Dynamic approach for micromagnetics close to the Curie temperature

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An increasing amount of research is focusing on the dynamic behavior of ferromagnetic materials at elevated temperatures. The motivations for this are manifold. A major imperative is the understanding of pulsed laser experiments on thin film samples, for example the all optical FMR experiments of Van Kampen et al. [1], and the higher laser power experiments of Beaurepaire et al. [2] who demonstrated complete demagnetization on a timescale of picoseconds. One of the main issues of the high-temperature magnetization dynamics is the rate of the magnetization relaxation due to different processes involving magnon, phonon and electron interactions that contribute to thermal spin disordering.

In conventional micromagnetism magnetic domain configurations are calculated based on a continuum theory for the magnetization which is assumed to be of constant length in time and space. Dynamics is usually described with the Landau-Lifshitz-Gilbert (LLG) equation the stochastic variant of which includes finite temperatures. Using simulation techniques with atomistic resolution we show that this conventional micromagnetic approach fails for higher temperatures since we find two effects which cannot be described in terms of the LLG equation: i) an enhanced damping when approaching the Curie temperature and, ii) a magnetization magnitude that is not constant in time. We show, however, that both of these effects are naturally described by the Landau-Lifshitz-Bloch equation which links the LLG equation with the theory of critical phenomena and turns out to be a more realistic equation for magnetization dynamics at elevated temperatures.

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Dynamics is usually described with the Landau-Lifshitz-Gilbert (LLG) equation which cannot correctly describe the transfer of high energy spin-waves from atomistic into the micromagnetic region. An alternative approach is the coarse graining model of Dobrovitški et. al. [7], which has the advantage of being able to link the length-scales but has been developed for simple systems only.

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The basis of most of theoretical investigations of thermal magnetization dynamics is a micromagnetic approach which considers the magnetization of a small particle or a discrete magnetic nanoelement as a vector of a fixed length (referred to here as a macro-spin) with the phenomenological Landau-Lifshitz-Gilbert (LLG) equation of motion augmented by a noise term $\xi$. However, contrary to atomic spins, there is no reason to assume a fixed magnetization length for nanoelements at non-zero temperature. For instance, the latter can decrease in time upon heating by a laser pulse. Hence, from the point of view of modeling of magnetization dynamics, there is a general need for further development of the micromagnetic theory in terms of its ability to deal with elevated temperatures.

Within this context we note the failure of micromagnetics in general to deal with the high frequency spin-waves which give rise to the variation of magnetization with temperature. It has been suggested to treat this problem using scaling approaches $\xi$. A similar problem arises in multi-scale modeling (with atomistic and micromagnetic discretizations to treat, for example, interfaces $\xi$) which can not correctly describe the transfer of high energy spin-waves from atomistic into the micromagnetic region. An alternative approach is the coarse graining model of Dobrovitški et. al. [7], which has the advantage of being able to link the length-scales but has been developed for simple systems only.

Some understanding of the pulsed laser experiments could indeed be obtained in terms of a micromagnetic approach taking into account, in an empirical way, the temperature variation of the intrinsic parameters, particularly the saturation magnetization $M_s$ and the anisotropy energy density $K$. Lyberatos and Guslienko have used this macro-spin model to investigate the response of nanoparticles during the Heat Assisted Magnetic Recording (HAMR) process. The validity of the macro-spin approach including the thermal variation of model parameters has further been investigated in Ref. [3] using an atomistic approach. This work demonstrates that, although the macro-spin model works well for temperatures far below the Curie temperature $T_c$, longitudinal fluctuations of the magnetization become important at elevated temperatures, which cannot be treated within the macro-spin model of the corresponding LLG equation of motion. The use of a macro-spin of fixed length places the same physical constraint on micromagnetics at temperatures close to $T_c$. Clearly, some approach to macro-spin dynamics beyond the LLG equation is needed.

A semi-phenomenological equation of motion for macro-spins allowing for longitudinal relaxation has been derived in Ref. [10] within the mean-field approximation (MFA) from the classical Fokker-Plank equation for individual spins interacting with the environment. This “Landau-Lifshitz-Bloch (LLB) equation” has been shown to be able to describe linear domain walls, a domain wall type with non-constant magnetization length. The validity of these results has been confirmed by measurements of the domain wall mobility in YIG crystals close to $T_c$. 

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In this letter we explore high-temperature dynamic properties using atomistic modeling. These simulations are still based on the LLG equation on the atomic level and, hence, do still not provide a microscopic description of the damping itself. Nevertheless they do include thermal degrees of freedom microscopically and encapsulate important phenomena associated with relaxation, including the thermodynamics of the phase transition and both, longitudinal and transverse macroscopic relaxation. We find an enhanced transverse relaxation when approaching the Curie temperature from below and a magnetization magnitude which is not constant in time. Both of these phenomena cannot be understood in terms of conventional micromagnetism but, comparing these predictions with a macro-spin model based on the LLB equation, we conclude that here these phenomena are indeed well described by the LLB equation.

For our atomistic simulations we use a model in which the dynamic behavior of classical spins $|s_i| = 1$ on lattice sites $i$ with magnetic moment $\mu_0$ is treated at the atomic level with the Langevin form of the LLG equation

$$\dot{s}_i = -\gamma [s_i \times H_i] - \gamma \alpha [s_i \times [s_i \times H_i]]$$

(1)

where $\gamma$ is the gyromagnetic ratio, and $\alpha$ is the damping parameter, $\alpha = 0.1$ in our simulations. The total field $H_i$ contains nearest-neighbor Heisenberg exchange (exchange constant $J$) and Zeeman contributions and it is augmented by a white-noise field $\zeta_i(t)$ with the correlator $\langle \zeta_\mu(t) \zeta_\nu(t') \rangle = \frac{2\alpha_0^2 J}{\mu_0^2} \delta_{ij} \delta_{\mu\nu} \delta(t-t')$, where $\mu, \nu = x, y, z$. For simplicity, the dipolar interaction is neglected as well as any crystalline anisotropy. A cubic lattice with periodic boundary conditions and system sizes of $48^3$ has been considered. In the calculations we first establish thermal equilibrium for a given temperature starting with all magnetic moments parallel to the $z$ axis and applying a field $H_z = 0.05J/\mu_0$. Then, to evaluate the transverse relaxation, all spins were simultaneously rotated by an angle of $30^\circ$. We have calculated the average spin polarization $m = (1/N) \sum_i (s_i)$ per lattice site which is proportional to the experimentally observed magnetization $M$.

Fig. 1a shows one transverse magnetization component as a function of time for different temperatures. The magnetization is normalized to its initial value and the magnetization magnitude shows a dip during the relaxation process which is well below its equilibrium value. A dynamic response of this type cannot be described in terms of the macro-spin LLG equation which conserves the absolute value of the magnetization, but is consistent with the LLB equation as will be discussed below.

It is interesting to note that our atomistic model with a constant microscopic damping parameter exhibits an increase in the effective macroscopic damping as observed experimentally [12]. We believe that this is due to magnon-magnon scattering processes which give rise to the initial decrease of the magnetization as the energy is transferred from the $k = 0$ (precessional) mode to higher order modes. This results in the enhanced transverse damping and in the dip of the magnetization, followed by the recovery to its equilibrium value as the spin waves decay.

Furthermore, we investigate the longitudinal relaxation time $\tau_\parallel$ from the initial relaxation of the fully ordered system to thermal equilibrium. The relaxation of the magnetization to equilibrium is found to be approximately exponential on longer time scales which defines the characteristic time $\tau_\parallel$. Fig. 1b shows the variation of the longitudinal relaxation time with temperature. The rapid increase close to $T_c$ is known as critical slowing down [13], a general effect characterizing second order phase transitions. Also shown in Fig. 1b is the perpendicular relaxation time $\tau_\perp$ determined as described above.
are dimensionless longitudinal and transverse damping
relaxation times from the atomistic modeling and the
Landau-Lifshitz-Bloch equation of motion \[10\]. This pro-
vides not only a deeper understanding of the phenomena
by macro-spin magnetization dynamics in terms of the
\[\dot{\mathbf{m}} = -\gamma (\mathbf{m} \times \mathbf{H}_{\text{eff}}) + \gamma \alpha_\parallel \frac{(\mathbf{m} \cdot \mathbf{H}_{\text{eff}}) \mathbf{m}}{m^2} - \gamma \alpha_\perp \frac{[\mathbf{m} \times [\mathbf{m} \times \mathbf{H}_{\text{eff}}]]}{m^2}, \]
(2)
where \(\mathbf{m} = \langle \mathbf{s} \rangle\) is the spin polarization and \(\alpha_\parallel\) and \(\alpha_\perp\) are dimensionless longitudinal and transverse damping parameters given by
\[\alpha_\parallel = \frac{2T}{T_{cMFA}}, \quad \alpha_\perp = \alpha \left[ 1 - \frac{T}{T_{cMFA}} \right] \]
(3)
for \(T < T_{cMFA}\) and the same with \(\alpha_\perp \Rightarrow \alpha_\parallel\) for \(T > T_{cMFA}\), where \(T_{cMFA}\) is the mean-field Curie temperature. Here, \(\alpha\) is the same damping parameter that enters Eq. \[10\]. The effective field \(\mathbf{H}_{\text{eff}}\) is assumed to be much weaker than the exchange interaction and it is given by
\[\mathbf{H}_{\text{eff}} = \mathbf{H} + \mathbf{H}_A + \begin{cases} \frac{1}{\chi_\parallel} \left( 1 - \frac{m^2}{m_0^2} \right) \mathbf{m}, & T \lesssim T_{cMFA} \\ \frac{m_0}{\mu_0} \frac{1}{\epsilon - 3m^2} \mathbf{m}, & T \gtrsim T_{cMFA} \end{cases} \]
(4)
Here \(\mathbf{H}\) and \(\mathbf{H}_A\) are applied and anisotropy fields and \(m_c\) is the zero-field equilibrium spin polarization in the MFA that satisfies the Curie-Weiss equation
\[m = B \left[ \beta (m_0 + \mu_0 H) \right] \]
(5)
with \(H = 0\) and \(\beta = 1 - T/T_{cMFA}\). \(B\) is the Langevin function, \(\beta = 1/(k_B T)\), and \(m_0\) the zero Fourier component of the exchange interaction related to \(T_{cMFA}\) as \(k_B T_{cMFA} = J_0/3\). In Eq. \[10\] \(\chi_\parallel \equiv \partial m(H, T)/\partial H\) is the longitudinal susceptibility at zero field that can be obtained from Eq. \[10\]. The anisotropy field \(\mathbf{H}_A\) due to the uniaxial anisotropy is related to the zero-field transverse susceptibility \(\chi_\perp\) as \(\mathbf{H}_A = (m_x \mathbf{e}_x + m_y \mathbf{e}_y)/\chi_\perp\)
\[10\]. The equilibrium solution of the LLB equation satisfies \(\mathbf{m} \times \mathbf{H}_{\text{eff}} = 0\) and \(\mathbf{m} \cdot \mathbf{H}_{\text{eff}} = 0\). For \(T \ll T_{cMFA}\) the longitudinal susceptibility \(\chi_\parallel\) becomes very small in which case it can be shown that \(m \approx m_c\). This means that the longitudinal relaxation vanishes and Eq. \[2\] reduces to the standard LLG equation with \(\alpha_\perp = \alpha\).

In the damping parameters \(\alpha_\parallel\) and \(\alpha_\perp\) of Eq. \[10\] \(\alpha\) is non-critical at \(T_{cMFA}\). Its temperature dependence cannot be established within our semi-phenomenological approach, so we assume it to be a constant, for the sake of comparison with the results of our atomistic simulations. The LLB equation also can be written in terms of the vector \(\mathbf{n} = \mathbf{m}/m_c\)
\[13\]. This form provides a link to the micromagnetic anisotropy constants but becomes inconvenient above \(T_c\) where \(m_c\) disappears.

In order to effect a comparison we analyse the relaxation rates derived from the LLB equation. Firstly we note from Eq. \[3\] a linear increase of \(\alpha_\parallel\) with \(T\), whereas the behavior of \(\alpha_\perp\) is non-monotonic, changing from a linear decrease below \(T_{cMFA}\) to a linear increase above \(T_{cMFA}\). However, it is important to note that \(\alpha_\parallel\) and \(\alpha_\perp\) are non-critical for all finite temperatures, and that the variation of \(\alpha_\perp\) is weak. With this background, we now consider the relaxation rates from the linearized LLB equation which have the form
\[\Gamma_\parallel = \frac{\gamma \alpha_\parallel}{\chi_\parallel(H, T)}, \quad \Gamma_\perp = \frac{\gamma \alpha_\perp}{\chi_\perp(H, T)}, \]
(6)
where \(\chi_\parallel(H, T)\) is the longitudinal susceptibility at nonzero field that follows from Eq. \[10\] or simply from \(\mathbf{m} \cdot \mathbf{H}_{\text{eff}} = 0\), in our approximation.

The longitudinal relaxation rate is, in general, very fast as \(\Gamma_\parallel \sim J_0\). Since \(\chi_\parallel(H, T)\) is large near \(T_{cMFA}\), \(\Gamma_\parallel\) shows critical slowing down which is a result of the critical behavior of \(\chi_\parallel(H, T)\) rather than the variation of \(\alpha_\parallel\). The transverse susceptibility for the isotropic model is simply given by \(\chi_\perp(H, T) = m(H, T)/H\) so that \(\Gamma_\perp \sim H\) is much smaller than \(\Gamma_\parallel\) below \(T_{cMFA}\). However, it increases with temperature, as was observed in the atomistic modeling presented above and its critically behavior close to \(T_c\) is \(\Gamma_\perp \sim 1/m(H, T)\). For temperatures below \(T_c\) a corresponding behavior was found for the line widths of FMR experiments
\[13\].
If one evaluates the macro-spin parameters directly from a region. However, this comparison could still be improved. The results are presented in Fig. 3. Comparison with Fig. 1 shows that the LLB equation reproduces essential physical processes which govern the magnetization dynamics at elevated temperatures and thus it can be used as an alternative to micromagnetics in this region. However, this comparison could still be improved if one evaluates the macro-spin parameters directly from an atomistic simulation. Furthermore, if the LLB equation is to be used as an alternative to micromagnetics, the corresponding parameters could as well be extracted from experiment.

In conclusion, performing atomistic simulations of thermal magnetization dynamics we observe an increase of the macroscopic transverse damping approaching the Curie temperature. This increase is determined by the thermal dispersion of magnetization and would exist independently from any other possible thermal dependence of internal damping mechanisms such as phonon-magnon coupling. This effect explains the broadening of the resonance line width in classical FMR experiments. Furthermore, the magnetization vector turns out not to be constant in length. Instead during relaxation one can observe a dip of the magnetization which is more pronounced when approaching the Curie temperature. Importantly, the observed dynamics is in agreement with the dynamics of a macro-spin described by the Landau-Lifshitz-Bloch equation which contains both longitudinal and transverse relaxation. This equation could serve in future as a basis for an improved micromagnetics at elevated temperature.

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At \( T = T_c^{\text{MFA}} \) the rates are given by

\[
\Gamma_\parallel \approx \frac{6}{5} \frac{\gamma \alpha J_0}{\mu_0} m_H^2, \quad \Gamma_\perp \approx \frac{2}{5} \frac{\gamma \alpha J_0}{\mu_0} m_H^2
\]

(7)

where \( m_H = [(5/3) (\mu_0 H/J_0)]^{1/3} \) is the induced magnetization at \( T_c^{\text{MFA}} \). Above \( T_c^{\text{MFA}} \) both rates merge:

\[
\Gamma_\parallel \approx \Gamma_\perp \approx \frac{2}{3} \frac{\gamma \alpha J_0}{\mu_0} \frac{T}{T_c^{\text{MFA}}} \left( \frac{T}{T_c^{\text{MFA}}} - 1 \right).
\]

(8)

Finally, in the presence of uniaxial anisotropy \( \Gamma_\perp \) is given by Eq. (8) with \( 1/\tilde{\chi}_\perp \left( H, T \right) = H/m \left( H, T \right) + 1/\tilde{\chi}_\perp \), where \( \tilde{\chi}_\perp \) is only weakly temperature dependent within mean-field theory below \( T_c^{\text{MFA}} \).

To compare the LLB results with the predictions of the atomistic model, Fig. 2 includes the inverse relaxation rates calculated using Eq. (6) with rescaled temperature to fit the exact value \( k_B T_c = 1.44 J \) for a simple cubic lattice. The agreement between Eq. (6) and the numerical results is remarkable given the MFA used in the derivation of Eq. (6).

Also, we have integrated numerically Eq. (2) for a macro-spin to give the time evolution of the magnetisation components for comparison with the numerical results of Fig. 1. The results are presented in Fig. 3. Comparison with Fig. 1 shows that the LLB equation reproduces essential physical processes which govern the magnetization dynamics at elevated temperatures and thus it can be used as an alternative to micromagnetics in this region. However, this comparison could still be improved if one evaluates the macro-spin parameters directly from an atomistic simulation.
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