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Ultrafast opto-magnetism

A M Kalashnikova, A V Kimel, R V Pisarev

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Contents

Abstract. In the last decade, a new area of research, referred to as femtomagnetism, has developed within the field of magnetism, which studies the excitation and control of magnetic medium dynamics on time scales comparable to or even much shorter than those of spin-lattice, spin-orbit, and exchange interactions. Among the many femtomagnetic processes studied to date, the opto-magnetic interaction of femtosecond laser pulses with media is of particular interest. This interaction is based on nondissipative Raman-type mechanisms and enables coherent spin dynamics to be efficiently and selectively excited and its parameters to be controlled. This review considers the key features of ultrafast opto-magnetic phenomena and how they relate to magneto-optical effects. A number of experimentally observed examples of ultrafast spin dynamics excited via opto-magnetic inverse Faraday and Cotton–Mouton effects are considered, and their microscopical nature is discussed. An experimental example is given demonstrating that combining ultrafast opto-magnetic phenomena with other laser-induced processes allows magnetization to be controlled on a picosecond time scale.

Keywords: ultrafast magnetic dynamics, femtosecond laser pulses, magnetically ordered dielectrics, magneto-optics

A M Kalashnikova, R V Pisarev Ioffe Physical-Technical Institute, Russian Academy of Sciences,
ul. Politekhnicheskaya 26, 194021 St. Petersburg, Russian Federation
E-mail: Kalashnikova@mail.ioffe.ru
A V Kimel Radboud University Nijmegen, Institute for Molecules and Materials, Heyendaalseweg 135, 6525 AJ Nijmegen, Netherlands;
Moscow State Technical University of Radio Engineering, Electronics and Automation,
prosp. Vernadskogo 78, 119454 Moscow, Russian Federation

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1. Introduction

In the last two decades, a new scientific field, called femtomagnetism, has been developed in the physics of the condensed matter and, in particular, in the physics of magnetically ordered media. This is the umbrella term for a broad class of phenomena that appear in magnetic media under the action of short (about 100 fs or less) and intense laser pulses and proceed on the time scale of less than characteristic spin–lattice relaxation time ~ 100 ps [1]. Femtomagnetism deals with nonequilibrium physical processes under conditions when the spin, the phonon, and, on shorter time scales, the electronic subsystems of the magnetically ordered media are not in thermodynamic equilibrium with each other.

The most detailed survey of work in the field of femtomagnetism is presented in [2]. The formation of this branch of physics of magnetic phenomena started from a number of studies in which it was shown that the action of a short powerful laser pulse drives a magnetically ordered medium into an excited state, in which the dynamics of the spin system and the subsequent relaxation differ significantly from well-known processes that have been examined earlier within the framework of classical thermodynamics. Specifically, the femtosecond laser pulses became the basic tool for studying the processes and mechanisms of demagnetization, magnetization emerging, and magnetization reversal in magnetic media, and also for developing methods of controlling ultrafast magnetic dynamics.

A large part of the phenomena that appear under the action of femtosecond laser pulses on magnetically ordered media are based on the absorption of the energy of the laser pulses. In the case of metals, the action of femtosecond pulses leads to a notable subpicosecond increase in the temperature of free electrons, which determines the subsequent dynamics both of the electron and of the phonon and spin subsystems. This ultrafast heating of an electron subsystem leads to

ultrafast demagnetization [3], i.e., to a decrease in the magnetization of the metal on timescales of $\gtrsim 100$ fs, depending on the efficiency of the interaction of free electrons and electrons that are responsible for the magnetization of the metal [4]. No strict microscopic theory of ultrafast laser-induced demagnetization has been developed so far, since it requires a description of the strongly nonequilibrium dynamics of the electron, phonon, and spin subsystems interacting with each other. In spite of the large number of unsolved problems, it is the ultrafast demagnetization that is the mechanism that made it possible for the first time to realize the all-optical switching of the magnetization in ferrimagnetic metals [5].

Another process that is based on absorption is the laser-induced excitation of coherent spin precession, which, in contrast to ultrafast demagnetization, is not limited to metals only. At present, several microscopic mechanisms of such excitation are known, which can be divided into thermal and nonthermal. Thermal mechanisms, as follows from the name, are those mechanisms that are based on the laser-induced increase in the effective temperature of one of the subsystems of the magnetic medium. Thus, ultrafast demagnetization in thin metallic films leads to a change in the demagnetizing fields; this can induce precession of the magnetization [6]. Furthermore, an increase in the effective temperature of the lattice can lead to a change in magnetocrystalline anisotropy, which, in turn, is an efficient mechanism for exciting spin precession both in metals [7] and in dielectrics [8]. The change in the magnetic anisotropy under the effect of laser pulses, which leads to the excitation of spin precession, can have a nonthermal nature as well. As examples of such processes, ultrafast photomagnetic effects in iron garnets can be mentioned [9–12], which consist in laser-induced change of the anisotropy as a result of charge transfer transitions between the ions which occupy different crystallographic positions and have different valences. This redistribution of the charge density can substantially change the magnetic anisotropy of the medium [13]. In contrast to the thermal-induced change in the anisotropy, photomagnetic effects manifest themselves through the appearance of transient laser-induced anisotropy axes.

In spite of a whole series of results which show that the absorption of the energy of femtosecond laser pulses makes it possible not only to efficiently excite magnetic dynamics, but also to control the magnetic state of the medium [5, 9, 14], thermal heating, which accompanies these processes, in many cases is considered to be a negative factor. Furthermore, the involvement of the excitation of other subsystems in the process, i.e., electrons and phonons, is also frequently considered a factor that complicates the optical excitation of the ultrafast magnetic dynamics. Therefore, a search is now underway for alternative mechanisms of excitation, which would make it possible to avoid the indicated limitations. For example, the possibility of exciting the spin dynamics by picosecond strain pulses [15, 16] or due to resonance magnetic-dipole transitions under the action of femtosecond pulses of terahertz radiation on the magnetic material [17–19] is actively being studied.

However, as will be considered in this article, femtosecond laser pulses also allow an interesting and important opportunity to directly excite the spin system due to so-called ultrafast opto-magnetic phenomena [9, 20–34]. These phenomena occupy a special place among the mechanisms of interaction of laser pulses with media, since, in contrast to the processes considered above, they are not based on optical absorption. The opto-magnetic action of light on the spins can be considered as the result of stimulated Raman-type processes [20, 23, 24]. This action has an impulsive nature, i.e., the disturbance of the spin system occurs only while the laser pulse acts on the medium. Therefore, ultrafast opto-magnetic phenomena represent an efficient mechanism to selectively excite the spin system [29]. An important feature of ultrafast opto-magnetic phenomena is their high sensitivity to the polarization of laser pulses [20, 21], which is absent for thermal mechanisms. Thus, ultrafast opto-magnetic phenomena provide an important degree of freedom for controlling the excitation of the spin system by means of a change in the polarization of femtosecond laser pulses, rather than only in the pulse intensity, central wavelength, or duration. Furthermore, the combination of ultrafast opto-magnetic phenomena with other laser-induced processes allows a unique opportunity of ultrafast control of the magnetic state of the medium [9, 14, 34].

The model media for the experimental observation of ultrafast opto-magnetic effects and related magnetic dynamics are the magnetically ordered dielectrics. The specific character of their electronic structure, the presence of an energy gap that ensures high optical transparency, the absorption of laser radiation on the localized d–d or f–f electron transitions that is weak in comparison with that observed in metals, and the absence of free electrons — all these factors open new opportunities for a detailed study of the interaction of femtosecond laser pulses with the spin system without introducing strong perturbation into other subsystems.

In this article we do not aim at presenting exhaustive survey of the results obtained in the field of the ultrafast laser-induced magnetic dynamics. We here consider only some studies united by the general problem of nondissipative or weakly dissipative interaction of femtosecond laser pulses with magnetically ordered media. In particular, in Section 2 we examine basic ultrafast opto-magnetic phenomena and their connection with magneto-optical effects. In Section 3, the principles of the experimental observation of the ultrafast laser-induced dynamics of the spin system are discussed. In Section 4, results concerning the experimental observation of ultrafast opto-magnetic phenomena and related magnetic dynamics are presented, and the microscopic mechanisms of ultrafast opto-magnetic effects are examined. Furthermore, we considered the problem of feasibility of a selective excitation of the spin system via such effects, i.e., such an excitation in which the energy and the angular momentum of the laser pulse are transferred exclusively into the spin system. In Section 5, the role of ultrafast opto-magnetic phenomena in controlling laser-induced phase transitions is discussed. In the conclusion, some prospects of further studies of ultrafast opto-magnetic phenomena as an important element of control of the magnetic state of media on pico- and subpicosecond time scales are presented.

2. Magneto-optical and opto-magnetic phenomena

The character of interaction of electromagnetic radiation of a frequency ω with a medium is determined, first of all, by the spectrum of electronic, phonon, and spin states and by their symmetry. In particular, the type of magnetic ordering of the

Table 1. Classification of magneto-optical and opto-magnetic phenomena.* For the magneto-optical phenomena, the corresponding components of the tensor of the dielectric susceptibility (2.3) are presented; for the opto-magnetic phenomena, the effective fields (2.4) are given.

Contribution to the Hamiltonian (2.2)	Magneto-optical effect (2.3)		Opto-magnetic effect (2.4)	
$\alpha_{ijk}E_iE_j^*M_k$	$\varepsilon_{ij}^{\mathrm{a}}=\alpha_{ijk}M_k$	Ferromagnetic Faraday effect	$H_k^{\mathrm{IFE}}=\alpha_{ijk}E_iE_j^*$	Inverse Faraday effect
$\alpha'_{ijk}E_iE_j^*L_k$	$\varepsilon_{ij}^{\mathrm{a}}=\alpha'_{ijk}L_k$	Antiferromagnetic Faraday effect	$h_k^{\mathrm{IFE}}=\alpha'_{ijk}E_iE_j^*$	Inverse Faraday effect
$\beta_{ijkl}E_iE_j^*M_kM_l$	$\varepsilon_{ij}^{\mathrm{s}}=\beta_{ijkl}M_kM_l$	Magnetic linear birefringence (Cotton–Mouton, or Voigt, effect)	$H_l^{\mathrm{ICME}}=\beta_{ijkl}E_iE_j^*M_k$	Inverse Cotton–Mouton effect
$\beta'_{ijkl}E_iE_j^*L_kL_l$	$\varepsilon_{ij}^{\mathrm{s}}=\beta'_{ijkl}L_kL_l$		$h_l^{\mathrm{ICME}}=\beta'_{ijkl}E_iE_j^*L_k$	
$\beta''_{ijkl}E_iE_j^*M_kL_l$	$\varepsilon_{ij}^{\mathrm{s}}=\beta''_{ijkl}M_kL_l$		$H_k^{\mathrm{ICME}}=\beta''_{ijkl}E_iE_j^*L_l$ $h_l^{\mathrm{ICME}}=\beta''_{ijkl}E_iE_j^*M_k$	
* Only effects that are observed upon the interaction of light with a nondissipative medium have been included.				

medium plays the decisive role in the formation of the magneto-optical and, as will be shown below, opto-magnetic responses. Let us examine, as an example, the magneto-optical response of a two-sublattice antiferromagnet which possesses weak ferromagnetism [35–37]. Its magnetic structure can be described by introducing ferromagnetic and antiferromagnetic vectors:

$$\mathbf{M}=\mathbf{M}_1+\mathbf{M}_2\,,\qquad \mathbf{L}=\mathbf{M}_1-\mathbf{M}_2\,, \tag{2.1}$$

where $\mathbf{M}_1$ and $\mathbf{M}_2$ are the magnetizations of the sublattices.

The Hamiltonian that describes the interaction of an electromagnetic wave with a magnetically ordered medium at an optical frequency ω can be presented as an expansion in the powers of the electric field of light $\mathbf{E}(\omega)$ and of the magnetic order parameters $\mathbf{M}$ and $\mathbf{L}$ [35–38]:

$$\mathcal{H}^{\mathrm{MO}}=-(\varepsilon_{ij}^0E_iE_j^*+\alpha_{ijk}E_iE_j^*M_k+\alpha'_{ijk}E_iE_j^*L_k +\beta_{ijkl}E_iE_j^*M_kM_l+\beta'_{ijkl}E_iE_j^*L_kL_l+\beta''_{ijkl}E_iE_j^*M_kL_l+\ldots)\,. \tag{2.2}$$

In expression (2.2), only terms of the lowest order of the expansion in $\mathbf{E}(\omega)$, which describe the linear optical and magneto-optical responses of the medium, are taken into account. The tensors $\hat{\varepsilon}^0$, $\hat{\alpha}$, and $\hat{\beta}$ correspond to the optical and magneto-optical susceptibilities. The number of nonzero distinct components of these tensors is uniquely determined by the elements of the spatial and time symmetry of the considered medium, i.e., by its crystallographic and magnetic point groups [39].

The response of the medium to the electric field of light is described by the tensor of the dielectric susceptibility, which connects the optical polarization $\mathbf{P}(\omega)$ induced in the medium with the electric field of the incident wave $\mathbf{E}(\omega)$:

$$\varepsilon_{ij}=-\frac{\partial\mathcal{H}^{\mathrm{MO}}}{\partial E_i\,\partial E_j^*}=\varepsilon_{ij}^{\mathrm{s}}+\varepsilon_{ij}^{\mathrm{a}}\,, \tag{2.3}$$

where $\varepsilon_{ij}^{\mathrm{s}}=\varepsilon_{ji}^{\mathrm{s}}$ and $\varepsilon_{ij}^{\mathrm{a}}=-\varepsilon_{ji}^{\mathrm{a}}$ are the symmetric and antisymmetric parts of the tensor, respectively. According to the Onsager principle, these contributions are even and odd with respect to time reversal. In the approximation of a nondissipative medium considered here, the symmetric and antisymmetric parts of the tensor ε_{ij} are real and imaginary, respectively.

Based on Eqns (2.2) and (2.3), we can write expressions for the components of the tensors of the dielectric susceptibility that correspond to the optical and magneto-optical responses of the medium. Thus, the first term in expansion (2.2) gives the contribution to the component of the tensor ε_{ij} that describes the crystallographic refraction, which is not connected with the magnetic order. The remaining terms describe the magneto-optical effects, which are linear, quadratic, and bilinear in the magnetic-order parameters. Expressions for the components of the dielectric-susceptibility tensor connected with different magneto-optical effects are given in Table 1.

Based on the effective Hamiltonian (2.2), expressions for inverse magneto-optical, or opto-magnetic, phenomena can also be obtained. The existence of such phenomena was first predicted in Ref. [40], where, based on a phenomenological analysis, an expression for the magnetization $\mathbf{M}^{\mathrm{IFE}}(0)=\alpha\mathbf{D}(\omega)\times\mathbf{D}^*(\omega)$ that is induced in the medium upon its irradiation by circularly polarized light was obtained. Here, $\mathbf{D}(\omega)$ is the vector of electric displacement in the medium, and α is the magneto-optical susceptibility, which also describes the magneto-optical Faraday effect. The magnetization thus induced possesses the following properties. It is collinear to the wave vector of the radiation, it changes sign when the polarization of the radiation changes from the right-circular to the left-circular, and its magnitude is proportional to the intensity of the radiation. The coefficient of proportionality α between the induced magnetization and the intensity of light is connected with the magneto-optical susceptibility, which also determines the ferromagnetic Faraday effect.

The appearance of magnetization in paramagnetic and diamagnetic media upon their irradiation by circularly polarized light was demonstrated experimentally in [41]. The authors of this study called the observed phenomenon 'the inverse Faraday effect' (IFE).

The quantum-mechanical theory of the inverse Faraday effect and of other opto-magnetic phenomena for thermodynamically equilibrium systems was developed in [38]. In particular, it was shown that circularly polarized light acts on the medium as an effective magnetic field directed along the wave vector of light: $\mathbf{H}^{\mathrm{IFE}}=-\partial\mathcal{H}^{\mathrm{MO}}/\partial\mathbf{M}$. When considering a thermodynamically equilibrium situation, such an effective field leads to the appearance of laser-induced magnetization, described above.

Thus, a femtosecond laser pulse must not induce magnetization [24] but, rather, a pulse of an effective magnetic field, whose direction is determined by the polarization of the radiation, and magnitude by the radiation intensity. As was shown in [21, 22], to describe the action of femtosecond laser pulses on the media that are characterized by the presence of two or several magnetic sublattices, i.e., ferrimagnets, antiferromagnets, and weak ferromagnets, several laser-induced fields should be introduced:

$$\mathbf{H}^{\mathrm{OM}} = -\frac{\partial \mathcal{H}^{\mathrm{MO}}}{\partial \mathbf{M}}\,, \tag{2.4a}$$

$$\mathbf{h}^{\mathrm{OM}} = -\frac{\partial \mathcal{H}^{\mathrm{MO}}}{\partial \mathbf{L}}\,. \tag{2.4b}$$

From Table 1, which contains the contributions to Hamiltonian (2.2) that are linear and quadratic in the ferromagnetic and antiferromagnetic vectors, as well as the phenomenological expressions for the effective laser-induced fields (2.4), it follows that opto-magnetic phenomena can be observed under the influence of not only circularly polarized pulses, which induce the effective field $\mathbf{H}^{\mathrm{IFE}}$, but also linearly polarized pulses, which create the field $\mathbf{H}^{\mathrm{ICME}}$. In contrast to the inverse Faraday effect, which appears in the first case, in the second case the inverse Cotton–Mouton effect (ICME) (or, otherwise, the inverse Voigt effect) can be observed [38]. An essential difference between these two phenomena lies in the fact that the inverse Faraday effect can be observed in para- and diamagnetic media, since the time symmetry of the circularly polarized optical pulse corresponds to the symmetry of the magnetic field. The inverse Cotton–Mouton effect is, on the contrary, possible only in the presence of a magnetic order or an external field.

To describe the dynamics of the spin system induced by ultrafast opto-magnetic phenomena, the Landau–Lifshitz equation [42] is used. In media with several magnetic sublattices, in particular, in the case of a two-sublattice antiferromagnet, it is possible to write the Landau–Lifshitz equation for each sublattice. However, in our opinion, a more convenient and more informative approach is that in which the modified Landau–Lifshitz equations are written for the ferromagnetic **M** and antiferromagnetic **L** vectors [36, 43]:

$$\frac{\mathrm{d}\mathbf{M}}{\mathrm{d}t} = -\gamma(\mathbf{M}\times\mathbf{H}^{\mathrm{eff}} + \mathbf{L}\times\mathbf{h}^{\mathrm{eff}})\,, \tag{2.5a}$$

$$\frac{\mathrm{d}\mathbf{L}}{\mathrm{d}t} = -\gamma(\mathbf{L}\times\mathbf{H}^{\mathrm{eff}} + \mathbf{M}\times\mathbf{h}^{\mathrm{eff}})\,, \tag{2.5b}$$

where $\mathbf{H}^{\mathrm{eff}} = -\partial(\mathcal{H}+\mathcal{H}^{\mathrm{MO}})/\partial\mathbf{M}$ and $\mathbf{h}^{\mathrm{eff}} = -\partial(\mathcal{H}+\mathcal{H}^{\mathrm{MO}})/\partial\mathbf{L}$ are the effective fields, and $\mathcal{H}$ is the Hamiltonian of the magnetic system. In general, the expressions for the effective fields $\mathbf{H}^{\mathrm{eff}}$ and $\mathbf{h}^{\mathrm{eff}}$ contain the effective exchange fields, the effective fields of magnetic anisotropy, and the external magnetic field. When considering ultrafast opto-magnetic phenomena, the time-dependent effective laser-induced fields $\mathbf{H}^{\mathrm{OM}}$ and $\mathbf{h}^{\mathrm{OM}}$ (Table 1) are introduced into the expressions for $\mathbf{H}^{\mathrm{eff}}$ and $\mathbf{h}^{\mathrm{eff}}$.

In Section 4, we will consider several experimental examples which demonstrate the basic features of magnetic dynamics excited in the magnetic media in the case of ultrafast opto-magnetic phenomena.

3. Experimental methods of studying ultrafast laser-induced dynamics in magnetic media

3.1 Femtosecond time-resolved magneto-optical spectroscopy

The registration of the dynamic response of a magnetically ordered medium to the action of a femtosecond laser pulse requires the use of procedures of detection that have the necessary subpicosecond time resolution. It is obvious that such a resolution is provided by the femtosecond optical pulses themselves. Magneto-optical effects, both linear [37, 44, 45] and nonlinear [46, 47], are known as an efficient tool for studying the volume, surface, and interfacial magnetic properties of various media. It is natural that, among the different methods in an experimental study of laser-induced dynamics in magnetically ordered media, the magneto-optical methods have obtained the widest use.

In the magneto-optical method of studies, laser-induced changes in the magnitude and direction of the magnetic order parameter are registered via the magneto-optical response of the medium in the near infrared and visible spectral ranges. During the measurements, the sample, which is usually placed in an external magnetic field that determines the equilibrium direction of the magnetization, is irradiated by femtosecond laser pump pulses. The pulse duration is from ≈ 200 fs to only several femtoseconds. The energy density in the pump pulse varies from several fractions of a mJ cm^{-2} for strongly absorbing media to several dozen mJ cm^{-2} for weakly absorbing materials. Less powerful (≈ 10 μJ cm^{-2}) probe pulses synchronized with pump pulses are used for measuring the magnitude of a magneto-optical effect as functions of the pump-probe time delay. The selection of a effect is determined, first, by the magnetic and magneto-optical properties of the sample under investigation. Thus, measurement of the Faraday effect for probe pulses makes it possible to obtain information concerning the dynamics of the component of the magnetization directed along the wave vector of the probe pulses. Changes of the magnetic linear birefringence induced by the pump pulses, on the contrary, indicate a change in the components of the magnetization or in the antiferromagnetic vector which are perpendicular to the direction of the propagation of the probe pulses. For the detection of magnetic dynamics in an absorbing medium, apart from the Faraday and Cotton–Mouton (Voigt) effects, i.e., circular and linear birefringence, the effects of circular and linear dichroism can be used as well. To study the laser-induced dynamics in highly absorbing media, time changes in the magnitude of the magneto-optical Kerr effects are measured [44], i.e., changes in the polarization of the probe pulses reflected from the sample.

Note the following feature of the detection of laser-induced dynamics in multisublattice magnetic media, for example, in antiferromagnets. In general, the possibility of investigating the static properties of antiferromagnets can apparently be provided by the magneto-optical effects that are quadratic in the magnetic order parameter, such as the magnetic linear birefringence (see Table 1). On the other hand, in order to register the laser-induced dynamics of antiferromagnets, both the quadratic and linear magneto-optical effects can be used [25, 29]. Indeed, in a compensated antiferromagnet the precession of the magnetizations of the sublattices leads not only to oscillations of the antiferromag-

netic vector **L**, but also to the emergence of nonzero dynamic magnetization [36]. Therefore, for the detection of laser-induced coherent spin precession in antiferromagnets measurements of the Faraday effect and magnetic linear birefringence for probe pulses allow obtaining complementary information. However, only the quadratic magneto-optical effects allow changes in the magnitude of the vector of antiferromagnetism to be detected as well, which are the consequence, for example, of incoherent laser-induced spin dynamics [29].

Modern laser systems make it possible to implement a tuning of the central photon energy of the femtosecond pulses in a broad spectral range. As was shown in some studies [26, 29, 48, 49], this degree of freedom significantly enhances the capability of the magneto-optical probing of ultrafast magnetic dynamics. Thus, the use of probe pulses in different spectral ranges can, in some cases, make it possible to distinguish the contributions to the measured signals which come from different magnetic subsystems [48]. This allows one to overcome partly such a substantial limitation on the method of magneto-optical probing as its spectral integration. However, it is the most interesting to study of the response of the magnetically ordered medium to the action of femtosecond laser pulses with different central photon energies. Such experiments, first, can reveal the microscopic mechanisms of the excitation of magnetic dynamics, since they make it possible to link the response of the medium to the specific features of its electronic structure. Second, when several spectral-dependent mechanisms of the excitation of spin dynamics exist, the spectral tuning of pump pulses opens the possibility of controlling contributions from these mechanisms. In Section 4.3, an example is given of the control of the coherent and incoherent spin dynamics in antiferromagnetic $KNiF_3$ by such tuning the central photon energy of the pump pulse.

It should be noted that the magneto-optical methods for studying laser-induced magnetic dynamics have a number of limitations. First, the excitation of the medium by a femtosecond laser pulse generally leads to a change of not only the magnetic parameters of the medium but also its optical properties. Optical and magneto-optical susceptibilities, which connect a change in the polarization of the probe pulses with changes in the magnetization of the medium, can differ from their steady-state values and possess their own dynamics. Therefore, the interpretation of the results of magneto-optical experiments can be ambiguous, e.g. in studies of the laser-induced dynamics in metals because of the intense absorption [50]. Second, in the case of media with a complex magnetic structure or in the case of heterostructures consisting of several magnetic layers, distinguishing among the dynamics of different components is a nontrivial problem and cannot be always solved [48, 51]. Therefore, at present, when studying laser-induced magnetic dynamics, some techniques that supplement magneto-optical methods are finding increasingly wider application. They include, first of all, X-ray magneto-optical measurements with a femtosecond time resolution, femtosecond terahertz spectroscopy, and spin-polarized photoemission. Nevertheless, magneto-optical methods remain an important tool to study ultrafast magnetic dynamics. This is due to a whole range of merits, such as the relative simplicity and availability, a good degree of spatial locality, and universality, which makes it possible to study the dynamics of the processes in media with various magnetic structures.

3.2 Single-pulse femtosecond time-resolved magneto-optical microscopy

An important step in the progress of methods of investigating laser-induced magnetic dynamics was the development of single-pulse time-resolved magneto-optical microscopy with a femtosecond time resolution. The main differences between this procedure and that described in Section 3.1 are the use of single pump and probe pulses instead of sequences of several hundred such pulses and obtaining a magneto-optical image with the aid of a probe pulse.

The main advantage of this procedure is the possibility of simultaneous studying the spatial and temporary dynamics of a magnetic system with a high resolution. This method was evidently used for the first time in [52] for the investigation of laser-induced demagnetization in permalloy. Subsequently, magneto-optical microscopy with a femtosecond time resolution has played an important role in the revealing of mechanisms of ultrafast optical switching of the magnetization in metals [53, 54].

In our opinion, the use of spatial resolution in the pump–probe measurements is especially important in the study of threshold processes. Indeed, in this case, the character of the laser-induced dynamics can substantially change within the limits of the region that is subjected to the action of a pump pulse, which, as a rule, has a Gaussian spatial distribution of intensity. In Section 4.3, we show that the method of time-resolved femtosecond magneto-optical microscopy significantly widens the opportunities to study laser-induced magnetic dynamics, in particular, in a vicinity phase transitions.

4. Ultrafast opto-magnetic phenomena in magnetic dielectrics

4.1 Experimental observation of ultrafast inverse Faraday and Cotton–Mouton effects

The excitation of spin dynamics via the ultrafast inverse magneto-optical Faraday effect was first investigated in the orthoferrite $DyFeO_3$ [20]. The rare-earth orthoferrites $RFeO_3$, where R is the yttrium ion Y^{3+} or a rare-earth ion, are crystallized in an orthorhombically distorted perovskite structure. At temperatures of the order of the Néel temperature $T_N \approx 650$ K, the magnetic moments of iron ions Fe^{3+} form four antiferromagnetically coupled sublattices $\mathbf{M}_{1-4}$ [55, 56]. The Dzyaloshinskii–Moriya interaction leads to a canting of the sublattices and to the appearance of a nonzero net magnetization; i.e., rare-earth orthoferrites are weak ferromagnets. To describe the magnetic structure of orthoferrites, vectors of ferromagnetism, $\mathbf{M} = \sum_i \mathbf{M}_i$, and antiferromagnetism, $\mathbf{L} = \mathbf{M}_1 - \mathbf{M}_2 + \mathbf{M}_3 - \mathbf{M}_4$, are introduced. At temperatures below 5 K, a magnetic order can appear in a rare-earth sublattice. Depending on the type of rare-earth ion, first-order or second-order reorientation phase transitions can be observed in the orthoferrites [55–57]. The dysprosium orthoferrite at temperatures above 51.6 K [55, 57] is a weak ferromagnet, in which the vectors **M** and **L** are oriented along the crystallographic c and a axes, respectively.

Figure 1 shows the result of a femtosecond magneto-optical 'pump–probe' experiment (see Section 3.1) on the excitation of a single crystal of $DyFeO_3$ by circularly polarized pulses with a duration of 200 fs with a central photon energy of 1.55 eV. The wave vector of the pump pulses was directed perpendicularly to the ac plane, in which the

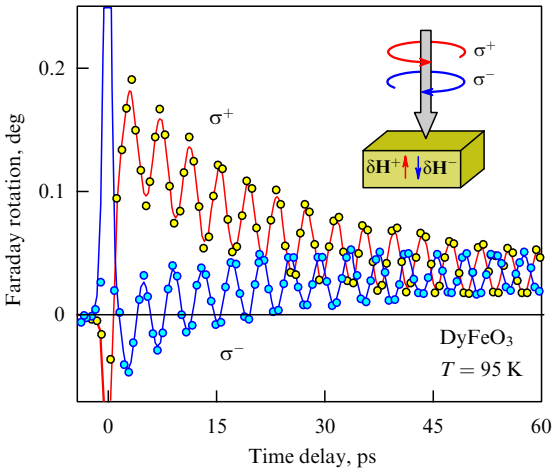

Figure 1. Dynamics of the rotation of the plane of polarization of probe pulses after the excitation of a weak ferromagnet $DyFeO_3$ by 200-femtosecond pump pulses with right-hand (σ^+) and left-hand (σ^-) circular polarization with the central photon energy equal to 1.55 eV and the energy density in the pulse equal to 30 mJ cm^{-2}. Inset: the action of a circularly polarized laser pulse is equivalent to the action of a short pulse of an effective magnetic field **H** [20].

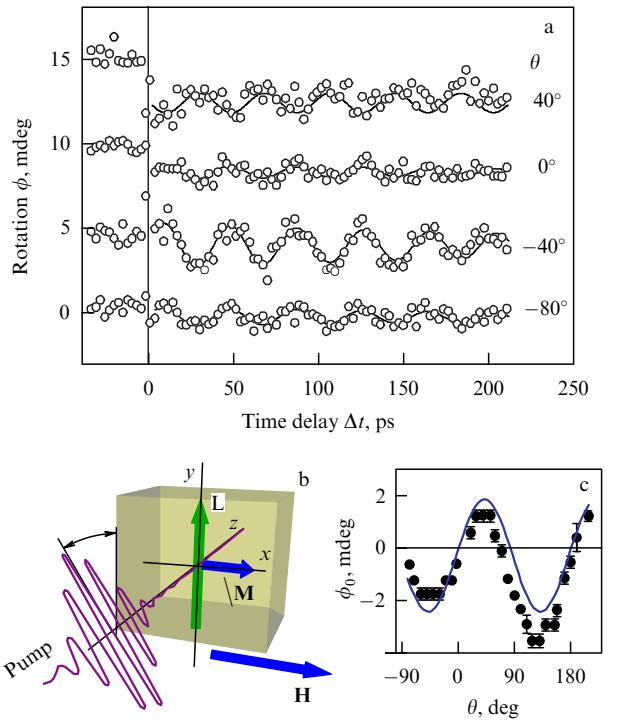

Figure 2. (a) Time dependence of the angle of rotation $\phi(t)$ of the plane of polarization of probe pulses after the action of linearly polarized 100-femtosecond pulses on the weak ferromagnet $FeBO_3$. (b) Geometry of the experiment: θ is the azimuthal angle, which describes the linear polarization of the pump pulses. (c) Dependence of the amplitude of laser-induced precession ϕ_0 on the azimuthal angle θ [22].

vectors **M** and **L** lie. As can be seen from Fig. 1, the action of pulses with right-hand and left-hand circular polarizations leads to the appearance of oscillations of the polarization of the probe pulses. The analysis of the frequency of these oscillations and of their temperature dependences has shown that they originate from the laser-induced precession of magnetization.

The most important result of this study was the observation that the pulses with right-hand and left-hand circular polarizations excite the precession with opposite initial phases. Thus, the process of excitation can be described as the action of a short pulse of a laser-induced effective field (2.4), i.e., as the ultrafast inverse Faraday effect. The effective field leads to the deviation of the magnetization from the equilibrium direction. The degree of this deviation can be found by substituting into the Landau–Lifshitz equation (2.5) the effective field (2.4), the magnitude and duration of which are determined by the intensity and duration of the laser pulse, as well as by the magneto-optical susceptibility of the medium. Based on the analysis of the amplitude of the excited precession, the magnitude of the effective laser-induced field was estimated to be of 0.03 T [20] for the experiment, which results are shown in Fig. 1. The action of a laser pulse with the central photon energy of 1.55 eV, duration of 200 fs, and energy density of 500 mJ cm^{-2} on the dysprosium orthoferrite should be equivalent to the action of a 200-fs pulse of an effective field (2.4) of strength 5 T.

A comparison of the magneto-optical and opto-magnetic effects indicates the possibility of exciting precession by linearly polarized femtosecond laser pulses. Indeed, according to Table 1, the emergence of an effective field that corresponds to the inverse Cotton–Mouton effect can be expected. The feasibility of exciting the precession of magnetization by linearly polarized pulses was first demonstrated experimentally for the weak ferromagnet $FeBO_3$ [21], which has a calcite rhombohedral crystal structure. The magnetic moments of the iron ions form two sublattices, $\mathbf{M}_1$ and $\mathbf{M}_2$, coupled antiferromagnetically. The Dzyaloshinskii–Moriya interaction leads, as in the case of orthoferrites, to a canting of the sublattices and to the emergence of a nonzero bulk magnetization. The static magnetic structure of the iron borate is described by two vectors, $\mathbf{M} = \mathbf{M}_1 + \mathbf{M}_2$ and $\mathbf{L} = \mathbf{M}_1 - \mathbf{M}_2$. This two-sublattice weak antiferromagnet is a model subject for the analysis of the excitation of magnetic precession on the basis of Eqns (2.4) and expressions for the effective laser-induced fields (see Table 1).

As can be seen in Fig. 2a, action of femtosecond linearly polarized laser pulses on the single crystal of the iron borate results in precession of the magnetization and of the antiferromagnetic vector. The initial phase of the precession possesses a periodic dependence (Fig. 2c) on the azimuthal angle (Fig. 2b) of the linear polarization of the pump pulses. A detailed examination carried out in [22] has shown that the excitation of the precession by linearly polarized femtosecond laser pulses can indeed be considered as a manifestation of the ultrafast inverse Cotton–Mouton effect (see Table 1). In this weak ferromagnet, $L \gg M$; therefore, in the process of the excitation of the precession via the opto-magnetic Cotton–Mouton effect, the effective field $\mathbf{h}^{\mathrm{OM}} = -\partial\mathcal{H}^{\mathrm{MO}}/\partial\mathbf{L}$ (2.4b) connected with the **L** terms in the expansion (2.1) plays a decisive role. A similar conclusion was made in [22] regarding the ultrafast inverse Faraday effect.

Thus, it was demonstrated, both experimentally and phenomenologically, that it is important to take into account the multisublattice magnetic structure for the interpretation of the laser-induced dynamics in magnetic dielectrics. To date, it has been shown in a number of experiments that the selective responses of magnetic sublattices to the excitation by femtosecond laser pulses play the most important role in the ultrafast dynamics of magnetization, not only in opto-magnetic [58], but also in other mechanisms of interaction [5, 59].

It was shown in a number of experimental studies that the ultrafast inverse Faraday and Cotton–Mouton effects make it possible to excite the precession of magnetization in dielectrics with different magnetic structures, such as ferrimagnets [9, 10, 27, 58, 60, 61], antiferromagnets [25, 28-30], weak ferromagnets [20–22], ferromagnets [32], and chiral magnets [31].

4.2 Microscopic mechanisms of ultrafast inverse opto-magnetic effects

Already in the first work concerning laser-induced excitation and control of the coherent precession of magnetization in the dielectric $DyFeO_3$ [20], an assumption was made that the microscopic mechanism of the process is the stimulated Raman scattering on magnons [62]. Figure 3 schematically shows such a process, in which two photons with frequencies ω and $\omega - \Omega$ and a magnon with frequency Ω are involved. In the above-described experiments, the period $2\pi/\Omega$ of coherent spin precession excited by femtosecond laser pulses varied from several dozen picoseconds to several nanoseconds, i.e., was much longer than the duration of the pump pulse. Thus, the spectral width of the pulse exceeds the frequency of the magnon. Therefore, there are pairs of photons present in the pump pulse which satisfy the condition for the appearance of stimulated Raman scattering leading to the excitation of coherent magnons. In [21], in order to emphasize the specific feature of this process during femtosecond excitation, the term *impulsive stimulated Raman scattering* (ISRS) *on magnons* was suggested, i.e., terminology employed for describing the laser-induced excitation of the coherent oscillations of a lattice was used [63, 64].

The phenomenological theory of ISRS on magnons for media with various types of magnetic ordering was developed in [23]. It should be noted that in [23] the process of interaction of femtosecond laser pulses with a magnetically ordered medium was considered using a classical Hamiltonian formalism for wave processes [65, 66]. In [22], the validity of the approaches based on effective fields formalism and the classical Hamiltonian one has been demonstrated for the case of the weak ferromagnet $FeBO_3$. The models used for developing the microscopic theory of the excitation of coherent spin precession by impulsive stimulated Raman scattering were considered in [67, 68].

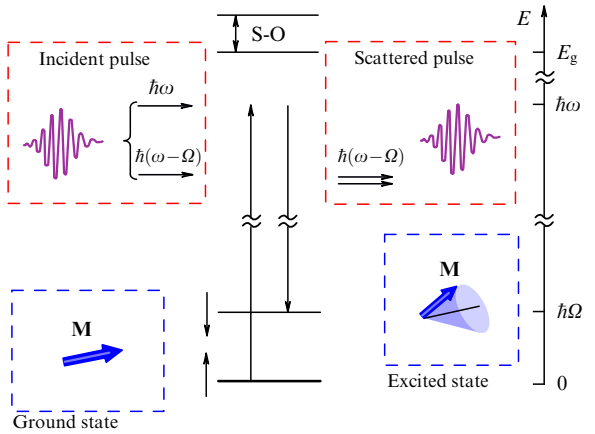

Figure 3. Schematic representation of the process of impulsive stimulated Raman scattering on the magnons. The femtosecond laser pulse with a central photon energy ω lower than the edge of the fundamental absorption ($\hbar\omega < E_g$) contains pairs of photons, the difference between frequencies of which corresponds to frequency Ω of the magnon in the medium. Such pairs of photons participate in the process of stimulated Raman scattering, which leads to the generation of coherent magnons with frequency Ω. S–O is the spin–orbital splitting of the excited states, which makes a contribution to the magneto-optical and opto-magnetic responses of the medium.

4.3 Selective excitation of magnetic dynamics via ultrafast opto-magnetic phenomena

The phenomenological analysis of effective laser-induced fields (see Table 1) and the theory of impulsive stimulated Raman scattering on magnons [28] are both based on the approximation of a nondissipative medium. However, in all the media examined above the optical absorption at the energy of photons of the pump pulses (typically, 1.55 eV) was from several dozen to several hundred cm^{-1}. The presence of finite absorption raises the question about the validity of the approach in which this absorption is neglected, and the answer can be obtained in an experimental study. Furthermore, in a number of studies (see, e.g., [17]) it was assumed that the presence of absorption, which unavoidably leads to a heating of the lattice, does not make it possible to obtain selective optical excitation, which affects exclusively the spin system.

An important experiment that confirmed the validity of this approach to the description of polarization-dependent excitation of coherent magnons in dielectrics was focused at the magnetic dynamics in the antiferromagnetic $KNiF_3$ [29]. This material, with an ideal cubic perovskite structure, is a model Heisenberg antiferromagnet with a Néel temperature $T_N = 246$ K [69]. The choice of this material for studying the problem of the role of absorption in the excitation of magnetic dynamics by ultrafast opto-magnetic effects was justified by its optical properties. The width of the energy gap in $KNiF_3$ is more than 6 eV [70], which substantially exceeds the typical values of approximately 3 eV, characteristic of the iron oxides discussed above. Figure 4a shows the spectrum of absorption of a single crystal of $KNiF_3$ [29]. As can be seen, in the spectral range of photon energies of 0.8–4.8 eV, this material has relatively weak (absorption coefficient of about 100 cm^{-1}) absorption bands due to the localized transitions between the 3d states of Ni^{2+} ions split by the crystalline field of the ligands [71]. Near a photon energy of approximately 2.2 eV, this material has a window of transparency, in which the absorption coefficient is very low.

The experimental investigation of laser-induced magnetic dynamics in $KNiF_3$ has shown that femtosecond laser pulses with a central photon energy of 2.2 eV in the transparency window efficiently excite coherent spin precession (Fig. 4b). The polarization dependences of the amplitude and phase of the precession are in good agreement with the model of the ultrafast inverse Faraday effect and ISRS. The results obtained made it possible to experimentally confirm the validity of the consideration of ultrafast opto-magnetic effects in the approximation of a nondissipative medium.

A comparison of the amplitudes of the spin precession in $KNiF_3$ excited by laser pulses with the central photon energy of 2.2 eV and with energies that correspond to the absorption bands (Fig. 4a) has shown that absorption due to d–d transitions does not lead to an increase in the efficiency of the excitation. At the same time, the absorption of part of the energy of laser pulses exerts a substantial influence on the efficiency and mechanism of the excitation of the incoherent spin dynamics. Indeed, the optical absorption in the dielectric oxides and fluorides of transition metals in the near infrared

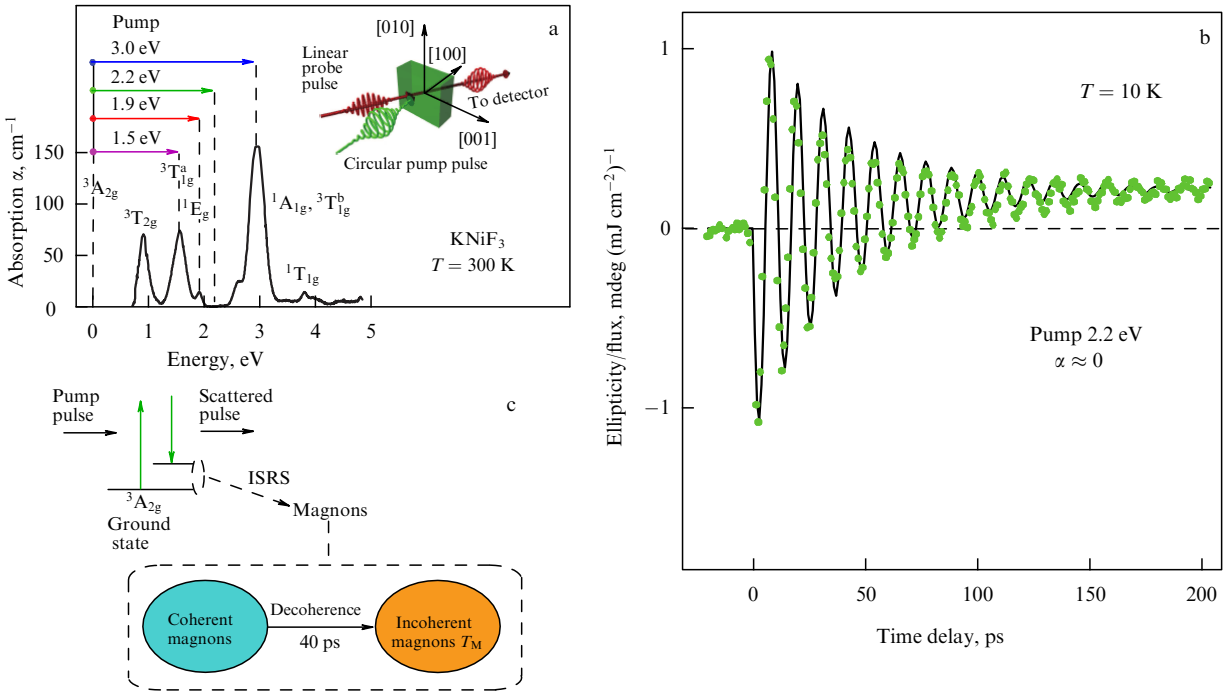

Figure 4. (a) Absorption spectrum of $KNiF_3$ at room temperature. The arrows show the photon energies of the pump pulses used for the excitation of the coherent spin precession. In the inset, the geometry of the experiment is presented. (b) Dynamics of the degree of ellipticity of the polarization of the probe pulses after the excitation of the antiferromagnet $KNiF_3$ by right-hand circularly polarized pump pulses with a duration of 35 fs with the central photon energy of 2.2 eV. The form of the signal is caused by the oscillations of the antiferromagnetic vector **L**. The difference in signal at delay times of more than 100 ps from the one observed at the negative values of the delay times reflects a change in the length of the antiferromagnetic vector **L**, i.e. the laser-induced demagnetization. (c) Schematic representation of the process of selective laser-induced excitation of the spin system [29].

and in visible parts of the spectral range is determined by the localized transitions between 3d states of ions. The excited states in the 3d shell are characterized by short lifetimes, on the order of several hundred femtoseconds. As a result, the absorbed energy of the laser pulse is transferred by means of nonradiative processes to the lattice, leading to the excitation of incoherent phonons, i.e., to a heating of the lattice. One of the consequences of the laser-induced heating of the lattice, i.e., of the phonon subsystem, is an increase in the spin temperature [10, 72–74], which appears as a result of the phonon–magnon interaction. The characteristic times of demagnetizing in the dielectrics are on the order of several hundred picoseconds.

In the experiments, an increase in the effective spin temperature is manifested as demagnetization, which can be considered as a variant of the incoherent laser-induced spin dynamics. At temperatures that are substantially lower than the temperature of the transition to the paramagnetic state, the degree of demagnetization is several fractions of a percent. This, together with a long characteristic time, is a fundamental difference between this process and laser-induced demagnetization in metals [3], which, in a number of cases, can reach 100%. Nevertheless, complete laser-induced demagnetization in the dielectrics as a result of phonon–magnon interaction can be realized in the range of temperatures close (within several kelvins) to the temperature of the transition to the paramagnetic state [10, 72].

Such a process of demagnetization with a characteristic time of approximately 100 ps was also observed in $KNiF_3$ as a slow laser-induced change in the ellipticity of the polarization of the probe pulses upon excitation by laser pulses with photon energies of 1.5, 1.9, and 3.0 eV. In spite of the expectations, the demagnetization was also observed upon excitation in the region of the transparency window near 2.2 eV (Fig. 4b). The characteristic time of this process differed significantly from the time of the demagnetization based on the phonon–magnon interaction, and was of 40 ps. The same value characterized the damping time of the coherent spin precession. The experimental data obtained indicate that an increase in the effective temperature of the spin system in the excitation of a dielectric by femtosecond pulses in the region of the transparency window occurs as a result of the decoherence of the magnons induced by the ultrafast inverse Faraday effect (Fig. 4c).

Thus, it has been shown in [29] that in the absence of optical absorption the excitation of the spin subsystem by femtosecond laser pulses can be selective: part of the energy of the laser pulse is transferred exclusively to the spin subsystem through the excitation of coherent precession due to ultrafast opto-magnetic effects. The subsequent decoherence of the spin precession leads to an increase in the effective spin temperature. It should be noted that the equality of the time of the decoherence of the spin precession to the time of an increase in the effective spin temperature serves as a criterion which allows one to determine whether a selective excitation of the spin system by a femtosecond laser pulse is realized [29].

5. Ultrafast opto-magnetic effects as a method for controlling laser-induced phase transitions

Although a laser pulse can lead to the excitation of coherent spin precession even in the absence of optical absorption, the latter, to a certain extent, is present in the majority of

media and can affect the process of interaction. Apart from the demagnetization we considered in Section 4.3, the laser-induced heating of the lattice can also change the strength of magnetic anisotropy [74]. As in the case of demagnetization, this effect must be manifested most vividly near the reorientation phase transitions caused by changes of the magnetocrystalline anisotropy [8].

The rare-earth orthoferrites $R\mathrm{FeO_3}$ which properties were described in Section 4.1 can serve as the model materials for studying the dynamics of the magnetization related to laser-induced phase transitions. The magnetic anisotropy in these materials has competing contributions from the single-ion anisotropy of iron ions $\mathrm{Fe^{3+}}$ and from the anisotropic $\mathrm{Fe^{3+}}-R^{3+}$ exchange. As a result, the different temperature dependences of these two contributions lead to the appearance of reorientation phase transitions, for example, such, upon which a reorientation of vectors **M** and **L** in the *ac* plane occurs (Fig. 5). The energy of the magnetic anisotropy of the orthoferrite in this case can be presented in the following form:

$$\Phi(T) = K_1(T)\sin^2\phi + K_2\sin^4\phi + \mathrm{const}\,,$$

where ϕ is the angle between the magnetization direction and the crystallographic c axis; and $K_1(T)$ and K_2 are the anisotropy parameters, the change in which leads to the appearance of reorientation phase transitions [55–57]. Depending on the sign of K_2 and on the relationship between $K_1(T)$ and K_2, reorientation phase transitions of the 1st or 2nd order can be observed in orthoferrites upon a change of the temperature. Below, we will discuss the case of $K_2 > 0$, in which the second-order phase transitions can occur [57].

As an example, let us consider the thulium orthoferrite $\mathrm{TmFeO_3}$, in which at temperatures below the Néel temperature $T_\mathrm{N} = 632$ K the anisotropy parameter is $K_1(T) > 0$. In this case, the magnetization **M** and the antiferromagnetic vector **L** are directed along the c and a axes, respectively, and the material is in the Γ_4 phase [56] (Fig. 5c). As the temperature decreases, at $T_2 = 91$ K a second-order phase transition occurs, corresponding to the condition $K_1(T_2) = 0$. In this case, an angular Γ_{24} phase becomes equilibrium one, in which the vectors **M** and **L** rotate in the *ac* plane (Fig. 5b). A further temperature decrease to 80 K leads to the appearance of a second phase transition of the 2nd order at $K_1(T_1) + 2K_2 = 0$. In this low-temperature Γ_2 phase, the vectors **M** and **L** are directed along the a and c axes, respectively (Fig. 5a). It should be noted that the type of rare-earth ion has an effect not only on the nature of the reorientation transitions but also on their temperatures [55].

In Ref. [8], it is shown that, upon the action of 100-fs laser pulses on a thulium orthoferrite in the Γ_2 or Γ_{24} phase, a spin reorientation on time scales of approximately 5 ps is observed. The closer the initial temperature of the sample to $T_1 = 80$ K, the more pronounced this process becomes. These processes disappear when the initial temperature of the sample is higher than $T_2 = 91$ K.

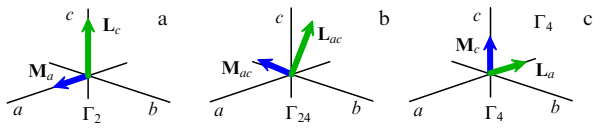

Figure 5. Reorientation second-order transitions in rare-earth orthoferrites: orientations of the ferromagnetic **M** and antiferromagnetic **L** vectors at temperatures (a) below the temperature T_1, (b) in the interval of $T_2 - T_1$, and (c) above the temperature T_2.

Thus, in Ref. [8] it was shown for the first time that the second-order spin-reorientation phase transition in a rare-earth orthoferrite can be induced by a femtosecond laser pulse and that the time after which the transition occurs is equal to several picoseconds. It should be noted that laser-induced reorientation phase transitions of both the 2nd and 1st order were observed in orthoferrites $\mathrm{ErFeO_3}$, $\mathrm{SmFeO_3}$, and $\mathrm{HoFeO_3}$, and also in a number of solid solutions of orthoferrites $(R_1, R_2)\mathrm{FeO_3}$, where R_1 and R_2 are different rare-earth ions [14, 34, 75–77].

In [76], the authors studied the effect of the initial temperature of the sample on time after which a laser-induced transition occurs in the erbium orthoferrite $\mathrm{ErFeO_3}$. It has been shown that the lower the initial temperature of the sample $(T_0 < T_1)$ and the less the energy density in the pumping pulse, the larger the delay between the moments of the action of the pump pulse and the emergence of the reorientation phase transition. This result made it possible to reveal important specific features of the mechanism of the laser-induced transition. As was expected, this thermal mechanism takes place due to the absorption of the laser-pulse energy at localized transitions in the 3d shell of iron ions. Due to the nonradiative transitions, the energy transfer to the lattice occurs on a relatively short time scale, on the order of several hundred femtoseconds. At the following stage, an increase in the temperature of phonons, i.e., of the lattice, leads to a change in the population of the sublevels of the rare-earth ions and, as a result, to a change in their magnetic moments. Specifically, it is this change that affects the anisotropic $R^{3+}-\mathrm{Fe^{3+}}$ exchange, which makes a contribution to the magnetic anisotropy. However, the characteristic time of the electron–phonon interaction for rare-earth ions is equal to several picoseconds, which, in particular, is reflected in the considerably narrower absorption lines of the rare-earth ions than the wide absorption bands of iron ions in the orthoferrites [78]. Thus, it is the time of the electron–phonon interaction, which amounts to several picoseconds for rare-earth ions, that determines at what time delay after the action of the laser pulse the phase transition occurs.

As was shown above, the action of a laser pulse on a rare-earth orthoferrite in the low-temperature Γ_2 phase leads to the appearance of a new phase, Γ_{24} or Γ_4, with a new direction of the anisotropy axis, which can be considered to be a method for ultrafast control of the direction of magnetization. However, in the absence of an external magnetic field or of another factor that will unambiguously determine the orientation of the magnetization in the new laser-induced phase, the above-considered ultrafast phase transition must lead to a multidomain state, in which two (in the Γ_4 phase) or four (in the Γ_{24} phase) types of magnetic domains can occur.

On the other hand, owing to the moderate optical absorption of orthoferrites in the near infrared part of the spectral range [78], the laser-induced phase transition and the excitation of precession due to ultrafast inverse magneto-optical effects can coexist in certain temperature ranges near the temperatures of phase transitions [75]. Therefore, the idea of using a balance between these two processes to control the

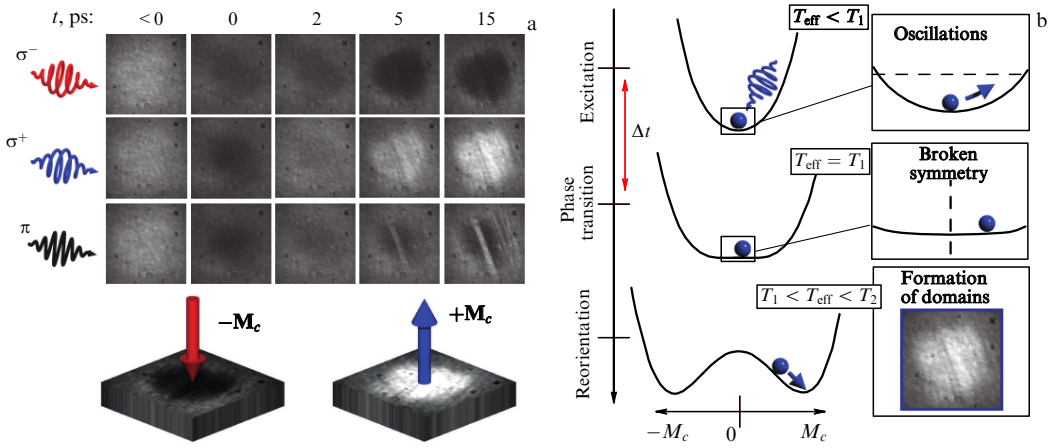

Figure 6. (a) Single-pulse magneto-optical images in a plate (91 μm thick) of a single crystal of $(Sm_{0.5}Pr_{0.5})FeO_3$ with the normal along the *c* axis, obtained for different time delays *t* after the action of femtosecond laser pulses with a circular (σ^+, σ^-) or linear (π) polarization. The initial temperature of the sample was below the temperature of the phase transition T_1. The light and dark regions correspond to opposite signs of the projection of the magnetization onto the normal to the sample (see images in the lower part of the figure). The gray regions correspond to the orientation of the magnetization in the plane of the sample. (b) Schematic representation of changes in the profile of the energy of magnetic anisotropy (from top to bottom) during the excitation; at the moment of the phase transition; and at the final state. Also shown are the positions of the magnetization at the corresponding moments of time [14].

reorientation phase transitions using circularly polarized laser pulses is of interest.

This possibility was tested experimentally in [14]. Using the method of single-pulse time-resolved magneto-optical microscopy (see Section 3.2), the authors investigated the time–spatial dynamics of magnetization in the solid-solution orthoferrite $(Sm_{0.5}Pr_{0.5})FeO_3$ upon its excitation by single circularly or linearly polarized pulses. The excitation and corresponding magneto-optical imaging were realized in the absence of an external magnetic field. The results of this experiment are presented in Fig. 6a, which show that a single femtosecond laser pulse induces a $\Gamma_2 \rightarrow \Gamma_{24}$ phase transition, occurring several picoseconds after the excitation, in complete agreement with the data of previous experiments. In the case of excitation by a linearly polarized pulse, the formation of a multidomain structure was clearly observed after 5 ps, as could be expected. However, upon excitation by pulses with right-hand (σ^+) or left-hand (σ^-) circular polarization, a single domain was formed with the direction of the magnetization unambiguously determined by the polarization of the laser pulse. Thus, in this experiment, the possibility of controlling the laser-induced reorientation phase transition was demonstrated for the first time.

The mechanism of such control is illustrated in Fig. 6b. On the one hand, the action of a laser pulse leads to the appearance of a phase transition, as was discussed in Section 4.2. This process is shown as a change in the profile of the anisotropy energy. On the other hand, under the influence of a circularly polarized pulse, an ultrafast inverse Faraday effect is observed (see Section 4), which leads to the impulsive excitation of the magnetization precession. In Fig. 6b, this is shown as a deviation of the magnetization direction from the equilibrium position at the time of excitation. Thus, at the moment when a laser-induced phase transition takes place, the degeneracy between the new minima is removed dynamically due to the precession of the magnetization. It should be noted that, according to the results of previous experiments, the typical deviation of the magnetization from the equilibrium position due to ultrafast inverse magneto-optical effects is equal to a few fractions of a degree or to a few degrees. However, even such a weak deviation proves to be sufficient to dynamically remove the degeneracy between the minima in the Γ_{24} phase.

Figure 6 illustrates a scenario, according to which the time delay at which the phase transition occurs somewhat shorter than half the period of the precession of magnetization excited by the ultrafast inverse Faraday effect. As was noted above, the time delay at which the phase transition occurs is determined by the initial temperature of the sample and by the energy of the exciting pulse [75]. For example, by decreasing the initial temperature of the sample, this time can be increased in such a way that it would become longer than half the period of precession. Accordingly, the picture depicted in Fig. 6b will change, and the system will proceed to another minimum of the anisotropy energy. Thus, by changing the initial temperature of the sample or the energy density in the pump pulse, it is possible to govern the direction of the magnetization in the laser-induced domain even upon excitation of the sample by single pulses with the same circular polarization. This was demonstrated experimentally in [14].

It should be noted that in this experiment control of the magnetization was possible due to the ultrafast inverse Faraday effect, since it is the dominating opto-magnetic effect in this material. In general, it can be expected that a similar mechanism for controlling the reorientation phase transition can also be realized for a linearly polarized pulse in a medium where the ultrafast inverse Cotton–Mouton effect prevails.

As was discussed in Section 4.2, the microscopic mechanism of ultrafast opto-magnetic effects is impulsive stimulated Raman scattering. From this viewpoint, the excitation of coherent magnons due to the ultrafast inverse Faraday effect, which forms the basis of the control of the reorientation phase transition, is analogous to the excitation of coherent phonons [63, 64]. In this connection, it is possible to expect that a similar scenario can be implemented for controlling the structural or ferroelectric transitions, in which the dynamic lifting the degeneracy will be achieved due to the impulsive polarization-dependent excitation of coherent phonons.

To conclude the consideration of the control of ultrafast reorientation transitions, it should be noted that the scenario examined above is effective in the case of second-order transitions. The control of laser-induced first-order transitions is also possible using ultrafast opto-magnetic phenomena, but it has specific features associated with the type of reorientation transition, as is shown in [34].

6. Conclusions

The purpose of this article was to show that ultrafast opto-magnetic phenomena are efficient tools for the excitation and control of the dynamics of a spin system. In these phenomena, a femtosecond laser pulse with a certain polarization acts on a magnetically ordered medium as a short pulse of an effective field. The direction and strength of this field are determined both by the parameters of the laser pulse (its intensity, spectral composition, duration, and polarization) and by the properties of the medium itself, first of all, by its magneto-optical susceptibility and by the type of magnetic ordering.

The results of studies of ultrafast opto-magnetic phenomena obtained in the last decade by different groups of researchers have shown that they are an efficient method for the excitation of magnetic dynamics in media with various types of magnetic ordering, from ferromagnets to chiral magnets. In this article, examples of several ultrafast opto-magnetic phenomena and their specific features in multi-sublattice magnetic media have been considered.

The most important feature of ultrafast opto-magnetic phenomena is the possibility to impulsively excite a spin system without energy absorption. This distinguishes them from the majority of other mechanisms of excitation of magnetic dynamics by femtosecond laser pulses. Undoubtedly, ultrafast opto-magnetic phenomena are an efficient mechanism for direct selective excitation and control of spin dynamics in various media, including optically transparent media.

The phenomenological theory of ultrafast opto-magnetic phenomena makes it possible to draw an analogy between these phenomena and magneto-optical effects. In this article, we considered phenomena that have been most studied to date, namely, the inverse Faraday and Cotton–Mouton effects, which make it possible to excite the coherent precession of magnetization and to control it by circularly and linearly polarized laser pulses. These phenomena are based on 'impulsive stimulated Raman scattering' (ISRS). However, the range of magneto-optical effects is, undoubtedly, considerably wider than that given in the table in Section 2. Thus, magnetic refraction, connected with the isotropic exchange interaction, manifests itself in the quadratic dependence of the refractive index on the magnetization of the medium [79–81]. The ultrafast inverse magnetorefractive effect can be used to change the exchange interaction and control it. Recently, this possibility was investigated experimentally in a number of weak ferromagnets — iron oxides [33]. It was shown that a femtosecond laser pulse acts as an effective field owing to the laser-induced change in the effective fields of isotropic and anisotropic exchange. Table 1, given in Section 2, can be extended with at least one additional ultrafast opto-magnetic phenomenon, namely, the inverse magnetorefractive effect, which offers the possibility of directly controlling exchange interaction on a femtosecond time scale.

It should be noted that recently Belotelov and Zvezdin [82] predicted, based on a phenomenological consideration, the inverse equatorial Kerr effect, which consists of the appearance of an effective magnetic field induced by laser pulses with a *p*-polarization and a nonzero angle of incidence. An important difference between the inverse equatorial Kerr effect and the inverse Faraday [20] and Cotton–Mouton [21] effects is that the necessary condition for its appearance is the presence of a nonzero imaginary part of the refractive index; i.e., this effect can be observed only in an absorbing medium. According to our information, no data on the experimental observation of this effect is available so far.

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