Introducing the Step Monte Carlo Method for Simulating Dynamic Properties

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In this work, a simple modification of the Monte Carlo algorithm is introduced, which is called step Monte Carlo (sMC). The sMC approach allows to simulate processes far from equilibrium and obtain information about the dynamic properties of the system under investigation. In the approach proposed, here the probability of accepting the final (trial) state depends on the activation energy, not on the relative energy between the final and initial state. This barrier height is probed on an ongoing basis, by generating intermediate states along the path connecting the initial and trial positions. Importantly, to calculate the activation energy, the model only requires knowledge of the Hamiltonian without having to introduce additional input parameters such as transition rates etc. The details of sMC are explained in the case of a simple spin model. The comparison of its results with the ones obtained within the frame of stochastic Landau-Lifshitz-Gilbert indicates the correctness of sMC. In this study’s opinion, the proposed method can be applied to simulate other processes, for example, dynamics of classical atoms and complex fluids, diffusion, nucleation, surface adsorption, and crystal growth processes.

1. Introduction

The Monte Carlo (MC) method has proved to be a valuable simulation tool in many branches of science such as physics, chemistry, biology, computer science, economics, finance, and engineering. In the field of condensed matter physics and materials science MC can be used to study, among many others, system of classical particles, classical spin systems, nucleation and crystal growth processes, polymer solutions, hopping transport, percolation, and fractals problems. The MC algorithm is the natural choice for studying the static properties of a system, where dynamical effects are not required. Then, the advantages of Monte Carlo are the relative ease of implementation and the rapid convergence to a steady state. In general, the theory of equilibrium properties is well developed for a wide variety of models and materials. Here, however, we shall consider situations far from equilibrium. For example, the MBE growth of thin films proceeds by both deposition and diffusion processes. The probability of an atom to hop to a nearest empty site, does not depend only on the relative energies of the configuration before and after the hopping event, but rather on the barrier height. The Kinetic Monte Carlo (KMC) method was developed for evolving systems dynamically from state to state. KMC simulates how the occupation of the sites changes over time in a system with known transition rates (these rates are inputs to the KMC algorithm).

In the modified MC approach proposed here (similarly to the KMC approach) the probability of accepting the final state depends on the activation energy, not on the relative energy between the final and initial state. However, the barrier height is calculated on an ongoing basis, by generating intermediate states with a predefined step \( \Delta \). Therefore, we name this method step Monte Carlo (sMC).

Importantly, the sMC method correctly takes into account the presence of various local barriers and it obeys the detailed balance condition, even if the system is not in equilibrium. As a result, the appropriate dynamics of the tested system is simulated. The details of sMC algorithm are explained for the case of magnetization process. To test the correctness of sMC, we compare its results with those obtained by stochastic Landau-Lifshitz-Gilbert (sLLG) approach.

2. Step Monte-Carlo Method

Here, we outline the details of step Monte Carlo method, that allows us to simulate processes far from equilibrium and obtain information about the dynamic properties of the system under investigation. The differences between sMC and standard MC are shown schematically in Figure 1 for the case of the magnetization process. In sMC, the probability of accepting the final magnetic direction \( \mathbf{m}_{\text{final}} \) depends on the activation energy (barrier height) \( E_{\text{AE}} \), not on the relative energy between the final and initial state \( E_{\text{final}} - E_{\text{initial}} \). To calculate the barrier height, we probe energies \( E_k = f(\mathbf{m}_k) \) of intermediate magnetic states \( \mathbf{m}_1, \mathbf{m}_2, \ldots, \mathbf{m}_{N-1} \). These intermediate states are generated on the arc connecting the initial \( \mathbf{m}_{\text{init}} = \mathbf{m}_1 \) and the final (trial) \( \mathbf{m}_{\text{final}} = \mathbf{m}_N \) magnetization direction. This means that \( \mathbf{m}_{k+1} \) is obtained using a predefined step \( \Delta \) in such a way that \( |\mathbf{m}_{k+1} - \mathbf{m}_k| \propto \Delta \). Details of the generation of intermediate states are shown in Figure 2. In general, there are many ways to generate intermediate states. The method described in Figure 2 produces intermediate states
Figure 1. Schematic illustration of (a) Monte Carlo and (b) step Monte Carlo (sMC) methods for the case of magnetization process (one Monte Carlo move). The magnetization direction \( m \) (bottom panels) can take any point on the unit sphere with energy \( E = f(m) \) (top panels). \( m_{\text{init}} \) and \( m_{\text{final}} \) are initial and final (trial) direction of magnetic moment with energy \( E_{\text{init}} \) and \( E_{\text{final}} \) respectively. In the sMC approach the probability of accepting the final random state \( m_{\text{final}} \) depends on the activation energy \( E_{\text{AE}} = E_{\text{max}} - E_{\text{init}} \), not on the relative energy \( E_{\text{final}} - E_{\text{init}} \). This barrier height is probed by generating intermediate magnetic states \( m_k \) \( (k = 2, 3, \ldots, N - 1) \) on the arc connecting the initial \( m_{\text{init}} \) and the final \( m_{\text{final}} \) magnetization direction with a predefined magnetization step \( \Delta \). This means that \( m_{k+1} \) is obtained in such a way that \( |m_{k+1} - m_k| \propto \Delta \).

that are not uniformly distributed on the arc connecting the initial and final magnetization states. This may cause some problems in the case of high values of the parameter \( \Delta \) resulting in strong variations in the intermediate magnetic energy \( E_k = f(m_k) \) (highly variable energy landscape). Therefore, as shown later, in simulations we choose very small value of \( \Delta = 10^{-3} \), which ensures correct calculation of the barrier height. In addition, more complex ways of generating intermediate states can be employed, such as those based on spherical coordinates.

After obtaining energies \( E_k \), the barrier height is obtained from \( E_{\text{AE}} = \max\{E_2, E_3, \ldots, E_N\} - E_{\text{init}} \), where \( E_N = E_{\text{final}} \). Finally, we proceed in a similar way to the Monte Carlo Metropolis algorithm for a classical spin system. The complete procedure is shown below:

1. Select a random atom \( j \) with the initial direction of the magnetic moment \( m_{\text{init}} \).
2. Choose randomly a new trial direction \( m_{\text{final}} \).
3. Generate intermediate magnetic states \( m_{1}, m_{2}, m_{3}, \ldots, m_{N-1} \) with the predefined step \( \Delta \) on the arc connecting the initial \( m_{\text{init}} = m_1 \) and the final \( m_{\text{final}} = m_N \) magnetization direction (see Figure 2).
4. Calculate energies \( E_{jk} = f(m_{jk}) \) and the activation energy \( E_{\text{AE}} = \max\{E_2, E_3, \ldots, E_N\} - E_{\text{init}} \).
5. The new trial direction \( m_{\text{final}} \) is accepted with the probability \( P = \exp\left(-\frac{E_{\text{final}}}{kT}\right) \), that is:
   a. Draw a random number \( r \) uniformly distributed between 0 and 1.
   b. If \( r \leq \exp\left(-\frac{E_{\text{final}}}{kT}\right) \), accept the new state, otherwise reject it.
6. Repeat the procedure.

One complete iteration of sMC consists of \( N_c \) such trial moves (one move corresponds to the whole procedure 1–5 presented above), where \( N_c \) is the number of atoms in the system. In sMC approach we use \( 4 \cdot 10^7 \) equilibration and averaging iterations.

Here for generation of the trial direction \( m_{\text{final}} \) we choose randomly one of the two different trial moves: Gaussian or random (c.f. Figures 3b, c and 4b in ref. [9]). The Gaussian trial move creates \( m_{\text{final}} \) by shifting the initial direction \( m_{\text{init}} \) on the unit sphere according to the formula:

\[
m_{\text{final}} = m_{\text{init}} + \sigma \frac{\Gamma}{|m_{\text{init}} + \sigma \Gamma|} \tag{1}
\]

Figure 2. Schematic illustration of the generation of intermediate states by shifting the magnetization direction on the unit sphere (on the arc connecting the initial \( m_{\text{init}} \) and the final \( m_{\text{final}} \) magnetization direction).
where $\Gamma$ is a 3D random vector characterized by the Gaussian distribution with a mean of zero and a standard deviation of 1. The width of the cone $\sigma_g$ takes the following form

$$\sigma_g = \frac{2}{5} \left( \frac{\pi^2}{k_B T} \right)^{1/5} \] [9] which aims to obtain a move acceptance rate of around 50%. The random trial move creates a random point on the unit sphere $m_{\text{final}} = \frac{\Gamma_{U}}{|\Gamma_{U}|}$, where $\Gamma_{U}$ is a 3D random vector with each components uniformly distributed between -1 and 1. In order to correctly (densely enough) arrange intermediate states meeting the following condition $m_{i,\text{init}} \cdot m_{i,\text{final}} \leq -0.97$ are rejected.

In our simulations, the total acceptance rate $r_{\text{accept}}$ was found to be strongly dependent on the input parameters. Its values were contained in the range from $r_{\text{accept}} \approx 0.17$ for small $T$, large $H$ and large anisotropy constant to a maximum value of $r_{\text{accept}} \approx 0.6$ for large $T$.

We can see that the implementation of the sMC algorithm is a little more difficult than the standard MC one. Also the execution time for one sMC iteration is longer than in MC case, especially for small values of $\Delta$. However, the advantage of using the new method is that MC provides proper Boltzman distribution of states in relation to the global energy minimum, whereas sMC (in the short run) provides Boltzman distribution of states relative to the local energy minimum. This guarantees the correct dynamics of sMC, as energy barriers much higher than $k_B T$ prevent from too fast global thermalization of spins.

It is important to note, that the sMC approach fulfills two general conditions for validity of Monte Carlo algorithms: ergodicity and the condition of detailed balance. Ergodicity expresses the requirement that all possible configurations of the system can be reached from any other configuration in a finite number of Markov steps. The detailed balance condition requires that, in the long run, the system approaches the correct thermal equilibrium. In sMC, for any two configurations $B$ and $C$ we have

$$\frac{W(B \rightarrow C)}{W(C \rightarrow B)} = e^{-(E_{C} - E_{B})/(k_B T)}$$

where $W(B \rightarrow C)$ describes the probability to go from state $B$ to state $C$ in one move of the Markov process. $p(B) = e^{E_B/(k_B T)} / Z$ is the Boltzmann probability distribution, in which $E_B$ is the energy of configuration $B$ and $Z$ is the partition function. $E_{\text{max}}$ corresponds to the energy of the barrier (see Figure 1 with $B = m_{\text{init}}$ and $C = m_{\text{final}}$). Equation (2) is also true in the case with no barrier, that is, when $E_B = E_{\text{max}}$ or $E_C = E_{\text{max}}$ (then sMC moves coincide with MC ones).

One should note, that the sMC approach presented here for spin systems can be generalized in a straightforward way to simulate other processes, for example diffusion, nucleation, surface adsorption, crystal growth processes, and dynamics of classical atoms. Then we just generate intermediate states along the line connecting the initial and final (trial) positions. Moreover, to simulate the crystal growth, our model only requires knowledge of the Hamiltonian without having to assume a specific discrete structure of the crystal lattice.

To test the correctness of sMC, we compare its results with those obtained by stochastic Landau-Lifshitz-Gilbert (sLLG) equation.\[9,21,24\] Using the sLLG approach, it was possible to simulate the dynamic properties of magnetic materials\[21\] including ferromagnetic hysteresis loops,\[9\] time evolution of a system of spins,\[9\] spin waves,\[22\] domain wall motion,\[24,27\] and magnetization switching processes.\[28,29\] The sLLG equation, applied at the atomic level, reads

$$\frac{\partial m_i}{\partial t} = -\frac{\gamma}{1 + \alpha C} [m_i \times H_{\text{eff}} + \alpha C m_i \times (m_i \times H_{\text{eff}})]$$

where $\gamma$ is the gyromagnetic ratio, $\alpha C$ is the damping parameter. The symbol $\mathbf{m}$ describes normalized $(|\mathbf{m}| = 1)$ direction of the local magnetic moment of atom $i$ and $H_{\text{eff}}$ corresponds to

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**Figure 3.** Investigated magnetic clusters composed of up to four magnetic ions (blue circles) coupled by a ferromagnetic exchange interaction (red lines).

**Figure 4.** The magnetization $M$ of single ion, computed as a function of the external magnetic field $H$ applied along the magnetic easy axis at $T = 0.04$ K. The simulations are performed using the sMC method with different steps $\Delta$. The standard MC approach is represented by $\Delta = 3$. 

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effective magnetic field acting on this spin, \( H_{\text{eff}} \) is calculated from the first derivative of the total spin Hamiltonian \( H \) and incorporates also the instantaneous thermal magnetic field \( H_{\text{th}} \), according to the following formula

\[
H_{\text{eff}} = -\frac{1}{\mu_s} \frac{\partial H}{\partial m_i} + H_{\text{th}}
\]

with

\[
H_{\text{th}} = \Gamma(t) \sqrt{\frac{2\alpha_s k_B T}{\gamma \mu_s \Delta t}}
\]

The instantaneous magnetic field \( H_{\text{th}} \) represents thermal effects (fluctuations) in the system, and allows to simulate the magnetization at a finite temperature \( T \). \( \Gamma(t) \) is a random vector in 3D space characterized by the Gaussian distribution with a mean of zero and a standard deviation of 1. \( \mu_s = g \mu_B S \) is an actual value of atomic moment with spin \( S = 2 \), electron g-factor \( g = 2 \) and \( \Delta t = 5 \times 10^{-5} \) ns is the time step of simulation. Additionally, in sLLG method we use \( \alpha_s = 1 \) and \( 2 \times 10^{10} \) equilibration and averaging iterations.

### 3. Results and Discussion

Having outlined the details of the step Monte Carlo method, we now proceed to a comparative study of the magnetization dynamics obtained within the frame of the sMC and standard sLLG approach. We analyze a simple model consisting of few interacting magnetic atoms possessing the uniaxial anisotropy along the \( z \) axis. The investigated small magnetic clusters are shown in Figure 3. The Hamiltonian of the system reads

\[
H = -k_z \sum_i m_i^z - \mu_s H \sum_i m_i - J \sum_{i<j} m_i m_j
\]

with appropriate terms relating to uniaxial anisotropy energy, Zeeman energy and the exchange coupling between atoms. Here \( k_z = 0.4 \text{ meV} \) is the anisotropy constant, \( J = 2 \text{ meV} \) describes the exchange parameter. The symbol \( \mathbf{H} \) corresponds to the external magnetic field.

In Figure 4, we present the magnetization \( M \) of a single ion, computed as a function of the external magnetic field \( H \) applied along the magnetic easy axis, namely \( \mathbf{H} \parallel \mathbf{z} \). The simulations are obtained using the sMC method with different steps \( \Delta \). To obtain magnetization curves \( M(H,T) \) we start simulations from the high field regime, where all spins are practically aligned along the direction of the applied \( H \). Next, the magnetic field is swept from 2.2 \( T \) to -2.2 \( T \) and back to 2.2 \( T \) in steps depending on the value of \( H \) (in the hysteresis region the steps of 0.005 \( T \) or less are used). The final state of the previous \( H \) is taken as the starting state of the next magnetic field.

In Figure 4 the standard (usual) MC approach is represented by \( \Delta = 3 \). Only in this case we observe the rapid convergence to equilibrium state with no hysteresis loop seen in \( M - H \) curve. By decreasing \( \Delta \) critical dynamic behaviors far from equilibrium emerge. That is, for a given very large number of iterations, hysteresis behavior is obtained in Figure 4 using sMC simulations with \( \Delta \leq 1 \).

Now the magnetization has to overcome the energy barrier for switching, what creates the hysteresis loop. By decreasing \( \Delta \) this barrier height is probed with higher resolution leading to the increase of the coercive field \( H_c \). However, for \( \Delta \leq 10^{-3} \) we observe saturation of \( H_c \) and a clear presence of thermal fluctuations. The fact that obtained results are practically independent of a predefined magnetization step \( \Delta \leq 10^{-3} \) indicates that the proposed here method is reliable and robust.

In Figure 5 the comparison of numerical results obtained using the sMC (\( \Delta = 10^{-3} \)) and sLLG methods. a) The magnetization \( M \) of a single ion with the magnetic field \( H \) applied at different angles \( \theta \) from the easy axis \( z \) at temperature \( T = 0.04 \text{ K} \). b) The magnetization \( M \) of a single ion at different temperatures \( T \) with the magnetic field \( H \) applied along the easy axis \( H \parallel \mathbf{z} \).
is even more beneficial for the sMC algorithm in the case of larger values of $\Delta$. In general, execution speed will depend on the specific problem being investigated. The sMC method requires significantly fewer (by at least two orders of magnitude) initialization and averaging iterations than sLLG approach. On the other hand, one sMC iteration (consisting of many intermediate steps) takes much more computational time than one sLLG iteration. Therefore, we expect sMC will be a particularly efficient algorithm for simulating magnetic systems involving long-range dipole–dipole interactions. In such systems, most of the time of one sMC or sLLG move is consumed in calculating the magnetic field originating from the surrounding magnetic dipoles. In such a case, the computational cost per iteration for both methods is almost equivalent. Since the sMC method requires significantly less iteration than sLLG, its benefits are obvious for systems with long range interactions. However, for most magnetic systems, the sLLG algorithm appears to be superior due to its ease of implementation. Similarly, for simulations of molecular systems, the well-established Molecular Dynamics (MD) seems to be the best numerical approach. The disadvantage of MD is that it is not straightforward and simple to maintain constant temperature during simulations. In contrast, MC simulations at finite temperature can be performed without the use of the various thermostats necessary in MD. The impact of such thermostats on simulation results is not entirely clear. In Monte Carlo calculations, temperature is taken into account in a straightforward and simplest possible way. Therefore, we expect that the main application (in the field of condensed matter physics) of the method proposed here would be simulations of dynamics of classical atoms or complex fluids at high temperature, for example: crystal growth or diffusion processes.

Finally, magnetizations for larger clusters are presented in Figure 6. At finite temperature, the coercive field increases slightly with the size of the cluster $N_c$, as expected. Again, the consistency between the sLLG and sMC results is very good, indicating a correct assumptions and implementation of the sMC algorithm.

Interestingly, the coercive field obtained from sLLG is slightly smaller than the one obtained from the sMC approach. This is especially noticeable for the magnetic field applied along the easy axis $H||z$ ($\theta = 0$). In our opinion, it is connected with the fact that in sMC algorithm the Landau-Lifshitz (LL) term is absent. This LL term producing precessional motion of magnetization along the effective magnetic field seems to be important in such switching processes.

4. Conclusion

Here, we present a modification of MC algorithm, named step Monte Carlo (sMC), that allows to simulate dynamic properties of various classical systems. In sMC the probability of accepting the final state depends on activation energy (barrier height), not on the relative energy between the final and initial state. This barrier height is probed by generating intermediate states with a predefined step $\Delta$. To calculate the activation energy, our model only requires knowledge of the Hamiltonian without having to introduce additional input parameters such as transition rates etc. Importantly, the sMC method correctly takes into account the presence of various local barriers. As a result, the appropriate dynamics of the tested system is simulated. To test the correctness of sMC, we simulate dynamic magnetic properties for interacting spin system and compare its results with those obtained by stochastic Landau-Lifshitz-Gilbert (sLLG) approach. The obtained excellent correspondence of both methods indicates the correctness of the proposed approach. In our opinion, sMC method can be applied to simulate other processes, for example dynamics of classical atoms and complex fluids, diffusion, and crystal growth processes.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

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[1] J. S. Liu, Monte Carlo Strategies in Scientific Computing, Springer Series in Statistics, Springer-Verlag, NY 2004.
[2] P. Glasserman, Monte Carlo Methods in Financial Engineering, Stochastic Modelling and Applied Probability, Springer Science+Business Media, NY 2003.
[3] T. Gerstner, P. Kloeden, Recent developments in computational finance : foundation, algorithms and applications, Interdisciplinary Mathematical Sciences, Singapore World Scientific, Singapore 2013.
[4] G. S. Fishman, Monte Carlo : Concepts, Algorithms, and Applications, Springer Series in Operations Research and Financial Engineering, Springer New York, NY 2003.
[5] D. Frenkel, B. Smit, Understanding Molecular Simulation : From Algorithms to Applications, Academic Press, San Diego 2002.
[6] A. R. Leach, Molecular Modelling : Principles and Applications, Longman, Harlow 1996.
[7] M. Sawicki, T. Devillers, S. Gałęski, C. Simserides, S. Dobkowska, B. Faina, A. Grois, A. Navarro-Quezada, K. N. Trohidou, J. A. Majewski, T. Dietl, A. Bonanni, Phys. Rev. B 2012, 85, 205204.
[8] C. Simserides, J. A. Majewski, K. N. Trohidou, T. Dietl, EPJ Web of Conferences 2014, 75, 01003.
[9] R. F. L. Evans, W. J. Fan, P. Chureemart, T. A. Ostler, M. O. A. Ellis, R. W. Chantrell, J. Phys.: Condens. Matter 2014, 26, 103202.
[10] H. Sato, P. Chureemart, F. Matsuura, R. W. Chantrell, H. Ohno, R. F. L. Evans, Phys. Rev. B 2018, 98, 214428.
[11] H. Sitter, Thin Solid Films 1995, 267, 37.
[12] P. J. C. M. van Hoof, W. J. P. van Enckevort, M. Schoutsen, J. Cryst. Growth 1998, 193, 679.
[13] D. L. Woodraska, J. A. Jaszcza, Surf. Sci. 1997, 374, 319.
[14] M. A. Żaluska-Kotur, F. Krzyzewski, S. Krukowski, R. Czernecki, M. Leszczyński, Crystal Growth & Design 2013, 13, 1006.
[15] M. S. Song, T. Koren, M. Żaluska-Kotur, R. Buczko, N. Avraham, P. Kacman, H. Shtrikman, H. Beidenkopf, Nano Letters 2021, 21, 10215.
[16] N. Urakami, M. Takasu, J. Phys. Soc. Jap. 1996, 65, 2694.
[17] K. Murayama, J. Kuwabara, Sol. State Comm. 1997, 103, 591.
[18] S. Ma, H. Xu, Y. Li, Z. Song, Physica B: Cond. Matter 2007, 398, 55.
[19] M. Dimakogianni, C. Simserides, G. P. Triberis, Phil. Mag. 2013, 93, 2729.
[20] D. Dhar, M. Barma, J. Phys. C: Solid State Phys. 1981, 14, L1.
[21] W. M. Young, E. W. Elcock, Proc. Phys. Soc. 1966, 89, 735.
[22] A. B. Bortz, M. H. Kalos, J. L. Lebowitz, J. Comp. Phys. 1975, 17, 10.
[23] R. F. L. Evans, U. Atxitia, R. W. Chantrell, Phys. Rev. B 2015, 91, 144425.
[24] Y. K. Edathumkandy, D. Sztenkiel, J. Magn. Magn. Mater. 2022, 562, 169738.
[25] S. A. Bender, H. Skarsvåg, A. Brataas, R. A. Duine, Phys. Rev. Lett. 2017, 119, 056804.
[26] F. Schlickeiser, U. Ritzmann, D. Hinzke, U. Nowak, Phys. Rev. Lett. 2014, 113, 097201.
[27] D. Hinzke, U. Nowak, Phys. Rev. Lett. 2011, 107, 027205.
[28] M. O. A. Ellis, R. W. Chantrell, Appl. Phys. Lett. 2015, 106, 162407.
[29] W. Scholz, T. Schrefl, J. Fidler, J. Magn. Magn. Mater. 2001, 233, 296.