Extended microscopic three temperature model, application to ultrafast spin dynamics of 4f spins in Gd

We investigate the ultrafast magnetization dynamics induced by fs laser pulses in Gd. In particular, we study the origin of the fluence dependence of the magnetization dynamics in Gd in the whole range of time scales, from sub-picosecond to nanoseconds. Fluence dependence investigations would require to solve numerically the equation of motion for the magnetization as well as the so-called two temperature model, which calculates the evolution of the electron and phonon temperatures.

Two-temperature model.– Here we present the simplest version of the two temperature model, which is well-known,

$$C_e(T_{\mathrm{e,z}})\frac{dT_{\mathrm{e}}(z)}{dt} = G_{\mathrm{e-p}}(T_{\mathrm{p}} - T_{\mathrm{e}}) + \nabla_z(k_{\mathrm{e}}\nabla_z T_{\mathrm{e}}(z)) + S(z,t) \tag{1}$$

$$C_p\frac{dT_{\mathrm{p}}(z)}{dt} = -G_{\mathrm{e-p}}(T_{\mathrm{p}} - T_{\mathrm{e}}) \tag{2}$$

where, C_{e} and C_{p} are the electron and phonon specific heats, k_e electronic conductivity, T_e and T_p the electron and phonon temperatures. $S(t,z)$ corresponds to the laser input power and it is give by the expression

$$S(z,t) = P_0 \exp\left(-\left(\frac{t-t_0}{\tau}\right)^2\right)\exp\left(-\frac{z}{\delta}\right) \tag{3}$$

For neatness, the other physical constants are described in table I.

Microscopic three temperature model.– For the modeling of the magnetization dynamics we have extended to $4f$ spins the microscopic three temperature model introduced by Koopmans. For completeness, the original M3TM reads

$$\frac{dm}{dt} = Rm\frac{T_p}{T_c}\left[1 - \frac{m}{\tanh(mT_c/T_e)}\right] = Rm\frac{T_p}{T_c}\left[1 - \frac{m}{B_{1/2}(mT_c/T_e)}\right], \tag{4}$$

where, $B_{1/2}(x) = \tanh(mT_c/T_e)$. The parameter R is related to the spin-flip probability a_{sf}, the electron-phonon coupling, and the phonon population as follows

$$R = \frac{8a_{\mathrm{sf}}G_{\mathrm{e-p}}T_C^2 V_{\mathrm{at}}}{(\mu_{\mathrm{at}}/\mu_B)E_D^2} \tag{5}$$

I. THE EXTENDED MICROSCOPIC 3TM

Here, we present the extended 3TM we have used to model the experimental data.

First, the equation of motion for the $4f$ spins is modelled by using the Brillouin function for $S = 7/2$ in Eq. (4).

$$\frac{dm}{dt} = Rm\frac{T_p}{T_c}\left[1 - \frac{m}{B_{7/2}(mT_c/T_e)}\right], \tag{6}$$

Second, the 2TM is extended by a new term which accounts for the heat absorbed by the spin system from the electron system.

$$C_e(T_{\mathrm{e,z}})\frac{dT_e(z)}{dt} = G_{\mathrm{e-p}}(T_{\mathrm{p}} - T_{\mathrm{e}}) + \nabla_z(k_{\mathrm{e}}\nabla_z T_{\mathrm{e}}(z)) + P_{\mathrm{s-e}} + S(z,t) \tag{7}$$

$$C_p\frac{dT_{\mathrm{p}}(z)}{dt} = -G_{\mathrm{e-p}}(T_{\mathrm{p}} - T_{\mathrm{e}}) \tag{8}$$

We use that this heat flow can be readily calculated from the time derivate of the Hamiltonian, $d\mathcal{H}/dt = M_s H_{\mathrm{eff}}$, which gives relation:

$$P_{\mathrm{s-e}} = \dot{M}_s H_{\mathrm{eff}}. \tag{9}$$

Physical Constant	Symbol	Value Used	Units
Electron specific heat coefficient (γ_e)	$C_e = \gamma_e T_e$	225	$\frac{\mathrm{J}}{\mathrm{m^3K^2}}$
Phonon specific heat	C_p	1.51×10^6	$\frac{\mathrm{J}}{\mathrm{m^3K}}$
Electron-phonon coupling	G_{ep}	2.5×10^{17}	$\frac{\mathrm{J}}{\mathrm{m^3K}}$
Thermal heat conductivity coefficient (κ_0)	$\kappa = \kappa_0 T_e/T_p$	11	$\frac{\mathrm{J}}{\mathrm{mKs}}$
Laser penetration depth	δ	40	nm
Laser pump fluence prefactor	P_0	4×10^{20}	$\frac{\mathrm{J}}{\mathrm{m^3s}}$
Laser pump temporal offset	t_0	210	fs
Laser pump temporal width	τ	70	fs

TABLE I. Table of parameters used in the two temperature model code.

The main remaining question in order to implement Eq. (9) into the 2TM is the meaning of H_{eff} within the Koopmans model. The Koopmans model is not based on the common technique of deriving the equation of motion from an equilibrium potential, for example, the simplest $F \sim m^2$ from which one can easily derive $H_{\mathrm{eff}} \sim dF/dm$. So, one needs to find out what would be the magnetic potential within the Koopmans model. This is why, here, we have to use a connection of the Koopmans model to the non-equilibrium finite temperature micromagnetism as given by the Landau-Lifshitz-Bloch equation of motion. At some point of the analytical derivation of the LLB equation, which I am not going to give details here, the Koopmans model (M3TM) have the same mathematical form. From there one can extract that

$$H_{\mathrm{eff}} = \frac{k_B T_e}{\mu_s} \frac{1 - B_{7/2}/m}{B'_{7/2}} m \tag{10}$$

here, $B_{7/2} = B_{7/2}(mJ_0/T_C)$ (k_B is Boltzmann constant), J_0 is basically the exchange interaction. In the easiest picture, when we assume that only interacts with z nearest neighbors, then $J_0 = zJ$. J_0 links to the Curie temperature of the material, $J_0(S^2/S(S+1))/3 = k_B T_C$.

At equilibrium, the effective field is zero, in Eq. (10) this happens when $m_e = B_{7/2}(m_e J_0/T_c)$, which is the well-known Curie-Weiss equation for the equilibrium magnetization within the mean-field approximation. At non-equilibrium, the effective field would be have the largest value for cases where the $B_{7/2}(mJ_0/T_c)$ and m are far one from each other. At lower fluences, the non-equilibrium effective field is rather small, whereas at high fluence is larger. This impacts the total energy flow from the electron system to the spin systems.

A. Link to FMR linewidth

By using the modified M3TM to fit experimental measurements of the magnetization dynamics of Gd we can estimate the value of the so-called spin-flip scattering rate a_{sf}. As we have seen before, we find an approximate value for $R = 0.184$ in Eq. (5).

The ferromagnetic-resonance (FMR) linewidth provides information of the scattering processes leading to transverse relaxation of the magnetization. One prominent question in the field of ultrafast spin dynamics is whether or not the transverse and longitudinal relaxation of the magnetization are related or not. First attempts to unify both relaxation mechanisms were made by Koopmans[1]

In a previous publication, we connected the M3TM for $S = 1/2$ to the Landau-Lifshitz-Bloch equation of motion for the magnetization. Interestingly, the LLB equation interpolates between the low temperature LLG equation usually used to estimate the FMR, and the Bloch equation to describe the relaxation of the longitudinal component of the magnetization. The LLB equation allows one to connect both relaxation rates.

We found that the relation between R and the Gilbert damping λ can be expressed as follows

$$\lambda = \frac{3R}{2\gamma} \frac{\mu_a}{k_B T_C} = 0.000475, \tag{11}$$

for Gd, we use $R = 0.184 \times 10^{-12}\mathrm{s}^{-1}$, $\gamma = 1.76 \times 10^{-11}$ $1/(\mathrm{Ts})^{-1}$, $\mu_a = 7.5\mu_B$, $k_B = 1.38 \times 10^{-23}$ J/K, $T_C = 293$ K. $\mu_B = 9.27 \times 10^{-24}$ J/T.

[1] Unifying Ultrafast Magnetization Dynamics B. Koopmans, J. J. M. Ruigrok, F. Dalla Longa, and W. J. M. de Jonge Phys. Rev. Lett. **95**, 267207 (2005)
[2] U. Atxitia and O. Chubykalo-Fesenko, Phys. Rev. Lett. B **84**, 144414 (2011)