead to the following main results:

• **Long-range magnetic ordering:**
Our study demonstrated for the first time that for irregular 2D island systems dipole couplings between in-plane magnetized islands lead to a stable (time-independent) collective magnetic order at finite temperatures considering thermal fluctuations. For film coverages Θ below the percolation threshold Θ_P, the used growth and magnetic parameters lead to ordering temperatures T_C of the long-range magnetic order in the range of 1 - 10 K induced by the dipole interaction. Such a magnetic ordering was previously calculated only for uniform spin systems [37, 38]. With increasing coverage, the exchange interaction couples single islands to magnetically aligned large clusters, resulting in a strong increase of T_C in the vicinity of Θ_P. This behavior has also been observed in experiments on Co/Cu(001) ultrathin films ('T_C-jump') [17, 133]. A comparison with results obtained by a simple mean-field theory (MFT) demonstrated the importance of thermal fluctuations for 2D systems of dipole-coupled islands. The neglect of fluctuations by using MFT leads to an overestimation of T_C by a factor 30.

For coverages $\Theta > \Theta_P$, the exchange coupling leads to a long-range ferromagnetic ordering with ordering temperatures T_C in the range of 100 - 300 K, depending on the connectivity of the system. The temperature dependence of the equilibrium film magnetization is governed by the internal island order and the thermal disturbance of the island spin alignmet.

• **Magnetic relaxation into equilibrium:**
For coverages $\Theta < \Theta_P$ and low temperatures, energy barriers due to single-island magnetic anisotropies lead to a slow relaxation behavior into equilibrium from an aligned state. For the assumed anisotropy values and a waiting time ~ 100 sec, we obtain blocking temperatures T_b which are an order of magnitude higher than the ordering temperature T_C of the long-range order.

Interestingly, an increase of the anisotropy constant K by a factor 10 leads to an increase of T_b by factors 2 to 5, depending on the coverage. This is in contrast to the simple Stoner-Wohlfarth model for coherent magnetization rotations of coagulated islands which predicts $T_b \propto K$. Here, the magnetic relaxation of the irregularly connected island system happens via internal island cluster excitations, in addition to coherent cluster rotations. In the investigated parameter range, mostly one or two islands in an island cluster are rotated in each step.

For systems without a percolating structure, $\Theta < \Theta_P$, we observe a very interesting effect of the dipole interaction on the relaxation behavior: Assuming *in-plane* magnetized islands, this coupling induces 10 % larger remanent nonequilibrium magnetizations and blocking temperatures for the used growth and magnetic parameters and thus a slower relaxation. Such a behavior was observed recently in experiments on 2D arrays of separated magnetic nanoparticles with random anisotropy axes [131]. Interestingly, we obtain this behavior also for temperatures much higher than the corresponding ordering temperatures T_C of the long-range magnetic order due to dipole coupling. For increasing ratios of the dipole energy to the single-island anisotropy, this effect becomes more pronounced, and relaxation evolves into the collectively ordered state. In contrast, for *out-of-plane* magnetized systems, the dipole coupling accelerates the magnetic relaxation and reduces the blocking temperatures.

For coverages $\Theta > \Theta_P$, generally a fast relaxation from the saturated state into the ferromagnetic state is obtained due to the strong exchange coupling. However, for coverages in vicinity of Θ_P, where the film is characterized by irregular island cluster formations, nonequilibrium effects due to anisotropy barriers are still pronounced. For increasing coverage and connectivity between the islands, T_b passes over to T_C.

• **Metastable multi-domain structure:**
In simulations of the time evolution of the magnetic domain structure during ultrathin film growth, we observe metastable micro-domains far from thermal equilibrium. We find this 'as-grown' magnetic structure in particular at coverages near the percolation threshold Θ_P where the systems are very irregularly connected. The metastable domain structure - with domain areas larger than isolated single-domain islands and smaller than equilibrium domains known for smooth films - is controlled and stabilized by the nonuniform nanostructure and the exchange coupling. Thus, the irregular film system represents a 'random ferromagnet' with a huge number of metastable collective states, separated by high energy barriers [27, 120]. We obtain a maximum of the average domain area as function of the growth temperature and a minimum of the average domain roughness which we define as the edge-to-area ratio of a domain. For the assumed growth and magnetic parameters the influence of single-island anisotropy barriers on the domain structure is small, but could be more distinct for larger island sizes. Also, the deposition rate of a growing film has only a minor influence on the metastable domains due to their strong temporal stability. Occurrence and stability of such 'frozen-in' nonequilibrium domains of growing thin films have not been investigated previously by theory. We suggest that the experimentally observed small 'as-grown' domains in rough Co/Au(111) films are of the described origin [3, 2, 154, 73].

• **Outlook:**
The results of our study lead to further interesting problems. For coverages $\Theta < \Theta_P$, it is interesting to study the magnetic domain structure in the low-temperature ordered phase, resulting from the inter-island dipole coupling. For this anisotropic coupling, in-plane and out-of-plane island magnetization directions have to be distinguished. For the latter case, we expect an antiferromagnetic alignment of the magnetization directions of neighbored, but still

separated islands, resulting in a long-range magnetic order with a vanishing overall magnetization. Furthermore, it is interesting to study the effect of the dipole interaction on the domain formation during film growth. Does a pre-ordering happen in systems without percolation, resulting in stripe domains for coverages $\Theta > \Theta_{\mathrm{P}}$? What is the role of the structural disorder, such as irregular island positions [79]? In addition, it is very tempting to study in more detail the combined effect of single-island anisotropy barriers and inter-island dipole couplings on the magnetic relaxation from an aligned state and to determine the corresponding relaxation laws. How does the crossover from a relaxation into disordered high-temperature states to a relaxation into collectively ordered low-temperature states happen? Also, the complicated relaxation behavior of irregularly coagulated films via internal island cluster excitations at higher temperatures can be investigated in more detail. Moreover, completely different magnetic nanostructures like chains of separated islands [160, 151] and the transition to one-dimensional stripes [58, 151], and to systems with two-dimensional percolation [151] can be examined which have been already realized by experimentalists. Here, we expect that the combined effect of the single-island anisotropy and the anisotropic dipole interaction enables a variety of different magnetic structures due to different possibilities for the island arrangements and the easy axes.

• **Improvements:**

Several possible extensions of the micromagnetic model are pointed out. In this study we have used a two-state model $S_i = \pm 1$ for the island magnetic moments. Noncollinear magnetic arrangements can be analysed by applying continuously varying island magnetization directions $\boldsymbol{S}_i$. The application of vector spins also allows for the calculation of the effect of an external magnetic field, the corresponding hysteresis loops, and the susceptibilities. In particular for a connected film, the movement of magnetic domain walls can be investigated. Additionally, the effect of magnetic anisotropies of different symmetries, e. g. a four-fold in-plane anisotropy, can be investigated.

In the near future a variety of magnetic systems with defined lateral nanostructures will be prepared in a more and more controlled manner, thus, there will be an increasing interest in the calculation of the corresponding magnetic ensemble properties at finite temperatures.