Longitudinal magnetization reversal in ferromagnets with Heisenberg exchange and strong single-ion anisotropy

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We analyze theoretically the novel pathway of ultrafast spin dynamics for ferromagnets with high enough single-ion anisotropy. This longitudinal spin dynamics includes the coupled oscillations of the modulus of the magnetization together with the quadrupolar spin variables, which are expressed through quantum expectation values of operators bilinear on the spin components. Even for a simple single-element ferromagnet, such dynamics can lead to a magnetization reversal under the action of an ultrashort laser pulse.

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I. INTRODUCTION

Which is the fastest way to reverse the magnetization of either a magnetic particle or a small region of a magnetic film? This question has attracted significant interest, both fundamental and practical, for magnetic information storage.[1] Intense laser pulses, with durations less than a hundred femtoseconds, are able to excite the ultrafast evolution of the total spin of a magnetically ordered system on a picosecond time scale, see, e.g., the reviews.[2–4] The limitations for the time of magnetization reversal come from the characteristic features of the spin evolution for a magnet with a concrete type of magnetic order. The dynamical time cannot be shorter than the characteristic period of spin oscillations T, $T = 2\pi/\omega_0$, where ω_0 is the magnetic resonance frequency. For ferromagnets, the frequency of standard spin oscillations (precession) is $\omega_{0,\mathrm{FM}} = \gamma H_\mathrm{r}$, where γ is the gyromagnetic ratio, and H_r is an effective field of relativistic origin, like the anisotropy field, which is usually less than a few Tesla. Thus, the dynamical time for Heisenberg ferromagnets cannot be much shorter than 1 ns. For antiferromagnets, all the dynamical characteristics are exchange enhanced, and $\omega_{0,\mathrm{AFM}} = \gamma\sqrt{H_\mathrm{ex} H_\mathrm{r}}$, where H_ex is the exchange field, $H_\mathrm{ex} = J/2\mu_\mathrm{B}$, J is the exchange integral, and μ_B is the Bohr magneton, see Ref. 5. The excitation of terahertz spin oscillations has been experimentally demonstrated for transparent antiferromagnets using the inverse Faraday effect or the inverse Cotton-Mouton effect.[6–11] The nonlinear regimes of such dynamics include the inertia-driven dynamical reorientation of spins on a picosecond time scale, which was observed in orthoferrites.[10,11]

The exchange interaction is the strongest force in magnetism, and the exchange field H_ex can be as strong as 10^3 T. The modulus of the magnetization is determined by the exchange interaction, and the direction of the magnetization is governed by relativistic interactions. It would be very tempting to produce a magnetization reversal by changing the modulus of the magnetization vector, i.e., via the *longitudinal* dynamics of $\mathbf{M}$. For such a process, dictated by the exchange interaction, the characteristic times could be of the order of the exchange time $\tau_\mathrm{ex} = 1/\gamma H_\mathrm{ex}$, which is shorter than 1 ps. Nevertheless, within the standard approach such dynamics is impossible. The evolution of the modulus of the magnetization $M = |\mathbf{M}|$ within the closed Landau-Lifshitz equation for the magnetization only (or the set of such equations for the sublattice magnetizations $\mathbf{M}_\alpha$) is purely dissipative.[12] This feature could be explained as follows: Two angular variables θ and φ describing the direction of the vector $\mathbf{M}$ within the Landau-Lifshitz equation determine the pair of conjugated Hamilton variables ($\cos\theta$ and φ are the momentum and coordinate, respectively). Also, the evolution of the single remaining variable $M = |\mathbf{M}|$, governed by a first-order equation can be only dissipative; see a more detailed discussion below. Moreover, the exchange interaction conserves the total spin of the system, and the relaxation of the total magnetic moment of any magnet can be present only when accounting for relativistic effects. Thus the relaxation time for the total magnetic moment is relativistic but it is exchange enhanced, as was demonstrated within the irreversible thermodynamics of the magnon gas.[12] Note here that the relaxation of the magnetization of a *single sublattice* for multisublattice magnets can be of purely exchange origin.[13] Recently, magnetization reversal on a picosecond time scale has been experimentally demonstrated for the ferrimagnetic alloy GdFeCo, see Refs. 13 and 14. These results can be explained within the concept of exchange relaxation, developed by Baryakhtar,[15] accounting for the purely exchange evolution of the sublattice magnetization.[16] Such an exchange relaxation can be quite fast, but its characteristic time is again longer than the expected "exchange time" $\tau_\mathrm{ex} = 1/\gamma H_\mathrm{ex}$.

Thus, the ultrafast mechanisms of magnetization reversal implemented so far are: the dynamical (inertial) switching possible for antiferromagnets,[10,11] and the exchange longitudinal evolution for ferrimagnets.[13,14,16] These are both quite fast, with a characteristic time of the order of picoseconds; but their characteristic times are longer than the "ideal estimate": the exchange time τ_ex.

In this work we present a theoretical study of the possibility of the *dynamical* evolution of the modulus of the magnetization for ferromagnets with high enough single-ion anisotropy that can be called *longitudinal spin dynamics*. For such a spin evolution, a *dynamical* magnetization reversal is possible even for a simple single-element ferromagnet. Longitudinal dynamics does not exist in Heisenberg magnets, and this dynamics cannot be described in terms of the Landau-Lifshitz equation, or using the Heisenberg Hamiltonian, which is bilinear over the components of spin operators for different spins, see more details below in Sec. II. The key ingredient of our theory is the inclusion of higher-order spin quadrupole variables. It is known that for magnets with atomic spin $S > 1/2$, allowing the presence of single-ion anisotropy, the spin dynamics is not described by a closed equation for spin *dipolar* variable $\langle \mathbf{S} \rangle$ alone (or magnetization $\mathbf{M} = -2\mu_B \langle \mathbf{S} \rangle$).[17–24] Here and below $\langle \cdots \rangle$ means quantum and (at finite temperature) thermal averaging. To be specific, we choose the spin-one ferromagnet with single-ion anisotropy, the simplest system allowing this effect. The full description of these magnets requires taking into account the dynamics of *quadrupolar*variables $S_{ik} = (1/2)\langle S_i S_k + S_k S_i \rangle$ that represent the quantum averages of the operators, bilinear in the spin components. Such magnets are of significant interest, both from the viewpoint of fundamental science and for applications, as can be seen from the recent reviews.[25,26] Our theory is based on the consistent semiclassical description of a full set of spin quantum expectation values (dipolar and quadrupolar) for the spin-one system, which was investigated by many authors from different viewpoints.[17–24] As we will show, the longitudinal dynamics of spin, including nonlinear regimes, can be excited by a femtosecond laser pulse. With natural accounting for the dissipation, the longitudinal spin dynamics can lead to changing the sign of the total spin of the system (longitudinal magnetization reversal).

II. MODEL DESCRIPTION

The Landau-Lifshitz equation was proposed many years ago as a phenomenological equation, and it is widely used for the description of various properties of ferromagnets. Concerning its quantum and microscopic basis, it is worth noting that this equation naturally arises using the so-called *spin coherent states*.[27,28] These states can be introduced for any spin S as the state with the maximum value of the spin projection on an arbitrary axis $\mathbf{n}$. Such states can be parametrized by a unit vector $\mathbf{n}$; the direction of the latter coincides with the quantum mean values for the spin operator $\langle \mathbf{S} \rangle = S\mathbf{n}$ (dipolar variables). This property is quite convenient for linking the quantum physics of spins to a phenomenological Landau-Lifshitz equation. The use of spin coherent states is most efficient when the Hamiltonian of the system is linear with respect to the operators of the spin components. If an initial state is described by a certain spin coherent state, its quantum evolution will reduce to a variation of the parameters of the state (namely, the direction of the unit vector $\mathbf{n}$), which are described by the classical Landau-Lifshitz equation. Thus, spin coherent states are a convenient tool for the analysis of spin Hamiltonians containing only operators linear on the spin components or their products on different sites. An important example is the bilinear Heisenberg exchange interaction, described by the first term in Eq. (1) below.

In contrast to the cases above, for the full description of spin-S states, one needs to introduce SU$(2S+1)$ generalized coherent states.[21–24] The analysis shows that spin coherent states are less natural for the description of magnets whose Hamiltonian contains products of the spin component operators at a single site. Such terms are present for magnets with single-ion anisotropy or a biquadratic exchange interaction. For such magnets, some nontrivial features, absent for Heisenberg magnets, are known. Among them we note the possibility of so-called quantum spin reduction; namely the possibility to have the value of $|\langle \mathbf{S} \rangle|$ less than its nominal value $|\langle \mathbf{S} \rangle| < S$, even for pure states at zero temperature. This was mentioned by Moriya,[29] as early as 1960. As an extreme realization of the effect of quantum spin reduction, we note the existence of the so-called spin nematic phases with a zero mean value of the spin in the ground state at zero temperature. In the last two decades, the interest on such states has been considerable, motivated by studies of multicomponent Bose-Einstein condensates of atoms with nonzero spin.[30–33]

A significant manifestation of quantum spin reduction is the appearance of an additional branch of the spin oscillations, which is characterized by the dynamics (oscillations) of the length of the mean value of spin without spin precession.[17,19–24] The characteristic frequency of this mode can be quite high (of the order of the exchange integral). For this reason, for a description of resonance properties or thermodynamic behavior of magnets, this mode is usually neglected, and the common impression is that the dynamics of magnetic materials with constant single-ion anisotropy $K < (0.2–0.3)J$ is fully described by the standard phenomenological theory. However, for an ultrafast evolution of the spin system under a femtosecond laser pulse, one can expect a lively demonstration of this longitudinal high-frequency mode. Thus, it is important to explore the possible manifestations of the effects of quantum spin reduction in the dynamic properties of ferromagnets.

The simplest model allowing spin dynamics with effects of quantum spin reduction is described by the Hamiltonian

$$H = -\frac{1}{2}\sum_{\mathbf{n},\boldsymbol{\ell}} \bar{J}\mathbf{S}_{\mathbf{n}}\mathbf{S}_{\mathbf{n}+\boldsymbol{\ell}} + \frac{K}{2}\sum_{\mathbf{n}} (S_{\mathbf{n},x})^2, \tag{1}$$

where $\mathbf{S}_{\mathbf{n}}$ is the spin-one operator at the site $\mathbf{n}$; $\bar{J} > 0$ is the exchange constant for nearest-neighbors $\boldsymbol{\ell}$, and $K > 0$ is the constant of the easy-plane anisotropy with the plane yz as the easy plane. The quantization axis can be chosen parallel to the z axis and $\langle \mathbf{S} \rangle = \langle S_z \rangle \mathbf{e}_z$. For the full description of spin $S = 1$ states, let us introduce SU(3) coherent states[21–24]

$$|\mathbf{u},\mathbf{v}\rangle = \sum_{j=x,y,z} (u_j + i v_j)|\psi_j\rangle, \tag{2}$$

where the states $|\psi_j\rangle$ determine the Cartesian states for $S = 1$ and are expressed in terms of the ordinary states $\{|\pm 1\rangle, |0\rangle\}$ with given projections ± 1, 0 of the operator S_z by means of the relations $|\psi_x\rangle = (|-1\rangle - |+1\rangle)/\sqrt{2}$, $|\psi_y\rangle = i(|-1\rangle + |+1\rangle)/\sqrt{2}$, $|\psi_z\rangle = |0\rangle$, with the real vectors $\mathbf{u}$ and $\mathbf{v}$ subject to the constraints $\mathbf{u}^2 + \mathbf{v}^2 = 1$, $\mathbf{u}\cdot\mathbf{v} = 0$. All irreducible spin averages, which include the dipolar variable $\langle \mathbf{S} \rangle$ (average value of the spin) and quadrupole averages S_{ik}, bilinear over the spin components, can be written through $\mathbf{u}$ and $\mathbf{v}$ as follows:

$$\langle \mathbf{S} \rangle = 2(\mathbf{u} \times \mathbf{v}),$$
$$S_{ik} = \frac{1}{2}\langle S_i S_k + S_k S_i \rangle = \delta_{ik} - u_i u_k - v_i v_k. \tag{3}$$

At zero temperature and within the mean-field approximation, the spin dynamics is described by the Lagrangian[24]

$$L = -2\hbar \sum_n \mathbf{v}_n(\partial \mathbf{u}_n/\partial t) - W(\mathbf{u},\mathbf{v}), \tag{4}$$

where $W(\mathbf{u},\mathbf{v}) = \langle \mathbf{u},\mathbf{v}|H|\mathbf{u},\mathbf{v}\rangle$ is the energy of the system.

We are interested in spin oscillations which are uniform in space, and hence we assume that the discrete variables $\mathbf{u}$ and $\mathbf{v}$ have the same values for all spins and are only dependent on time. The frequency spectrum of linear excitations, which consists of two branches, can be easily obtained on the basis of the linearized version of the Lagrangian (4). In the general case, the system of independent equations for $\mathbf{u}$ and $\mathbf{v}$, taking into account the aforementioned constrains $\mathbf{u}^2 + \mathbf{v}^2 = 1, \mathbf{u}\cdot\mathbf{v} = 0$, consists of four nonlinear equations, describing two different regimes of spin dynamics. One regime is similar to that for an ordinary spin dynamics treated on the basis of the Landau-Lifshitz equation; it corresponds to oscillations of the spin direction. The second regime corresponds to oscillations of the modulus of the magnetization $\langle \mathbf{S} \rangle = S(t)\mathbf{e}_z$, with the vectors $\mathbf{u}$ and $\mathbf{v}$ rotating in the xy plane perpendicular to $\langle \mathbf{S} \rangle$. This mode of the spin oscillations corresponds to the longitudinal spin dynamics. It is convenient to consider these two types of dynamics separately. Particular nonlinear longitudinal solutions, with $\langle \mathbf{S} \rangle = s(t)\mathbf{e}_z$ and $u_z = 0,\ v_z = 0$, were found in Refs. 34 and 35. Note here that the longitudinal dynamics is much faster than the standard transversal one, and the standard spin precession (described by the Landau-Lifshitz equation) at a picosecond time scale just cannot develop. Therefore, these two regimes, longitudinal and transverse, can be treated independently, and we limit ourselves only to the longitudinal dynamics with $\langle \mathbf{S} \rangle = s(t)\mathbf{e}_z$ and $u_z = 0,\ v_z = 0$.

III. LONGITUDINAL SPIN DYNAMICS

To describe the longitudinal spin dynamics, it is convenient to introduce new variables: the spin modulus $s = 2|\mathbf{u}||\mathbf{v}| = 2uv$ and angular variable γ, with

$$\mathbf{u} = u(\mathbf{e}_x \cos\gamma - \mathbf{e}_y \sin\gamma), \quad \mathbf{v} = v(\mathbf{e}_x \sin\gamma + \mathbf{e}_y \cos\gamma). \tag{5}$$

In this representation $\langle S_z \rangle = s$, and the nontrivial quadrupolar variables are

$$\langle S_x S_y + S_y S_x \rangle = \sqrt{1-s^2}\sin 2\gamma,$$
$$\langle S_y^2 - S_x^2 \rangle = \sqrt{1-s^2}\cos 2\gamma, \tag{6}$$

with all other quantum averages being either zero (as the transverse spin components $\langle S_{x,y}\rangle$ or $S_{xz},\ S_{yz}$) or trivial, independent on s and γ, as $\langle S_z^2\rangle = 1$. The mean-field energy, written per one spin through the variables $s,\ \gamma$, takes the form

$$W(s,\gamma) = -\frac{J}{2}s^2 - \frac{K}{4}\sqrt{1-s^2}\cos 2\gamma, \tag{7}$$

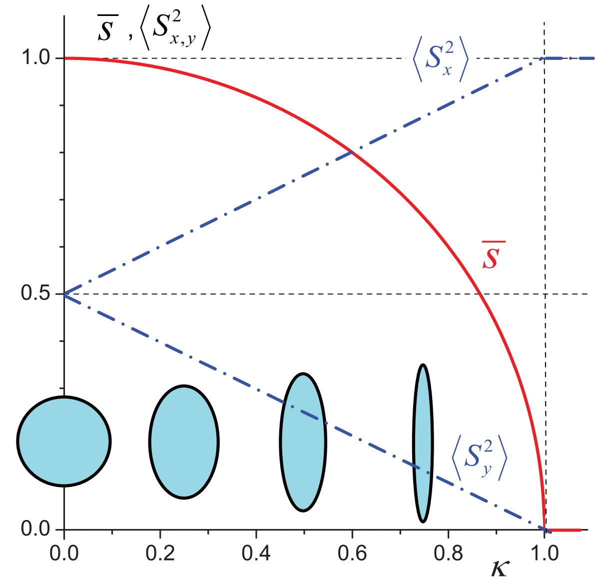


FIG. 1. (Color online) The ground state parameters $\langle S_x^2\rangle$ and $\langle S_y^2\rangle$ (blue dash-dot line) and $\bar{s} = \langle S_z\rangle$ (red solid line) as the function of κ. In the lower part of the figure the graphic presentation of the variables $\langle S_x^2\rangle$ and $\langle S_y^2\rangle$ showing the shape of the cross section of the quadrupolar ellipsoid in the xy plane for four values of κ; from left to right $\kappa = 0, 0.25, 0.5$, and 0.75.

where $J = \bar{J}Z$, Z is the number of nearest neighbors. The ground state at $\sqrt{1-s^2} > 0$ corresponds to $\cos 2\gamma = 1$, with the mean value of the spin $s = \pm\bar{s}$, $\bar{s} = \sqrt{1-\kappa^2} < 1$, that is a manifestation of quantum spin reduction at nonzero anisotropy. Here we introduce the dimensionless parameter $\kappa = K/4J$.

The values of the variables in the ground state are the following:

$$\langle {S_z}^2\rangle = 1,\ \langle {S_x}^2\rangle = \frac{1}{2}(1+\kappa),\ \langle {S_y}^2\rangle = \frac{1}{2}(1-\kappa). \tag{8}$$

The dependencies on κ for the ground state parameters $\langle S_x^2\rangle$, $\langle S_y^2\rangle$, and $\langle S_z\rangle$, together with the schematic graphic image of the xy cross section of the quadrupolar ellipsoid are shown in Fig. 1. The quadrupolar variables can be graphically illustrated by using a *quadrupolar ellipsoid*, which is a three-axial ellipsoid with the directions of the main axis (chosen to have $\langle S_1 S_2\rangle = 0$): $\mathbf{e}_3 = \mathbf{e}_z$ and $\mathbf{e}_1$, $\mathbf{e}_2$ and with the half-axes of the ellipsoid equal to $\langle S_1^2\rangle$, $\langle S_2^2\rangle$, and $\langle S_3^2\rangle = \langle S_z^2\rangle = 1$. This is a typical picture of ferro-quadrupolar ordering, known for many magnets,[25] and we are dealing with the coupled dynamics of spin dipolar variables and quadrupolar variables. For $\kappa \geqslant 1$, the state with zero magnetization (quadrupolar state), and with the values $\langle {S_z}^2\rangle = \langle {S_x}^2\rangle = 1$ and $\langle {S_y}^2\rangle = 0$, is realized.

For the variables s and γ, the Lagrangian can be written as

$$L = \hbar s \frac{\partial\gamma}{\partial t} - W(s,\gamma), \tag{9}$$

and $\hbar s$ and γ play the role of canonical momentum and coordinate, respectively, with the Hamilton function $W(s,\gamma)$. The physical meaning of the above formal definitions is quite clear: The angular variable γ describes the transformation of the quadrupolar variables under rotation around the z axis, with $\hbar s$ being the projection of the angular momentum on this axis. The Hamiltonian equations are

$$\hbar \frac{\partial \gamma}{\partial t} = \frac{\partial W(s,\gamma)}{\partial s}, \quad \hbar \frac{\partial s}{\partial t} = -\frac{\partial W(s,\gamma)}{\partial \gamma}. \tag{10}$$

The possible stationary solutions of the dynamical equations (10) ($s = \text{const}$, $\gamma = \text{const}$) are determined by the stationary points of the energy function, i.e., by the conditions $\partial W(s,\gamma)/\partial s = 0$ and $\partial W(s,\gamma)/\partial \gamma = 0$. The minimum of the energy corresponds to the ground state of the system. Note the presence of other stationary solutions, at $s = 1$, $\cos 2\gamma = 0$; and at $s = 0$, $\sin 2\gamma = 0$, which are unstable. These stationary points of the energy manifest themselves in the phase plane as saddle points and as singular phase trajectories at $s = 1$.

A. Small oscillations

Let us now start with the description of the dynamics of small-amplitude oscillations. After the linearization around the ground state, the equation leads to a simple formula for the frequency of longitudinal oscillations

$$\hbar\omega_l = 2J\bar{s} = 2J\sqrt{1-\kappa^2}, \tag{11}$$

which are in fact coupled oscillations of the projection of the spin and quadrupolar variables, see Fig. 2.

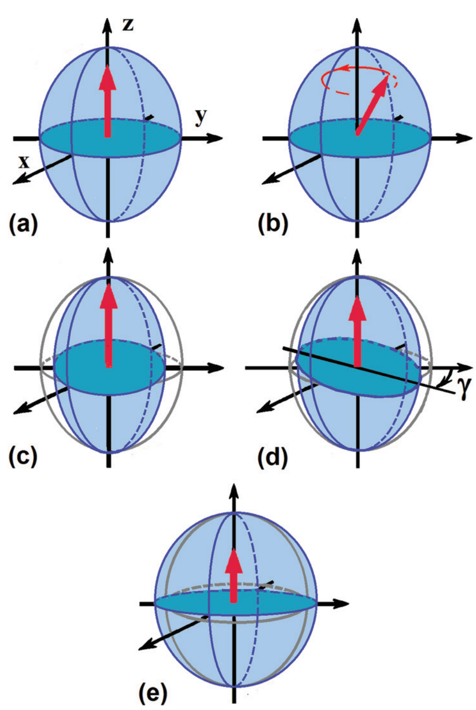


FIG. 2. (Color online) Graphic presentation of the variables s and γ and their evolution. The thick red arrow represents the mean value of the spin. The quadrupolar variables are shown by the quadrupolar ellipsoid, see the text, drawn in blue. (a) The ground state and (b) the standard transverse dynamics, i.e., the spin precession. (c)–(e) Transient values of the variables for longitudinal oscillations. (c) and (e) Correspond to the longest and shortest length of the spin, and at the moment depicted in (d) the spin length equals to its equilibrium value, but the quadrupolar ellipsoid is turned on the angle γ with respect to the x axis. (c)–(e) The shape of the unperturbed ellipsoid is shown in light gray.

One can see that, for a wide range of values of the anisotropy constant, like $\kappa < 0.2$–0.8, this frequency ω_l is of the order of $(1.8$–$1.2)J/\hbar$, i.e., ω_l is comparable to the exchange frequency $J/\hbar$. Thus the longitudinal spin dynamics is expected to be quite fast. In contrast, standard transversal oscillations for a purely easy-plane model (1) are gapless (they acquire a finite gap when accounting for a magnetic anisotropy in the easy plane, which is usually small). Thus the essential difference in the frequencies of these two dynamical regimes is clearly seen.

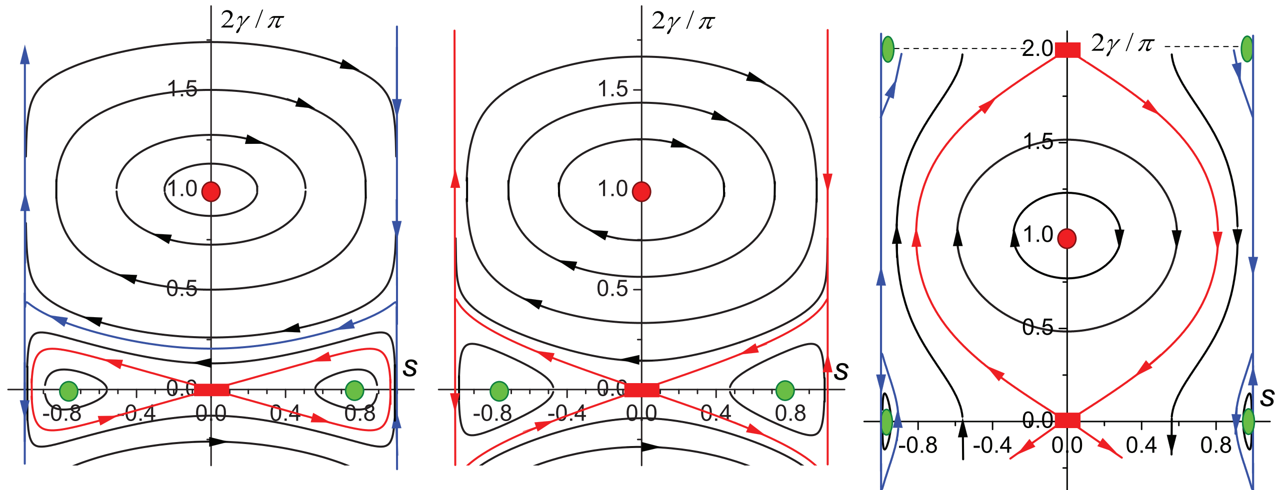
At a first glance, there is a contradiction between the concept of longitudinal dynamics caused by single-ion anisotropy and the result present in Eq. (11): The value of ω_l is still finite for vanishing anisotropy constant K; and it is even growing to the value $2J/\hbar$ when $\kappa \to 0$. This can be explained as follows: In the limit $\kappa \to 0$ the spin-quadrupole moment is simply zero according to the formula (6). Hence the free rotation of the quadrupole ellipsoid is a free rotation of a quantity of zero size, and the actual resonance does not exist in this limit. More formally, on a phase plane with coordinates (s,γ) this dynamics is depicted by vertical straight lines parallel to the γ axis, and the mean value of spin does not change, as can be seen by comparing Figs. 3(a) and 3(c) below. We will discuss this feature in more detail later with the analysis of nonlinear oscillations.

B. Nonlinear dynamics and phase plane analysis

Before considering damped oscillations, it is instructive to discuss dissipationless nonlinear longitudinal oscillations. It is convenient to present an image of the dynamics as a "phase portrait" on the plane momentum-coordinate (s, γ), which shows the behavior of the system for arbitrary initial conditions. The phase trajectories in the plane without dissipation can be found from the condition $W(s,\gamma) = \text{const}$.

The energy (7) has an infinite set of minima, with $s = \pm\bar{s}$ and $\gamma = \pi n$, with equal energies (green ellipses in Fig. 3), and an infinite set of maxima at $s = 0$ and $\gamma = \pi/2 + \pi n$ (red ellipses in Fig. 3), here n is an integer. Only the minima with $s = \bar{s}$ and $s = -\bar{s}$ are physically different; equivalent extremes with different values of n correspond to the equal values of the observables and are completely equivalent. The minima on the phase plane correspond to foci with two physically different equilibrium states with antiparallel orientation of spin $s = \pm\bar{s}$, and $\langle S_y^2 - S_x^2\rangle = \sqrt{1-\bar{s}^2}$, $\langle S_z^2\rangle = 1$, $\langle S_xS_y + S_yS_x\rangle = 0$. The saddle points are located at the values $s = 0$ and $\gamma = \pi n$. The lines with $s = \pm 1$ are singular; these correspond to degenerate motion with γ linear in time $\gamma = \pm tJ/\hbar$; the points at these lines where $d\gamma/dt$ change sign can be treated as nonstandard saddle points.

The shape of the phase trajectories, i.e., the characteristic features of oscillations, varies when changing the anisotropy parameter κ. Note first the general trend, the relative amplitude of the changes of the spin and γ depends on κ: The bigger κ is, the larger values of the change of spin are observed. The topology of the phase trajectories change at the critical value of the anisotropy parameter κ. At small $\kappa < 1/2$, the trajectories with infinite growing γ are present, and the standard separatrix trajectories connect together different saddle points, see Fig. 3(c). As mentioned above; only such trajectories are present at the limit $\kappa \to 0$, but, in fact, even for



(a) $\kappa = 0.6$ (b) $\kappa = \kappa_C = 0.5$ (c) $\kappa = 0.2$

FIG. 3. (Color online) Phase plane representations for dissipation-free nonlinear longitudinal spin oscillations for different values of the parameter κ; $\kappa = 0.6, 0.5$, and 0.2 for (a), (b), and (c), respectively. The green and red ellipses present the minima and maxima, respectively; the standard saddle points are depicted by red rectangles, while the standard separatrix trajectories are drawn by red lines. The singular trajectories with $s = \pm 1$ and the separatrix trajectories entering the nonstandard saddle points on these lines $s = \pm 1$ are shown by blue lines in (a) and (c). For the critical value $\kappa = 0.5$, all the separatrix trajectories and the singular trajectories with $s = \pm 1$ organize a common net; and on the corresponding frame (b) all of them are presented by red lines.

the small value of $\kappa = 0.2$ used in Fig. 2(c), the main part of the plane is occupied by the trajectories that change the spin. At the critical value $\kappa = 1/2$, the separatrix trajectories connect the saddle points at $s = 0$, $\gamma = 0$ and the degenerated saddle points at the singular lines with $s = \pm 1$. At larger anisotropy $\kappa > 1/2$, as in Fig. 3(a), the only separatrix loops entering the same saddle point are present.

The minima, saddle point, and the separatrix are the key ingredients for the switching between the ground states with $s = \bar{s}$ and $s = -\bar{s}$. The case of large anisotropy $\kappa > 1/2$ looks like the standard one for the switching phenomena, whereas for small anisotropy the situation is more complicated.

C. Damped longitudinal motion

Damping is a crucial ingredient for the dynamical switching between different, but equivalent in energy, states. The high-frequency mode of longitudinal oscillations have high-enough relative damping; as was found from microscopic calculations,[34] the decrement of the longitudinal mode $\Gamma = \lambda\omega_l$, where $\lambda \sim 0.2$. To account for the damping in the dynamic equations for s and γ, it is useful to consider a different parametrization of the longitudinal dynamics. Let us now introduce a unit vector $\boldsymbol{\sigma} = \sigma_1 \mathbf{e}_1 + \sigma_2 \mathbf{e}_2 + \sigma_3 \mathbf{e}_3$ with components

$$\sigma_3 = s,\ \sigma_1 = \langle S_y^2 - S_x^2 \rangle, \quad \sigma_2 = \langle S_1 S_2 + S_2 S_1 \rangle. \tag{12}$$

Being written through $\boldsymbol{\sigma}$, the equation of motion takes the form of the familiar Landau-Lifshitz equation

$$\hbar \frac{\partial \boldsymbol{\sigma}}{\partial t} = [\boldsymbol{\sigma} \times \mathbf{h}_{\text{eff}}] + \mathbf{R}, \quad \mathbf{h}_{\text{eff}} = -\frac{\partial W}{\partial \boldsymbol{\sigma}}, \tag{13}$$

where $\mathbf{h}_{\text{eff}}$ can be treated as an effective field for longitudinal dynamics, and the relaxation term $\mathbf{R}$ is added. The equation of motion with $\mathbf{R} = 0$ is fully equivalent to the Hamilton form of the equation found from (9), but the form of the dissipation is more straightforward in unit-vector presentation. The choice of the damping term in a standard equation for the motion of the transverse spin is still under debate.[15,36] But here the damping term can be written in the simplest form, as in the original paper of Landau and Lifshitz, $\mathbf{R} = \lambda[\mathbf{h}_{\text{eff}} - \boldsymbol{\sigma}(\mathbf{h}_{\text{eff}}\boldsymbol{\sigma})]$. The arguments are as follows: (i) this form gives the correct value of the decrement of linear oscillations $\Gamma = \lambda\omega_l$; and (ii) it is convenient for analysis, because it keeps the condition $\boldsymbol{\sigma}^2 = 1$. Finally, the equations of motion with the dissipation term of the aforementioned form are

$$\begin{aligned} \hbar \frac{ds}{dt} &= -\frac{\partial W}{\partial \gamma} - \lambda(1 - s^2)\frac{\partial W}{\partial s}, \\ \hbar \frac{d\gamma}{dt} &= \frac{\partial W}{\partial s} - \frac{\lambda}{(1 - s^2)}\frac{\partial W}{\partial \gamma}. \end{aligned} \tag{14}$$

These equations describe the damped counterpart of the nonlinear longitudinal oscillations discussed in the previous subsection and present as phase portraits in Fig. 3. The character of the motion at not-too-large λ can be qualitatively understood from energy arguments. The trajectories of damped oscillations in any point of the phase plane approximately follow the nondamped [described by equation $W(s,\gamma) = \text{const}$] ones, but cross them passing from larger to smaller values of W, see Figs. 4 and 6. It happens that for the case of interest, the dynamics is caused by the time-dependent stimulus. An action of the stimulus on the system can be described by adding the corresponding time-dependent interaction energy ΔW to the system Hamiltonian $W \to W(s,\gamma) + \Delta W(s,\gamma,t)$. Within this dynamical picture, ΔW produces an "external force" driving the system far from equilibrium.

The analysis is essentially simplified for a pulselike stimulus of a short duration Δt (much shorter than the period of motion $\omega_l \Delta t \ll 1$). In this case, the role of the pulse is reduced to the creation of some nonequilibrium state, which then evolves as some damped nonlinear oscillations described by the "free"

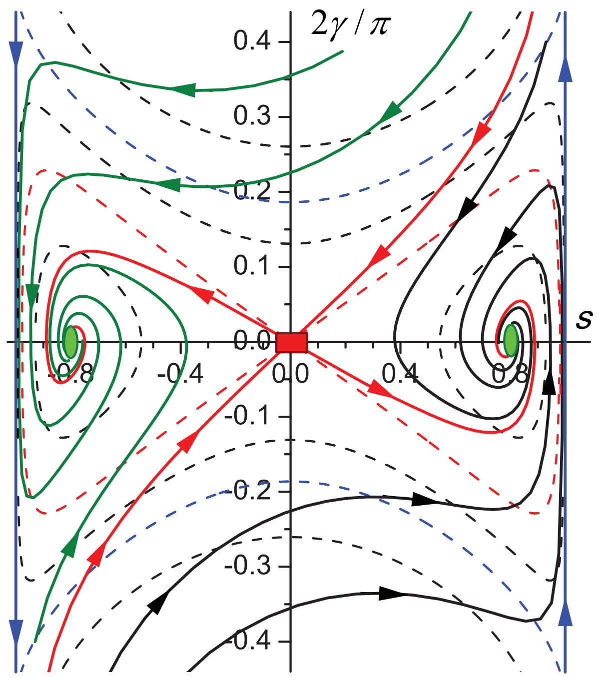


FIG. 4. (Color online) Phase plane representation of damped longitudinal spin oscillations for $\kappa = 0.6$. Here and in Fig. 6, the dashed lines (obtained analytically before) represent the phase trajectories without dissipation, while the full lines are trajectories for dissipation constant $\lambda = 0.2$, found numerically. The separatrix lines are drawn by red curves.

Eqs. (14) with $\Delta W = 0$. The phase plane method, which shows the behavior of the system for arbitrary initial conditions, is the best tool for the description of such an evolution.

First, let us start with the analysis for high-enough anisotropy. The corresponding phase portrait is present in Fig. 4. The general property of the phase plane is that the phase trajectories cannot cross each other; they can only merge at the saddle points. Thus the trajectories coming to different minima are stretched between two separatrix lines entering the same saddle point from different directions, as shown in Fig. 4. From this it follows that any initial state with arbitrary nonequilibrium values of spin $s(+0)$, but without deviation of γ from its equilibrium value, evolve to the state with the same sign of the spin as for $s(+0)$, and no switching occurs. However, if the initial condition is above the separatrix trajectory, entering the saddle point, the evolution will move the system to the equivalent minimum with the sign of the spin opposite to the initial one $s(+0)$, realizing the switching.

The switching can be realized for different initial conditions, which correspond to a high enough deviation from the initial state. The smaller deviation of s is present, the larger deviation of γ is necessary. For the case of high anisotropy $\kappa > \kappa_C = 1/2$, even if the initial value of s equals to its equilibrium value $s(0) = \bar{s} = \sqrt{1-\kappa^2}$, the switching can be observed, but for quite large values of $\gamma(0)$. However, the "minimal" deviation of the ground state corresponds to both $s(0) < \bar{s}$ and $\gamma(0) \neq 0$. Note that, except some short initial stage, $t < (2\text{–}4)t_{\text{ex}}$, the behavior of damped oscillations is very common for these two regimes. The time evolution for two characteristic initial conditions are shown in Fig. 5.

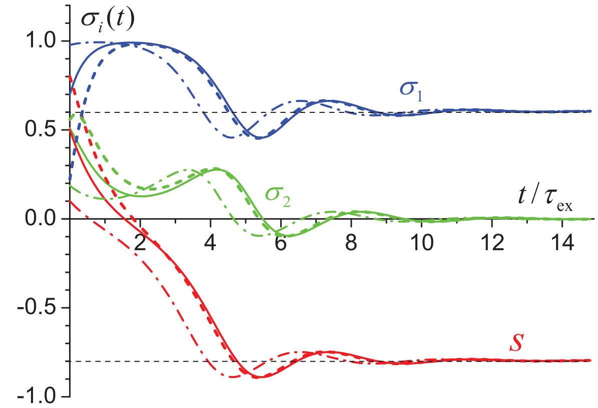


FIG. 5. (Color online) Time evolution of the spin s and quadrupolar variables $\sigma_{1,2}$, see Eq. (6), describing the switching for the magnet with $\kappa = 0.6$. The data for two characteristic initial conditions, corresponding, to relatively small (or large) initial deviation of s and relatively large and (or small) initial deviation of γ are present by full and dash-dot lines, respectively. For concrete calculations, the values $s(0) = 0.5$, $\gamma(0) = 0.1\pi$ and $s(0) = 0.1$, $\gamma(0) = 0.03\pi$ are used in these cases. The dash lines demonstrate the switching for the equilibrium initial value of spin $s(0) = \sqrt{1-\kappa^2} = 0.8$ and large enough $\gamma(0) = 0.19\pi$. The values of spin s (red lines) and $\sigma_1 = \langle S_y^2 - S_x^2 \rangle$ (blue lines) are going to their finite equilibrium values $-\sqrt{1-\kappa^2}$ and $\kappa = 0.6$, whereas $\sigma_2 \to 0$ (green lines).

Figure 6 shows the phase plane for Eqs. (14) for the more complicated case of low anisotropy $\kappa < \kappa_C = 0.5$, demonstrating several possible scenarios of the switching of the sign of the spin value during such dynamics. Note once more the qualitative difference from the case of small anisotropy: When the anisotropy constant is lower than the critical value $\kappa_C = 0.5$, the initial deviation of s is necessary for switching, which can be seen comparing Figs. 4 and 6. Here the separatrix trajectories for the damped motion can be monitored from their maxima, and the full picture of the behavior can be understood only when including a few equivalent foci with $\gamma = 0, \pm\pi, \pm 2\pi$, etc., with different, but equivalent in energy, values of the spin $s = \pm\bar{s}$, $\bar{s} = \sqrt{1-\kappa^2}$, and different saddle points, located at $\gamma = 0, \pm\pi, \pm 2\pi$. As for high anisotropy, the trajectories evolving to different minima are located between two branches of the separatrix lines, but now this "separatrix corridor" is organized by separatrix lines entering different saddle points. The switching phenomenon is also possible, but the process involves a few full turns of the variable γ.

The general rule for any anisotropy can be formulated as follows: For realizing spin switching, one needs to start from the states just above the separatrix line entering the saddle point from positive values of s. The larger is the deviation of the initial value of the spin from the equilibrium, the smaller value of $\gamma(0)$ would realize the switching. The asymptotic behavior of the separatrix trajectories at $\gamma, s \to 0$, is important for this analysis, and can be easily found analytically as

$$\left(\frac{\gamma}{s}\right)_{\text{separ}} = R_{\text{separ}} = \frac{1}{8\kappa}[\lambda(1+3\kappa) + \sqrt{\lambda^2(1+3\kappa)^2 + 16\kappa(1-\kappa)}]. \quad (15)$$

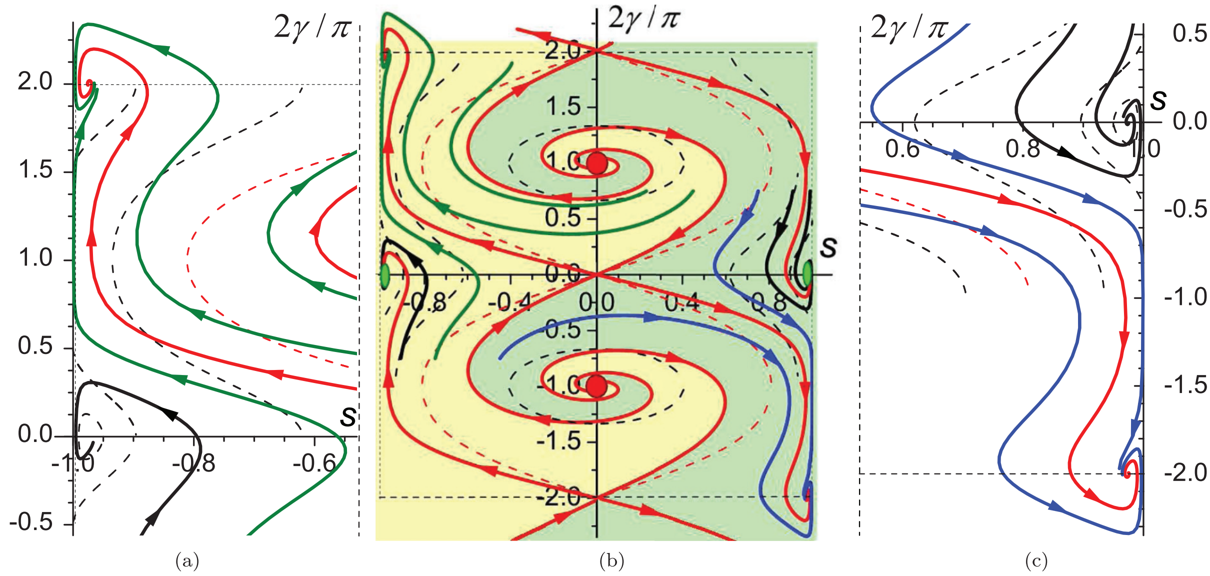


FIG. 6. (Color online) Phase plane for the damped spin evolutions for low anisotropy $\kappa = 0.2$. The central frame shows the full diagram; left and right panels demonstrate the details of the behavior near the equilibrium values $s = -\bar{s}$ and $s = \bar{s}$, respectively. On this frame, the regions colored by green and yellow correspond to different basins of attraction with initial values leading to the equilibrium states with $s = \bar{s}$ and $s = -\bar{s}$, respectively.

From the asymptotic equation (15), the corresponding ratio $R_{\text{separ}} = \gamma(0)/m(0)$ is smaller for small values of λ; but even when $\lambda \to 0$, it exceeds the value $R_{\text{separ}}(\lambda = 0) = 0.5\sqrt{(1-\kappa)/\kappa}$. However, for finite damping and small $\kappa \to 0$, the value of $R_{\text{separ}} = \lambda/4\kappa \to \infty$, when $\kappa \to 0$. The separatrix becomes almost vertical in this limit, and the reversal is basically prohibited. Thus, the switching could occur for nonzero values of $\kappa > \lambda \sim 0.2$.

IV. INTERACTION OF THE LIGHT PULSE ON THE SPIN SYSTEM: CREATION OF THE INITIAL STATE FOR SWITCHING

Let us now consider the longitudinal spin evolution caused by a specific stimulus: a femtosecond laser pulse. As mentioned above, the optimal initial state for the switching should include the deviation of the spin value, "spin quenching," and, simultaneously, a nonzero deviation of the quadrupolar variable γ. We now briefly discuss how both conditions could be realized.

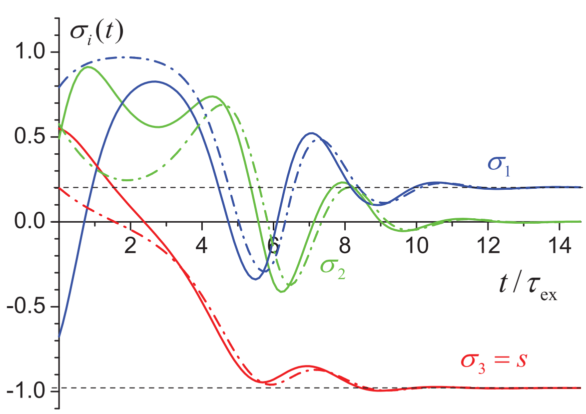


FIG. 7. (Color online) The same as in Fig. 5, but for small anisotropy $\kappa = 0.2$. Here the full and dash-dot lines show the behavior for minimal deviation $s(0) = 0.55$, with the large value of $\gamma(0) = 0.4\pi$ and the small values $s(0) = 0.2$, $\gamma(0) = 0.1\pi$, respectively. Note here much larger amplitude of the sign-alternating oscillations of σ_1 within the initial stage of evolution, that corresponds with the change of γ on π, see Fig. 6.

A. Spin quenching

In the literature, the laser-induced ultrafast quenching of the magnetic moment is mainly associated with transition metals or their alloys.[13,37–39] Moreover, the microscopic scenario of this effect is based on d-shell itinerant electrons.[40] (Note here the recent work[41] where a scenario for such a quenching has been proposed for rare-earth metals.) However, the theory developed here can be applied to different materials, either conducting or insulating. The quenching of the magnetic order is known for many nonmetallic compounds, insulators, and semiconductors. Note the observation of this effect for a number of spinel-type chromium chalcogenides,[42] either metallic or insulating, e.g., insulating ferrimagnetic $FeCr_2S_4$, $CoCr_2S_4$, and insulating ferromagnetic $CdCr_2S_4$. It has been mentioned that the quenching becomes faster for magnets with high anisotropy, which are interesting for our consideration. Femtosecond demagnetization has also been found for ferromagnetic semiconductors InMnAs and GaMnAs.[43,44] The large magneto-optic response, witnessing a drastic change of the order parameter within the first few hundreds of femtoseconds, has been observed for insulating antiferromagnets nickel oxide nickel NiO (Ref. 8) and Cr_2O_3.[45] A fast (over a time scale of 300 fs) and strong suppression of the magnetic order under the action of a femtosecond laser pulse has been found recently in copper oxide CuO.[46] Whereas the mechanisms of these effects is not as clear as for transition metals, their existence for different compounds is undoubtful. Note as well the recent observation of femtosecond suppression of the antiferromagnetic order in manganite $Pr_{0.7}Ca_{0.3}MnO_3$, exhibiting colossal magnetoresistance.[47]

B. Control of quadrupolar variables

The next nontrivial question is: How can we create a deviation of the quadrupolar variable γ from its equilibrium value? To find out how to do this, let us consider possible mechanisms of light interaction with quadrupolar variables of the magnets with nonsmall single-ion anisotropy.

The interaction of the spin system of magnetically ordered media and light is described by the Hamiltonian (as above, written per spin) $\Delta W = \bar{\varepsilon}_{ij} v_0 E_i(t) E_j^*(t)/16\pi$, where v_0 is the volume per spin, $E_i(t)$ is the time-dependent amplitude of the light in the pulse, $\bar{\varepsilon}_{ij} = d(\omega\varepsilon_{ij}^{(\mathrm{spin})})/d\omega$, $\varepsilon_{ij}^{(\mathrm{spin})}$ is the spin-dependent part of the dielectric permittivity tensor, and ω is the frequency of light. For the longitudinal dynamics considered here, circularly polarized light propagating along the z axis acts on the z component of the spin via the standard inverse Faraday effect, with the antisymmetric part of $\bar{\varepsilon}_{ij}^{(a)}$ as $\bar{\varepsilon}_{xy}^{(a)} = -\bar{\varepsilon}_{xy}^{(a)} = s\alpha_{\mathrm{F}}$, giving an interaction of the form

$$\Delta W_{\mathrm{circular}} = s\frac{\alpha_{\mathrm{F}} v_0}{16\pi}|E_{\mathrm{circ}}|^2\sigma, \tag{16}$$

where E_{circ} is the (complex) amplitude, and σ describes the pulse helicity: $\sigma = \pm 1$ for right-handed and left-handed circularly polarized laser pulses. To describe qualitatively the result of the action of the light pulse, let us now assume that the pulse duration τ_{pulse} is the shortest time of the problem. If the pulse duration is shorter than the period of spin oscillations, the real pulse shape can be replaced by the Dirac δ function $|E_{\mathrm{circ}}|^2 \to E_{\mathrm{p}}^2\tau_{\mathrm{pulse}}\delta(t)$, where $E_{\mathrm{p}}^2 = \int |E_{\mathrm{circ}}|^2 dt/\tau_{\mathrm{pulse}}$ characterizes the pulse intensity. (Note that this approximation is still qualitatively valid even for any comparable values of τ_{pulse} and $2\pi/\omega$.) Then, using Eqs. (14) one can find the effect produced by the pulse. Within this approximation, the action of a pulse leads to an instantaneous deviation of the variable γ from its equilibrium value, which then evolves following the nonperturbed equations of motion (14). Keeping in mind that before the pulse action the system is in equilibrium, $s(-0) = \bar{s}$ and $\gamma(-0) = 0$, it is straightforward to find the values of these variables (s and γ) after the action of the pulse $s(+0)$ and $\gamma(+0)$. For our purposes, the nonequilibrium value of the quadrupolar variable γ is important:

$$\gamma(+0) = -\frac{\alpha_{\mathrm{F}}}{16\pi\hbar}E_{\mathrm{p}}^2 v_0 \tau_{\mathrm{pulse}}\sigma. \tag{17}$$

The cumulative action of the circularly polarized pulse, including an essential reduction of the spin value (caused either by thermal or nonthermal mechanisms) and the deviation of γ described by (17) could lead to the evolution we are interested here, switching the spin of the system. Note that the effective field associated with the inverse Faraday effect for transparent insulating magnets reaches a few Tesla.[2] The theory of the ultrafast inverse Faraday effect for metals has been developed by Popova and coauthors[48] and the experiment shows huge values, up to 10 T for GdFeCo, see Fig. 2 of Ref. 49. Note here that, for standard spin reduction, the polarization of the light pulse is not essential,[37–39] whereas the values of $\gamma(+0)$ are opposite for right- and left-handed circularly polarized pulses. These features are characteristic of the effect described here. Note the recent experiment where the role of circular polarization in spin switching for GdFeCo alloy was mentioned, but the authors[49] have attributed it to magnetic circular dichroism.

V. CONCLUDING REMARKS

First, let us now compare the approach developed in this article with previous results on subpicosecond spin evolution. The first experimental observation of demagnetization for ferromagnetic metals under femtosecond laser pulses shows that the magnetic moment can be quenched very fast to small values, much faster than 1 ps.[37–39] These effects are associated with a new domain of the physics of magnets, *femtomagnetism*,[50] and its analysis is based on the microscopic consideration of spins of atomic electrons,[51,52] or itinerant electrons.[40] Not discussing this fairly promising and fruitful domain of magnetism, note that, to the best of our knowledge, no effects of magnetization reversal during this "femtomagnetic stage" has been reported in the literature (see, however, the recent article[47]). For example, the subpicosecond quenching processes for the ferromagnetic alloy GdFeCo are responsible for the creation of a far-from-equilibrium state, but the evolution of this state, giving the spin reversal, can be described within the standard set of equations for the sublattice magnetizations.[16]

In contrast, here we propose some pathway to switch the sign of the magnetic moment during extremely short times, of the order of the exchange time. It is shown here that the spin dynamics for magnets with nonsmall single-ion anisotropy can lead to the switching of the sign of the magnetic moment via the longitudinal evolution of the spin modulus together with quadrupolar variables (concretely, the quantum expectation values of the operators bilinear over the spin components S_x and S_y). It is worth to stress here that the "restoring force" for this dynamics is the *exchange interaction*, and the characteristic time is the exchange time. However, to realize this scenario, one needs to have a significant coupling of the spin dipole variable s and spin quadrupole variables. Obviously this effect is going beyond the standard picture of spin dynamics based on any closed set of equations for the spin dipolar variables alone. Note that our approach based on the full set of variables for the atomic spin is "more macroscopic" than the "femtomagnetic" approach,[40,51,52] dealing with electronic states.

We have found this type of switching within the concrete model (1) with spin one, Heisenberg exchange, and strong enough single-ion anisotropy. Such an anisotropy is known for the numerous magnets based on anisotropic ions of transition elements such as Ni^{2+}, Cr^{2+}, Fe^{2+}. Note the first spin-one system $CsNiF_3$ investigated thoroughly with respect to its nonlinear properties,[53] with the ratio of the anisotropy (estimated with quantum corrections) to exchange integral as high as 0.3. A better candidate is nickel fluosilicate hexahydrate $NiSiF_6{\cdot}6H_2O$, with spin-one Ni^{2+} ions, coupled by the isotropic ferromagnetic exchange interaction and subject to high single-ion anisotropy. For this compound, the strong effect of the quantum spin reduction is known, with its strength dependent on the pressure: The value of K/J is growing with the pressure P resulting in the value $\langle S\rangle = 0.6$ at $P = 6$ kbar and leading to the transition to the nonmagnetic state with $\langle S\rangle = 0$ at $P \sim 10$ kbar.[54,55] Other necessary conditions are the following: The possible materials should be very susceptible to magnetization quenching, as well as they should have a sizable Faraday effect. Unfortunately, we are not aware of the corresponding properties for the aforementioned materials.

A number of recent experiments were done with rare-earth transition-metals alloys.[13,14,49,56] Note here a rich variety of nonlinear spin dynamics observed for thin films of the FeTb alloy under the action of femtosecond laser pulses.[56] However, the theory developed here for a simple one-sublattice ferromagnet cannot be directly applied for the description of such compounds. Ferromagnetic order with high easy-plane anisotropy is present for many heavy rare-earth elements, such as Tb and Dy at low temperatures.[57] This feature is known both for bulk monocrystals,[57] and for thin layers and superlattices, see Ref. 58 and references wherein. Rare-earth metals have nonzero values of both spin and orbital momentum, forming the total angular momentum of the ion, and for their description the theory needs some modifications.

We conclude this article with some discussions of possible generalizations of the model and also pointing out some other magnets which probably could be used for realizing such kind of dynamics. Note first that our model neglects some physical interactions, for example, magnetoelastic coupling, which can be strong for magnets with high anisotropy. The detailed discussion of such effects is far beyond the scope of our article. Note only that many of the previous theories (either numerical or analytical) describing the laser-induced spin reorientation for GdFeCo alloys also do not consider this interaction,[13,14,16,41] whereas the rare-earth compounds are associated with giant magnetostriction.[57] In this work only spin-one ferromagnets with high single-ion anisotropy were considered. Nevertheless, we believe that the switching pathway presented here is more general than the concrete model discussed in this work

Going beyond the concrete model (1), let us stress that the main necessary conditions for the realization of this type of dynamics is the presence of both nonsaturated value of spin (quantum spin reduction) and significant values of nondipolar (quadrupolar) variables in the ground state, which are coupled. As a consequence of their coupling, the longitudinal dynamical mode with an exchange "restoring force" appears. All of the above features are characteristics of the so-called *ferromagnetic-quadrupolar states*, known for many rare-earth and actinide compounds with nonconventional magnetic order, see the review articles.[25,26] The cause of such states and modes cannot only be single-ion anisotropy, but non-Heisenberg exchange interaction as well. Such interactions include biquadratic exchange $(\vec{S}_1 \cdot \vec{S}_2)^2$, and for higher spins $S > 1$ the terms $(\vec{S}_1 \cdot \vec{S}_2)^n$, with $2 < n \leqslant 2S$ can be present as well. All these interactions could lead to spin reduction and the formation of ferromagnetic-quadrupolar states. They also couple the spin dipole and multipole variables, giving rise to longitudinal modes. The above features are known for either integer spin-two models,[59] or half-odd three-halves spin models.[20,33] Moreover, the Hamilton structure of the dynamical equations for these coupled variables is expected to be the same as for spin-one systems (10) [of course, the form of the Hamiltonian can differ from the one in Eq. (9)]. The reason is the following: From both classical and quantum mechanics it is known that the angular momentum and the angle of rotation around its direction are Hamilton-conjugated variables. Thus one can expect the possibility of the mode with coupled rotation of the quadrupolar ellipsoid around its principal axis, coinciding with the spin direction, and the oscillation of the spin value along this direction. Of course, this is only a qualitative argument. A detailed analysis of concrete models with spin $S > 1$ is of interest, but it is far beyond the scope of this article. The effects of spin switching caused by dipolar-quadrupolar longitudinal spin dynamics can be present for magnets with high values of the atomic angular momentum, which are in ferromagnetic-quadrupolar states. It is difficult to identify the best candidate for the realization of these effects of spin switching among the large variety of nonconventional magnets. A possible candidate could be the intriguing URu_2Si_2, which has been a puzzle for decades.[26] This material has a phase transition from a magnetic phase to a phase with so-called hidden order,[60] and a mode with longitudinal spin oscillations in the vicinity of this transition.[61]

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