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Ultrafast Spin Dynamics: The Effect of Colored Noise

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Recent experimental results have pushed the limits of magnetization dynamics to pico- and femtosecond time scales. This ultrafast dynamics occurs in extreme conditions of strong and rapid fields and high temperatures. This situation requires a new description of magnetization dynamics, taking into account that the electron correlation time could be of the order of the inverse spin frequency. For this case we introduce a thermodynamically correct phenomenological Landau-Lifshitz-Miyasaki-Seki approach. We demonstrate the effect of the noise correlation time on the ultrafast demagnetization rate.

One of the fundamental questions of modern solid state physics is how rapidly magnetization can respond to an external excitation. The recent development of time-resolved pump-probe experimental techniques using x-ray spectroscopy based on synchrotron radiation [1] and the Stanford linear accelerator (SLAC) [2,3] has allowed for the investigation of magnetization dynamics on the picosecond time scale. The use of powerful femtosecond lasers [4–6] has pushed this limit down to the femtosecond time scale. The physical processes underlying the response of the magnetization on this ultrashort time scale are complicated and far from being understood, but clearly involve the excitation and consequent nonequilibrium interaction of electron, phonon and spin subsystems. Spin dynamic processes on this time scale occur under extreme conditions remarkably different from those typical for dynamics at longer time scales. First, the three subsystems (electron, phonon, and spin) are not in equilibrium with each other. Second, spin dynamic processes occur under very strong fields with different sources. In particular, in the experiments using the SLAC the magnitude of the external field can be as large as 20 T. The strongly nonhomogeneous magnetization processes are driven by the exchange field, having a magnitude $\gtrsim$100 T. Finally, in the laser-induced magnetization dynamics the effective temperature is increased up to and often above the Curie temperature [4,5].

Atomistic spin models have proved to be a powerful approach to model ultrafast magnetization dynamics [7,8]. For example, in the case of laser-induced magnetization changes, spin models provide important physical insight into the spin-reordering process, establishing the linear character of the demagnetization during the subpicosecond regime and predicting the origin of different recovery rates in the picosecond regime. The basis of these models is the stochastic Landau-Lifshitz (LL) equation for each localized magnetic moment $\mathbf{s}_i$:

$$\dot{\mathbf{s}}_i = \gamma[\mathbf{s}_i \times \mathbf{H}_i] - \gamma\alpha[\mathbf{s}_i \times (\mathbf{s}_i \times \mathbf{H}_i)]. \quad (1)$$

Here $\mathbf{H}_i$ is the local effective field which includes Zeeman, exchange, anisotropy and magnetostatic contributions, augmented with a stochastic field $\xi_i(t)$ with the following properties for both components and different spin sites:

$$\langle \xi_i(t) \rangle = 0, \qquad \langle \xi_i(t)\xi_j(t') \rangle = \frac{2\alpha k_B T}{\gamma \mu_s} \delta(t - t')\delta_{ij}. \quad (2)$$

Here T is the temperature, γ is the gyromagnetic ratio, μ_s is the magnetic moment, and α is the parameter describing the coupling to the bath system. The basis of this equation is the separation of time scales, assuming that the heat bath (phonon or electron system) acts much faster than the spin system. In this case, the bath degrees of freedom can be averaged out and replaced by a stochastic field with white noise correlation functions. The coefficient in front of the delta function in Eq. (2) is determined by the fluctuation-dissipation theorem. The assumption of white noise is therefore invalid for magnetization dynamics occurring on a time scale comparable to the relaxation time of the electron system. The typical correlation time for the electron system in metals is of the order of 10 fs [1]. Such magnetization dynamics time scale is now commonly achieved by applying femtosecond laser pulses. A further limitation of this approach comes from the fact that characteristic frequencies of the magnetization process are now also of the order of the time scale of the noise (electron) variable. Therefore, for modelling the ultrafast magnetization experiments the approach Eq. (1) could break down.

The aim of the present Letter is to present a classical formalism beyond the white noise approximation to form a strong physical basis for models of ultrafast magnetization dynamics in extreme conditions. First, we introduce the formalism and show that our approach is consistent with the equilibrium Boltzmann distribution and coincides with the previous atomistic approach for small correlation times for the bath variable. As main implications of the correlated noise approach, we discuss the influence of noise correlations on the most relevant characteristics of magnetization dynamics: the longitudinal and transverse relaxation times. Finally, we model the laser-induced demagnetization rate for materials with different noise correlation times.

The standard generalization of the white noise to include correlations is the Ornstein-Uhlenbeck stochastic process [9]. However, on implementing this process within LL dynamics, we found, in agreement with previous work [10], that the Boltzmann equilibrium distribution is not recovered. The deviations invariably correspond to precessional frequencies of the order of the inverse correlation time. A suitable approach has been found in the work of Miyazaki and Seki [11] who generalized the Langevin equation for one spin to a non-Markovian case. The approach has been introduced for one spin at high temperatures, neglecting the interactions with other spins and assuming that their role is to provide the bath environment. In the present Letter we generalize this approach to a many spin case, similar to the standard way of Eq. (1) where the applied field is substituted by the local field. We assume that the bath variable is due to external sources such as electrons. Consequently, the applicability of the model is limited to the time scale where the notion of the quasistationary electron temperature is valid. The other assumption made in our approach is that the spin is connected locally to the bath. Consequently, the set of equations for magnetization dynamics [in the following called Landau-Lifshitz-Miyazaki-Seki (LLMS)] reads:

$$\dot{\mathbf{s}}_i = \gamma[\mathbf{s}_i \times (\mathbf{H}_i + \boldsymbol{\eta}_i)], \quad \dot{\boldsymbol{\eta}}_i = -\frac{1}{\tau_c}(\boldsymbol{\eta}_i - \chi \mathbf{s}_i) + \mathbf{R}_i, \quad (3)$$

with the fluctuation-dissipation theorem for the bath variable: $\langle \mathbf{R}_i(t) \rangle = 0$; $\langle \mathbf{R}_i(t)\mathbf{R}_j(t')\rangle = (2\chi k_B T/\tau_c)\delta_{ij}\delta(t - t')$. The parameter χ describes the coupling of the bath variable to the spin. The precession term in the first equation of the set (3) has the same form as in the Eq. (1). However, the damping is now described by the second equation in this set where also the bath variable adjusts to the direction of the spin due to the interaction with it. In the limit $\tau_c \to 0$ the stochastic LL Eq. (1) is recovered [11]. This also provides a relation between the damping and the coupling constants as $\alpha = \gamma\chi\tau_c$, giving a more precise physical sense to the LL damping constant at atomistic level.

First of all we investigated the equilibrium properties for an ensemble of noninteracting spins. In all cases of large fields, temperatures and correlation times, the correct Boltzmann distribution is obtained at equilibrium (see inset in Fig. 1).

Now we turn to the multispin system. First of all, we prove that the stochastic equations (3) for a multispin system are consistent with the standard equilibrium properties. For this purpose we use the formalism of the Onsager kinetic coefficient method applied in Ref. [12] for the LL multispin system (1). The system (3) is linearized near equilibrium and represented in a general form of the Langevin equation:

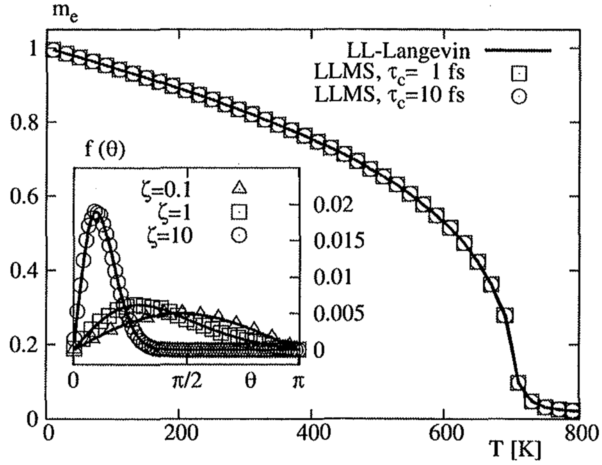

FIG. 1. Equilibrium magnetization as a function of temperature for a system of $\mathcal{N} = 32^3$ interacting spins, integrating the LLMS equation with different correlation times and integrating the LL equation. The inset shows distribution functions for a noninteracting spin system modeled within the LLMS approach for different values of the reduced field $\zeta = \mu_s H/k_B T$ ($\gamma H \tau_c = 1.76$) and correlation time $\tau_c = 10$ fs. The solid line in the inset represents the Boltzmann distribution.

$$\frac{dx_i}{dt} = -\sum_j \gamma_{ij} X_j + r_i \quad \langle r_i(t)\rangle = 0; \quad \langle r_i(0) r_j(t)\rangle = \mu_{ij}\delta(t). \quad (4)$$

Here x_i stands for small deviations of the stochastic variables $\mathbf{s}_i$ or $\boldsymbol{\eta}_i$ from their equilibrium values, X_i represent their thermodynamically conjugate variables and $\mu_{ij} = \gamma_{ij} + \gamma_{ji}$. For the spin variable we have: $X_j = -(\mu_s/k_B T)H_j$, where H_j is the internal field corresponding to a particular lattice site and spin component. Unlike Eq. (1), the first equation in (3) contains only a precessional term and, therefore, the corresponding kinetic coefficients are antisymmetric in spin components, giving for this equation $\mu_{ij} = 0$. Taking into account the generalization of the internal energy to include the bath variable as $F(\{\mathbf{s}_i\}, \{\boldsymbol{\eta}_i\}) = F_0(\{\mathbf{s}_i\}) + \sum_i [\boldsymbol{\eta}_i^2/(2\chi) - \boldsymbol{\eta}_i \mathbf{s}_i]$, where $F_0(\{\mathbf{s}_i\})$ is the internal energy without the bath variable, the conjugate variable to the bath one is $\mathbf{X}_j = (\boldsymbol{\eta}_j - \chi \mathbf{s}_j)/(k_B T \chi)$. Therefore, the corresponding matrix of the kinetic coefficients is diagonal and for the second equation we obtain $\mu_{ij} = (2k_B T\chi/\tau_c)\delta_{ij}$. Consequently, we have proven that the set of multispin equations (3) is consistent with the equilibrium properties.

In our simulations for the multispin system we use a Heisenberg Hamiltonian on a cubic system of 32^3 spins (with magnetic moment $\mu_s = 1.45\mu_B$) and with nearest-neighbor interactions only (exchange constant J). The Curie temperature is $T_c = 700$ K with $k_B T_c \approx 1.44J$. The coupling parameter χ was chosen to give the LL damping parameter $\alpha = 0.01$. In Fig. 1 we present calculations of the equilibrium magnetization as a function of temperature for spin systems with different values of the correlation times. Independence of the equilibrium properties on the correlation time, and the agreement with calculations using the LL equation with uncorrelated noise demonstrates our generalization of the LLMS equation to multispin systems. Consequently, the LLMS equation provides a basis for the phenomenological description of magnetization dynamics in extreme situations of high temperatures, and large and rapidly varying external fields. The advantage of the approach is also that the fluctuation-dissipation theorem is not applied directly to the spin variable. Therefore, the bath variable (for example electrons) and the spin system need not be in equilibrium with each other.

Next we discuss the most important implications of this new approach to ultrafast dynamics. It is known that during the excitation with spatially inhomogeneous fields in the Terahertz range [1,3] and also during laser-induced magnetization dynamics [4,5], strong local disordering of the spin system occurs. The dynamics in these cases is governed by field or temperature excited high-frequency spin waves which are responsible for the effective damping. The important dynamical feature then is the rate of magnetization recovery. During these processes two types of relaxation could be distinguished. The first one known as longitudinal relaxation is responsible for linear magnetization recovery, i.e., the magnetization magnitude. During the laser-induced demagnetization, the longitudinal relaxation is responsible for the femtosecond demagnetization. The second one is the transverse relaxation when the magnetization vector relaxes to the direction parallel to the internal field via magnetization precession. The longitudinal relaxation time increases with the temperature while the transverse relaxation time has minimum at T_c [13].

To simulate the longitudinal relaxation, we start with the initial condition $\{s_i^z\} = 1.0$ and observe the system to relax at given temperature T. The obtained relaxation curves are then fitted to exponential decay to extract the longitudinal relaxation rate. The longitudinal relaxation time, normalized to the uncorrelated case, is presented in Fig. 2 as a function of the noise correlation time. The longitudinal time calculated by means of the LL approach (1) is of the order of 10 fs ($\tau_\parallel^0 = 28$ fs at $T = 300$ K). For correlation time $\tau_c \lesssim 1$ fs the uncorrelated approach gives the same results as the LLMS one. However, one can see that $\tau_c \simeq 10$–100 fs gives a dramatic increase of the longitudinal relaxation time. The effect is less pronounced at higher temperature since in this case the temperature contributes to the loss of correlations.

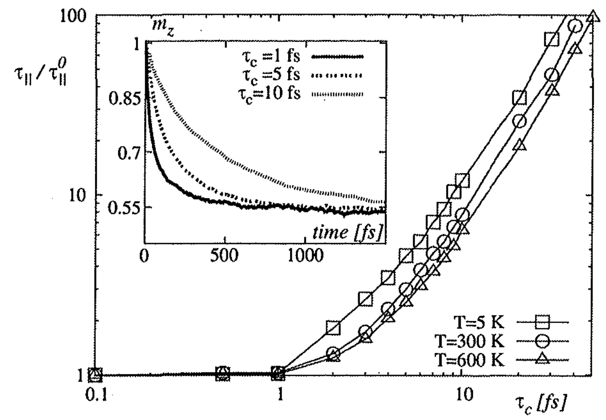

FIG. 2. Longitudinal relaxation time (normalized to the uncorrelated case) as a function of the correlation time for various temperatures calculated within the LLMS approach. The inset shows longitudinal relaxation for various correlation times and $T = 600$ K.

Next, we investigate the transverse relaxation in Fig. 3. The transverse relaxation time is defined by the magnetization precession and normally is much slower than the longitudinal one. For one spin $\tau_\perp = \tau_\perp^0[1 + (\omega_H \tau_c)^2]$ [11], where ω_H is the field-dependent precessional frequency. Consequently, the influence of the correlation time on the transverse relaxation may be expected only for the strong applied field for which $\omega_H \sim \tau_c^{-1}$ and, thus, could be relevant for SLAC experiments. To show the influence of the noise correlation on the transverse relaxation, we model the precessional dynamics at strong applied

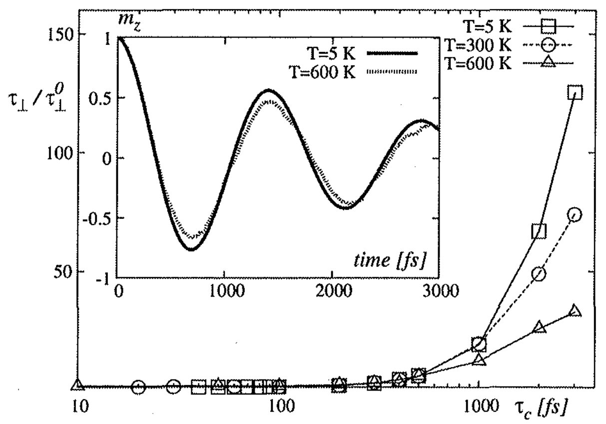

FIG. 3. Transverse relaxation time (normalized to the uncorrelated case) as a function of the correlation time for various temperatures calculated within the LLMS approach. The inset shows transverse relaxation for two temperatures $T = 5$ K and $T = 600$ K and $\tau_c = 10$ fs.

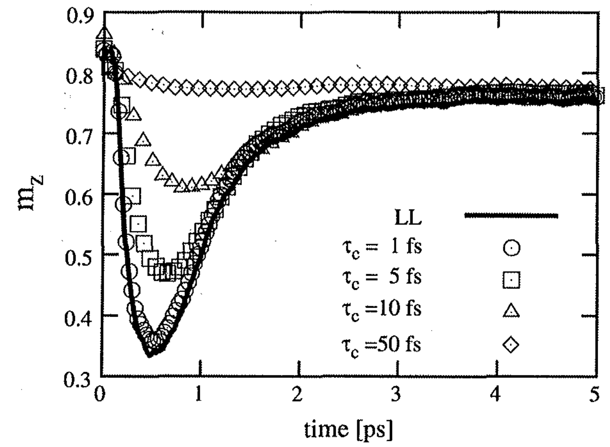

FIG. 4. Laser-induced demagnetization as a function of time for various noise correlation times τ_c modeled within LLMS and LL approaches.

field $H = 24.85$ T ($0.05J/\mu_s$). In this case, the spin system was first equilibrated at given temperature and applied field. After that the whole system was rotated to an angle 30° and the relaxation to the direction parallel to the applied field was observed. For this particular strong applied field, the correlation times $\tau_c \gtrsim 100$ fs are necessary in order to see their influence on the transverse relaxation.

To demonstrate the immediate physical consequence of the correlated noise for a more concrete experimental situation, we present in Fig. 4 the modelling results for the laser-induced demagnetization for various noise correlation times and constant damping parameter $\alpha = 0.05$. We suppose that the magnetization dynamics is produced by a Gaussian laser pulse with 50 fs duration and the fluency 33.7 $\mathrm{mJ/cm^2}$. Similar to Ref. [7], we assume that the photon energy is transferred to the electrons and lattice but the magnetization is directly coupled to the electron temperature T_e. The latter is calculated within the two-temperature model [14] with the electron and lattice specific heat constants $C_e = 700\ J/\mathrm{m^3\,K^2} \times T_e$, $C_l = 3 \times 10^6\ J/\mathrm{m^3\,K}$ and the coupling constant $G_{el} = 8 \times 10^{17}\ \mathrm{W/m^3\,K}$. Our results clearly show strong impact of the noise correlation time on the degree of demagnetization during the laser-induced process. Namely, materials with smaller correlation time τ_c demagnetized faster. This could be true, for example, for d electrons in metals with large scattering rate or for f electrons in rare earths which have strongly relativistic nature.

In conclusion, the standard phenomenological approach to model spin dynamics has been generalized to the non-Markovian case. This approach is necessary in the extreme situations of large characteristic magnon frequencies occurring during ultrafast magnetization processes. The advantages of the new approach are the following. (i) The memory (correlation) effects arising from the fact that the bath variable responds to the spin direction are taken into account. This corresponds to the situation when the bath variable is not in equilibrium with the spin system. (ii) The fluctuation-dissipation theorem is not applied to the spin systems as in the standard LL approach. (iii) At equilibrium the Boltzmann distribution is recovered. The price for this new approach is the use of two phenomenological constants: the phenomenological damping parameter α for the LL approach is substituted by two phenomenological parameters in the LLMS approach: the correlation time τ_c and the coupling constant χ. Several processes may be important in determining these constants, as for example, the spin-orbit coupling, momentum relaxation, scattering rate and dephasing time of conduction electrons. As in the LL approach, these parameters will be material specific and their physical origins should be clarified on the basis of first-principle approaches. We have shown that the ultrafast magnetization dynamics is strongly influenced by these parameters which stresses the necessity of first-principle models, capable to clarify their physical origins. Finally, we hope that in the future the direct comparison of experimental ultrafast demagnetization rates with the predictions of the present theory would allow us to extract the values of the correlation times in different materials.

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[1] J. Stöhr and H. C. Siegmann, *Magnetism* (Springer-Verlag, Berlin, 2006) and references therein.
[2] I. Tudosa *et al.*, Nature (London) **428**, 831 (2004).
[3] C. Stamm *et al.*, Phys. Rev. Lett. **94**, 197603 (2005).
[4] E. Beaurepaire *et al.*, Phys. Rev. Lett. **76**, 4250 (1996).
[5] B. Koopmans *et al.*, Phys. Rev. Lett. **85**, 844 (2000).
[6] J. Hohlfeld *et al.*, Phys. Rev. Lett. **78**, 4861 (1997).
[7] N. Kazantseva *et al.*, Europhys. Lett. **81**, 27 004 (2008).
[8] N. Kazantseva *et al.*, Phys. Rev. B **77**, 184428 (2008).
[9] J. M. Sancho and M. San Miguel, Phys. Rev. A **26**, 1589 (1982).
[10] H. Hasegawa, Physica A (Amsterdam) **387**, 1404 (2008).
[11] K. Miyazaki and K. Seki, J. Chem. Phys. **108**, 7052 (1998).
[12] O. A. Chubykalo *et al.*, J. Magn. Magn. Mater. **266**, 28 (2003).
[13] O. Chubykalo-Fesenko *et al.*, Phys. Rev. B **74**, 094436 (2006).
[14] G. Zhang *et al.*, in *Spin Dynamics in Confined Magnetic Structures I*, edited by B. Hillebrands and K. Ounadjela (Springer-Verlag, Berlin, 2002), p. 245.