Chapter 4

Monte Carlo Simulation

Due to the complexity of the problem, treating irregular spin systems with competing interactions in thermal equilibrium and nonequilibrium, we are forced to use Monte Carlo (MC) simulations, since present analytical methods can not cope equivalently with such systems. The MC method is widely applied in statistical physics, since it is a powerful tool for the (in principle) *exact* numerical calculation of thermodynamic properties of interacting many-particle systems. On one hand, *equilibrium* MC simulations allow for the study of thermodynamic equilibrium properties, e. g. phase transitions of systems with many competing interactions, taking into account statistical fluctuations in contrast to simplified mean-field treatments, as presented in Sec. F.2. On the other hand, under certain conditions also the nonequilibrium stochastic time evolution of interacting many-particle systems can be calculated with the help of *kinetic* MC simulations. In the present thesis, the MC method is used for the investigation of the long-range magnetic ordering and the magnetic relaxation of nanostructured films during growth described by the micromagnetic model (Eq. (3.20). For this purpose, the island spin flip rates, given by Eqs. (3.37) and (3.39), are applied. For details and a foundation of the MC method the reader is referred to standard textbooks [10, 59, 13, 92].

In **Sec. 4.1**, a brief introduction into the MC method is given. The equilibrium and the kinetic MC method, as applied in our magnetic calculations, are explained. In **Sec. 4.2**, a single-spin flip algorithm and a newly developed cluster-spin-flip algorithm for the application of the MC method is presented. Finally, in **Sec. 4.3**, the accuracy of the method is discussed.

4.1 Monte Carlo method

The contact of the interacting island spin system to a heat bath with finite temperature T leads to random spin reversals. This stochastic pro-

cess represents a so-called *Markov chain* $\boldsymbol{X}_i \rightarrow \boldsymbol{X}_{i+1} \rightarrow \ldots$ of spin states $\boldsymbol{X} = \{S_1, \ldots, S_Z\}$ in the phase space of the system, with $S_i = \pm 1$ and Z being the number of spins. Each state $\boldsymbol{X}_{i+1}$ of this process follows from the preceding state $\boldsymbol{X}_i$ by a transition probability per time step (transition rate) $\Gamma(\boldsymbol{X}_i \rightarrow \boldsymbol{X}_{i+1})$. The dynamics of the system can be described by *Markov's master equation*

$$\frac{dP(\boldsymbol{X},t)}{dt} = -\sum_{\boldsymbol{X}'} \Gamma(\boldsymbol{X} \rightarrow \boldsymbol{X}')\ P(\boldsymbol{X},t) + \sum_{\boldsymbol{X}'} \Gamma(\boldsymbol{X}' \rightarrow \boldsymbol{X})\ P(\boldsymbol{X}',t) \quad (4.1)$$

for the probability $P(\boldsymbol{X},t) := P(\boldsymbol{X}_i)$ to find the system at discrete time t, i. e. in the ith configuration, to be in state $\boldsymbol{X}$. It can be shown that if the transition rates $\Gamma(\boldsymbol{X}_i \rightarrow \boldsymbol{X}_{i+1})$ fulfil the condition of *detailed balance*

$$\Gamma(\boldsymbol{X} \rightarrow \boldsymbol{X}')\ P_{\mathrm{eq}}(\boldsymbol{X}) = \Gamma(\boldsymbol{X}' \rightarrow \boldsymbol{X})\ P_{\mathrm{eq}}(\boldsymbol{X}') \quad , \quad (4.2)$$

the Markov process converges towards thermodynamical equilibrium, where the system is found to be in state $\boldsymbol{X}$ with probability

$$P_{\mathrm{eq}}(\boldsymbol{X}) = \mathcal{Z}^{-1} \exp(-E(\boldsymbol{X})/k_{\mathrm{B}}T) \quad , \quad (4.3)$$

and where

$$\frac{dP_{\mathrm{eq}}(\boldsymbol{X})}{dt} \equiv 0 \quad . \quad (4.4)$$

Here, $E(\boldsymbol{X})$ is the energy of spin state $\boldsymbol{X}$, given by the micromagnetic model (Eq. (3.20); $\mathcal{Z}$ is the partition function

$$\mathcal{Z} = \sum_{\boldsymbol{X}} \exp(-E(\boldsymbol{X})/k_{\mathrm{B}}T) \quad . \quad (4.5)$$

Detailed balance is a sufficient, but not necessary condition to reach the equilibrium distribution. In addition, *ergodicity* has to be fulfilled, stating that any state of the system is accessible from any other state.

In MC simulations, Markov chains of high-probability spin states are generated for the calculation of equilibrium and relaxational properties, which will be explained in the following.

4.1.1 Equilibrium MC method for long-range order

We use the *equilibrium* MC method [106, 10] for the calculation of the equilibrium overall magnetization of the island spin system, which characterizes the long-range magnetic order.

We define the magnetization M of a system in spin state $\boldsymbol{X}$ by

$$M(\boldsymbol{X}) := \frac{\sum_i m_i(\Theta,T)\ N_i(\Theta)\ S_i}{\sum_i N_i(\Theta)} \quad , \quad (4.6)$$

which is the component along the easy axis, and where $m_i(\Theta, T)$ is the *internal* island magnetization at coverage Θ and temperature T (see Appendix F.1), N_i the island size, and $S_i = \pm 1$ the island spin state. For a canonical ensemble, the expectation value of the magnetization in thermodynamical equilibrium $\langle M_{\rm eq} \rangle$ is calculated by

$$\langle M_{\rm eq} \rangle = \mathcal{Z}^{-1} \sum_{\boldsymbol{X}} M(\boldsymbol{X}\,) \exp(-E(\boldsymbol{X})/k_B T) = \sum_{\boldsymbol{X}} M(\boldsymbol{X})\; P_{\rm eq}(\boldsymbol{X}) \quad . \quad (4.7)$$

In the importance sampling MC approach, the complicated and often impossible summation over all states $\boldsymbol{X}$ of the phase space is avoided. Rather, this method samples over a number of most important states of the phase space. For this purpose, one starts from an initial spin configuration $\boldsymbol{X}_1$ (at time t_1) and successively, by application of transition rates $\Gamma(\boldsymbol{X} \rightarrow \boldsymbol{X}')$, one generates a Markov chain of spin states, which relaxes into states of high probability at thermodynamical equilibrium. Reaching a high-probability state at time t_2, the subsequent states are assumed to occurr according the Boltzmann distribution $P_{\rm eq}(\boldsymbol{X})$. Then, Eq. (4.7) can be reduced to the simple arithmetic average

$$\langle M_{\rm eq} \rangle \approx \overline{M} = \frac{1}{t_3 - t_2} \sum_{t=t_2+1}^{t_3} M(\boldsymbol{X}, t) \quad , \quad (4.8)$$

where the sum runs over a large number of states in the Markov chain. Thus, Eq. (4.8) represents an average over the MC time intervall $(t_3 - t_2)$ of the equilibrated system.

In the present case of a *nonuniform* spin system, one additionally has to average over a large number G of different structural realizations of the unit cell, obtaining

$$[\langle M_{\rm eq} \rangle]_{\rm av} = \frac{1}{G} \sum_{g=1}^{G} \langle M_{\rm eq} \rangle_g \quad . \quad (4.9)$$

As already mentioned, the sufficient prerequisite for reaching the equilibrium distribution $P_{\rm eq}(\boldsymbol{X})$ in the Markov process is that the applied transition rates $\Gamma(\boldsymbol{X} \rightarrow \boldsymbol{X}')$ obey the condition of detailed balance (Eq. (4.2)) and ergodicity. In literature, many single-spin-flip and cluster-spin-flip algorithms with different *efficiencies* due to different equilibration times are known to fulfil these conditions [92]. Both, the Arrhenius-type and the Metropolis-type rates for island spin flips, Eqs. (3.37) and (3.39), fulfil these criteria. However, for MC simulations of equilibrium properties, we exclusively apply the latter for *all* values of the reduced effective field $|h_i^{\rm eff}|$, since then the relaxation of the system into equilibrium is not hindered by anisotropy energy barriers, resulting in a better efficiency.

4.1.2 Kinetic MC method for relaxation process

Under certain conditions, the MC method can be interpreted dynamically, where the computer-generated Markov chain of spin states $\boldsymbol{X}$ represents the 'true' nonequilibrium *stochastic* time evolution of a system towards equilibrium, although MC simulations do not solve the equations of motion. For this purpose, a proper connection between MC time t and physical time t' is needed. A foundation of this *kinetic* MC method, which most of the time has been applied to the study of atomic diffusion processes, can be found in Refs. [171, 50, 94, 92].

The *deterministic* trajectory of the interacting island spin system through phase space can be described by the corresponding set of coupled Landau-Lifshitz-Gilbert (LLG) equations (Eq. (D.1)), where the probability of a state develops according to Liouville's equation. Note that a numerical integration of the LLG equations is restricted to time scales of the order of the spin precession time $\tau_{\rm pr} \approx 10^{-9}$ sec. However, the characteristic time τ for island magnetization reversal over an energy barrier can range up to seconds or even years so that a numerical integration of the LLG equations for the study of such slow relaxation behavior is not feasible. This problem can be avoided by describing island magnetization reversal as a thermally activated *stochastic* process, induced by the degrees of freedom on short time scales. In this case, Liouville's equation reduces to Markov's master equation (Eq. (4.1)) for the stochastic time evolution, which can be solved by the MC method [92, 50].

Most importantly, for a proper connection of MC time t to a physical time t', the transition rates $\Gamma(\boldsymbol{X} \rightarrow \boldsymbol{X}')$ have to be based on a reasonable *dynamical model*. For the characteristic time τ of island magnetization reversals, this is achieved by the Néel-Brown model (Eq. (3.14)). This model results in the Arrhenius-type and the Metropolis-type rates (Eqs. (3.37) and (3.39)) which are applied by us to kinetic MC simulations, depending on the value of the reduced effective field $|h_i^{\rm eff}|$.[1]

The expectation value of the nonequilibrium magnetization at MC time t for a given film structure is defined by [13, 12]

$$\langle M(t) \rangle = \sum_{\boldsymbol{X}} P(\boldsymbol{X}, t)\, M(\boldsymbol{X}) \quad , \tag{4.10}$$

where $P(\boldsymbol{X}, t)$ is the solution of the master equation, Eq. (4.1). Using the MC method, the relaxation of $\langle M(t) \rangle$ is calculated by averaging over $R \gg 1$

[1] Recently, an alternative method for MC simulations with physical time step quantification was proposed for vector spins $\boldsymbol{S}$. The main idea of this novel approach is to compare the spin fluctuations appearing in a MC time step with the fluctuations within a given time scale resulting from the linearized stochastic LLG equation [117, 116, 28].

statistically independent MC runs r, applying

$$\langle M(t)\rangle \approx \overline{M}(t) = \frac{1}{R}\sum_{r=1}^{R} M_r(t) \quad . \tag{4.11}$$

Each MC run starts from the same initial spin state $\boldsymbol{X}_1$, and different Markov chains of states are generated by application of different sequences of random numbers. $M_r(t)$ is monitored after MC time t. For a nonuniform film structure, additionally a structural averaging $[\ldots]_{\mathrm{av}}$ has to be performed, as described by Eq. (4.9).

4.2 Monte Carlo algorithms

In the present study, two different algorithms are used for the application of the MC method: a conventional single-spin-flip algorithm, and a newly developed cluster-spin-algorithm which yields an improved relaxational behavior for irregularly connected island systems which are preferably investigated in this study.

4.2.1 Single-spin-flip algorithm

In conventional single-spin-flip (SSF) algorithms, e. g. the heat-bath or Metropolis algorithm, Markov chains of configurations $\boldsymbol{X}$ in phase space are generated, using SSF transition rates and computer-generated pseudo-random numbers.

In the present thesis, the following SSF algorithm is applied for the realization of the MC method:

(1) Specify an initial spin state $\boldsymbol{X}_1 = \{S_1, \ldots, S_Z\}$ in phase space.

(2) Choose a random island spin S_i out of the system using a uniformly distributed random number r_1.

(3) Calculate the reduced effective field $|h_i^{\mathrm{eff}}|$, using Eq. (3.35).

(4) Calculate the corresponding probability

$$P_i = \kappa\, \Delta t\, \Gamma_i \tag{4.12}$$

for an island flip $S_i \rightarrow -S_i$ in the MC time step Δt, using Eqs. (3.36) and (3.37) or Eqs. (3.38) and (3.39).

(5) Generate a second uniform random number $r_2 \in (0, 1)$.

(6) If $r_2 \leq P_i$, perform the spin flip $S_i \rightarrow -S_i$; if $r_2 > P_i$, leave S_i unchanged.

(7) Goto step (2).

After steps (2) – (7) have been applied Z times, where Z is the number of spins in the system, the MC time step $\Delta t = 1$ MC step/spin (MCS) has passed, and the system transforms from the old configuration $\boldsymbol{X}_i$ to the new configuration $\boldsymbol{X}_{i+1}$ in the Markov chain. Within kinetic simulations, Δt is adjusted to the physical time step $\Delta t' = \kappa\,\Delta t$ by the prefactor κ appearing in Eq. (4.12). The assumption of equidistant MC time steps Δt leads to a good approximation of the solution of Markov's master equation (Eq. (4.1)), where time t is rather a continuous stochastic variable [12, 1].

4.2.2 Cluster-spin-flip algorithm

Conventional SSF algorithms can become very unfavorable for the calculation of equilibrium properties, since large clusters of aligned spins are updated very inefficiently by probing single spins. For example, such spin clusters appear in a uniform system near the critical temperature, where the correlation length diverges, and in random magnets or spin glass systems [11] at low temperatures. Therefore, sophisticated cluster-spin-flip (CSF) algorithms have been developed to avoid critical slowing down [162, 175, 7] or to reduce low-temperature relaxation times, e. g. in frustrated systems [96, 141] or ferrofluids [35]. To our knowledge, cluster algorithms have not been applied to kinetic MC simulations yet.

In context of the present study, the exclusive consideration of SSF updates of irregularly connected island spin systems yields an extremely inefficient and unrealistically slow relaxation behavior towards thermodynamic equilibrium at low and intermediate temperatures. For example, in a large coverage range of the bilayer growth mode, see Fig. 2.6, p. 30, most of the magnetic islands are connected to neighboring islands and thus form large island clusters. In such a cluster, the strong exchange interaction favors an aligned arrangement of island spins S_i. Within the two-state model $S_i = \pm 1$, *subsequent* single-spin flips in the cluster are strongly hindered by the exchange energy. Thus, a rotation of the entire island cluster becomes very unlikely, and the role of the dipole interaction and the anisotropy on the cluster rotation is strongly blurred by the exchange coupling. For *efficient* MC calculations of equilibrium properties and for *improved* kinetic simulations of the relaxation behavior, *coherent* rotations of the island spins in such clusters have to be taken into account, since such processes may be more probable. In other words, coherent rotations enable more favorable paths in the 'rough' energy landscape of the randomly connected island system.

In the remainder of this subsection, we propose a new CSF algorithm which consideres such coherent processes *in addition* to internal cluster excitations. Within this algorithm, **every MC update** consists of two steps.

In **step (I)**, a cluster of connected island spins is constructed and then, in **step (II)**, a coherent flip of this cluster is probed:

(I) A cluster $\mathcal{C}_\nu$, consisting of ν connected island spins S_i, is constructed by the following scheme:

(1′) Choose a random spin S_i, representing the first (smallest) spin cluster $\mathcal{C}_1 = \{S_i\}$.

(2′) Add a random second spin S_j which is connected to spin S_i ($L_{ij} \neq 0$), resulting in the second cluster $\mathcal{C}_2 = \{S_i, S_j\}$.

(3′) Construct subsequently larger spin clusters $\mathcal{C}_\nu$ by iteration of step (2′): add a randomly chosen spin to the preceeding cluster $\mathcal{C}_{\nu-1}$, provided that this spin is connected to at least one of the $\nu - 1$ spins of $\mathcal{C}_{\nu-1}$.

(4′) Continue this construction procedure, until either no additional adjacent spins are present,[2] or until a maximum allowed number $\lambda_{\max}$ of spins in the cluster is reached. From this procedure, a set of $\lambda \leq \lambda_{\max}$ clusters $\{\mathcal{C}_1, \ldots, \mathcal{C}_\lambda\}$ is obtained.

(5′) Out of this set, choose one cluster $\mathcal{C}_\nu$ with weight ω_ν for probing.

(II) Next, all ν spins of the chosen cluster $\mathcal{C}_\nu$ are probed for a coherent flip in the usual way, according to steps $(3) - (7)$ of the SSF algorithm given in the previous subsection. For the calculation of the reduced effective field h_ν^{eff} and the transition probability P_ν first Eq. (3.41), then Eqs. (3.47) and (3.37), or Eqs. (3.48) and (3.39) have to be applied.

We point out that not only the largest possible spin cluster $\mathcal{C}_\lambda$ can be probed for flipping, rather all spin clusters out of the constructed set of clusters are considered. Note that the spins within a spin cluster need not to be parallel. The CSF algorithm satisfies the condition of detailed balance. This is guaranteed by the fact that the probabilities for construction and choice of the cluster $\mathcal{C}_\nu$ are the same for both flip directions. Ergodicity is maintained since any spin state can be reached within a single MC time step due to the allowance of single-spin flips.

The MC time step $\Delta t = 1$ MCS is defined by the usual condition that Z spins have been probed. In step (5′), different kinds of weights ω_ν can be chosen in principle, subject to the condition $\sum_\nu \omega_\nu = 1$. However, probing a cluster $\mathcal{C}_\nu$ containing ν spins considers the fraction ν/Z of the system in a single update. To ensure that probing of large clusters does not dominate the relaxation process in kinetic simulations, we assign the weight

$$\omega_\nu = 1/\nu \tag{4.13}$$

[2] In the present study, this is especially the case for film coverages $\Theta < \Theta_{\text{P}}$.

for choosing $\mathcal{C}_\nu$ out of the set $\{\mathcal{C}_1, \ldots, \mathcal{C}_\lambda\}$. This definition implies that within a MC time step no additional relaxation channels are opened by the consideration of cluster-spin flips.

Note that the proposed algorithm for spin-cluster updates can be applied to many different nonuniform spin systems such as random magnets and spin glasses. For example, *atomic* spins of an irregular magnetic film can be treated by this method.

4.3 Accuracy of the Monte Carlo method

The accuracy of the MC calculations is determined by the following limitations:

- Finite-size effects due to the usage of finite (500×500)-unit cells with periodic boundary conditions, containing 625 or 1250 island spins. At phase transitions, where the correlation length diverges, a 'rounding' instead of a singularity of thermodynamical quantities is obtained. We use periodic boundary conditions for the simulation of an extended film system.
- Statistical error due to averaging over:
  - a finite number $(t_3 - t_2)$ of Markov states in equilibrium calculations (Eq. (4.8)),
  - a finite number R of runs in kinetic calculations (Eq. (4.11)),
  - a finite number G of structural realizations (Eq. (4.9)) in equilibrium and kinetic calculations.
- Usage of computer-generated pseudo-random numbers, which must be uniform, uncorrelated and of extremely long period. We make use of the well tested random-number generators `ran2` (Fortran 77) and `ran` (Fortran 90), taken from Refs. [135, 136].

In *kinetic* simulations using the CSF algorithm, the relaxation time depends on the weight $\omega_\nu = 1/\nu$ (Eq. (4.13)) and the definition of a MC time step. Both need for sound justification and thus have 'model character', but still the CSF algorithm leads to much more realistic results than the SSF for irregularly connected systems.