Relaxation of Strongly Suppressed Modulus of Magnetization Following Ultrafast Demagnetization

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A longitudinal nonlinear relaxation of magnetization following the ultrafast demagnetization is considered. As demonstrated, the fastest regime occurs for a small deviation of the magnetization from the equilibrium value. However, the relaxation time of strongly suppressed magnetization is substantially increased due to a nonlinearity of magnetic system.

Рассмотрена продольная нелинейная релаксация намагниченности после сверхбыстрого размагничивания. Наиболее быстрый режим релаксации реализуется при небольших отклонениях намагниченности от своего равновесного состояния. Однако характерное время релаксации сильно замедленной системы существенно увеличивается за счет нелинейности магнитной системы.

Key words: ferromagnets, femtosecond laser pulse, ultrafast demagnetization, relaxation effects.

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1. INTRODUCTION

Intense femtosecond laser pulses are able to manipulate the magnetic order of condensed matter [1] on a time scale pertinent to the characteristic time of magnetization motion in the exchange field (picosec-
ond). This time is much shorter than a characteristic time of the standard transversal dynamics of magnetization (nanoseconds), which originates from magnetic interactions (Zeeman energy, magnetic anisotropy, demagnetizing field). The achieved laser induced demagnetization times are typically 100–300 fs for ferromagnets such as Ni [2]. Quite non-trivial behaviour, e.g., magnetization reversal, have been experimentally observed for broad variety of materials, including rare-earth and transition metals alloy [3–6] and some engineered magnetic materials, e.g., rare earth-free Co–Ir-based synthetic ferrimagnets [7].

Thus, the analysis of the picosecond, longitudinal evolution of the non-equilibrium distributions of magnetization with strong reduction of the magnetization has become increasingly important.

A longitudinal evolution of magnetization can be described using the Landau–Lifshitz equation with a relaxation term proposed by Bar’yakhtar [8–10] also called LLBar equations [11–13]. The LLBar equations are well suited for description of non-uniform states, like magnetic solitons [14, 15] and Bloch points [16], and give the explanation of the reversal effects [17, 18]. These equations provide an explanation of recent experiments on magnetization recovery in laser-pumped Ni–Ru–Fe heterostructures [19], where the importance of the nonlocal character of the magnetization recovery is established [13].

2. FORMULATION OF THE PROBLEM

The longitudinal relaxation of the homogeneous distribution of the magnetization with strong reduction of the magnetization following ultrafast demagnetization is considered.

The femtosecond laser excitation deposits energy directly into the 3d-electron system, subsequently generating mobile s–p-electrons [20, 21]. After electron thermalization that happens approximately in some 300 femtoseconds, highly nonequilibrium magnetization distribution appears. Since either the duration of a laser pulse used in the experiment and the thermalization time are significantly shorter than the characteristic time of the longitudinal evolution of the magnetization (of a few picoseconds), the analysis can be performed considering the evolution of the magnetization outside the time interval of the pulse duration. In this case, a strongly non-equilibrium state created by the laser pulse plays the role of the initial condition for the LLBar equations.

For our case, since we consider homogeneous distribution of the magnetization, the exchange term, which retains the total magnetization of a sample, can be disregarded (compare with [13, 17]), and the evolution of the magnetization is determined by relativistic processes. Then the LLBar equations take the form of the Landau–Lifshitz–Bloch equation [22]:

\[
\frac{\partial \mathbf{m}}{\partial t} = -\gamma \mathbf{m} \times \mathbf{H}_{\text{eff}} - \alpha \frac{\partial \mathbf{m}}{\partial T} + \mathbf{D},
\]

where \(\mathbf{m}\) is the magnetization, \(\gamma\) is the gyromagnetic ratio, \(\mathbf{H}_{\text{eff}}\) is the effective field, \(\alpha\) is the damping constant, and \(\mathbf{D}\) is a phenomenological term.
\[
\frac{\partial \mathbf{M}}{\partial t} = -\gamma [\mathbf{M} \times \mathbf{H}_{\text{eff}}] + \gamma \hat{\lambda}_r M_0 \mathbf{H}_{\text{eff}}
\] (1)

where \(\mathbf{M}\) is a magnetization of a ferromagnet, \(\gamma\) is the gyromagnetic ratio, \(\mathbf{H}_{\text{eff}} = -\partial F/\partial \mathbf{M}\) is an effective magnetic field, \(F\) is a free energy, \(\hat{\lambda}_r\) is a dimensionless relaxation tensor of the relativistic nature. Consideration of classical spins interacting with a thermal bath modelled by stochastic Langevin fields demonstrates that the Landau–Lifshitz–Bloch equation valid at all temperatures [22–24]. Note that the relaxation term \(\sim \hat{\lambda}_r \mathbf{H}_{\text{eff}}\) was first introduced by Bar'yakhtar [8] and gives the possibility to describe both the nonlinear, longitudinal relaxation of \(\mathbf{M}\), and the transversal relaxation of the magnetization. In the linear approximation nearby the equilibrium value of magnetization, this relaxation term can be split on the standard transversal Gilbert term and longitudinal term for the length of the magnetic moment [8].

Since, during the relaxation of the magnetization toward the equilibrium value, the field \(\mathbf{H}_{\text{eff}}\) is parallel to the magnetization, only the modulus of the magnetization \(M = |\mathbf{M}|\) enters the equation. We adopt the Landau model for the free energy
\[
F = \frac{1}{8\chi_0 M_0^2} \left( M^2 - M_0^2 \right)^2,
\] (2)

where \(M_0\) is the (temperature-dependent) equilibrium value of the magnetic moment of the bulk material, \(\chi_0 = dM/dH\) is a longitudinal magnetic susceptibility of a material in the equilibrium state and within the zero external magnetic field. Substituting the explicit form of the effective field
\[
H_{\text{eff}} = -\frac{(M^2 - M_0^2) M}{2\chi_0 M_0^2}
\] (3)

into Eq. (1), dividing Eq. (1) by \(M_0\), and introducing dimensionless variables, the nonlinear equation is derived for the evolution of \(M\):
\[
\frac{dm}{d\tau} = \frac{1}{2} m(1 - m^2)
\] (4)

where \(m(\tau) = M/M_0\) is a dimensionless magnetization, \(\tau\) is a dimensionless time measured in units of
\[
t_0 = \frac{\chi_0}{\gamma M_0 \hat{\lambda}_r}.
\] (5)

This formula shows that higher rate of a ‘stiffness’ of the spin system (smaller values of \(\chi_0\)) and higher rate of the coupling between the spin
system and the thermal bath (the value of $\lambda_r$) lead to a faster dynamics of the magnetization. Simple estimates give that, for nickel, the characteristic time $t_0$ of a longitudinal dynamics is of the order of a few picoseconds; see [25] for details. Note that the characteristic time $t_0$ is of relativistic origin but exchange enhanced ($\chi_d \approx 1/J$ where $J$ is an exchange integral) [8].

3. NONLINEAR RECOVERY OF MAGNETIZATION FOR DIFFERENT VALUES OF $m_0$

After the pulse action and the electron thermalization occur, the value of magnetization is reduced to $m(0) = m_0 < 1$. Then, the magnetization recovers to its equilibrium value $m = 1$. Integrating Eq. (4), the dependence of the magnetization on time within the demagnetized region can be presented as follows:

$$m(\tau) = m_0 / [m_0^2 + (1 - m_0^2) \exp(-\tau)]^{1/2}. \quad (6)$$

Figure 1 presents the time evolution of the relative change of the magnetization:

$$\Delta m(\tau)/\Delta m = (m(\tau) - m_0)/(m(\infty) - m_0) \quad (7)$$

Fig. 1. The time evolution of $\Delta m(\tau)/\Delta m$ derived from the solution of Eq. (4) for different values of $m_0$. Dashed lines are the approximations of $\Delta m(\tau)/\Delta m$ with $1 - \exp(-\tau/\tau_r)$ where corresponding values of $\tau_r$ are found from Fig. 2 ($\tau_r = 1.09$ for $m_0 = 0.9$ and $\tau_r = 1.8$ for $m_0 = 0.5$). The dotted line indicates the relaxation time $\tau_r$. 
for the following initial values of $m_0 = 0.9, 0.5, 10^{-1}, 10^{-2}, 10^{-4}$, where $m(\tau)$ is defined by Eq. (6). The value $m_0 = 0.9$ can be realized for weak intensity of the laser pulse; $m_0 = 0.5$ was realized in [2] and corresponds to the effective temperature of the spin system lower than the Curie temperature. Values of $m_0$ close to zero can be realized for strong intensity of the laser pulse, as has been done in [5—7]. Figure 2 presents the relaxation time $\tau_r$, defined as the time, for which the relative deviation of the magnetization from the equilibrium value decays by $e$ (Euler’s constant) times.

One can see that the fastest regime is realized for small deviation of the magnetization from the equilibrium value: $m_0 \to 0$. In the linear approximation on the small parameter $(1 - m_0)$, Eq. (6) can be reduced to

$$\frac{\Delta m(\tau)}{\Delta m} = 1 - \exp(-\tau/\tau_r)$$

(8)

where $\tau_r = 1$ (in units of $t_0$). With decreasing of $m_0$ (increasing of the power of the laser pump), the relaxation time increases. Figure 1 demonstrates that, for the initial values $m > 0.5$, the evolution of $\Delta m(\tau)/\Delta m$ can be also approximated by a simple exponential dependence (8), but with larger values of $\tau_r$ found from Fig. 2. For strong excitation of the spin system ($m < 0.5$), the evolution of magnetization should be describes by the strongly nonlinear dependence (6). In the vicinity of the unstable point $m_0 = 0$, the $H_{\text{eff}}$ approaches to zero, and the relaxation time diverges like $\tau_r \propto \ln(1/m_0)$. 

Fig. 2. The dependence of the relaxation time $\tau_r$ on the initial value of the magnetization $m_0$ in the demagnetized region ($\tau_r$ is the solution of $\Delta m(\tau_r)/\Delta m = 1 - 1/e$).
4. CONCLUSION

The relaxation time of highly nonequilibrium uniform states created by the femtosecond laser pulse within the relativistic relaxation approximation substantially depends on the initial value of the magnetization after the electron thermalization occurred in the demagnetized region. For small deviation of the magnetization from the equilibrium value (for small power of the laser pump), the relaxation time takes minimal value \( t_0 \). In this case, the longitudinal dynamics is described by the linearized Landau–Lifshitz–Bloch equation \[ \frac{\partial M}{\partial t} = \left( M_0 - M \right)/t_0 \] [26]. With increasing of the power of the laser pump, the relaxation time increases, and this process can no longer be described by linearized equations. For high power of the laser pump, when the magnetization in the demagnetized region is strongly suppressed, the relaxation time logarithmically diverges.

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