nature materials

SUPPLEMENTARY INFORMATION

DOI: 10.1038/NMAT2593

# The paradoxical diversity of ultrafast laser-induced demagnetization reconciled

B. Koopmans,* G. Malinowski, and F. Dalla Longa
*Department of Applied Physics, center for NanoMaterials (cNM)*
*Eindhoven University of Technology, P.O.Box 513, 5600 MB Eindhoven, The Netherlands*

D. Steiauf and M. Fähnle
*Max-Planck-Institut für Metallforschung Stuttgart,*
*Heisenbergstraße 3, 70569 Stuttgart, Germany*

T. Roth, M. Cinchetti, and M. Aeschlimann
*Department of Physics and Research Center OPTIMAS, University of Kaiserslautern,*
*Erwin-Schrödinger-Straße 46, 67663 Kaiserslautern, Germany*

(Dated: November 29, 2009)

## I. RATE EQUATION WITHIN THE MICROSCOPIC THREE-TEMPERATURE MODEL

In deriving the microscopic version of the 3TM, we use a model Hamiltonian very similar to the one we introduced in Ref. [1], but with an explicit phonon system interacting with the electrons, rather than only considering impurity scattering like in the main part of Ref. [1]. The first part of the derivation is also described in the PhD thesis by Dalla Longa [2]. The explicit way we write the final differential equations, making the system simple to calculate, has not been published before.

We aim for the simplest model system that possibly could describe the interacting electron, spin and lattice (phonon) systems in enough detail to make contact with experiments on different materials and under variable conditions (temperature, laser fluence). Thus, we consider a Fermi sea of spinless electrons with a constant density of states $D_F$, described by Bloch functions $\left|\vec{k}\right\rangle = N^{-1/2}\sum_j \exp(i\vec{k}\cdot\vec{r}_j)\left|u_j\right\rangle$ on a lattice of $N$ sites, where $\vec{k}$ is a reciprocal lattice vector, and $u_j$ is a local orbital of site $j$ at position $\vec{r}_j$. A separate spin bath is defined, obeying Boltzmann statistics and described by a total number of $N_s \equiv ND_s$ equivalent two-level systems with an exchange splitting $\Delta_{\rm ex}$ that depends in a self-consistent way on the average spin moment $\overline{S}$, i.e., using a mean-field (Weiss) description: $\Delta_{\rm ex} = J\overline{S}$, where the exchange energy $J$ is related to the Curie temperature via $k_B T_C = J/2$. Throughout this Supplementary information spin operators are defined in units of $\hbar$, i.e. $S_z = \pm 1/2$, and we drop the index $z$. For the phonon system we start by using an Einstein model of identical oscillators obeying Bose-Einstein statistics, with excitation energy $E_p$, and $D_p$ oscillators per atomic site. Later, we will include a more realistic description of the phonon dispersion, within the Debye model, in an effective way.

Our corresponding model Hamiltonian, defining the relevant interactions, reads:

$$\mathcal{H} = \mathcal{H}_e + \mathcal{H}_s + \mathcal{H}_p + \mathcal{H}_{ee} + \mathcal{H}_{ep} + \mathcal{H}_{es}, \qquad (1)$$

where

$$\mathcal{H}_e = E_k \sum_k c_k^\dagger c_k, \qquad (2)$$

$$\mathcal{H}_s = \Delta_{\rm ex} \sum_{j=1}^{N_s} S_{z,j}, \qquad (3)$$

$$\mathcal{H}_p = E_p \sum_q^{ND_p} (\frac{1}{2} + a_q^\dagger a_q), \qquad (4)$$

$$\mathcal{H}_{ee} = \frac{U}{N^2} \sum_k \sum_{k'} \sum_{k''} \sum_{k'''} c_{k'''}^\dagger c_{k''}^\dagger c_{k'} c_k, \qquad (5)$$

$$\mathcal{H}_{ep} = \frac{\lambda_{ep}}{N} \sum_k \sum_{k'} \sum_q^{ND_p} c_k^\dagger c_{k'} (a_q^\dagger + a_q), \qquad (6)$$

$$\mathcal{H}_{es} = \sqrt{\frac{a_{\rm sf}}{D_s}} \frac{\lambda_{ep}}{N^{3/2}} \sum_k \sum_{k'} \sum_q^{ND_p} \sum_j^{N_s} c_k^\dagger c_{k'} (s_{j,+} + s_{j,-})(a_q^\dagger + a_q), \qquad (7)$$

where $c_k^\dagger$ ($c_k$) are creation (annihilation) operators for electrons in state $k$, $a_q^\dagger$ ($a_q$) are similar operators for phonons in state $q$, and $s_{j,+}$ ($s_{j,-}$) are spin raising (lowering) operators at site $j$. $E_k$ is the energy of electron in state $k$, $U$ is the screened Coulomb interaction, $\lambda_{ep}$ is the e-p coupling constant, and $a_{\rm sf}$ the Elliott-Yafet scattering probability, as discussed in the main Article. The first three terms in the Hamiltonian describe the separate electron, spin and phonon system, respectively. The fourth term describes e-e scattering. It will be assumed to be efficient enough ($U \to \infty$), to keep the electron system internally in thermal equilibrium throughout our analysis. This means we can assume the electron occupation

*Electronic address: B.Koopmans@tue.nl

DOI: 10.1038/NMAT2593

to be described by a Fermi-Dirac distribution function at all times. The fifth term describes the e-p scattering (both emission and absorption of phonons). Finally, the last term is the crucial one for the magnetization dynamics. It describes the process of a spin-flip upon phonon emission or absorption during e-p scattering.

In our description of the experiment, we start with all three sub-systems at ambient temperature $T_0$, and suddenly at $t = 0$ increase the electron temperature to $T_e(0) = T_0 + \Delta T_e(0)$. The successive dynamics is treated by setting up Boltzmann rate equations for the three systems, using Fermi's golden rule, and integrating over all scattering (emission, absorption) processes. We first consider the interaction between the electrons and lattice. Integrating over all phonon emission and absorption processes, we find:

$$\dot{N}_p = \frac{\pi}{\hbar} D_F^2 E_p \lambda_{ep}^2 \left[ \coth\left(\frac{E_p}{2k_B T_e}\right) - \coth\left(\frac{E_p}{2k_B T_p}\right) \right], \tag{8}$$

where $N_p$ is the phonon occupation number according to the Bose-Einstein distribution, related to the phonon temperature via $N_p = (e^{E_p/k_B T_p} - 1)^{-1}$. The population rate can be converted to a temperature rate. Then, in the low fluence approximation, the e-p equilibration time is given by:

$$\tau_E = \frac{T_p(\infty) - T_p(0)}{\dot{T}_p} = \frac{T_e(\infty) - T_e(0)}{\dot{T}_e}, \tag{9}$$

where $T_p(\infty) = T_e(\infty)$ is the temperature of electrons and lattice after equilibration. Assuming $c_p \gg c_e$, and solving for the lowest order in $E_p/k_B T$, one finds:

$$\tau_E = \frac{\hbar \pi k_B T_e}{6 D_F D_p E_p \lambda_{ep}^2}. \tag{10}$$

Alternatively, using an ordinary two-temperature model in terms of coupling constant $g_{ep}$, a constant $C_p$ and $C_e = \gamma T_e$ (see Methods section), yields within the same approximations:

$$\tau_E = \frac{\gamma T_e}{g_{ep}}. \tag{11}$$

For an electronic system, with constant density of states $D_F$, one has

$$\gamma = \frac{\pi^2 D_F k_B^2}{3 V_{\mathrm{at}}}. \tag{12}$$

Thus, equating the two expressions for $\tau_E$ allows us to relate the phenomenological ('macroscopic') e-p coupling constant $g_{ep}$ to microscopic parameters:

$$g_{ep} = \frac{2\pi D_F^2 D_p E_p k_B}{\hbar V_{\mathrm{at}}} \lambda_{ep}^2. \tag{13}$$

By neglecting the spin specific heat, the electron and phonon dynamics is not affected by the spin system, and the above treatment remains valid after including interactions with the spins.

Next, we perform a similar analysis for the spin system. The resulting microscopic expression for the spin rate reads:

$$\dot{S} = -\frac{2\pi a_{\mathrm{sf}} \lambda_{ep}^2 D_F^2 D_p k_B \Delta_{\mathrm{ex}} T_p}{\hbar E_p D_s} \left(1 + 2S \coth\left(\frac{\Delta_{\mathrm{ex}}}{2k_B T_e}\right)\right), \tag{14}$$

where $\Delta_{\mathrm{ex}}$ is the exchange splitting (see below). In order to have as few microscopic parameters in our final differential equation, we substitute Eq. 13, and rewrite the differential equation for the average spin moment (Eq. 14) into one for the normalized magnetic moment. Further using $S = -m/2$, $\mu_{\mathrm{at}}/\mu_B = D_s$ (where $m = M/M_s$ is the normalized magnetic moment at finite temperature), and realizing that within the Weiss model $\Delta_{\mathrm{ex}}/(2k_B T_C) = m$, yields:

$$\frac{dm}{dt} = Rm \frac{T_p}{T_C} \left(1 - m \coth\left(\frac{mT_C}{T_e}\right)\right) \tag{15}$$

with

$$R = \frac{4 a_{\mathrm{sf}} g_{ep} k_B T_C^2 V_{\mathrm{at}}}{(\mu_{\mathrm{at}}/\mu_B) E_p^2}, \tag{16}$$

which equals Eq. (3) used in our Article, except for the substitution $E_p^2 = E_D^2/2$. This last step is obtained after inspection of the different role played by $E_p$ in the e-p energy equilibration and demagnetization process. The energy rate during e-p equilibration scales linearly in $E_p$ (Eq. 13), while the rate of demagnetization scales in an inverse way, i.e., $\propto E_p^{-1}$ (Eq. 14). Thereby, phonons with a smaller energy can be relevant for demagnetization, while the e-p equilibration is dominated by large energy (zone-edge) phonons. To treat this difference in an effective way, we consider a Debye model for the phonon system (a linear dispersion, i.e., a phonon density of states that scales quadratic in energy, and with a maximum phonon energy of $E_D$). Averaging over this density of states, we replace

$$E_p \to \overline{E_p} = \frac{\int_0^{E_D} E \cdot E^2 dE}{\int_0^{E_D} E^2 dE} = \frac{3}{4} E_D \tag{17}$$

in Eq. 13, and

$$E_p^{-1} \to \overline{E_p^{-1}} = \frac{\int_0^{E_D} E^{-1} \cdot E^2 dE}{\int_0^{E_D} E^2 dE} = \frac{3}{2} E_D^{-1} \tag{18}$$

in Eq. 14. Finally, inserting these DOS-averaged values into Eq. 16, leads to

$$R = \frac{4 a_{\mathrm{sf}} g_{ep} k_B T_C^2 V_{\mathrm{at}} \overline{E_p^{-1}}}{(\mu_{\mathrm{at}}/\mu_B) \overline{E_p}} = \frac{8 a_{\mathrm{sf}} g_{ep} k_B T_C^2 V_{\mathrm{at}}}{(\mu_{\mathrm{at}}/\mu_B) E_D^2}, \tag{19}$$

which completes our derivation.

## II. DATA ANALYSIS

### Nickel and cobalt, general

Throughout our analysis, we use literature values as indicated in Table I. Here, we address specific procedures used to fit the TRMOKE time traces for Ni and Co in Fig. 3 of the main Article.

In order to increase computational speed during the least-squares fitting process, we divide the ferromagnetic layer (10 nm in Fig. 3a, 15 nm in the others) in five separate slabs. We verified that using a finer mesh size did not substantially change the results. We chose an optimal absorption depth of $\lambda = 15\,\mathrm{nm}$.

Heat diffusion to the substrate is taken care of in a pragmatic way, by adding a slab (heat sink) with a large ('infinite') heat capacity and a fitting parameter $\kappa'$ to adjust heat diffusion. Since we're only interested in the onset of heat leakage and not in the complete thermal profile that develops over tens of ps, this just provides enough freedom for correcting data in the 1-5 ps regime with a single parameter. Furthermore, the underlayer having a much larger optical penetration depth than the ferromagnetic film, it is assumed not to be heated by the laser pulse.

There are two parameters that cannot be obtained from ordinary equilibrium experiments: $g_{ep}$ and the ratio $C_p/\gamma$. The value of $g_{ep}$ can be adjusted to reproduce the low fluence e-p equilibration time $\tau_E \sim 0.4\,\mathrm{ps}$. More specifically, we used the fitted value in the low fluence limit for Ni (Fig. 3a, main Article). Values of $C_p$ are fitted from the TRMOKE traces, upon the constraint that the total heat capacity at 300 K keeps equal to its literature value, i.e., adjusting $\gamma$ at the same time.

In fitting data, we use the outcome of the three coupled differential equations (M3TM), and convolute the final result with a gaussian with a half-width-at-$1/e$ $\Gamma$. To deduce the corresponding value of the demagnetization time $\tau_M$, we redo the simulation for the iterated parameters, while setting $\Gamma = 0$. $\tau_M$ is than obtained according to exactly the same procedure as used in our theoretical analysis, i.e., determining the time at which the drop of the magnetization towards its lowest value is completed by 63% (i.e., $1-1/e$).

| | Ni | Co | Gd |
|---|---|---|---|
| $\rho$ [kg m$^{-3}$] | 8900 | 8860 | 7895 |
| $\kappa$ [J (s m K)$^{-1}$] | 90.7 | 100 | 10.5 |
| $C$ [J (kg K)$^{-1}$] | 445 | 421 | 230 |
| $C_p$ [$10^6$ J m$^{-3}$ K$^{-1}$] | 2.33 [a] | 2.07 [b] | 1.78 [c] |
| $g_{ep}$ [$10^{18}$ J (s m$^3$ K)$^{-1}$] | 4.05 [d] | 4.05 [e] | 0.21 [c] |
| $E_D$ [eV] | 0.036 | 0.0357 | 0.0173 |
| $\mu_{at}$ [$\mu_B$] | 0.62 | 1.72 | 7.55 [f] |
| $T_C$ [K] | 627 | 1388 | 297 |
| $a_{sf}$ [1] | 0.185 [a] | 0.150 [b] | 0.08 [g] |
| $R$ [ps$^{-1}$] | 17.2 | 25.3 | 0.092 |

TABLE I: Parameters used in the modeling, taken from the CRC Handbook of Chemistry and Physics, 89th edition, ed. by D.R. Lide (2008), unless specified otherwise. [a]: Fitted from TRMOKE experiment, Fig. 3b. [b]: Id., Fig. 3c. [c]: To match $T_e$ and $T_p$ transients of Ref. [3]. [d]: Fitted by adjusting $\tau_M$ in Fig. 3a. [e]: Assumed to be equal to Ni. [f]: From Ref. [4]. [g]: Fitted from data in Ref. [5]. The last row lists the value of $R$, as calculated from Eq. 4 in the Methods section, and using parameters as listed in the table.

### Nickel, low fluence

In order to account for state-filling effects in this degenerate pump-probe experiment, we add a gaussian peak at zero delay time to account for coherent/state filling effects [6]. The amplitude $A_0$ of this gaussian is treated as an adjustable parameter, while its width is assumed equal to the broadening $\Gamma$. Fitting values of $\Gamma$, $A_0$, $g_{ep}$, $C_p$ and $\kappa'$ with a least-squares routine yields a spin flip probability of $0.19 \pm 0.03$. The blue dashed line in Fig. 3a in our Article represents the fitted result upon setting $A_0 = 0$, i.e., representing the genuine magnetization dynamics.

### Nickel and cobalt, high fluence

These experiments were performed with a non-degenerate pump-probe scheme. As a consequence, we find no trace of an instantaneous state-filling contribution, and thus can set $A_0$ to zero. We find that $\Gamma$ hardly effects the outcome of the results, and fix $\Gamma = 0.10\,\mathrm{ps}$ throughout the analysis. The e-p coupling constant $g_{ep}$ is assumed to be equal to the Ni low fluence data. Also the other materials specific parameters $C_p$ ($\gamma$) and $\kappa'$ are assumed equal for measurements at different laser fluence. Fitting the set of experimental curves this way, yields a separate value of $a_{sf}$ for each value of the fluence. However, we find no systematic dependence of $a_{sf}$ on the laser fluence, and obtain average values of $a_{sf} = 0.185 \pm 0.015$ and $0.150 \pm 0.015$ for Ni and Co, resp.

### Gadolinium analysis

In order to obtain an estimate of $a_{sf}$ for gadolinium, we use recent data from Bovensiepen, Melnikov, Wietstruk, and coweorkers. They performed time-resolved XMCD with different time-resolution, ranging from 50 ps [3], via 12 ps (using low-$\alpha$ mode, [5]) to $\sim 100$ fs (using fs-slicing, [5]). The authors observed a two-step demagnetization process described by an initial decrease by $\Delta m_1 \approx 25\%$ within $\tau_1 = 1.0 \pm 0.2$ ps, and a final demagnetization towards $\Delta m_f \sim 0.5$ at a time scale $\tau_M = 40 \pm 10\,\mathrm{ps}$. We again applied a least-squares fitting routine, similar to the one employed for the Ni and Co data, to achieve a best fit of our theoretical model to all four experimental parameters ($\tau_M$, $\tau_1$, $\Delta m_1$ and $\Delta m_f$). Like for the Ni and Co case, we use a simplified heat diffusion approach, using a Gd film of 10 nm and fitting $\kappa'$ to reproduce the experimentally observed $m(t)$ for long time delays (up to 200 ps). As to fitting $g_{ep}$ we followed two approaches. Either we used a best fit of $g_{ep}$ to get optimal agreement with the electron temperature profiles as reported by Bovensiepen and Melnikov [3, 4] or we fitted it to get

DOI: 10.1038/NMAT2593

optimal agreement with $\tau_1 = 1.0$ ps. Both approaches yielded quite similar results. Other parameters used are given in Table I. Following the procedure outlined above, we obtained as a best fit to the experimental data: $a_{\rm sf} = 0.08 \pm 0.02$. We emphasize that within our theory it is thus possible to explain all four parameters defining the type-II demagnetization process without introducing additional mechanisms and/or parameters.

**TbFe analysis**

As discussed in our Article, very recently Kim et al. reported on a two-step demagnetization process in a ferrimagnetic $Tb_{35}Fe_{65}$ alloy, as measured by TRMOKE [7]. Also in that case, the duration of initial fast demagnetization corresponded to the e-p equilibration time, as explicitly measured by transient reflectivity. We applied the same fitting procedure as for Gd to the TRMOKE data. As shown in Fig. 4c of our main Article, we found a perfect fit, with a fitted result $a_{\rm sf}/\mu_{\rm at} = 0.056\mu_B^{-1}$. Note that due to the fact that magnetic moments at Fe and Tb atoms are aligned in a *ferri*magnetic (antiparallel) fashion, it is unclear which value of $\mu_{\rm at}$ one should choose. If we associate $\mu_{\rm at}$ in the M3TM with the average of the absolute value of the magnetic moments at each atomic site (i.e., use the 'staggered' magnetization rather than the net magnetization), we obtain $\mu_{\rm at} = 0.67|\mu_{\rm Fe}| + 0.33|\mu_{\rm Tb}| = 3.4\mu_B$, and consequently $a_{\rm sf} = 0.19$. Alternatively, we could have used the averaged magnetic moment per atom, leading to $\mu_{\rm at} = 0.67|\mu_{\rm Fe}| - 0.33|\mu_{\rm Tb}| = 0.5\mu_B$ and $a_{\rm sf} = 0.03$. We emphasize that the range of $a_{\rm sf}$ found this way is again of the same order of magnitude as for Ni, Co and Gd. However, because our theory is not designed for such ferrimagnetic systems we refrain from too strict conclusions. Nevertheless, the preliminary analysis may show a glimpse of the universality of type II dynamics, and certainly defines a highly interesting route for future research.


[1] Koopmans, B., Ruigrok, J.J.M., Dalla Longa, F. & de Jonge, W.J.M. Unifying ultrafast magnetization dynamics. *Phys. Rev. Lett.* **95,** 267207 (2005).

[2] Dalla Longa, F., Laser-induced magnetization dynamics –an ultrafast journey among spins and light pulses–. *PhD thesis, Eindhoven University of Technology, Eindhoven, The Netherlands* (2008); http://alexandria.tue.nl/extra2/200810818.pdf.

[3] Melnikov, A. *et al.* Nonequilibrium magnetization dynamics of gadolinium studied by magnetic linear dichroism in time-resolved 4f core-level photoemission. *Phys. Rev. Lett.* **100,** 107202 (2008).

[4] Bovensiepen, U. Coherent and incoherent excitations of the Gd(0001) surface on ultrafast timescales. *J. Phys. Cond. Mat.* **19,** 083201 (2007).

[5] Wietstruk, M., Kachel, T., Pontius, N., Stamm, C., Drr, H., Eberhardt, W., Melnikov. A., Bovensiepen, U., Gahl, C., & Weinelt, M. Ultrafast magnetization dynamics in Gd studied by time resolved XMCD. *International Conference on Ultrafast Surface Dynamics 6*, Kloster Banz, Germany, July 20-25, 2008; Wietstruk, M., et al., submitted.

[6] Dalla Longa, F., Kohlhepp, J.T., de Jonge, W.J.M. & Koopmans, B. Influence of photon angular momentum on ultrafast demagnetization in nickel. *Phys. Rev. B* **75,** 224431 (2007).

[7] Kim, J.W., Lee, K.-D., Jeong, J.-W. & Shin, S.-C. Ultrafast spin demagnetization by nonthermal electrons of TbFe alloy film. Appl. Phys. Lett. 94, 192506 (2009).