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Fundamentals and perspectives of ultrafast photoferroic recording

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ABSTRACT

The ability to switch ferroics (ferro-, ferri-, antiferromagnets, ferroelectrics, multiferroics) between two stable bit states is one of the keystones of modern data storage technology. Due to many new ideas, originating from fundamental research during the last 50 years, this technology has developed in a breath-taking fashion. Finding a conceptually new way to control ferroic state of a medium with the lowest possible production of heat and at the fastest possible timescale is a new challenge in fundamental condensed matter research. Controlling ferroic state of media by light is a promising approach to this problem. Photomagnetism and photoferroelectricity have long been intriguing and the development of femtosecond laser sources made this approach even more appealing. Laser pulse is the shortest stimulus in contemporary experimental physics of condensed matter. While commercial lasers are able to produce pulses with duration of the order of tens of femtosecond, advanced laser sources can generate intense pulses of light even at the sub-femtosecond timescale. Seeking understanding a response of magnetically-ordered media to ultrashort excitation led to foundation of new research field of ultrafast magnetism, discoveries of all-optical magnetic switching in various metallic and dielectric materials. Despite obvious analogies between magnetically-ordered and ferroelectric materials, the issue of the ultrafast switching of the order parameter in the latter class of ferroics has been given very little attention. This raises an obvious question about the possibility of optical switching of the spontaneous polarization in ferroelectrics and the prospects of information recording in ferroelectrics by means of light. Here we briefly review the main findings of earlier studies of optical control of spontaneous magnetization and polarization, highlight recent developments of ultrafast magnetism and magnetic recording with femtosecond laser pulses, and discuss a new field of ultrafast ferroelectricity. Analyzing the literature, we derive the most promising strategies for optical recording in ferroic media and speculate about applicability of the strategy proven to be efficient in magnetically-ordered media, to ferroelectrics and multiferroics.

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1. Introduction

A crystal is provisionally referred to as being "ferroic" [1] when it has two or more orientational states of order parameter in the absence of magnetic field, electric field, and mechanical stress and can shift from one to another of these states by means of one or combination of several external stimuli. Due to this fundamental property ferroics is a natural, cheap and reliable mean to store information. Ferroelectric, antiferroelectric, ferro- and antiferromagnetic materials are particular examples of ferroics, and have already found their applications in memory devices [2,3]. The main principle of these devices is based on the unique ability of ferroics to be switched between two energetically equivalent, stable (bit) states. Although there are many other alternative ways to store information, and flash memory or solid state drives are outperforming recording on ferroics in some of applications, magnetic memories will remain competitive in a foreseeable feature. Moreover, amount of data to be stored increases from 30 to 40 percent per year [4], and one can anticipate that the demand for faster, denser, more energy efficient and cheaper ferroic data storage will be high for many more years to come. Even a magnetic tape, which one can rarely see outside of black-and-white movies, has in fact never gone away [4]. For instance, in 2015 the Information Storage Industry Consortium, an organization that includes HP Consortium, IBM, Oracle and Quantum, along with a number of academic research group, released what it called "The International Magnetic Tape Storage Roadmap" [http://www.insic.org/].

The reorientation of the ferroic order parameter, in general, and recording, in particular, implies that a corresponding external field, or any other stimulus that acts as an effective field, assists the order parameter to overcome the potential barrier ΔU separating two bit states. In order to reliably store information on ferroic medium, the height of this barrier should satisfy the criterion to prevent thermal fluctuations between the bit states. In magnetically-ordered media the height of the barrier is defined by the energy of magnetic anisotropy $\Delta U = KV$, where K is the anisotropy constant and V is the bit volume. Accordingly, the bit stability is defined by the superparamagnetic limit [5], which yields the condition $\Delta U \geq 60 k_B T$ for ensuring at least 10 years of information storage (k_B is the Boltzmann constant, T is the temperature).

Aiming at increasing areal density of data storage, industries eventually approached fundamental limits known as trilemma of magnetic recording: bit size-stability-writability. To ensure reliable storage of data, decreasing the bit volume V one must increase the energy of magnetic anisotropy KV and thus increase the coercive field of the recording medium H_c. Upon increasing the coercive field, it becomes increasingly difficult to write a bit, because writing requires a magnetic field larger than the coercive field $H > H_c$. It was proposed that the trilemma could be resolved with the help of light. The coercivity of many materials is temperature dependent. If the temperature of a magnetized object is temporarily raised above the Curie point, its coercivity reduces. State-of-the-art Heat Assisted Magnetic Recording (HAMR) uses this property of magnetic materials and reduces the coercivity using laser-induced heating. In this way, bits are recorded in a much smaller area and using a much smaller magnetic field than would otherwise be possible. Apart from heating by light, an electromagnetic radiation at microwave frequencies is also a mean to assist switching in magnetic media and the stimulus in microwave-assisted magnetic recording (MAMR).

In ferroelectrics, in contrast, a superparaelectric limit is currently a lesser problem. For instance, estimates for a unit cell of a ferroelectric $PbTiO_3$ give the barrier height ΔU being 4–8 times larger than $k_B T$, suggesting that even a few single cells can act as a bit [6]. However, minimum bit size is defined in reality by the size-induced phase transition to a paraelectric state due to depolarization effect [7,8]. This along with a problem of a polarization fatigue [9] has held back the development of ferroelectric memories [2]. Nevertheless, ferroelectric memory is still under active development fueled by progress in fabrication of thin and ultrathin films supporting ferroelectricity, demonstration of fatigue-free ferroelectrics, and nondestructive read-out of local polarization [10–13].

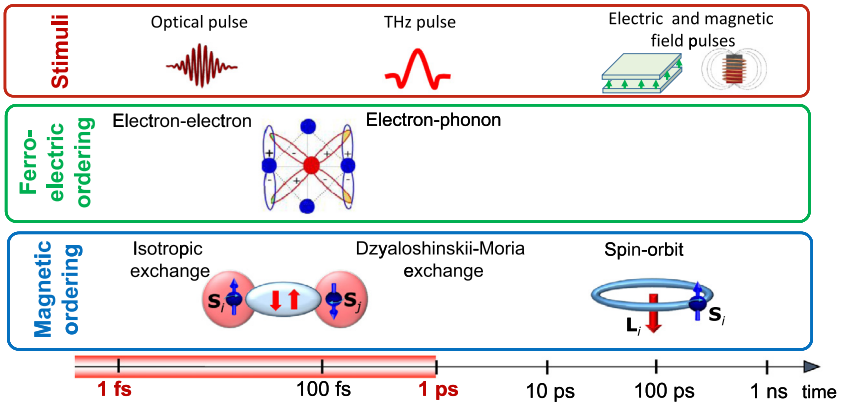

Fig. 1. Pictorial diagram of the main interactions and relevant timescale responsible for the magnetic and ferroelectric ordering. Coupling parameters responsible for interactions between spins, electrons, and lattice are functions of time at the timescale comparable to duration of optical, IR or THz laser pulses. As a result, dynamics of ferroics at the timescale below $\sim$ 1 ps is poorly understood.

It is well established from the studies of conventional magnetization dynamics, that the most efficient and intrinsically fast way to switch a ferroic order parameter is to coherently excite a specific mode, which transforms according to the same irreducible representation as the order parameter. For instance, magnetization of a ferromagnet can be switched via coherent excitation of homogeneous spin precession, or a ferromagnetic resonance mode [14]. Reorientation of antiferromagnetic vector in an antiferromagnet relies on an excitation of one of antiferromagnetic resonance modes, which is a soft mode in the particular material [15]. In ferroelectrics of displacive and order–disorder types [16,17], it is suggested that coherent excitation of the soft phonon mode can result in switching of the spontaneous polarization [18]. Beyond the conventional classification of ferroelectricity, there is a class of electronic ferroelectrics where electronic degrees of freedom and electronic correlations are directly responsible for spontaneous polarization [19,20]. It is suggested in literature, that, if ferroelectricity is electronic, there should be a ferroelectric resonance [19,20]. In a close analogy with ferromagnets, one may expect that the switching between two bit states of electronic ferroelectric can be launched via excitation of this resonance.

How can a femtosecond laser pulse switch the electric and magnetic order parameter between stable states? Can the switching be also femtosecond fast? Let us consider a proposal for optical control of spins in a magnetically ordered medium as an example. The energy of light-matter interaction is mainly given by the energy of interaction of the electric field of light with electric charges i.e. electric dipoles. Direct interaction of the magnetic field of light with spins is known to be much weaker and can be neglected at least in the visible spectral range. Although the effect of the electric field of light on electric dipoles is the strongest perturbation in light-matter interaction, it conserves electron spin. In order to understand how interaction of the electric field of light with an electron, for instance, can affect electron spin, we note that spin corresponds to angular momentum, and changing the direction of a spin $\mathbf{S}$ should satisfy the conservation law

$$\frac{d\mathbf{S}}{dt} = -\gamma \mathbf{S} \times \mathbf{H}_{\text{eff}}, \tag{1}$$

where $\mathbf{H}_{\text{eff}}$ is the effective magnetic field and $\gamma = 28$ GHz/Tesla is the gyromagnetic ratio for the electron without spin-orbit interaction. Thus, the stronger the effective magnetic field, the faster the spin reorients. The effective field $\mathbf{H}_{\text{eff}}$ can be obtained from the electron energy U as $\mu_0 \mathbf{H}_{\text{eff}} = -dU/d\mathbf{S}$, if the spin-dependent part of U is not zero. In magnetic media the effective magnetic fields originating from the spin–orbit and the exchange interactions reach 10 T and 100–1000 T, respectively. These fields are much larger than those generated by electromagnets in conventional experiments and correspond to the Larmor precession with periods of 3 ps and 300–30 fs, respectively. Trying to understand how to harness these fields for optical control of magnetization, one unavoidably faces probably the most fundamental problem of ultrafast magnetism: In a strongly non-equilibrium state, prepared by ultrafast laser excitation, the spin–orbit interaction, the exchange coupling between the spins as well as the spin dynamics driven by these interactions are all time-dependent and, as a result, poorly understood. In ferroelectrics, time-dependent electron–electron and electron–phonon interactions in the laser-excited state can be employed for changing spontaneous polarization. In Fig. 1 we sketch the main interactions and relevant timescales responsible for the magnetic and ferroelectric ordering and compare them to durations of laser pulses in optical and THz ranges.

Thermodynamically there are a lot of similarities between media possessing spontaneous magnetization and spontaneous polarization, and both types of ferroic media can be adequately discussed in terms of corresponding order parameters, domain structure and critical temperature. It is interesting to examine to which extent this analogy holds in the case of ferroic recording, especially at an ultrafast timescale. Switching order parameter in magnetically-ordered media is different from switching in ferroelectrics due to totally different physical origin of the orders. Magnetic order originates

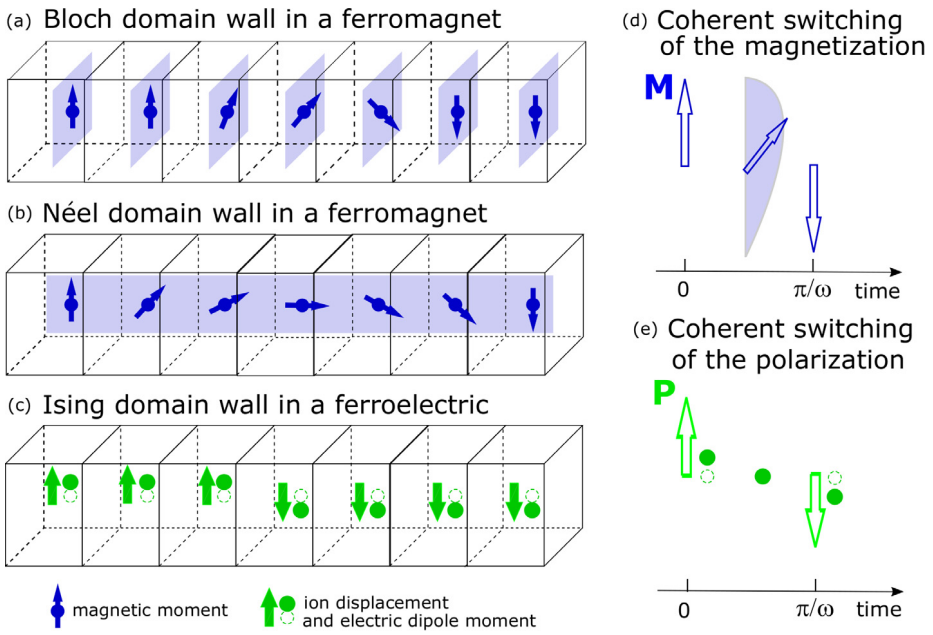

Fig. 2. Types of domain walls between 180°-domains in magnetically-ordered and ferroelectric materials. Panels (a,b) show the Bloch (a) and the Néel (b) walls in a ferromagnet, and (c) — the Ising wall in an ferroelectric. Pathway for the fast coherent switching of the magnetization **M** (d), and of the polarization **P** (e) via excitation of a corresponding large-amplitude magnon or phonon mode mode with a frequency ω.

from an isotropic short-range exchange interaction between spins. The magnetic anisotropy, i.e. the direction in which the spins are aligned, is typically defined by a weaker spin–orbit interaction. The spontaneous magnetization thus can be reversed without changing its absolute value via rotation of local magnetic moments. In displacive and order–disorder ferroelectrics the order parameter is intrinsically linked to particular crystallographic directions. Contrary to magnets, the order and the anisotropy of a ferroelectric are defined by the very same interactions. Therefore, the reversal of a spontaneous ferroelectric polarization proceeds though the linear reversal of local electric dipole moments.

The fundamental difference between reorientation of order parameter in (anti-)ferromagnets and (anti-)ferroelectrics can be demonstrated using the examples of domain walls existing in these ferroic media. In magnetically-ordered materials spatial reorientation of the local magnetic moments between two domains results in a wall of a Bloch- or Néel-type [21]. In both cases, the local magnetic moments continuously rotate in space. In the most common case, the energy of the exchange interaction is much larger than the energy of magnetic anisotropy and the rotation of the magnetic moments occurs on a scale up to 10–100 nanometers (see Fig. 2(a,b)). In ferroelectrics, domain walls are predominantly of Ising-type [22–25]. The walls can be atomically-thin and include an abrupt change of magnitude of local electric dipole moments without a change of their orientation (Fig. 2(c)). From this difference in space it can be readily seen that the switching of the order parameters in time for these two types of ferroics must be also different (Fig. 2(d,e)). Moreover, the difference in the origins of ferroelectric polarization and magnetization results in a distinct behavior of these ferroics in the vicinity of their Curie temperatures. In particular, mean-field field approximation generally fails to describe magnets near an order–disorder phase transition, but appears to be much more satisfactory for ferroelectrics [26–28]. Ultrafast heating of electrons, lattice and spins with eventual crossing the Curie point is one of the basic concepts in physics of ultrafast laser-induced demagnetization [29] and heat-assisted magnetic recording [30]. Therefore it is an interesting question if similar concepts can be applied for optical control of ferroelectrics.

An important class of ferroics is comprised by those possessing two or more order parameters in the same phase. In particular, in magnetoelectric multiferroics magnetic and ferroelectric order parameters coexist within a certain temperature range and are mutually coupled [31–33]. Can one anticipate a switching of ferroelectric polarization mediated by an optical switching of magnetic order parameter and vice versa in a multiferroic medium?

To summarize, optical switching of order parameter in ferroics raises several fundamentally intriguing questions. In this review we summarize the main findings of earlier studies of optical switching of ferroics, and discuss the most recent advances in this field achieved using femtosecond laser pulses. Analyzing the literature, we derive the most promising strategies of ultrafast optical recording on ferroic media. One of our main goals is to draw parallels between recordings on different types of ferroics. As ferroelectric order and good electric conductivity are mutually exclusive, here we discuss laser-driven processes in metals only briefly, and refer a reader to extended reviews dealing with this subject [34–37]. The review mainly focuses on semiconductors and dielectrics. Based on this discussion we speculate about applicability of the strategies for optical control of order parameters in ferroelectrics and multiferroics. The review is organized as

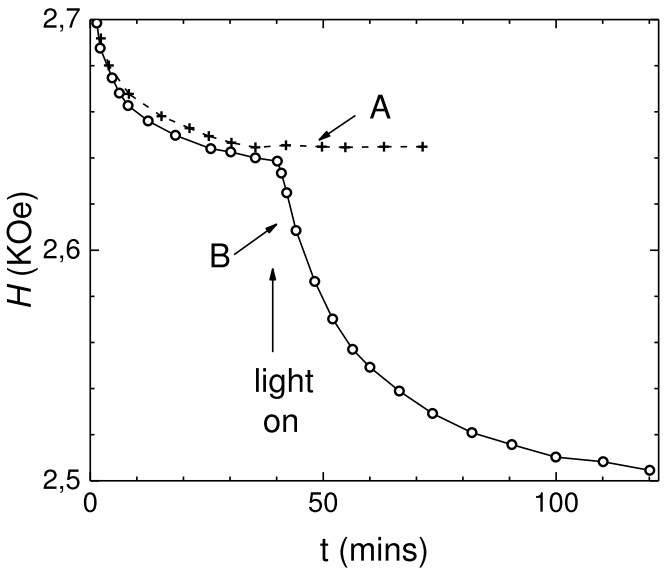

Fig. 3. Effect of unpolarized light on the magnetic anisotropy of $Y_3Fe_{4.9}Si_{0.1}O_{12}$ detected by measuring the field of ferromagnetic resonance as a function of time [45]. Curve A shows the dynamics of the resonance field without any effect of light. Curve B shows that photoexcitation launches further changes of the field on a timescale of hours. To the best of our knowledge this is the first experimental observation of effect of light on magnetic properties.
Source: The figure is taken from Ref. [45].

follows: after introducing in Section 1 the problem of ultrafast photoferroic switching, we review in Section 2 earlier studies of optical control of magnetically-ordered and ferroelectric materials. Section 3 is devoted to thermodynamics of photoferroic switching. Section 4 describes microscopic mechanisms of optical control of ferroics. Section 5 discusses the routes for photoferroic recording, and Section 6 speculates about perspectives of photoferroics, including multiferroics, for research and technology.

2. Earlier studies of photoferroics and photoferroic recording

To the best of our knowledge, the term "photomagnetism" was for the first time mentioned in literature back in 1934 [38,39]. The paper reported about dependence of photocurrents in paramagnetic Cu_2O upon an applied magnetic field and hence had very little to do with control of magnetic order. The discovery of the Ruderman–Kittel–Kasuya–Yosida (RKKY) mechanism of the exchange interaction mediated by conduction electrons [40–42] naturally led to speculations about optical control of magnetism in magnetic semiconductors by injecting photocarriers [43,44]. Although photoinduced changes of magnetic anisotropy [45–48], magnetic permeability [49–52], mobility of domain walls [53,54], magnetostriction [55], and even exchange interaction [56] were soon observed experimentally, the effects of light on magnetic properties appeared to be even more pronounced in dielectrics than in semiconductors.

Magnetic dielectric Si-doped yttrium iron garnet $Y_3Fe_5O_{12}$ (YIG) became one of the most popular compounds in these studies. For instance, [45] reports about an effect of near-infrared light in the range of wavelength from 800 nm to 2200 nm on the strength of magnetic anisotropy of the material. The light was unpolarized and the induced effect was observed at low temperature (20 K) on a timescale of hours (Fig. 3). In the same material, laser illumination could also irreversibly change magnetic permeability and coercive field [49]. After the irradiation in order to restore the initial "dark" state, the sample had to be heated and cooled down in darkness. The effect was developing on a timescale of minutes. However, experiments with pulsed excitation showed that pulses as short as 10^{-4} s or less are also able to cause the changes.

Similar photoinduced change of magnetic permeability were also reported for a magnetic semiconductor Ga-doped spinel $CdCr_2Se_4$ [57]. In experiments the material was irradiated with unpolarized white light of a fluence of 10^{-2} W/cm^2 at temperature of 77 K, which is below the Curie temperature $T_C = 130$ K of the compound. Fig. 4 shows that after light is switched on the magnetic permeability rapidly drops on a timescale of seconds. After the light is switched off, the permeability recovers to the initial state on a timescale of minutes. The same experiments performed at 4.2 K yielded similar results with much slower recovery process. The observed light-induced modification of the magnetic properties (Figs. 3, 4) were explained as a result of photo-activation of Fe^{2+} ions at tetrahedrally and octahedrally cites in the garnet [49], or Ga^{3+} ions in the spinel [57]. As we discuss below in this Section and in Section 5, such centers play a crucial role in photomagnetism at both long and ultrashort timescales.

Soon after the first experiments with unpolarized light it was realized that light polarization provides an additional and very important degree of freedom. For instance, [47] reports about experimental studies of the effect of polarized

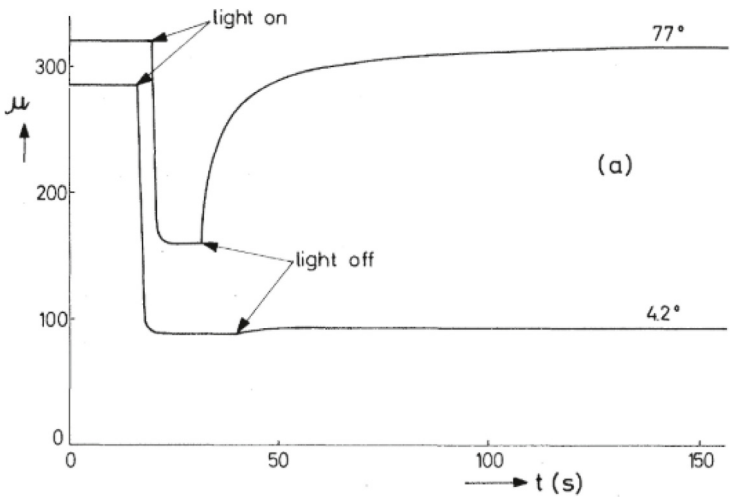

Fig. 4. Effect of unpolarized light on magnetic permeability in Ga-doped $CdCr_2Se_4$ measured at two different temperatures of 77 and 4.2 K [57]. *Source:* The figure is taken from Ref. [57].

light on the magnetic anisotropy of Si-doped YIG. Using a torque magnetometry, it was shown that light with different linear polarizations may have different effects on the magnetic anisotropy of the sample.

In [52] it was also shown that the magnetic permeability of Pb-doped YIG could be changed by illumination with low intensity X-ray radiation. Photomagnetic effects were reported for many other magnetic dielectrics such as $FeBO_3$ [58], Ru-doped $Li_{0.5}Fe_{2.5}O_4$ spinel [51], Ni-Zn-Co ferrite [50], $EuCrO_3$ [56], and magnetic semiconductor EuS [59].

While the first reports about effects of light on magnetic properties of media were found interesting mainly from fundamental point of view, soon it was realized that the photomagnetic effect can be employed to control magnetic domains in the materials and thus to record information. To the best of our knowledge, the first successful demonstration of photomagnetic recording with polarized light was reported in 1981 [60]. Using $Y_3Fe_{4.96}Si_{0.04}O_{12}$ (110) single crystal plate as a recording medium, it was possible to write information with a CW laser beam at the wavelength of 1.15 μm. The crystal had two easy axes of magnetic anisotropy in the sample plane. Without any magnetic field applied the magnetization of the plate could be in one of four metastable states along the easy axes. Excitation of the medium with a linearly polarized light resulted in a reorientation of the magnetization to the state with a smallest angle between the magnetization and the polarization of light (Fig. 5). Changing the polarization of light, it was possible to write and erase information. The similar phenomenon was demonstrated in Co-doped YIG and employed for rearranging the domain pattern [61]. Underlying mechanism of the photomagnetic writing is due to doping with Si^{4+} and Co^{2+} ions which changes the magnetic anisotropy of the garnet. It was proposed that absorption of linearly polarized light results in a charge transfer between the dopants and Fe^{3+} ions, altering the anisotropy. Selection rules of these charge transfer transitions show that the effect must depend on the polarization of light. In Section 5 we discuss how the photomagnetic effect at the ultrafast timescale can be used to realize a "cold", i.e. nearly non-dissipative, writing of information in a garnet film.

The idea of optical control of ferroelectricity emerged in a way similar to photomagnetism. The discovery of ferroelectricity in $A_VB_{VI}C_{VII}$ semiconductors, such as SbSI [62,63], led to theoretical proposals to control their ferroelectric properties by photo-injection of free electrons from the valance to the conduction band [64]. Soon the predictions were confirmed experimentally in [65] by showing that optical illumination can shift the Curie temperature of SbSI (Fig. 6). An effect of photostriction on ferroelectricity was also reported in the same compound [66]. However, such a sensitivity to photo-excitation was found to be present in other types of ferroelectrics as well, and is not limited to the specific group of $A_VB_{VI}C_{VII}$ semiconductors. In [67], for instance, a similar effect of light illumination on the Curie temperature was reported for a dielectric ferroelectric $BaTiO_3$ doped with different transition metal ions.

In a certain analogy to photomagnetism based on photoexcitation of dopants, changing spontaneous polarization via photoionization and bleaching of impurity centers in a ferroelectric host can also be considered [68]. If an impurity in a ferroelectric has an electric dipole moment, excitation of the impurities should lead to a redistribution of charges and a change of overall spontaneous polarization. It was shown for $LiNbO_3$ single crystals doped with 0.26% Cr^{3+} that excitation with light resonant with $d-d$ transitions in Cr^{3+} yields a change of the dipole moment of the impurity [69]. The role of impurity centers was verified by showing that no such effect was present in undoped $LiNbO_3$.

The possibility of light-assisted recording on lanthanum-doped lead zirconium titanate ceramics was reported in [70]. The mechanism of the recording was based on combined action of electric field and light. Photo-assisted switching of the spontaneous electric polarization in important ferroelectric $LiNbO_3$ was reviewed in [71].

From all these examples it is seen that light is able to affect ferroic order, and one can even write information using ferroics as a recording medium. The first reviews about earlier studies of optical control of magnetism and ferroelectricity

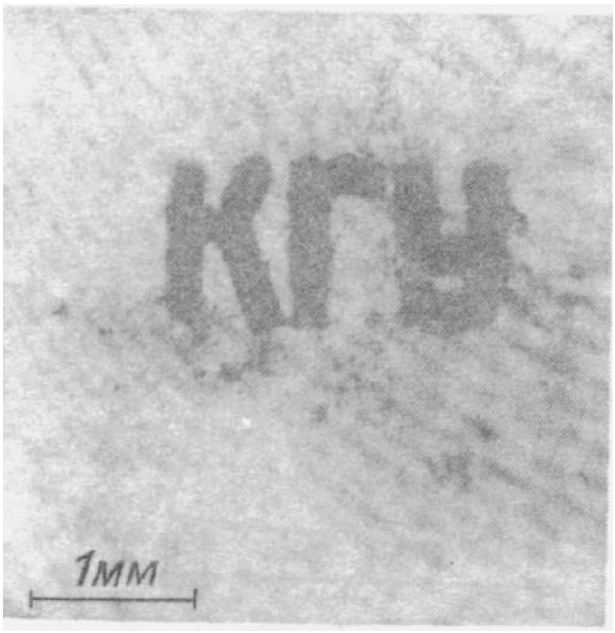

Fig. 5. Photomagnetic recording on $Y_3Fe_{4.96}Si_{0.04}O_{12}$ [60]. The dark letters are formed by the domains with magnetization switched from its original direction (white area) by linearly polarized light. To the best of our knowledge this is the first photomagnetic recording ever reported in literature.
Source: The figure is taken from Ref. [60].

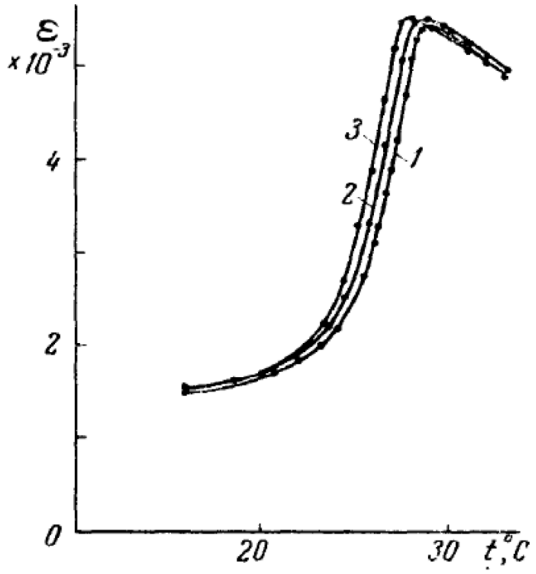

Fig. 6. Experimental demonstration of a light-induced shift of the Curie point in the ferroelectric semiconductor SbSI due to photocarriers [65]. Dielectric permittivity at 0.1 GHz was measured as a function of temperature in the dark (curve 1), and upon illumination (curves 2, 3) corresponding to different concentrations of photocarriers concentration.
Source: The figure is taken from Ref. [65].

were published already 30 years ago [68,72]. With the development of femtosecond laser sources this research topic acquired a new boost. Laser pulse is among the shortest stimuli in contemporary experimental physics of condensed matter. Therefore photoferroics in combination with femtosecond light sources opened up intriguing opportunities for both fundamental studies of ultimately fast processes in spontaneously ordered states of matter and the fastest ever recording for future technologies.

3. Thermodynamics of photoferroic switching

3.1. Order parameters and corresponding fields

Thermodynamic treatment of ferroics starts with introduction of order parameter. For instance, in the case of ferromagnets the order parameter is the magnetization

$$\mathbf{M} = \sum_k \frac{\mathbf{S}_k}{V}, \tag{2}$$

where $\mathbf{S}_k$ is the magnetic moment of kth ion constituting the lattice of the ferromagnet. In the case of antiferromagnets which consist of two antiferromagnetically coupled magnetic sublattices, the order parameter is the antiferromagnetic, or Néel, vector

$$\mathbf{L} = \mathbf{M}_1 - \mathbf{M}_2, \tag{3}$$

where the magnetizations $\mathbf{M}_1$ and $\mathbf{M}_2$ are defined for each of the sublattices. In the case of ferroelectric the order parameter is the electric polarization $\mathbf{P}$

$$\mathbf{P} = \sum_k \frac{\mathbf{d}_k}{V}, \tag{4}$$

where $\mathbf{d}_k$ is the dipole moment of the kth elementary cell of the ferroelectric. Similarly to antiferromagnets, a vector describing antiferroelectric state can be also introduced as

$$\mathbf{N} = \mathbf{P}_1 - \mathbf{P}_2. \tag{5}$$

According to the first law of thermodynamics for a change of the internal energy of a system we have $dU = dW + dQ$, where dQ is the heat supplied to the system, and dW is the work done on the system. The Clausius' theorem states that $dQ \leq TdS$, where T is the temperature and S is the entropy. It means that $dU \leq dW + TdS$. The equation implies that for an isolated system, the internal energy can only decrease, i.e. $dU \leq 0$. It means that after the system reaches the thermodynamic equilibrium, the internal energy is at its minimum corresponding to $\partial U/\partial \mathbf{\Lambda} = 0$, where $\mathbf{\Lambda}$ states for an order parameter.

The work performed on a medium of unit volume by an external magnetic field $\mathbf{H}$ is given as $dW = \mu_0 \mathbf{H} d\mathbf{M}$, where μ_0 is the fundamental constant. Assume that a medium is brought out of the equilibrium by tilting the magnetization from the orientation corresponding to the minimum of the internal energy. The process of relaxation of the system towards the thermodynamic equilibrium is practically the motion of the magnetization in the effective magnetic field $\mathbf{H}_M$

$$\mu_0 \mathbf{H}_M = -\frac{\partial U}{\partial \mathbf{M}}. \tag{6}$$

Similar field can be also defined for ferroelectric using the expression for the work performed by electric field $dW = \mathbf{E} d\mathbf{P}$

$$\mathbf{E}_P = -\frac{\partial U}{\partial \mathbf{P}}, \tag{7}$$

Therefore, if a light pulse changes the internal energy such that $\partial U/\partial \mathbf{M} \neq 0$, it may trigger magnetization dynamics and thus can, in principle, steer the magnetization into another stable bit-state. Similarly, light can trigger dynamics of $\mathbf{P}$, if light changes the internal energy such that $\partial U/\partial \mathbf{P} \neq 0$. Can light generate the effective fields $\mathbf{H}_M$, $\mathbf{E}_P$?

3.2. Coupling of electromagnetic field of light to an order parameter

Light is an electromagnetic wave. The electric $\mathbf{E}^\omega$ and magnetic $\mathbf{H}^\omega$ fields of an electromagnetic wave can directly couple to the electric polarization $\mathbf{P}$ and the magnetization $\mathbf{M}$, respectively. The work performed by the electromagnetic field on a medium in a general case should be written as $dW = \mathbf{E}^\omega d\mathbf{P} + \mu_0 \mathbf{H}^\omega d\mathbf{M}$. Although it is less obvious, but magnetic field of an electromagnetic wave can also couple to the antiferromagnetic Néel vector. In order to derive a corresponding expressions, one has to analyze spin dynamics in antiferromagnets induced by a time-dependent magnetic field, as discussed in Section 3.4.

The strongest force in the interaction of light and matter is the action of electric field on electric dipoles. The electric field of light can also couple to the magnetization, if one takes into account effects of the second order with respect to the electric field of light. The work performed by the electric field of light $\mathbf{E}^\omega$ on a medium in the electric dipole approximation is $dW = \mathbf{E}^\omega d\mathbf{P}^\omega$, where $d\mathbf{P}^\omega$ is the vector of electric polarization induced by the electric field of light. For simplicity we assume that the electric polarization is a linear function of the electric field.

$$P_k^\omega = \epsilon_0 \chi_{kl} E_l^\omega, \tag{8}$$

where ϵ_0 is the fundamental constant and χ_{kl} is the optical susceptibility tensor. From $W = \int dW = \frac{1}{2}\epsilon_0 \chi_{kl} E_k^{\omega *} E_l^{\omega} + \text{const}$ one finds the expression for the internal energy of a medium irradiated by light

$$U = U_0 + \frac{1}{2}\epsilon_0 \chi_{kl} E_k^{\omega *} E_l^{\omega}, \tag{9}$$

where U_0 is the part of the internal energy that does not depend on the electric field of light. If the medium was in a thermodynamic equilibrium before the action of light, it means that $\partial U_0/\partial \mathbf{M} = 0$ and $\partial U_0/\partial \mathbf{P} = 0$. It is also seen that light can generate an effective magnetic $\mathbf{H}_M$ or electric $\mathbf{E}_P$ field, if the optical susceptibility is a function of $\mathbf{M}$ or $\mathbf{P}$, respectively, and $\partial \chi_{kl}/\partial \mathbf{M} \neq 0$ or $\partial \chi_{kl}/\partial \mathbf{P} \neq 0$.

In order to reveal how the optical susceptibility depends on the magnetization, it is convenient to assume that there is no irreversible energy transfer from light to medium during light-matter interaction, i.e. the light is only refracted, but not absorbed. This assumption is called approximation of no-dissipations. In theory it means that electromagnetic wave excites electric dipoles at a non-resonant frequency [73,74]. It can be shown that if dissipations are absent, the susceptibility is a Hermitian tensor $\chi_{kl} = \chi_{lk}^*$ [75]. Moreover, it also means that the entropy during light-matter interaction does not change $dS = 0$ and two directions of time must be equivalent. Applying operation of time-reversal to Eq. (9) one obtains

$$\chi_{kl}(M) = \chi_{lk}(-M), \tag{10}$$

Further it is convenient to express the optical susceptibility tensor as a sum of symmetric and antisymmetric parts $\chi_{kl} = \chi_{kl}^a + \chi_{kl}^s$, where $\chi_{kl}^a = -\chi_{lk}^a$ and $\chi_{kl}^s = \chi_{lk}^s$. Based on the definition and Eq. (10) the following must hold $\chi_{kl}^a(M) = \chi_{lk}^a(-M) = -\chi_{kl}^a(-M)$ and $\chi_{kl}^s(M) = \chi_{lk}^s(-M) = \chi_{kl}^s(-M)$. Therefore, if the parts of the optical susceptibility tensor are expressed in terms of Taylor series, a half of the terms can be omitted:

$$\chi_{kl}(M)^a = i\alpha'_{klm} M_m + i\alpha''_{klmnq} M_m M_n M_q + \cdots \tag{11}$$

$$\chi_{kl}(M)^s = \chi_{kl}(0)^s + \beta'_{klmn} M_m M_n + \cdots \tag{12}$$

Here α'_{klm}, α''_{klmnq}, β'_{klmn} are phenomenological tensors whose properties are dictated by the space–time symmetry. Limiting the expansion to the second order with respect to the magnetization, one obtains the expression for the internal energy of the medium

$$U = U_0 + \frac{1}{2}\epsilon_0 \chi_{kl}^s(0) E_k^* E_l + i\frac{1}{2}\epsilon_0 \alpha'_{klm} E_k^* E_l M_m + \frac{1}{2}\epsilon_0 \beta'_{klmn} E_k^* E_l M_m M_n. \tag{13}$$

The term of the internal energy proportional to $\mathbf{M}$ renders the direct magneto-optical Faraday effect also known as magnetic circular birefringence. The term proportional to $\mathbf{MM}$ renders the direct magneto-optical Cotton–Mouton (Voigt) effect, or magnetic linear birefringence. In both cases, the magnetization affects the polarization of light.

The linear and quadratic terms of magnetization in Eq. (13) are also responsible for the inverse opto-magnetic Faraday and Cotton–Mouton effects. Let us take the Cartesian coordinate system with the axes x, y, z, the simplest case of an isotropic medium, and right-handed circularly monochromatic light $\sigma^{(+)}$ at the frequency ω. The light propagates along the z-axis and the components of the electric field of light along the x and y axes are $E_x^{\omega} = E_0^{\omega} \exp(i\omega t)$ and $E_y^{\omega} = -iE_0^{\omega} \exp(i\omega t)$, where E_0^{ω} is the amplitude of the electromagnetic wave. Eq. (13) shows that if the medium is excited by right-handed circularly polarized light $\sigma^{(+)}$, two orientations of the magnetization $\mathbf{M}$ along the z-axis (parallel, M_z+, and antiparallel, M_z-, to the axis) correspond to different internal energies. In particular, if $\alpha_{xyz} > 0$, it means that $U(M_z-; \sigma^{(+)}) < U(M_z+; \sigma^{(+)})$. If the ferroic medium was initially in the state M_z+, but had enough time to reach thermodynamic equilibrium, i.e. the minimum of U, under illumination, the medium will eventually switch to the state M_z-. Similar considerations can be repeated for left-handed circularly polarized light. The effective magnetic field that breaks the degeneracy between M_z+ and M_z- must be expressed using Eqs. (6), (13) and can be seen as the inverse Faraday effect [76,77].

If the symmetry of the medium is such that $\beta'_{xxxx} \neq \beta'_{xxyy}$, exposing the medium to linearly polarized light with the polarization plane along the x-axis will induce a magnetic anisotropy. In this case the state with the magnetization along the x-axis would have a different internal energy than the state with the magnetization along the y-axis. The effective magnetic field that breaks the degeneracy between mutually perpendicular states of the magnetization can also be expressed using Eqs. (6), (13) and can be seen as the inverse Cotton–Mouton effect [78,79].

In a ferroelectric material the internal energy takes a form

$$U = U_0 + \frac{1}{2}\epsilon_0 \chi_{kl}^s(0) E_k^{\omega *} E_l^{\omega} + \frac{1}{2}\epsilon_0 \eta'_{klm} E_k^{\omega *} E_l^{\omega} P_m + \frac{1}{2}\epsilon_0 \eta''_{klmn} E_k^{\omega *} E_l^{\omega} P_m P_n, \tag{14}$$

where η'_{klm} and η''_{klmn} are phenomenological tensors. The third term gives rise to the electro-optical Pockels effect, where the electric polarization in a medium makes the later optically anisotropic. Optical rectification and difference frequency generation are also particular examples originating from this term [80]. From symmetry considerations η'_{klm} has non-zero components only in noncentrosymmetric media, including ferroelectrics. The last term in the equation yields the electro-optical Kerr effect allowed in media of any symmetry.

To summarize, we show that the effect of optical excitation can be expressed in terms of effective fields $\mathbf{H}_M$ and $\mathbf{H}_P$ acting on the order parameters. These fields can drive switching of the order parameters between stable bit-states. According to Eqs. (6), (7), (13), these fields do depend on the polarization of light and present only during the excitation by light [81]. The duration of such pulses of the effective magnetic or electric field is, thus, significantly shorter than the period of the spin precession in ferro- and even antiferromagnets and shorter than the soft phonon mode period in ferroelectrics. Therefore, the effective field should exert an impulsive driving force on the corresponding order parameter. We note that in a frequency domain this means that the spectrum of the femtosecond laser pulse is broad enough for excitation of coherent magnons or phonons at the center of the Brillouin zone. This process is known as impulsive Stimulated Raman scattering of light by coherent magnons [81,82] or phonons [83,84]. However, as the present review is dedicated to switching of order parameter, it is more convenient to consider the interaction of light with matter in time domain.

Several papers reported about polarization dependent all-optical switching of order parameter with ultrashort laser pulses in ferro-, ferri-, and antiferromagnetic materials. Does it mean that the switching occurs at the timescale of the laser excitation? The answer on this question depends on the role of dissipations in the interaction of light and matter, as well as on a character of kinetics of the order parameter in response to the laser excitation.

3.3. Role of dissipations in nonlinear coupling of light to an order parameter

If dissipations cannot be neglected, the effect of light on order parameter in ferroics can be present on a timescale much longer than the duration of the light pulse. The main reason for it is that dissipations, i.e. irreversible energy transfer from light to matter, increase the entropy of the medium which will decrease again only after sufficiently long time needed to cool the medium down. For instance, if dissipations cannot be neglected in the case of a ferroic with order parameter $\mathbf{\Lambda}$, not only refraction, but also the absorption coefficient starts to depend on both polarization of light and the order parameter of the medium. It means that $\partial Q/\partial \mathbf{\Lambda} \neq 0$, and the dissipations result in an additional term in the effective field generated by light. This means that the light-induced effective field may be present much longer than the duration of the laser pulse. Naturally, details of evolution of the effective field following excitation depend on a particular microscopic process underlying the emergence of the field. Several particular cases of such long-living effective field and their origins are considered in the following Section.

3.4. Response of the order parameter to fields

In order to understand how order parameters respond to optically generated effective fields or the fields of the electromagnetic wave, one has to consider equations of motion for a particular ferroic order parameter. In case of a ferromagnet, the magnetization dynamics in an effective magnetic field $\mathbf{H}_M$ is described by the Landau–Lifshitz–Gilbert equation:

$$\dot{\mathbf{M}} = -\gamma \mathbf{M} \times \tilde{\mathbf{H}}_M + \frac{\alpha}{M} \mathbf{M} \times \dot{\mathbf{M}}, \tag{15}$$

where $\tilde{\mathbf{H}}_M$ is the effective magnetic field which consists of the equilibrium contribution $\mathbf{H}_M^0$ and the light-induced one $\mathbf{H}_M$; α is dimensionless constant of Gilbert damping. Consequently, since we start from the equilibrium state with $\mathbf{M} \| \mathbf{H}_M^0$, the initial torque acting on the magnetization because of the light-induced effective field is given by $-\gamma \mathbf{M} \times \mathbf{H}_M$. The equation assumes that the length of the net magnetization vector is conserved.

In order to reveal the response of an antiferromagnet to external stimuli, we consider the case of a compensated antiferromagnet having two equivalent antiferromagnetically coupled sublattices. The magnetizations of the two sublattices are $\mathbf{M}_1$ and $\mathbf{M}_2$. It is convenient to express the magnetizations in terms of unit vectors $\mathbf{m}_1 = \mathbf{M}_1/M_0$ and $\mathbf{m}_2 = \mathbf{M}_2/M_0$, where $M_0 = |\mathbf{M}_1| = |\mathbf{M}_2|$. The motion of the magnetizations in each of the sublattices obeys the Landau–Lifshitz–Gilbert equation i.e.

$$\dot{\mathbf{m}}_1 = -\gamma \mathbf{m}_1 \times \tilde{\mathbf{H}}_1 + \alpha \mathbf{m}_1 \times \dot{\mathbf{m}}_1, \tag{16}$$

$$\dot{\mathbf{m}}_2 = -\gamma \mathbf{m}_2 \times \tilde{\mathbf{H}}_2 + \alpha \mathbf{m}_2 \times \dot{\mathbf{m}}_2, \tag{17}$$

where $\tilde{\mathbf{H}}_1, \tilde{\mathbf{H}}_2$ are the effective magnetic fields experienced by the magnetizations of the first and the second sublattice, respectively. Similarly to the net magnetization $\mathbf{m} = (\mathbf{m}_1 + \mathbf{m}_2)/2$, here, in order to describe antiferromagnetism, we also introduce antiferromagnetic vector $\mathbf{l} = (\mathbf{m}_1 - \mathbf{m}_2)/2$. According to the definition $\mathbf{m}^2 + \mathbf{l}^2 = 1$ and $\mathbf{ml} = 0$. Next to time-dependent applied magnetic field $\mathbf{H}(t)$ and the field of magnetic anisotropy $\mathbf{H}_A(\mathbf{l})$, the magnetic sublattices also experience the exchange field $H_E \sim 10^3$ T. Therefore for the effective magnetic fields $\tilde{\mathbf{H}}_1$ and $\tilde{\mathbf{H}}_2$ one can write

$$\tilde{\mathbf{H}}_1 = H_E \mathbf{l} + \mathbf{H}_t, \tag{18}$$

$$\tilde{\mathbf{H}}_2 = -H_E \mathbf{l} + \mathbf{H}_t, \tag{19}$$

where for simplicity $\mathbf{H}_t = \mathbf{H}(t) + \mathbf{H}_A(\mathbf{l})$, i.e. it is assumed that the applied field $\mathbf{H}(t)$ is time-dependent, while the anisotropy field $\mathbf{H}_A(\mathbf{l})$ depends on $\mathbf{l}$.

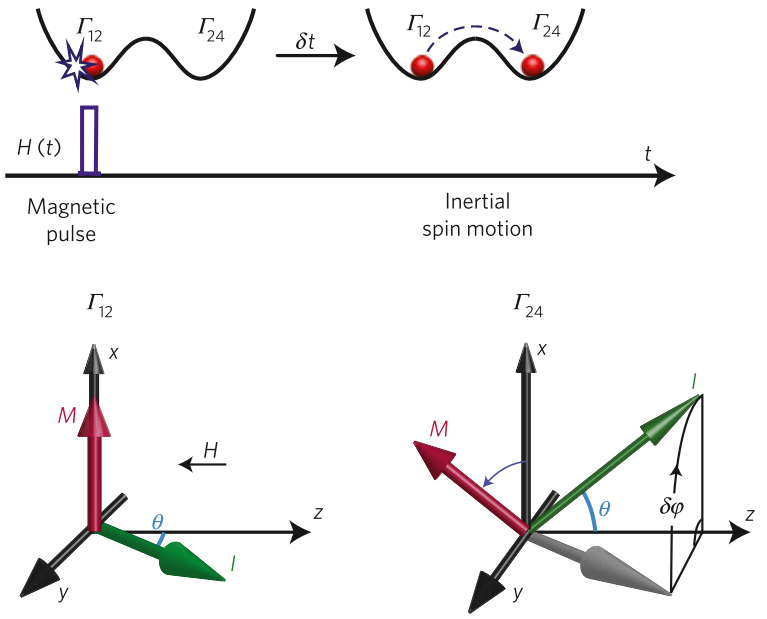

Fig. 7. Switching spins over the potential barrier in $HoFeO_3$ employing inertia of spins in an antiferromagnet. The effective magnetic field pulse duration is much shorter than time required to overcome the potential barrier. This is a canted antiferromagnet with a non-zero magnetization. Optical excitation promotes a transition from Γ_{12} to Γ_{24} phase during which the magnetization **M** and the antiferromagnetic vector **L** rotate over 90 degrees [15].
Source: The figure is taken from Ref. [15].

Eqs. (16), (17) can be rewritten as follows

$$\dot{\mathbf{l}} = \gamma(H_E\mathbf{m}\times\mathbf{l} - \mathbf{l}\times\mathbf{H}_t) + \alpha(\mathbf{m}\times\dot{\mathbf{l}} + \dot{\mathbf{m}}\times\mathbf{l}), \tag{20}$$

$$\dot{\mathbf{m}} = -\gamma\mathbf{m}\times\mathbf{H}_t + \alpha\mathbf{l}\times\dot{\mathbf{l}}. \tag{21}$$

Finding vector products of both sides of Eq. (20) and vector **l**, one gets

$$\mathbf{m} = \frac{1}{H_E}(\mathbf{H}_t - \mathbf{l}(\mathbf{H}_t\mathbf{l}) - \frac{1}{\gamma}\mathbf{l}\times\dot{\mathbf{l}}). \tag{22}$$

It is seen that the net magnetization of a compensated antiferromagnet can be induced not only by an external magnetic field, but also by coherent motion of spins so that in motion $\mathbf{l}\times\dot{\mathbf{l}} \neq 0$. Substituting Eq. (22) into Eq. (21) one obtains [85]

$$\frac{d}{dt}[\mathbf{l}\times\dot{\mathbf{l}}] = \gamma\dot{\mathbf{H}}(t) + \gamma^2 H_E\mathbf{m}\times\mathbf{H}_t - \alpha\gamma H_E\mathbf{l}\times\dot{\mathbf{l}} - \frac{d}{dt}(\mathbf{l}(\mathbf{H}_t\mathbf{l})). \tag{23}$$

Hence dynamics of the antiferromagnetic vector can also be launched by an external magnetic field, if the latter is not constant i.e. $\dot{\mathbf{H}}(t) \neq 0$. This effect was in detail described theoretically in several papers [85–88].

This consideration shows that applied or light-induced effective magnetic field can trigger ferro- and antiferromagnetic resonance modes, which can lead to switching of the corresponding order parameter. However, in antiferromagnets, the switching of the order parameter can follow a different route compared to ferro- and ferrimagnetic materials. By comparing switching the order parameter between two metastable states to transferring a particle over a potential barrier from one minimum to another (Fig. 7), one can notice that the inertia of motion can be employed for switching. Even if the interaction between an external stimulus and the particle is so short that the coordinate of the particle hardly changes during the interaction time, as long as the particle acquires sufficient linear momentum and kinetic energy it may overcome the potential barrier afterwards. Nevertheless, the Landau–Lifshitz–Gilbert excludes any inertial mechanisms. The Landau–Lifshitz equation implies that the rate of the magnetization change $\dot{\mathbf{M}}$ is proportional to the torque. In antiferromagnets, the equation of motion is of the second order with respect to time and therefore spin dynamics in antiferromagnets must show signatures of inertia. For instance, it must be possible to switch spins of antiferromagnets with stimuli much shorter than those required for non-inertial switching. Such a switching of spins between two potential minima was reported for antiferromagnetic $HoFeO_3$[15].

To describe nonlinear dynamics of the ferroelectric order parameter **P** a simple model [89,90] based on a double well energy potential $U(Q_P)$ is usually used:

$$U = -\frac{aQ_P{}^2}{2} + \frac{bQ_P{}^4}{4}, \tag{24}$$

where Q_P is atomic displacement, proportional to the particular component of the order parameter **P** (in other words, this phonon mode is transformed according to the same irreducible representation of the crystal symmetry group as the appropriate component of the order parameter **P**), a is the oscillation frequency, b is the distance between two minima of Eq. (24) at $T < T_C$, where T_C is the Curie temperature of the ferroelectric. a and b are assumed to be positive.

Dynamics of the $Q_P(t)$ is determined by the following equation

$$\ddot{Q}_P + 2\Gamma\dot{Q}_P - aQ_P + bQ_P{}^3 = \beta E(t), \tag{25}$$

where Γ is the damping rate ($\Gamma \sim 0.3 - 0.5$ ps^{-1} [90,91]). The right side of Eq. (25) describes the driving force exerted by the applied electric field, β is the coupling factor.

4. Microscopic mechanisms of coupling of light with spins and charges in ferroics

Microscopically light-matter interaction relies on an excitation of electronic, phononic and magnonic resonances with the help of both electric and magnetic components of light. They lead to non-equilibrium population of excited electronic states, generation of coherent as well as incoherent phonons and magnons. However, the coherent excitations eventually lose their coherence, and the thermodynamic temperature of the medium increases as a result of the increase of non-coherent quasi-particles population. In the following we discuss how these excitations can facilitate switching of the order parameters in ferroics with the help of light.

4.1. Light-spin coupling

Light is an electromagnetic wave and the magnetic field of light can interact with spins and affect the magnetic order. Obviously, for the most efficient control of spins with the magnetic field of electromagnetic radiation, the spectrum of the magnetic field must have components at the frequencies of the mode involved in the reorientation of the order parameter.

Off-resonant coupling of short THz pulses to spins in ferromagnet was demonstrated in Ref. [92]. As expected, the coupling was not efficient, and immediately after the action of the THz pulse the spins returned back to the initial position (Fig. 8). Indeed, resonant excitation of spins in antiferromagnets with the help of THz pulse was demonstrated for NiO in [93]. As a result of the excitation, a part of the energy of the THz pulse was transferred to spins and the latter kept oscillating at the frequency of the antiferromagnetic resonance long after the action of the THz pulse (Fig. 9). It is important to note, that here the resonant coupling between the magnetic field of the THz pulse and the antiferromagnetic vector relies on the time-derivative of the field (Eq. (23)) as discussed in paragraph 3.4.

From thermodynamics of light-matter interactions considered in Section 3 it is seen that interaction of the electric field of light with electrons can also result in a generation of effective magnetic fields. In order to understand microscopic processes responsible for coupling of light to spins eventually leading to generation of coherent magnons, one has to realize that, if no magnetic field is applied, the spin-dependent part of the potential energy of an electron in a medium U is defined by three main interactions:

- The exchange interaction $J\mathbf{S}_i\mathbf{S}_j$ is responsible for the coupling between the spins of ith and jth electrons with effectively overlapping wave-functions. It is isotropic and, in majority of magnetically-ordered media, is the strongest interaction.
- The Dzyaloshinskii–Moriya interaction $\mathbf{D}(\mathbf{S}_i \times \mathbf{S}_j)$, or antisymmetric exchange interaction, is a two-cites interaction between neighboring spins $\mathbf{S}_i$ and $\mathbf{S}_j$ taking into account spin–orbit interaction. The interaction is responsible for occurrence of non-collinear magnetic structures.
- The spin–orbit interaction $\lambda\mathbf{SL}$ responsible for the coupling of the spin **S** and the orbital momenta **L**. This term, in particular, gives rise to the single-ion magnetocrystalline anisotropy. The spin–orbit interaction is also eventually responsible for the Dzyaloshinskii–Moriya interaction. While this interaction is typically weaker than the exchange interaction, in particular cases of high-Z ions (e.g. lanthanides) with large orbital momenta, spin–orbit interaction competes or even dominates over the exchange between neighboring spins.

The effective magnetic field experienced by a spin $\mathbf{S}_k$ is given by $\mathbf{H}_M \sim -\partial U/\partial\mathbf{S}_k$. The effective field originating from the exchange and the Dzyaloshinskii–Moriya terms depends on spin structure, and the most universal way to excite spins is to employ the third term related to the spin–orbit interaction. Although the electric field of light cannot affect the spin of electron directly, an excitation of an optical transition changes the orbital state of electron and thus generates an effective magnetic field $\mathbf{H}_M$ acting on the spin.

If light excites electronic transitions off-resonantly and thus no irreversible energy transfer from light to matter takes place, the situation must be treated under non-dissipative approximation (Section 3) implying that the effective magnetic

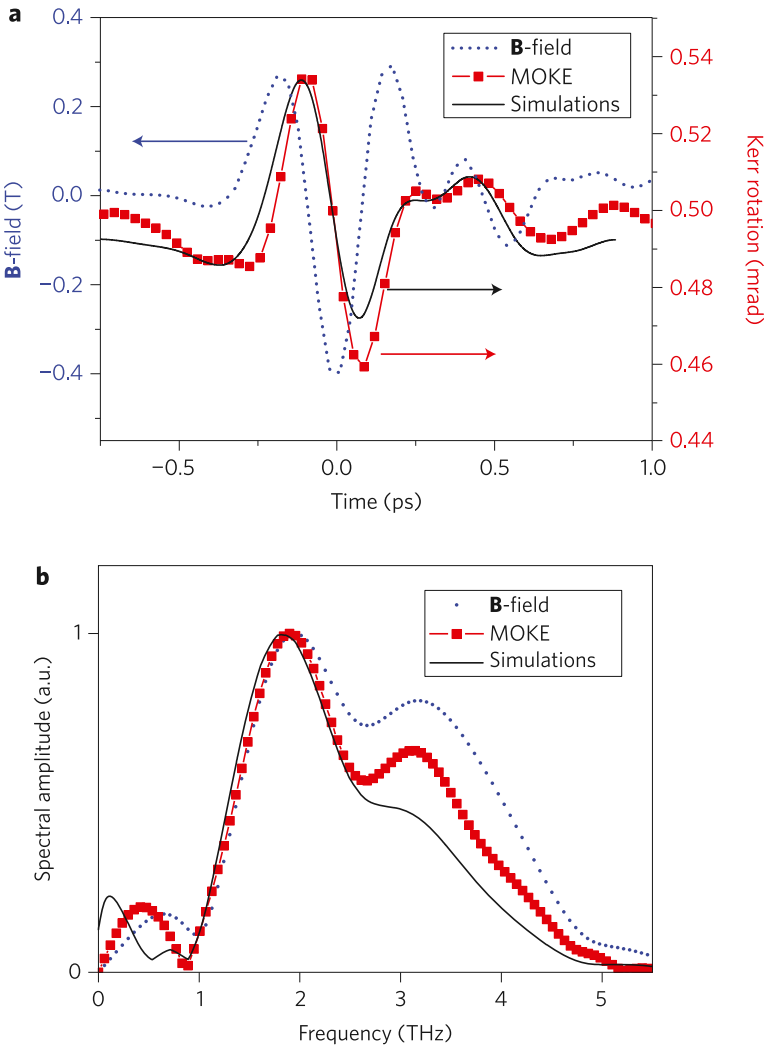

Fig. 8. Magnetization dynamics triggered by strong, nearly single cycle THz pulses as a result of off-resonant pumping of ferromagnetic resonance in Co [92]. (a) Femtosecond magnetization dynamics represented by the MOKE signal (red curve) and initiated by the phase-stable single-cycle terahertz magnetic field (blue dotted curve) follow the terahertz field oscillations. (b) The corresponding spectral intensities of the terahertz pulse and MOKE response are almost identical, which illustrates that the coupling between the terahertz field and magnetization does not rely on any resonant excitation or spin waves. The magnetization vector evolution calculated using the Landau-Lifshitz equation reflects the observed ultrafast magnetization dynamics well in both temporal and spectral domains [92].
Source: The figure is taken from Ref. [92].

field is present in the medium only during the action of the laser pulse. In this case, during the optical excitation the wave-function of the excited electron becomes a time-dependent superposition of wave-functions of stationary states. Hence the effective spin–orbit interaction becomes different from the one in the ground state. The fact that femtosecond laser pulse can act on exchange coupled spins as an equally short effective magnetic field was for the first time demonstrated in [94] for a weak ferromagnetic dielectric $DyFeO_3$ (Fig. 10(a)) and later shown to be present in a large class of magnetic dielectrics, including ferrimagnets and compensated antiferromagnets (for a review see, e.g., [34,81]).

Experiments on laser-induced coherent spin dynamics in compensated antiferromagnetic dielectric NiO allow to demonstrate the effect of the time derivative of an effective field on the spin system (Eq. (23)). Ref. [93] showed that linear coupling of the THz magnetic field to the magnetizations of the sublattices triggers coherent spin oscillations at the frequency of antiferromagnetic resonance in NiO (Fig. 9). Similar coherent spin oscillations could also be excited via nonlinear mechanism of coupling of light to the magnetization of the sublattices [96] (Fig. 11). In both cases, the role of a driving force is played by time derivative of the effective magnetic field $\partial\mathbf{H}/\partial t$, and in both cases observation of the coherent spin oscillations in the antiferromagnet was based on measurements of the direct magneto-optical Faraday effect. Although for unperturbed compensated antiferromagnet the net magnetization and the magneto-optical Faraday

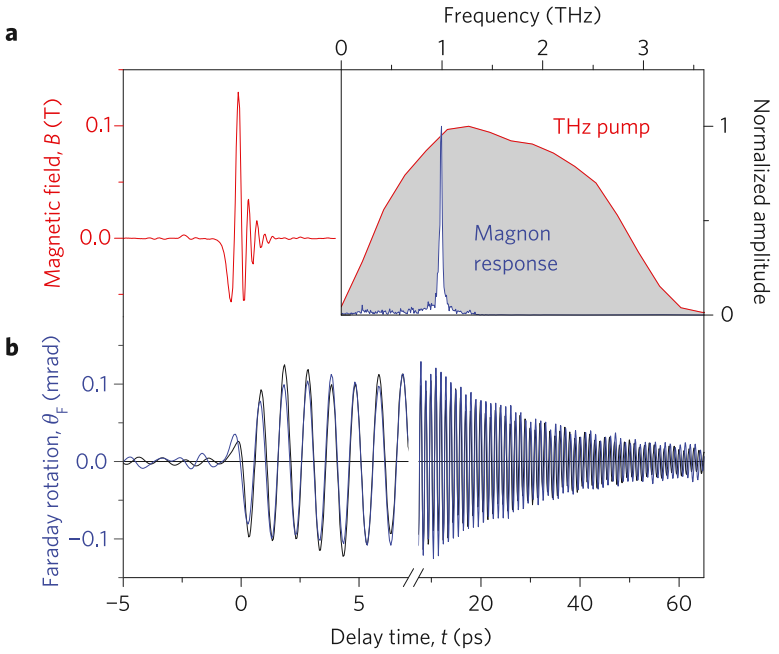

Fig. 9. Excitation of the antiferromagnetic resonance mode in NiO by nearly single cycle THz pulse [93]. The mode is revealed by measuring dynamics of the magneto-optical Faraday effect $\mathbf{l} \times \dot{\mathbf{l}}$-term (Eq. (22)) of the magnetization. The spins are excited with by the rapidly changing THz magnetic field. The dynamics is probed by detecting $\mathbf{l} \times \dot{\mathbf{l}}$-term of the magnetization with the help of the magneto-optical Faraday effect [93].
Source: The figure is taken from Ref. [93].

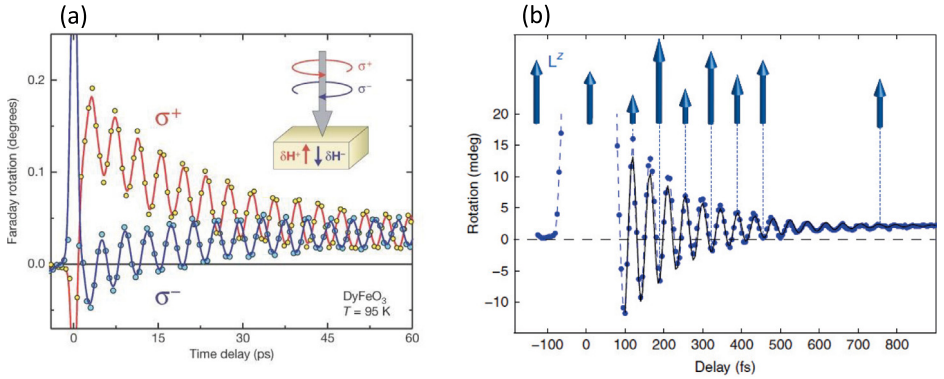

Fig. 10. Spin dynamics triggered by femtosecond laser pulses in magnetic dielectric as a result of inverse opto-magnetic effects via off-resonant pumping of electronic transitions. Panel (a) shows optically detected magnetization dynamics excited by circularly polarized light in antiferromagnetic $DyFeO_3$ employing the inverse Faraday effect and optical control of the spin–orbit interaction [94]. Panel (b) shows optically detected oscillations of the length of the antiferromagnetic vector in $KNiF_3$ triggered as a result of laser-induced change of the exchange interaction. This effect of light generates two coherent magnons at the edge of the Brillouin zone. The length of the antiferromagnetic vector oscillates as long as the magnons maintain the mutual coherence [95].
Source: The figures are taken from Refs. [94,95].

effect are zero, both the magnetization and the magneto-optical Faraday effect emerge due to the $\mathbf{l} \times \dot{\mathbf{l}}$-term in Eq. (21) upon excitation of the dynamics of $\mathbf{l}$.

A resonant excitation of specific electronic transitions can be even more efficient tool to control spins in magnets. If light excites real electronic transitions, dissipations cannot be neglected. The effective magnetic field generated by light can be present long after the action of the laser pulse and stay as long as the lifetime of the electrons in the excited state. Examples of dissipative effects of light on magnetic system are photoinduced magnetic anisotropy due to photo-activation of anisotropic centers such as Co^{2+} in the case of iron garnet [97,98] and Tm^{3+} ions in the case of $TmFeO_3$ [99], optical spin-transfer torque [100] and optical spin orbit-torque [101] shown for magnetic semiconductors.

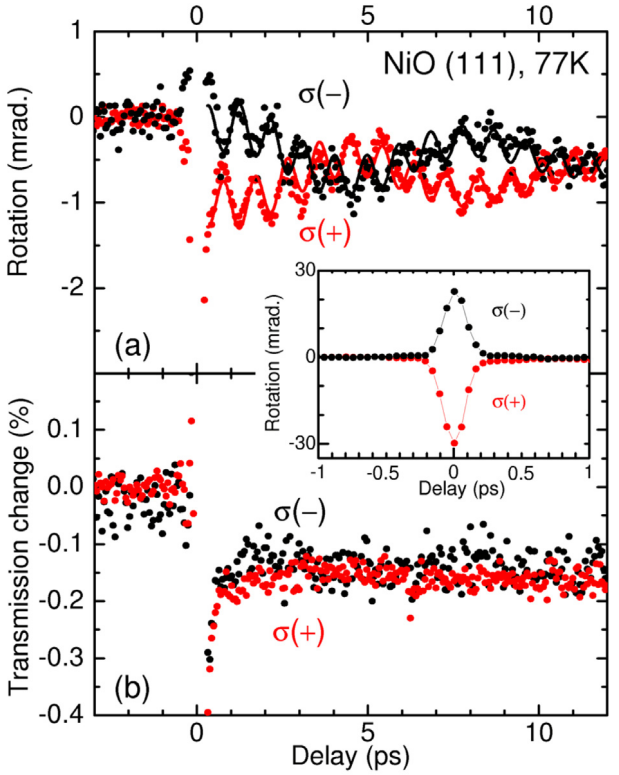

Fig. 11. Excitation of the antiferromagnetic resonance mode in the antiferromagnetic dielectric NiO by a circularly polarized laser pulse [96]. The mode is revealed by measuring dynamics of the magneto-optical Faraday effect proportional to the $\mathbf{l} \times \dot{\mathbf{l}}$-term of the magnetization (Eq. (22)). The spins are excited by the rapidly changing effective magnetic field generated due to the inverse Faraday effect $\dot{\mathbf{H}}_M$ (Eq. (23)). Circularly polarized laser pulses of opposite helicities excite oscillations of opposite phases.
Source: The figure is taken from Ref. [96].

Similarly to the optical control of spins mediated by one-site spin–orbit interaction in the laser-excited state, one can anticipate that light can couple to spins via optical control of the two-cites exchange and the Dzyaloshinskii–Moriya interaction. Light effectively changes the wave-function of the excited electron and the new wave-function can be expressed in terms of time-dependent superposition of the wave-functions of the stationary states. Hence the quantum mechanical interatomic exchange interaction becomes different from the one in the unperturbed state and must be a function of time. Thermodynamically, the change of the exchange interaction by light can be described in terms of inverse magneto-refractive effect [102]. If one considers isotropic exchange interaction, making this interaction stronger or weaker with the help of light does not lead to collective reorientation of spins, i.e. reorientation of order parameter. Instead, a decrease of the exchange interaction, for instance, should eventually result in a reduction of the absolute value of the net order parameter. This has been demonstrated in [95] where a sudden change of the exchange energy in an antiferromagnet via non-resonant excitation resulted in a generation of magnons at the edge of the Brillouin zone with the frequency ω_m. As long as these magnons remained coherent, the length of the order parameter, i.e. antiferromagnetic Néel vector $\mathbf{L}$, oscillated at the frequency equal to $2\omega_m$. Upon decoherence of the magnons, the oscillations damped, and the order parameter reduced its absolute value (Fig. 10(b)). Hence laser-induced change of the exchange interaction can result in an eventual demagnetization even after off-resonant optical excitation. Such a demagnetization can be realized on the timescale of the quarter of magnon period at the edge of the Brillouin zone $\pi/2\omega_m$. For instance, in $KNiF_3$ (Fig. 10(b)) this time is as short as $\sim$10 fs [95]. If the light-induced perturbation of electrons wave-functions is present longer, the effect of reduced/increased exchange interactions would be more pronounced. In contrast to collinear antiferromagnets, in antiferromagnets with magnetic sublattices canted either due to the Dzyaloshinskii–Moriya interaction or an external magnetic field, an ultrafast effect of light on the exchange coupling between the sublattices results in coherent spin oscillations at the frequency of antiferromagnetic resonance [102,103].

Effect of light on the exchange interaction via resonant pumping of electronic transitions must have both a longer life time and higher efficiency compared to the off-resonant case. For instance, theory predicts that photo-doping of a Mott insulator can quench the exchange parameter by as much as 10% [104]. Further studies of ultrafast laser-induced dynamics in Mott insulators revealed that the results of simulations using nonequilibrium dynamical mean-field theory are rather different from the experimentally observed dynamics [105]. In particular, the results of the previous computational

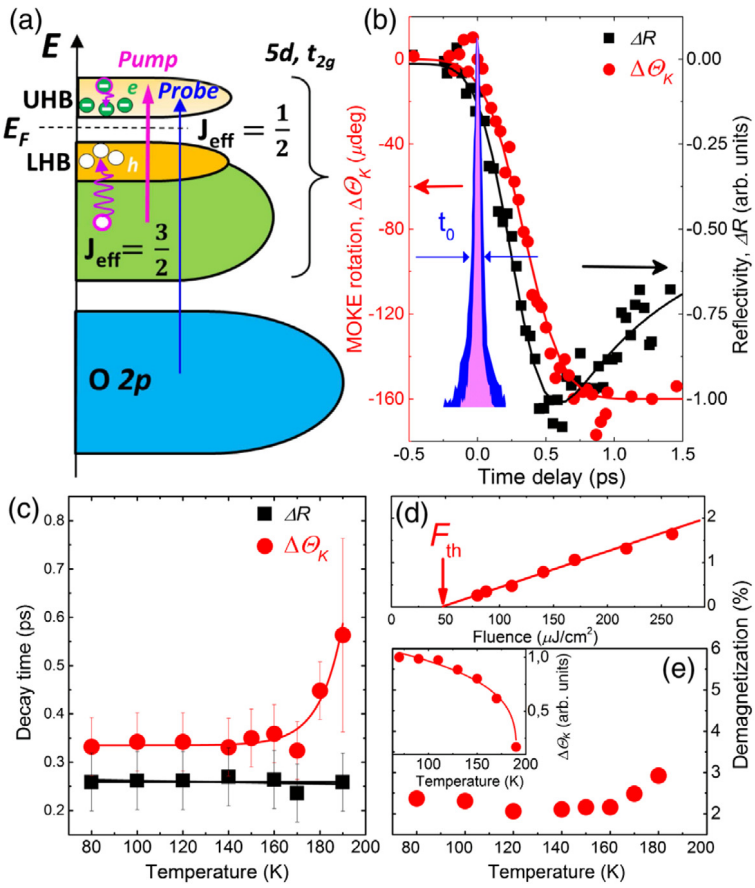

Fig. 12. Laser-driven melting of the antiferromagnetic ordering in a Mott-insulator Sr_2IrO_4 [105]. (a) Schematic band structure of Sr_2IrO_4. The pink arrow indicates the optical transition launched by the pump pulse with a photon energy of 1.55 eV. Open circles and circles with a dash indicate photoinduced holes *h* and electrons *e*, respectively. Pink spring illustrates the process of the impact ionization and relaxation of the photoinduced carriers to the bottom of the upper (UHB) and lower (LHB) Hubbard bands. The solid blue line denotes charge-transfer transition between oxygen 2*p* and iridium 5*d* ions probed with the laser pulses pulse having a photon energy of 3.1 eV. (b) Left axis: Time-resolved transient magneto-optical Kerr effect signal triggered by the pump pulse. Right axis: Time-resolved transient reflectivity signal triggered by the pump pulse. Solid lines are fits to the data. The fits are represented by an exponential decay function multiplied by the error function. Sample temperature is 80 K and the pump fluence is 100 μJ/cm^2. The blue and violet lines show the autocorrelation functions of the probe and pump pulses, respectively. (c) Temperature dependence of the magnetization decay time (red circles) and time of the fast rising edge in the reflectivity signal (black squares). (d) Value of the pump-induced demagnetization as a function of the pump fluence. (e) Value of the pump-induced demagnetization as a function of the sample temperature. The inset shows the magnitude of the transient pump-induced MOKE rotation as a function of the sample temperature. The fluence of the pump pulse is 250 μJ/cm^2.
Source: The figure is taken from Ref. [105].

studies and success in fabrication of new materials motivated experimental investigation of ultrafast laser-induced dynamics of magnetization in a novel spin–orbit Mott insulator Sr_2IrO_4. This Mott-insulator is an antiferromagnet with canted magnetic sublattices and the net magnetic moment. Using the transient magneto-optical Kerr effect sensitive to the net magnetization, it was revealed that photodoping by femtosecond laser pulses with photon energy above the Mott bandgap (Fig. 12(a)) launches melting of the antiferromagnetic order seen as ultrafast demagnetization by 2–3% with a characteristic time of 300 fs (see Fig. 12(b)). The work propose a phenomenological model which is based on Onsager's formalism and accounts for the photogenerated electron–hole pairs using the concepts of holons and doublons.

Instead of using virtual or real electronic excited states to generate a light-induced effective field, one may employ coherent lattice oscillations, or phonons. In order to control magnetic order parameter via coherent phonons, one needs to match the symmetries of the lattice and the spin excitations. For instance, elliptical lattice vibration may generate an effective magnetic field [106] which would dynamically change the symmetry of the ground state of electrons. Although the electron remains in the ground state, due to coherent lattice vibrations the orbital momentum of the ground state changes affecting the spin of the electron. Such a scenario has been recently realized in a weak ferromagnet — $ErFeO_3$ orthoferrite. Pumping $ErFeO_3$ single crystal with femtosecond laser pulses in the mid-infrared spectral range it was possible to excite spin oscillations by generating phonon-mediated effective magnetic fields [107]. Generation of such a

field was enabled by resonant excitation of two IR-active optical phonon modes of slightly detuned frequencies (16.17 THz and 17.03 THz). Symmetry-wise the effect of the mid-infrared light can be described in a way similar to the inverse opto-magnetic effects, where instead of the electric field of light the expression for the effective magnetic field $H_c = i\alpha_{abc}Q_{ua}Q_{ub}$ contains normal coordinates Q_u of the phonon modes.

It is important to stress, that the effect of phonon-induced effective field was found to be significantly weaker that the one from the optically-induced field. For instance, for pump-fluence of 20 mJ/cm^2 the effective magnetic field in [106] was estimated to be around 36 mT, which is at least an order of magnitude smaller than the typical estimates obtained in the near-infrared experiments, including those with $ErFeO_3$ [108]. In order to understand why the optically induced effective magnetic fields can depend on the photon energy, one can consider an electronic transition between two states e and g. The resonance of this electronic transition is at the frequency ω_{eg} and the transition is excited by the electric field at the frequency ω. Ref. [109] showed that antisymmetric and symmetric parts of the optical susceptibility vary with the frequency as $\chi^a_{kl} \sim \omega/(\omega^2 - \omega^2_{eg})$ and $\chi^s_{kl} \sim \omega_{eg}/(\omega^2 - \omega^2_{eg})$, respectively. First of all, it is seen that upon a reduction of ω_{eg} the susceptibility χ^s_{kl} decreases. Secondly, if ω_{eg} is not equal to zero, one can anticipate that upon a decrease of the frequency of light effective magnetic field generated by circularly polarized electric fields $\sim \chi^a_{kl}$ will become weaker than those induced by linearly polarized radiation χ^s_{kl}. Smaller effective magnetic field generated by circularly polarized light at THz frequency in comparison with the field generated by linearly polarized light can be also illustrated in terms of a simple model. The effective magnetic field generated by circularly polarized light can be modeled by circular currents of electrons driven by light [110]. The current breaks the degeneracy between spins pointing parallel and antiparallel with respect to the wavevector of light, respectively. The strength of the field and splitting between two spin states is proportional to the speed of the electron and thus to the frequency of light. Hence the splitting naturally vanishes upon approaching zero frequency. Linearly polarized light, on the contrary, breaks the degeneracy in the sample plane and thus induces an axis of magnetic anisotropy in otherwise isotropic medium. The corresponding effective magnetic field is proportional to the square of the electric field of light and remains strong even for electric fields at zero frequency [111].

The effective magnetic field generated by the elliptically-polarized transverse acoustical phonons was reported to be the origin of spin polarization in a semiconductor quantum well separated by a 30 nm thick barrier from a ferromagnetic metallic layer [112]. In this work it was suggested that the phonons in the ferromagnet acquire ellipticity due to their coupling to the magnons, propagate into the quantum well, and thus create a polarization of heavy holes spins there. The effect which can be seen as an acoustic analogy of the Stark effect, reveals the possibilities to use optically generated acoustic coherent phonons [113] for dynamical long-range coupling between different magnetic and nonmagnetic species in heterostructures.

Laser-generated acoustic phonons, or picosecond strain pulses, are also able to affect magnetism employing the mechanisms of magnetostriction [114,115]. In antiferromagnetic $FeBO_3$ it was shown that optically generated strain can cause a large amplitude coherent spin dynamics in a strongly anharmonic regime via modulating magnetic anisotropy of the medium [116]. Theoretical proposal for strain-mediated optically induced magnetic switching [117], and developed approaches for generation of acoustic pulses with a broad spectrum up to THz frequencies and large amplitudes [113] show the potential of ultrafast optical control of magnetism via optically-induced strain.

From a general point of view, ultrafast lattice excitations, both in form of acoustic and optical phonons, should have a direct effect on the exchange interactions, since changing distances and angles between particular ions in a solids should yield change of the wavefunctions overlap and, thus, the exchange integrals. In [118] it was considered theoretically, that an effective overlap of the wave-functions of the adjacent spins can be changed via a lattice expansion, e,g, strain. Since strain can be optically generated, the suggested pathway to control exchange interactions can be investigated experimentally as well. Effect of the laser-driven optical polar phonon mode on the exchange interaction in antiferromagnetic Cr_2O_3 was analyzed theoretically in [119]. Here the antiferromagnetic ordering type was shown to depend on the distance between two Cr ions. Employing the effects of nonlinear phononics, the distance can increase due to a rectified phononic field. It was suggested theoretically that magnon excitation can also affect the exchange interaction in a medium [120,121].

Both resonant and off-resonant excitations of electric-dipole electronic transition are accompanied by a change of orbital magnetic quantum number. In principle, it means that light induces an orbital magnetic moment. If the transition is excited off-resonantly, the induced magnetization is present only during the laser pulse [122]. Resonant excitation can induce long-living orbital magnetization. However, it is still not clarified, how efficiently such a laser-induced magnetization can couple to spin order in a ferroic material. An effect of optically-induced transient magnetization was discussed in the study of ultrafast laser-induced magnetization dynamics in $DyFeO_3$. This is an antiferromagnet with weak ferromagnetism in which the magnetic ordering is due to exchange coupling between spins of Fe^{3+}, while the spins of Dy^{3+} ions are in a paramagnetic state being magnetized only partially. Using femtosecond laser pulses as a pump and THz emission spectroscopy as a probe of ultrafast magnetization dynamics in Ref. [123] it was shown that a resonant optical pumping of $f-f$ electronic transitions in the Dy^{3+} ions magnetizes these partially ordered Dy ions by almost 1%. Interestingly, this laser-induced magnetization did not result in excitation of the exchange coupled spins of Fe^{3+} ions, and even weakened the effects of light on the spin order. In fact, it was shown that the resonant photoinduced magnetization in the Dy^{3+} subsystem and the off-resonant excitation of spin waves in the Fe^{3+} subsystem are intrinsically competing processes.

4.2. Light-charge coupling

Here we turn to a discussion of possible impacts of ultrashort laser pulses on ferroelectric ordering. As follows from the thermodynamic consideration, one expects to alter ferroelectric polarization by driving a particular optical phonon mode. There are several approaches to drive coherent optical phonons by light. A direct way is to excite a medium in the THz and mid-IR range resonantly with the phonon mode of interest (see e.e. a recent review [124]). In this way only IR-active modes can be excited. This limitation can be lifted by means of phonon–phonon coupling enabling the frequency conversion, which is a key tool in a 'nonlinear' phononics [125]. For reviews on this emerging field we refer to [126,127].

Alternatively, difference- and sum-frequency generation processes can be used to drive a specific phonon mode. Both processes can be either of photonic or ionic origin [128]. The difference-frequency generation with laser pulses is realized via a so-called impulsive stimulated Raman scattering. Here the photons at optical frequencies are scattered non-elastically and drive excitation of a particular phonon mode [129,130] with the efficiency defined by the Raman selection rules, as well as by spectral and temporal profiles of the optical pulse [131]. The counterpart of such process is the ionic scattering (IRS), where the IR-driven phonons are scattered producing a phonon at the difference-frequency [125,132,133]. Furthermore, sum-frequency generation on phonons can be used to access phonons, which do not couple to THz or IR light directly [128,134]. Thus, a short pulse in the THz, IR or optical spectral range can enable a range of mechanisms allowing driving coherent optical phonons of different frequencies and symmetries.

In searching for possible mechanism of coupling of light to charges, it is instructive to consider microscopic origin of ferroelectricity, as we did above for magnetically-ordered media. The most common mechanisms are:

- Pseudo Jahn–Teller effect (PJTE), or second order Jahn–Teller effect [135], which describes the noncentrosymmetric distortion of centrosymmetric ground state as a result of its mixing with the low-lying noncentrosymmetric excited state. $BaTiO_3$ is an example of materials which ferroelectricity originates from PJTE.
- Rotations and tilts of ligand complexes, which lead to ferro- or antiferroelectric ordering depending on particular crystal structure. Ferroelectricity in hexagonal manganites is attributed to these rotations. These rotations combined with PJTE give rise to antiferroelectricity in $PbZrO_3$.
- Long-range Coulomb interactions. These interactions are seen as a partial analogy of the exchange interaction in magnetically-ordered materials, as they play important role in stabilizing parallel alignment of local dipole moments emerged within the unit cells.

Therefore, apart from driving specific phonon modes coupled to the ferroelectric polarization, excitation of a ferroelectric material by light creating a time-dependent superposition of ground and excited electronic states, may have a strong impact on ferroelectric ordering, for instance, via modifying PJTE.

Modulation of ferroelectric polarization by coherent phonons. The idea to employ coherent phonons to mediate coupling of light to a spontaneous polarization in ferroelectric has been a subject of experimental and theoretical studies for more than 20 years. In ferroelectrics there is a particular optical phonon mode possessing the symmetry of the order parameter. This mode softens at the Curie temperature (so-called soft phonon mode). Below T_C, a coherent excitation of this mode will launch oscillations of the order parameter. If the amplitude of the oscillations is larger than the potential barrier separating two states with opposite directions of spontaneous polarization, the switching to another bit state can occur. Aiming to find the most efficient driving force for phonon modes, different strategies were considered theoretically. It was proposed to employ stimulated Raman scattering of light on phonons [18], nonlinear conversion of excited phonons and anharmonism of the modes [136], and direct i.e. resonant pumping of the soft-mode with THz-pulses [90,137]. Several experiments claimed that femtosecond laser pulse can excite phonon mode associated with the ferroelectric polarization in a number of displacive and order–disorder ferroelectrics [138–141].

THz-driven excitation of the structural dynamics and associated dynamics of the ferroelectric polarization was reported in [142] for the case of single domain $BaTiO_3$ film. Intense THz pulses additionally enhanced by a split-ring resonator were employed for excitation. Time-resolved X-ray scattering technique was used to monitor the atomic displacements. Observed in these experiments structural dynamics consisted of several steps. At longer timescale the strain- and heating effects were observed. During the THz pulse impact the modulation of atomic displacements along the direction of ferroelectric polarization was observed reaching $\sim$10% of the build-in polarization. This modulation was ascribed to an effective rotation of the ferroelectric polarization, which originated from incoherent in-plane displacements of Ti atoms along the electric field of the THz pulse. However, no coherent excitation of the polar phonon mode was detected in these experiments although the spectrum of the excitation contained the frequency of the mode.

The most direct way to affect the ferroelectric polarization would be to drive the soft phonon mode using THz electric field being collinear with the polarization. Recently it was shown that THz pulses can indeed excite coherent atomic oscillations at the frequency of the soft phonon mode [90,143]. Fig. 13(a) shows the phonon mode in a ferroelectric $Sn_2P_2S_6$ excited resonantly by intense THz pulse and probed via ultrafast X-ray diffraction [90]. The initial motion of Sn^{2+} ions can be reversed by changing the initial phase of the THz pulse by 180 deg. Softening of the excited phonon mode upon approaching the transition to paraelectric phase was observed (Fig. 13(b)) along with an enhancement of the coupling factor. The atomic displacements and relative spontaneous polarization changes were found to be of 7.5 and 8%, respectively. The amplitudes are not sufficient for the switching.

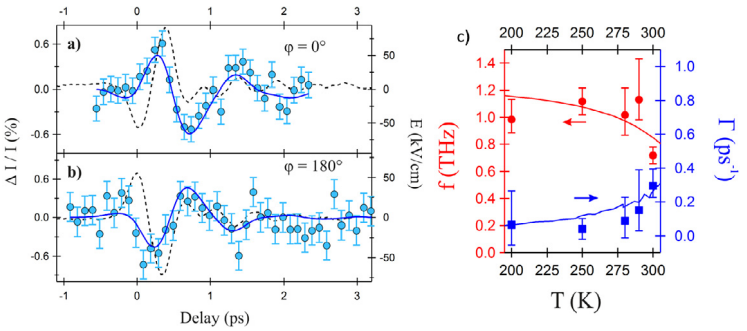

Fig. 13. Resonant excitation of the soft phonon mode in ferroelectric $Sn_2P_2S_6$ by an intense nearly single-cycle THz pulse. (a) Evolution of the diffracted peak intensity (symbols) in the upon excitation with the THz pulses of opposite polarity (dashed line shows the pulse profile) at $T = 300$ K. (b) Temperature dependences of the THz-driven mode frequency (red symbols) and damping factor (blue symbols) indicating the mode softening towards the transition to the paraelectric phase [90].
Source: The figure is taken from Ref. [90].

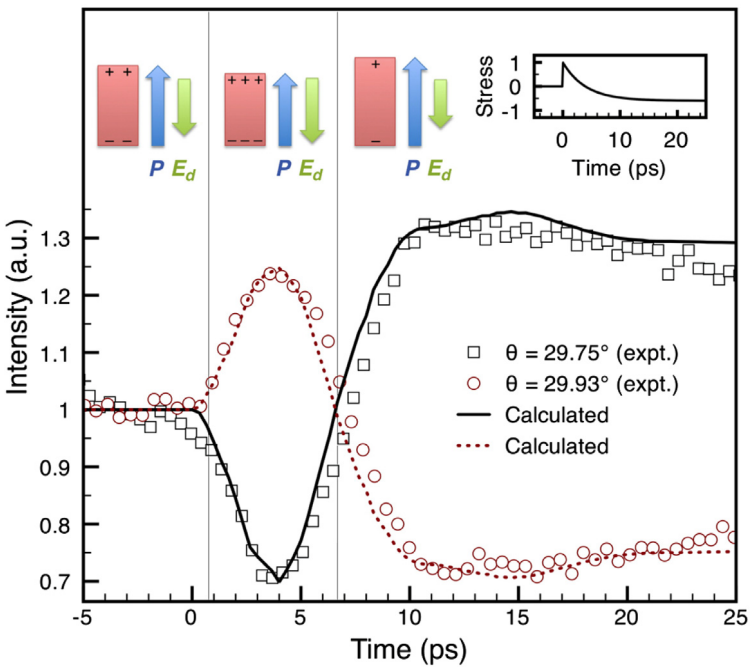

Fig. 14. Experimental evidence of the laser-induced decrease and increase of tetragonality in a thin film of $PbTiO_3$ resulting from the ultrafast dynamics of photogenerated carriers and screening fields [144]. Here the changes of tetragonality result from ultrafast screening of depolarizing field due to optically generated charges, as shown schematically in inset.
Source: The figure is taken from Ref. [144].

The soft phonon mode associated with the ferroelectric polarization can be excited by short laser pulses via several mechanisms. The amplitudes of the excited modes are, however, rather small, and switching to another bit state without any additional stimulus is still elusive. It was also argued that due to a small charge-to-mass ratio of the displaced ions resonant coupling of electric field of light pulses to a phonon mode is in principle not very efficient [145]. Nevertheless, as discussed in Section 5, continuous experimental and theoretical studies make us believe that ultrafast stable switching of spontaneous electric polarization between two bit states will be demonstrated in the nearest feature.

Electron mediated depolarization of ferroelectrics. The possibility to control the magnitude of the ferroelectric polarization via optical excitation of electronic transition has been discussed from very first experiments on photoferroics [68]. For instance, spontaneous polarization in thin ferroelectric films is partially screened by the depolarizing fields associated with surface changes. If such a ferroelectric is excited by light with photon energy exceeding the bandgap, the photogenerated carriers can affect screening fields. The impact of the photocarriers on the dynamics of ferroelectric properties was studied in a thin film of $PbTiO_3$ [144]. In the experiments thin $PbTiO_3$ films were excited by 80-fs laser pulses with

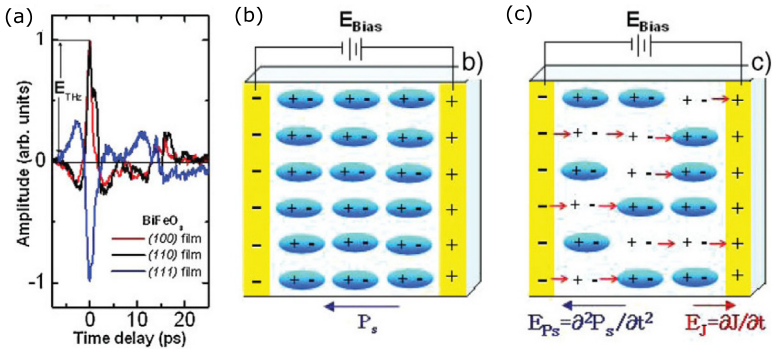

Fig. 15. (a) THz emission waveform ascribed to the laser-induced dynamics of the ferroelectric polarization in a multiferroics $BiFeO_3$ films with different orientations. (b) The schematic representation of the equilibrium ferroelectric state and the optically excited state where partially depolarized electric dipoles give rise to depolarization and photocurrents [146].
Source: The figure is taken from Ref. [146].

the central wavelength of 400 nm. The 60-fs X-ray pulses were used to probe evolution of the tetragonality — a signature of the ferroelectricity. The main observation (Fig. 14) was a decrease of tetragonality, which occurs on 5 ps timescale. It was followed by a lattice expansion at timescale of 10 ps and nanosecond relaxation. Since depolarizing fields play a crucial role in ferroelectric nanostructures, such an ultrafast photovoltaic effect should be the most pronounced there. Furthermore, it would be interesting to investigate the ultrafast photovoltaicity in ferroelectric nanostructures in a vicinity of a size-induced transition to paraelectric state.

Alternative route to achieve ultrafast depolarization in a ferroelectric via optical excitation of electrons has been considered in [146], where the impact on intrinsic ferroelectric polarization was believed to be achieved. The paper reports about THz emission from the multiferroic $BiFeO_3$ films upon excitation by femtosecond laser pulses with the central wavelength of 400 nm. The key observation in this experiment was a difference in THz emission from films with different orientations (Fig. 15(a)). It was shown that for the efficient THz excitation the material must be excited above the bandgap [147]. It was suggested, that the dynamics of the THz response can be associated with a change of the spontaneous polarization on a timescale of 1–2 ps. The authors suggested that the underlying mechanism of the spontaneous polarization dynamics is photoexcitation of localized electronic transitions in Fe ions, leading to temporal changes in distortion of the FeO_6 octahedra and reduction of the ferroelectric polarization (Fig. 15(b)). The authors pointed out that the density of the photoexcitation in the described experiments was too low to persistently depolarize the ferroelectric [147].

Understanding of the effect of the excitation of electronic transitions on structural dynamics was recently advanced by studying time-resolved X-ray diffraction in $Ca:SrTiO_3$ following the above-bandgap excitations [148]. Fig. 16(a) shows evolution of one of the X-ray diffraction peaks associated with the antiferrodistortive rotations of the oxygen octahedra ϕ. Using expression for the double energy potential (Eq. (24)) with carrier concentration-dependent frequency and repulsive force, the observed reduction of ϕ was successfully reproduced (Fig. 16(b)).

Thus, exciting real electronic transitions and realizing photodoping at the ultrafast timescale have an important effect on ultrafast lattice dynamics, both via changing the depolarizing fields (ultrafast photovoltaic effect), and via changing the potential energy profile. These effects may be used to facilitate switching between different ferroelectric states, as we discuss in Section 5.

Electronic ferroelectrics. In the examples considered above the ferroelectricity originates from ionic displacements, and an excitation of a particular phonon mode, i.e. lattice distortions, is required for modulation of a ferroelectric polarization. Direct coupling of the electric field of THz, IR or visible light to the order parameter in such ferroelectrics seems to be intrinsically weak due to small charge to mass ratio of ions forming the lattice. Instead, electronic ferroelectrics, where spontaneous polarization originates from charge redistribution rather than from ionic displacements [19], typically demonstrate larger susceptibility to the electric fields. As a result, it is expected that they should possess a stronger coupling of the "electronic" ferroelectric polarization to the electric field of light. This difference in light-ferroelectric polarization coupling manifests itself, for example, in stronger nonlinear optical signals obtained from electronic ferroelectrics [149]. Most of the discovered so far electronic ferroelectric are organic. Among inorganic solids, electronic ferroelectricity is present in the type II multiferroics with polarization being induced by magnetic ordering [32,33,150,151]. So far, the most attention was paid to photoexcitation of electronic organic ferroelectrics.

Ultrafast neutral-to-ionic transition. The organic compound tetrathiafulvalene-*p*-chloranil (TTF-CA) became the model material to study optically-induced changes of electronic ferroelectricity. This compound is built from chains of donor (D:TTF) and acceptor (A:CA) molecules. At $T_C = 81$ K the transition to the ferroelectric phase occurs as a result of the fractional charge transfer from A to D molecule. In [152] it is suggested that for the optical control of polarization of

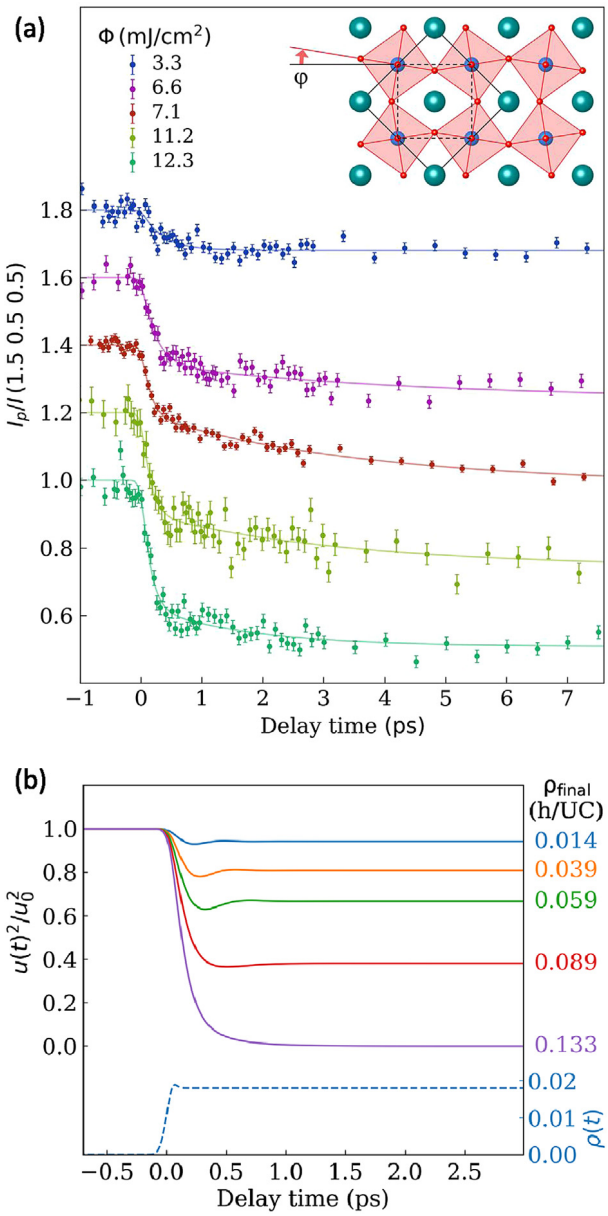

Fig. 16. Experimental evidence of the laser-induced decrease of the oxygen octahedra rotations (see inset) associated with the antiferrodistortive phase transition in Ca:$SrTiO_3$ [148]. Panels (a) and (b) show experimental results and results of calculations, respectively, for different levels of photodoping. Slower dynamics seen in (a) is associated with laser-induced heating.
Source: The figure is taken from Ref. [148].

this organic ferroelectric crystal one can rely on a cooperative phenomenon — laser-driven ultrafast local charge transfer yielding a change in valency of large number of molecules. In order to reveal the related structural changes associated with changes of the ferroelectricity, the impact of 300 fs near-IR laser pulse on TTF-CA was investigated in [153] using time-resolved X-ray diffraction. Studies confirmed the suggested earlier scenario for emergence of the laser-induced ferroelectric phase [152]. In the experiments [153], laser-induced nanodomains forming three-dimensional chains yielded additional peaks in X-ray diffraction (Fig. 17(a)). In [152] using time-resolved spectroscopic optical studies it was shown that light causes a destruction of ferroelectricity at a similar timescale (Fig. 17(b)). Thus, optically-induced charge transfer facilitates both the transition from the ferroelectric to the paraelectric phase and vice versa. Dynamics of this process at shorter timescale was reported in [154]. Based on temperature dependence of the transient reflectively in the ionic phase, it was suggested that the charge transfer induces nucleation of the neutral phase within 2 ps. It is followed either

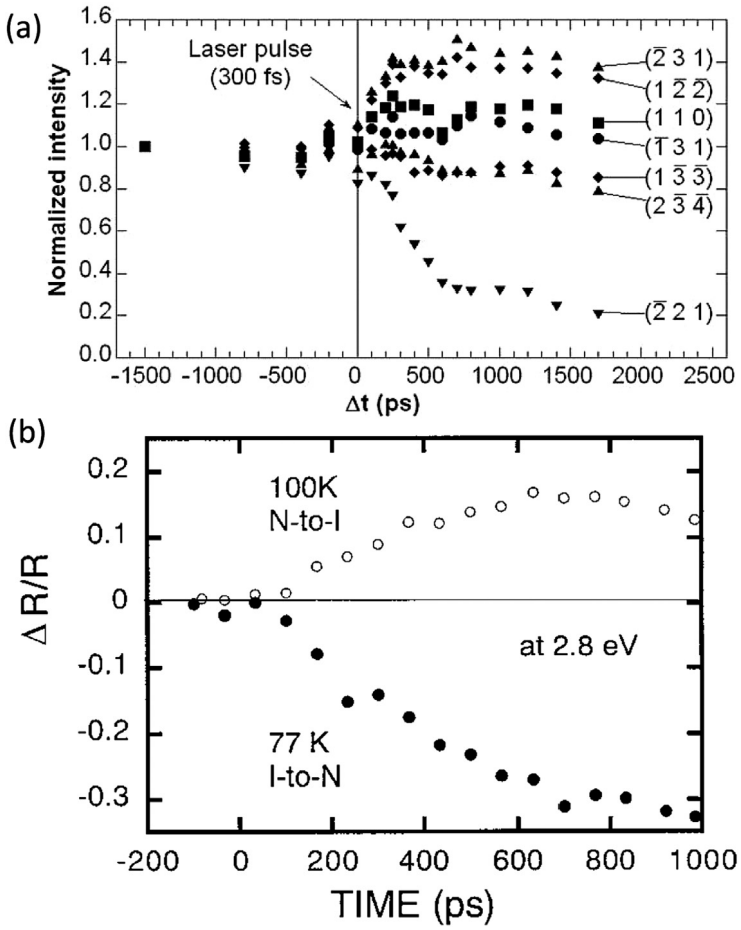

Fig. 17. Evidences of the laser-induced ferroelectricity of TTF-CA organic crystal. (a) Evolution of the X-ray diffraction peaks following photo-excitation of TTF-CA in a neutral state revealing emergence of the metastable ionic (ferroelectric) state at a subnanosecond scale [153]. (b) Photo-induced transitions between neutral and ionic phases and vice versa detected by means of time-resolved reflectivity measurements [152].
Source: The figures are taken from Refs. [152,153].

by formation of macroscopic neutral phase domain within if the sample temperature is close to T_C or by the relaxation at lower temperatures.

In [155] it was also suggested to use short THz pulses to trigger the charge transfer and to induce ferroelectricity in the neutral phase of TTF-CA crystal. In the experiment, the ferroelectric polarization was probed by measuring the Second Harmonic Generation and reflectivity for the probe pulses. During the overlap of the THz pump and the probe pulses, the signal first followed the electric field of the driving THz pulse and afterwards revealed oscillations (Fig. 18 (a–c)). These observations are explained in terms of a change transfer between D and A molecules driven by the THz electric-field. The charge transfer induces a motion of domain walls between nanoscale neutral and ionic (i.e. charged) domains. This motion is suggested to be intrinsically fast, because of primary electronic nature of ferroelectricity in the ionic domains. The oscillations in the signal originate from the coherent oscillations, or breathing, of the neutral-ionic domain wall (NIDW) pairs. In [155] it was estimated that the THz-induced polarization amounts to about 17% of the polarization below T_C.

THz excitation was suggested to lead to modulation of the electronic polarization in TTF-CA (Fig. 18(d–g)) also as a result of charge transfer within DA dimers [145]. Despite many reports about modulation of the ferroelectric polarization in electronic ferroelectrics, the reported light-induced changes were in general short-living. No switching was reported, but it was emphasized that using second harmonic generation as probe of ferroelectricity can seriously complicate interpretation of experimental results [156,157].

Ultrafast changes of ferroelectricity via melting of change ordering. Ferroelectric based on Bis(ethylenedithio)-tetrathiafulvalene (BEDT-TTF) molecules is an example of electronic ferroelectric with the polarization due to charge ordering. In these compounds the charge ordering occurs below the transition temperature T_C yielding the net ferroelectric polarization. Depending on particular structure, these compounds upon the transition to the charge-ordered state

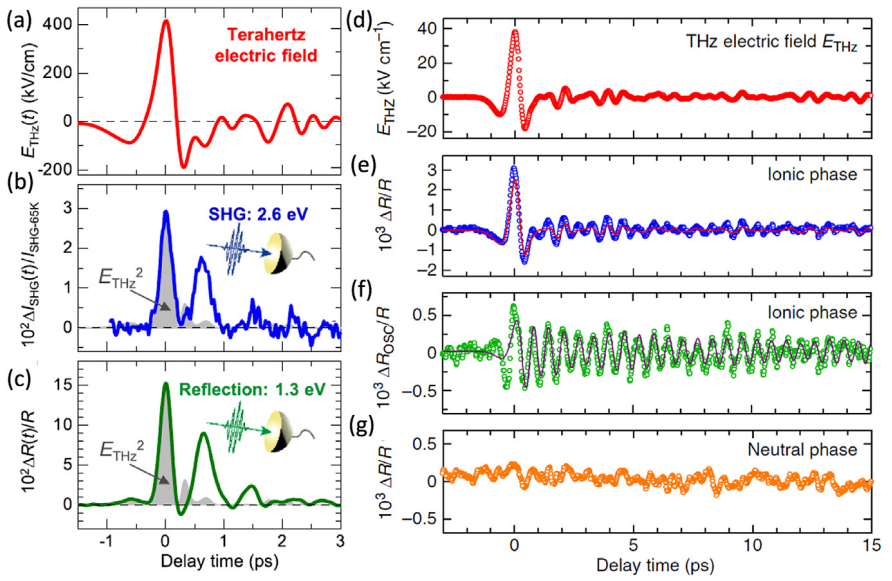

Fig. 18. THz-induced response of TTF-CA in the neutral and ferroelectric phase [145,155]. Single-cycle THz electric field (a) drives the changes of SH intensity (b) and reflectivity (c) of TTF-CA in the neutral phase, which include coherent component associated with NIDW pairs oscillations. Excitation with the single-cycle THz electric field of the TTF-CA in the ferroelectric phase (d) yields changes in the reflectivity within 1 ps ascribed to the ferroelectric polarization modulation (e) followed by long-living coherent oscillations (f) due to molecular vibrations. No such response was found upon excitation of the neutral phase (g).
Source: The figures are taken from Refs. [144,155].

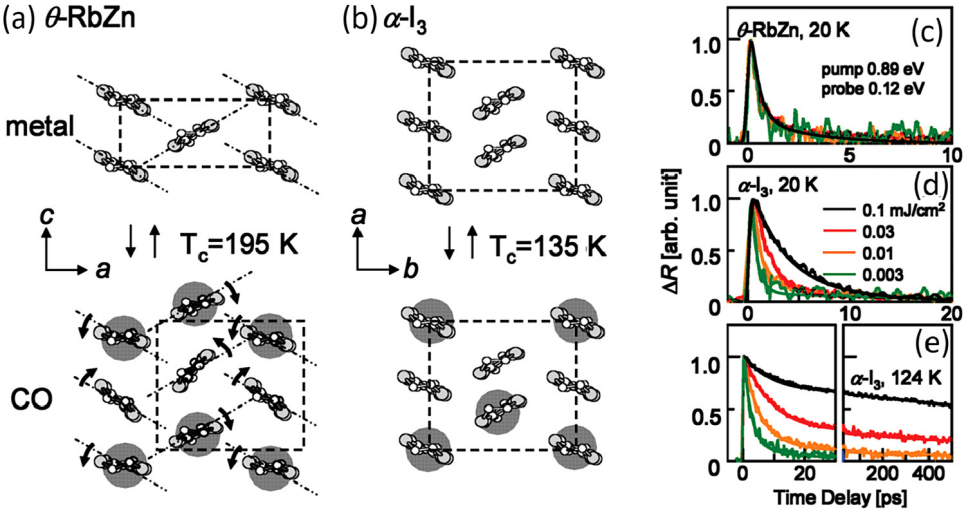

Fig. 19. Ultrafast laser-induced melting of the change ordering in BEDT-TTF-based electronic ferroelectrics [158]. Electronic and crystal structures of (a) θ-RbZn and (b) α-I_3 compounds above and below T_C. Transient reflectivity changes in (c) θ-RbZn and (d,e) α-I_3 showing different relaxation dynamics of the nonequilibrium metallic state.
Source: The figure is taken from Ref. [158].

demonstrate different degree of structural changes. Thus, in θ-(BEDT-TTF)$_2$RbZn(SCN)$_4$ [θ-RbZn] the charge ordering is accompanied by pronounced structural changes including molecular rotations (Fig. 19(a)). In contrast, in α-(BEDT-TTF)$_2$I$_3$ [α-I$_3$] the structural changes are less pronounced (Fig. 19(b)). Thus, these compounds are interesting materials to study ultrafast laser-driven dynamics in ferroelectrics. It is also remarkable that the transition to the ordered ferroelectric state in these compounds is also accompanied by the metal–insulator transition.

In [158] transient reflectivity of these two ferroelectrics was studied upon excitation by 200 fs laser pulses with photon energy above the bandgaps. As expected, the ultrafast insulator–metal transition was observed, indicating melting of the charge-order. However, it was also found that the photoinduced metallic phase was rapidly relaxing in the θ-RbZn

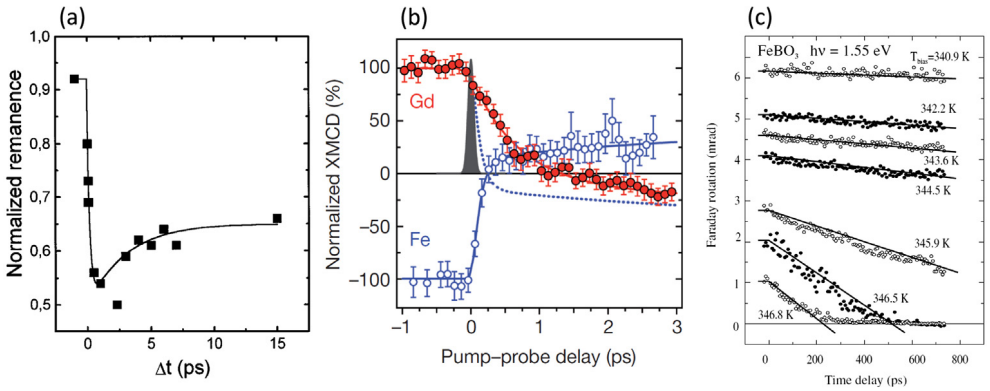

Fig. 20. Examples of ultrafast laser-induced demagnetization in (a) ferromagnetic metal Ni [29], (b) ferrimagnetic metallic alloy GdFeCo [162], (c) and antiferromagnetic dielectric $FeBO_3$ with a weak ferromagnetism [163].
Source: The figures are taken from Refs. [29,162,163].

(Fig. 19(c)), while longer relaxation was evident in α-I_3 (Fig. 19(d)). It was argued that in α-I_3 the laser excitation yields condensation of the induced metallic nanoareas into macroscopic domains, which then relax on a longer timescale. In θ-RbZn such a macroscopic domain formation is prevented by the fact, that metallic phase implied molecular rearrangement and costed more energy. This experiment shows that purely electronic ferroelectrics may be promising candidates for photoinduced switching of polarization.

4.3. Laser-induced heating

Energy of light deposited into a ferroic material inevitably causes temperature increase and a partial melting of ferroic order. Using femtosecond laser pulses even such a seemingly trivial effect as laser-induced heating can lead to counter-intuitive consequences in ferroics.

Magnetically ordered media. In thermodynamics, demagnetization is a result of a heat deposited into the spin system and an increase of the amplitude of spin fluctuations, which eventually leads to melting of magnetic order. Being proposed for explanation of very first experiments on ultrafast magnetism [29], three-temperature-model is still the main tool for description of ultrafast laser-induced demagnetization in metals. Even before the discovery of ultrafast collapse of magnetic order in ferromagnetic Ni, three-temperature-model was employed to describe a less surprising sub-nanosecond laser-induced demagnetization in magnetic metals [159]. The model describes a metallic magnet as three interconnected reservoirs of energy and angular momentum. First reservoir states for spins and its temperature defines the net magnetization of the medium. Second reservoir represents the conduction electrons. It is assumed that light interacts only with the electrons, deposits energy to the reservoir and increases its temperature. Low heat capacity of this reservoir allows subpicosecond heating of electrons to temperatures exceeding 1000 K i.e. far above the Curie point of most magnets. Third reservoir stays for the lattice. It has the largest heat capacity and eventually accepts most of the heat deposited into the medium. In the model, magnetization dynamics is described in terms of dynamics and balance of temperatures of electrons, lattice and spins.

Three-temperature-model was able to describe first experiments on ultrafast laser-induced demagnetization under assumption that electrons efficiently transfer heat to spins. Understanding the emergence and ultrafast dynamics of electron–spin correlations has become the key for understanding the fundamentals of ultrafast magnetism. Further experimental studies of ultrafast magnetization dynamics in other compounds led to observation of a large diversity of timescales of ultrafast demagnetization (Fig. 20(a,b)), but upgrades of the three-temperature-model were able to explain even the diversities that seem to be paradoxical [160]. Up to now, the simplistic three-temperature model was rather sufficient to explain ultrafast demagnetization of ferromagnets such as Ni, Fe, Co, NiFe, Gd and Tb. However, attempts to apply the model to ferrimagnetic alloys such as GdFeCo and GdCo revealed substantial shortcomings [161]. The most serious concern about validity of the three-temperature model is supported by experimental studies of laser-induced demagnetization in X-ray spectral range. It was shown that despite antiferromagnetic exchange interaction between the spins of two magnetic sublattices of ferrimagnetic GdFeCo, the magnetizations of the rare-earth (Gd) and transition metal (FeCo) sublattices are collapsed at different timescales (Fig. 20(b)) [162]. Consequently, it is also not correct to assign one temperature to all the spins in the ferrimagnet and an additional reservoir must be added to the model.

The temperature that can be reached by the reservoir of free electrons as a result of optical excitation depends on the electron mobility in the excited state. In an extreme case of dielectrics, for instance, even electrons excited into the conduction band are not mobile and cannot acquire a large kinetic energy. Therefore, a control of magnetism via laser-induced heating in dielectrics can be efficiently described by two-temperature model, where light first deposits energy to the lattice and the demagnetization occurs on a timescale of spin-lattice interaction [163].

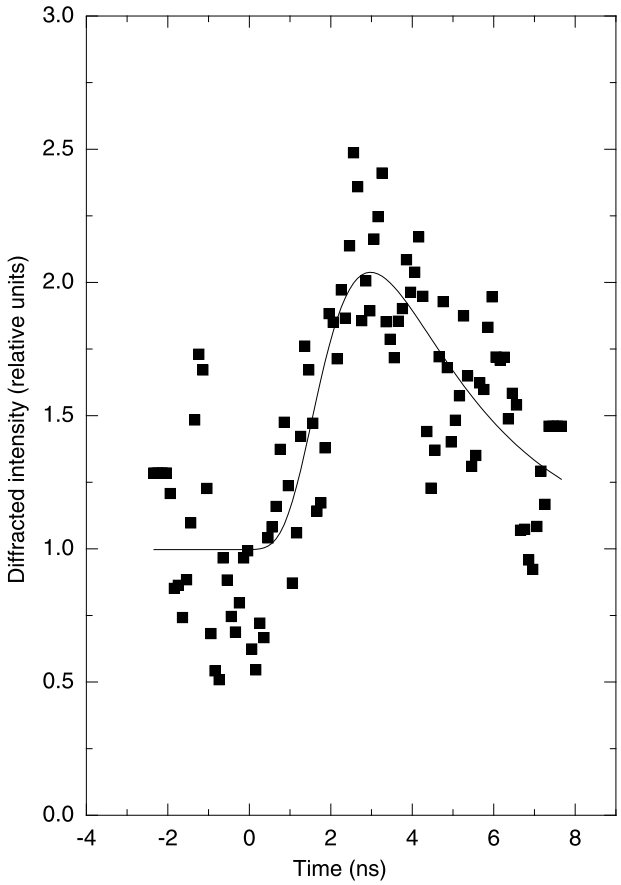

Fig. 21. Experimental evidence of the laser-induced depolarization in a ferroelectric DKDP [168]. The material was pumped with femtosecond laser pulses at the central wavelength of 800 nm and the laser-induced effect on ferroelectricity was probed using X-ray diffraction with 100 ps resolution. *Source:* The figure is taken from Ref. [168].

Although in first experiments three-temperature model discussed only the effect of temperature on the net magnetization, soon it was realized that magnetic anisotropy is also sensitive to temperatures of electrons and phonons. Ultrafast lattice heating results in a change of magnetic anisotropy. In [164] it was shown that the magnetic anisotropy of metallic Co changes upon a relaxation of the initial hot electron distribution and the lattice temperature increase. Similar results have been demonstrated in a number of other metals (Fe [165], FeGa [166]). In dielectrics the changes of magnetic anisotropy were also shown to occur on a timescale corresponding to the lattice temperature increase [167].

Ferroelectrics. Since ferroelectricity is incompatible with metallic properties, one cannot rely on free electrons as a reservoir that mediates optical control of ferroelectricity. Upon interaction with ferroelectrics light can generate phonons. The phonons thermalize and this process leads to a lattice temperature increase as well as melting of ferroic order. The effect of laser-induced lattice heating on ferroelectric order parameter of deuterated potassium dihydrogen phosphate (DPDP) was studied in [168] using time resolved X-ray diffraction as a probe of ferroelectricity. The authors observed that upon excitation of the DPDP sample in the ferroelectric phase by a laser pulse, the intensity of the diffraction peak associated with the paraelectric phase increases (Fig. 21). It was found that the laser pulse heats the lattice by about 5 K and thus lowers the barrier between ferroelectric and paraelectric states.

Effect of the laser-induced heating on the ferroelectricity in the material with nominally displacive transition to the paraelectric phase was reported in [169]. Using the same X-ray diffraction technique, the authors investigated evolution of the tetragonal distortion and the ionic displacements responsible to the ferroelectric polarization in $BaTiO_3$. It was found that the laser excitation of the $BaTiO_3$ powder results in a decrease of the tetragonal distortions within 100 ps (Fig. 22). The effect of the tetragonal distortions corresponds to a temperature increase about 5 K towards the paraelectric phase.

To summarize, one can highlight a similarity between dielectric magnetically-ordered media and ferroelectrics. Hence one can also expect similarities between scenarios for optical recording on magnetic dielectric and ferroelectrics. However, it is also possible that mechanisms of laser-induced switching of magnetization discovered in metals can inspire discoveries of novel routes for switching of polarization in ferroelectrics. Therefore, in the following Section we also review ultrafast magnetic switching in metals and speculate about possible mechanisms of switching in ferroelectrics.

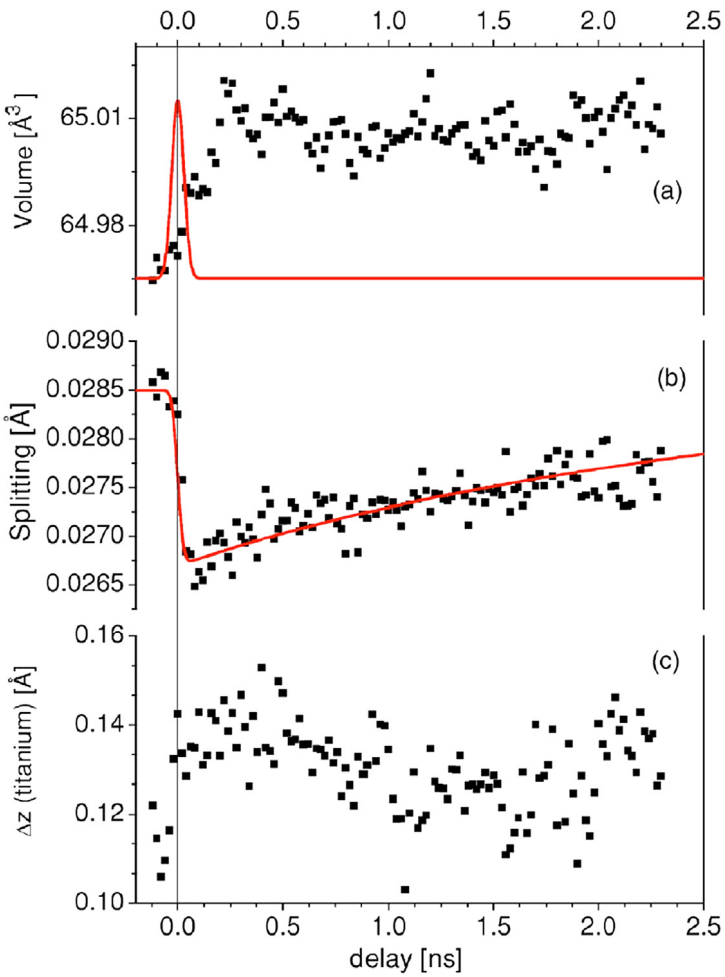

Fig. 22. Laser-induced evolution of the unit cell volume, tetragonal splitting and the Ti ions displacements upon laser-induced excitation of $BaTiO_3$ at $T = 284$ K [169]. While the tetragonal splitting (b) reduces with the experimental resolution of 100 ps, the unit cell volume changes on a longer timescale (a), and the reduction of Ti ions displacements could not be detected (c).
Source: The figure is taken from Ref. [169].

5. Routes for switching of order parameter by light

5.1. Photoferroic domain wall motion

Thermodynamically, order parameter in ferroics is a first derivative of thermodynamic potential. According to the classification of Ehrenfest, switching polarization, magnetization, antiferromagnetic vector in ferroics between metastable states can be seen as first order phase transition. Kinetics of such phase transitions includes two steps: (a) emergence of the nuclei's i.e. domains of the new phase; (b) growth of the domains corresponding to the new phase. Hence understanding the mechanisms of photoinduced change of ferroic domain patterns can be the key to ultrafast and energy efficient photoferroic recording.

The discovery of photomagnetic recording [60] was followed by a number of theoretical works with a particular attention to effects of light on domain structure of polydomain ferroics. Readjustment of domain structure of magnetic and ferroelectric materials was studied theoretically in [170,171]. In both cases, it was important that different magnetic or ferroelectric domains interact with polarized light differently. For instance, due to magnetic circular dichroism, circularly polarized light propagating through a magnetic material will be absorbed differently in the domains with the magnetization parallel and antiparallel with respect to the wave-vector of light. In ferroelectrics, circular dichroism could be achieved due to gyrotropy. In particular, $Pb_3Ge_3O_{11}$, triglycine sulfate (TGS), Rochelle salt are examples of gyrotropic ferroelectrics for which the sign of optical activity depends on the direction of the ferroelectric polarization and thus changes from domain to domain. According to the Kramers–Kronig ratio, optical activity must result in circular dichroism and thus absorption of light will differ in domains with opposite ferroelectric polarizations. Interestingly,

earlier theories completely ignored optically induced heating. It was assumed that absorption of light can efficiently change exchange parameters in magnetic materials or recharge impurities in ferroelectrics. Different absorption would mean that in adjacent domains the order parameter changes differently and the domain walls move in the direction defined by the polarization of light. It was also proposed that the effect of light on the exchange parameters in magnetic materials can be employed to control domain structure and cause a collapse of bubble domains [172]. Recent theoretical studies propose that effect of light on the exchange interaction can be employed to induce even more complex spin textures such as anisotropic skyrmions [173]. Although the fact that light can change the exchange parameters was proven experimentally [102], if the effect of light on the exchange parameters can be large enough to facilitate such a dramatic reconstruction of domain pattern is still an open question.

However, it seems that even if these complex effects are not present in ferroic media, dichroism in ferroic domains must lead to domain wall motion simply due to different heating of different domains by light. It is known, for instance, that in ferro- and antiferromagnetic media a temperature gradient will move domain walls towards hotter region [174]. Indeed, magnetic circular dichroism and laser-induced heating were employed to explain experimental observations of optical control of the net order parameter in polydomain ferromagnets Co/Pt, FePt [175,176] and ferrimagnetic iron garnet [177]. Mechanisms allowing laser-induced control of domain structure are also a subject of discussion in antiferromagnets. Laser-induced heating has been shown to be the main mechanism in controlling the domain structure of antiferromagnets with light in MnF_2 [178] and $TbMnO_3$ [179].

At the same time, experimental observations of optical control of the net order parameter in polydomain ferromagnetic $CdCr_2Se_4$ [180,181] and FePt [182] were explained without relying on heating. In particular, it was suggested that absorption of circularly polarized light can induce magnetization with the direction defined by the helicity of light. However, if this mechanism is efficient, photo-induced magnetization must be observed in both polydomain and single domain materials. Recently it was reported that circularly polarized subpicosecond laser pulses are able to excite oscillations of spins in $CdCr_2Se_4$ and the phase of the oscillations was defined by the helicity of light. The maximum of the oscillations amplitude was observed at the energy of one of maxima of optical absorption of $CdCr_2Se_4$ [183]. Despite several experimental reports of laser induced spin dynamics in FePt [184,185], no helicity dependent magnetization dynamics in monodomain FePt samples was reported until now. Hence the validity of the assumption of a large laser-induced helicity dependent magnetization in FePt is still to be confirmed.

Very unusual effect of light on domain structure was observed in Ni-doped $FeBO_3$. This is antiferromagnetic material with canted sublattices due to the Dzyaloshinskii–Moriya interaction. Illumination of the material with light of a rather low intensity in an external magnetic field results in a dynamic instability of the domain structure. In particular, the whole pattern starts to move in the direction perpendicular to the magnetic field [186]. Photoactivation of Ni-dopants seems to be crucial for the phenomenon and optically induced heating does not seem to play for the mechanism of the dynamic instability a decisive role.

Recently the possibility to move domain walls with electric currents, THz radiation and a combination of both was shown for antiferromagnetic CuMnAs [187]. The total magneto-resistance of the polydomain sample was modified by nearly single-cycle pulses of THz radiation and a train of such pulses were shown to result in a change of resistance. Based on the fact that the THz-induced switching was dependent on polarization and the direction of switching was fully determined by the polarization of the THz pulses, it was proposed that the switching must occur due to the torque acting from the electric current on the Néel vector. For the very same reason it was argued that the switching is completed within the duration of a pulse, (a picosecond or less). However, it is also clear that owing to the magneto-resistance used for the detection of antiferromagnetism, heat dissipated during the flow of electrical current through such a sample will depend on the angle between the current and the Néel vector of the antiferromagnetic domain. In the case of 90°-magnetic domains, taking magneto-resistance at the level of 0.1% and absorbed pump energy density of 1 kJ/cm^3, one finds that electric current at a THz frequency must result in a temperature gradient of 0.1 K over an antiferromagnetic domain wall. Such a gradient must push the domain wall towards hotter regions [174]. The domain wall motion will change the net Néel vector and thus the total magneto-resistance of the sample in a polarization-dependent way.

There is also an ongoing research on optical control of ferroelectric domains. For instance, in [188] optically-induced strain and flexoelectric effect [189] was shown to control the coupled ferroelectric and magnetic domains in $BiFeO_3$. Photovoltaic effect was also shown to enable manipulating ferroelectric domains and domain walls in lead-free ferroelectric ceramic [190]. However, to the best of our knowledge, no studies of ferroelectric domain walls dynamics in response to ultrafast optical excitation have been reported.

It looks that the only way to prove and reliably interpret the ultrafast impact of light on the domain wall is to measure the dynamics of the effect. For instance, in [177,181] the dynamics of the laser-induced polarization dependent domain wall motion was successfully detected in polydomain in iron garnet films with temporal resolution of 1 ns. In particular, it was shown that the domain wall moves on a timescale which is much longer than duration of the pump pulse (5 ns) and the net magnetization continues increasing up to 120 ns. Such a slow magnetization dynamic is typical for processes induced by laser-induced heating of magnetic dielectrics. It seems that recent developments of experimental techniques do have the potential to reveal ultrafast effects of light on ferroic domains and mechanisms of polarization dependent domain wall motion which do not rely on heat. Even using visible light and the phenomenon of diffraction on magnetic domains it was possible to detect domain wall motion with spatial and temporal resolution down 5 nm and 1 ns, respectively [191]. The advanced X-ray and XUV sources open up a plethora of opportunities of these studies in the nearest future [192].

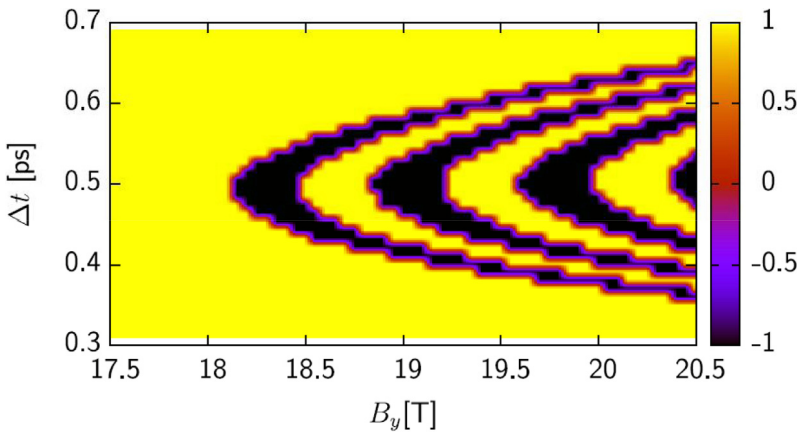

Fig. 23. Proposal for switching antiferromagnetic vector **L** in NiO by THz laser pulse without applied magnetic field [198]. Results of simulations show that either system relaxed back into its initial equilibrium state (yellow areas), or relaxes to the state with switched **L** (black areas). The switching depends on pulse duration Δt and the strength B_y of the THz magnetic field orthogonal to the antiferromagnetic vector $\mathbf{L}\|z$ [198]. Color scheme indicates the value of the normalized antiferromagnetic vector $\mathbf{l}_z$.
Source: The figure is taken from Ref. [198].

5.2. Routes for switching of magnetic order parameter in a single domain

Routes. Intuitively, the fastest way to reverse magnetization is to apply the magnetic field perpendicular to the magnetization. In the simplest case of isotropic medium, the field will launch a precession in the plane perpendicular the field. If the field is present only during half a period of the precession, the magnetization will be reversed much faster and independently on damping [14].

The aim to achieve the fastest possible magnetization reversal motivated experiments performed with picosecond pulses of magnetic field generated by short bunches of relativistic electrons due to the Ampere's law. Experimental design was such that the generated magnetic field was directed perpendicular to the magnetization of a metallic ferromagnetic Co sample [193–195]. The experiments showed that it is possible to switch magnetization with magnetic field pulse as short as 2.3 ps. However, the switching was not homogeneous and the areas with the switched and unswitched magnetization were found. It was also realized that picosecond magnetic field is accompanied by a rather high electric field with the strength up to 10 MV/cm. It was suggested that this field can substantially affect the process of magnetic switching by changing the magnetic anisotropy of the medium [196], and may be responsible for inhomogeneous switching. These experiments were performed without any temporal resolution, and no actual dynamics of the ultimately fast switching was studied. The dynamics of the processional switching was eventually revealed in time-resolved experiments using less intense but longer pulses with duration of about 100 ps [14,197].

Computational studies claimed that the efficiency of resonant coupling of THz magnetic field to antiferromagnetic order parameter can be sufficient to cause a switching in NiO [198]. This theoretical proposal has not been confirmed experimentally yet, but revealed a characteristic periodicity in conditions required for the switching versus the THz field strength and duration (see Fig. 23).

Heat-assisted switching in external magnetic field. How can light steer magnetization reversal? Currently envisaged paradigm of next-generation hard-disk-drive technology – heat-assisted magnetic recording (HAMR) – employs light-assisted magnetization reversal in antiparallel magnetic field [199,200].

Recently, a novel scenario of heat-assisted magnetic recording in perpendicular magnetic field was demonstrated for iron garnet. Instead of thermally leveraging the magnetization to assist the process of reversal, it was conjectured that one could rather thermally leverage the growth-induced magnetocrystalline uniaxial anisotropy field [201]. Taking iron garnet film with a strong out-of-plane magnetic anisotropy and applying the magnetic field in the sample plane, one sets the magnetization in a state defined by the interplay of the applied magnetic field and the field of magnetic anisotropy. It is known that in iron oxides absorption of light can cause a redistribution of electrons among energy states, resulting in a subpicosecond reduction of the magnetocrystalline anisotropy, while the demagnetization is negligible. This effect leads to an emergence of a transient effective torque [167], inducing precession of the magnetization around the new orientation of the effective magnetic field. In [201] this scenario has been realized in Bi:YIG upon excitation with laser pulses with the central wavelength of 400 nm allowing a significant reduction of magnetic anisotropy. The cone angle of the magnetization precession appeared to be large enough to cause the magnetic switching within one half of a precessional period (Fig. 24).

It is important to note that the switching described above results in formation of concentric rings with switched/non-switched magnetization (Fig. 24). Such pattern is, in fact, the characteristic feature of coherence-mediated switching of order parameter in ferroics. Such patterns were found, for instance, in experiments with switching of magnetization by

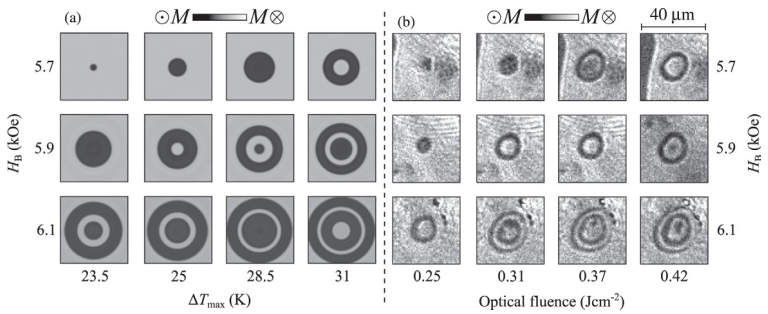

Fig. 24. Single-shot switching of the magnetization in Bi-substituted yttrium iron garnet in an applied magnetic field via laser-driven reduction of magnetic anisotropy [201]. (a) Snapshots of the numerically calculated spatial distributions of the out-of-plane component of magnetization, taken 3.5 ns after the instantaneous modification of the magnetocrystalline anisotropy field. The images were obtained by varying the strength of the in-plane bias magnetic field $\mathbf{H}_B$ and the maximum temperature change ΔT_{max}, as indicated. (b) Typical experimental raw snapshots of the out-of-plane component of magnetization, acquired 3.5 ns after exposure to the ultrashort optical pulse. These snapshots were obtained with different optical fluences and magnetic field strengths as indicated [201].
Source: The figure is taken from Ref. [201].

ultrafast magnetic field described above [193–195]. Patterns similar to those shown in Fig. 24 were also obtained in computational studies of switching in antiferromagnetic NiO (see Fig. 23).

To explain a formation of the pattern of switched and not-switched areas in experiments with heat-assisted switching in garnets (Fig. 24), one has to note that, upon excitation, the total effective magnetic field, given by a superposition of the effective field of the magnetic anisotropy and the applied magnetic field, changes and the magnetization starts to precess around new equilibrium orientation. The frequency of the precession depends on the total effective magnetic field. Due to Gaussian spatial distribution of the intensity in the laser beam, the strength of the total effective magnetic field also has a Gaussian distribution. In order to understand how such a laser-induced inhomogeneous pattern of magnetic anisotropy affects the final state of the magnetization the authors of Ref. [201] performed simulations using the Landau–Lifshitz–Gilbert equation. In the simulations, the effective magnetic field was composed of an out-of-plane uniaxial temperature-dependent anisotropy field and a spatially uniform in-plane bias magnetic field. In order to mimic the thermal load delivered by an ultrashort optical pulse with a Gaussian intensity profile, the temperature of the medium was instantaneously increased and subsequently restored on a nanosecond timescale, across a Gaussian distribution in the sample plane. The simulations allowed to observe the precessional reversal of magnetization, as shown in the top left panel of Fig. 24. Increasing either the external magnetic field and/or the maximum temperature change leads to formation of "bullseye" domain patterns of magnetization, whereby the radially symmetric distribution of magnetization alternately has positive and negative projections on the normal to the sample. If upon recovery of the magnetic anisotropy the magnetization has precessed for an odd (even) number of half-periods, the magnetization is rendered switched (unswitched). Thus, in the described experiments this bullseye pattern emerges because temporarily diminished anisotropy field has a spatial distribution. It is in contrast to the experiments with the pulsed magnetic fields [193–195], where the pattern is likely to originate from interplay between spatial distribution of the switching magnetic field and electric field induced change of the anisotropy. This feature of the coherent switching is clearly a general one for all types of ferroics. Therefore, attempting to observe the laser-driven switching, one may need to keep this feature in mind and perform the experiments allowing imaging of the switched area.

Finally, we note that, despite the fact that the iron garnet films are known to have exceptionally small damping, the experiments surprisingly revealed that the laser-induced magnetization precession robustly leads to switching of the magnetization within just one half of a period. Exploring this route of the switching the authors [201] concluded that the damping is a function of the precession amplitude. For small amplitudes of the spin precession the damping is found to be indeed small. Upon an increase of the precession amplitude the damping increases dramatically reaching anomalously large values (Fig. 24).

As a matter of fact, this scenario of switching steers the magnetization along the ultimately fastest route. Moreover, unlike the HAMR approach, this alternative route of the heat-assisted switching does not require heating the material up to the Curie temperature, it does not require destroying magnetization and no energy flow from lattice to spins is involved. Therefore, the route is not only faster, but also more energy efficient than the one followed in the conventional HAMR. The switching can be realized in materials with a weak spin-lattice interaction, such as iron oxides and iron garnets, in particular. Among the disadvantages of this scenario of magnetization switching is the fact that the magnetization reverses after every laser-excitation event independently on the polarity of the applied magnetic field.

Heat-assisted switching without external magnetic field. Periodic switching conditions and ring-like pattern were also experimentally revealed in (Sm,Pr)FeO_3 orthoferrite upon excitation with single circularly polarized laser pulses when no external magnetic field was applied [108]. The work reported about dynamics of spin-reorientation phase transition triggered by circularly polarized light. Magnetic anisotropy of the rare-earth orthoferrites is characterized by a high

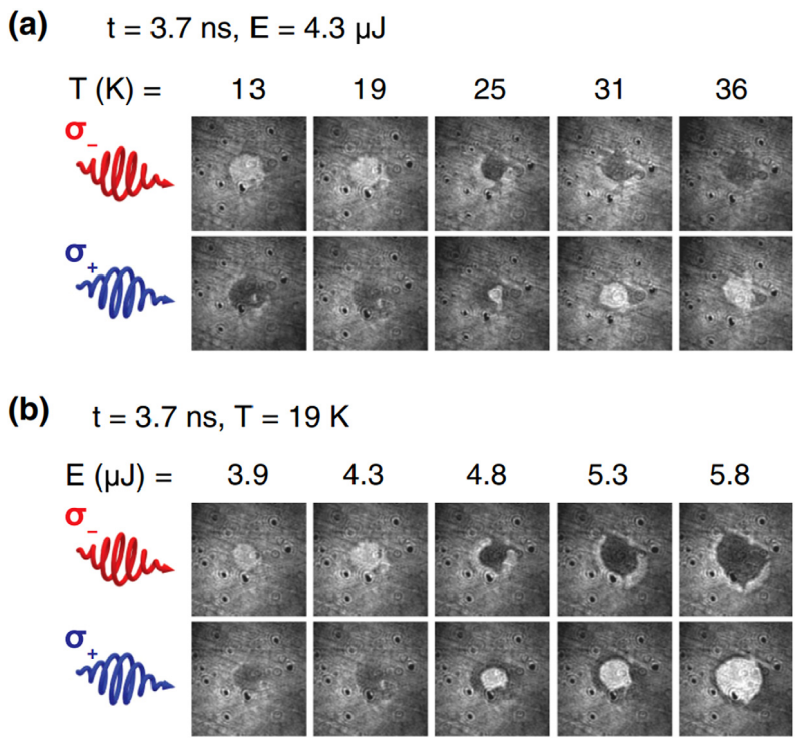

Fig. 25. Switching of magnetization in (Sm,Pr)FeO_3 via spin-orientation transition triggered by single circularly-polarized laser pulses [108]. Images show the magnetic state of the sample at 3.7 ns after the excitation with a right-handed ($\sigma_{(+)}$) or left-handed ($\sigma_{(-)}$) circularly polarized pulse. The orientation of magnetization can be controlled by (a) an initial sample temperature T (a), and (b) pump pulse energy E. Depending on the combination of T and E, pulses of the same helicity can write domains with opposite magnetization [108].
Source: The figure is taken from Ref. [108].

sensitivity to a temperature change yielding spin-reorientation transitions [202]. In particular, the orthoferrite is an antiferromagnet with weak ferromagnetism. A heating of (Sm,Pr)FeO_3 from 98 K to 130 K results in a rotation of the antiferromagnetically coupled spins over $\pi/2$ angle. The spin orientation is also accompanied by a rotation of the net magnetization over $\pi/2$ angle. The orientation of the sample studied in Ref. [108] was such that below 98 K the magnetization was in the sample plane, and above 130 K it was pointing along the sample normal. A conventional temperature increase resulted in a formation of a multi-domain state with nearly equal areas covered by the domains with the magnetization "up" and "down", respectively. However, it was found that the heating resulting from excitation with a single femtosecond circularly polarized light pulse led to a formation of a single domain with the magnetization defined by the helicity of light (see images on the left in Fig. 25(a,b)).

The results, in particular, unexpected sensitivity to the laser pulse polarization, were explained by a two-fold effect of the laser pulse. First, due to laser-induced heating the magnetic anisotropy changes and a new potential barrier grows up separating two states with the magnetization "up" and "down", respectively. Second, due to the coupling of light to spins via inverse opto-magnetic effects, femtosecond laser pulse triggers spin oscillations at the frequency of the spin resonance and with the phase defined by the helicity of light (Fig. 25). Depending on mutual timing between the period of the spin oscillations and the growth of the potential barrier, when the barrier becomes larger than the amplitude of the spins oscillations the spins got caught by one or another potential minimum. Changing the helicity of light one changes the phase of the oscillations and this results in reversal of the magnetization in the final state. Changing the pump fluence and the temperature of the sample, one changes the growth time of the barrier and thus can affect the switching conditions. Similarly to the case of a ferrimagnet garnet, in the experiments with the antiferromagnetic (Sm,Pr)FeO_3 Gaussian distribution of the intensity of the laser beam resulted in a ring-like structure of the magnetization in the final state (see images on the right in Fig. 25(b)). Therefore, owing to such combination of effects, the route of the phase transition can be controlled through three independent parameters, the pulse polarization, the initial temperature and the laser fluence. Similarly, one can combine the ultrafast inverse Cotton–Mouton effect and laser-induced heating over the spin reorientation transition, as demonstrated in another rare-earth orthoferrite $DyFeO_3$ [203].

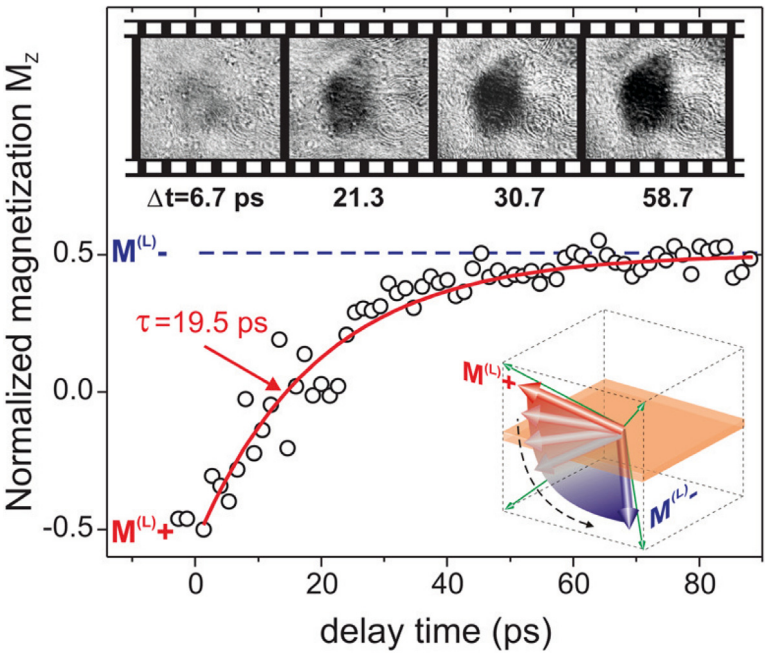

Fig. 26. Laser-induced all-optical switching of the magnetization in Co-doped yttrium iron garnet by linearly-polarized 40-fs laser pulses, as observed by femtosecond single-shot imaging [98]. Lower part of the figure shows the temporal evolution of the out-of-plane projection of the sample magnetization in the center of the images following excitation. Inset depicts the switching trajectory. The switching is precessional and occurs within 19.5 ps.
Source: The figure is taken from Ref. [98].

5.3. Heat-free switching without external magnetic field

From the first experiments on photo-magnetism it was realized that light can not only destroy, but also strengthen or modify magneto-crystalline anisotropy of magnetically-ordered media (see Section 2). For instance, in Si-doped and Co-doped iron garnets it was shown that effect of light on magnetic anisotropy can change the domain pattern [61]. Recently this effect of light on the magnetic anisotropy was employed to realize ultrafast all-optical magnetic recording on a dielectric medium without any involvement of magnetic field and nearly no heating. In particular, the experiments were performed with Co-doped YIG thin lm (YIG:Co) with (001) plane of the sample. In this material, the Co dopants are responsible for strong magnetocrystalline anisotropy, making cube diagonals ⟨111⟩ to be eight easy magnetization directions. Moreover, Co-dopant are responsible for a dramatic increase of the damping in the iron garnet. First, in the experiment the sample was brought in a magnetic state with two domains having the magnetizations along $[1\bar{1}1]$ and $[\bar{1}\bar{1}\bar{1}]$ axes, respectively. The domains corresponding to these states were visualized with the help of polarizing microscope. Due to a slight miscut of the sample from the pure (001) orientation, the domains with the magnetizations along $[1\bar{1}1]$ and $[\bar{1}\bar{1}\bar{1}]$ axes were seen as large bright and small dark areas [204]. Afterwards the material was excited by a single 40 femtosecond laser pulse with polarization along [100] crystallographic direction. For laser pulses with intensities above the threshold value such an excitation led to a reversal of the magneto-optical contrast. The magnetization from the $[1\bar{1}1]$ state was switched to the $[11\bar{1}]$ state and thus formed large bright domain on the magneto-optical image, while the magnetization from the $[\bar{1}\bar{1}\bar{1}]$ state was switched to the [111] state forming small bright domain. It was shown that the switching is accompanied by record low energy dissipations. Using femtosecond single shot imaging it was possible to reveal that after the laser excitation the magnetization switches to another metastable state via heavily damped precession (see Fig. 26).

Ref. [205] computationally studied routes of the switching between different states in the iron garnet with photo-induced magnetic anisotropy. It was shown that the switching process can be controlled not only by light with polarizations along [100] and [010] crystallographic axes of the garnet, but also light with polarizations along [110] and $[1\bar{1}0]$ can steer the magnetization to different metastable states. The direction of the switching appeared to be also sensitive to the pump fluence and the damping. The same study revealed the switching with two pairs of polarizations (either along [100] and [010] or along [110] and $[1\bar{1}0]$ crystallographic axes) has maxima of the efficiency at very different wavelengths of light. If the switching by a pair of polarizations along [100] and [010] axes has the maximum efficiency at the wavelength of 1300 nm, the switching with the second pair is the most efficient at 1140 nm. It was shown that the transition at 1.1 eV corresponds to electronic transition from state $^4T_1$ to state $^4T_2$ in Co^{2+} ions in octahedral

sites. A broader peak at 0.95 eV corresponds the transition from $^4A_2$ to $^4T_1$ in Co^{2+} ions in tetrahedral sites. Therefore, after bringing all the findings together, it becomes clear that the effect of photo-magnetic recording is based on optical excitation of $d-d$ transitions in Co^{2+} ions in tetrahedral and octahedral cites. While unperturbed Co-doped garnet has four axes of magnetic anisotropy, the laser excitation strengthens different axes depending on the polarization and causes reorientation of the magnetization to another stable state. Recent experiments revealed that the maximum frequency at which magnetic bits can be written and erased can reach 20 GHz, which is the highest frequency demonstrated in magnetic recording until now [206]. Another interesting research direction is related to studies of photomagnetic recording in a broader class of materials. The key element of photomagnetic recording is the need for strongly anisotropic, optically susceptible ions in media such as Co^{2+}. Next to Co:YIG, the examples of suitable materials are ferrites (garnet, spinel, ortho- and hexaferrites, ferroborates, and magnetite) and perovskites. For instance, a strong contribution to single ion magnetic anisotropy is typical for 3*d* (e.g., Mn^{2+}, Mn^{3+}, Cr^{2+}, Cr^{3+}, Fe^{2+}, Co^{2+}, Co^{3+}, Ni^{2+}, Ni^{3+}), 4*d* (Ru^{3+}, Ru^{4+}), 5*d* (Ir^{3+}, Ir^{4+}), and 4*f* (Ce^{3+}, Tb^{3+}) elements. Hence there is a plethora of opportunities for further studies of new materials for ultrafast photomagnetism and photomagnetic recording in the nearest future.

5.4. Magnetic switching via strongly non-equilibrium state

Principally different route for laser-induced magnetization reversal via a strongly non-equilibrium state was discovered in metallic ferrimagnets. This reversal in zero applied magnetic field does not involve precession and proceeds via a state with quenched magnetization. The discovery of all-optical helicity dependent magnetic switching in ferrimagnetic GdFeCo alloys [207] triggered a search for the mechanisms responsible for the effect. GdFeCo is a ferrimagnet with two antiferromagnetically coupled sublattices comprised by spins of Gd and FeCo, respectively. GdFeCo is a metal, and ultrafast spin dynamics in GdFeCo can be conveniently described in terms of intuitive three temperature model (see Section 4), where the metallic magnet is mimicked as three interconnected reservoirs, free electrons, lattice, and spins. In the visible spectral range it is safe to assume that light first excites the electrons and increases their kinetic energy, i.e. their temperature. If the laser pulse duration τ is much shorter than the electron–lattice interaction $\tau_{el} \sim 2$ ps, i.e. $\tau \ll \tau_{el}$, the electronic temperature increase goes far above the Curie temperature. Spins of Gd and FeCo, being immersed in a hot reservoir of free electrons, start losing their order. It was found that due to larger spin of Gd-atoms, the order of spins of Gd sublattice is melted slower, than the order of Fe-spins [208]. Hence soon after the laser excitation the ferrimagnet arrives to a strongly non-equilibrium state with nearly demagnetized Fe-sublattice and slightly reduced magnetization of Gd. In this way laser excitation prepares a strongly non-equilibrium state. It concludes the first stage of all-optical switching of magnetization in ferrimagnets.

In parallel, due to electron–phonon interaction the temperature of electrons decreases upon transfer of energy to the lattice. If the electron temperature is reduced below the Curie temperature, magnetization of the fully demagnetized Fe-sublattice must recover to a nonzero value. Relaxation to an equilibrium state is the second stage of the reversal process. In [209] it was shown that conservation of angular momentum favors the route of the relaxation accompanied by reversal of the net magnetization. If GdFeCo has out-of-plane magnetic anisotropy, due to magnetic circular dichroism light of opposite helicities is absorbed differently in the medium. This allows to switch the magnetization in a helicity dependent way.

From the description of the mechanism it is clear that preparation of the strongly non-equilibrium state with fully demagnetized Fe-sublattice is crucial for the whole process of the reversal. Moreover, it is also clear that laser-pulses longer than electron–lattice interaction $\tau_{el} \sim 2$ ps would not result in such a dramatic overheating of the free electrons [210]. Therefore the magnetization reversal triggered by both laser and current pulses with duration τ longer than 10 ps were found to be counter-intuitive [210,211]. As a result, these observations even led to statements about insolvency of the mechanism of the switching via a strongly non-equilibrium state. We would like to note that making such statements one overlooks the fact that three temperature model does not always adequately represent a ferrimagnet. Two very different magnetic sub-lattices are better represented not by one, but by two interconnected reservoirs. The exchange interaction between the spins of Fe and Gd defines the characteristic time of interaction between the reservoirs τ_{Gd-Fe}. The necessary requirement for preparation of a strongly non-equilibrium state would imply the validity of one of the two conditions, $\tau < \tau_{Gd-Fe}$ or $\tau < \tau_{el}$. Ref. [212] explores the possibilities to generate in GdFeCo a strongly non-equilibrium state with nearly demagnetized Fe-sublattices and non-zero magnetization of Gd for laser pulses with duration $\tau < \tau_{Gd-Fe}$. It is shown that the mechanism of the switching via the strongly non-equilibrium state can indeed work for pulse durations above 10 ps. We note that the microscopic processes which take place in the strongly nonequilibrium state of metals are still under debate. For instance, the superdiffusive spin transport is also considered as a possible mechanism behind the switching [213]. Nevertheless, even for such alternative mechanism, the presence of different magnetic constituents coupled antiferromagnetically remains crucial.

5.5. Switching of the ferroelectric polarization

As it is discussed in Section 4, although pumping ferroelectrics with an intense THz pulse can efficiently excite coherent motion of ions in ferroelectrics at the frequency of the soft phonon mode, achieved amplitudes of the ionic oscillations are not large and the total change of the ferroelectric polarization is about 10% at the THz electric field amplitude of

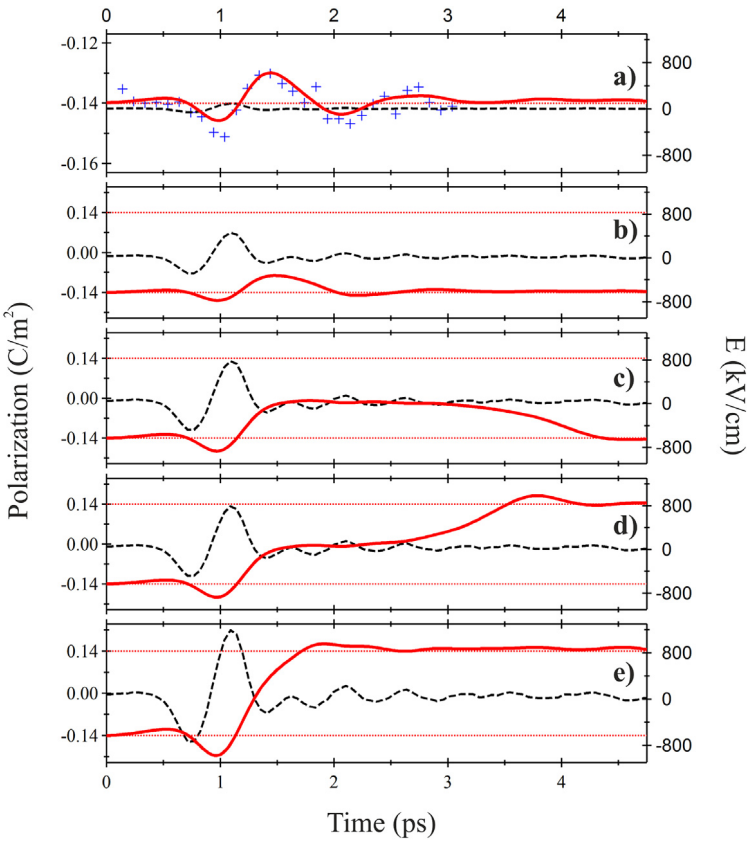

Fig. 27. Proposal for the switching of the ferroelectric polarization in $Sn_2P_2S_6$ via resonantly driving a soft phonon mode by a single-cycle THz pulse [90]. (a) Experimentally observed and (b-e) calculated dynamics of ferroelectric polarization (left scale) driven by a single-cycle THz pulse of different electric field amplitudes E (right scale). Symbols are the experimental data, red solid lines are the results of calculations and black dashed lines are the electric field of the THz pulse.
Source: The figure is taken from Ref. [90].

290 kV/cm (Fig. 13). In [90] simulations for increased THz amplitudes were performed (Fig. 27(b–e)) to verify, if this can be the way to achieve the polarization switching. It is shown that THz field exceeding 790 kV/cm should drive the system over the potential barrier and enable switching (Fig. 27(d)). At higher THz field amplitude reaching ∼1 MV/cm the switching is coherent and occurs within a single period of the phonon mode (Fig. 27(e)). Such fields are available already nowadays [214–216].

An alternative approach for switching of ferroelectrics relies on nonlinear phononics. Instead of directly driving soft phonon mode, recent theoretical works [89,217] suggested an alternative route to manipulate the ferroelectric polarization on ultrafast timescales. A coherent excitation of the ferroelectric soft mode could be achieved indirectly, by exciting a vibrational mode in mid-infrared spectral range, which is anharmonically coupled to the soft mode. In [91] this scenario of reversal of the ferroelectric polarization has been realized experimentally for $LiNbO_3$. It was argued that this reversal can be induced in both directions. Although the authors claimed the navigation of the ferroelectric polarization over the potential barrier, they did not succeed to stabilize the reversed state. It is suggested that the use of thin films or other types of fabricated structures that isolate the reversed volume may improve the lifetime. One can expect a successful writing if the same technique could also be applied to materials with lower coercive fields. It is also interesting to explore mechanisms of writing in ferroelectrics similar to those realized in Heat Assisted Magnetic Recording for metallic ferromagnets, but without involvement of heat. In ferroelectrics an additional optical pulse can cause a depolarization and thus decrease the barrier separating two stable states such that coherent excitation of the soft mode will be able to promote the switching.

6. Outlook: perspectives of photoferroic recording

It is expected that research interest to ultrafast photoferroics in the coming years will only increase. Since laser pulse is the shortest stimulus in contemporary condensed matter physics, photoferroics allow one to explore magnetism and ferroelectricity at the unprecedentedly fast timescale. In particular, using femtosecond or even attosecond laser pulses

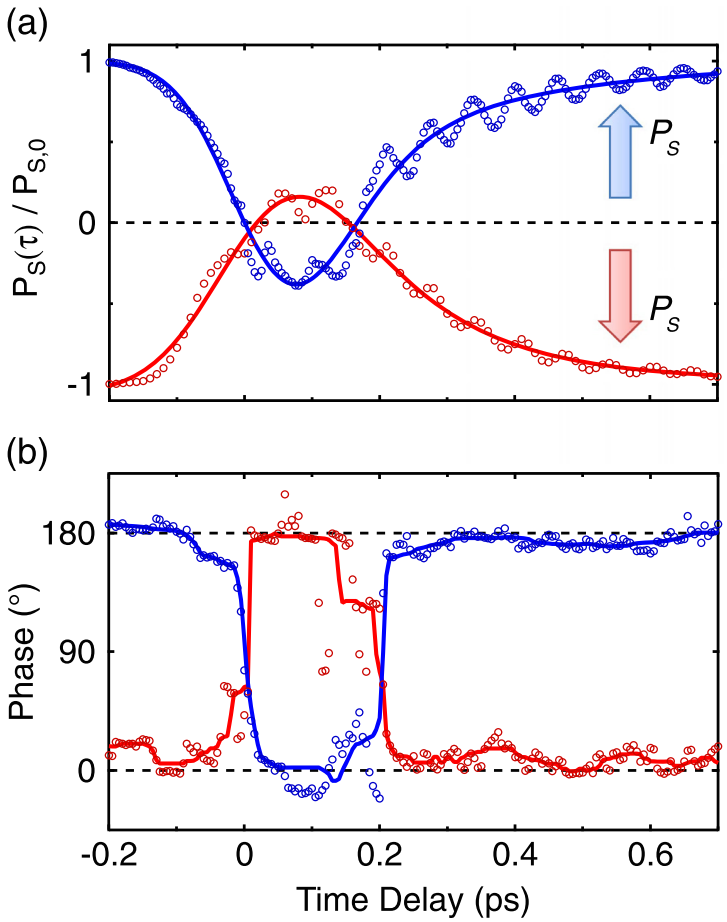

Fig. 28. Transient switching of the ferroelectric polarization in $LiNbO_3$ enabled mid-TR pumping and nonlinear phononic interactions [91]. Dynamics of normalized ferroelectric polarization in $LiNbO_3$ is deduced from time-resolved measurements of the second harmonic generation (a) after pumping the medium with a short mid-infrared pulse with the central frequency of 19 THz. The curves represent data for two opposite initial polarization states. Panel (b) shows the phase of the electric field of the second harmonic proving the switching of the spontaneous polarization.
Source: The figure is taken from Ref. [91].

it is possible to study the collective phenomena of switching of the magnetization or the ferroelectric polarization on a timescale pertinent to characteristic timescales of flipping of a single spin or motion of a single atom. In ferroics these times are defined by the periods of magnons or phonons at the edge of the Brillouin zone, respectively.

Discovery of fundamental limits on speed of recording and accompanying energy dissipations is among the ultimate goals of studies of ultrafast photoferroics. Since two stable bit states of order parameter in a ferroic medium have equal entropies and equal energies, according to (quasi-equilibrium) thermodynamics, a switching between these states can be realized even with zero production of heat. However, in this case, the switching must be a reversible process which takes an infinitely long time [218]. Hence within thermodynamics, ultrafast and least dissipative switching of the ferroic order parameter seems to be mutually exclusive. Pushing the dynamics of electrons, lattice, and spins into a regime in which thermodynamic theories fail, finding non-thermodynamic ways to control the ferroic state of media with the lowest possible production of heat and simultaneously at the fastest possible timescale is the ultimate challenge for fundamental and applied research. In particular, it is proposed that ultrafast optical control of magnetization in spintronic devices can be a competitive and energy efficient solution for the design of Magnetic Random Access Memory [219].

In ultrafast magnetism, one of the first steps in this direction revealed temporal and spectral fingerprints of picosecond and all-coherent, i.e. non-dissipative, navigation of spins over the potential barrier [220]. No recording was demonstrated, but mimicking the process experimentally and using computational analysis it was shown that ultrafast and least dissipative spin switching must have a characteristic phase slip in the time-domain and an asymmetric splitting of the spectrum of the collective spin resonance. To achieve the required conditions this experiment had to employ intense nearly single cycle THz pulses and plasmonic antenna. Both these advanced approaches represent very promising directions of research in the nearest future.

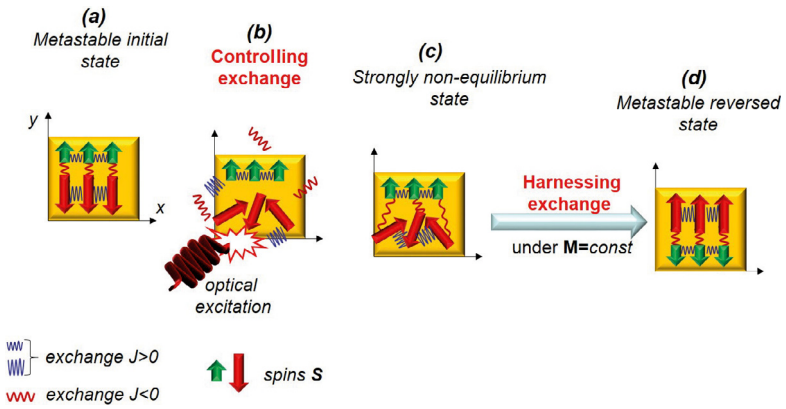

Fig. 29. Controlling and harnessing the exchange interaction for the fastest reversal of the magnetization M. (a) Initial state; (b) In contrast to heating light selectively switches off chosen exchange interactions causing a disorder in a part of the magnet; (c) the light is gone, the exchange interactions are restored and the medium is turned into a strongly non-equilibrium state. The relaxation from this state is driven by the exchange fields up to 1000 T. If during the relaxation the net angular momentum is conserved, $M = const$, the relaxation is accompanied by magnetization reversal. (d) Final state. Such a switching does not require any heat.

From the very first experiments with intense THz pulses it was argued that such pulses must be the most energy efficient mean to control magnetization [93]. As a matter of fact, ultrafast photomagnetic recording has so far been demonstrated using femtosecond laser pulses in the visible spectral range. However, employing THz pulses of the same energy one increases photon flux of the excitation by factor of 1000. Therefore in the search for the fundamental limits on speed of ferroic recording and accompanying dissipations, experimental studies with THz pulses are among the most appealing ones.

We also believe that in the coming years a significant attention will be paid to the search of new photoferroic materials or heterostructures. It is important to note that although laser excitation results in a heating of absolutely any material, the heat-driven mechanisms of all-optical magnetic switching were first discovered and so far have been reported exclusively for metals. Using the thermodynamic approach of [75], we note that in order to reorient the magnetization laser-induced heating should result in a dramatic change of the internal energy U. According to the definition of the internal energy, for the temperature dependence one finds $U(T) = U(0) + \int_0^T C dT$, where C is the heat capacity. The force with which the electric field of light acts on the electrons is the strongest perturbation in light-matter interaction and in the visible spectral range it is safe to assume that light first excites the electrons. Therefore, metallic magnets, where the electrons have the strongest contribution to the total heat capacity, seem to be the best candidates to facilitate heat-driven mechanisms of all-optical magnetic switching. Nevertheless, photoinduced melting of magnetic order can, in principle, be achieved avoiding an increase of the kinetic energy, i.e. heating, of free electrons. An effect of light on the exchange interaction was claimed in several experimental studies. Photo-doping has been suggested to cause a modification of the exchange interaction in the Mott insulators and magnetic semiconductors, while pumping charge transfer electronic transitions was shown to have an effect on the strength of the superexchange interaction in magnetic oxides. Altering the exchange interaction with the help of light can help us to mimic heating or even cooling spins in dielectric media with no free electrons. If the effect of light on the exchange interaction is strong and selective to one of the magnetic sublattices of a ferrimagnet, one can anticipate a scenario of magnetic switching with the help of light as shown in Fig. 29.

Note that in addition to a strong effect of light on the exchange interaction, this scenario requires a very large efficiency of spin-lattice relaxation at stage (b) and a very low spin-lattice coupling at stage (c). While at stage (b) one needs to create conditions for ultrafast demagnetization of one of the sublattice, stage (c) will follow the required scenario if the spin-lattice relaxation is not effective and the total magnetic moment is conserved. The required tuning of the spin-lattice relaxation can be achieved optically. For instance, in the case of Eu-compounds a $d-f$ transition transfers the Eu^{2+} ion from state $4f^7$ to state $4f6d^5$ causing thus a huge change of the spin–orbit interaction.

Another recording scheme can be based on a synthetic ferrimagnet with two antiferromagnetically coupled layers (Fig. 30). In the initial state the magnetizations of the layers are almost equal and thus the coercive field of the ferrimagnet diverges. An enhancement of the exchange interaction by a laser pulse in one of the layers increases its magnetization on a timescale of the exchange-driven spin dynamics. This process leads to a reduction of the coercive field and a bit can be recorded by applying a moderate magnetic field. The suggested scheme is very similar to HAMR. However, in contrast, in the considered approach the reduction of the coercive field does not require any heating, as it results directly from the virtual states induced by the electric field of the light pulse.

A similar recording scheme can be realized in ferrimagnets which are brought in the vicinity of the compensation temperature. Several rare-earth containing iron oxides, such $ErFeO_3$ or $Dy_3Fe_5O_{12}$, do have such a temperature. With the help of selective pumping of the rare-earth ions one can effectively change the exchange parameters in the compound, mimic a temperature change and thus promote the magnetization reversal.

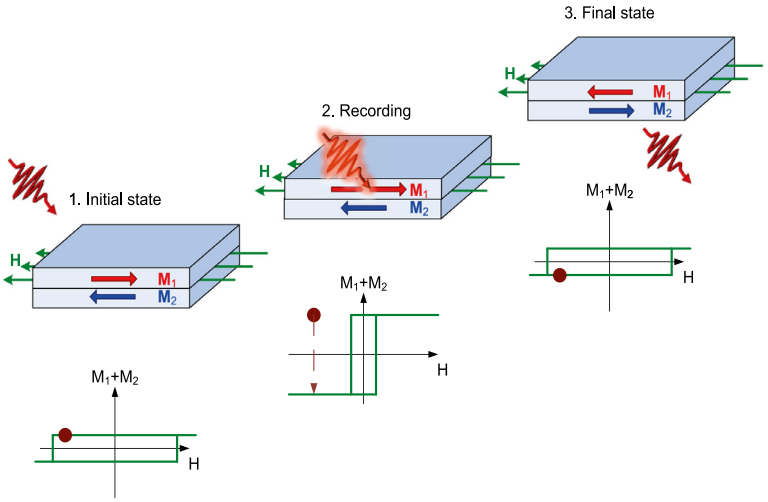

Fig. 30. A proposed scenario of magnetic recording based on the optical control of exchange energy. The two antiferromagnetically coupled layers constitute an element of the data storage medium. In the initial state the magnetizations of the layers are almost equal and the coercive field of the artificial ferrimagnet diverges. The laser pulse acts on the upper magnetic layer and increases its magnetization $\mathbf{M}_1$ thereby diminishing the coercive field. The laser-induced transient magnetic configuration becomes unstable in the applied field **H**. Thus, the magnetizations of the layers reverse. The recording does not involve absorption of the laser pulse. The brown circle represents the position of the system in the phase space.

As discussed in the present review, switching ferroelectric order parameter with optical or near-infrared laser pulses is obviously more challenging than already demonstrated switching magnetization or antiferromagnetic vector in magnets. Ultrafast recording on a ferroelectric medium with the help of ultrashort pulses of light is one of the challenges for future studies. Here, in our opinion, one needs to pay particular attention to electronic ferroelectrics, especially those based on charge ordering. Ultrafast laser-driven melting of charge-order demonstrated in both ferroelectric (see Section 4.2) and non-ferroelectric materials [221], may play the role similar to the ultrafast demagnetization in magnetic recording with light, and reduce the barrier separating two stable states. Coherent excitation of the soft mode using optical rectification effect or THz pulse can promote the reversal over the barrier lowered via a partial quenching of the polarization.

Although pumping ferroelectrics with intense THz pulses so far has not led to effects sufficient for switching ferroelectric polarization, we note that the ferroelectric $Sn_2P_2S_6$ used in some of these experiments, is a special class of materials — ferroelectric semiconductors. These materials are ferroelectric in the ground state, but optical excitation of electrons from the valence to the conduction band generates mobile charge carriers that can efficiently screen the ferroelectric polarization, and thus decrease the barrier that separates two states with the opposite orientations of the polarization. Effect of photocarriers on a potential energy profile opens up a promising pathway to assist the switching of the ferroelectric polarization via soft mode excitation. One can anticipate that these materials allow a scenario of optical switching similar to the one shown in Fig. 25. In this scenario the first pump THz pulse excites phonon mode coupled to the ferroelectric order parameter with the amplitude which is not sufficient to overcome the potential barrier. The second pump pulse excites electronic transitions from the valance to the conduction band and lowers the barrier. Similarly to [108] tuning the timing between the period of phonon oscillations and the recovery of the barrier it should be possible to stabilize the polarization either in one or in another potential minimum.

Hence it is believed that ultrafast optical recording on ferroelectric media must be possible and will be demonstrated in the nearest future. Possibly, the combination of two stimuli, or the two-fold effect of femtosecond laser pulses would be required for switching. It would be also important to pay attention to a possibility to realize the switching of the ferroelectric in an external electric field which is assisted by optical excitation.

When discussing ferroics it is important to consider such class of materials as magnetoelectric multiferroics. One may anticipate that the presence of coupled magnetic and ferroelectric ordering can, for instance, facilitate response of spontaneous polarization to light. And, in contrast, stronger coupling of light to electronic degree of freedom in a multiferroic may allow more efficient manipulation of magnetic ordering by light.

A class of magnetoelectric multiferroics is comprised by two large groups — single phase and composite multiferroics. In the first group the two order parameters coexist in a same material under the same conditions. The drawback of many of such materials is a moderate or even weak magnetoelectric coupling, which is explained by the fact that different ions of the medium contribute to the magnetism and ferroelectricity (type I multiferroics), since *d*-electrons contributing to magnetic ordering make ferroelectric distortions unfavorable [222]. Therefore, a lot of attention is paid nowadays to type II multiferroics where the ferroelectric ordering is *induced* by the magnetic ordering [32]. Specific noncollinear spin structures is fundamental feature of type II multiferroics. The peculiarities of the magnetic structure force displacements

of magnetic ions resulting in emergence of spontaneous electric polarization [32]:

$$\mathbf{P} \propto [(\mathbf{M} \cdot \partial)\,\mathbf{M} - \mathbf{M}\,(\partial \cdot \mathbf{M})]\,. \tag{26}$$

Naturally, such materials are highly promising for the ultrafast optical manipulation of ferroelectric polarization, since laser-induced perturbation of spin ordering is expected to affect the ions displacements. There are numerous reports on ultrafast electron, phonon and lattice dynamics in various multiferroics, however only limited number of works address the problem of coupled dynamics of two order parameters. Therefore, below we review several works, which report on attempts to manipulate spin ordering in a type II multiferroics with perspective for the switching of the associated ferroelectric polarization.

$TbMnO_3$ is the type II multiferroic being actively studied nowadays. Below 27 K the Mn^{3+} spins form a cycloid within the (bc)—crystallographic plane, and a spontaneous ferroelectric polarization along the c-axis emerges [223]. After theoretical proposal of the switching with the help of picosecond optical pulses [224], the problem was experimentally investigated in [225]. Intense THz pulses were used as pump and were tuned to the resonance with the electromagnon excitation in $TbMnO_3$, i.e. electric-dipole active spin excitations directly connected to the magnetoelectric coupling [226]. The THz-induced dynamics in the medium was probed using ultrafast X-ray diffraction. It was estimated that the detected signals correspond to amplitude of spin-cycloid plane rotation equal to 4.2 ± 0.4^{o}.

Alternative approach to ultrafast control of the spin cycloid has been examined in [227], where the 100 fs near-infrared laser pulses (1.55 eV) were used to excite $TbMnO_3$, and the resonant soft X-ray diffraction was employed to monitor the laser-induced changes of the spin cycloid. An intuitive picture that laser-induced heating of a spin system above 27 K would yield changes of the wavevector of the spin cycloid, was found to contradict experimental results. Instead, the melting of the long-range magnetic ordering at the timescale of $\sim$22 ps was observed, but the wavevector of the cycloid remained unchanged. It was suggested that the changes of the cycloid require higher magnon group velocities which would allow the localized laser-induced excitation propagate and affect the spatial distribution of spins.

The ongoing research on ultrafast switching of magnetic and ferroelectric ordering in type II multiferroics may profit from recent findings of ultrafast optical changes of the exchange and the Dzyaloshinskii–Moriya interactions in rare-earth orthoferrites [102] discussed in Section 4.1. There are also theoretical proposals for inducing multiferrocity via laser-induced emergence of the Dzyaloshinskii–Moriya coupling [228], and for the laser-induced change of D/J ratio in the antiferromagnetic spin–orbit Mott insulator [229].

Alternative way to have the coexisting ferroelectric polarization and magnetic ordering in the same structure is realized in so-called composite multiferroics. These are 1-, 2-, or 3D heterostructures which combine the coupled magnetically-ordered and ferroelectric constituents [230–232].

For instance, it is well established that strain transferred from a ferroelectric to a ferromagnet in such a structure effectively couples the two order parameters [233], enabling, for instance, efficient electric field induced switching of the magnetization [234]. It is thus appealing to use such structures, in which the order parameter in one of the constituents is sensitive to photoexcitation, while another order parameter is controlled via associated strain changes. In [235] the first attempt to go along this line was reported for a composite structure with photostrictive matrix containing magnetostrictive columns. Illumination of the matrix with a nanosecond light source modified the strain in the structure, and, therefore, changed the effective anisotropy of the magnetic constituents, enabling change of the magnetic state of the latter. In this respect it is important to mention that excitation of a piezoelectric heterostructure by femtosecond laser pulses may enable generation of record high picosecond strains of amplitudes up to 2% [236]. Ferroelectrics also allow generation of large-amplitude strains due to femtosecond photoexcitation [237,238] Therefore, ultrafast experiments with multiferroic composites with strain-mediated coupling seem to be very promising.Theoretical consideration of the laser-driven strain and the related dynamics in a multiferroic composite is developed in [239], neglecting, however, laser-induced changes of such parameters of composite multiferroics as magnetization, magnetoelastic coupling constants etc.

It may appear promising to use composite multiferroics to achieve yet elusive optical recording on a ferroelectric via advanced methods of optical control of magnetism. Here one can employ the strain generated during the laser-induced magnetization dynamics [240–242]. For instance, Ref. [240] showed that the second optical harmonic generation linked to the spontaneous polarization of the ferroelectric $Ba_{0.1}Sr_{0.9}TiO_3$ (BSTO) could be strongly modified due to photoexcitation and rapid decrease of the magnetization in the adjustment ferromagnet $La_{0.7}Ca_{0.3}MnO_3$ (LCMO) (Fig. 31).

Based on these experimental evidence and general considerations, one can envisage a scenario with two-fold effect of the laser pulses on composite multiferroics — when the direct photoexcitation of the ferroelectric is assisted by the strain launched simultaneously in the magnetic constituent. We would like to stress that, since a technology of composite multiferroics based on metallic magnetic layers and dielectric ferroelectric is well established, it seems to be a straightforward idea to use ultrafast demagnetization, which can reach 100%, for launching large amplitude strain into adjustment ferroelectric [241,242]. Here it would be also provisional to employ laser-generated shear strain [166,242] instead of longitudinal one typically obtained in such experiments.

In memory applications one should also take into account not only writing time, but a time required for the whole write-read cycle. In state-of-the-art devices the time required for a written bit to get settled into a readable state is on a timescale of nanoseconds. In the conventional ferroic memory the switching process often proceeds via nucleation of a phase with the reversed magnetization or electric polarization. The nucleation is followed by domain wall motion.

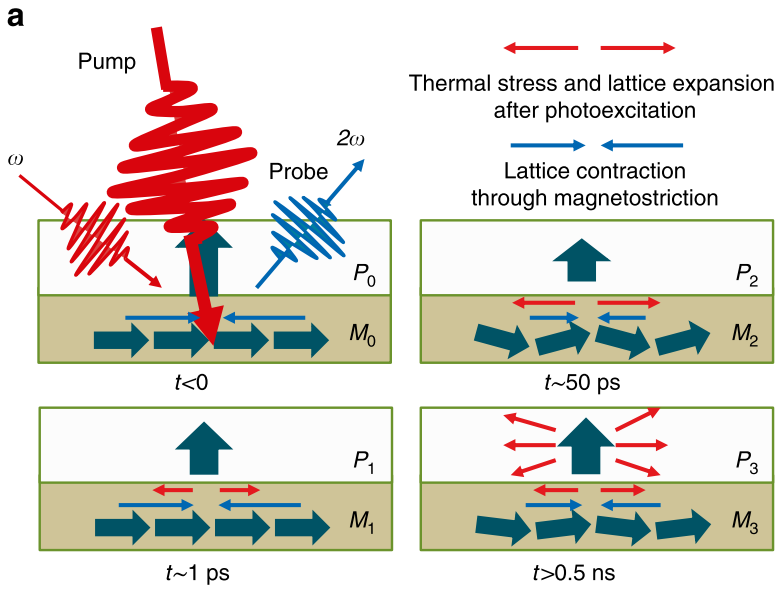

Fig. 31. A scenario for indirect laser-driven control of ferroelectric polarization via ultrafast changes of magnetization in a composite multiferroic LCMO/BSTO [240].
Source: The figure is taken from Ref. [240].

In ferroelectric memories [2,12] such a switching mechanism is also partly responsible for the polarization fatigue problem [9]. As we discussed above, laser excitation enables switching of the order parameter in ferroics via a soft mode excitation. In the case of such a switching, the magnetization is reoriented between two stable bit states via precessional motion. The settling time in this case is defined by the period of the precession and the Gilbert damping [14]. The ability of ultrashort laser pulses to trigger large amplitude magnetization precession in iron garnets [97,98,201], which are known for the small Gilbert damping, revealed an anomalous enhancement of the Gilbert damping during precessional switching of the magnetization [201]. The gain in the settling time provided by the laser-induced switching can be even more striking in ferroelectrics, since the electric field driven switching via nucleation and domain wall motion would be replaced by the coherent switching with the switching times of just a few picoseconds [90,91]. Furthermore, switching of a ferroic order parameter via non-equilibrium state is fundamentally different from the conventional switching and the physical and technological limits for the read out time in this case are still to be understood. Such a switching was realized in a metallic ferrimagnet and the read out was successfully realized after 30 ps [243].

State-of-the-art all-electrical memory devices have already revealed the trend indicating that switching energy scales with switching time at long timescales, and electrical switching faster than 1 ns results in a high energy penalty [244]. It is thus interesting to explore if the demonstrated or suggested pathways of laser-induced ferroic recording can lead to a technology for faster and nearly nondissipative switching. The lowest theoretical limit on energy dissipation Q when manipulating one bit of information is defined by the Landauer principle [245] as $Q = k_B T \ln 2$. At or below room temperature, Q is of the order of millielectronvolt, which by the uncertainty principle entails picosecond timescales for minimally dissipative switching. Thus, precessional switching of magnetization triggered by a single-cycle THz pulse with millielectronvolt photon energies and subpicosecond duration promises to be the fastest and least-dissipative route [220].

Clearly, one of the obstacles on a way to the photoferroic recording based on various mechanisms described in the present review, is the relatively small amplitudes of excited spin or lattice dynamics, achieved in most of the experiment. Similarly to magnets, the effect of light on a ferroelectric medium can be enhanced using principles of plasmonics [246–248] and photonics [249,250]. Plasmonic antennas were already successfully employed, for instance, for enhancement of THz-induced SHG in $BiFeO_3$ [156] and for enhancement of impact of THz field on structural dynamics in $SrTiO_3$ [251], and for achieving a 10% modulation of the atomic displacements associated with the ferroelectric polarization during the THz pulse excitation of $BaTiO_3$ [142].

Magnetophotonic structures, i.e. the structures with artificial photonic bandgaps containing magnetic layers [250], were recently used to demonstrate localization and enhancement of ultrafast inverse Faraday effect in dielectric garnets [252]. Here the interference effects in the magnetic layer placed between two specially designed Bragg mirrors allowed to localize the electric field of femtosecond laser pulses within a certain depth of the magnetic layer and, thus to observe strongly enhanced amplitude of the magnetization precession driven via ultrafast inverse Faraday effect.

It is worth noting that, along with concentrating the electromagnetic field and enhancing its power density, plasmonic and photonic structures enable nanoscale localization of the photoinduced impact in lateral as well as in tangential directions. From the very first demonstrations of photoferroic recording it was realized that one of the main drawbacks

of this approach is a relatively low recording density, which is density intrinsically limited by the wavelength of light and the diffraction limit. Thus, the ability of nanooptics to overcome the diffraction limit is crucial for practical applications of photoferroic recording. In particular, spatial localization of the femtosecond laser pulses using plasmonic nanoantenna enabled optical switching of TbGdFe alloy at the submicron scale [253]. Here, using plasmonic antennas and light with the wavelength of 800 nm, it was possible to record magnetic domains as small as 40 nm. From the point of view of light localization, it would be important to understand whether the switched ferroelectric domain would be more stable in such a case, which could help to resolve the difficulty occurred in experiments with $LiNbO_3$ (Fig. 28) where the switched ferroelectric polarization could not be preserved [91] . Thus, it can be anticipated, that the further progress in ultrafast photoferroic recording would largely benefit from recent achievements in the fields of plasmonics and photonics. Combination of photoferroics and plasmonics as well as nanoscale optical recording on ferric media are the subjects for fundamental studies with clear implications in technology.

Finally, we anticipate that in the coming years increasingly more attention will be paid to possibilities to improve functionality of spintronics devices with the help of photoferroics and laser pulses [254,255].

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References

[1] K. Aizu, Possible species of ferromagnetic, ferroelectric, and ferroelastic crystals, Phys. Rev. B 2 (1970) 754–772, http://dx.doi.org/10.1103/PhysRevB.2.754, URL https://link.aps.org/doi/10.1103/PhysRevB.2.754.
[2] J.F. Scott, Ferroelectric Memories, Springer, 2000.
[3] M.L. Plumer, J. Van Ek, D. Weller, The Physics of Ultra-High-Density Magnetic Recording, Springer Science and Business Media, 2012.
[4] M. Lantz, Tape storage mounts a comeback, IEEE Spectrum 55 (9) (2018) 1–3, 55 (2018) 32.
[5] C.P. Bean, J.D. Livingston, Superparamagnetism, J. Appl. Phys. 30 (4) (1959) S120–S129, http://dx.doi.org/10.1063/1.2185850, arXiv:https://doi.org/10.1063/1.2185850.
[6] C. Lichtensteiger, M. Dawber, J.-M. Triscone, in: K.M. Rabe, C.H. Ahn, J.-M. Triscone (Eds.), Physics of Ferroelectrics, Springer, 2007, pp. 305–338.
[7] A. Rüdiger, T. Schneller, A. Roelofs, S. Tiedke, T. Schmitz, R. Waser, Nanosize ferroelectric oxides – tracking down the superparaelectric limit, Appl. Phys. A 80 (6) (2005) 1247–1255, http://dx.doi.org/10.1007/s00339-004-3167-z.
[8] M.D. Glinchuk, A.V. Ragulya, V.A. Stephanovich, Nanoferroics, Springer, 2013.
[9] A.K. Tagantsev, I. Stolichnov, E.L. Colla, N. Setter, Polarization fatigue in ferroelectric films: Basic experimental findings, phenomenological scenarios, and microscopic features, J. Appl. Phys. 90 (3) (2001) 1387–1402, http://dx.doi.org/10.1063/1.1381542, arXiv:https://doi.org/10.1063/1.1381542.
[10] C. A.-Paz de Araujo, J.D. Cuchiaro, L.D. McMillan, M.C. Scott, J.F. Scott, Fatigue-free ferroelectric capacitors with platinum electrodes, Nature 374 (1995) 627.
[11] C.H. Ahn, K.M. Rabe, J.-M. Triscone, Ferroelectricity at the nanoscale: Local polarization in oxide thin films and heterostructures, Science 303 (5657) (2004) 488–491, http://dx.doi.org/10.1126/science.1092508, arXiv:https://science.sciencemag.org/content/303/5657/488.full.pdf. URL https://science.sciencemag.org/content/303/5657/488.
[12] N. Setter, D. Damjanovic, L. Eng, G. Fox, S. Gevorgian, S. Hong, A. Kingon, H. Kohlstedt, N.Y. Park, G.B. Stephenson, I. Stolitchnov, A.K. Taganstev, D.V. Taylor, T. Yamada, S. Streiffer, Ferroelectric thin films: Review of materials, properties, and applications, J. Appl. Phys. 100 (5) (2006) 051606, http://dx.doi.org/10.1063/1.2336999, arXiv:https://doi.org/10.1063/1.2336999.
[13] S. Streiffer, D. Fong, Phase transitions in nanoscale ferroelectric structures, MRS Bull. 34 (11) (2009) 832–837, http://dx.doi.org/10.1557/mrs2009.233.
[14] T. Gerrits, H.A.M. van den Berg, J.B. Hohlfeld, T. L. Rasing, Ultrafast precessional magnetization reversal by picosecond magnetic field pulse shaping, Nature 418 (2002) 509.
[15] A.V. Kimel, B.A. Ivanov, R.V. Pisarev, P.A. Usachev, A. Kirilyuk, T. Rasing, Inertia-driven spin switching in antiferromagnets, Nature Phys. 5 (10) (2009) 727.
[16] W. Cochran, Crystal stability and the theory of ferroelectricity, Adv. Phys. 9 (36) (1960) 387–423, http://dx.doi.org/10.1080/00018736000101229, arXiv:https://doi.org/10.1080/00018736000101229.
[17] J.C. Slater, Theory of the transition in KH_2PO_4, J. Chem. Phys. 9 (1) (1941) 16–33, http://dx.doi.org/10.1063/1.1750821, arXiv:https://doi.org/10.1063/1.1750821.
[18] S. Fahy, R. Merlin, Reversal of ferroelectric domains by ultrashort optical pulses, Phys. Rev. Lett. 73 (1994) 1122–1125, http://dx.doi.org/10.1103/PhysRevLett.73.1122, URL https://link.aps.org/doi/10.1103/PhysRevLett.73.1122.
[19] T. Portengen, T. Östreich, L.J. Sham, Theory of electronic ferroelectricity, Phys. Rev. B 54 (1996) 17452–17463, http://dx.doi.org/10.1103/PhysRevB.54.17452, URL https://link.aps.org/doi/10.1103/PhysRevB.54.17452.
[20] S. Tomić, M. Dressel, Ferroelectricity in molecular solids: a review of electrodynamic properties, Rep. Progr. Phys. 78 (9) (2015) 096501, http://dx.doi.org/10.1088/0034-4885/78/9/096501.
[21] A. Hubert, R. Schäfer, Magnetic Domains, Springer, Berlin Heidelberg, 1998.
[22] A.K. Tagantsev, L.E. Cross, J. Fousek, Domains in ferroic crystals and thin films, Springer, 2010.
[23] J. Padilla, W. Zhong, D. Vanderbilt, First-principles investigation of 180° domain walls in $BaTiO_3$, Phys. Rev. B 53 (1996) R5969–R5973, http://dx.doi.org/10.1103/PhysRevB.53.R5969, URL https://link.aps.org/doi/10.1103/PhysRevB.53.R5969.
[24] D. Lee, R.K. Behera, P. Wu, H. Xu, Y.L. Li, S.B. Sinnott, S.R. Phillpot, L.Q. Chen, V. Gopalan, Mixed Bloch-Néel-Ising character of 180° ferroelectric domain walls, Phys. Rev. B 80 (2009) 060102, http://dx.doi.org/10.1103/PhysRevB.80.060102, URL https://link.aps.org/doi/10.1103/PhysRevB.80.060102.
[25] A. Angoshtari, A. Yavari, Atomic structure of steps on 180° ferroelectric domain walls in $PbTiO_3$, J. Appl. Phys. 108 (8) (2010) 084112, http://dx.doi.org/10.1063/1.3501050, arXiv:https://doi.org/10.1063/1.3501050.
[26] M.E. Lines, A.M. Glass, Principles and Applications of Ferroelectrics and Related Material, OUP Oxford, 2001.
[27] B.A. Strukov, A.P. Levanyuk, Ferroelectric Phenomena in Crystals, Springer-Verlag, Berlin-Heidelberg, 1998.

[28] J. Scott, Absence of critical exponents in ferroelectrics: experiments of Hilczer and theory of Levanyuk and Sigov, Phase Transit. 89 (2016) 645.
[29] E. Beaurepaire, J.-C. Merle, A. Daunois, J.-Y. Bigot, Ultrafast spin dynamics in ferromagnetic nickel, Phys. Rev. Lett. 76 (1996) 4250–4253, http://dx.doi.org/10.1103/PhysRevLett.76.4250, URL https://link.aps.org/doi/10.1103/PhysRevLett.76.4250.
[30] M.H. Kryder, E.C. Gage, T.W. McDaniel, W.A. Challener, R.E. Rottmayer, G. Ju, Y.-T. Hsia, M.F. Erden, Heat assisted magnetic recording, Proc. IEEE 96 (11) (2008) 1810–1835.
[31] M. Fiebig, Revival of the magnetoelectric effect, J. Phys. D: Appl. Phys. 38 (8) (2005) R123–R152, http://dx.doi.org/10.1088/0022-3727/38/8/r01.
[32] S.-W. Cheong, M. Mostovoy, Multiferroics: a magnetic twist for ferroelectricity, Nature Mater. 6 (2007) 13, http://dx.doi.org/10.1038/nmat1804.
[33] A.P. Pyatakov, A.K. Zvezdin, Magnetoelectric and multiferroic media, Phys.-Usp. 55 (6) (2012) 557–581, http://dx.doi.org/10.3367/ufne.0182.201206b.0593.
[34] A. Kirilyuk, A.V. Kimel, T. Rasing, Ultrafast optical manipulation of magnetic order, Rev. Mod. Phys. 82 (2010) 2731–2784, http://dx.doi.org/10.1103/RevModPhys.82.2731, URL https://link.aps.org/doi/10.1103/RevModPhys.82.2731.
[35] A. Kirilyuk, A.V. Kimel, T. Rasing, Laser-induced magnetization dynamics and reversal in ferrimagnetic alloys, Rep. Prog. Phys. 76 (2) (2013) 026501, http://dx.doi.org/10.1088/0034-4885/76/2/026501.
[36] M.S. El Hadri, M. Hehn, G. Malinowski, S. Mangin, Materials and devices for all-optical helicity-dependent switching, J. Phys. D: Appl. Phys. 50 (13) (2017) 133002.
[37] A.V. Kimel, M. Li, Writing magnetic memory with ultrashort light pulses, Nature Rev. Mater. (2019) 1.
[38] I.K. Kikoin, M.M. Noskov, Photoelectromagnetic effect, Phys. Zs. Sowjetunion 5 (1934) 586.
[39] I.K. Kikoin, S.D. Lazarev, Photoelectromagnetic effect, Sov. Phys. Usp. 21 (4) (1978) 297–308, http://dx.doi.org/10.1070/pu1978v021n04abeh005538.
[40] M.A. Ruderman, C. Kittel, Indirect exchange coupling of nuclear magnetic moments by conduction electrons, Phys. Rev. 96 (1954) 99–102, http://dx.doi.org/10.1103/PhysRev.96.99, URL https://link.aps.org/doi/10.1103/PhysRev.96.99.
[41] T. Kasuya, A theory of metallic ferro- and antiferromagnetism on Zener's model, Progr. Theoret. Phys. 16 (1) (1956) 45–57, http://dx.doi.org/10.1143/PTP.16.45, arXiv:http://oup.prod.sis.lan/ptp/article-pdf/16/1/45/5266722/16-1-45.pdf.
[42] K. Yosida, Magnetic properties of Cu-Mn alloys, Phys. Rev. 106 (1957) 893–898, http://dx.doi.org/10.1103/PhysRev.106.893, URL https://link.aps.org/doi/10.1103/PhysRev.106.893.
[43] B.V. Karpenko, A.A. Berdyshev, Exchange interaction via current carriers in ordered semiconducting magnets, Sov. Phys.—Solid State 5 (1964) 2215.
[44] A.A. Berdyshev, Ferromagnetic semiconductors in which the exchange coupling occurs through the conduction electrons, Sov. Phys.—Solid State 8 (1966) 1104.
[45] R.W. Teale, D.W. Temple, Photomagnetic anneal, a new magneto-optic effect, in Si-doped yttrium iron garnet, Phys. Rev. Lett. 19 (16) (1967) 904.
[46] R.F. Pearson, A.D. Annis, P. Kompfner, Photomagnetic anneal properties of silicon-doped yttrium iron garnet, Phys. Rev. Lett. 21 (27) (1968) 1805.
[47] J.F. Dillon Jr., E.M. Gyorgy, J.P. Remeika, Photoinduced magnetic anisotropy and optical dichroism in silicon-doped yttrium iron garnet, Phys. Rev. Lett. 22 (13) (1969) 643.
[48] P.J. Flanders, C.D. Graham Jr., J.F. Dillon Jr., E.M. Gyorgy, J.P. Remeika, Photoinduced changes in the crystal anisotropy of Si-doped YIG, J. Appl. Phys. 42 (4) (1971) 1443–1445.
[49] U. Enz, H. Van der Heide, Two new manifestations of the photomagnetic effect, Solid State Commun. 6 (6) (1968) 347–349.
[50] T. Merceron, P. Bernstein, Photomagnetic effect in Ni-Zn ferrite with small amount of cobalt, Phys. Status Solidi a 35 (2) (1976) 681–686.
[51] H.D. Jonker, Photomagnetic effects in single-crystal Ru-doped lithium ferrite, J. Solid State Chem. 10 (2) (1974) 116–121.
[52] K. Hisatake, K. Ohta, N. Ichinose, H. Yokoyama, X-ray induced decrease of permeability in YIG single crystals with pb impurity, Phys. Status Solidi a 26 (1) (1974) K79–K82.
[53] J. Haisma, J.M. Robertson, U. Enz, Direct observation of light-induced Bloch wall pinning, Solid State Commun. 10 (11) (1972) 1021–1024.
[54] F.K. Lotgering, Photomagnetic effects and disaccommodation in Co-doped YIG, J. Phys. Chem. Solids 36 (11) (1975) 1183–1191.
[55] V.G. Veselago, N.V. Vorob'eva, R.A. Doroshenko, Photoinduced change in magnetostriction in yttrium iron garnet, JETP Lett. 45 (8) (1987) 512.
[56] E.I. Golovenchits, B.D. Laikhtman, V.A. Sanina, Long-lived, magnetically ordered state in $EuCrO_3$ excited by optical pumping, JETP Lett. 31 (1980) 223.
[57] W. Lems, P.J. Rijnierse, P.F. Bongers, U. Enz, Photomagnetic effect in a chalcogenide spinel, Phys. Rev. Lett. 21 (24) (1968) 1643.
[58] D.E. Lacklison, J. Chadwick, J.L. Page, Photomagnetic effect in ferric borate, J. Phys. D: Appl. Phys. 5 (4) (1972) 810.
[59] M.M. Afanasiev, M.E. Kompan, I.A. Merkulov, Increase of Curie temperature of magnetic semiconductors by illumination, JETP Lett. 23 (1976) 570.
[60] V.G. Kovalenko, E.S. Kolezhuk, P.S. Kuts, Photomagnetic recording of information, Soviet Tech. Phys. Lett. 7 (1981) 435–437.
[61] A. Stupakiewicz, A. Maziewski, I. Davidenko, V. Zablotskii, Light-induced magnetic anisotropy in Co-doped garnet films, Phys. Rev. B 64 (2001) 064405, http://dx.doi.org/10.1103/PhysRevB.64.064405, URL https://link.aps.org/doi/10.1103/PhysRevB.64.064405.
[62] E. Fatuzzo, G. Harbeke, W.J. Merz, R. Nitsche, H. Roetschi, W. Ruppel, Ferroelectricity in SbSi, Phys. Rev. 127 (6) (1962) 2036.
[63] R. Nitsche, H. Roetschi, P. Wild, New ferroelectric V. VI. VII compounds of the SbSI type, Appl. Phys. Lett. 4 (12) (1964) 210–211.
[64] V.M. Fridkin, Some effects due to electron-phonon interaction in phase transitions occurring in a semiconductor ferroelectric, JETP Lett. 3 (1966) 161.
[65] L.M. Belyaev, I.I. Groshik, V.V. Lyakhovitskaya, V.N. Nosov, V.M. Fridkin, Photosensitive phase transition in the ferro-electric semiconductor sbsi, JETP Lett. 6 (1967) 16.
[66] S. Ueda, I. Tatsuzaki, Y. Shindo, Change in the dielectric constant of SbSI caused by illumination, Phys. Rev. Lett. 18 (12) (1967) 453.
[67] G. Godefroy, P. Jullien, L. Cai, Photoconduction in doped $BaTiO_3$ single crystals, Ferroelectrics 13 (1) (1976) 309–312.
[68] V.M. Fridkin, Photoferroelectrics, Springer, 1979.
[69] A.M. Glass, D.H. Auston, Excited state dipole moments of impurities in polar crystals, Opt. Commun. 5 (1) (1972) 45–49.
[70] P. Peercy, C. Land, Optical image storage in ion implanted PLZT ceramics, Nucl. Instrum. Methods 182–183 (1981) 787–796, http://dx.doi.org/10.1016/0029-554X(81)90811-9, URL http://www.sciencedirect.com/science/article/pii/0029554X81908119.
[71] T. Volk, M. Wöhlecke, Lithium Niobate: Defects, Photorefraction and Ferroelectric Switching, Springer Series in Materials Science, Springer Berlin Heidelberg, 2008, URL https://books.google.ru/books?id=IcNpNnyWCUwC.
[72] V.F. Kovalenko, É.L. Nagaev, Photoinduced magnetism, Sov. Phys. Usp. 29 (4) (1986) 297.
[73] P.S. Pershan, Nonlinear optical properties of solids: Energy considerations, Phys. Rev. 130 (1963) 919–929, http://dx.doi.org/10.1103/PhysRev.130.919, URL https://link.aps.org/doi/10.1103/PhysRev.130.919.
[74] V. Lakhno, E. Nagaev, Nondissipative photoferromagnetism of magnetic semiconductors, Sov. J. Exp. Theor. Phys. 47 (1978) 1105.
[75] L.D. Landau, E.N. Lifshitz, Electrodynamics of Continious Media, Pergamon, Oxford, 1984.
[76] L.P. Pitaevskii, Electric forces in a transparent medium, Sov. Phys.—JETP 12 (1961) 1008.

[77] J.P. van der Ziel, P.S. Pershan, L.D. Malmstrom, Optically-induced magnetization resulting from the inverse Faraday effect, Phys. Rev. Lett. 15 (1965) 190–193, http://dx.doi.org/10.1103/PhysRevLett.15.190, URL https://link.aps.org/doi/10.1103/PhysRevLett.15.190.

[78] P.S. Pershan, J.P. van der Ziel, L.D. Malmstrom, Theoretical discussion of the inverse faraday effect, Raman scattering, and related phenomena, Phys. Rev. 143 (1966) 574–583, http://dx.doi.org/10.1103/PhysRev.143.574, URL https://link.aps.org/doi/10.1103/PhysRev.143.574.

[79] B.A. Zon, V.Y. Kupershmidt, G.V. Pakhomov, Urazbaev, Optically-induced magnetization resulting from the inverse Faraday effect, JETP Lett. 45 (1987) 272.

[80] Y.R. Shen, The Principles of Nonlinear Optics, Wiley, 2002.

[81] A.M. Kalashnikova, A.V. Kimel, R.V. Pisarev, Ultrafast opto-magnetism, Phys.-Usp. 58 (10) (2015) 969–980, http://dx.doi.org/10.3367/ufne.0185.201510j.1064.

[82] V.N. Gridnev, Phenomenological theory for coherent magnon generation through impulsive stimulated Raman scattering, Phys. Rev. B 77 (2008) 094426, http://dx.doi.org/10.1103/PhysRevB.77.094426, URL https://link.aps.org/doi/10.1103/PhysRevB.77.094426.

[83] L. Dhar, J.A. Rogers, K.A. Nelson, Time-resolved vibrational spectroscopy in the impulsive limit, Chem. Rev. 94 (1) (1994) 157–193, http://dx.doi.org/10.1021/cr00025a006, arXiv:https://doi.org/10.1021/cr00025a006.

[84] R. Merlin, Generating coherent THz phonons with light pulses, Solid State Commun. 102 (2) (1997) 207–220, http://dx.doi.org/10.1016/S0038-1098(96)00721-1, URL http://www.sciencedirect.com/science/article/pii/S0038109896007211.

[85] A.K. Zvezdin, Dynamics of domain walls in weak ferromagnets, JETP Lett. 29 (10) (1979) 553–557.

[86] V.G. Bar'yakhtar, B.A. Ivanov, M.V. Chetkin, Dynamics of domain walls in weak ferromagnets, Phys.-Usp. 28 (7) (1985) 563–588, http://dx.doi.org/10.1070/PU1985v028n07ABEH003871, URL https://ufn.ru/en/articles/1985/7/b/.

[87] A.K. Zvezdin, A. Mukhin, Novel nonlinear dynamical effects in antiferromagnets, Bull. Lebedev Physical Institute 12 (1981) 10–15.

[88] A.F. Andreev, V.I. Marchenko, Symmetry and the macroscopic dynamics of magnetic materials, Sov. Phys. Usp. 23 (1) (1980) 21–34, http://dx.doi.org/10.1070/pu1980v023n01abeh004859.

[89] A. Subedi, A. Cavalleri, A. Georges, Theory of nonlinear phononics for coherent light control of solids, Phys. Rev. B 89 (2014) 220301, http://dx.doi.org/10.1103/PhysRevB.89.220301, URL https://link.aps.org/doi/10.1103/PhysRevB.89.220301.

[90] S. Gruebel, J.A. Johnson, P.B.C. Dornes, A. Ferrer, V. Haborets, L. Huber, T. Huber, A. Kohutych, T. Kubacka, S.O.M. M. Kubli, J. Rittmann, J.I. Saari, Y. Vysochanskii, G. Ingold, S.L. Johnson, Ultrafast X-ray diraction of a ferroelectric soft mode driven by broadband terahertz pulses, arXiv:1602.05435.

[91] R. Mankowsky, A. von Hoegen, M. Först, A. Cavalleri, Ultrafast reversal of the ferroelectric polarization, Phys. Rev. Lett. 118 (2017) 197601, http://dx.doi.org/10.1103/PhysRevLett.118.197601, URL https://link.aps.org/doi/10.1103/PhysRevLett.118.197601.

[92] C. Vicario, C. Ruchert, F. Ardana-Lamas, P.M. Derlet, B. Tudu, J. Luning, C.P. Hauri, Off-resonant magnetization dynamics phase-locked to an intense phase-stable terahertz transient, Nature Photon. 7 (9) (2013) 720.

[93] T. Kampfrath, A. Sell, G. Klatt, A. Pashkin, S. Mährlein, T. Dekorsy, M. Wolf, M. Fiebig, A. Leitenstorfer, R. Huber, Coherent terahertz control of antiferromagnetic spin waves, Nature Photon. 5 (1) (2011) 31.

[94] A.V. Kimel, A. Kirilyuk, P.A. Usachev, R.V. Pisarev, A.M. Balbashov, T. Rasing, Ultrafast non-thermal control of magnetization by instantaneous photomagnetic pulses, Nature 435 (2005) 655.

[95] D. Bossini, S. Dal Conte, Y. Hashimoto, A. Secchi, R.V. Pisarev, T. Rasing, G. Cerullo, A.V. Kimel, Macrospin dynamics in antiferromagnets triggered by sub-20 femtosecond injection of nanomagnons, Nature Commun. 7 (2016) 10645.

[96] T. Satoh, S.-J. Cho, R. Iida, T. Shimura, K. Kuroda, H. Ueda, Y. Ueda, B.A. Ivanov, F. Nori, M. Fiebig, Spin oscillations in antiferromagnetic NiO triggered by circularly polarized light, Phys. Rev. Lett. 105 (2010) 077402, http://dx.doi.org/10.1103/PhysRevLett.105.077402, URL https://link.aps.org/doi/10.1103/PhysRevLett.105.077402.

[97] F. Atoneche, A.M. Kalashnikova, A.V. Kimel, A. Stupakiewicz, A. Maziewski, A. Kirilyuk, T. Rasing, Large ultrafast photoinduced magnetic anisotropy in a cobalt-substituted yttrium iron garnet, Phys. Rev. B 81 (21) (2010) 214440.

[98] A. Stupakiewicz, K. Szerenos, D. Afanasiev, A. Kirilyuk, A.V. Kimel, Ultrafast nonthermal photo-magnetic recording in a transparent medium, Nature 542 (2017) 71.

[99] S. Baierl, M. Hohenleutner, T. Kampfrath, A.K. Zvezdin, A.V. Kimel, R. Huber, R.V. Mikhaylovskiy, Nonlinear spin control by terahertz-driven anisotropy fields, Nature Photon. 10 (11) (2016) 715.

[100] P. Němec, E. Rozkotová, N. Tesařová, F. Trojánek, E. De Ranieri, K. Olejník, J. Zemen, V. Novák, M. Cukr, P. Malỳ, et al., Experimental observation of the optical spin transfer torque, Nature Phys. 8 (5) (2012) 411.

[101] N. Tesařová, P. Němec, E. Rozkotová, J. Zemen, T. Janda, D. Butkovičová, F. Trojánek, K. Olejník, V. Novák, P. Malỳ, et al., Experimental observation of the optical spin–orbit torque, Nature Photon. 7 (6) (2013) 492.

[102] R.V. Mikhaylovskiy, E. Hendry, A. Secchi, J.H. Mentink, M. Eckstein, A. Wu, R.V. Pisarev, V.V. Kruglyak, M.I. Katsnelson, T. Rasing, et al., Ultrafast optical modification of exchange interactions in iron oxides, Nature Commun. 6 (2015) 8190.

[103] R.R. Subkhangulov, A.B. Henriques, P.H.O. Rappl, E. Abramof, T. Rasing, A.V. Kimel, All-optical manipulation and probing of the $d-f$ exchange interaction in eute, Sci. Rep. 4 (2014) 4368.

[104] J.H. Mentink, M. Eckstein, Ultrafast quenching of the exchange interaction in a Mott insulator, Phys. Rev. Lett. 113 (2014) 057201, http://dx.doi.org/10.1103/PhysRevLett.113.057201, URL https://link.aps.org/doi/10.1103/PhysRevLett.113.057201.

[105] D. Afanasiev, A. Gatilova, D.J. Groenendijk, B.A. Ivanov, M. Gibert, S. Gariglio, J. Mentink, J. Li, N. Dasari, M. Eckstein, et al., Ultrafast spin dynamics in photodoped spin-orbit mott insulator Sr_2Iro_4, Phys. Rev. X 9 (2) (2019) 021020.

[106] Y.T. Rebane, Faraday effect produced in the residual ray region by the magnetic moment of an optical phonon in an ionic crystal, J. Exp. Theor. Phys. 57 (1983) 1356.

[107] T.F. Nova, A. Cartella, A. Cantaluppi, M. Först, D. Bossini, R.V. Mikhaylovskiy, A.V. Kimel, R. Merlin, A. Cavalleri, An effective magnetic field from optically driven phonons, Nature Phys. 13 (2) (2017) 132.

[108] J.A. De Jong, I. Razdolski, A.M. Kalashnikova, R.V. Pisarev, A.M. Balbashov, A. Kirilyuk, T. Rasing, A.V. Kimel, Coherent control of the route of an ultrafast magnetic phase transition via low-amplitude spin precession, Phys. Rev. Lett. 108 (15) (2012) 157601.

[109] A. Zvezdin, A. Popov, K.I. Turkmenov, The magnetooptical anisotropy of rare-earth crystals, Fiz. Tverd. Tela 28 (6) (1986) 1760–1767.

[110] R. Hertel, Theory of the inverse Faraday effect in metals, J. Magn. Magn. Mater. 303 (1) (2006) L1 – L4, http://dx.doi.org/10.1016/j.jmmm.2005.10.225, URL http://www.sciencedirect.com/science/article/pii/S0304885305010747.

[111] N.A. Spaldin, M. Fiebig, The renaissance of magnetoelectric multiferroics, Science 309 (5733) (2005) 391–392, http://dx.doi.org/10.1126/science.1113357, arXiv:https://science.sciencemag.org/content/309/5733/391.full.pdf, URL https://science.sciencemag.org/content/309/5733/391.

[112] V.L. Korenev, M. Salewski, I.A. Akimov, V.F. Sapega, L. Langer, I.V. Kalitukha, J. Debus, R.I. Dzhioev, D.R. Yakovlev, D. Muller, C. Schroder, H. Hovel, G. Karczewski, M. Wiater, T. Wojtowicz, Y.G. Kusrayev, M. Bayer, Long-range $p--d$ exchange interaction in a ferromagnet–semiconductor hybrid structure, Nature Phys. 12 (2016) 85.

[113] O. Matsuda, M.C. Larciprete, R.L. Voti, O.B. Wright, Fundamentals of picosecond laser ultrasonics, Ultrasonics 56 (2015) 3–20, http://dx.doi.org/10.1016/j.ultras.2014.06.005, URL http://www.sciencedirect.com/science/article/pii/S0041624X14001541.

[114] A.V. Scherbakov, A.S. Salasyuk, A.V. Akimov, X. Liu, M. Bombeck, C. Brüggemann, D.R. Yakovlev, V.F. Sapega, J.K. Furdyna, M. Bayer, Coherent magnetization precession in ferromagnetic (Ga,Mn)as induced by picosecond acoustic pulses, Phys. Rev. Lett. 105 (2010) 117204, http://dx.doi.org/10.1103/PhysRevLett.105.117204, URL https://link.aps.org/doi/10.1103/PhysRevLett.105.117204.

[115] J.-W. Kim, M. Vomir, J.-Y. Bigot, Ultrafast magnetoacoustics in nickel films, Phys. Rev. Lett. 109 (2012) 166601, http://dx.doi.org/10.1103/PhysRevLett.109.166601, URL https://link.aps.org/doi/10.1103/PhysRevLett.109.166601.

[116] D. Afanasiev, I. Razdolski, K.M. Skibinsky, D. Bolotin, S.V. Yagupov, M.B. Strugatsky, A. Kirilyuk, T. Rasing, A.V. Kimel, Laser excitation of lattice-driven anharmonic magnetization dynamics in dielectric $FeBO_3$, Phys. Rev. Lett. 112 (14) (2014) 147403.

[117] O. Kovalenko, T. Pezeril, V.V. Temnov, New concept for magnetization switching by ultrafast acoustic pulses, Phys. Rev. Lett. 110 (26) (2013) 266602.

[118] A. Szilva, M. Costa, A. Bergman, L. Szunyogh, L. Nordström, O. Eriksson, Interatomic exchange interactions for finite-temperature magnetism and nonequilibrium spin dynamics, Phys. Rev. Lett. 111 (12) (2013) 127204.

[119] M. Fechner, A. Sukhov, L. Chotorlishvili, C. Kenel, J. Berakdar, N.A. Spaldin, Magnetophononics: Ultrafast spin control through the lattice, Phys. Rev. Mater. 2 (2018) 064401, http://dx.doi.org/10.1103/PhysRevMaterials.2.064401, URL https://link.aps.org/doi/10.1103/PhysRevMaterials.2.064401.

[120] M. Stamenova, S. Sanvito, Dynamical exchange interaction from time-dependent spin density functional theory, Phys. Rev. B 88 (10) (2013) 104423.

[121] A. Secchi, S. Brener, A.I. Lichtenstein, M.I. Katsnelson, Non-equilibrium magnetic interactions in strongly correlated systems, Ann. Physics 333 (2013) 221–271.

[122] V.V. Pavlov, R.V. Pisarev, S.G. Nefedov, I.A. Akimov, D.R. Yakovlev, M. Bayer, A.B. Henriques, P.H.O. Rappl, E. Abramof, Magnetic-field-induced crossover from the inverse Faraday effect to the optical orientation in EuTe, J. Appl. Phys. 123 (19) (2018) 193102, http://dx.doi.org/10.1063/1.5027473, arXiv:https://doi.org/10.1063/1.5027473.

[123] R.V. Mikhaylovskiy, T.J. Huisman, A.I. Popov, A.K. Zvezdin, T. Rasing, R.V. Pisarev, A.V. Kimel, Terahertz magnetization dynamics induced by femtosecond resonant pumping of Dy^{3+} subsystem in the multisublattice antiferromagnet $DyFeO_3$, Phys. Rev. B 92 (9) (2015) 094437.

[124] P. Hamm, M. Meuwly, S.L. Johnson, P. Beaud, U. Staub, Perspective: THz-driven nuclear dynamics from solids to molecules, Struct. Dyn. 4 (6) (2017) 061601, http://dx.doi.org/10.1063/1.4992050, arXiv:https://doi.org/10.1063/1.4992050.

[125] M. Forst, C. Manzoni, S. Kaiser, Y. Tomioka, Y. Tokura, R. Merlin, A. Cavalleri, Nonlinear phononics as an ultrafast route to lattice control, Nat. Phys. 7 (2011) 854.

[126] R. Mankowsky, M. Forst, A. Cavalleri, Non-equilibrium control of complex solids by nonlinear phononics, Rep. Progr. Phys. 79 (6) (2016) 064503, http://dx.doi.org/10.1088/0034-4885/79/6/064503.

[127] D. Nicoletti, A. Cavalleri, Nonlinear light–matter interaction at terahertz frequencies, Adv. Opt. Photon. 8 (3) (2016) 401–464, http://dx.doi.org/10.1364/AOP.8.000401, URL http://aop.osa.org/abstract.cfm?URI=aop-8-3-401.

[128] D.M. Juraschek, S.F. Maehrlein, Sum-frequency ionic Raman scattering, Phys. Rev. B 97 (2018) 174302, http://dx.doi.org/10.1103/PhysRevB.97.174302, URL https://link.aps.org/doi/10.1103/PhysRevB.97.174302.

[129] Y.-X. Yan, K.A. Nelson, Impulsive stimulated light scattering. I. General theory, J. Chem. Phys. 87 (1987) 6240, http://dx.doi.org/10.1063/1.453733; Impulsive stimulated light scattering. II. Comparison to frequency-domain light-scattering spectroscopy, J. Chem. Phys. 87 (1987) 6257, http://dx.doi.org/10.1063/1.453454.

[130] R. Merlin, Generating coherent THz phonons with light pulses, Solid State Commun. 102 (1997) 207.

[131] K. Imasaka, R.V. Pisarev, L.N. Bezmaternykh, T. Shimura, A.M. Kalashnikova, T. Satoh, Excitation of multiple phonon modes in copper metaborate CuB_2O_4 via nonresonant impulsive stimulated Raman scattering, Phys. Rev. B 98 (2018) 054303, http://dx.doi.org/10.1103/PhysRevB.98.054303, URL https://link.aps.org/doi/10.1103/PhysRevB.98.054303.

[132] R.F. Wallis, A.A. Maradudin, Ionic Raman effect. II. The first-order ionic Raman effect, Phys. Rev. B 3 (1971) 2063–2075, http://dx.doi.org/10.1103/PhysRevB.3.2063, URL https://link.aps.org/doi/10.1103/PhysRevB.3.2063.

[133] T.P. Martin, L. Genzel, Ionic Raman scattering and ionic frequency mixing, Phys. Status Solidi (b) 61 (2) (1974) 493–502, http://dx.doi.org/10.1002/pssb.2220610214, arXiv:https://onlinelibrary.wiley.com/doi/pdf/10.1002/pssb.2220610214. URL https://onlinelibrary.wiley.com/doi/abs/10.1002/pssb.2220610214.

[134] M. Kozina, M. Fechner, P. Marsik, T. van Driel, J.M. Glownia, C. Bernhard, M. Radovic, D. Zhu, S. Bonetti, U. Staub, M.C. Hoffmann, Terahertz-driven phonon upconversion in $SrTiO_3$, Nat. Phys. 15 (2019) 387.

[135] I.B. Bersuker, B.G. Vekhter, The vibronic theory of ferroelectricity, Ferroelectrics 19 (1) (1978) 137–150, http://dx.doi.org/10.1080/00150197808237842, arXiv:https://doi.org/10.1080/00150197808237842.

[136] D.M. Juraschek, M. Fechner, N.A. Spaldin, Ultrafast structure switching through nonlinear phononics, Phys. Rev. Lett. 118 (2017) 054101, http://dx.doi.org/10.1103/PhysRevLett.118.054101, URL https://link.aps.org/doi/10.1103/PhysRevLett.118.054101.

[137] T. Qi, Y.-H. Shin, K.-L. Yeh, K.A. Nelson, A.M. Rappe, Collective coherent control: Synchronization of polarization in ferroelectric $PbTiO_3$ by shaped THz fields, Phys. Rev. Lett. 102 (2009) 247603, http://dx.doi.org/10.1103/PhysRevLett.102.247603, URL https://link.aps.org/doi/10.1103/PhysRevLett.102.247603.

[138] T.P. Dougherty, G.P. Wiederrecht, K.A. Nelson, M.H. Garrett, H.P. Jensen, C. Warde, Femtosecond resolution of soft mode dynamics in structural phase transitions, Science 258 (5083) (1992) 770–774, http://dx.doi.org/10.1126/science.258.5083.770, arXiv:https://science.sciencemag.org/content/258/5083/770.full.pdf.

[139] T.P. Dougherty, G.P. Wiederrecht, K.A. Nelson, M.H. Garrett, H.P. Jenssen, C. Warde, Femtosecond time-resolved spectroscopy of soft modes in structural phase transitions of perovskites, Phys. Rev. B 50 (1994) 8996–9019, http://dx.doi.org/10.1103/PhysRevB.50.8996, URL https://link.aps.org/doi/10.1103/PhysRevB.50.8996.

[140] A. Cavalleri, S. Wall, C. Simpson, E. Statz, D.W. Ward, K.A. Nelson, M. Rini, R.W. Schoenlein, Tracking the motion of charges in a terahertz light field by femtosecond X-ray diffraction, Nature 1442 (2006) 664.

[141] K.A. Brekhov, K.A. Grishunin, D.V. Afanas'ev, S.V. Semin, N.E. Sherstyuk, G.K. Kitaeva, E.D. Mishina, T. Rasing, A.V. Kimel, Photoinduced dynamics and femtosecond excitation of phonon modes in ferroelectric semiconductor $Sn_2P_2S_6$, JETP Lett. 102 (6) (2015) 372–377.

[142] F. Chen, Y. Zhu, S. Liu, Y. Qi, H.Y. Hwang, N.C. Brandt, J. Lu, F. Quirin, H. Enquist, P. Zalden, T. Hu, J. Goodfellow, M.-J. Sher, M.C. Hoffmann, D. Zhu, H. Lemke, J. Glownia, M. Chollet, A.R. Damodaran, J. Park, Z. Cai, I.W. Jung, M.J. Highland, D.A. Walko, J.W. Freeland, P.G. Evans, A. Vailionis, J. Larsson, K.A. Nelson, A.M. Rappe, K. Sokolowski-Tinten, L.W. Martin, H. Wen, A.M. Lindenberg, Ultrafast terahertz-field-driven ionic response in ferroelectric $BaTiO_3$, Phys. Rev. B 94 (2016) 180104, http://dx.doi.org/10.1103/PhysRevB.94.180104, URL https://link.aps.org/doi/10.1103/PhysRevB.94.180104.

[143] X. Li, T. Qiu, J. Zhang, E. Baldini, J. Lu, A.M. Rappe, K.A. Nelson, Terahertz field–induced ferroelectricity in quantum paraelectric $SrTiO_3$, Science 364 (6445) (2019) 1079–1082, http://dx.doi.org/10.1126/science.aaw4913, arXiv:https://science.sciencemag.org/content/364/6445/1079.full.pdf. URL https://science.sciencemag.org/content/364/6445/1079.

[144] D. Daranciang, M.J. Highland, H. Wen, S.M. Young, N.C. Brandt, H.Y. Hwang, M. Vattilana, M. Nicoul, F. Quirin, J. Goodfellow, T. Qi, I. Grinberg, D.M. Fritz, M. Cammarata, D. Zhu, H.T. Lemke, D.A. Walko, E.M. Dufresne, Y. Li, J. Larsson, D.A. Reis, K. Sokolowski-Tinten, K.A. Nelson, A.M. Rappe, P.H. Fuoss, G.B. Stephenson, A.M. Lindenberg, Ultrafast photovoltaic response in ferroelectric nanolayers, Phys. Rev. Lett. 108 (2012) 087601, http://dx.doi.org/10.1103/PhysRevLett.108.087601, URL https://link.aps.org/doi/10.1103/PhysRevLett.108.087601.

[145] T. Miyamoto, H. Yada, H. Yamakawa, O. H., Ultrafast modulation of polarization amplitude by terahertz fields in electronic-type organic ferroelectrics, Nature Commun. 4 (2013) 2586.

[146] D.S. Rana, I. Kawayama, K. Mavani, K. Takahashi, H. Murakami, M. Tonouchi, Understanding the nature of ultrafast polarization dynamics of ferroelectric memory in the multiferroic $BiFeO_3$, Adv. Mater. 21 (28) (2009) 2881–2885, http://dx.doi.org/10.1002/adma.200802094, arXiv:https://onlinelibrary.wiley.com/doi/pdf/10.1002/adma.200802094. URL https://onlinelibrary.wiley.com/doi/abs/10.1002/adma.200802094.

[147] K. Takahashi, N. Kida, M. Tonouchi, Terahertz radiation by an ultrafast spontaneous polarization modulation of multiferroic $BiFeO_3$ thin films, Phys. Rev. Lett. 96 (2006) 117402, http://dx.doi.org/10.1103/PhysRevLett.96.117402, URL https://link.aps.org/doi/10.1103/PhysRevLett.96.117402.

[148] M. Porer, M. Fechner, E.M. Bothschafter, L. Rettig, M. Savoini, V. Esposito, J. Rittmann, M. Kubli, M.J. Neugebauer, E. Abreu, T. Kubacka, T. Huber, G. Lantz, S. Parchenko, S. Grübel, A. Paarmann, J. Noack, P. Beaud, G. Ingold, U. Aschauer, S.L. Johnson, U. Staub, Ultrafast relaxation dynamics of the antiferrodistortive phase in Ca doped $SrTiO_3$, Phys. Rev. Lett. 121 (2018) 055701, http://dx.doi.org/10.1103/PhysRevLett.121.055701, URL https://link.aps.org/doi/10.1103/PhysRevLett.121.055701.

[149] T. Lottermoser, D. Meier, R.V. Pisarev, M. Fiebig, Giant coupling of second-harmonic generation to a multiferroic polarization, Phys. Rev. B 80 (2009) 100101, http://dx.doi.org/10.1103/PhysRevB.80.100101, URL https://link.aps.org/doi/10.1103/PhysRevB.80.100101.

[150] J. Hemberger, P. Lunkenheimer, R. Fichtl, H.-A. Krug von Nidda, V. Tsurkan, A. Loidl, Relaxor ferroelectricity and colossal magnetocapacitive coupling in ferromagnetic $CdCr_2S_4$, Nature 434 (2015) 364, http://dx.doi.org/10.1038/nature03348.

[151] N. Ikeda, H. Ohsumi, K. Ohwada, K. Ishii, T. Inami, K. Kakurai, Y. Murakami, K. Yoshii, S. Mori, Y. Horibe, H. Kitô, Ferroelectricity from iron valence ordering in the charge-frustrated system $LuFe_2O_4$, Nature 436 (2015) 1136, http://dx.doi.org/10.1038/nature04039.

[152] S.-y. Koshihara, Y. Takahashi, H. Sakai, Y. Tokura, T. Luty, Photoinduced cooperative charge transfer in low-dimensional organic crystals, J. Phys. Chem. B 103 (14) (1999) 2592–2600, http://dx.doi.org/10.1021/jp984172i, arXiv:https://doi.org/10.1021/jp984172i.

[153] E. Collet, M.-H. Lemée-Cailleau, M. Buron-Le Cointe, H. Cailleau, M. Wulff, T. Luty, S.-Y. Koshihara, M. Meyer, L. Toupet, P. Rabiller, et al., Laser-induced ferroelectric structural order in an organic charge-transfer crystal, Science 300 (5619) (2003) 612–615.

[154] S. Iwai, S. Tanaka, K. Fujinuma, H. Kishida, H. Okamoto, Y. Tokura, Ultrafast optical switching from an ionic to a neutral state in tetrathiafulvalene-*p*-chloranil (TTF-CA) observed in femtosecond reflection spectroscopy, Phys. Rev. Lett. 88 (2002) 057402, http://dx.doi.org/10.1103/PhysRevLett.88.057402, URL https://link.aps.org/doi/10.1103/PhysRevLett.88.057402.

[155] T. Morimoto, T. Miyamoto, H. Yamakawa, T. Terashige, T. Ono, N. Kida, H. Okamoto, Terahertz-field-induced large macroscopic polarization and domain-wall dynamics in an organic molecular dielectric, Phys. Rev. Lett. 118 (2017) 107602, http://dx.doi.org/10.1103/PhysRevLett.118.107602, URL https://link.aps.org/doi/10.1103/PhysRevLett.118.107602.

[156] F. Chen, J. Goodfellow, S. Liu, I. Grinberg, M.C. Hoffmann, A.R. Damodaran, Y. Zhu, P. Zalden, X. Zhang, I. Takeuchi, A.M. Rappe, L.W. Martin, H. Wen, A.M. Lindenberg, Ultrafast terahertz gating of the polarization and giant nonlinear optical response in $BiFeO_3$ thin films, Adv. Mater. 27 (41) (2015) 6371–6375, http://dx.doi.org/10.1002/adma.201502975, arXiv:https://onlinelibrary.wiley.com/doi/pdf/10.1002/adma.201502975. URL https://onlinelibrary.wiley.com/doi/abs/10.1002/adma.201502975.

[157] K. Grishunin, V. Bilyk, N. Sherstyuk, V. Mukhortov, A. Ovchinnikov, O. Chefonov, M. Agranat, E. Mishina, A.V. Kimel, Transient Second Harmonic Generation Induced by Single Cycle THz pulses in $Ba_{0.8}Sr_{0.2}TiO_3$/MgO, Sci. Rep. 9 (2019) 697.

[158] S. Iwai, K. Yamamoto, A. Kashiwazaki, F. Hiramatsu, H. Nakaya, Y. Kawakami, K. Yakushi, H. Okamoto, H. Mori, Y. Nishio, Photoinduced melting of a stripe-type charge-order and metallic domain formation in a layered BEDT-TTF-based organic salt, Phys. Rev. Lett. 98 (2007) 097402, http://dx.doi.org/10.1103/PhysRevLett.98.097402, URL https://link.aps.org/doi/10.1103/PhysRevLett.98.097402.

[159] M.B. Agranat, S.I. Ashitkov, A.B. Granovskii, G.I. Rukman, Interaction of picosecond laser pulses with the electron, spin, and phonon subsystems of nickel, J. Exp. Theor. Phys. 59 (4) (1984) 804.

[160] B. Koopmans, G. Malinowski, F. Dalla Longa, D. Steiauf, M. Fähnle, T. Roth, M. Cinchetti, M. Aeschlimann, Explaining the paradoxical diversity of ultrafast laser-induced demagnetization, Nature Mater. 9 (3) (2010) 259.

[161] A. Mekonnen, A.R. Khorsand, M. Cormier, A.V. Kimel, A. Kirilyuk, A. Hrabec, L. Ranno, A. Tsukamoto, A. Itoh, T. Rasing, Role of the inter-sublattice exchange coupling in short-laser-pulse-induced demagnetization dynamics of GdCo and GdCoFe alloys, Phys. Rev. B 87 (18) (2013) 180406.

[162] I. Radu, K. Vahaplar, C. Stamm, T. Kachel, N. Pontius, H.A. Dürr, T.A. Ostler, J. Barker, R.F.L. Evans, R.W. Chantrell, et al., Transient ferromagnetic-like state mediating ultrafast reversal of antiferromagnetically coupled spins, Nature 472 (7342) (2011) 205.

[163] A.V. Kimel, R.V. Pisarev, J. Hohlfeld, T. Rasing, Ultrafast quenching of the antiferromagnetic order in $FeBO_3$: Direct optical probing of the phonon-magnon coupling, Phys. Rev. Lett. 89 (2002) 287401, http://dx.doi.org/10.1103/PhysRevLett.89.287401, URL https://link.aps.org/doi/10.1103/PhysRevLett.89.287401.

[164] J.-Y. Bigot, M. Vomir, L. Andrade, E. Beaurepaire, Ultrafast magnetization dynamics in ferromagnetic cobalt: The role of the anisotropy, Chem. Phys. 318 (1) (2005) 137–146, http://dx.doi.org/10.1016/j.chemphys.2005.06.016, Molecular nanoscience. URL http://www.sciencedirect.com/science/article/pii/S030101040500248X.

[165] E. Carpene, E. Mancini, D. Dazzi, C. Dallera, E. Puppin, S. De Silvestri, Ultrafast three-dimensional magnetization precession and magnetic anisotropy of a photoexcited thin film of iron, Phys. Rev. B 81 (2010) 060415, http://dx.doi.org/10.1103/PhysRevB.81.060415, URL https://link.aps.org/doi/10.1103/PhysRevB.81.060415.

[166] V.N. Kats, T.L. Linnik, A.S. Salasyuk, A.W. Rushforth, M. Wang, P. Wadley, A.V. Akimov, S.A. Cavill, V. Holy, A.M. Kalashnikova, A.V. Scherbakov, Ultrafast changes of magnetic anisotropy driven by laser-generated coherent and noncoherent phonons in metallic films, Phys. Rev. B 93 (2016) 214422, http://dx.doi.org/10.1103/PhysRevB.93.214422, URL https://link.aps.org/doi/10.1103/PhysRevB.93.214422.

[167] L.A. Shelukhin, V.V. Pavlov, P.A. Usachev, P.Y. Shamray, R.V. Pisarev, A.M. Kalashnikova, Ultrafast laser-induced changes of the magnetic anisotropy in a low-symmetry iron garnet film, Phys. Rev. B 97 (2018) 014422, http://dx.doi.org/10.1103/PhysRevB.97.014422, URL https://link.aps.org/doi/10.1103/PhysRevB.97.014422.

[168] J. Larsson, P. Sondhauss, O. Synnergren, M. Harbst, P. Heimann, A. Lindenberg, J. Wark, Time-resolved X-ray diffraction study of the ferroelectric phase-transition in DKDP, Chem. Phys. 299 (2) (2004) 157–161, http://dx.doi.org/10.1016/j.chemphys.2003.11.019, URL http://www.sciencedirect.com/science/article/pii/S0301010403006505.

[169] K. Istomin, V. Kotaidis, A. Plech, Q. Kong, Dynamics of the laser-induced ferroelectric excitation in $BaTiO_3$ studied by X-ray diffraction, Appl. Phys. Lett. 90 (2) (2007) 022905, http://dx.doi.org/10.1063/1.2430773, arXiv:https://doi.org/10.1063/1.2430773.

[170] G.M. Genkin, I.D. Tokman, V.V. Zil'berg, Photomagnetization of polydomain magnetic insulators by circularly polarized light, Solid State Commun. 44 (5) (1982) 631–633.

[171] G.M. Genkin, I.D. Tokman, N.V. Shedrina, Readjustment of ferroelectric domain structure under the action of polarized light, Sov. Phys. J. 27 (6) (1984) 455–458, https://doi.org/10.1007/BF00901860.

[172] G. Genkin, I. Tokman, Bubble collapse of magnetic domains under the circular-polarized light effect, Fiz. Tverd. Tela 25 (7) (1983) 2206–2208.

[173] D. Yudin, D.R. Gulevich, M. Titov, Light-induced anisotropic skyrmion and stripe phases in a Rashba ferromagnet, Phys. Rev. Lett. 119 (14) (2017) 147202.
[174] S. Selzer, U. Atxitia, U. Ritzmann, D. Hinzke, U. Nowak, Inertia-free thermally driven domain-wall motion in antiferromagnets, Phys. Rev. Lett. 117 (10) (2016) 107201.
[175] R. Medapalli, D. Afanasiev, D.K. Kim, Y. Quessab, S. Manna, S.A. Montoya, A. Kirilyuk, T. Rasing, A.V. Kimel, E.E. Fullerton, Multiscale dynamics of helicity-dependent all-optical magnetization reversal in ferromagnetic Co/Pt multilayers, Phys. Rev. B 96 (22) (2017) 224421.
[176] M.O.A. Ellis, E.E. Fullerton, R.W. Chantrell, All-optical switching in granular ferromagnets caused by magnetic circular dichroism, Sci. Rep. 6 (2016) 30522.
[177] M.V. Gerasimov, M.V. Logunov, A.V. Spirin, Y.N. Nozdrin, I.D. Tokman, Time evolution of domain-wall motion induced by nanosecond laser pulses, Phys. Rev. B 94 (2016) 014434, http://dx.doi.org/10.1103/PhysRevB.94.014434, URL https://link.aps.org/doi/10.1103/PhysRevB.94.014434.
[178] T. Higuchi, M. Kuwata-Gonokami, Control of antiferromagnetic domain distribution via polarization-dependent optical annealing, Nature Commun. 7 (2016) 10720.
[179] S. Manz, M. Matsubara, T. Lottermoser, J. Büchi, A. Iyama, T. Kimura, D. Meier, M. Fiebig, Reversible optical switching of antiferromagnetism in $TbMnO_3$, Nature Photon. 10 (10) (2016) 653.
[180] G.M. Genkin, Y.N. Nozdrin, I.D. Tokman, V.N. Shastin, Direct observation of photo-magnetization of the ferromagnet $CdCr_2Se_4$ by circularly-polarized light, JETP Lett. 35 (4) (1982) 199–202.
[181] G.M. Genkin, Y.N. Nozdrin, A.V. Okomel'Kov, I.D. Tokman, Magnetization of a garnet film through a change in its multidomain structure under circularly polarized light, Phys. Rev. B 86 (2) (2012) 024405.
[182] R. John, M. Berritta, D. Hinzke, C. Müller, T. Santos, H. Ulrichs, P. Nieves, J. Walowski, R. Mondal, O. Chubykalo-Fesenko, et al., Magnetisation switching of FePt nanoparticle recording medium by femtosecond laser pulses, Sci. Rep. 7 (1) (2017) 4114.
[183] A. Pogrebna, S. Barsaume, R.R. Subkhangulov, A.V. Telegin, Y.P. Sukhorukov, A.V. Chzhan, T. Rasing, A.V. Kimel, Spectral tunability of laser-induced spin dynamics in the ferromagnetic semiconductor $CdCr_2Se_4$, Phys. Rev. B 98 (21) (2018) 214427.
[184] S. Mizukami, S. Iihama, N. Inami, T. Hiratsuka, G. Kim, H. Naganuma, M. Oogane, Y. Ando, Fast magnetization precession observed in $L1_0$-FePt epitaxial thin film, Appl. Phys. Lett. 98 (5) (2011) 052501.
[185] J. Becker, O. Mosendz, D. Weller, A. Kirilyuk, J.C. Maan, P.C.M. Christianen, T. Rasing, A. Kimel, Laser induced spin precession in highly anisotropic granular $L1_0$ FePt, Appl. Phys. Lett. 104 (15) (2014) 152412.
[186] Y.M. Fedorov, A.A. Leksikov, A.E. Aksenov, Light-induced dynamic instability of the domain structure in $FeBO_3$:Ni, JETP Lett. 37 (3) (1983) 161–164.
[187] K. Olejník, T. Seifert, Z. Kašpar, V. Novák, P. Wadley, R.P. Campion, M. Baumgartner, P. Gambardella, P. Němec, J. Wunderlich, et al., Terahertz electrical writing speed in an antiferromagnetic memory, Sci. Adv. 4 (3) (2018) eaar3566.
[188] Y.-D. Liou, Y.-Y. Chiu, R.T. Hart, C.-Y. Kuo, Y.-L. Huang, Y.-C. Wu, R.V. Chopdekar, H.-J. Liu, A. Tanaka, C.-F. Chen, Chien-Te anc Chang, L.H. Tjeng, Y. Cao, V. Nagarajan, Y.-H. Chu, Y.-C. Chen, J.-C. Yang, Deterministic optical control of room temperature multiferroicity in bifeo3 thin films, Nature Mater. 18 (2019) 5805873, http://dx.doi.org/10.1038/s41563-019-0348-x.
[189] P. Zubko, G. Catalan, A.K. Tagantsev, Flexoelectric effect in solids, Annu. Rev. Mater. Res. 43 (1) (2013) 387–421, http://dx.doi.org/10.1146/annurev-matsci-071312-121634, arXiv:https://doi.org/10.1146/annurev-matsci-071312-121634.
[190] G. Vats, Y. Bai, D. Zhang, J. Juuti, J. Seidel, Optical control of ferroelectric domains: Nanoscale insight into macroscopic observations, Advanced Optical Materials 7 (2019) 1800858, http://dx.doi.org/10.1002/adom.201800858, arXiv:https://onlinelibrary.wiley.com/doi/pdf/10.1002/adom.201800858. URL https://onlinelibrary.wiley.com/doi/abs/10.1002/adom.201800858.
[191] M.V. Gerasimov, S.V. Ilin, M.V. Logunov, S.A. Nikitov, A.V. Spirin, A.N. Chaldyshkin, A magneto-optical setup for studying the time evolution of nanoscale domain-wall displacements under pulsed magnetization, Instrum. Exp. Tech. 60 (5) (2017) 716–721.
[192] L. Müller, S. Schleitzer, C. Gutt, B. Pfau, S. Schaffert, J. Geilhufe, C. von Korff Schmising, M. Schneider, C.M. Günther, F. Büttner, F. Capotondi, E. Pedersoli, S. Düsterer, H. Redlin, A. Al-Shemmary, R. Treusch, J. Bach, R. Frömter, B. Vodungbo, J. Gautier, P. Zeitoun, H. Popescu, V. Lopez-Flores, N. Beaulieu, F. Sirotti, N. Jaouen, G. Malinowski, B. Tudu, K. Li, J. Lüning, H.P. Oepen, M. Kiskinova, S. Eisebitt, G. Grübel, Ultrafast dynamics of magnetic domain structures probed by coherent free-electron laser light, Synchrotron Radiat. News 26 (6) (2013) 27–32, http://dx.doi.org/10.1080/08940886.2013.850384, arXiv:https://doi.org/10.1080/08940886.2013.850384.
[193] C.H. Back, D. Weller, J. Heidmann, D. Mauri, D. Guarisco, E.L. Garwin, H.C. Siegmann, Magnetization reversal in ultrashort magnetic field pulses, Phys. Rev. Lett. 81 (1998) 3251–3254, http://dx.doi.org/10.1103/PhysRevLett.81.3251, URL https://link.aps.org/doi/10.1103/PhysRevLett.81.3251.
[194] C.H. Back, R. Allenspach, W. Weber, S.S.P. Parkin, D. Weller, E.L. Garwin, H.C. Siegmann, Minimum field strength in precessional magnetization reversal, Science 285 (5429) (1999) 864–867, http://dx.doi.org/10.1126/science.285.5429.864, arXiv:https://science.sciencemag.org/content/285/5429/864.full.pdf. URL https://science.sciencemag.org/content/285/5429/864.
[195] I. Tudosa, C. Stamm, A.B. Kashuba, F. King, H.C. Siegmann, J. Stöhr, G. Ju, B. Lu, D. Weller, The ultimate speed of magnetic switching in granular recording media, Nature 428 (6985) (2004) 831.
[196] S.J. Gamble, M.H. Burkhardt, A. Kashuba, R. Allenspach, S.S.P. Parkin, H.C. Siegmann, J. Stöhr, Electric field induced magnetic anisotropy in a ferromagnet, Phys. Rev. Lett. 102 (2009) 217201, http://dx.doi.org/10.1103/PhysRevLett.102.217201, URL https://link.aps.org/doi/10.1103/PhysRevLett.102.217201.
[197] S. Kaka, S.E. Russek, Precessional switching of submicrometer spin valves, Appl. Phys. Lett. 80 (16) (2002) 2958–2960.
[198] S. Wienholdt, D. Hinzke, U. Nowak, THz switching of antiferromagnets and ferrimagnets, Phys. Rev. Lett. 108 (24) (2012) 247207.
[199] D. Weller, O. Mosendz, G. Parker, S. Pisana, T.S. Santos, $L1_0$ FePtX–Y media for heat-assisted magnetic recording, Phys. Status Solidi (a) 210 (7) (2013) 1245–1260.
[200] W. Challener, C. Peng, A. Itagi, D. Karns, W. Peng, Y. Peng, X. Yang, X. Zhu, N. Gokemeijer, Y.-T. Hsia, et al., Heat-assisted magnetic recording by a near-field transducer with efficient optical energy transfer, Nature Photon. 3 (4) (2009) 220.
[201] C.S. Davies, K.H. Prabhakara, M.D. Davydova, K.A. Zvezdin, T.B. Shapaeva, S. Wang, A.K. Zvezdin, A. Kirilyuk, T. Rasing, A.V. Kimel, Anomalously damped heat-assisted route for precessional magnetization reversal in an iron garnet, Phys. Rev. Lett. 122 (2019) 027202, http://dx.doi.org/10.1103/PhysRevLett.122.027202, URL https://link.aps.org/doi/10.1103/PhysRevLett.122.027202.
[202] K.P. Belov, A.K. Zvezdin, A.M. Kadomtseva, R.Z. Levitin, Spin-reorientation transitions in rare-earth magnets, Sov. Phys. Usp. 19 (7) (1976) 574–596, http://dx.doi.org/10.1070/pu1976v019n07abeh005274.
[203] D. Afanasiev, B.A. Ivanov, A. Kirilyuk, T. Rasing, R.V. Pisarev, A.V. Kimel, Control of the ultrafast photoinduced magnetization across the morin transition in $DyFeO_3$, Phys. Rev. Lett. 116 (9) (2016) 097401.
[204] A. Maziewski, Unexpected magnetization processes in YIG+ Co films, J. Magn. Magn. Mater. 88 (3) (1990) 325–342.
[205] A. Stupakiewicz, K. Szerenos, M.D. Davydova, K.A. Zvezdin, A.K. Zvezdin, A. Kirilyuk, A.V. Kimel, Selection rules for all-optical magnetic recording in iron garnet, Nature Commun. 10 (1) (2019) 612.
[206] K. Szerenos, A. Kimel, A. Maziewski, A. Kirilyuk, A. Stupakiewicz, Fundamental limits on the repetition rate of photomagnetic recording, Phys. Rev. Appl. 12 (2019) 044057, http://dx.doi.org/10.1103/PhysRevApplied.12.044057, URL https://link.aps.org/doi/10.1103/PhysRevApplied.12.044057.

[207] C.D. Stanciu, F. Hansteen, A.V. Kimel, A. Kirilyuk, A. Tsukamoto, A. Itoh, T. Rasing, All-optical magnetic recording with circularly polarized light, Phys. Rev. Lett. 99 (2007) 047601, http://dx.doi.org/10.1103/PhysRevLett.99.047601, URL https://link.aps.org/doi/10.1103/PhysRevLett.99.047601.

[208] T. Ostler, J. Barker, R. Evans, R. Chantrell, U. Atxitia, O. Chubykalo-Fesenko, S. El Moussaoui, L. Le Guyader, E. Mengotti, L. Heyderman, F. Nolting, A. Tsukamoto, A. Itoh, D. Afanasiev, B. Ivanov, A. Kalashnikova, K. Vahaplar, J. Mentink, A. Kirilyuk, T. Rasing, A. Kimel, Ultrafast heating as a sufficient stimulus for magnetization reversal in a ferrimagnet, Nature Commun. 3 (2012) 666, http://dx.doi.org/10.1146/annurev-matsci-071312-121634, URL https://doi.org/10.1038/ncomms1666.

[209] J.H. Mentink, J. Hellsvik, D.V. Afanasiev, B.A. Ivanov, A. Kirilyuk, A.V. Kimel, O. Eriksson, M.I. Katsnelson, T. Rasing, Ultrafast spin dynamics in multisublattice magnets, Phys. Rev. Lett. 108 (2012) 057202, http://dx.doi.org/10.1103/PhysRevLett.108.057202, URL https://link.aps.org/doi/10.1103/PhysRevLett.108.057202.

[210] J. Gorchon, R.B. Wilson, Y. Yang, A. Pattabi, J.Y. Chen, L. He, J.P. Wang, M. Li, J. Bokor, Role of electron and phonon temperatures in the helicity-independent all-optical switching of GdFeCo, Phys. Rev. B 94 (18) (2016) 184406.

[211] Y. Yang, R.B. Wilson, J. Gorchon, C.-H. Lambert, S. Salahuddin, J. Bokor, Ultrafast magnetization reversal by picosecond electrical pulses, Sci. Adv. 3 (11) (2017) e1603117.

[212] C.S. Davies, T. Janssen, Mentink, A. Tsukamoto, A.V. Kimel, A.F.G. van der Meer, A. Stupakiewicz, A. Kirilyuk, Blueprint for deterministic all-optical switching of magnetization, arXiv:1904.11977.

[213] C.E. Graves, A.H. Reid, T. Wang, B. Wu, S. de Jong, K. Vahaplar, I. Radu, D.P. Bernstein, M. Messerschmidt, L. Müller, R. Coffee, M. Bionta, S.W. Epp, R. Hartmann, N. Kimmel, G. Hauser, A. Hartmann, P. Holl, H. Gorke, J.H. Mentink, A. Tsukamoto, A. Fognini, J.J. Turner, W.F. Schlotter, D. Rolles, H. Soltau, L. Strüder, Y. Acremann, A.V. Kimel, A. Kirilyuk, T. Rasing, J. Stöhr, A.O. Scherz, H.A. Dörr, Nanoscale spin reversal by non-local angular momentum transfer following ultrafast laser excitation in ferrimagnetic GdFeCo, Nature Mater. 12 (2013) 293, http://dx.doi.org/10.1038/nmat3597.

[214] F. Junginger, A. Sell, O. Schubert, B. Mayer, D. Brida, M. Marangoni, G. Cerullo, A. Leitenstorfer, R. Huber, Single-cycle multiterahertz transients with peak fields above 10 MV/cm, Opt. Lett. 35 (15) (2010) 2645–2647, http://dx.doi.org/10.1364/OL.35.002645, URL http://ol.osa.org/abstract.cfm?URI=ol-35-15-2645.

[215] C. Vicario, A.V. Ovchinnikov, S.I. Ashitkov, M.B. Agranat, V.E. Fortov, C.P. Hauri, Generation of 0.9-mJ THz pulses in DSTMS pumped by a Cr:Mg_2SiO_4 laser, Opt. Lett. 39 (23) (2014) 6632–6635, http://dx.doi.org/10.1364/OL.39.006632, URL http://ol.osa.org/abstract.cfm?URI=ol-39-23-6632.

[216] C. Vicario, C. Ruchert, C. Hauri, High field broadband THz generation in organic materials, J. Modern Opt. 62 (18) (2015) 1480–1485, http://dx.doi.org/10.1080/09500340.2013.800242, arXiv:https://doi.org/10.1080/09500340.2013.800242.

[217] A. Subedi, Proposal for ultrafast switching of ferroelectrics using midinfrared pulses, Phys. Rev. B 92 (2015) 214303, http://dx.doi.org/10.1103/PhysRevB.92.214303, URL https://link.aps.org/doi/10.1103/PhysRevB.92.214303.

[218] M. Madami, D. Chiuchiù, G. Carlotti, L. Gammaitoni, Fundamental energy limits in the physics of nanomagnetic binary switches, Nano Energy 15 (2015) 313–320.

[219] D. Sander, S.O. Valenzuela, D. Makarov, C.H. Marrows, E.E. Fullerton, P. Fischer, J. McCord, P. Vavassori, S. Mangin, P. Pirro, B. Hillebrands, A.D. Kent, T. Jungwirth, O. Gutfleisch, C.G. Kim, A. Berger, The 2017 magnetism roadmap, J. Phys. D: Appl. Phys. 50 (36) (2017) 363001.

[220] S. Schlauderer, C. Lange, S. Baierl, T. Ebnet, D.C.V.C.P. Schmid, A.K. Zvezdin, A.V. Kimel, R.V. Mikhaylovskiy, R. Huber, Temporal and spectral fingerprints of ultrafast all-coherent spin switching, Nature 569 (2019) 383–387.

[221] S. de Jong, R. Kukreja, C. Trabant, N. Pontius, C.F. Chang, T. Kachel, M. Beye, F. Sorgenfrei, C.H. Back, B. Bräuer, W.F. Schlotter, J.J. Turner, O. Krupin, M. Doehler, D. Zhu, M.A. Hossain, A.O. Scherz, D. Fausti, F. Novelli, M. Esposito, W.S. Lee, Y.D. Chuang, D.H. Lu, R.G. Moore, M. Yi, M. Trigo, P. Kirchmann, L. Pathey, M.S. Golden, M. Buchholz, P. Metcalf, F. Parmigiani, W. Wurth, A. Föhlisch, H.A. Schüßler-Langeheine, Speed limit of the insulator–metal transition inmagnetite, Nature Mater. 12 (2013) 882, http://dx.doi.org/10.1038/nmat3718.

[222] N.A. Hill, Why are there so few magnetic Ferroelectrics?, J. Phys. Chem. B 104 (2000) 6694, http://dx.doi.org/10.1021/jp000114x.

[223] T. Kimura, T. Goto, H. Shintani, K. Ishizaka, T. Arima, Y. Tokura, Magnetic control of ferroelectric polarization, Nature 426 (2003) 55, http://dx.doi.org/10.1038/nature02018.

[224] M. Mochizuki, N. Nagaosa, Theoretically predicted picosecond optical switching of spin chirality in multiferroics, Phys. Rev. Lett. 105 (14) (2010) 147202.

[225] T. Kubacka, J.A. Johnson, M.C. Hoffmann, C. Vicario, S. De Jong, P. Beaud, S. Grübel, S.-W. Huang, L. Huber, L. Patthey, et al., Large-amplitude spin dynamics driven by a THz pulse in resonance with an electromagnon, Science 343 (6177) (2014) 1333–1336.

[226] A. Pimenov, A.A. Mukhin, V.Y. Ivanov, V.D. Travkin, A.M. Balbashov, A. Loidl, Possible evidence for electromagnons in multiferroic manganites, Nature Phys. 2 (2006) 97, http://dx.doi.org/10.1038/nphys212.

[227] J.A. Johnson, T. Kubacka, M.C. Hoffmann, C. Vicario, S. de Jong, P. Beaud, S. Grübel, S.-W. Huang, L. Huber, Y.W. Windsor, E.M. Bothschafter, L. Rettig, M. Ramakrishnan, A. Alberca, L. Patthey, Y.-D. Chuang, J.J. Turner, G.L. Dakovski, W.-S. Lee, M.P. Minitti, W. Schlotter, R.G. Moore, C.P. Hauri, S.M. Koohpayeh, V. Scagnoli, G. Ingold, S.L. Johnson, U. Staub, Magnetic order dynamics in optically excited multiferroic $TbMnO_3$, Phys. Rev. B 92 (2015) 184429, http://dx.doi.org/10.1103/PhysRevB.92.184429, URL https://link.aps.org/doi/10.1103/PhysRevB.92.184429.

[228] M. Sato, S. Takayoshi, T. Oka, Laser-driven multiferroics and ultrafast spin current generation, Phys. Rev. Lett. 117 (2016) 147202, http://dx.doi.org/10.1103/PhysRevLett.117.147202, URL https://link.aps.org/doi/10.1103/PhysRevLett.117.147202.

[229] J.M. Losada, A. Brataas, A. Qaiumzadeh, Ultrafast control of spin interactions in honeycomb antiferromagnetic insulators, Phys. Rev. B 100 (2019) 060410, http://dx.doi.org/10.1103/PhysRevB.100.060410, URL https://link.aps.org/doi/10.1103/PhysRevB.100.060410.

[230] Y. Wang, J. Hu, Y. Lin, C.-W. Nan, Multiferroic magnetoelectric composite nanostructures, NPG Asia Mater. 2 (2010) 61–68, http://dx.doi.org/10.1038/asiamat.2010.32.

[231] C.-W. Nan, M.I. Bichurin, S. Dong, D. Viehland, G. Srinivasan, Multiferroic magnetoelectric composites: Historical perspective, status, and future directions, J. Appl. Phys. 103 (3) (2008) 031101, http://dx.doi.org/10.1063/1.2836410.

[232] Z. Chu, M.J.P. Asl, S. Dong, Review of multi-layered magnetoelectric composite materials and devices applications, J. Phys. D: Appl. Phys. 51 (24) (2018) 243001, URL http://stacks.iop.org/0022-3727/51/i=24/a=243001.

[233] T.H.E. Lahtinen, J.O. Tuomi, S. van Dijken, Pattern transfer and electric-field-induced magnetic domain formation in multiferroic heterostructures, Adv. Mater. 23 (28) (2013) 3187–3191, http://dx.doi.org/10.1002/adma.201100426, URL https://onlinelibrary.wiley.com/doi/abs/10.1002/adma.201100426.

[234] R. Lo Conte, J. Gorchon, A. Mougin, C.H.A. Lambert, A. El-Ghazaly, A. Scholl, S. Salahuddin, J. Bokor, Electrically controlled switching of the magnetization state in multiferroic $BaTiO_3$/CoFe submicrometer structures, Phys. Rev. Mater. 2 (2018) 091402(R), http://dx.doi.org/10.1103/PhysRevMaterials.2.091402.

[235] H.-J. Liu, L.-Y. Chen, Q. He, C.-W. Liang, Y.-Z. Chen, Y.-S. Chien, Y.-H. Hsieh, S.-J. Lin, E. Arenholz, C.-W. Luo, Y.-L. Chueh, Y.-C. Chen, Y.-H. Chu, Epitaxial photostriction–magnetostriction coupled self-assembled nanostructures, ACS Nano 6 (8) (2012) 6952–6959, http://dx.doi.org/10.1021/nn301976p.

[236] P.J.S. van Capel, D. Turchinovich, H.P. Porte, S. Lahmann, U. Rossow, A. Hangleiter, J.I. Dijkhuis, Correlated terahertz acoustic and electromagnetic emission in dynamically screened InGaN/GaN quantum wells, Phys. Rev. B 84 (2011) 085317, http://dx.doi.org/10.1103/PhysRevB.84.085317, URL https://link.aps.org/doi/10.1103/PhysRevB.84.085317.
[237] M. Lejman, G. Vaudel, I.C. Infante, P. Gemeiner, V.E. Gusev, B. Dkhil, P. Ruello, Giant ultrafast photo-induced shear strain in ferroelectric $BiFeO_3$, Nature Commun. 5 (2014) 4301.
[238] D. Schick, M. Herzog, H. Wen, P. Chen, C. Adamo, P. Gaal, D.G. Schlom, P.G. Evans, Y. Li, M. Bargheer, Localized excited charge carriers generate ultrafast inhomogeneous strain in the multiferroic $BiFeO_3$, Phys. Rev. Lett. 112 (2014) 097602, http://dx.doi.org/10.1103/PhysRevLett.112.097602, URL https://link.aps.org/doi/10.1103/PhysRevLett.112.097602.
[239] C. Jia, N. Zhang, A. Sukhov, J. Berakdar, Ultrafast transient dynamics in composite multiferroics, New J. Phys. 18 (2) (2016) 023002, http://dx.doi.org/10.1088/1367-2630/18/2/023002.
[240] Y.M. Sheu, S.A. Trugman, L. Yan, Q.X. Jia, A.J. Taylor, R.P. Prasankumar, Using ultrashort optical pulses to couple ferroelectric and ferromagnetic order in an oxide heterostructure, Sci. Rep. 5 (2014) 5832.
[241] A.H. Reid, X. Shen, P. Maldonado, T. Chase, E. Jal, P.W. Granitzka, K. Carva, R.K. Li, J. Li, L. Wu, T. Vecchione, T. Liu, Z. Chen, D.J. Higley, N. Hartmann, R. Coffee, J. Wu, G.L. Dakovski, W.F. Schlotter, H. Ohldag, Y.K. Takahashi, V. Mehta, O. Hellwig, A. Fry, Y. Zhu, J. Cao, E.E. Fullerton, J. Stöhr, P.M. Oppeneer, X.J. Wang, H.A. Dürr, Beyond a phenomenological description of magnetostriction, Nature Commun. 9 (2018) 388, http://dx.doi.org/10.1038/s41467-017-02730-7.
[242] C. Dornes, Y. Acremann, M. Savoini, M. Kubli, M.J. Neugebauer, E. Abreu, L. Huber, G. Lantz, C.A.F. Vaz, H. Lemke, E.M. Bothschafter, M. Porer, V. Esposito, L. Rettig, M. Buzzi, A. Alberca, Y.W. Windsor, P. Beaud, U. Staub, D. Zhu, S. Song, J.M. Glownia, S.L. Johnson, The ultrafast Einstein–de Haas effect, Nature 565 (2019) 209, http://dx.doi.org/10.1038/s41586-018-0822-7.
[243] K. Vahaplar, A.M. Kalashnikova, A.V. Kimel, D. Hinzke, U. Nowak, R. Chantrell, A. Tsukamoto, A. Itoh, A. Kirilyuk, T. Rasing, Ultrafast path for optical magnetization reversal via a strongly nonequilibrium state, Phys. Rev. Lett. 103 (2009) 117201, http://dx.doi.org/10.1103/PhysRevLett.103.117201, URL https://link.aps.org/doi/10.1103/PhysRevLett.103.117201.
[244] A.V. Kimel, M. Li, Writing magnetic memory with ultrashort light pulses, Nature Mater. Rev. 4 (2019) 189, http://dx.doi.org/10.1038/s41578-019-0086-3.
[245] R. Landauer, Irreversibility and heat generation in the computing process, IBM J. Res. Dev. 5 (3) (1961) 183–191, http://dx.doi.org/10.1147/rd.53.0183.
[246] P. Mühlschlegel, H.-J. Eisler, O.J.F. Martin, B. Hecht, D.W. Pohl, Resonant optical antennas, Science 308 (5728) (2005) 1607–1609, http://dx.doi.org/10.1126/science.1111886, arXiv:https://science.sciencemag.org/content/308/5728/1607.full.pdf. URL https://science.sciencemag.org/content/308/5728/1607.
[247] M.A. Seo, H.R. Park, S.M. Koo, D.J. Park, J.H. Kang, O.K. Suwal, S.S. Choi, P.C.M. Planken, G.S. Park, N.K. Park, Q.H. Park, D.S. Kim, Terahertz field enhancement by a metallic nano slit operating beyond the skin-depth limit, Nature Photon. 3 (2009) 152, http://dx.doi.org/10.1038/nphoton.2009.22.
[248] L. Novotny, B. Hecht, Principles of Nano-Optics, Cambridge University Press, 2012.
[249] J.D. Joannopoulos, R. Meade, J. Winn, Photonic Crystals, Princeton University Press, Princeton NJ, 1995.
[250] M. Inoue, R. Fujikawa, A. Baryshev, A. Khanikaev, P.B. Lim, H. Uchida, O. Aktsipetrov, A. Fedyanin, T. Murzina, A. Granovsky, Magnetophotonic crystals, J. Phys. D: Appl. Phys. 39 (8) (2006) R151–R161, http://dx.doi.org/10.1088/0022-3727/39/8/r01.
[251] M. Kozina, M. Pancaldi, C. Bernhard, T. van Driel, J.M. Glownia, P. Marsik, M. Radovic, C.A.F. Vaz, D. Zhu, S. Bonetti, U. Staub, M.C. Hoffmann, Local terahertz field enhancement for time-resolved X-ray diffraction, Appl. Phys. Lett. 110 (8) (2017) 081106, http://dx.doi.org/10.1063/1.4977088, arXiv:https://doi.org/10.1063/1.4977088.
[252] M.A. Kozhaev, A.I. Chernov, D.A. Sylgacheva, A.N. Shaposhnikov, A.R. Prokopov, V.N. Berzhansky, A.K. Zvezdin, V.I. Belotelov, Giant peak of the inverse Faraday effect in the band gap of magnetophotonic microcavity, Sci. Rep. 8 (2018) 11435, http://dx.doi.org/10.1038/s41598-018-29294-w.
[253] T.-M. Liu, T. Wang, A.H. Reid, M. Savoini, X. Wu, B. Koene, P. Granitzka, C.E. Graves, D.J. Higley, Z. Chen, et al., Nanoscale confinement of all-optical magnetic switching in TbFeCo-competition with nanoscale heterogeneity, Nano Lett. 15 (10) (2015) 6862–6868.
[254] B. Zhang, Y. Xu, W. Zhao, D. Zhu, X. Lin, M. Hehn, G. Malinowski, D. Ravelosona, S. Mangin, Energy-efficient domain-wall motion governed by the interplay of helicity-dependent optical effect and spin-orbit torque, Phys. Rev. Appl. 11 (2019) 034001, http://dx.doi.org/10.1103/PhysRevApplied.11.034001, URL https://link.aps.org/doi/10.1103/PhysRevApplied.11.034001.
[255] B. Zhang, Y. Xu, W. Zhao, D. Zhu, H. Yang, X. Lin, M. Hehn, G. Malinowski, N. Vernier, D. Ravelosona, S. Mangin, Domain-wall motion induced by spin transfer torque delivered by helicity-dependent femtosecond laser, Phys. Rev. B 99 (2019) 144402, http://dx.doi.org/10.1103/PhysRevB.99.144402, URL https://link.aps.org/doi/10.1103/PhysRevB.99.144402.