Can spin-lattice dynamics model hysteresis loops?

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I. INTRODUCTION

For basic research and applications, magnetic studies are of fundamental relevance as they are an important tool for revealing otherwise hidden structural, thermodynamic and physicochemical properties of a material. Hysteresis loops, for instance, are usually measured to characterize the dynamic behavior of bulk samples that exhibit microstructures like grain boundaries, defects, etc, and multiple domains [1][2]. Furthermore, such measurements are also performed for low-dimensional magnets such as magnetic nanoparticles (NPs) or thin films [3]. The power generated by a sample subject to an alternating magnetic field can be directly determined from the shape of a hysteresis loop. In fact, the Specific Absorption Rate (SAR), defined as the absorbed energy per unit of mass, is proportional to the area of this curve [4]. In magnetic hyperthermia [5–7], for instance, SAR provides the heating efficiency of a specific type of magnetic NP.

There are different classical models that are used to describe magnetic hysteresis [1][8]. Perhaps, the most popular of them is that of Stoner-Wohlfarth (SW) [9][10]. This model is relatively simple to evaluate numerically, allowing easy comparison with experimental results. However, the model includes an important number of approximations. Among them, it is considered that the material behaves as a single magnetic domain. This is known as the macrospin approximation, which implies that below a certain length scale, typically on the order of a few nanometers, the internal structure of the material is neglected. In the SW model the Hamiltonian is the sum of a Zeeman term and an uniaxial anisotropy term. Hysteretic behavior arises as a consequence of a simple zero-temperature dynamic protocol: as the external field varies with time, the magnetization can either change continuously while the system remains at an energy minimum, or it can jump abruptly to another value when that minimum becomes unstable. In addition, although in the original model thermal fluctuations were not considered, further approaches have attempted to incorporate temperature effects [11]. Fortunately, micromagnetic simulations allow to overcome many of these limitations [12][13]. Thus, it is possible to study a system made up of a large number of interacting macrospins with a Hamiltonian including any complex energetic term. In particular, the equilibrium or nonequilibrium magnetization dynamics of such model can be calculated by numerically solving the phenomenological stochastic Landau-Lifshitz-Gilbert (sLLG) equation [14]. From first principles it can be demonstrated that this same theoretical framework is valid to carry out atomistic spin dynamic (ASD) simulations [15][16]. This simulation method has been successfully applied to the modeling of hysteresis loops [17][20]. Another alternative to include temperature effects in hysteresis loops is to use Metropolis Monte Carlo (MC) simulations for a fixed lattice [21][22].

More recently, there has been increased interest in studying more realistic models that take into account both spin and lattice degrees of freedom, and the cou-
pling between them [23][27]. These spin-lattice dynamics (SLD) simulations can use a Langevin bath for the lattice and sLLG for the spins [28]. Although this method is very powerful, its implementation can be computationally demanding and, as the complexity of a system increases, the number of parameters that characterize it also increases, and therefore the practical use of this numerical scheme may not be feasible.

In this work we use SLD calculations with sLLG spin dynamics to simulate the hysteresis behavior of a ferromagnetic material. We use physical parameters that are typical for bulk bcc iron. We focus on determining optimal simulation parameters that allow to obtain reliable results at low temperatures. Also, we explore how the hysteresis loops depend on the different physical properties that characterize these magnetic systems: anisotropy, exchange, damping, and lattice vibrations. The paradigmatic SW model is taken as a reference to achieve this goal. The paper is organized as follows. In Sec. II the theoretical models used and the simulation details are presented. In Sec. III the main results of this study are shown. Finally, a summary and the conclusions of the work are drawn in Sec. IV. In order to simplify several parametric studies, as a first approach we run spin dynamics simulations with the positions of the atoms fixed at their ideal equilibrium values. The influence of the spin-lattice coupling is explored and discussed in the last part of Sec. III.

II. METHODS

A. SLD Model

Let us consider an ensemble of \( N \) atoms each endowed with a classical magnetic moment. The Hamiltonian of the model is

\[
\mathcal{H} = \sum_{i=1}^{N} \frac{|\mathbf{p}_i|^2}{2m_i} + \sum_{i,j,i\neq j} V(r_{ij}) + \mathcal{H}_\text{mag},
\]

(1)

The first term in Eq. (1) accounts for the kinetic energy of the \( N \) atoms of the system where \( \mathbf{p}_i \) and \( m_i \) represent, respectively, the linear momentum and mass of the \( i \)-th atom \( (m_i = 55.845 \text{ u for iron}) \). The second term is the interatomic potential describing the interactions between pairs of atoms separated by a distance \( r_{ij} \). As usual in molecular dynamics simulations of metals, we use a classical embedded atom model potential which describes well a broad spectrum of iron properties [29]. The interatomic cutoff distance for this potential was set to 0.57 nm.

The coupling among the spin and lattice degrees of freedom is provided through the last term in Eq. (1), a magnetic Hamiltonian defined as

\[
\mathcal{H}_\text{mag} = -\mu_0 \mu H \cdot \sum_{i=0}^{N} \mathbf{s}_i - \sum_{i,j,i\neq j} J(r_{ij}) \mathbf{s}_i \cdot \mathbf{s}_j + \mathcal{H}_\text{ani}. \quad (2)
\]

Here, \( \mathbf{s}_i \) is a classical unitary vector representing the spin of \( i \)-th atom. The first term in Eq. (2) is the Zeeman energy, the interaction of each spin with an external uniform magnetic field \( \mathbf{H} \), where \( \mu = 2.2 \mu_B \) is the atomic magnetic moment for iron \( (\mu_B \) is the Bohr magneton) and \( \mu_0 \) is the vacuum permeability constant. Note that the Zeeman energy within LAMMPS is defined slightly differently than in Eq. 2. The second term is just a Heisenberg Hamiltonian describing the interaction between spins, where \( J(r_{ij}) \) is an interatomic distance-dependent exchange coupling which is defined as the following Bethe-Slater curve [30][31].

\[
J(r_{ij}) = 4\alpha \left( \frac{r_{ij}}{\delta} \right)^2 \left( 1 - \gamma \left( \frac{r_{ij}}{\delta} \right)^2 \right) e^{-\left( \frac{r_{ij}}{\delta} \right)^2} \Theta(R_c - r_{ij}), \quad (3)
\]

being \( \Theta(R_c - r_{ij}) \) the Heaviside step function and \( R_c \) the cutoff distance. The coefficients in Eq. (3) must be fitted to ab-initio data. In the present work, we have set \( \alpha = 25.498 \text{ meV} \), \( \gamma = 0.281 \), and \( \delta = 0.1999 \text{ nm} \) which fits very well the ab-initio data from Pajda et. al [32]. In addition, we choose \( R_c = 0.35 \text{ nm} \).

Finally, the last term in Eq. (2) is responsible for computing the magneto-crystalline anisotropy. In this work we consider one of two, either uniaxial \( \mathcal{H}_\text{uni} \) or cubic \( \mathcal{H}_\text{cub} \) anisotropy. The corresponding expressions are given in Eqs. (4) and (5).

\[
\mathcal{H}_\text{uni} = -K_1 \sum_{i=1}^{N} (\mathbf{s}_i \cdot \mathbf{n})^2 \quad (4)
\]

and

\[
\mathcal{H}_\text{cub} = \sum_{i=1}^{N} K_1 [(\mathbf{s}_i \cdot \mathbf{n}_1)^2(\mathbf{s}_i \cdot \mathbf{n}_2)^2 + (\mathbf{s}_i \cdot \mathbf{n}_3)^2 + \mathbf{s}_i \cdot \mathbf{n}_1]^2] - K_2 (\mathbf{s}_i \cdot \mathbf{n}_1)^2 (\mathbf{s}_i \cdot \mathbf{n}_2)^2 (\mathbf{s}_i \cdot \mathbf{n}_3)^2. \quad (5)
\]

Here, the unit vectors \( \mathbf{n}_1, \mathbf{n}_2, \) and \( \mathbf{n}_3 \) lie along the three crystallographic directions [100], [010], and [001], respectively. In Eq. (4), \( \mathbf{n} \) is also a unit vector that in general could point along any of these axes. The first term in the cubic anisotropy energy above is defined with different sign within LAMMPS. \( K_1 \) and \( K_2 \) are the magneto crystalline anisotropy constants which we set \( K_1 = 35 \mu \text{eV/atom} \) and \( K_2 = 3.6 \mu \text{eV/atom} \) (equivalents to volumetric anisotropies \( K_{1V} = 470 \text{ kJ/m}^3 \) and \( K_{2V} = 46 \text{ kJ/m}^3 \)). In order to make comparisons, we have used the same value of \( K_1 \) in both anisotropy equations. Since this constant is positive (and also \( K_2 > 0 \)), then the easy axes of magnetization in Eqs. (4) and (5) are given by the unit vector defined above. Note that the values of \( K_1 \) and \( K_2 \) are ten times larger than those usually used to model bulk bcc iron [3]. However, such larger anisotropy magnitude has been considered for Fe NP, and we also consider bulk values for selected runs. As we will discuss later, using a large anisotropy in the simulations helps to quickly stabilize the magnetization of the system. In addition, to
speed up our calculations we have neglected long-range dipolar interactions in the magnetic Hamiltonian. Since we simulate small systems (see below), this approximation should not affect the validity of our results.

**B. Simulation details**

We perform SLD simulations using the SPIN package of the software LAMMPS [33, 34]. The evolution of the system is described by two coupled Langevin equations, one for the spin and another one for the lattice degrees of freedom. In particular, the former is the well known sLLG equation introduced by Brown [14].

Each Langevin equation has a damping term and a random force (or torque) which are connected through the “fluctuation-dissipation” theorem. We have chosen the damping constants equal to $\lambda_s = 0.5$ (for the spin degrees of freedom) and $\lambda_L = 1.0 \ \text{s}^{-1}$ (for the lattice). We use separate Langevin thermostats for the lattice and spin subsystems, but both are set to the same temperature $T = 10 \ \text{K}$. For a more detailed description of these SLD simulations see Ref. [28].

The hysteresis loops were calculated applying an alternating magnetic field $H$, and averaging the curves over up to ten different cycles. The field is varied discretely: remains constant during certain simulation time, and then jumps to reach the value given by a function $H_{\text{max}}\cos(2\pi ft)$. $H_{\text{max}}$ is set to be larger than the expected saturation field, and jumps do not have the same magnitude along the field, given that the simulations time at each field value is kept constant.

The simulation time step was set to 0.1 fs and each cycle took around 2–8 ns, giving MHz frequencies. In particular, we have simulated field frequencies of $f_0$, $f_0/2$, $f_0/4$, and $f_0/8$, with $f_0 = 500 \ \text{MHz}$. These values are equivalent to sweeping rates of approximately $2.2 \times 10^9 \ \text{T/s}$, $1.1 \times 10^9 \ \text{T/s}$, $0.55 \times 10^9 \ \text{T/s}$ and $0.275 \times 10^9 \ \text{T/s}$ respectively. We note that, in general, in experiments hysteresis loops are obtained using fields that change almost continuously, and the measurement of the magnetic moment of a sample can take up to several microseconds. Those time scales are well beyond the feasibility of SLD, or of other simulation methods like ASD since typically, simulations only reach nanosecond scale with sweep rates similar to ours [17, 19]. Nevertheless, as we show in the next section, the hysteresis loops quickly converge to a limiting curve as the frequency is decreased.

Simulations were run for different angles, $\phi = 0^\circ$, $45^\circ$, and $90^\circ$, between the external field $H$ and the easy anisotropy axes. In practice, this was done by changing the uniaxial anisotropy axis directions while keeping $H$ aligned in the [001] direction (z axis). For uniaxial anisotropy, for instance, for $\phi = 90^\circ$ or $45^\circ$ the unit vector $\hat{n}$ was oriented along the [100] or [101] direction. Most simulations where run with magnetic uniaxial anisotropy according to Eq. [4].

In all cases, we use cubic samples of dimensions $(10 \times 10 \times 10) a_0^3$ ($a_0 = 0.286 \ \text{nm}$ is the lattice parameter of bcc Fe), with periodic boundary conditions in all directions. The resulting system contains 2000 atoms. To show that this size is large enough to capture the main hysteresis properties of the model, we also run a few simulations using a larger system with $(15 \times 15 \times 15) a_0^3$ cells (6750 atoms), as shown in the Supplemental Material (SM).

Finally, we have to mention that the total magnetization $M$ as well as all components $M_x$, $M_y$, and $M_z$, will be expressed normalized to the ideal Fe bulk saturation magnetization $M_s$. Saturation magnetization is given by the maximum magnetic moment per unit volume. For bulk iron, in the volume of a bcc unit cell ($a_0^3$) there are 2 spins with magnetic moment $\mu = 2.2 \mu_B$ and, therefore $M_s = \frac{2 \times 2.2 \mu_B}{a_0^3} \approx 1730 \ \text{kA/m}$. In this way, $M = 1.0$ means $M = 1730 \ \text{kA/m}$.

**III. RESULTS AND DISCUSSION**

In this section, we first analyze how the computational parameters, such as the simulation run time and the field frequency, affect the final outcome of the calculations. The aim is to establish optimal parameters that allow minimizing the computational cost without affecting the reliability of the calculations. Next, we focus on studying how the hysteresis loops can change when different physical properties like anisotropy, exchange interaction, and damping parameter, are modified. In a first stage, we analyze these effects without coupling the lattice vibrations to the the spins dynamics, i.e. we consider for most cases spin dynamics simulations with the atoms fixed at their ideal-lattice positions. The influence of coupling the lattice vibrations is analyzed on the final part of this section, where we have run full SLD simulations.

**A. Effect of simulation time**

We start analyzing the effect of the simulation run time $t_{\text{sim}}$. This quantity represents the interval during which the external field $H$ is held constant before increasing or decreasing it by a given amount. It is basically the total time of simulation used to calculate each individual discrete point of a hysteresis loop. After changing the external field, the magnetic moments must relax to a new stable configuration and therefore $t_{\text{sim}}$ must be large enough to allow this process to occur. It is important, then, to determine an optimal value of this quantity that allows to achieve this goal.

In principle, one can imagine an individual spin precessing around the field direction, with a frequency which increases with the field magnitude. For Fe, the resulting Larmor frequency gives a period of 36 ps for a 1T field, and several precession periods are needed to describe magnetization evolution. However, for damped dynamics, the frequency remains the same but the spin will spiral down towards the field direction, allowing shorter
the region around the switching field, before is varied a fluctuation period is observed, specially in M

tion, stabilization of the magnetization along the field direc-

value, so for 90 ps before increasing or decreasing it to the next 
curve and right axis) is maintained at a constant value 
tions we use uniaxial anisotropy. As it can be noticed by 
Unless otherwise stated, for this and subsequent calcula-
culations of hysteresis loops [13, 17–19, 35, 36]. We have 
late the hysteresis loops for the cases of 
φ 

shorter simulation times can be employed.

ever, thanks to the high damping values discussed below, 
limitations of atomistic simulation approaches. Instead, 
it is not possible to calculate hysteresis loops at typical 
energies, from now on we set t_{sim} = 90 ps.

B. Hysteresis loops and the effect of field frequency

As it was argued in Section II, using SLD simulations 
it is not possible to calculate hysteresis loops at typical 
very low experimental frequencies, due to intrinsic time 
limitations of atomistic simulation approaches. Instead, 
we are limited to working with high frequencies in the 
order of MHz. However, high frequency hysteresis loops 
appear to converge to a limit curve as the frequency is 
decreased and, as the following analysis shows, this limit 
loop should not be so different from those measured ex-
perimentally or calculated theoretically. Following this 
line of argumentation, we calculate the hysteresis loops 
for different frequencies. In Fig. 4 we show how the area 
of the hysteresis loops depends on the field frequency for 
the cases with φ = 0° and φ = 45°. Note that we ex-
clude the case φ = 90° from this analysis, since for this 
angle the curves do not present a hysteretic behavior. 
The curves in Fig. 4 exhibit a typical crossover from a 
high dynamic regime (for which the area of the loops is 
large) to a low dynamic regime (a slow variation of the area) [37].

Given that the simulation time for a given field value 
is always the same and, in principle, long enough to cover 
spin relaxation for high damping, higher frequency would 
only mean larger changes in the discrete field values. The 
larger the field jumps, the larger the change across the 
energy landscape, and the system might not adapt fast 
ough within the 90 ps of simulated time, decreasing spin 
switching probability and generating wider loops.

Figure 1. Time evolution of the components of the magnetization throughout an entire cycle (a) and half a cycle (b). The field value at each time is also included (green dashed line and right axis). Note that each field value is kept constant for 90 ps (9 × 10^3 steps) of simulation time. In (b), the average value of M_z ( M_z ) (blue dotted line), obtained from the last 30 ps of simulation, is also added for comparison.

simulations [15]. Figure 1 shows the time evolution of the components of the normalized magnetization along the three Cartesian axes, M_x, M_y, M_z, corresponding to directions [100], [010], and [001], respectively, during the simulation of an entire loop for the case of φ = 90°.

Unless otherwise stated, for this and subsequent calculations we use uniaxial anisotropy. As it can be noticed by inspection of this figure, the applied field (green dashed curve and right axis) is maintained at a constant value for 90 ps before increasing or decreasing it to the next value, so t_{sim} = 90 ps. We are interested here in the stabilization of the magnetization along the field direction, M_z. During the first moments after the field value is varied a fluctuation period is observed, specially in the region around the switching field, before M_z reaches a stable value approximately after 45 ps. This is better appreciated in Fig. 1(b), where a zoom in the region around the first magnetization switching is displayed. In this figure, we have also included the average value of M_z, ( M_z ), which is obtained in our simulations from the last 30 ps of each step, where a well stabilized magnetization is observed. We note that t_{sim} = 90 ps is similar to simulation times used in micromagnetic and ASD calculations of hysteresis loops [13, 17, 19, 35, 36]. We have checked that longer simulation times do not significantly affect the final results, as it is shown in Fig. S3 in the SM. It is crucial that our magnetization dynamics is well described near the switching field values. For fields of 0.5 T, the precession period is ~72 ps, and one would need a few ns of simulation time for low damping. How-
ever, thanks to the high damping values discussed below, shorter simulation times can be employed.

Using t_{sim} = 90 ps and frequency f_0/4, we calculate the hysteresis loops for the cases of φ = 0° and φ = 90°. These curves, along with snapshots showing typical spin configurations at different stages of the process, are shown in Fig. 2. As we can see, the system behaves qualitatively according to what the SW model predicts, i.e., all spins are roughly in sync like a single macrospin [9, 10]. This is expected for low-temperature simulations, but deviations would occur at higher temperatures. Still, Fig. 2 shows that there is a roughly Gaussian distribution of spin values, an appreciable deviation from the simpler macrospin assumption. The spin orientation histograms in this figure correspond to the state of the system described by the snapshot (c) of the φ = 90° case of Fig. 2 (bottom right).

Another observation is the behavior of the magnetization components around H ≈ 0 (being H the modulus of H) for the case of φ = 90°. In Fig. 1 we can see that when this field takes values close to zero, M_z ≈ 0 (and also M_y ≈ 0) while M_x takes values close to saturation. This means that, at this stage, all spins are aligned approximately along the x direction (the easy axis of magnetization for the uniaxial anisotropy), showing that the simulation reproduces well the expected behavior for φ = 90°. The snapshots in Fig. 2 (right panels) confirm this interpretation.

Since these tests confirm that our simulations are consistent with some general physical properties of the system, from now on we set t_{sim} = 90 ps.
Figure 2. Top images: resulting hysteresis loops obtained at the converged frequency for $\phi = 0^\circ$ (top left) and $\phi = 90^\circ$ (top right). Bottom images: snapshots of a fraction of the system, showing typical spin configurations at different stages of the hysteresis loop for the case of $\phi = 0^\circ$ (bottom left) and $\phi = 90^\circ$ (bottom right). Each snapshot, (a), (b), (c) and (d), correspond to the points marked on the hysteresis curves (upper images). In the snapshots, the atoms are represented as red spheres and the spins are colored according to their orientation along the field direction ($S_z$). Each cubic cell represents a fraction of the system with a volume of $(8 \text{ Å})^3$, extracted from the center of the original system.

The magnitude of the jumps around the region of the switching field are $\sim 0.3 \text{ T}$ for $f_0$ and $\sim 0.06 \text{ T}$ for $f_0/4$. Therefore, the stabilization of the frequency for $\phi = 0^\circ$ occurs when the field jumps are about one order of magnitude lower than the value of the converged coercive field.

For $\phi = 45^\circ$, we found convergence at a frequency of $f_0/2$ while for the $\phi = 0^\circ$ case it is not until a frequency of $f_0/4$ that the loop area reaches a stable value. At these frequencies the area of the loops coincides (within the statistical errors) with the predictions of the SW model. Figure S2 in the SM shows the corresponding hysteresis curves calculated at several different frequencies for the case of $\phi = 0^\circ$, evidencing convergence as frequency goes down. This is similar to results showing convergence as loop time increases in ASD simulations [38] for simulation times similar to our SLD simulations. This analysis shows that, although the convergence frequencies found in this study are much higher than those typically used in experiments, they are low enough to allow the magnetization to equilibrate with the field direction at each field step. This means that, at this rates, the ballistic regime is avoided and, therefore, our simulated hysteresis loops should not present large discrepancies with experiments, as discussed for example by Westmoreland et. al [19].

In Fig. 5 we compare the hysteresis loops calculated for $\phi = 0^\circ$, $\phi = 45^\circ$, and $\phi = 90^\circ$ at a single frequency $f_0/4$, with those of the SW model [3]. As we can see, a good agreement is observed in all cases. Nevertheless, it is important to stress that we do not expect that the simulation curves to fit exactly to the theoretical ones. Unlike the SW model (for which the macrospin approximation is considered at $T = 0$), the SDL simulations are carried out at low but finite temperature for many magnetic atomic moments which, along a hysteresis cycle, do not rotate coherently in a perfect way (see Fig. 3). For example, for the $\phi = 0^\circ$ case, we do not obtain a rectangular curve because thermal fluctuations and also the
existence of many degrees of freedom, make it easier for the magnetization to change its orientation at a external field value slightly lower than predicted by the SW model.

C. Effect of anisotropy

According to the SW model, the coercive field and, therefore, the area of the hysteresis loop when the external field is applied along the easy axis (φ = 0°), is proportional to the anisotropy constant $K_1$ in Eq. (1). We have explored this dependency in our SLD simulations by reducing and increasing the original anisotropy constant value (throughout this test called $K_1^*$) by a factor of 10. Figure 6 (a) shows that the resulting simulated loops qualitatively exhibit the expected behavior, that is, the loop broadens (narrow) according to the increase (reduction) of the anisotropy constant. This is further confirmed in Fig. 6 (b) where it can be seen that the loop area follows an approximately linear behavior as a function of $K_1/K_1^*$, deviating slightly from the SW model prediction only for small values of this constant.

These deviations arise from the combination of two effects. On one hand, this is due to the fact that simulated loops are “rounded” near the point of spin reversal, deviating from perfect rectangular loops as explained above. The discrepancy with the SW model area become increasingly important as the loop narrows. On the other hand, as it was mentioned in the methods section, large anisotropy helps stabilizing the magnetization of the system [18]. For the case of low anisotropy ($K_1 = 0.1K_1^*$) there is poor stabilization of $M_z$ during the 90 ps of simu-
ulation of each field step (specially near the region of magnetization switching), as seen in Figure S6. Much longer simulation times would be required for low anisotropy values. Given that our smallest anisotropy is the Fe bulk anisotropy, we could compare with some experimental results. MOKE measurements of Fe(100) appear to give an slightly lower coercive field than our simulation estimate [39], although those measurements were obtained at higher temperatures and for multi-domain samples.

We have also analyzed the influence of anisotropy symmetry. Loops calculated with SLD simulations for systems with uniaxial and cubic anisotropies, Eqs. 4 and 5 are shown in Fig. 7 for the case $\phi = 0^\circ$. We compare these curves with the corresponding one for the SW model with uniaxial anisotropy. The reason is that at $T = 0$ K both simulation curves should coincide with the latter. Our results show that for the case of $\phi = 0^\circ$ the hysteresis loop does not depend on the type of anisotropy. Therefore, the small differences between the curves shown in Fig. 7 can be attributed to the effect of temperature, which is more pronounced in the case of cubic anisotropy. The area of the loop is slightly smaller for cubic anisotropy, similar to the results reported in ref. [35]. Usov and Peschany [40] considered the SW model for uniaxial and cubic anisotropies, for randomly oriented nanoparticles. They report that the maximum normalized coercive field in both cases is 1, in agreement with our result. For a random collection of anisotropy orientations, the resulting coercive field for cubic anisotropy was $\sim 0.68$ of the one for uniaxial.

It is more interesting to analyze the difference between uniaxial and cubic anisotropies, when the field is applied in the [111] direction. For this field orientation, we consider a SW model whose Hamiltonian has an arbitrary anisotropy term. We start by applying an external field strong enough such that there is a single energy minimum for this Hamiltonian, for which the magnetization $M$ points in the direction of $H$. Then, we decrease the intensity of the field using small jumps. At each step we use a steepest descent method to try to escape from the energy minimum, so that if it becomes unstable, a new minimum can be reached. Using this simple algorithm, we have simulated the zero temperature dynamics of the SW model with both uniaxial and cubic anisotropies. As shown in Fig. 8 in this case the corresponding hysteresis loops for the SW model are very different. However, as expected, the loops calculated with the SLD simulations agrees very well with the curves for the SW model for both, cubic and uniaxial anisotropies. In the case of cubic anisotropy, while the remanent magnetization for
both curves (SW and SLD) is $\pm 1/\sqrt{3}$, there is a qualitative difference between them for field values greater than the coercive one: a small hysteresis behavior present in the loop of the SW model is not well reproduced in the SLD simulation. The reason for this is that the energy barrier separating these states is of approximately 0.3 $\mu$eV/atom, several orders of magnitude smaller than $K_B T \sim 8.6 \times 10^{-3}$ eV, where $K_B$ is the Boltzmann constant. Only a simulation at much lower temperature would be able to reproduce this behavior. The cubic case has a lower coercive field, which could be somewhat expected from results for a collection of NP with random orientation\[40].

\[\phi \approx 54.7^\circ \]
\[f_0/4\]

Figure 8. Hysteresis loops for an external field applied along the [111] direction ($\phi \approx 54.7^\circ$) and frequency $f_0/4$, for cubic and uniaxial anisotropy. The loops obtained with SLD are compared to the predictions of an extension of the SW model that incorporates cubic anisotropy (red short-dotted line), and to the SW with uniaxial anisotropy (green dot-dashed line).

D. Effect of exchange interactions

Exchange energy does not play a role in SW, and within that framework there should be no effect from the exchange interaction on the simulated hysteresis loops. To test this within the SLD framework, we have run simulations with different values of the function $J(r_{ij})$. In Fig. 9 we compare the curves obtained using the original exchange $J(r_{ij})^*$, with the ones obtained by increasing or reducing this interaction by a factor of 10. As it can be seen, loops are nearly unaffected by these changes. Increasing the magnitude of the exchange interaction is expected to give higher values of magnetization and broader loops\[41], but for low temperatures (as in this case) this effect is expected to be very small. Larger differences should be noticed at higher temperatures where thermal fluctuations start to play a major role in the magnetization behavior. Nevertheless, even at this low temperatures, a few subtle effects can be noticed: (i) for the simulation with $J(r_{ij}) = 0.1 \times J(r_{ij})^*$ a small but consistent reduction of saturation magnetization is observed, and (ii) for $J(r_{ij}) = 10 \times J(r_{ij})^*$ the loop becomes more rectangular. This last effect is also observed in other theoretical approaches based on the SW model which include exchange interactions through a mean field approach as in Refs.\[42, 43\]. Lower/higher exchange will lead to lower/higher Curie temperature and spin ordering will be modified. For the same value of external field, the histogram of spin values is narrower and with a higher mean value for the higher exchange, facilitating the macrospin behavior, leading to a larger saturation field and helping with the sudden spin flip which causes a more square loop.

Figure 9. Simulated hysteresis curves for the case of $\phi = 0^\circ$ considering different magnitudes of the exchange coupling.

E. Effect of damping parameter

The value of the Gilbert damping parameter in the sLLG equation is usually chosen to be in the range 0.01–1. Experimental estimates for Fe are around 0.001. However, the possibility of increasing typical spin damping values for numerical convenience is often explored. In SLD simulations a value of $\lambda_s = 0.1$ was used in ref.\[26\]. ASD\[44\] and micromagnetic\[13, 35\] simulations use values in the range 0.1–1.

In general, as the damping increases, there is a faster energy loss, and the relaxation time required for the system to reach a steady state decreases, with spins aligned with the preferred orientation determined by the external field and the anisotropy. Therefore, it is possible to use higher frequencies to calculate the hysteresis loops, which become narrower as damping grows. In the same
Figure 10. (a) Hysteresis curves for the case of $\phi = 90^\circ$ with a field frequency of $f_0$ and with damping parameters of $\lambda_s = 0.1$ (dashed line) and $\lambda_s = 0.5$ (full line). The plot shows several cycles for each case. (b) Time evolution of the components of the magnetization throughout the simulation of half a cycle for the cases considered in (a). The field value at each time is also included (green dashed line and right axis). Note that each field value is kept constant for $90 \text{ ps} \left(9 \times 10^5 \text{ steps} \right)$ of simulation time.

In most of the simulations presented here we use a larger damping value, $\lambda_s = 0.5$. Using lower damping is possible but would require lower frequencies as discussed below. The parameter $\lambda_s$ is related to the optimal value of the simulation run time $t_{\text{sim}}$.

In micromagnetic simulations, it has been proposed to use both very high damping and high loop sweep rates (SR), in order to achieve computational efficiency [13]. The rationale behind this proposal was as follows. The magnitude of the coercive field is a function of the measurement time, and the attempt frequency for spin flips. This measurement time can be related to SR, and the flip frequency can be assumed proportional to damping. Therefore, loops calculated with the same value of SR/$\lambda_s$ would have the same coercive field which gives the loop width. Based on this, they were able to use high SR in their simulations to obtain hysteresis loops that closely match those obtained at very low SR, by employing extremely high damping values, and large time steps. Recently, this methodology was successfully applied to simulate hysteresis of iron oxide magnetic nanoparticles with application to hyperthermia [30].

In Fig. 10 we use $f_0$ for the case with $\phi = 90^\circ$. For $\lambda_s = 0.5$ the hysteresis loop in (a) is reasonably close to the SW prediction, and the magnetization versus time in (b) indicates reasonable convergence. However, for $\lambda_s = 0.1$, the loop in (a) is poorly defined, as expected from the lack of convergence towards stable magnetization values shown at the top of panel (b). The possibility of reducing the simulation time in micromagnetic simulations by increasing both, the SR and the damping, leaving constant the ratio SR/$\lambda_s$, would be equivalent, in our case, to keep constant the ratio $f/\lambda_s$. However, SLD simulations at high damping might depart from the desired dynamics and should be treated with care. In our simulations, by choosing $\lambda_s = 0.5$, we were able to set $t_{\text{sim}} = 90 \text{ ps}$ and use $f_0/4$ as discussed at the beginning of this section. For lower values of damping, smaller frequencies would be required, significantly increasing the computational costs. Following the previous scaling, in instance, for $\lambda_s = 0.1$, around 400 ps ($4 \times 10^6 \text{ steps} \right)$ are required to stabilize magnetization at each field value, in contrast with the 90 ps ($9 \times 10^5 \text{ steps} \right)$ that are required for $\lambda_s = 0.5$. We have run some simulations for such a low damping and nanosecond steps, and verified that this indeed the case, explaining the lack of convergence in Fig. 10(a).

F. Effect of coupling the lattice dynamics

All the results presented up to this point correspond to frozen-lattice simulations, meaning that the lattice vibrations are not included in the dynamics. We now analyze the effect of full SDL calculations actually coupling the spin and lattice degrees of freedom, which we refer as moving-lattice simulations. In Fig. 11 we compare the simulated hysteresis loops for the case of $\phi = 0^\circ$ obtained with (moving-lattice) and without (frozen-lattice) spin-lattice coupling at different frequencies. The former gives lower coercivity and narrower loops in comparison with the frozen-lattice case. This effect is more pronounced for higher frequencies. This is consistent with the fact that in a system that incorporates new degrees of freedom (in this case those of the lattice), the energy barrier that must be overcome to reverse the magnetization must decrease. In other words, when a reversal field is applied, a moving lattice provides additional paths for the magne-
zation relaxation process. However this effect is small at a frequency $f_0/4$ where stable magnetization is easily achieved for the chosen sweep rate, showing that lattice dynamics contributions are not very important at this low temperatures ($T = 10$ K), as expected. Larger differences are expected as the temperature increases. As it was previously shown for iron NP at equilibrium, these contributions would become significant at temperatures higher than 300 K [26, 45]. The equilibrium magnetization for a moving lattice is always smaller than that of a frozen lattice for all temperatures [26]. In addition, in the present approach the anisotropy contributions do not depend on the lattice. Larger differences between moving and frozen lattice simulations might be observed for Hamiltonians that include anisotropy terms which are sensitive to lattice details, like orientation, vibrations, and atomic volume [46, 47].

**IV. SUMMARY AND CONCLUSIONS**

We use Spin-Lattice Dynamic (SLD) simulations to calculate the hysteresis loops of a bulk ferromagnetic system at a low temperature. SLD can include temperature effects and lattice defects in a relatively simple way, complementing spin-dynamic simulations and micromagnetic simulations.

In order to validate our SLD results, we compare to the Stoner-Wolfhart (SW) model, which is widely used in experimental and simulation studies. In the SW model, there is no lattice, no temperature, and no exchange interactions. It assumes uniaxial anysotropy and Zeeman energy contributions, and spins in the volume are assumed to behave like a single macrospin. We present comparisons for different angles between the external field and the anisotropy axis. We analyze the effects of several parameters like simulation run time, field frequency, damping, exchange, anisotropy and lattice-vibrations. We find that loops obtained with SLD agree very well with those calculated with SW, provided that several parameters are chosen with care.

Given the short timestep required to integrate the atomic degrees of freedom, usually $\sim 0.1$ fs - 1 fs, one needs relatively long simulations to achieve a stable magnetization at a given value of the applied magnetic field. Typically, this would involve several precession periods. Using the Larmor frequency for Fe gives a period of $\sim 36$ ps for a field of 1T. In order to speed-up spin-dynamics convergence, a Gilbert damping $\lambda = 1.0$ is used in most ASD simulations [17, 20] and values much larger than 1 have been used in micromagnetic simulations of hysteresis loops [13]. In our simulations, we find that for a given damping, as frequency decreases, the loop area also decreases until reaching a constant value. For bulk iron, and a Gilbert damping of 0.5, we find that frequencies of 125 MHz or lower provide such a constant loop area, for simulation times of 90 ps for each field value. Values between 30 and 200 ps (depending on the system under study and the Gilbert damping employed) are also used for ASD simulations [17, 18, 20]. For the case where the anisotropy axis and the field direction are perpendicular, $\phi = 90^\circ$, a converged loop can be found at even higher frequencies. Agreement between SLD and SW suggests that loops using this limiting frequency would not be much different from the experimental or theoretical estimates. This result partially solves the problem related to the computational costs in atomistic simulations of hysteresis loops, allowing to reproduce experimental hysteresis loops, running SLD simulations at much higher frequencies than those typically used in experiments.

From our simulations at a temperature of 10 K, spins behave almost like a macrospin, but there is some expected spread in magnetization values, associated with thermal effects and exchange interactions. The exchange energy, which is much larger than Zeeman or anisotropy energies, does not significantly affect hysteresis loops at low temperature. The coercive field depends linearly on
the magnitude of the uniaxial anisotropy, as in the SW model. Cubic anisotropy and uniaxial anisotropy produce nearly the same loops for the external field aligned with an easy anisotropy axis. However, there are important differences for the case of $\phi \approx 54.7^\circ$ misalignment, where the applied field lies along the [111] direction. For this case, cubic anisotropy leads to a lower coercivity value.

When the atoms vibrations are coupled to spin dynamics, we find that lattice dynamics contributions depend on the loop frequency, but they are very small, as expected for such a low temperature. SLD simulations of hysteresis loops at higher temperature, or including anisotropy terms sensitive to lattice details [46, 47], are expected to behave differently than the simulations presented here.

Future SLD simulations can be applied to hysteresis loops for systems of technological interest, including nanoparticles [48] and bulk systems with defects [20, 45, 49–51], providing information which can inform micromagnetic and atomistic spin dynamics simulations. Large computer cluster allow for simulations of increasingly larger systems by distributing spatial scales, but time scale cannot be split amongst different parallel processes. Therefore, new approaches for accelerated dynamics [52], bypassing this time scale problem, will be required for more efficient simulations of hysteresis loops.

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[1] G. Bertotti, Hysteresis in Magnetism (Academic Press, Elsevier, London, 1998).
[2] I. Mayergoyz, Mathematical Models of Hysteresis and their Applications (Academic Press, Elsevier, London, 2003).
[3] B. D. Cullity and C. D. Graham, Introduction to magnetic materials (John Wiley & Sons, New Jersey, 2011).
[4] B. Mehdaoui, A. Meffre, J. Carrey, S. Lachaize, L.-M. Lacroix, M. Gougeon, B. Chaudret, and M. Respaud, Optimal size of nanoparticles for magnetic hyperthermia: a combined theoretical and experimental study, Advanced Functional Materials 21, 4573 (2011).
[5] Z. Shaterabadi, G. Nabiyouni, and M. Soleymani, Physics responsible for heating efficiency and self-controlled temperature rise of magnetic nanoparticles in magnetic hyperthermia therapy, Progress in biophysics and molecular biology 133, 9 (2018).
[6] Q. A. Pankhurst, J. Connolly, S. K. Jones, and J. Dobson, Applications of magnetic nanoparticles in biomedicine, Journal of physics D: Applied physics 36, R167 (2003).
[7] Z. Hedayatnasab, F. Abnisa, and W. M. A. W. Daud, Review on magnetic nanoparticles for magnetic nanofluid hyperthermia application, Materials & Design 123, 174 (2017).
[8] F. Liorzou, B. Phelps, and D. Atherton, Macroscopic models of magnetization, IEEE Transactions on Magnetics 36, 418 (2000).
[9] E. C. Stoner and E. Wohlfarth, A mechanism of magnetic hysteresis in heterogeneous alloys, Philosophical Transactions of the Royal Society of London. Series A, Mathematical and Physical Sciences 240, 599 (1948).
[10] C. Tannous and J. Gieraltowski, The Stoner–wohlfarth model of ferromagnetism, European journal of physics 29, 475 (2008).
[11] L. Lanci and D. V. Kent, Introduction of thermal activation in forward modeling of hysteresis loops for single-domain magnetic particles and implications for the interpretation of the day diagram, Journal of Geophysical Research: Solid Earth 108 (2003).
[12] G. Bertotti, I. Mayergoyz, and C. Serpico, Nonlinear magnetization dynamics in nanosystems (Elsevier, Oxford, 2008).
[13] R. Behbahani, M. L. Plumer, and I. Saika-Voivod, Coarse-graining in micromagnetic simulations of dynamic hysteresis loops, Journal of Physics: Condensed Matter 32, 35LT01 (2020).
[14] W. F. Brown, Thermal fluctuations of a single-domain particle, Physical Review 130, 1677 (1963).
[15] R. F. Evans, W. J. Fan, P. Chureemart, T. A. Ostler, M. O. Ellis, and R. W. Chantrell, Atomistic spin model simulations of magnetic nanomaterials, Journal of Physics: Condensed Matter 26, 103202 (2014).
[16] O. Eriksson, A. Bergman, L. Bergqvist, and J. Hellsvik, Atomistic Spin Dynamics (Oxford University Press, Oxford, 2017).
[17] D. Böttcher, A. Ernst, and J. Henk, Atomistic magnetization dynamics in nanostructures based on first principles calculations: application to co nanoislands on cu (111), Journal of Physics: Condensed Matter 23, 296003 (2011).
[18] J. Alzate-Cardona, D. Sabogal-Suárez, R. Evans, and E. Restrepo-Parra, Optimal phase space sampling for monte carlo simulations of heisenberg spin systems, Journal of Physics: Condensed Matter 31, 095802 (2019).
[19] S. C. Westmoreland, C. Skelland, T. Shoji, M. Yano, A. Kato, M. Ito, G. Hrkac, T. Schrefl, R. F. Evans, and R. W. Chantrell, Atomistic simulations of α-fe/nd2fe14b magnetic core/shell nanocomposites with enhanced energy product for high temperature permanent magnet applications, Journal of Applied Physics 127, 133901 (2020).
[20] S. Jenkins, R. W. Chantrell, and R. F. Evans, Atom-
istic origin of the athermal training effect in granular irmn/cofe bilayers, Physical Review B 103, 104419 (2021).

[21] T. Tajiri, H. Deguchi, M. Mito, K. Konishi, S. Miyahara, and A. Kohno, Effect of size on the magnetic properties and crystal structure of magnetically frustrated dynn 2 o 5 nanoparticles, Physical Review B 98, 064409 (2018).

[22] R. Essajai, Y. Benhouria, A. Rachadi, M. Qjani, A. Mzerd, and N. Hassanain, Shape-dependent structural and magnetic properties of fe nanoparticles studied through simulation methods, RSC advances 9, 22057 (2019).

[23] T. Shimada, K. Ouchi, I. Ikeda, Y. Ishii, and T. Kitanura, Magnetic instability criterion for spin–lattice systems, Computational Materials Science 97, 216 (2015).

[24] P.-W. Ma, S. Dudarev, and C. Woo, Spilady: A parallel cpu and gpu code for spin–lattice magnetic molecular dynamics simulations, Computer Physics Communications 207, 350 (2016).

[25] X. Wu, Z. Liu, and T. Luo, Magnon and phonon dispersion, lifetime, and thermal conductivity of iron from spin-lattice dynamics simulations, Journal of Applied Physics 123, 085109 (2018).

[26] G. Dos Santos, R. Aparicio, D. Linares, E. Miranda, J. Tranchida, G. Pastor, and E. Bringa, Size-and temperature-dependent magnetization of iron nanoclusters, Physical Review B 102, 184426 (2020).

[27] G. dos Santos, R. Meyer, R. Aparicio, J. Tranchida, E. M. Bringa, and H. M. Urbassek, Spin-lattice dynamics of surface vs core magnetization in fe nanoparticles, Applied Physics Letters 119, 012404 (2021).

[28] J. Tranchida, S. Plimpton, P. Thibadeau, and A. P. Thompson, Massively parallel symplectic algorithm for coupled magnetic spin dynamics and molecular dynamics, Journal of Computational Physics 372, 406 (2019).

[29] H. Chamati, N. Papanicolaou, Y. Mishin, and D. Papaconstantopoulos, Embedded-atom potential for fe and its application to self-diffusion on fe (1 0 0), Surface Science 600, 1793 (2006).

[30] T. Kaneyoshi, Introduction to amorphous magnets (World Scientific Publishing Company, Singapore, 1992).

[31] K. Yosida, D. C. Mattis, and K. Yosida, THEORY OF MAGNETISM.: Edition en anglais, Vol. 122 (Springer Science & Business Media, Berlin, 1996).

[32] M. Pajda, J. Kudrnovský, I. Turek, V. Drchal, and P. Bruno, Ab initio calculations of exchange interactions, spin-wave stiffness constants, and curie temperatures of fe, co, and ni, Physical Review B 64, 174402 (2001).

[33] S. Plimpton, Fast parallel algorithms for short-range molecular dynamics, Journal of Computational Physics 117, 1 (1995).

[34] http://lammps.sandia.gov/doc/fix_nve_spin.html

[35] D. Aurélio and J. Vepravová, Understanding magnetization dynamics of a magnetic nanoparticle with a disordered shell using micromagnetic simulations, Nanomaterials 10, 1149 (2020).

[36] R. Behbahani, M. L. Plumer, and I. Saika-Voivod, Multiscale modelling of magnetostatic effects on magnetic nanoparticles with application to hyperthermia, Journal of Physics: Condensed Matter 33, 215801 (2021).

[37] M. I. Dölz, S. D. Calderón Rivero, H. Pastoriza, and F. Romá, Magnetic hysteresis behavior of granular manganese la6.7ca0.3mno3 nanotubes, Physical Review B 101, 174425 (2020).

[38] P. Chureemart, R. Evans, R. Chantrell, P.-W. Huang, K. Wang, G. Ju, and J. Chureemart, Hybrid design for advanced magnetic recording media: Combining exchange-coupled composite media with coupled granular continuous media, Physical Review Applied 8, 024016 (2017).

[39] J. Bansmann, M. Getzlaff, C. Westphal, and G. Schönhense, Surface hysteresis curves of fe (110) and fe (100) crystals in ultrahigh vacuum—evidence of adsorbate influences, Journal of magnetism and magnetic materials 117, 38 (1992).

[40] N. Usov and S. Peschany, Theoretical hysteresis loops for single-domain particles with cubic anisotropy, Journal of magnetism and magnetic materials 174, 247 (1997).

[41] H. Kachkachi, A. Eziz, M. Nogues, and E. Tronc, Surface effects in nanoparticles: application to maghemite-fe o, The European Physical Journal B 14, 681 (2000).

[42] D. L. Atherton and J. Beattie, A mean field stoner-wohlfarth hysteresis model, IEEE transactions on magnetics 26, 3059 (1990).

[43] H.-w. Zhang, S.-y. Zhang, B.-g. Shen, and H. Kronmüller, The magnetization behavior of nanocrystalline permanent magnets based on the stoner–wohlfarth model, Journal of magnetism and magnetic materials 260, 352 (2003).

[44] R. F. Evans, Atomic spin dynamics, in Handbook of Materials Modeling: Applications: Current and Emerging edited by W. Andreoni and S. Yip (Springer, Cham, 2020) pp. 427–448.

[45] R. Meyer, G. dos Santos, R. Aparicio, E. M. Bringa, and H. M. Urbassek, Influence of vacancies on the temperature-dependent magnetism of bulk fe: A spin-lattice dynamics approach, Computational Condensed Matter , e00662 (2022).

[46] P. Nieves, J. Tranchida, S. Arapan, and D. Legut, Spin-lattice model for cubic crystals, Physical Review B 103, 094437 (2021).

[47] W. Dednam, C. Sabater, A. E. Botha, E. B. Lombardi, J. Fernández-Rossier, and M. J. Caturla, Spin-lattice dynamics simulation of the einstein–de haas effect, Computational Materials Science 209, 111359 (2022).

[48] A. L. S.isch, and P. Bender, Embracing defects and disorder in magnetic nanoparticles, Advanced Science 8, 2002682 (2021).

[49] D. Palanisamy, A. Kovács, O. Hegde, R. E. Dunin-Borkowski, D. Raabe, T. Hickel, and B. Gault, Influence of crystalline defects on magnetic nanodomains in a rare-earth-free magnetocrystalline anisotropic alloy, Physical Review Materials 5, 064403 (2021).

[50] L. Han, Z. Rao, I. R. Souza Filho, F. Maccari, Y. Wei, G. Wu, A. Ahmadian, X. Zhou, O. Gutfeisch, D. Ponge, et al., Ultrastrong and ductile soft magnetic high-entropy alloys via coherent ordered nanoprecipitates, Advanced Materials 33, 2102139 (2021).

[51] M. Bersweiler, E. P. Sinaga, I. Peral, N. Adachi, P. Benner, N.-J. Steinke, E. P. Gilbert, Y. Todaka, A. Michels, and Y. Oba, Revealing defect-induced spin disorder in nanocrystalline ni, Physical Review Materials 5, 044409 (2021).

[52] S. J. Plimpton, D. Perez, and A. F. Voter, Parallel algorithms for hyperdynamics and local hyperdynamics, The Journal of Chemical Physics 153, 054116 (2020).
SUPPLEMENTARY MATERIAL

This supplementary material presents extra figures intended to expand the discussion and provide extra evidence and support the points discussed in the main text.

A. Average curve

Figure S1 presents an example of how the resulting hysteresis loops were obtained. The resulting curve was obtained by averaging several individual cycles.

B. Effect of frequency and loop convergence

Figure S2 shows the hysteresis loops at different frequencies for the case where the anisotropy axis and the direction of the external field form an angle of $\phi = 0^\circ$. The plot evidence that the loops narrow when the frequency is decreased until the convergence frequency is found and the resulting loop coincides with the Stoner Wohlfarth prediction (green curve).

C. Increased simulation time

As it is argued in the main text, a simulation time of $t_{sim} = 90$ ps for each field value is sufficient to produce reliable hysteresis loops. Figure S3 shows that increasing this time by a factor of approximately 2 has no appreciable effect in the shape of the resulting curves.

D. Size effects

Figure S4 compares two individual hysteresis loops obtained for a bcc lattice in a cubic box of volume $V = (10a_0)^3$ (the system under study) and a larger system with $V = (15a_0)^3$. The simulations correspond to the case of $\phi = 0^\circ$ and frequency $f_0/4$.

Figure S5 compares the fluctuations of $M_z$, the component of the magnetization along the field direction, during several field steps for three different system volumes. Both figures show that simulating a system with $V = (10a_0)^3$ is sufficient to avoid large size-effects.
E. Anisotropy and magnetization fluctuations

As it was argued in the Methods section of the main text, we have used mainly uniaxial anisotropy with an anisotropy constant equal to $K^*_1 = 35\ \mu\text{eV/atom}$, ten times larger than the Fe bulk value. The main reason for this is that, using a large anisotropy in the simulations helps to quickly stabilize the magnetization of the system for the $\phi = 0^\circ$ case, and therefore, less simulation time for each field value is required. Figure S6 shows this effect by comparing the time-evolution of the components of the magnetization for two different anisotropy intensities.
Figure S6. Comparison of the fluctuations of the components of the magnetization for the simulations with $\phi = 0^\circ$, frequency $f_0/4$ and different anisotropy constants, during the same number of applied field steps. The Gilbert damping in these simulations is $\lambda_g = 0.5$. 