

Evolution of a demagnetized spin state toward equilibrium

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The temporal evolution of the nonequilibrium nearly homogeneous ferromagnetic state is analyzed within the general phenomenological spin-dynamics theory proposed by Baryakhtar, which employs the concepts of both local (relativistic) and nonlocal (exchange) spin relaxation. We demonstrate that strongly inhomogeneous spin dynamics can emerge on nanometer length scales. The mechanism responsible for this inhomogeneous dynamics is related to instabilities of the demagnetized spin state. These instabilities qualitatively modify the relaxation process toward equilibrium and may result in a dramatic underestimation of the characteristic time and amplitude of the spin dynamics. This finding can be exploited for the development of a new type of source for coherent spin-wave generation.

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I. INTRODUCTION

From the first discovery of sub-picosecond demagnetization [1] and the demonstration of magnetization reversal [2,3], the manipulation of spin systems of various magnets by ultrashort laser pulses has become an important topic of fundamental physics promising for applications [4–11]. The ability to generate and control spin waves in magnetic materials opened up exciting opportunities for spintronics or magnonic technologies. Among methods of magnon excitation we would like to mention optical means, where an ultrashort laser pulse pushes the magnetic system out of equilibrium [12–14]. In this case, the wave-vector distribution of the excited spin waves in reciprocal space is determined by the Fourier transform of the spatial intensity distribution of the pump spot.

Within the first (sub-picosecond) stage, a common behavior observed in many magnets is laser-induced demagnetization, whereas the follow-up evolution (picosecond relaxation of the magnetization vector $\mathbf{M}$ to the equilibrium) can be quite nontrivial. In the works where for a strongly focused spot of light it was possible to observe the real picture of the spatiotemporal evolution under the action of a solitary pulse, the authors reported on the development of strongly inhomogeneous states. Along with the propagation of spin waves far away from the light spot [12–14], the authors observed the formation of domains [15], ring structures localized near the spot [13], and the formation of phases with inverted spin orientation in the system of quasi-equilibrium magnons

of high density [16,17]. Ultrafast magnetization dynamics in new materials, such as van der Waals heterostructures, monolayer ferromagnets, kagome ferromagnets, are investigated [5,7,10,11]. Nonthermal pathways for ultrafast manipulation of ultrafast magnetization dynamics are explored [6,9].

In recent years, analyzing the dynamics of spatially inhomogeneous, nonequilibrium magnetization states has been of particular interest due to the development of experimental methods that pertain to the study of femtosecond spin dynamics, with a spatial resolution in the nanometer region [18–21]. The authors map quenched magnetic domains and localized spin structures in Co/Pd multilayers with a sub-wavelength spatial resolution down to 16 nm, and stroboscopically trace the local magnetization dynamics with 40 fs temporal resolution [20]. A counterintuitive, and therefore often overlooked, strongly inhomogeneous spin dynamics on spatial scales much smaller than the pump spot is discovered [21]. The strongly inhomogeneous magnetization oscillations originate from the different ultrafast dynamics of competing magnetocrystalline and shape anisotropies [21]. Very recently, femtosecond extreme ultraviolet microscopy with sub-wavelength 13.5-nm spatial resolution has been introduced, enabling direct real-space imaging of domains in ferri- and ferromagnetic thin films during laser-induced demagnetization [22].

In the theoretical description of the relaxation of weakly inhomogeneous nonequilibrium spin states toward equilibrium, local longitudinal relaxation is naturally dominant [1,23–25]. For strongly inhomogeneous nonequilibrium spin states, nonlocal longitudinal relaxation becomes important [3,26]. Homogeneous nonequilibrium spin states with external pumping demonstrate longitudinal instabilities, leading to formation of phases with inverted spin orientation [16,17]. The transversal dynamics is believed to be nonessential for this type of process.

Here, we examine theoretically the relaxation of strongly demagnetized spin states towards equilibrium within the

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Baryakhtar phenomenological spin-dynamics theory, accounting for both longitudinal and transversal dynamics as well as local and nonlocal relaxation mechanisms. We start with the investigation of the stability of the homogeneously demagnetized state. We find that nonlocal relaxation can lead to both longitudinal and, counterintuitively, transversal instabilities. We numerically analyze the nonlinear evolution of initially small perturbations. Within purely longitudinal dynamics, instead of a nearly homogeneous relaxation, the system can evolve via an intermediate domainlike state toward equilibrium. Transversal instabilities can qualitatively alter the relaxation scenario, giving rise to strongly inhomogeneous spin dynamics with characteristic length scales ranging from the order of a nanometer to several nanometers, which may be relevant for spin-wave generation.

II. PHENOMENOLOGICAL DESCRIPTION OF STRONGLY NONEQUILIBRIUM MAGNETIZATION DYNAMICS

The most direct way to describe ultrafast magnetization dynamics on the nanoscale is to use atomistic spin dynamics simulations on an atomic level [9,27]. However, this approach limits analysis capabilities and is associated with significant difficulties in computation. Thus, the development of a phenomenological approach suitable for describing ultrafast magnetization dynamics on the nanoscale is of great importance. The Landau-Lifshitz equation [28] with the Landau-Lifshitz (or equivalent Gilbert) relaxation term [28,29] preserves the magnetization length. This fact limits the applicability of the Landau-Lifshitz equation to the dynamics of the unit magnetization vector, as already noted in the original work [28]. The Landau-Lifshitz-Bloch equation [30–32] does not account for nonlocal relaxation processes [33]. The approach of Tserkovnyak *et al.* is efficient only within the linear approximation, i.e., for small deviations of the magnetization from its equilibrium value [34].

For a description of the ultrafast dynamics on the nanoscale the Landau-Lifshitz-Baryakhtar (LLBar) equation was proposed [26]. The LLBar equation explained an intriguing laser-induced magnetization dynamics of a Ni layer magnetization in a Ni-Ru-Fe heterostructure [26,35], where the nonlocal (exchange) relaxation turned out to be dominant. The LLBar equation explained [3] the switching effect (change in the direction of the sublattice magnetizations under the action of a laser pulse with a duration of less than 100 fs) in ferromagnetic alloy of rare-earth and transition metals GdFeCo [36]. The LLBar equation has also been used to describe the formation of new phase inclusions in systems of high-density quasiequilibrium magnons [16,17]. It was established long ago that the use of the LLBar equation is important in describing the relaxation of various magnetic solitons [37–43].

III. THE LANDAU-LIFSHITZ-BARYAKHTAR EQUATION

In 1984, Baryakhtar [44–46] proposed to use Onsager relations and general symmetry arguments for construction of the magnetization dynamic equations. This equation, referred to as the LLBar equation, operates with the magnetization (generalized coordinates) and the effective magnetic field

(generalized forces) and can be written as

$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma[\mathbf{M} \times \mathbf{H}_{\text{eff}}] + \gamma M_0 \hat{\alpha}_l \mathbf{H}_{\text{eff}} - a^2 \gamma M_0 \hat{\alpha}_{nl} \Delta \mathbf{H}_{\text{eff}}. \quad (1)$$

Here, γ is the gyromagnetic ratio, $\mathbf{H}_{\text{eff}} = -\delta\Phi/\delta\mathbf{M}$ is an effective magnetic field, Φ is a thermodynamic potential, $\mathbf{M}_0$ ($M_0 = |\mathbf{M}_0|$) is the temperature-dependent equilibrium value of the magnetization, a is the lattice constant, $\hat{\alpha}_l$ is a local (relativistic) relaxation tensor, $\hat{\alpha}_{nl}$ is a nonlocal (exchange) relaxation tensor. The LLBar equation comprises two distinct relaxation terms, one of relativistic origin and an exchange relaxation term. The local relaxation term $\propto \mathbf{H}_{\text{eff}}$ describes the local transfer of angular momentum to the thermal bath. The nonlocal relaxation term $\propto \Delta \mathbf{H}_{\text{eff}}$ can be interpreted as magnon diffusion (nonlinear spin current); such diffusion processes induce spatial redistribution of magnons. Since general principles (Onsager relations, exchange interaction symmetry, symmetry of the crystal lattice) that are valid for temperatures both below and above the Curie temperature were used to derive the LLBar equation, the equation proposed by Baryakhtar is also valid for all temperatures.

When longitudinal magnetic susceptibility and $\hat{\alpha}_{nl} = 0$ (the longitudinal and nonlocal relaxations are disregarded), the LLBar equation in the form (1) reduces to the Landau-Lifshitz equation with the standard Gilbert relaxation term. When $\hat{\alpha}_l$ is chosen in a way that the longitudinal evolution is determined by the longitudinal relaxation constant $\alpha_{\parallel}$ [30–32], whereas the transversal dissipative evolution is determined by the transversal local relaxation constant (the Gilbert damping), and neglecting the nonlocal relaxation term (for sufficiently smooth spatial changes to the magnetization), the LLBar equation reduces to the Landau-Lifshitz-Bloch equation [30]. In the linear approximation, close to the equilibrium value of magnetization, the local term $\hat{\alpha}_l \mathbf{H}_{\text{eff}}$ can be split into the standard transversal Gilbert term and longitudinal term for the length of the magnetic moment [44]. We note that the concepts of the exchange relaxation, introduced by Baryakhtar, and nonlocal relaxations (see Tserkovnyak *et al.* [34]) are in fact similar. In the linear approximation, according to how much the magnetization deviates from the equilibrium value, the LLBar equation reduces to the equations of Tserkovnyak *et al.* [34].

The LLBar equation has no alternatives for the extreme regimes of the magnetization dynamics, considered in this paper. Admittedly, the generality of the Bar'yakhtar equation has its price: the analysis becomes much more complicated. Thus, the Bar'yakhtar equation has been used when there were no simpler alternatives.

The LLBar equation contains nonlinear terms with $\nabla^2 \mathbf{M}$, as well as the term with $\nabla^4 \mathbf{M}$. Most of the rigorous mathematical methods, developed for the nonlinear differential equation of the second-order, cannot be extended to higher-order equations [47,48]. For example, the eigen-value problem are well-understood for the second-order equations (for example, in connection with quantum mechanical calculations), but are still far from complete understanding for the fourth-order equation. The fourth-order equation can be interpreted as two systems with the second-order equations. Interactions of these two systems can lead to much more rich physics, compared

with “simple” second-order systems. Numerical schemes that provide accurate approximations of both the magnetization vector and the effective magnetic field within the LLBar equation are investigated [49]. In fact, systematic research on the LLBar equation is just beginning.

IV. THE MODEL

The analysis of the previous section demonstrates that for the description of ultrafast, strongly nonlinear magnetization dynamics on the nanoscale within a phenomenological approach, the LLBar equation has no alternatives.

We consider a quasi one-dimensional evolution of the magnetization $\mathbf{M}(x, t)$ for our problem. Adopting the Landau model, the thermodynamic potential of the demagnetized ferromagnetic state can be presented in the form

$$\Phi = \frac{1}{8\chi_{\parallel}M_0^2}(|\mathbf{M}|^2 - M_0^2)^2 + \frac{A}{2}\left(\frac{\partial\mathbf{M}}{\partial x}\right)^2 - (\mathbf{H} \cdot \mathbf{M}), \quad (2)$$

where $\chi_{\parallel}$ is the static equilibrium value of the parallel susceptibility of the material (below the critical temperature $\chi_{\parallel} \ll 1$), A is an inhomogeneous exchange constant, and $\mathbf{H}$ is an external magnetic field. The first two terms in Eq. (2) have an exchange nature: the first term determines the energy of a homogeneous magnetization distribution, and the second term describes the energy of spatial inhomogeneity. Note that these two terms in Eq. (2) are isotropic. Surprisingly, due to the high symmetry of the exchange interaction, even weak anisotropic interactions (e. g., external magnetic field) may significantly modify evolution scenarios.

For simplicity, we assume that both local and nonlocal relaxation tensors are isotropic. In the following, we adopt the Gilbert damping constant as an estimate for the local relaxation tensor, $\hat{\alpha}_l = \alpha_G$. An estimate of the nonlocal relaxation constant α_{nl} can be obtained from Refs. [26,50]. In the experiment on ultrafast magnetization dynamics in Ni-Ru-Fe heterostructures, the ratio of the nonlocal and local relaxation constants is estimated as $\alpha_{nl}/\alpha_G \approx 200$ [26]. An estimation of α_{nl} can also be made from the experimental data on spin-wave resonance in confined permalloy samples, where the importance of the nonlocal contribution to the relaxation of magnon modes was established for magnetic nanoparticles of sizes up to 100 nm [50]. For the spin-wave length $\lambda_0 \approx 150$ a, the contribution from the nonlocal relaxation is about 20% of the local relaxation. Consequently, equating the local and nonlocal relaxation terms in the LLBar equation, the condition for the estimation of α_{nl} can be formulated as $0.2\alpha_G\mathbf{H}_{\text{eff}} = a^2\alpha_{nl}\partial^2\mathbf{H}_{\text{eff}}/\partial x^2$. Assuming $\mathbf{H}_{\text{eff}} \propto \exp(i(2\pi/\lambda_0)x)$, the estimation for the nonlocal relaxation constant is $\alpha_{nl}/\alpha_G = 0.2\lambda_0^2/(2\pi a)^2 \approx 114$, which is approximately two times smaller compared with the estimation from the experiment on ultrafast magnetization dynamics in Ni-Ru-Fe heterostructures [26].

Substituting the thermodynamic potential (2) into the LLBar equation, introducing dimensionless variables, one obtains the explicit form of an equation for the modeling of the magnetization dynamics

$$\frac{\partial\mathbf{m}}{\partial\tau} = -[\mathbf{m} \times \mathbf{h}_{\text{eff}}] + \alpha_G\mathbf{h}_{\text{eff}} - \alpha_{nl}\frac{\partial^2\mathbf{h}_{\text{eff}}}{\partial\xi^2}, \quad (3)$$

where the normalized effective field $\mathbf{h}_{\text{eff}} = \mathbf{H}_{\text{eff}}\gamma t_{\text{ex}}/a^2$ ($t_{\text{ex}} = a^2/\gamma AM_0$) is given by

$$\mathbf{h}_{\text{eff}} = \frac{1 - \mathbf{m}^2}{(\xi_0/a)^2}\mathbf{m} + \frac{\partial^2\mathbf{m}}{\partial\xi^2} + \mathbf{h}. \quad (4)$$

Here, the dimensionless coordinate ξ is measured in units of the lattice constant a , $\mathbf{m}$ is a normalized magnetization, $\mathbf{h} = \gamma t_{\text{ex}}\mathbf{H}$ is a normalized external magnetic field

$$\xi = \frac{x}{a}, \quad \tau = \frac{t}{t_{\text{ex}}}, \quad \mathbf{m} = \frac{\mathbf{M}}{M_0}, \quad \xi_0 = \sqrt{2\chi_{\parallel}A}. \quad (5)$$

The dimensionless time τ is measured in units of $t_{\text{ex}} = a^2/\gamma AM_0$. t_{ex} is the characteristic time scale of the fastest magnetic dynamics - a spin motion in an exchange effective field; for the typical magnetic materials t_{ex} is of the order of tens of femtoseconds. The value of ξ_0 , entering into the normalized effective field, for temperatures far from the Curie temperature is of the order of a lattice constant, but becomes larger in the vicinity of the critical temperature [26].

In the following, we assume that initially the system is in the equilibrium state with the normalized magnetization equal to its equilibrium value $|\mathbf{m}| = 1$. Then, due to, for example, laser impulse the value of the normalized magnetization is reduced. The rate of the demagnetization depends on the intensity of a laser pulse. In fact, an ideally homogeneous demagnetized state cannot be realized in practice. Small inhomogeneities always exist. Then, we consider the evolution of this state towards equilibrium by solving of the LLBar equation.

V. HOMOGENEOUS EVOLUTION SCENARIO

Here, we consider the longitudinal relaxation of the homogeneous distribution of the magnetization with a strong reduction of the magnetization following ultrafast demagnetization [26]. We suppose that the magnetization length is homogeneously suppressed to a value $m(0)$. Mathematically, this strongly nonequilibrium state plays the role of the initial condition for the LLBar equation. Since we consider homogeneous distribution of the magnetization, the nonlocal relaxation term can be disregarded and the evolution of the magnetization is driven by the local relaxation term $\alpha_G\mathbf{h}_{\text{eff}}$. During the relaxation of the magnetization toward the equilibrium value the effective field is parallel to the magnetization, therefore the precession term in the LLBar vanishes. This leads to the differential equation $\partial\mathbf{m}/\partial\tau = -\alpha_G(a/\xi_0)^2(1 - \mathbf{m}^2)\mathbf{m} + \mathbf{h}$ (here, $\mathbf{h}$ is oriented along $\mathbf{m}$), describing a nonlinear pendulum with a cubic nonlinearity. A nonzero $\mathbf{h}$ shifts the equilibrium value of the magnetization. However, the analysis shows that its influence in this case is negligible. Therefore, the solution for the magnetization length can be well-approximated by the solution with $\mathbf{h} = 0$:

$$m(\tau) = \frac{m(0)}{\sqrt{m^2(0) + (1 - m^2(0))\exp(-\tau/\tau_h)}}, \quad (6)$$

where $\tau_h = \xi_0^2/2a^2\alpha_G$ is a longitudinal equilibration time of a weakly $(1 - m(0)) \ll 1$ nonequilibrium homogeneous state and τ_h being of relativistic origin, but exchange enhanced [44]. The value of a longitudinal equilibration time $\tau_{ht_{\text{ex}}}$ is about $1/\alpha_G$ times slower than the fastest motion in magnetism t_{ex} ;

for typical parameters of transition ferromagnets $\tau_h t_{\text{ex}}$ is of the order of picoseconds, which corresponds to the microscopic theory based on the nonequilibrium thermodynamics of the magnon gas [23].

VI. INSTABILITIES OF THE DEMAGNETIZED STATE

Our ultimate aim is to understand how small deviations from the homogeneous state change the evolution compared to the homogeneous evolution scenario, considered in the previous section. In this section we investigate the stability of the homogeneously demagnetized state. We present the solution of the LLBar equation in the form

$$\mathbf{m}(\xi, \tau) = \mathbf{e}_z m_z(\xi, \tau) + \mathbf{e}_x m_x(\xi, \tau) + \mathbf{e}_y m_y(\xi, \tau), \quad (7)$$

where $m_z(\xi, \tau) = [m(\tau) + \Delta m_z(\xi, \tau)]$, $m(\tau)$ describes the homogeneous nonlinear evolution of the magnetization to the equilibrium state, considered in the previous section, and defined by Eq. (6). Here, the components m_x , m_y , Δm_z are deviations from the homogeneous evolution. Analyzing the stability, we assume that m_x , m_y , Δm_z are small. We present the solution for m_x , m_y , Δm_z in the form of plane waves

$$m_x(\xi, \tau) = A \cos(\omega\tau - k\xi) e^{\lambda_\perp \tau}, \quad (8)$$

$$m_y(\xi, \tau) = A \sin(\omega\tau - k\xi) \cdot e^{\lambda_\perp \tau}, \quad (9)$$

$$\Delta m_z(\xi, \tau) = B \cos(\kappa\xi) e^{\lambda_\parallel \tau}. \quad (10)$$

Here, A and B are initial amplitudes of the transversal and longitudinal deviations, k and κ are wave numbers of the transversal and longitudinal deviations, ω describes the dispersion law, $\lambda_\perp$, $\lambda_\parallel$ are the increments of transversal and longitudinal instability, respectively. Note that the phase velocity of the longitudinal perturbation is zero. The difference in the form of solutions for the longitudinal and transversal evolution reflects the fundamental difference between them: while the longitudinal evolution of magnetization can be only dissipative, the transversal one can also be nondissipative (includes some oscillations). Inserting Eqs. (8), (9), (10) into the LLBar equation, we find

$$\omega = m(\tau)(ka)^2, \quad (11)$$

$$\lambda_\perp = -\frac{1}{2\tau_h} \left\{ \left[\frac{\alpha_{\text{nl}}}{\alpha_G} (m^2(\tau) - 1) + \frac{\xi_0^2}{a^2} \right] (ka)^2 + \frac{\alpha_{\text{nl}}}{\alpha_G} \frac{\xi_0^2}{a^2} (ka)^4 \right\}, \quad (12)$$

$$\lambda_\parallel = -\frac{1}{2\tau_h} \left\{ (3m^2(\tau) - 1) + \left[\frac{\alpha_{\text{nl}}}{\alpha_G} (3m^2(\tau) - 1) + \frac{\xi_0^2}{a^2} \right] (\kappa a)^2 + \frac{\alpha_{\text{nl}}}{\alpha_G} \frac{\xi_0^2}{a^2} (\kappa a)^4 \right\}. \quad (13)$$

Let us analyze the instability increments of Eqs. (12), (13). Figures 1 and 2 show the instability increments for the transversal $\tau_h \cdot \lambda_\perp$ and longitudinal $\tau_h \cdot \lambda_\parallel$ perturbations for different demagnetized values $m(\tau)$, the realistic ratio of the nonlocal and local relaxation constants $\alpha_{\text{nl}}/\alpha_G = 100$ (see the estimation above) and $\xi_0 = a$. For comparison, the dashed lines show the case $\alpha_{\text{nl}} = 0$. We note the key role of the nonlocal relaxation; when the nonlocal relaxation vanishes ($\alpha_{\text{nl}} = 0$), $\lambda_\perp$ and $\lambda_\parallel$ are negative and both transversal and

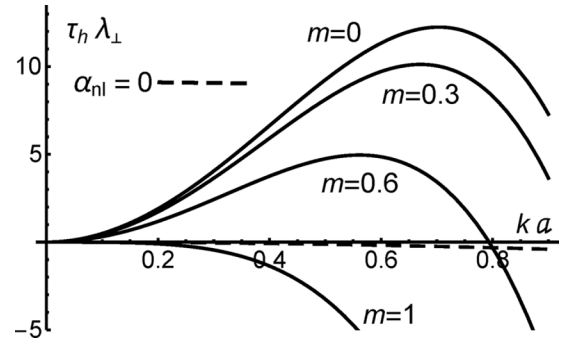


FIG. 1. The transversal instability increment $\tau_h \lambda_\perp$ for different values of $m(\tau) = 0, 0.3, 0.6, 1$, the ratio of the nonlocal and local relaxation constants $\alpha_{\text{nl}}/\alpha_G = 100$ and $\xi_0 = a$. The transversal perturbations for the chosen parameters become unstable for $m < \sqrt{1 - 10^{-4}}$. For comparison, the dashed line shows the case $\alpha_{\text{nl}} = 0$.

longitudinal perturbations decay. For realistic values of α_{nl} , the instability increments can become positive by the spin current. As the local relaxation changes the total magnetization of the sample, the local contribution to the instability increment of the longitudinal perturbations tends to a finite value for $k \rightarrow 0$, whereas the nonlocal contribution to the instability increment vanishes for $k \rightarrow 0$ (the nonlocal relaxation preserves the total magnetization of the sample).

When the system is in equilibrium ($m(\tau) = 1$), instability increments are negative. For transversal perturbations, Eq. (12) reduces to the standard expression for the damping increment of spin waves $-\lambda_\perp = (\alpha_G + \alpha_{\text{nl}}(ka)^2)\omega$, where the magnon frequency $\omega = (ka)^2$ [Eq. (11)]. The first term can be calculated within the Gilbert damping term from the standard Landau-Lifshitz equation and the second term is a specific nonlocal contribution to relaxation [24,34,41,44]. This asymptotic behavior is in accordance with the well-known result of the microscopic theory by Dyson [24]. For longitudinal perturbations, two additional damping terms, which are proportional to $1/\chi_\parallel$, appear $-\lambda_\parallel = (2a^2/\xi_0^2)(\alpha_G + \alpha_{\text{nl}}(ka)^2) + (\alpha_G + \alpha_{\text{nl}}k^2)\omega$ (note that $\xi_0^2 \propto \chi_\parallel$). In the limit of the homogeneous distribution ($k \rightarrow 0$), $-\lambda_\parallel \rightarrow 1/\tau_h$, which is in agreement with Eq. (6).

When the system is demagnetized ($m(\tau) < 1$), the nonequilibrium of the system comes into play. Equation (13)

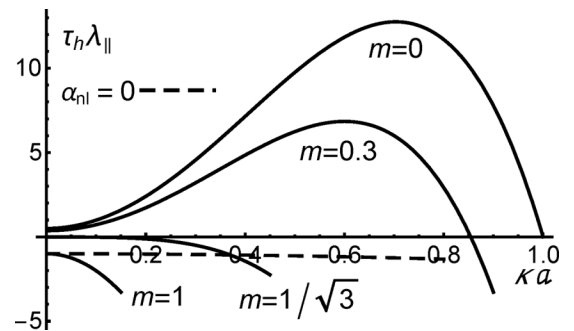


FIG. 2. The longitudinal instability increment $\tau_h \lambda_\parallel$ for different values of $m(\tau) = 0, 0.3, 1/\sqrt{3}, 1$, the ratio of the nonlocal and local relaxation constants $\alpha_{\text{nl}}/\alpha_G = 100$ and $\xi_0 = a$. For comparison, the dashed line shows the case $\alpha_{\text{nl}} = 0$.

shows that if the demagnetization rate exceeds some critical value, which depends on the nonlocal relaxation constant, the longitudinal perturbations become unstable. Similar longitudinal instabilities are founded within the Ginzburg-Landau theory of nonequilibrium relaxation, when a system is brought into a state far from thermal equilibrium by a sudden change of independent variables [51]. Longitudinal instabilities of the demagnetized spin state are also observed for the case of external pumping [16,17]. For transversal perturbations, when the coefficient in front of k^2 changes its sign to negative, $m^2(\tau) < 1 - \alpha_G \xi_0^2 / \alpha_{nl} a^2$, the transversal perturbations become unstable. Note a key role of the nonlocal relaxation in this unexpected finding, for $\alpha_{nl} = 0$, transversal perturbations always decay. In the following, we show that these instabilities (especially transversal) lead to a variety of qualitative effects in the field of ultrafast nanomagnetism.

VII. NONLINEAR ANALYSIS

To understand the role of the instabilities discussed in the previous section, we numerically analyze a further nonlinear evolution of initially small perturbations of the demagnetized state. We consider a system of size L . We chose the boundary conditions as follows: the first and second derivatives equal to zero. We present initial conditions for the LLBar equation in the form

$$m_x(\xi, 0) = A \exp\left[\frac{(\xi - L/2)^2}{\Delta^2}\right], m_y(\xi, 0) = 0, \quad (14)$$

$$m_z(\xi, 0) = m_0 + B \exp\left[\frac{(\xi - L/2)^2}{\Delta^2}\right], \quad (15)$$

where m_0 is a magnetization length value after the demagnetization process, A and B are the amplitudes of transversal and longitudinal inhomogeneities (located in the center of the sample), Δ is a characteristic size of these inhomogeneities. In fact, an ideally homogeneous state cannot be practically achieved, small inhomogeneities always exist. Since the initial phase of the transversal perturbation is not essential for demonstration purposes, for simplicity the y component of the transversal perturbation is set to zero. For our calculations, we use the following parameters: $L = 65$ (a nanoparticle with a size of about 20 nanometers), $\Delta = 7$, $m_0 = 0.1$ (the sample is demagnetized by 90%, which is consistent with experimentally accessible values [22]). For the Gilbert constant, a typical value for permalloy is taken $\alpha_G = 0.005$. For the nonlocal relaxation constant, which is key for instabilities, the minimum value from the above estimates is chosen $\alpha_{nl}/\alpha_G = 100$. In the numerical simulation, the external magnetic field $\mathbf{h}$ is oriented along the z axis. Typical values of $h_z = |\mathbf{h}| = 0.005$ (corresponding to approximately $|\mathbf{H}| \approx 1$ T), $h_z = 0.01$ ($|\mathbf{H}| \approx 2$ T), $h_z = 0.015$ ($|\mathbf{H}| \approx 3$ T), and $h_z = 0$ are used.

A. Purely longitudinal evolution

First, we consider a purely longitudinal inhomogeneity $A = 0$ and $B = -0.05$ for two limiting cases $\alpha_{nl} = 0$ and $\alpha_{nl}/\alpha_G = 100$, and $h_z = 0.005$. Figure 3 shows the evolution of this state when the nonlocal relaxation is neglected ($\alpha_{nl} = 0$) and $h_z = 0.005$. In this case, the evolution of the magnetization in each point of this distribution is driven by a local

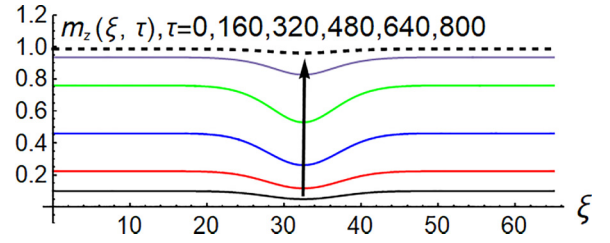


FIG. 3. The purely longitudinal evolution of $m_z(\xi, \tau)$ for $\alpha_G = 5 \times 10^{-3}$, $\alpha_{nl} = 0$, the amplitude of the longitudinal inhomogeneity $B = -0.05$, shown for successive times computed with a time step $\Delta\tau = 160$. Vertical arrow indicates the direction of time evolution. Note, equilibration time in the linear approximation for the homogeneous scenario $\tau_h = 1/\alpha_G = 200$. The plot is calculated for $h_z = 0.005$. The data for $h_z = 0, 0.01, 0.015$ are visually indistinguishable from those presented in this figure.

value of the effective field. The evolutions for $h_z = 0.005$ and $h_z = 0, 0.01, 0.015$ are practically indistinguishable on the plot. Therefore, this evolution can be described by Eq. (6) with a space-dependent value of $m(0)$.

Figures 4, 5 present the evolution of the same initial distribution but with a realistic value of $\alpha_{nl}/\alpha_G = 100$ for $h_z = 0.005$. When the nonlocal relaxation is taken into account, we see another scenario. This initial nonuniform deviation can be presented as a sum of harmonics with amplitudes determined via the Fourier transform. At the initial stage, these harmonics grow in accordance with the instability increments (12), (13). When the nonlinearity comes into play, the harmonics start to interact, only one harmonic wins, occupies a large portion of the region, which ultimately leads to the formation of a domainlike structure (see Fig. 4). The characteristic period of this domain structure can be determined by identifying the harmonic mode that exhibits the maximum instability. The period of this domainlike structure, determined from Fig. 4, is approximately $\Delta\xi \approx 14$. The wave length that maximizes the longitudinal instability increment for $m_0 = 0.1$, is about 11. Consequently, the period estimated

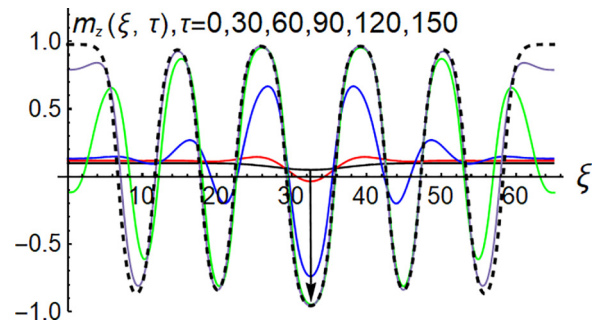


FIG. 4. The purely longitudinal evolution of $m_z(\xi, \tau)$ for the same initial distribution as in Fig. 3 (the amplitude of the longitudinal inhomogeneity $B = -0.05$), but with realistic nonlocal relaxation ($\alpha_G = 5 \times 10^{-3}$, $\alpha_{nl}/\alpha_G = 100$), shown for successive times computed with a time step $\Delta\tau = 30$. Vertical arrow indicates the direction of time evolution. The plot is calculated for $h_z = 0.005$. The data for $h_z = 0, 0.01, 0.015$ are nearly identical to those presented in this figure.

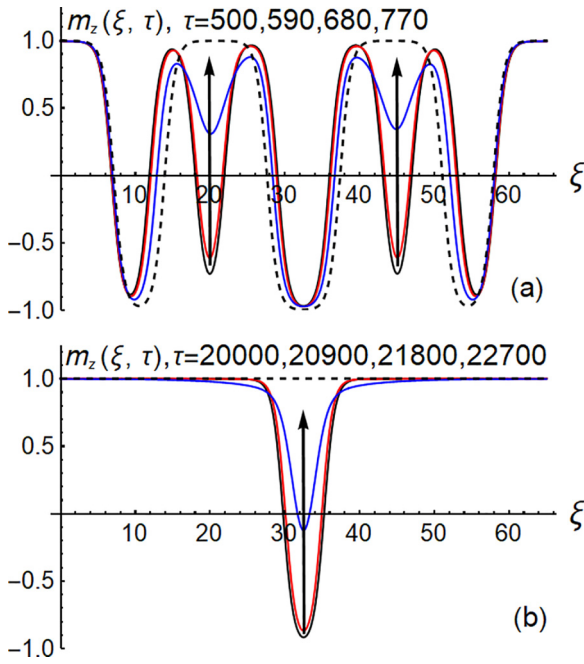


FIG. 5. The subsequent longitudinal evolution (a continuation of Fig. 4) of $m_z(\xi, \tau)$ for the same initial distribution as in Fig. 3 (the amplitude of the longitudinal inhomogeneity $B = -0.05$), but with realistic nonlocal relaxation ($\alpha_G = 5 \times 10^{-3}$, $\alpha_{nl}/\alpha_G = 100$). In contrast to the initial evolution presented in Fig. 4, the relaxation of the domainlike structure towards equilibrium is extremely sensitive to the external magnetic field. For $h_z = 0.005$, the relaxation of the domainlike structure takes about $\tau = 22\,500$. (a) The first stage of the increase of the period of the domainlike structures in the time interval from 500 up to 770 shown for successive times computed with a time step $\Delta\tau = 90$. (b) The final stage of the relaxation to the homogeneous state from 20 000 up to 22 770 shown for successive times computed with a time step $\Delta\tau = 900$. Vertical arrows indicate the direction of time evolution. For values $h_z = 0.01$, 0.015 , the scenario is similar. Yet, for $h_z = 0.01$, the equilibration occurs about two times faster $\tau \approx 1.14 \times 10^4$, for $h_z = 0.015$, $\tau \approx 0.8 \times 10^4$.

on the basis of the instability argument is approximately 20% smaller than that from Fig. 4. This underestimation can be associated with the fact that the maximum of the instability shifts to a longer wavelength as m_0 increases. This process of the domainlike structure formation is practically the same for $h_z = 0$, 0.01 , 0.015 and $h_z = 0.005$. Note that similar domainlike structures (but for systems with an external pumping) that originate from the longitudinal instabilities of the homogeneous solution are studied by Sugakov [16,17].

Later, this periodic domainlike structure very slowly (compared to the time of its formation) relaxes to the equilibrium state via an increase in its period. Contrary to the formation of the domainlike structure, this process is strongly affected by h_z . Figure 5 demonstrates this relaxation towards equilibrium for $h_z = 0.005$. From $\tau = 150$ to $\tau = 500$, this domainlike structure is practically unchanged. In the period from $\tau = 500$ to $\tau = 770$, first increase in the period occurs [Fig. 5(a)]. At $\tau \approx 1 \times 10^4$, the second increase of the period occurs (not shown). Figure 5(b) shows the final stage of the relaxation. Here, the last negative peak decreases its amplitude to zero and the system reaches the equilibrium.

Therefore, the process of the equilibration of the domainlike structure for $h_z = 0.005$ takes about $\tau \approx 2.2 \times 10^4$. For values $h_z = 0.01$, 0.015 , the scenario is similar. Yet, the equilibration time of the domainlike structure is strongly affected by h_z : for $h_z = 0.01$, the equilibration occurs about two times faster $\tau \approx 1.14 \times 10^4$; for $h_z = 0.015$, about three times faster $\tau \approx 0.8 \times 10^4$. For $h_z = 0$, our numerical simulations (not shown) demonstrate that equilibration generally proceeds via the similar scenario, through an increase in the period. However, the equilibration time depends on the boundary conditions and on the ratio of the system size to the period of the domainlike structure, and is at least an order of magnitude longer. Even at $\tau \approx 2.5 \times 10^5$, the negative peaks still persist.

Note that in the presence of external pumping, such a domainlike structure can be stable within a pure longitudinal picture (the transversal instabilities are not taken into account in these works) [16,17].

B. Longitudinal and transversal evolution

When the recovery of magnetization to equilibrium in strongly demagnetized systems is studied, the common opinion is that only the longitudinal evolution is relevant [3,16,17,26,33,35,51]. Here, we go beyond this seemingly natural assumption and consider both transverse and longitudinal evolution. We show that accounting for transversal evolution is important and can completely change the scenario of recovery to equilibrium.

For modeling, we use the same two cases for relaxation constants as in the previous section: $\alpha_{nl}/\alpha_G = 100$, $\alpha_G = 0.005$ and the case of negligible nonlocal relaxation $\alpha_{nl} = 0$, $\alpha_G = 0.005$. The longitudinal perturbation here is the same as in the previous section $B = -0.05$, but the amplitude of the transversal perturbation $A = 0.05$. Calculations were performed for $h_z = 0.005$.

Figures 6, 7, 8 show the evolution of these initial conditions. The nonuniform deviations at the initial stage grow according to the instability increments (12), (13) and occupy the entire sample. When the amplitudes of the spin-wave oscillations become sufficiently large, they start to interact with each other; but instead of practically stationary domainlike structures within a purely longitudinal evolution scenario, we observe strongly inhomogeneous, nonlinear longitudinal and transversal spin oscillations (Fig. 6). To complete the picture of this nonlinear dynamics, we additionally calculate the evolution of the magnetization modulus $|\mathbf{m}|$ (Fig. 8), using the data from Fig. 6. Comparison of Figs. 3 and 8 demonstrates that the nonlocal relaxation substantially enhances the relaxation of the magnetization modulus (with the nonlocal relaxation approximately 5 times faster). The physical picture of this phenomenon is as follows: due to instabilities caused by nonlocal relaxation, the motion of each component of the magnetization becomes faster, leading to more intense local relaxation. Figures 6 and 8 show that while the modulus of the magnetization almost reaches its equilibrium value (the magnetization modulus reaches a value of 0.9 at about $\tau \approx 150$), the components of the magnetization still strongly oscillate. Figure 7 demonstrates the subsequent evolution of the magnetization towards equilibrium. For $\tau = 5000$, we

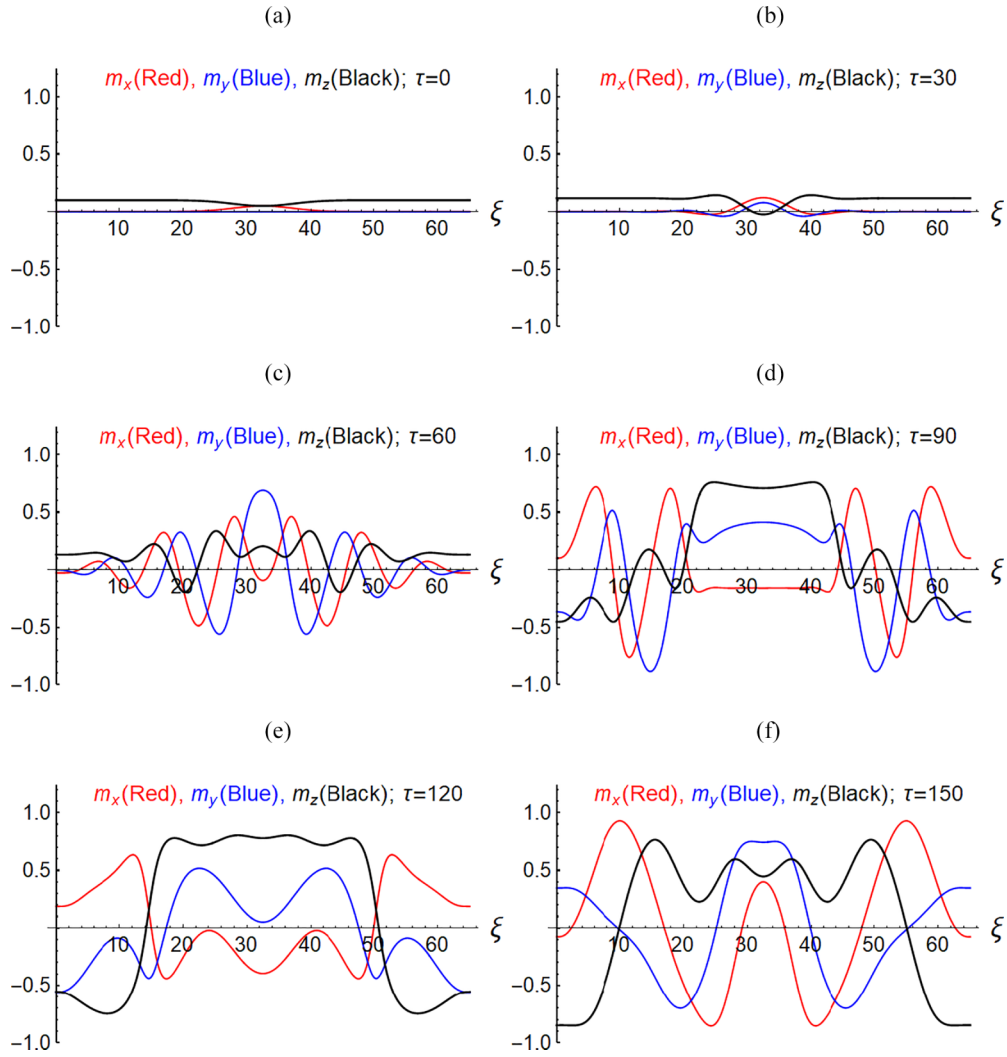


FIG. 6. Time evolutions of the magnetization m_x (red), m_y (blue), m_z (black) shown for successive times computed with a time step $\Delta\tau = 30$ [(a) $\tau = 0$, (b) $\tau = 30$, (c) $\tau = 60$, (d) $\tau = 90$, (e) $\tau = 120$, (f) $\tau = 150$], for $\alpha_{nl}/\alpha_G = 100$, $\alpha_G = 5 \times 10^{-3}$, and the amplitudes of transversal and longitudinal inhomogeneities $A = 0.05$, $B = -0.05$. The plot is calculated for $h_z = 0.005$. The scenario for $h_z = 0, 0.01, 0.015$ is similar. The evolution of the magnetization modulus $|\mathbf{m}(\xi, \tau)| = \sqrt{m_x^2 + m_y^2 + m_z^2}$ is presented in Fig. 8.

see that the m_z is separated by the symmetry of the system (a nonzero h_z) and has a value larger than m_x and m_y . For $\tau = 3 \times 10^4$, m_z almost reaches its equilibrium value 1, but the components m_x, m_y are still visible with the amplitudes of

the order of 0.1. The numerical simulation demonstrates that the scenario described above and the corresponding relaxation times are practically independent on the magnetic field (again, $h_z = 0.005, 0.01, 0.015$ and $h_z = 0$ were used).

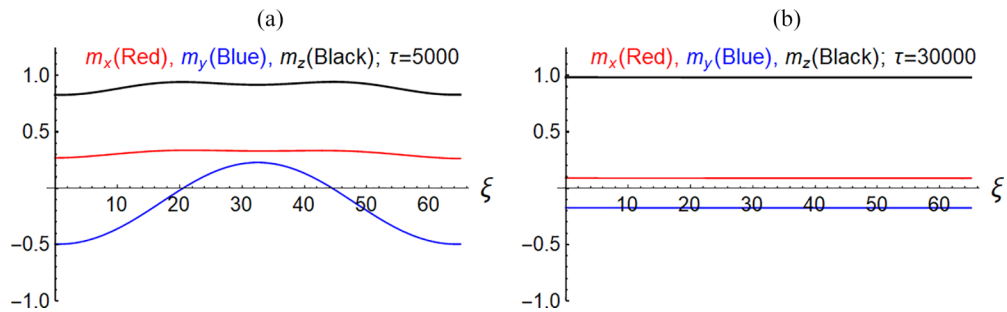


FIG. 7. Time evolutions of the magnetization m_x (red), m_y (blue), m_z (black) shown for (a) $\tau = 5000$, (b) $\tau = 30000$, $\alpha_{nl}/\alpha_G = 100$, $\alpha_G = 5 \times 10^{-3}$, and the amplitudes of transversal and longitudinal inhomogeneities $A = 0.05$, $B = -0.05$. The plot is calculated for $h_z = 0.005$. The magnetization modulus $|\mathbf{m}(\xi, \tau)| = \sqrt{m_x^2 + m_y^2 + m_z^2}$ for $\tau = 5000$ is presented in Fig. 8.

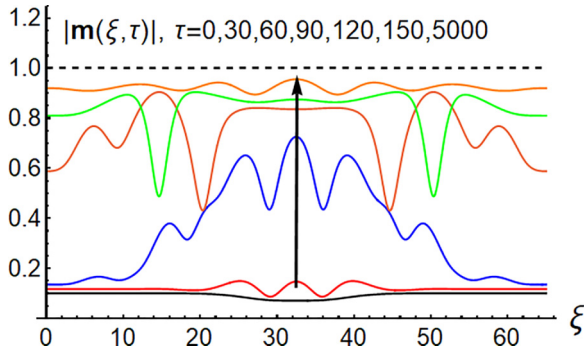


FIG. 8. Time evolutions of the magnetization modulus $|m(\xi, \tau)| = \sqrt{m_x^2 + m_y^2 + m_z^2}$, computed based on the data from Figs. 6, 7. Black corresponds to Fig. 6(a) ($\tau = 0$), Red—Fig. 6(b) ($\tau = 30$), Blue—Fig. 6(c) ($\tau = 60$), Wine—Fig. 6(d) ($\tau = 90$), Green—Fig. 6(e) ($\tau = 120$), Orange—Fig. 6(f) ($\tau = 150$), Dashed Black—Fig. 7(a) ($\tau = 5000$), respectively. The plot is calculated for $h_z = 0.005$. The data for $h_z = 0, 0.01, 0.015$ are visually indistinguishable from those presented in this figure. Vertical arrow indicates the direction of time evolution.

VIII. CONCLUSIONS

We have considered the relaxation of the strongly demagnetized spin state to equilibrium. It is commonly believed that only longitudinal relaxation is important in the equilibration process. We show that when the demagnetization rate exceeds a certain critical value, a realistic strength of nonlocal relaxation leads to both longitudinal and, curiously, transversal instabilities. These transversal instabilities completely change the scenario of magnetization relaxation towards equilibrium. Contrary to expectations, small transversal perturbations grow and strongly inhomogeneous nonlinear spin dynamics may emerge on spatial scales much smaller than the demagnetized region. In contrast to Ref. [21], where the strong inhomogeneity of spin dynamics is a result of anisotropic interactions, the energy in our case is isotropic.

The characteristic length scale of the inhomogeneities in our case ranges from the order of a nanometer to several nanometers. Very recently, femtosecond extreme ultraviolet microscopy with sub-wavelength 13.5-nm spatial resolution has been introduced [22], approaching the required resolution for our case. But access to femtosecond spin dynamics at nanometer scale (as in our case) has not yet been reported [22]. As noted in Ref. [21], strongly inhomogeneous spin dynamics even on spatial scales much larger than in our case (submicrons) is counterintuitive, and therefore often overlooked. Still, our predictions show that these instabilities also affect the evolution of the averaged magnetization dynamics (in our example, the magnetic moment of a particle of about 20-nm in size). Therefore, methods with spatial resolution of the order of 20 nm can be exploited (see Refs. [20,22] for short overview of experimental methods with such resolution); in this case, for a proper analysis, all components of the averaged magnetization vector must be measured simultaneously and the relaxation constants should be known.

Transversal instabilities prevent both purely longitudinal evolution and the formation of domainlike structures. However, the possibility of realizing such domainlike structures remains a subject for future investigation, possibly in highly anisotropic ferromagnets.

Figures 6 and 8 indicate the possibility of a new type of spin-wave generator based on transverse instabilities. If one ‘opens’ the boundaries that confine the spin oscillations within the particle, the spin waves begin to propagate outward. In spin-wave excitation methods [12–14], the distribution of the excited spin waves in wave-vector space is determined by the Fourier transform of the spatial intensity distribution of the pump spot. In our case, since the maximum instability rate depends on the demagnetization rate, the wave vectors of the generated spin waves can instead be tuned rather by the demagnetization rate.

DATA AVAILABILITY

The data that support the findings of this article are openly available [52].

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