Spin-Lattice Dynamics Simulation of Magnon-Phonon-Electron Heat Transfer on the Million Atom Scale

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We develop an atomistic spin-lattice-dynamics model for simulating energy relaxation in magnetic materials. The model explicitly solves equations of motion for the atoms and spins, and includes interaction with electron excitations. We apply the model to simulate the dynamics of propagation of a compressive elastic wave in iron. We find that coupling between the lattice, spin, and electron degrees of freedom does not have an appreciable effect on the velocity of the wave. At the same time, dissipative lattice-electron-spin interactions entirely dominate the dynamics of attenuation of the wave in the material.

Modelling interactions between magnetic and atomic degrees of freedom is necessary for understanding the dynamics of energy relaxation in magnetic materials, which play a significant part in applications ranging from micro-magnetic devices to radiation damage phenomena in steels. Spin dynamics (SD) and molecular dynamics (MD) have been widely applied to explore the evolution of magnetic and atomic degrees of freedom. The problem of treating SD and MD within a unified framework, involving also electron excitations, remains outstanding. For example, Radu et al.[1] described the dynamics of energy relaxation in a Fe-Gd alloy using SD, modelling electron and lattice subsystems by the heat transfer equations and including spin-electron interactions via a fluctuating term. The model does not make it possible to treat mechanical response of the lattice to magnetic excitations.

Spin-lattice dynamics (SLD) simulations[2], on the other hand, follow the evolution of magnetic and atomic degrees of freedom. This makes it possible to explore the dynamics of energy transfer between fluctuating magnetic moments and atomic vibrations at elevated temperatures. By introducing fluctuation and dissipation terms into SLD equations of motion, and by mapping them onto the three-temperature model (3TM)[3], we are able to develop a self-consistent treatment of interactions and energy transfer between spin, lattice and electron subsystems. In this paper, we outline the model and illustrate its range of applications by simulating a compressive wave propagating in ferromagnetic iron.

Assuming that magnetic properties of a material are described by the Heisenberg model, a Hamiltonian, including lattice, spin and electron subsystems, can be written as[2]

$$\mathcal{H} = \mathcal{H}_l + \mathcal{H}_s + \mathcal{H}_e, \tag{1}$$

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where

$$\mathcal{H}_l = \sum_i \frac{\mathbf{p}_i^2}{2m} + U(\mathbf{R}), \tag{2}$$

$$\mathcal{H}_s = -\frac{1}{2} \sum_{i,j} J_{ij}(\mathbf{R}) \, \mathbf{S}_i \cdot \mathbf{S}_j, \tag{3}$$

and $\mathcal{H}_e$ is the Hamiltonian of conduction electrons. $J_{ij}(\mathbf{R})$ is a coordinate-dependent exchange coupling function, and $\mathbf{S}_i$ is an atomic spin vector. Equations of motion for atomic coordinates, momenta and the spin vectors have the form[2].

$$\frac{d\mathbf{R}_k}{dt} = \frac{\mathbf{p}_k}{m} \tag{4}$$

$$\frac{d\mathbf{p}_k}{dt} = -\frac{\partial U}{\partial \mathbf{R}_k} + \frac{1}{2} \sum_{i,j} \frac{\partial J_{ij}}{\partial \mathbf{R}_k} \mathbf{S}_i \cdot \mathbf{S}_j - \frac{\gamma_l}{m} \mathbf{p}_k + \mathbf{f}_k \tag{5}$$

$$\frac{d\mathbf{S}_k}{dt} = \frac{1}{\hbar} \left[\mathbf{S}_k \times (\mathbf{H}_k + \mathbf{h}_k) - \gamma_s \mathbf{S}_k \times (\mathbf{S}_k \times \mathbf{H}_k) \right]. \tag{6}$$

Here $\mathbf{H}_k = \sum_i J_{ik} \mathbf{S}_i$ is the effective exchange field acting on spin $\mathbf{S}_k$. γ_s and γ_l are the damping constants for the spin and lattice degrees of freedom, respectively. $\mathbf{h}_k$ and $\mathbf{f}_k$ are the delta-correlated fluctuating field and force, satisfying the conditions $\langle \mathbf{h}_k(t) \rangle = 0$, $\langle \mathbf{f}_k(t) \rangle = 0$; and $\langle h_{i\alpha}(t) h_{j\beta}(t') \rangle = \mu_s \delta_{ij} \delta_{\alpha\beta} \delta(t - t')$ and $\langle f_{i\alpha}(t) f_{j\beta}(t') \rangle = \mu_l \delta_{ij} \delta_{\alpha\beta} \delta(t-t')$. Subscripts α and β denote the Cartesian components of a vector.

We assume that fluctuation and dissipation terms in Eq. 5 and 6 describe interaction with the electron subsystem, similarly to how interaction between atoms and electrons was introduced in MD simulations in Refs. 4. Using the fluctuation-dissipation theorem[5], we write the fluctuation-dissipation relation for the lattice[5] and spin[2,6] subsystem as $\mu_l = 2\gamma_l k_B T_e$ and $\mu_s = 2\gamma_s \hbar k_B T_e$, respectively.

The local electron temperature T_e satisfies a diffusion equation, where the heat transfer terms couple it to the dynamically evolving lattice and spin subsystems.

$$C_e \frac{dT_e}{dt} = \nabla(\kappa_e \nabla T_e) - G_{el}(T_e - T_l) - G_{es}(T_e - T_s). \tag{7}$$

Here C_e is the electron specific heat and κ_e is thermal conductivity. Coefficients G_{el} and G_{es} in (7) can be found analytically by mapping the above equations onto the 3TM, and by evaluating the change of energy for each subsystem via Eqs. 1-6. After some algebra, we find that $G_{el} = 3k_B\gamma_l/m$ and $G_{es} = 2k_B\gamma_s\langle \mathbf{S}_k \cdot \mathbf{H}_k \rangle/\hbar$. The local lattice temperature T_l is calculated from the local kinetic energy of atoms, whereas the local spin temperature T_s is evaluated using the equation $2k_B T_s = \langle|\mathbf{S}_k \times \mathbf{H}_k|^2\rangle/\langle \mathbf{S}_k \cdot \mathbf{H}_k \rangle$ derived in previous work[7].

The spin-lattice-electron (SLE) model becomes fully self-consistent once the Langevin equations of motion for the spin and lattice degrees of freedom, and the diffusion equation for electrons are coupled to each other via the fluctuation, dissipation, and the energy transfer terms. If there is no external source of energy, the SLE model describes a closed system.

To illustrate applications of the model, we simulate a compressive elastic wave propagating in ferromagnetic iron. For comparison, we also perform an MD simulation describing the same wave, assuming that the atomic system evolves conservatively with no interaction with the spin and electron subsystems.

Simulations were performed using samples containing $30\times30\times550$ body-centred cubic (BCC) unit cells with the coordinate axes parallel to the [100], [010] and [001] crystallographic directions. Each sample contained in total 990000 atoms (and spins). Interatomic and exchange potentials for BCC ferromagnetic iron were used[11]. At the start of each simulation, all the samples were thermalized to 300K, with temperature homogeneously distributed through the sample. Simulations of electron temperature were performed using the intrinsic "finite difference" grid associated with linked cells defined within the MD and SLD integration algorithms. Spin, lattice and electron temperatures T_s, T_l and T_e were treated as parameters associated with each linked cell, where they were evaluated as averages over atoms belonging to a cell. By defining temperatures of each subsystems in this way, we are able to formulate a finite difference algorithm for solving Eq. 7.

Practical simulations showed that an efficient and accurate numerical integration algorithm was required to accurately compute the energy change associated with each time step. Applying an algorithm[8,9] based on the Suzuki-Trotter decomposition[10], the symplectic nature of which guarantees the accumulation of small numerical error[10], proves essential for the feasibility of simulations. All the simulations were performed using a GPU-enabled computer program run on Nvidia GTX480.

A compressive wave propagating in the [001] direction was initiated using a method proposed by Holian *et al.*[14]. For a system with periodic boundary condition imposed along all the axes, we introduced the initial symmetric impact by shrinking the simulation box uniaxially. This makes the boundaries on the left and right to move inwards with velocities $\pm u_p$, which is equivalent to applying the action of two pistons moving with velocities $\pm u_p$.
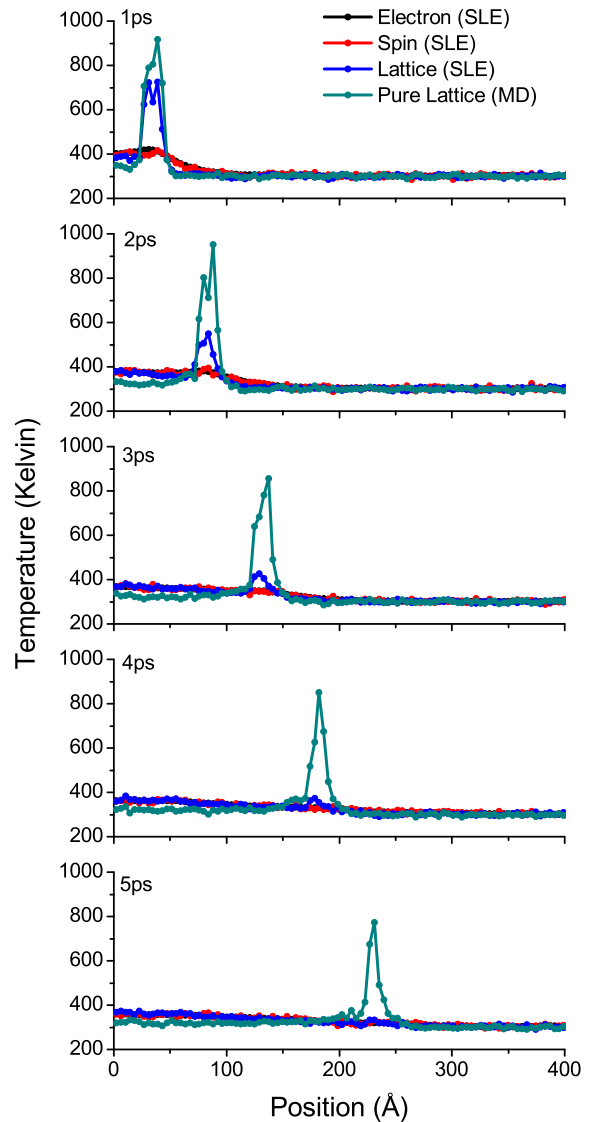

FIG. 1. Average T_s, T_e and T_l predicted by a SLE simulation for link cells at z at time $t = 1$ to 5 ps. The average T_l derived from a pure MD simulation is also shown for comparison. Samples are constructed with sizes of $30 \times 30 \times 550$ unit cells of body-centered cubic structure along [100], [010] and [001] directions. Prior to simulation the samples are thermally equilibrated to 300K. A compressive elastic wave moving in the [001] direction is created by shrinking the system size uniaxially, to make the left and the right boundaries of the system move inwards at speed $\pm u_p$. The simulation corresponds to $u_p = 500$m/s continuing for 0.5ps. The velocity of the compressive wave is $u_s \approx 4500$m/s. The wave attenuates much faster in a SLE simulation, due to the dissipation of energy into spin and electron excitations.

Instead of compressing the simulation sample continuously, we applied such artificial compression on the simulation box for only 0.5ps, with $u_p = 500$m/s. During the simulations, there were no further geometric transformation of the sample after 0.5ps. The formation of a transformation wave propagating through the sample

after an elastic precursor is often observed in shock wave simulations[15]. The small lateral size of the current sample prevents dislocation nucleation and the occurrence of plastic deformation.

Fig. 1 shows the evolution of T_s, T_l and T_e, treated as functions of time and coordinate in the direction of propagation of the wave, found in SLE simulations, and the evolution of lattice temperature T_l found in a pure MD simulation. Each point in the figure corresponds to the average temperature of atoms in a linked cell situated at a particular position in the direction of propagation of the wave. Since the magnitude of T_l is by definition related to the kinetic energy of atoms, the peak of T_l indicates the position of the compressive wave. In both SLE and MD simulations the compressive wave moves away from the edge of the cell inwards at a constant velocity $u_s \approx 4500\text{m/s}$, which is close to the speed of a longitudinal sound wave propagating in a single crystal of iron in the [001] direction[13] $c_{[001]} = \sqrt{C_{11}/\rho} \approx 5300\text{m/s}$.

The main difference between the SLE and MD simulations is the rate of attenuation of the wave, seen in Fig. 1 as the rate at which temperature at the wave front decreases as the wave propagates through the crystal. This attenuation occurs much faster in the SLE simulation in comparison with the MD case. The electron subsystem absorbs energy from the lattice via the dissipation term, and transfers energy into the spin subsystem via the fluctuation term. The effect of direct spin-lattice coupling is much weaker than the sequence of lattice-electron and electron-spin interactions. Fig. 1 shows that gradually all the energy initially stored in the compressive wave gets fully dissipated. The temperatures of spin, lattice, and electron subsystems rise behind the front of the wave. In the pure lattice system, modelled by MD, energy dissipates via phonon-phonon interactions, and the rate of dissipation is significantly slower than that found in an SLE simulation. While this conclusion is general, the actual rate of energy dissipation predicted by the SLE model depends on the choice of parameters γ_s and γ_l which should vary subjecting to different experimental conditions. In the current simulation, they were chosen by matching results of simulations to experiments[16] on ultra-fast pulsed laser heating of magnetic materials[3].

In conclusion, we have developed a spin-lattice-electron model for simulating energy relaxation involving lattice, spin and electron degrees of freedom in a magnetic material. The model explicitly solves equations of motion for the atoms and spins, and includes interaction with electron excitations, which are modeled by a diffusion equation. We apply the model to simulate the dynamics of propagation of a compressive elastic wave in iron. We find that interaction between the lattice, electron and magnetic degrees of freedom has a significant effect on the rate of attenuation of the wave propagating through the material. The new method offers a way of including the effect of magnetic and electronic excitations in simulations of shock-induced plastic deformation and defect production in magnetic materials, which so far remained inaccessible to direct atomistic modelling[15].

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