## Ultrafast magnetization dynamics at very high magnetic fields and elevated temperatures

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The role of external magnetic fields is often neglected in the field of ultrafast magnetization dynamics induced by femtosecond laser pulses. Here it is shown theoretically that very high magnetic fields can substantially modify ultrafast magnetization dynamics at elevated temperatures. Magnetic fields speed up the ultrafast magnetization dynamics. Notably, we predict that the observed two-step demagnetization dynamics in some materials transition to one-step demagnetization dynamics for high magnetic fields. The description of the dynamics is based on the Landau-Lifshitz-Bloch equation of motion. Analytical expressions for the longitudinal and transversal magnetization relaxation time as a function of the magnetic field are provided.

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#### I. INTRODUCTION

Ultrafast spin dynamics in magnetic materials have been routinely investigated by femtosecond laser pulses since in 1996 Beaurepaire and co-workers uncovered the possibility to use them to break the magnetic order in ferromagnetic Ni thin films [1]. A range of microscopic spin-related mechanisms have been proposed to explain the observed subpicosecond magnetic order break down; electron-phonon spin-flip scattering [2], nonequilibrium electron-magnon interaction [3], super diffusive spin dependent electron transport [4], and many others, which have been well summarized recently by Illg et al. [5]. Yet it remains a theoretical and experimental challenge to disentangle the particular role of each of those mechanisms in the demagnetization processes. In the quest of further insights into the underlying physics behind the demagnetization dynamics, a range of experiments have been devised to disentangle the importance of the proposed mechanisms, examples include variation of the laser fluence [6], light polarity [7], ambient temperature [2,8-10], and multilayers with insulating/conductor substrate to avoid/promote the effect of superdiffusive spin currents [11,12].

Interestingly, by varying the initial temperature up to the Curie temperature, there exists a transition from a demagnetization/remagnetization dynamics-type I-to a twostep demagnetization dynamics-type II. This transition has been observed in Ni [8] and FePt [13]. Direct comparison of those observations to theoretical models, such as the Landau-Lifshitz-Bloch equation [6] or the microscopic threetemperature model [8], has revealed that this transition is related to the so-called critical slowing down of the spin dynamics close to the phase transition. Surprisingly, it has been recently demonstrated that such a critical behavior also drives the out-of-equilibrium laser induced magnetic phase transition in nickel within 20 fs [14]. At those time scales the electron temperature reaches its maximum temperature, and in principle, its value should increase monotonously with the power of the laser, however, it shows a clear shift at the point the magnetic specific heat diverges. This divergence could be strongly reduced by external magnetic fields.

However, the role of the external magnetic field is often neglected and barely used as active stimulus in the field of ultrafast spin dynamics [15,16]. It is simply used to restore initial conditions in spectroscopic, pump-probe measurements. At those timescales, the spin dynamics is defined by the misalignment between spins against the strong exchange interaction. This creates internal magnetic fields at the atomic level with characteristic timescales of tens of femtoseconds. In comparison, the timescales accessible by magnetic fields are usually of the order of tens of picoseconds. Hence, it is sensible to assume that magnetic fields barely affect the subpicosecond spin dynamics. Only recently Haag et al. [17] have succinctly tackled the problem by investigating the effect of ultrahigh magnetic fields (up to 1000s of Tesla) using first-principles computer calculations. In particular, they estimated the value of the spin mixing at ultrahigh external fields. Within the framework of electron-phonon mediated spin-flip mechanism, spin mixing is related to the demagnetization rate. The authors showed that the spin mixing is nearly independent of the external magnetic fields, and thus any possible modification of the ultrafast spin dynamics should come from the modification of the spin wave spectrum. Along this line and as mentioned before, recent observations of the ultrafast electron temperature increase and spin dynamics in Ni [14] suggest that at short times high-energy magnon excitations store part of the energy from the laser, which is subsequently transferred to lower energy excitations. This mechanism would depend directly on the energy gap of the spin wave spectrum, which can be controlled by the magnetic field. Further theoretical and experimental investigations on the interplay between temperature and high magnetic fields effects could help in the understanding of the ultrafast magnetization dynamics.

An adequate framework that correctly accounts for the modifications of the spin wave spectrum are the atomistic spin dynamics based on the Landau-Lifshitz equation [18]. While atomistic spin dynamics is a useful method for modeling of experiments, insights about the physics behind computer simulations can only be gained from their macroscopic counterparts, e.g., the Landau-Lifshitz-Bloch (LLB) equation [19]. This is the reason why in this work we investigate the effect of very

high magnetic fields on the magnetization dynamics at the macroscopic level using the LLB framework [20]. However, in the standard derivation of the LLB equation of motion the external magnetic fields are assumed to be small in comparison to the exchange interaction. Thus, in this work we also present a correction to the standard LLB equation to account for high magnetic fields. By using this framework, we show that the presence of magnetic fields during ultrafast heating speeds up the magnetization dynamics. For starting temperatures close to the critical temperature  $T_C$ , the presence of high magnetic fields prevents the critical slowing down associated with the phase transition. As a direct consequence, we predict that the transition from type I to type II dynamics can be reversed by the application of a sufficiently high magnetic field. As an illustrative example of such a transition, we apply the developed model to a realistic situation, the magnetization dynamics of gadolinium after the application of a femtosecond laser pulse. Gadolinium is both the prototypical Heisenberg and type II ferromagnet. We show that high magnetic fields can convert gadolinium into a type I material. Experimental verification of such predictions would shed light into the role of the critical behavior of the magnetic system in the ultrafast dynamics, as well as into the fundamentals of the laser induced magnetic phase transitions.

## II. THE LANDAU-LIFSHITZ-BLOCH APPROACH TO DESCRIBE MAGNETIZATION DYNAMICS

The LLB equation is a quite recent approach that has been shown to adequately describe the magnetization dynamics at elevated temperatures [19,20]. Differently to the low temperature Landau-Lifshitz equation, the LLB equation does not conserve the length of the magnetization vector, rather it allows for longitudinal fluctuations of the magnetization in space and time. The LLB equation has not only been widely used in the context of ultrafast spin dynamics [6,9,13,21] but also to describe many other phenomena; spin torque transfer [22–24], vortex core reversal [25,26], and spin caloritronic effects [27,28]. Therefore, a number of fields benefit from further improvements of the LLB framework.

The description of the ultrafast magnetization dynamics using the LLB model has been shown so far successful [20]. Direct comparison to experiments include; fluence and thickness dependence of the ultrafast spin dynamics of Ni thin films [6], electron- and phonon-mediated demagnetization in Gd thin films [9], FePt demagnetization dynamics [13], as well as all-optical switching behaviors in FePt nanoparticles [29]. Moreover, the LLB model has been shown to be equivalent to the microscopic three-temperature model proposed by Koopmans and co-workers, which has been used to model optically driven ultrafast magnetization dynamics of Ni and Co [2] and hot electron induced demagnetization in Co/Pt [30]. Those works were made in the presence of negligible magnetic fields, in comparison here we investigate the ultrafast magnetization dynamics in the presence of high magnetic fields.

## A. Dynamical equation

The equation of motion of the thermally averaged spin polarization of an isolated magnetic moment in the sole presence magnetic field H reads [19]

$$\frac{d\mathbf{m}}{dt} = \gamma [\mathbf{m} \times \mathbf{H}] - \Gamma_{\parallel} \left( 1 - \frac{\mathbf{m} \mathbf{m}_0}{m^2} \right) \mathbf{m} - \Gamma_{\perp} \frac{[\mathbf{m} \times [\mathbf{m} \times \mathbf{H}]]}{m^2}.$$
(1)

The first term describes the precession around  $\mathbf{H}$ , the second term the relaxation of the magnetization length, and the third term describes the transverses relaxation of the magnetization towards equilibrium. The relaxation frequencies in Eq. (1) are defined as

$$\Gamma_{\parallel} = \Lambda_N \frac{1}{\xi_0} \frac{L(\xi_0)}{L'(\xi_0)}, \quad \Gamma_{\perp} = \frac{\Lambda_N}{2} \left( \frac{\xi_0}{L(\xi_0)} - 1 \right).$$
(2)

Here the relaxation frequencies  $\Gamma_{\parallel(\perp)}$  have two contributions. First,  $\Lambda_N = 2\lambda k_{\rm B}T/(\mu_0/\gamma)$  is the rate ( $\lambda$ ) at which the thermal energy  $k_{\rm B}T$  is distributed into the magnetic system defined by  $\mu_0/\gamma$ , where  $\gamma$  is the gyromagnetic ratio and  $\mu_0$  is the atomic magnetic moment. Second, in Eqs. (2), information about the thermodynamic field  $\xi_0 = \beta \mu_0 H$  is contained implicitly.  $L(\xi_0)$ is the Langevin function which defines the transient effective field towards which the nonequilibrium magnetization relaxes,  $m_0 = L(\xi_0)$ . At equilibrium, it reduces to  $m_0 = m_e = L(\xi_e)$ . The derivative is defined,  $L'(x) \equiv dL/dx$ . Interestingly, for demagnetization processes involving spin flip from majority to minority spin bands, the rate  $\lambda$  depends directly from the spin mixing, and it has been shown to be nearly independent of the magnetic fields, up to 1000 T [17].

Equation (1) is valid for an isolated spin in the presence of a magnetic field, however this approach can be easily extended for ferromagnets within a mean-field approximation (MFA) framework. One only needs to substitute  $\mathbf{H} \rightarrow \mathbf{H}_{\text{MFA}}$ , where  $\mathbf{H}_{\text{MFA}}$  is the MFA effective field acting on  $\mathbf{m}$ . For a isotropic Heisenberg model with only first nearest neighbors (z) exchange interaction J ( $J_0 = zJ$ ) and magnetic field, the Hamiltonian can be expressed as  $\mathcal{H} = -J \sum_{\langle ij \rangle} \mathbf{S}_i \mathbf{S}_j - \mu_0 H \sum_i \mathbf{S}_i$ , here  $\mathbf{S}_i$  is the normalized classical spin vector at lattice site *i*. Thus,  $\xi_0 = \beta \mu_0 H_{\text{MFA}} = \beta (J_0 m + \mu_0 H)$  includes the effect of the magnetic field in the relaxation rates in Eq. (1) through the highly nonlinear expression in Eqs. (2).

#### B. Dynamics of the magnetization modulus

For ultrafast demagnetization processes, where the magnetic field is directed along the z axis, the dynamics is described by

$$\frac{dm_z}{dt} = \Gamma_{\parallel}(m_z - m_{z,0}),\tag{3}$$

where  $m_{z,0} = L(\xi_0)$ , with  $\xi_0 = \beta(J_0m_z + \mu_0H)$ , and  $\Gamma_{\parallel}$  given by Eqs. (2). Equation (3) can be used to illustrate the effect of very high fields in the ultrafast magnetization dynamics. To simplify the problem, we assume that the effect of the laser pulse is to provide a heat bath to the magnetic system defined by the electron temperature  $T_e$ . Further simplification is to assume a steplike form for the electron temperature  $T_e = T_{\text{room}}$ for time t < 0, here  $T_{\text{room}}$  stands for room temperature. The fast increase on the electron temperature is minicked by a step pulse of 1 ps, and a maximum temperature  $T_{\text{max}}$ . After the pulse action is gone the electron temperature goes back to a final temperature  $T_e = T_f$ , which is higher than the initial



FIG. 1. Temperature driven transition from type I to type II dynamics. This transition occurs when the final electron temperature is larger than the corresponding transient temperature of the magnetic state.

 $T_{\text{room}}$ . This model for the electron temperature captures the main aspects of the ultrafast magnetization dynamics triggered by laser pulses. For instance, when the magnetic system starts close to  $T_C$  it has has been shown that the demagnetization dynamics slows down [2,8]. Increasing the laser fluence, and thus the transient electron temperature, has shown a to produce a similar behavior [6,13]. Interestingly, this slowing down of the magnetization dynamics causes the transition from type I—subpicosecond demagnetization/ps remagnetization—to type II—two-step demagnetization dynamics; an initial subpicosecond demagnetization followed by picosecond demagnetization process.

To illustrate this purely thermal effect we use the LLB equation, in the  $H \rightarrow 0$  limit, to calculate the magnetization dynamics after the application of a heat pulse of temperature  $T_{\rm p}$ , for different final temperatures  $T_{\rm final}$ . In Fig. 1 we reproduce the transition from type I to type II dynamics. For final temperature near the critical temperature ( $T_C = 650$  K) the transient temperature of the magnetic state stays below the final temperature owning to the high spin specific heat close to  $T_C$ . Consequently, at the moment the remagnetization dynamics should take over, the temperature of the magnetic system is still below the electron temperature. Thus, instead of a remagnetization process (type I dynamics) it continues demagnetizing (type II dynamics) until both subsystem temperatures equalized [31]. This transition between type I and type II dynamics has also been reported in FePt [13] which is one of the potential candidates for ultradense magnetic recording media due to its high anisotropy in the  $L1_0$  phase. Thus, the critical slowing down could set a speed limit in ultrafast heat-assisted magnetization reversal [32].

Differently to the effect of increasing ambient temperature, when one increases the value of the magnetic field, a transition from type I to type II dynamics is observed. To illustrate this transition, we calculate the magnetization dynamics after the application of a heat pulse by solving the LLB equation (3). We fix the heat pulse temperature  $T_p = 700$  K and the final temperature  $T_{\text{final}} = 600$  K. As depicted in Fig. 2, when the magnetic field is low, H = 1 T, the transition from type I to type II still occurs, similar to Fig. 1. As the strength of the magnetic



FIG. 2. Field driven transition from type II to type I dynamics. The final temperature is fixed and the strength of the magnetic field is varied up to 40 T. The peak demagnetization depends on the magnetic field strength, and the two-step demagnetization transits (type II) to a demagnetization/remagnetization process (type II).

field is increased we can observe that the magnetization starts to recover instead of following a second demagnetization. That is transit from type II to type I dynamics. At the same time, we can observe that the maximum demagnetization and the magnetization recovered at 4 ps depend on the magnetic field value. This is related to the field dependence of equilibrium magnetization as we can observe in Fig. 3. The magnetization relax towards those values, which are magnetic field dependent. Although stronger effects are expected for even larger magnetic fields, we restrict ourselves to values (40 T) which are presently accessible.

# C. Application of the model to Gd; transition from type II to type I

To show how robust our predictions are in comparison to real situations, we model the dynamics of Gd magnetization within a realistic model for the electron temperature dynamics. We use Gd since it is the benchmark material for both Heisenberg ferromagnet and type II dynamics. Gd is the ferromagnetic rare earth with the highest Curie temperature  $T_C = 293$  K, and where magnetic properties are determined mostly by the seven 4 *f* electrons, strongly localized at the ion core, providing an atomic moment of  $\mu_{4f} = 7 \mu_B$ . Plus, some contribution from the 5*d*6*s* delocalized electrons  $\mu_{5d6s} = 0.55 \mu_B$ . So far experimental observations in Gd have only observed a two-step



FIG. 3. Field dependence of the equilibrium magnetization as a function of the magnetic field.



FIG. 4. (Up) Electron temperature dynamics. (Bottom) Dynamics of Gd magnetization for four different magnetic fields H. One can observe a clear transition from two-step—type II—to one demagnetization dynamics—type I—dynamics.

demagnetization behavior, that is, type II dynamics. Models describing these dynamics include atomistic spin dynamics [10], the microscopic three-temperature model [2], and the LLB model [9]. All the experiments have been performed in the presence of a small external magnetic field. Here we shortly present the effect of high magnetic fields on the dynamics of the Gd magnetization, from subpicosecond timescale to hundreds of picoseconds.

We use the so-called two-temperature model to describe the dynamics of the electron and lattice energy are modeled by the atomic layer resolved two-temperature model (2TM)

$$C_{\rm e}(T_{\rm e})\frac{dT_{\rm e}}{dt} = G_{\rm ep}(T_{\rm e} - T_{\rm ph}) + \nabla_z(k_e \nabla_z T_e) + P(t), \quad (4)$$

$$C_{\rm ph}\frac{dT_{\rm ph}}{dt} = -G_{\rm ep}(T_{\rm e} - T_{\rm ph}),\tag{5}$$

where electron-phonon coupling  $G_{ep} = 2.5 \times 10^{17}$  W(K m<sup>3</sup>)<sup>-1</sup>,  $C_e(T_e) = \gamma_e T_e$ , with  $\gamma_e = 2.25 \times 10^2$  J m<sup>-3</sup> K<sup>-2</sup>,  $C_{ph} = 1.51 \times 10^6$  J m<sup>-3</sup> K<sup>-1</sup>,  $k_e = k_{e,0}T_e/T_{ph}$ , where  $k_{e,0} = 11$  W(K m)<sup>-1</sup>. These parameters have been used in a number of works, and it has been demonstrated to be sufficient to describe the dynamics of the electron temperature [33]. The magnetization dynamics is calculated using Eq. (3) for each atomic layer. In our calculations we consider a thin film of 100 nm. A similar model was already used by Sultan *et al.* [9] to directly compare experimental observations to the LLB model of the dependence of demagnetization dynamics with the initial temperature of Gd thin films, in the absence of high magnetic fields.

Furthermore, here we link the LLB equation to the microscopic three-temperature model (M3TM) by considering that the intrinsic damping parameter is temperature dependent,  $\lambda = \lambda_0 (T_{\rm ph}/T_{\rm e})$ , a connection demonstrated in Ref. [34]. We show in the upper panel of Fig. 4 the dynamics of the electron temperature. Initially, the electron temperature increases steeply up to a peak temperature of 2000 K at only 200 fs, once the energy input from the laser is gone, the electron system cools down by releasing energy to the colder phonon system, as described by Eqs. (4) and (5). During this process

the first demagnetization step happens, we note here that a similar demagnetization is observed when we used a simple temperature step pulse of 1 ps, see Fig. 1. As we can see in Fig. 4, for magnetic fields up to 20 T, the first demagnetization is followed by a second one, that is, a type II dynamics. However, as we increase the magnetic field H, the relaxation time of the second demagnetization process reduces, and for 40 T it transitions to one-step demagnetization dynamics. Very high magnetic field experiments are in a very early stage, however we expect that this theoretical prediction could soon be tested.

## III. THE CLOSED LLB EQUATION IN THE PRESENCE OF HIGH FIELDS

In contrast to the longitudinal spin dynamics, the transverse magnetization dynamics, usually seen as a damped precession around a well defined equilibrium direction, is not usually considered to be within the realm of ultrafast magnetization dynamics. The reason behind lies in the relatively low magnetic energy (and thus frequency) of the physical quantities involved in the precession dynamics, namely, the anisotropy and the magnetic field. Those two parameters commonly define the precession frequencies, which are of the order of sub-GHz. However, for high anisotropy materials and under high magnetic fields, the precession frequencies could reach the THz regime. This high-speed precession has been recently reported using the so-called optical FMR technique in FePt, where frequencies over 400 GHz were found. The optical FMR is based on the temperature changes of the anisotropy field produced by a femtosecond laser pulse which in turn temporarily changes the equilibrium state driving the magnetization to precess towards it. A correct account of the temperature and magnetic field dependence of the transverse relaxation time is therefore important to properly analyze experimental data. For example, for granular FePt, there are differing values of the Gilbert damping parameter in the literature, Becker et al. (using up to 7 T) measured a damping constant of 0.1 using an optical FMR technique [15], whereas Alvarez et al. found a value of 0.055 using standard FMR in a broad frequency range [35].

In order to further investigate the effect of high magnetic field into the spin dynamics, we present a closed form of the LLB equation (1), namely with relaxation rates independent of m, which enable us to derive analytical expression for the relaxation rates,

$$\frac{d\mathbf{m}}{dt} = \gamma [\mathbf{m} \times \mathbf{H}] - \alpha_{\parallel} \frac{(\mathbf{m} \cdot \mathbf{H}_{\text{eff}})\mathbf{m}}{m^2} - \alpha_{\perp} \frac{[\mathbf{m} \times [\mathbf{m} \times \mathbf{H}]]}{m^2}.$$
(6)

The effective field  $\mathbf{H}_{\text{eff}}$  is given by

$$\mathbf{H}_{\rm eff} = \frac{1}{\widetilde{\chi}_{\parallel}(H,T)} \left(\frac{m^2 - m_e^2}{m_e^2}\right) \mathbf{m},\tag{7}$$

here  $m_e = L(\xi_e)$  (Fig. 3) and  $\tilde{\chi}_{\parallel}(H, T)$  is the temperature and field dependent longitudinal susceptibility (Fig. 5),

$$\widetilde{\chi}_{\parallel}(H,T) = \frac{\mu}{J_0} \frac{L'(\xi_0)\beta J_0}{1 - L'(\xi_0)\beta J_0},$$
(8)



FIG. 5. Longitudinal susceptibility as a function of temperature and magnetic field.

 $\alpha_{\parallel}$  and  $\alpha_{\perp}$  are dimensionless longitudinal and transverse damping parameters and are given by

$$\alpha_{\parallel} = 2\lambda \frac{T}{3T_C(H)}, \quad \alpha_{\perp} = \lambda \left[1 - \frac{T}{3T_C(H)}\right]$$
(9)

for  $T < T_C$ , and the same for with  $\alpha_{\perp} \Rightarrow \alpha_{\parallel}$  for  $T > T_C$ . Here  $\lambda$  is the damping parameter that describes the coupling to the heat bath at the atomic level. In the MFA one can link the values of the magnetic field and the exchange interaction to the Curie temperature  $T_C(H)$  through the relation  $3k_BT_C(H) =$  $J_0 + \mu_0 H$ . Thus, the effect of the magnetic field on the longitudinal and transverse damping parameters is to shift the critical temperature towards higher values due to the magnetic fields. These parameters could also be calculated through multiscale frameworks based on hierarchical spin models, from experimental data, or more refined than MFA theoretical models [20].

Next, in order to understand the magnetization dynamics coming out after a laser pulse, we restrain ourselves to the limit of small deviations from thermal equilibrium. We consider that the initial magnetization *m* is close to the equilibrium value  $m_e$ . In this way  $m - m_e$  is considered as a small parameter. This assumption is necessary when one wants to derive analytical expressions for the characteristic relaxation times. This implies that in the following we consider that the temperature of the thermostat is kept constant during the process. In the linear regime the longitudinal dynamics described by Eq. (6) reduces to  $m - m_e \approx \exp(-\Gamma_{\parallel}t)$ , where

$$\Gamma_{\parallel} = \frac{\gamma \alpha_{\parallel}}{\widetilde{\chi}_{\parallel}(H,T)}.$$
(10)

At low temperatures,  $\Gamma_{\parallel} \sim J_0 + \mu_0 H$ , therefore the longitudinal relaxation rate is very fast. The presence of a magnetic field H speeds up the longitudinal relaxation, with  $\Gamma_{\parallel}(H)/\Gamma_{\parallel}(0) =$  $1 + \mu_0 H/J_0$ . The ratio  $\mu_B/J_0 \approx 7.6 \times 10^{-6} \text{ T}^{-1}$ , thus magnetic fields of the order of the exchange fields, namely 10<sup>4</sup> to  $10^5$  T, will be necessary to speed up the magnetization dynamics. As temperature increases, the exchange field effectively reduces. As we can see in Fig. 6, at intermediate temperatures  $T = 0.5T_C$  and  $0.7T_C$ , the speed up with H of the relaxation time  $\tau_{\parallel} = 1/\Gamma_{\parallel}$  is already appreciable. At elevated temperatures, the longitudinal susceptibility  $\tilde{\chi}_{\parallel}(H = 0, T)$  becomes rather large for small H (Fig. 5), and thus the longitudinal relaxation rate  $\Gamma_{\parallel}(H = 0, T) \rightarrow 0$ . This effect is known as the critical slowing down of the magnetization. In Fig. 1 we have already shown that this is the reason behind the transition



FIG. 6. Longitudinal relaxation time of small deviations of the magnetization modulus from the equilibrium as a function of the magnetic field H, for four temperatures, see. Eq. (10).

from type I to type II spin dynamics. Under the presence of high magnetic fields, this critical behavior is suppressed due to the *H* induced magnetization  $m_H = [(5/3)(\mu_0 H/J_0)]^{1/3}$ , and therefore the longitudinal relaxation rate at  $T_C$  is finite and depends on the magnetic field  $\Gamma_{\parallel} \cong (6/5)\gamma\lambda(J_0/\mu_0)m_H^2$ . This means that the longitudinal spin dynamics is faster as the strength of the magnetic field increases (Fig. 6). Notably, near  $T_C$  we observe that the relaxation time of the magnetization can speed up by almost an order of magnitude for magnetic fields of H = 50 T, values accessible in current experimental set ups in high magnetic field labs.

Thus, near  $T_C$  the magnetic field suppresses the critical behavior, and thus the transition from type I to type II dynamics can be correspondingly reversed back by the application of a strong enough magnetic field, as presented in Figs. 2 and 4. This observation is consistent with the fact that the application of a magnetic field increases the Curie temperature, which translates to an effective temperature reduction.

Apart from the longitudinal dynamics, the transverse dynamics is also altered by magnetic fields, within the LLB equation the relaxation of the transverse dynamics is defined by  $\Gamma_{\perp} = \gamma \alpha_{\perp} / \tilde{\chi}_{\perp}(H, T)$ , where the transverse susceptibility, in absence of anisotropy contribution (isotropic model), is simple given by  $\tilde{\chi}_{\perp}(H, T) = m(H, T)/H$ , which translates into  $\Gamma_{\perp} \sim H$ . However, due to the presence of a high magnetic field, the value of m(H, T) could be significantly different to m(0, T), specially around  $T_C$ ,  $m_H = [(5/3)(\mu H/J_0)]^{1/3}$ , which gives  $\Gamma_{\perp} \approx \gamma H \alpha_{\perp} H^{1/3} J_0^{2/3}$ . This expression should be useful to interpret experimental observation at very high magnetic fields and elevated temperature, rather than the relaxation rate coming out from the low-temperature Landau-Lifshitz equation  $\Gamma_{\perp} = \lambda \gamma H$ .

To summarize, very high magnetic fields speed up the ultrafast magnetization dynamics. This speed up becomes apparent in the temperature region near the critical temperature. There, in absence of magnetic fields, the critical slowing down of the magnetic fluctuations slows significantly the ultrafast magnetization dynamics. In contrast, in the presence of high magnetic fields, we predict a transition from relatively slow to fast magnetization dynamics. This comes out as a result of the removal of the critical behavior by the presence of very high magnetic fields. Notably, we have demonstrated that Gd—prototypical type II—dynamics should present a transition to type I dynamics for magnetic fields of around 40 T. Although experiments on ultrafast magnetization dynamics

under very high magnetic fields are in a very early stage, one would expect a possible observation of the transition in the future. This would evidence the thermal nature of the ultrafast spin dynamics driven by femtosecond laser pulses. These results have been obtained by using the Landau-Lifshitz-Bloch equation of motion for the magnetization dynamics. The LLB equation-based models are recent, however it has already been utilized in a number of physical problems, such as ultrafast-spin dynamics, spin caloritronics, and so on, that could benefit for the theoretical developments presented in the present work. Importantly, dynamics of antiferromagnetically coupled spin systems, such as antiferromagnet or ferrimagnets,

- E. Beaurepaire, J.-C. Merle, A. Daunois, and J.-Y. Bigot, Ultrafast Spin Dynamics in Ferromagnetic Nickel, Phys. Rev. Lett. 76, 4250 (1996).
- [2] B. Koopmans, G. Malinowski, F. Dalla Longa, D. Steiauf, M. Fähnle, T. Roth, M. Cinchetti, and M. Aeschlimann, Explaining the paradoxical diversity of ultrafast laser-induced demagnetization, Nat. Mater. 9, 259 (2010).
- [3] E. G. Tveten, A. Brataas, and Y. Tserkovnyak, Electron-magnon scattering in magnetic heterostructures far out of equilibrium, Phys. Rev. B 92, 180412 (2015).
- [4] M. Battiato, K. Carva, and P. M. Oppeneer, Superdiffusive Spin Transport As a Mechanism of Ultrafast Demagnetization, Phys. Rev. Lett. 105, 027203 (2010).
- [5] C. Illg, M. Haag, and M. Fähnle, Ultrafast demagnetization after laser irradiation in transition metals: *Ab initio* calculations of the spin-flip electron-phonon scattering with reduced exchange splitting, Phys. Rev. B 88, 214404 (2013).
- [6] U. Atxitia, O. Chubykalo-Fesenko, J. Walowski, A. Mann, and M. Münzenberg, Evidence for thermal mechanisms in laserinduced femtosecond spin dynamics, Phys. Rev. B 81, 174401 (2010).
- [7] F. Dalla Longa, J. T. Kohlhepp, W. J. M. de Jonge, and B. Koopmans, Influence of photon angular momentum on ultrafast demagnetization in nickel, Phys. Rev. B 75, 224431 (2007).
- [8] T. Roth, A. J. Schellekens, S. Alebrand, O. Schmitt, D. Steil, B. Koopmans, M. Cinchetti, and M. Aeschlimann, Temperature Dependence of Laser-Induced Demagnetization in Ni: A Key for Identifying the Underlying Mechanism, Phys. Rev. X 2, 021006 (2012).
- [9] M. Sultan, U. Atxitia, A. Melnikov, O. Chubykalo-Fesenko, and U. Bovensiepen, Electron-and phonon-mediated ultrafast magnetization dynamics of Gd (0001), Phys. Rev. B 85, 184407 (2012).
- [10] B. Frietsch, J. Bowlan, R. Carley, M. Teichmann, S. Wienholdt, D. Hinzke, U. Nowak, K. Carva, P. M. Oppeneer, and M. Weinelt, Disparate ultrafast dynamics of itinerant and localized magnetic moments in gadolinium metal, Nat. Commun. 6, 8262 (2015).
- [11] A. J. Schellekens, W. Verhoeven, T. N. Vader, and B. Koopmans, Investigating the contribution of superdiffusive transport to ultrafast demagnetization of ferromagnetic thin films, Appl. Phys. Lett. **102**, 252408 (2013).
- [12] E. Turgut, C. La-o-vorakiat, J. M. Shaw, P. Grychtol, H. T. Nembach, D. Rudolf, R. Adam, M. Aeschlimann, C. M. Schneider, T. J. Silva, M. M. Murnane, H. C. Kapteyn, and

are expected to shown a much richer and exotic dynamics than ferromagnets in the presence of high magnetic fields. The LLB model for those materials in the limit of low magnetic fields already exists, therefore future works will address this issue.

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S. Mathias, Controlling the Competition Between Optically Induced Ultrafast Spin-Flip Scattering and Spin Transport in Magnetic Multilayers, Phys. Rev. Lett. **110**, 197201 (2013).

- [13] J. Mendil, P. Nieves, O. Chubykalo-Fesenko, J. Walowski, T. Santos, S. Pisana, and M. Münzenberg, Resolving the role of femtosecond heated electrons in ultrafast spin dynamics, Sci. Rep. 4, 3980 (2014).
- [14] P. Tengdin, W. You, C. Chen, X. Shi, D. Zusin, Y. Zhang, C. Gentry, A. Blonsky, M. Keller, P. M. Oppeneer, H. C. Kapteyn, Z. Tao, and M. M. Murnane, Critical behavior within 20 fs drives the out-of-equilibrium laser-induced magnetic phase transition in nickel, Sci. Adv. 4, eaap9744 (2018).
- [15] J. Becker, O. Mosendz, D. Weller, A. Kirilyuk, J. C. Maan, P. C. M. Christianen, Th. Rasing, and A. Kimel, Laser induced spin precession in highly anisotropic granular 110 FePt, Appl. Phys. Lett. **104**, 152412 (2014).
- [16] J. Becker, A. Tsukamoto, A. Kirilyuk, J. C. Maan, T. Rasing, P. C. M. Christianen, and A. V. Kimel, Ultrafast Magnetism of a Ferrimagnet Across the Spin-Flop Transition in High Magnetic Fields, Phys. Rev. Lett. 118, 117203 (2017).
- [17] M. Haag, C. Illg, and M. Fähnle, Influence of magnetic fields on spin-mixing in transition metals, Phys. Rev. B 90, 134410 (2014).
- [18] O. Eriksson, A. Bergman, J. Hellsvik, and L. Bergqvist, Atomistic Spin Dynamics: Foundations and Applications (Oxford University Press, Oxford, 2017).
- [19] D. A. Garanin, Fokker-Planck and Landau-Lifshitz-Bloch equations for classical ferromagnets, Phys. Rev. B 55, 3050 (1997).
- [20] U. Atxitia, D. Hinzke, and U. Nowak, Fundamentals and applications of the Landau–Lifshitz–Bloch equation, J. Phys. D 50, 033003 (2016).
- [21] D. Hinzke, U. Atxitia, K. Carva, P. Nieves, O. Chubykalo-Fesenko, P. M. Oppeneer, and U. Nowak, Multiscale modeling of ultrafast element-specific magnetization dynamics of ferromagnetic alloys, Phys. Rev. B 92, 054412 (2015).
- [22] C. Schieback, D. Hinzke, M. Kläui, U. Nowak, and P. Nielaba, Temperature dependence of the current-induced domain wall motion from a modified Landau-Lifshitz-Bloch equation, Phys. Rev. B 80, 214403 (2009).
- [23] A. J. Ramsay, P. E. Roy, J. A. Haigh, R. M. Otxoa, A. C. Irvine, T. Janda, R. P. Campion, B. L. Gallagher, and J. Wunderlich, Optical Spin-Transfer-Torque-Driven Domain-Wall Motion in a Ferromagnetic Semiconductor, Phys. Rev. Lett. **114**, 067202 (2015).

- [24] T. Janda, P. E. Roy, R. M. Otxoa, Z. Soban, A. Ramsay, A. C. Irvine, F. Trojanek, R. P. Campion, B. L. Gallagher, and P. Nemec, Inertial displacement of a domain wall excited by ultrashort circularly polarized laser pulses, Nat. Commun. 8, 15226 (2017).
- [25] K. M. Lebecki, D. Hinzke, U. Nowak, and O. Chubykalo-Fesenko, Key role of temperature in ferromagnetic bloch point simulations, Phys. Rev. B 86, 094409 (2012).
- [26] K. M. Lebecki and U. Nowak, Ferromagnetic vortex core switching at elevated temperatures, Phys. Rev. B 89, 014421 (2014).
- [27] D. Hinzke and U. Nowak, Domain Wall Motion by the Magnonic Spin Seebeck Effect, Phys. Rev. Lett. 107, 027205 (2011).
- [28] F. Schlickeiser, U. Ritzmann, D. Hinzke, and U. Nowak, Role of Entropy in Domain Wall Motion in Thermal Gradients, Phys. Rev. Lett. 113, 097201 (2014).
- [29] R. John, M. Berritta, D. Hinzke, C. Müller, T. Santos, H. Ulrichs, P. Nieves, J. Walowski, R. Mondal, O. Chubykalo-Fesenko, J. McCord, P. M. Oppeneer, U. Nowak, and M. Münzenberg, Magnetisation switching of FePt nanoparticle recording medium by femtosecond laser pulses, Sci. Rep. 7, 4114 (2017).

- [30] N. Bergeard, M. Hehn, S. Mangin, G. Lengaigne, F. Montaigne, M. L. M. Lalieu, B. Koopmans, and G. Malinowski, Hot-Electron-Induced Ultrafast Demagnetization in Co/Pt Multilayers, Phys. Rev. Lett. 117, 147203 (2016).
- [31] J. Kimling, J. Kimling, R. B. Wilson, B. Hebler, M. Albrecht, and D. G. Cahill, Ultrafast demagnetization of FePt: Cu thin films and the role of magnetic heat capacity, Phys. Rev. B 90, 224408 (2014).
- [32] C.-H. H. Lambert, S. Mangin, B. S. D. Ch S. Varaprasad, Y. K. Takahashi, M. Hehn, M. Cinchetti, G. Malinowski, K. Hono, Y. Fainman, M. Aeschlimann, and E. E. Fullerton, All-optical control of ferromagnetic thin films and nanostructures, Science 345, 1337 (2014).
- [33] U. Bovensiepen, Coherent and incoherent excitations of the Gd(0001) surface on ultrafast timescales, J. Phys.: Condens. Matter 19, 083201 (2007).
- [34] U. Atxitia and O. Chubykalo-Fesenko, Ultrafast magnetization dynamics rates within the Landau-Lifshitz-Bloch model, Phys. Rev. B 84, 144414 (2011).
- [35] N. Álvarez, G. Alejandro, J. Gómez, E. Goovaerts, and A. Butera, Relaxation dynamics of ferromagnetic FePt thin films in a broad frequency range, J. Phys. D 46, 505001 (2013).