Theoretical Investigation of Non-Equilibrium Intra-Atomic Spin Transfer in Rare-Earth Atoms

Atomistic classical spin dynamics have been used in the past successfully to describe the magnetization dynamics on short timescales. However, due to their intrinsic classical character, they fail at predicting some basic thermodynamic properties such as the specific heat of the spin system or the magnon power spectrum for a larger temperature range. We aim to implement and test a quantum thermostat to investigate non-equilibrium spin dynamics.

In the state-of-the-art experiments of ultrafast magnetization dynamics researchers are hoping to track the spin transfer between localized 4f and delocalized 5d6s spins in rare-earth materials. From a theory point of view, simulations are otherwise used to help interpret highly non-equilibrium experimental observations [1]. However, the classical spin dynamics simulations which are typically used have been shown to be deficient in describing the thermal statistics of magnons [2] (Fig. 1). Barker and Bauer have recently included a quantum thermostat in spin dynamics simulations to calculate equilibrium thermodynamics of magnetic materials [3].

In this research visit I aimed to learn about this technique, implement the method and eventually extend it to non-equilibrium scenarios.

Atomistic spin dynamics are based on a classical Heisenberg model and the numerical solution the Landau-Lifshitz-Gilbert equation for classical spins:

$$\frac{\partial \boldsymbol{S}_i}{\partial t} = -\frac{\gamma}{(1+\lambda_i)} \boldsymbol{S}_i \times [\boldsymbol{H}_i + \lambda_i(\boldsymbol{S}_i \times \boldsymbol{H}_i)]$$

where $\boldsymbol{S}_i$ is a spin at lattice site 'i', γ is the gyromagnetic ratio, λ is a damping factor and $\boldsymbol{H}_i = (1/\mu_{s,i})(\partial \mathrm{H}/\partial \boldsymbol{S}_i) + \boldsymbol{\zeta}_i(t)$ is an effective field with $\mu_{s,i}$ as the magnetic moment, H the spin Hamiltonian and $\boldsymbol{\zeta}_i(t)$ is a stochastic field determined by the thermostat.

It is the stochastic thermostat term which we change to include quantum statistics, obeying the quantum fluctuation dissipation theorem

$$\varphi(\omega, T) = \frac{\hbar\omega}{\exp(\hbar\omega/k_B T) - 1}$$

The stochastic term must therefore have the correlations

$$\langle \boldsymbol{\zeta}_{i\alpha}(t) \rangle = 0$$

$$\langle \boldsymbol{\zeta}_{i\alpha}(t) \boldsymbol{\zeta}_{j\beta}(t') \rangle = 2\lambda_i \delta_{i\alpha} \delta_{j\beta} \varphi(t, t', T)/\mu_{s,i}$$

The difficulty in implementation comes from the fluctuation dissipation theorem being defined in frequency (ω) but the spin dynamics being solved in real time.

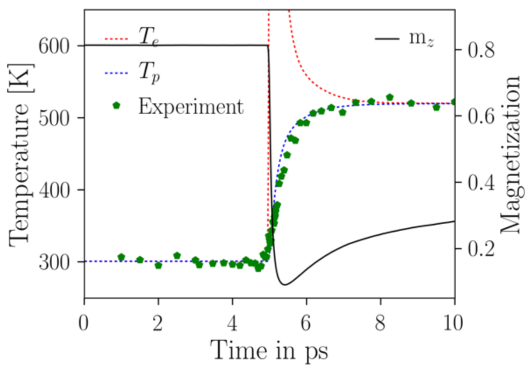

Fig. 1 A comparison of the phonon temperature and simulated ultrafast heating (electron, phonon and magnetization) of Ni using classical statistics. Although the comparison looks favorable, the extracted magnon specific heat is much too large.

We have now implemented the quantum thermostat and are performing calculations to test how it works in non-equilibrium situations with ultra-fast laser heating.

References

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