Vinamax: a macrospin simulation tool for magnetic nanoparticles

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Abstract We present Vinamax, a simulation tool for nanoparticles that aims at simulating magnetization dynamics on very large timescales. To this end, each individual nanoparticle is approximated by a macrospin. Vinamax numerically solves the Landau–Lifshitz equation by adopting a dipole approximation method, while temperature effects can be taken into account with two stochastic methods. It describes the influence of demagnetizing and anisotropy fields on magnetic nanoparticles at finite temperatures in a space- and time-dependent externally applied field. Vinamax can be used in biomedical research where nanoparticle imaging techniques are under development, e.g., to validate other higher-level models and study their limitations.

Keywords Magnetic nanoparticles · Macrospin · Micromagnetism · Simulations

1 Introduction

In recent years, many biomedical applications based on nanotechnology [27] in general and magnetic nanoparticles [31, 32] specifically have emerged. Examples of applications under development are disease detection [24], targeted drug delivery [1] and hyperthermia [23]. All of these depend upon an accurate knowledge of the spatial distribution of the magnetic particles [11, 17] and thus require a nanoparticle imaging technique [4, 28]. Some promising imaging techniques are magnetic particle imaging (MPI) [20], magnetorelaxometry [38] and electron paramagnetic resonance (EPR) [12]. While each method has its distinct advantages, none of them is able to quantitatively reconstruct the spatial particle distribution in vivo. For example, while MPI has a submillimeter resolution, it still has difficulties obtaining the absolute values of the MNP distribution [37]. This is in contrast to EPR which can accurately determine the total MNP amount in the sample, but currently only allows to recover the 1D distribution [12]. Inadequate knowledge of the spatial distribution of the nanoparticles results in suboptimal applications and even a decrease in patient safety and comfort. The development of an adequate technique is challenging, in part, due to an insufficient understanding of the collective magnetic behavior of the particles.

However, numerically investigating these particles from first principles (i.e., micromagnetically [8]) can lead to an understanding of this collective behavior and consequently an improved performance of aforementioned applications. To the best of our knowledge, there exists no simulation software that both are accurate on the smallest timescales at which micromagnetic dynamics take place, and still are able to simulate the large timescales involved in experiments. Therefore, we have developed Vinamax [36]: a broad simulation tool in which individual nanoparticles are represented by single macrospins. Vinamax numerically simulates the magnetic dynamics by solving a phenomenological equation, including relaxation and damping, just like the Landau–Lifshitz equation [25]. It considers demagnetizing and anisotropy fields and takes into account externally applied fields that can be space and time dependent. To be able to simulate large ensembles of nanoparticles,
the demagnetizing interaction is solved efficiently using a dipole approximation method [33]. This contrasts approaches in which the demagnetizing interaction is not taken into account [3] or cut off after a short distance, e.g., when considering aggregates of nanoparticles [2]. Additionally, thermal effects [9, 18] can be taken into account by two different approaches: First, a stochastic field term can be added to the effective field or second, magnetic nanoparticles are switched at stochastic time intervals, equivalent to the first approach. The “first-principles” approach adopted by Vinamax makes it an ideal tool to validate other higher-level models and/or investigate their limitations.

The paper is organized as follows. In Sect. 2, the Landau–Lifshitz-like equation with the additional stochastic term is described in detail together with the algorithm used to calculate the demagnetization interaction. Section 3 demonstrates the validity of the software by comparing simulation results to the well-established micromagnetic software MuMax3 [35], which is a broad micromagnetic simulation software. The largest differences between both softwares are that mamax3 calculates the demagnetizing field using a fast Fourier transform method on a grid of cubic cells. This approach has the drawback in nanoparticle simulations that also the empty space between the nanoparticles (which can be more than 99 % of the simulated volume) is taken into this calculation, which severely limits the number of nanoparticles that can be simulated. We were able to validate the Vinamax implementation on simple problems, that only take into account a few nanoparticles, by comparing the results with MuMax3. Also in Sect. 3, the equivalency of the two different approaches to include thermal effects is demonstrated and a magnetorelaxometry simulation is considered and compared to the moment superposition model [15, 16], which is typically used to describe a magnetorelaxometry experiment [19]. Hereby, we illustrate the usefulness of Vinamax in this field as magnetorelaxometry is a well-known technique for the imaging of magnetic nanoparticles in biomedical applications. Finally, some concluding remarks about Vinamax are given in Sect. 4.

2 Methods

2.1 Micromagnetic theory

In the micromagnetic framework, the magnetization is described as a continuum vector field \( \mathbf{M}(\mathbf{r}, t) \). In the following, the space and time dependence of the magnetization vector field is no longer explicitly shown in the equations. The nanoparticles under consideration are assumed to be uniformly magnetized [30] (i.e., their size is sufficiently small to be single-domain particles). Because all spins within each particle lie parallel to each other, Vinax can further simplify the continuum approximation by describing every nanoparticle as one single macrospin. This implies that the exchange interactions do not have to be evaluated. The magnetic dynamics of each nanoparticle are described by a Landau–Lifshitz-like equation (Eq. (1)) including a relaxation and damping term:

\[
\tau = \frac{d\mathbf{m}}{dt} = -\frac{\gamma_0}{1+\alpha^2} (\mathbf{m} \times \mathbf{B}_{\text{eff}} + \alpha \mathbf{m} \times \mathbf{m} \times \mathbf{B}_{\text{eff}})
\]

In this equation, the torque \( \tau \), due to the effective field \( \mathbf{B}_{\text{eff}} \) felt by each nanoparticle, equals the time derivative of \( \mathbf{m} \), where \( \mathbf{m} \) denotes the magnetization vector normalized with respect to the saturation magnetization \( \mathbf{M} = \mathbf{m} \mathbf{M}_{\text{sat}} \). Furthermore, \( \gamma_0 \) denotes the gyromagnetic ratio \( 1.7595 \times 10^{11} \) rad/Ts and \( \alpha \) is the dimensionless Gilbert damping constant. Equation (1) is solved numerically by time stepping it with timestep \( \Delta t \), which is described in further detail in Sect. 2.2.

The field \( \mathbf{B}_{\text{eff}} \) is the sum of the different effective field terms that influence the magnetization:

\[
\mathbf{B}_{\text{eff}} = \mathbf{B}_{\text{ext}} + \mathbf{B}_{\text{anis}} + \mathbf{B}_{\text{demag}} + \mathbf{B}_{\text{therm}}
\]

(2)

The different terms in Eq. (2) are described in more detail below.

2.1.1 External field

\( \mathbf{B}_{\text{ext}} \) is an externally applied field that can be both space and time dependent.

2.1.2 Anisotropy field

In Vinax, the nanoparticles are assumed to have uniaxial anisotropy which is the case for the iron oxide nanoparticles used in biomedical applications [32]. The field \( \mathbf{B}_{\text{anis}} \) affecting a particle due to this anisotropy is the derivative of \( \mathbf{m} \) of the micromagnetic anisotropy energy term [13] (4).

\[
\mathbf{B}_{\text{anis}} = \frac{2K_{\text{u1}}}{\mathbf{M}_{\text{sat}}} (\mathbf{m} \cdot \mathbf{u}) \mathbf{u}
\]

(3)

\[
E_{\text{anis}} = K_{\text{u1}} \left[ 1 - (\mathbf{M} \cdot \mathbf{u})^2 \right]
\]

(4)

The anisotropy constant \( K_{\text{u1}} \) and the anisotropy axis \( \mathbf{u} \) can be chosen freely. \( \mathbf{u} \) can be set to a predefined direction or to uniformly distributed random directions for all nanoparticles. To this end, two random numbers \( \phi \) and \( \theta \) are generated with the following distribution:

\[
\phi = 2\pi \rho_1
\]

(5)

\[
\theta = 2 \arcsin(\sqrt{\rho_2})
\]

(6)

where \( \rho_{1,2} \) denote two uncorrelated, uniformly distributed random numbers in the interval \([0.0, 1.0]\). These spherical
coordinates are then mapped to their cartesian counterparts using the well-known relations below:

\[
x = \sin(\theta) \cos(\phi) \tag{7}
\]

\[
y = \sin(\theta) \sin(\phi) \tag{8}
\]

\[
z = \cos(\theta) \tag{9}
\]

2.1.3 Demagnetizing field

The demagnetizing field (or the magnetostatic interaction) results from the dipole–dipole interaction of the different particles. Its contribution to the effective field is given by Eq. (10) [13], where \(i\) loops over all particles, \(\mu_0 = 4\pi \times 10^{-7} \text{Tm/A}\), \(r_i\) denotes the distance from each particle to the point at which the demagnetizing field is evaluated, and \(V_i\) is the volume of each particle.

\[
B_{\text{demag}} = \mu_0 \sum_i V_i M_{\text{sat},i} \left[ \frac{(m_i \cdot r_i) r_i}{r_i^3} - \frac{m_i}{r_i^3} \right] \tag{10}
\]

2.1.4 Dipole approximation method

To speed up the evaluation of the demagnetizing field, we have implemented a dipole approximation method. This method is based on a multipole approximation described by Tan [33]. We will briefly describe our adapted implementation, but for a detailed explanation we refer to Tan [33].

In Vinamax, the user has to define a world, which is a cube that encloses all the particles in the simulation. This cube is then subdivided into eight subnodes, which are further subdivided until every node contains at most one particle. Within this tree, we then calculate the center of magnetization (CM) for every node. The center of magnetization (CM) is the center of mass of the particles in the node weighted with the magnetic moment of each particle. This is shown in Eq. (11), where \(\sum_i\) denotes a sum over all particles in the node, and \(r_i\) is the position of particle \(i\).

\[
R_{\text{CM}} = \frac{1}{\sum_i V_i M_{\text{sat},i}} \sum_i r_i V_i M_{\text{sat},i} \tag{11}
\]

In the evaluation of the total demagnetizing field, we take into account the contribution per node of the field \(B_d\) as if all the particles in a node were compressed into one dipole in the CM of that node, using Eq. (12), where \(r\) denotes the distance between the CM of the node and the point at which the field is evaluated. Before every timestep, the magnetization \(M = \sum_i V_i M_{\text{sat},i} m_i\) of each node is updated based on the magnetization of every particle within the nodes.

\[
B_d = \mu_0 \frac{3(M \cdot r) r - M}{r^3} \tag{12}
\]

For every particle, the demagnetizing field working on it is evaluated with the algorithm described in Fig. 1, copied from Tan [33] with minor adaptations to our implementation.

**Fig. 1** The algorithm used to evaluate the demagnetizing field, adapted from Tan [33].
In the high barrier limit, Eq. (13) leads to switching between two states with a minimal energy, separated by an energy barrier of height \( \Delta E \). The probability that a particle does not switch during a certain time \( \Delta t \) is given by \( \frac{dP_{\text{not}}}{dt} = -P_{\text{not}} \). In a constant field, this leads to

\[
\ln P_{\text{not}} = \int_0^{\Delta t} f \, dt \Rightarrow P = 1 - \exp(-f \Delta t)
\]

The next switching time for a particle can thus be generated with \( t = -\frac{1}{f} \ln (1 - P) \) by generating a random number \( P \), uniformly distributed between 0 and 1 [26]. When the simulation reaches this time, the magnetization of the particle is switched to its opposite direction and a new switching time is generated.

### 2.2 Time integration schemes

Vinamax provides the user with a wide range of different methods to numerically integrate Eq. (1) [10]

- Euler’s method
- Heun’s method
- Third-order Runge–Kutta method
- Fourth-order Runge–Kutta method
- Dormand–Prince (fifth order, adaptive step)
- Fehlberg method (sixth order, adaptive step)
- Fehlberg method (seventh order, adaptive step)
- The adaptive timestep methods incorporate a lower-order scheme, which provides an estimate of the error on the obtained results. Using Eq. (17) [21], in which \( O \) stands for the order of the solver, it is then possible to estimate an optimal timestep \( dt_{\text{opt}} \) based on the error tolerance \( \epsilon \), which can be set by the user.

\[
dt_{\text{opt}} = dt_{\text{current}} \left( \frac{\epsilon}{dt_{\text{current}} \tau_{\text{max}}} \right)^{(1/O)}
\]

Note that adaptive timesteps cannot be used when using the stochastic thermal field, as the size of this field depends on the timestep (Eq. 13), which turns Eq. (17) unstable. Because the thermal fields should be uncorrelated in time [9], the timestep should not be set smaller than 1e-13 s. Table 1 gives an overview of the different properties for the two implementations for thermal effects.

### 2.3 Particle size distribution

As the distribution of nanoparticle diameters \( D \) often follows a lognormal distribution [6, 38], the diameter of the particles in Vinamax can also be drawn from such a distribution. Equation (18) shows the probability density

\[
f_0 = \frac{\alpha y}{1 + \alpha^2} \sqrt{\frac{H^2 M}{2\pi k_B T}}
\]

with \( H_k = 2K_{u1}/M_s \) equal to the coercive field.

This algorithm only requires work of \( O(N\log(N)) \), while the direct pairwise evaluation of the demagnetizing field (from now on called brute force method) between \( N \) particles (Eq. 10) scales as \( O(N^2) \). These statements are validated in Sect. 3.
function \( P(D) \) for a lognormal distribution with \( \mu \) the mean diameter and \( \sigma \) the standard deviation of the logarithm of the diameter distribution.

\[
P(D) = \frac{1}{\sqrt{2\pi} \sigma D} \exp \left( -\frac{\ln^2(D/\mu)}{2\sigma^2} \right)
\]  \( (18) \)

3 Results

The use of the macrospin approximation in Vinamax has the advantage that exchange interactions do not have to be evaluated. However, this has the drawback that the micromagnetic standard problems [22], which do incorporate exchange interactions, cannot be solved by Vinamax. In this section, we therefore simulate some simple, yet general, problems and compare their solution with the micromagnetic simulation software \( \text{MuMax3} \) [35].

3.1 Problem one: precession and damping

In this first problem, an isotropic single particle is considered at 0 K. It is initialized with the magnetization along the \( x \)-direction, and an external field of 10 mT is applied along the \( z \)-direction. The magnetization gyrates around this axis with a frequency of 28 GHz/T and slowly damps toward the \( z \)-axis.

In Fig. 2, the results of the simulation are shown. The oscillations in the \( x \)- and \( y \)-components of the magnetization have a frequency of 0.28 GHz, which is in agreement with both the theoretically expected value and the result obtained with \( \text{MuMax3} \). The steady increase of the \( z \)-component of the magnetization corresponds with the damping toward the \( z \)-axis (\( \alpha = 0.02 \)).

3.2 Problem two: magnetostatic interaction

The aim of the second problem is to show that the brute force method to calculate the demagnetizing field is implemented correctly. To this end, at 0 K, two isotropic nanoparticles relax in the presence of an external field of 1 mT along the \( x \)-axis. The same simulation is also repeated without calculating the demagnetizing field to see that this problem is suited to validate the implementation; i.e., to see that, the demagnetizing field has an influence.

The nanoparticles have a diameter of 32 nm and a saturation magnetization of 860,000 A/m. The damping constant \( \alpha \) was set to 0.1.
The brute force evaluation of the demagnetizing field in this problem and that the simulation results correspond well with each other.

### 3.3 Problem three: dipole approximation method

This example shows the agreement between the dipole approximation implementation and the brute force implementation. The same problem is also solved without taking this interaction into account so to illustrate that it is of importance in this system.

In this problem, 256 isotropic nanoparticles with a diameter of 32 nm and saturation magnetization 860,000 A/m are created with a spatially uniform distribution in a cube with a side of 2 μm with their magnetization along the z-axis. They relax at 0 K in the presence of an external field of 1 mT along the x-axis (α = 0.1). In Fig. 4, the results of these simulations are visualized, and we can conclude that there is a good correspondence between the dipole approximation method and the brute force method.

We have also investigated the performance of the different methods for evaluating the demagnetizing field in Vinamax. Figure 5 shows the scaling behavior of the time it takes to calculate one timestep with the brute force evaluation of the demagnetizing field versus the dipole approximation method. All simulations were performed on an Intel Core i7-3770 CPU @ 3.40 GHz. The brute force method scales as $O(N^2)$. The dipole approximation method used a β of 0.7 as this was determined to be sufficiently low value [21]. This method is slower for smaller numbers of particles, but scales as $O(N \log(N))$. From 512 particles on, the dipole approximation method is faster than the brute force calculation, and for large numbers of particles, this results in an enormous speedup of the simulation. Note that we adapted the volume of the simulation to the number of particles to keep the concentration of the particles constant.

### 3.4 Application: thermal switching time distribution and magnetization relaxation

In a ensemble of single-domain particles with uniaxial anisotropy, there exist two ground states along the anisotropy axis for every particle. Due to thermal fluctuations, the magnetization of each particle will switch between these two directions at random intervals. The total magnetization of the ensemble will thus slowly relax toward 0. The rate of this relaxation is given by the Néel relaxation time [29] $\tau_N$, Eq. (19).

$$\tau_N = \tau_0 \exp \left( \frac{K_{\text{el}} V}{k_B T} \right)$$  \hspace{1cm} (19)

In this equation, $\tau_0$ is an extinction time with values between $10^{-8}$ and $10^{-12}$ s [5, 38] and is related to $f_0$ (Eq. (15)) by $\tau_0 = \frac{1}{f_0}$. First, we look at the distribution of switching times of uniaxial particles. As can be seen in Fig. 6, both approaches [a stochastic thermal field (Sect. 2.1.5) and stochastic switching (Sect. 2.1.6)] show good agreement in the
distribution of switching times. Therefore, we use the stochastic switching implementation to look at the result of many of these switches on the magnetization.

The magnetization of an ensemble of nanoparticles with a narrow size distribution is described by the moment superposition model [15, 16] (Eq. 20).

\[ m = \int_V m_0 \exp \left( -\frac{t}{\tau_N(V)} \right) P(V)dV \]  

(20)

which equals \( m = m_0 \exp \left( -\frac{t}{\tau_N} \right) \) for particles of equal size. In this model, the magnetization at the start of the relaxation is determined by Eq. (21) which describes a magnetization process of time \( t_{\text{mag}} \) in a field \( H \), with \( L(V, H, T) \) the Langevin function (Eq. (22)).

\[ m_0 = L(V, H, T) \left[ 1 - \exp \left( -\frac{t_{\text{mag}}}{\tau_N(H)} \right) \right] \]  

(21)

\[ L(V, H, T) = \coth \left( \frac{M_s V \mu_0 H}{K_B T} \right) - \frac{K_B T}{M_s \mu_0 H} \]  

(22)

Furthermore, during the magnetization of the ensemble, \( \tau_N(H) \) differs from \( \tau_N \) in the absence of an external field, and is given by Eq. (23), where the factor 0.82 results from the random orientation of the anisotropy axes [15].

\[ \tau_N(H) = \tau_0 \exp \left( \frac{K_{\text{u1}} V}{k_B T} \left( 1 - \frac{0.82 \mu_0 H M_s}{K_{\text{u1}}} \right) \right) \]  

(23)

We have simulated three ensembles of nanoparticles with different size distributions, as shown in Fig. 7a. Each ensemble was magnetized for 1 s in a field of 1,600 A/m as described by the moment superposition model. The magnetic relaxation of the sample was then calculated by the moment superposition model (black lines in Fig. 7b) and simulated by Vinamax. The particles had a saturation magnetization of 400,000 A/m, a uniaxial anisotropy of 10,000 J/m³ and were simulated at 300 K with \( \alpha = 0.05 \).

The results are shown in Fig. 7b and show that there is in general a good correspondence between both models. The two curves coincide exactly for the particles with an infinitely sharp diameter distribution. There is a small deviation between both models for wider diameter distributions. This results from the fact that in Vinamax \( \tau_0 \) has a particle volume dependency (Eq. 15), while this is not accounted for in the moment superposition model.

4 Discussion

The aim of this paper is to present Vinamax: A numerical software package that performs micromagnetic simulations of magnetic nanoparticles, approximated by macrospins. We have validated Vinamax in various problems against other micromagnetic software (MuMAX3) and analytical results. Each problem shows that a different part of Vinamax is implemented correctly and the results correspond well to the expected ones. In Sect. 3.4, Vinamax is then used to simulate a relaxometry experiment and the resulting magnetization curve corresponds with the one obtained with the moment superposition model, which is typically used to describe these experiments [15, 16]. A small deviation was seen in the case of a broad distribution of nanoparticles which is due to the fact that in Vinamax the attempt frequency is volume dependent, while this is considered constant in the moment superposition model.

Due to the dipole approximation method that scales as \( O(N \log(N)) \) with \( N \) the number of particles, it is possible
to simulate systems with large amounts of particles, and on large timescales. We emphasize that Vinamax can be used as a research tool in biomedical applications, where we especially aim at nanoparticle imaging techniques, such as relaxometry where the collective effect of nanoparticles still is not completely understood. In this domain, Vinamax could, for example, be used to investigate the effect of the dipolar interaction on the relaxation curves and to validate or further refine existing models.

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