A thermodynamically self-consistent non-stochastic micromagnetic model for the ferromagnetic state

Mykola Dvornik, Arne Vansteenkiste, and Bartel Van Waeyenberge
DyNaMat Lab, Ghent University, Krijgslaan 281/S1, 9000 Ghent, Belgium

In this work, a self-consistent thermodynamic approach to micromagnetism is presented. The magnetic degrees of freedom are modeled using the Landau-Lifshitz-Bloch (LLB) theory, which unifies and extends the previously developed approaches within a single framework. Although the model is phenomenological, it allows for a consistent physical interpretation of the spin relaxation terms and their coupling to electron and phonon systems.

Micromagnetism typically relies on the phenomenological theory developed by Landau and Lifshitz[1] that is used to fit experimental data on a wide range of magnetization dynamics problems. However, this theory cannot be applied to ultrafast magnetization phenomena and spintronics, since it cannot account for changes of the magnetization vector length (so-called longitudinal magnetization dynamics)[2–5] and non-local exchange damping[6–9]. The Landau-Lifshitz-Bloch (LLB) equation [13, 14] to describe magnetic response at elevated temperatures.

We rely on Baryakhtar’s theory [17, 19] that derives the following equation for the magnetization dynamics and relaxation:

\[ \frac{\partial \mathbf{M}}{\partial t} = -\gamma_{LL} \mathbf{M} \times \mathbf{H} + \hat{\lambda}(\mathbf{M})\mathbf{H} - \frac{\partial \mathbf{H}}{\partial x_p} \partial^2 \mathbf{H} \frac{\partial \mathbf{H}}{\partial x_q} \]  

(1)

where \( \mathbf{M} \), \( \gamma \), \( \mathbf{H} \) are the magnetization vector, the positively defined gyromagnetic ratio and the internal effective field, respectively. The terms in the right-hand side of the equation describe the magnetic torque, the local relativistic and the non-local exchange relaxations, respectively. The LLB equation is the special case of eq. (1).

The internal field is given by \( \mathbf{H} = -\frac{\partial G}{\partial \mathbf{M}} = \mathbf{H}^{MM} + \mathbf{H}^{M} \), where \( w \) is the Gibbs free energy density of the magnetic medium, \( \mathbf{H}^{MM} \) and \( \mathbf{H}^{M} \) denote micromagnetic and longitudinal effective fields. The longitudinal field (historically referred to as “molecular” field) is the micromagnetic representation of the microscopic exchange field that arises from interaction of the spins with other quasi-particles, namely electrons and phonons. It drives the spin system to thermodynamic equilibrium via angular momentum transfer. This field consists of two parts \( \mathbf{H}^{M} = \mathbf{H}_{s-e}^{M} + \mathbf{H}_{s-p}^{M} \), i.e. a spin-electron \( (s-e) \) and a spin-phonon \( (s-p) \) contribution [20]. The field implicitly accounts for the spin fluctuations and optical magnons that cannot be resolved on the micromagnetic scale. It
has the following phenomenological form\[13\]
\[
\mathbf{H}_i = \frac{n k_B T_i}{\mu_0 M_s} B_f(m \frac{\partial}{\partial t} - m) - m \frac{m}{B_f(m \frac{\partial}{\partial t})} m
\]  
(2)
where \(T_i\) is the temperature of the corresponding quasiparticles \((i = s - e, s - p)\). \(n\), \(M_s\) and \(J\) are the number density of atoms, zero-temperature saturation magnetization and atomic moment, respectively and \(m = M/M_s\). \(\theta = \frac{3}{4} k_B T_c\), where \(T_c\) is the Curie temperature of the material. \(B_f\) denotes the Brillouin function for the given atomic moment. Eq. (2) accounts only for competition between the microscopic exchange and thermal fluctuations energies. The contribution of the Zeeman interaction is explicitly included into the model.

Although eq. (1) and eq. (2) successfully describe the ferromagnetic-to-paramagnetic phase transition, the opposite transition cannot be modeled without explicitly considering short-order spin fluctuations\[21\], e.g. by introducing a stochastic form of the model. This regime lies beyond the scope of the present study.

The relativistic relaxation tensors \(\lambda\) and \(\lambda_{pq}(\mu)\) describe the strength of the relativistic and exchange dissipations, respectively. In contrast to the Landau-Lifshitz and Landau-Lifshitz-Bloch models, the relativistic relaxation tensor obeys the crystallographic and magnetic symmetries of the system. So let us expand it into powers of \(\mathbf{M}\) around the highest symmetry magnetic state \(M = 0\)[19], i.e.

\[
\tilde{\lambda}(\mathbf{M}) = \tilde{\lambda}_{\parallel} + \tilde{\mu}_{pq} M_p M_q + O(M^4)
\]  
(3)
The relativistic relaxation tensors \(\tilde{\lambda}_{\parallel}\) and \(\tilde{\mu}_{pq}\) mimic the crystallographic symmetry of the system over the corresponding spatial indices. Substituting of eq. (3) into eq. (1) and assuming at least a uniaxial symmetry of the media leads to

\[
\frac{\partial \mathbf{m}}{\partial t} = -\gamma_{LL} \mathbf{M} \times \mathbf{H} + \\
+ \tilde{\lambda}_{\parallel} \mathbf{H} - \tilde{\mu}_{pq} \mathbf{M}_p \mathbf{M}_q - \tilde{\lambda}_{\parallel} (\mathbf{M} \cdot \mathbf{H}) \mathbf{M} - \tilde{\mu}_{\perp} \mathbf{M} \times \tilde{\mu}_{\perp} \mathbf{M} \times \mathbf{H}
\]  
(4)
where \(\tilde{\mu}_{\perp}\) and \(\tilde{\mu}_{\parallel}\) are decomposed from \(\tilde{\mu}_{pq}\) to separate conservative and non-conservative second-order relaxations, respectively. Let us rewrite all the relaxation tensors via dimensionless coupling constants

\[
\tilde{\lambda}_{\parallel} = -\gamma_{LL} M_s \alpha_{\parallel} \hat{\nu}
\]
\[
\tilde{\lambda}_{\parallel} = -\gamma_{LL} M_s \alpha_{\parallel} \hat{\nu}
\]
\[
\tilde{\mu}_{\perp} = -\frac{\gamma_{LL} M_s}{\alpha_{\parallel}} \hat{\nu}
\]
\[
\tilde{\mu}_{\parallel} = -\frac{\gamma_{LL} M_s}{\alpha_{\parallel}} \hat{\nu}
\]
(5)
where \(a\) is the lattice constant, \(\hat{\nu}\) describes crystallographic symmetry of the media, while \(\alpha_{\parallel}\), \(\alpha_{\parallel}\), \(\mu_{\perp}\) and \(\mu_{\parallel}\) define strength of the corresponding relaxations. Substitution of (5) into (1) we finally arrive at

\[
\frac{1}{\gamma_{LL}} \frac{\partial \mathbf{m}}{\partial t} = -\mathbf{m} \times \mathbf{H} + R_{s-e} + R_{s-p}
\]

\[
R_{s-e} = \alpha_{\parallel} \hat{\nu} \mathbf{H} - \alpha_{\parallel} \hat{\nu} \mathbf{A} \mathbf{H}
\]
\[
R_{s-p} = \mu_{\perp} \hat{\nu} (\mathbf{m} \cdot \mathbf{H}) \mathbf{m} - \mu_{\perp} \mathbf{m} \times \hat{\nu} (\mathbf{m} \times \mathbf{H})
\]  
(6)
The important property of the expansion (3) is that it separates the zeroth-order paramagnetic relaxation \(R_{s-e}\) (independent of \(\mathbf{m}\)) from the higher-order magnonic relaxation \(R_{s-p}\) (that depends on \(\mathbf{m}\)). If we neglect the non-local relaxation and assume that the length of the magnetization vector is conserved, then eq. (6) reduces to the Landau-Lifshitz equation with a damping constant that is the sum of Baryakhtar’s local relaxation constants \(\alpha_{LL} = \alpha_{\parallel} + \mu_{\perp}\).

Baryakhtar has pointed out that the expansion of the relaxation tensors (3) is analogous to the expansion of the Gibbs free energy of a magnetic medium. So the quadratic relaxation terms in eq. (6) should describe relaxation due to the spin-orbit and, thereby, spin-phonon coupling\[22\]. This conclusion is also consistent with the spin-electron-lattice model developed by Ma et al.\[15\] who showed the spin-phonon coupling is at least a four-spin correlation function.

At the same time the zeroth-order relativistic paramagnetic relaxation is independent of the magnetic configuration, and so cannot be attributed to spin-orbit coupling. According to Overhauser\[23\], the main relativistic contribution to the relaxation in paramagnets is the dipolar (spin-spin) spin-electron coupling. This interaction is independent on the direction of the magnetization and is isotropic for a cubic lattice, consistent with the symmetry properties of the zero-order relativistic relaxation term in eq. (3).

According to the review of Fähnle and Zhang\[8\], the non-local exchange damping in metals of a form similar to eq. (6) is due to spin-electron s-d exchange interaction\[26\]. This is also consistent with the conclusions of Overhauser\[25\] for paramagnets. So to summarize, the zeroth-order and higher-order relaxation terms in eq. (6) must describe spin-electron and spin-lattice couplings, respectively. This classification is the first important feature of the proposed micromagnetic model.

The second problem addressed by this model is the compliance with the energy conservation law. To couple the LLBar model to the corresponding heat transfer equations, we calculate the rate of the energy density change due to the different dissipations. We emphasize that the Gibbs free energy depends on the entropy of the system, i.e. \(w = w(s)\), where \(s\) is the entropy density.
So the rate of the energy density change at the constant entropy density is given by the following expression

\[
\frac{d\varphi}{dt} = \frac{\partial \varphi}{\partial t} + \frac{\partial w_s}{\partial s} \frac{\partial s}{\partial t} - 2 \frac{\partial q}{\partial t} \tag{7}
\]

where \(\frac{\partial \varphi}{\partial t} = -\gamma_{LL} n k_B \theta m \mathbf{R}_i\) describes change of the internal energy density due to the the molecular field \([27]\) and \(\frac{\partial q}{\partial t} = \frac{1}{2} \frac{\partial w_s}{\partial s} \frac{\partial s}{\partial t} = \frac{1}{2} \gamma_0 M_i \mathbf{HR}_i\) is the Baryakhtar’s dissipative function. The indices denote the type of the relaxation channel. Finally, we assume that the thermodynamics of the electrons and phonons is governed by a 2T model

\[
C_e(T_e) \frac{\partial T_e}{\partial t} = k_i \Delta T_e + G_{e-p}(T_p - T_e) - \frac{d\varphi_{e-p}}{dt}
\]

\[
C_p(T_p) \frac{\partial T_p}{\partial t} = k_p \Delta T_p + G_{e-p}(T_e - T_p) - \frac{d\varphi_{e-p}}{dt} \tag{8}
\]

where \(T_i, C_i, k_i\) and \(G_{e-p}\) are the temperature, volume-specific heat capacity, heat conductance and macroscopic electron-phonon coupling constant, respectively. The volume-specific heat capacity of the phonons is estimated from the Debye distribution and DOS of electrons, but this approach lies beyond the scope of the present study. Finally, the system of coupled equations \([43, 45]\) forms a thermodynamically self-consistent non-stochastic micromagnetic model. It is worth noting, that our model correctly reproduces the specific heat of spins given by the mean-field approximation in the temperature range of \([0, T_c]\).

We would like to emphasize that neither LLB nor M3T nor the proposed model account for the angular momentum conservation law and thereby cannot be strictly used to identify the microscopic scattering mechanisms responsible for the ultrafast heat-induced demagnetization. This can only be achieved using full spin-electron-phonon model in the spirits of the one used by Ma et al. \([15]\).

Now we apply our model to the problem of the ultrafast laser heating of Nickel that was systematically investigated by Roth et al. \([28]\). Hereafter we refer to the data acquired at fixed ambient temperature (of \(T_{amb} = 300\) K) using varying laser fluence as fluence data, while the data acquired at fixed laser fluence (of \(F_0 = 35\) J/m\(^2\)) using varying ambient temperature as the temperature data. In the simulations we neglect the direct spin-phonon relaxation mechanism, i.e. \(\mu_\perp = \mu_\parallel \equiv 0\), since the spin-orbit interaction is vanishing in Nickel. We also neglect any heat-transport, i.e. \(k_e = k_p \equiv 0\), since (a) sample thickness is assumed to be comparable to the skin-depth, (b) the ratio between the diameters of the pump and probe spots used in the experiments is around 250 : 1 and (c) the heat transfer is much slower than the local longitudinal magnetization dynamics. The values of the Debye temperature \(\theta_D = 390\) K, \(\gamma = 4.51 \cdot 10^{-3}\) J/K\(^2\)mol\(^{-1}\) and \(T_C = 633\) K are all extracted from Ref. \([29]\). The atomic moment of Nickel is \(J = \frac{1}{2}\). The laser pulse is assumed to be Gaussian with FWHM 50 fs. The light absorption \(A\), \(G_{e-p}\) and \(\alpha_p\) were all estimated using the Levenberg-Marquardt least-squares fitting algorithm from the SciPy set of libraries \([30]\). The solutions to the system of equations \([43, 45]\) are calculated using the in-house developed open-source hotspin micromagnetic solver \([31]\).

Our analysis shows that for the fluence data, the estimated values of the aforementioned parameters were significantly different from those estimated from the temperature data. In a private communication, the authors of the experimental study confirmed that (a) the temperature and fluence data were acquired from different samples and (b) the fluence data acquisition was performed without any means of the temperature control leading to the accumulation of the residual heat. We account for this effect by introducing the ad-hoc linear dependence of the ambient temperature on the laser fluence, i.e. \(T_{amb} = (8.89 K J^{-1} m^2) F_0\).

The best fits of our model to the experimental data are shown in Fig. 1 and Fig. 2 for the temperature and fluence data, respectively. The correlation between

![Figure 1: Temperature dependence of the longitudinal magnetization dynamics. The solid lines represent the best fits of the proposed micromagnetic model to the experimental data from Ref. 28 (shown as symbols).](image-url)
the zero-order relativistic relaxation and electron-phonon coupling constants is shown in Fig. 3. The fit shows almost no correlation between these two quantities suggesting that the ultra-fast angular momentum transfer happens only within the spin-electron subsystem. This is in contrast to M3T model that assumes that the angular momentum is transferred to the lattice and predicts a linear dependence between (longitudinal) damping and electron-phonon coupling constants. So our findings question the widely accepted hypothesis of the Elliot-Yafet scattering dominants in the ultrafast magnetization dynamics, consistent with ab-initio calculations performed by Ilg et al. 32. However, the importance of the electron-phonon coupling in the ultrafast magnetization dynamics should not be underestimated, since it provides the ultrafast energy dissipation channel (possibly via the inelastic electron-phonon scattering) and, thereby, maintains the magnetization recovery.

For the temperature data, a gradual decrease of the electron-phonon coupling constant is observed, consistent with the electron DOS calculations from Ref. 33. In contrast, for the fluence data the opposite effect is observed. We believe this is artificial, since the exact experimental conditions are unknown in this case. This observation urges higher quality measurements with finer steps in both laser fluence and ambient temperature.

The fit gives the following bounds for the relevant parameters: $\alpha_{\parallel} = (1.34 \pm 0.24) \cdot 10^{-2}$ consistent with $\alpha_{LLB} = 1.30 \cdot 10^{-2}$ estimated from Q-band FMR measurements 32. $A = 0.19 \pm 0.05$ consistent with $A = 0.21$ measured at $T_{amb} = 4.2 K$ using continuous excitation of 1 $\mu m$ wavelength 32. Unfortunately, there is no consistent data on the value of the macroscopic electron-phonon coupling constant, since it is typically estimated indirectly, e.g. using the three-temperature model where thermodynamic parameters are assumed to be constant 3. Nevertheless, we estimated that $G_{e\rightarrow p}(T_e) \in [0.91, 1.97] \cdot 10^{13} W K^{-1} mol^{-1}$ that is below the upper bound of $G_{e\rightarrow p}(0) = 2.13 \cdot 10^{13} W K^{-1} mol^{-1}$ provided by the electron DOS calculations 33.

In contrast to the LLB and M3T models, we assume that the relaxation constants are independent of the electron and (or) phonon temperatures. In fact, the fitting shows a weak temperature dependence of the zero-order relativistic damping constant for $T_{amb} \leq 400 K$, with prominent enhancement for $T_{amb} = 480 K$. This might suggest an activation (or significant enhancement) of an alternative relaxation mechanism, e.g. Elliot-Yafet scattering or non-local damping. The fact that the value of the electron-phonon coupling constant is also enhanced (contradictory to the electron DOS calculations) might indeed be a sign of the Elliot-Yafet relaxation mechanism that pops-up only when spins acquire temperature close to the Curie point. The influence of the non-local exchange damping on the observed enhancement can only be estimated using atomistic simulations.

In conclusion, we propose a model that solves two major problems of finite-temperature micromagnetism: (i) it provides physical interpretation of the relaxation terms and (ii) it fully complies with the energy conservation law. The model could be readily used to (a) quantitatively estimate the LLBar-specific relaxation constants from the experimental data and (b) to explore applications of spin caloritronics and heat-assisted magnetic
Authors would like to acknowledge Dr. Mirko Cinchetti for sharing the experimental data and the details on its acquisition routines.

*Electronic address: Mykola.Dvornik@ugent.be; URL: http://dynamat.ugent.be

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