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Spin-lattice dynamics model for magnon-phonon-electron heat transfer on a 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 atoms and spins, and includes interaction with electron excitations. We apply the model to simulate the dynamics of propagation and attenuation of a compressive elastic wave in iron. We find that interaction 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 spin-lattice-electron interactions dominate the dynamics of attenuation of the wave in the material. [doi:10.1063/1.3673859]

Modeling interaction between magnetic and atomic degrees of freedom is necessary for describing the dynamics of energy relaxation in magnetic materials, which plays 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 applied to explore the evolution of magnetic and atomic degrees of freedom. The problem of treating SD and MD within a unified framework, also including the treatment of electron excitations, remains outstanding. For example, Radu *et al.*[1] described energy relaxation in a Fe-Gd alloy using SD, modeling electron and lattice subsystems by the heat transfer equations and including spin-electron interactions through a fluctuation term. The range of validity of such a model is limited, since it cannot describe the mechanical response of the lattice to magnetic excitations. *Ab initio* MD[2] and SD[3] can in principle treat the combined dynamics of atomic and magnetic degrees of freedom, but at present applications of such models are limited to system containing no more than a few hundred atoms.

Spin-lattice dynamics (SLD) simulation,[4] using a numerically efficient Suzuki-Trotter decomposition (STD) algorithm,[5] follows the evolution of magnetic and atomic degrees of freedom for systems containing in excess of 10^6 magnetic atoms. 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 the SLD equations, and by mapping them onto the three-temperature model (3TM),[6] it appears possible to formulate a self-consistent dynamic model for energy transfer between the spin, lattice and electron subsystems. In this paper, we outline the model and illustrate its applications by simulating a compressive wave propagating in ferromagnetic iron.

Assuming that magnetic properties of the material are described by the Heisenberg model, a Hamiltonian, including lattice, spin and electron subsystems, can be written as[4]

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

where

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

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

and $\mathcal{H}_e$ is the Hamiltonian of conduction electrons. Here, $\mathbf{p}_i$ is the momentum of atom i, $U(\{\mathbf{R}_i\})$ is the potential energy of interaction between atoms, $J_{ij}(\{\mathbf{R}_i\})$ is the coordinate-dependent exchange coupling function, and $\mathbf{S}_i$ is an atomic spin vector. We treat the variables describing the lattice and spin parts of the Hamiltonian explicitly, by solving the relevant equations of motion, whereas the electron subsystem is treated phenomenologically using the notion of spatially-dependent effective electron temperature.

The equations of motion for the atomic coordinates, momenta and spin vectors have the form:[4]

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

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

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

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 $\langle \mathbf{h}_k(t)\rangle = 0$, $\langle \mathbf{f}_k(t)\rangle = 0$, $\langle h_{i\alpha}(t)h_{j\beta}(t')\rangle$

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$= \mu_s \delta_{ij}\delta_{\alpha\beta}\delta(t-t')$ 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.

The fluctuation and dissipation terms in Eqs. (5) and (6) describe interaction with conduction electrons. This is similar to how interaction between atoms and electrons is included in molecular dynamics.[7,8] Using the fluctuation-dissipation theorem, we write the fluctuation-dissipation relations for the lattice[9] and spin[4,10] subsystems 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 in which the heat transfer terms couple it to the dynamically evolving lattice and spin subsystems:

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

Here, C_e is the electron specific heat and κ_e is electron thermal conductivity. Analytical expressions for G_{el} and G_{es} in Eq. (7) can be found by mapping the above equations onto the 3TM, and by equating the rates of energy change for each subsystem through Eqs. (1)–(6). 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 $2k_B T_s = \langle \sum_k |\mathbf{S}_k \times \mathbf{H}_k|^2\rangle / \langle \sum_k \mathbf{S}_k \cdot \mathbf{H}_k\rangle$ derived in Ref. 11.

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 the electrons are coupled to each other through the fluctuation, dissipation and the energy transfer terms. If there is no external source of energy, the SLE model describes a closed, energy conserving, system.

To illustrate applications of the model, we simulate a compressive elastic wave propagating in ferromagnetic iron. For comparison, we also perform *pure lattice* MD simulations describing the same wave, assuming that the atomic system evolves conservatively and that there is no interaction with the spin and electron subsystems. Simulations were performed using samples containing $30 \times 30 \times 550$ body-centered cubic (BCC) unit cells with the coordinate axes parallel to the [100], [010] and [001] crystallographic directions. Each sample involved a total of 990 000 atoms (and spins). Interatomic and exchange potentials for BCC ferromagnetic iron were parameterized in Refs. 12 and 13. At the start of each simulation, all the samples were thermalized to 300 K, with temperature homogeneously distributed through the sample. Simulations of electron temperature were performed using an intrinsic "finite difference" grid associated with the linked cells defined within the MD and SLD integration algorithms. The spin, lattice and electron temperatures T_s, T_l and T_e were treated as variables associated with each linked cell, where they were evaluated as averages over the atoms belonging to a cell. By defining the temperature of each interacting subsystem in this way, we formulate a finite difference algorithm for solving Eq. (7).

Practical simulations showed that an efficient and accurate numerical integration scheme was required to accurately compute the energy change associated with each integration time step. The coupled equations of motion for the atomic coordinates, momenta and spins, coupled to the diffusion equation for the electron temperature through the Langevin stochastic terms, were integrated using an algorithm[5,14] based on the Suzuki-Trotter decomposition.[15] Its symplectic nature guarantees the accumulation of small numerical error, which proves essential for the feasibility of simulations. The time step is chosen as approximately one tenth of the inverse Larmor frequency and is equal to 1 fs. All the simulations were performed using a GPU-enabled computer program run on Nvidia GTX480. In terms of thermodynamic classification, the spin-lattice part of the simulation is performed using an effective canonical ensemble. The temperature of the thermostat (electrons) is treated as a variable quantity, which is self-consistently adjusted during the simulation through the fluctuation and dissipation coupling between the discrete atomic and spin subsystems on the one hand, and the continuous electron temperature subsystem on the other hand. The combined system has no external source of energy, and the combined simulation model corresponds to a microcanonical ensemble.

A compressive wave propagating in the [001] direction was initiated using a method proposed by Holian *et al.*[17] For a system with periodic boundary conditions applied along all the axes, we introduced the initial symmetric impact by uniaxially shrinking the simulation box. This makes the boundaries on the left and right move inwards with velocities $\pm u_p$, which is equivalent to applying the action of two pistons moving with velocities $\pm u_p$.

Instead of compressing the simulation box continuously, we applied this artificial compression for only 0.5 ps, taking $u_p = 500$ m/s. During the simulation, there was no further geometric transformation applied to the sample after 0.5 ps. The formation of a transformation wave propagating through the sample following the elastic precursor stage is often observed in shock wave simulations.[18] The small lateral size of the current sample prevents dislocation nucleation and the occurrence of plastic deformation.

Figure 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 defined via the kinetic energy of atoms, the peak of T_l indicates the position of the compressive wave front corresponding to the highest velocity of moving atoms. In both SLE and MD simulations the compressive wave moves away from the edge of the cell at a constant velocity $u_s \approx 4500$ m/s, which is close to the speed of a longitudinal sound wave propagating in a single crystal of iron in the [001] direction[16] $c_{[001]} = \sqrt{C_{11}/\rho} \approx 5300$ m/s.

The main difference between the SLE and MD simulations is the rate of attenuation of the wave. It can be seen in Fig. 1 that temperatures at the wave front decreases as the wave propagates through the crystal. This attenuation occurs much faster in an SLE simulation in comparison with the MD one. The electron subsystem absorbs energy from the

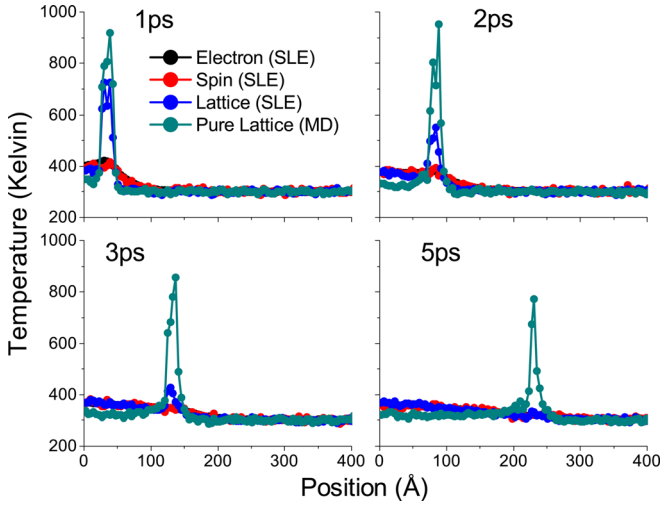

FIG. 1. (Color online) Average T_s, T_e and T_l predicted by an SLE simulation for the linked cells situated at various points in the direction of propagation of the wave at times $t = 1$ to 5 ps. Values of T_l found in a corresponding MD simulation are also shown for comparison. The rate of attenuation of the wave is found to be much higher in SLE in comparison with MD simulations, reflecting the effect of energy dissipation and energy exchange between the lattice subsystem, on the one hand, and the spin and conduction electron subsystems, on the other.

lattice through the dissipation term, and transfers energy into the spin subsystem through the fluctuation term. The effect of direct spin-lattice coupling is weaker than the sequence of lattice-electron and electron-spin interactions. Figure 1 shows that, gradually, all the energy initially stored in the compressive wave is fully dissipated. The temperatures of spin, lattice and electron subsystems rise behind the front of the wave. In a pure lattice system, modeled by MD, energy dissipates through phonon-phonon interactions, and the rate of dissipation is significantly slower than that 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 vary depending on particular experimental conditions. We treat γ_s and γ_l as phenomenological constants, the values of which reflect the complex kinetics of energy exchange between the lattice, spin and electron subsystems. In simulations described in this work, values of γ_s and γ_l were chosen by matching the results of simulations to experiments[19] on ultra-fast pulsed laser heating of magnetic materials.[6] Our use of the two parameters γ_s and γ_l for describing energy exchange between atomic, spin and electron subsystems is closely related to the notion of the three-temperature model,[1,6] which is known to describe a broad range of experimental data characterizing various materials.

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 described 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 electron, atomic 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 modeling.[18]

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