Nonlinear optical effects due to magnetization dynamics in a ferromagnet

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(Received 24 October 2023; revised 12 December 2023; accepted 13 December 2023; published 10 January 2024)

We theoretically consider magnetization dynamics in a ferromagnetic slab induced by the magnetic field of a strong femtosecond laser pulse. The longitudinal geometry, in which the initial magnetization lies in both the plane of incidence and the sample plane, is studied. The magnetization oscillations at the optical wave frequency are calculated with the use of the Kapitza pendulum approach taking into account that the optical frequency is much greater than the magnetization oscillation eigenfrequency. We study the reflection of the electromagnetic wave from a ferromagnet and show that this laser-induced low-frequency magnetization dynamics leads to the appearance of the second-order nonlinearity in the Maxwell's equations, which in turn gives rise to both the second harmonic generation (SHG) and rectification effect. Although the amplitude of the magnetization oscillations is small, the considered effect may be responsible for the SHG with the efficiency comparable to that of nonmagnetic SHG from metal surfaces. Our estimations show that the suggested mechanism may explain the recent experiments on magnetization induced modulation of the SHG intensity in a "forbidden" $P_{\text{in}}P_{\text{out}}$ combination of incident and reflected waves in cobalt/heavy metal systems, where it can be even more pronounced due to the spin current flow through the ferromagnet/heavy metal interface.

DOI: 10.1103/PhysRevB.109.024411

I. INTRODUCTION

Nonlinear optical effects such as second harmonic generation (SHG) or rectification attract a lot of attention for the last decades. On one hand, this is governed by the fact that these effects may exist only in noncentrosymmetric systems. As a result, these nonlinear optical phenomena, and first of all the SHG probe, provide a powerful method for studying the properties of surfaces and interfaces where the inversion symmetry is broken [1,2]. On the other hand, the rectification effect is an efficient mechanism for the THz wave generation under the excitation by femtosecond laser pulses [3–6]. A special research direction here is the magnetization-induced phenomena in systems containing magnetic materials. For instance, THz sources based on ferromagnet/heavy metal multilayers are widely studied [7,8]. Magnetization brings new symmetry properties to a medium [9], which in turn leads to the appearance of the nonlinear-optical analogues of the magnetooptical Kerr and Faraday effect, and even to a number of new ones. Among others, recently the so named "forbidden" magnetization-induced SHG intensity effect was observed in ferromagnet/heavy metal systems such as Co/Pt, Co/Ta, etc., multilayers, which consists in variation of the *p*-polarized SHG intensity by longitudinal dc magnetic field [10–12].

Symmetry analysis of the nonlinear-optical interactions [9] does not take into account the effects of (expected) magnetization dynamics induced by the electromagnetic wave. They are usually supposed to be small as the optical frequency exceeds substantially the eigenfrequency of magnetization oscillations. However, if a strong femtosecond optical pulse is considered with the electric field of the order of 1 MV/cm or greater, the frequency ratio is of the order of 10^{-4}, while the magnetic field of the optical wave is relatively strong. Therefore, one can expect that the effects that appear due to magnetization dynamics may be comparable to those provided by static magnetization due nonlinearity at a ferromagnet surface.

In this work we study these effects and compare the theoretical results with the data of recent experiments. The magnetization dynamics caused by the magnetic field of the incident laser radiation is described in the framework of the Landau-Lifshitz-Gilbert equation solved within the Kapitza pendulum approach. The Maxwell equations are then solved with the assumptions of small gyrotropic terms of the dielectric constant and small magnetization oscillation magnitude. We obtain both double frequency electric field and zero-frequency (rectified) electric field, or the electric current in a ferromagnet induced by the electromagnetic wave. Finally, a boundary problem is solved. We suppose that the electromagnetic wave is incident at the surface of a ferromagnet magnetized in the longitudinal geometry. The estimations show that the suggested mechanism may explain the recent experiments [10–12]: strong effects reported for a cobalt/heavy

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metal system may be explained by enhanced dissipation due to spin current from a ferromagnet to heavy metal [13,14]. THz generation via such a magnetization dynamic is also discussed.

II. THEORETICAL APPROACH

We start with the Landau-Lifshitz-Gilbert equation for a uniform magnetic medium placed in an alternating magnetic field:

$$\frac{d\mathbf{M}}{dt} = -\frac{\omega_M}{M_s}[\mathbf{M}\times\mathbf{H}+\mathbf{h}'] + \frac{\alpha\omega_M}{M_s^2}[\mathbf{M}\times[\mathbf{M}\times\mathbf{H}+\mathbf{h}'], \tag{1}$$

where $\mathbf{M}$ is the magnetization, M_s its saturation value, $\mathbf{H}$ is the external magnetic field, $\omega_M = \gamma M_s$ is the magnetization oscillation characteristic frequency, γ is the gyromagnetic ratio, α is the dimensionless Gilbert damping constant of the considered medium, and $\mathbf{h}' = \mathbf{h}'_0 \cos\omega t$ is the alternating magnetic field of the optical wave inside the medium. We then take into account that the optical frequency is much larger than the magnetic system eigenfrequency, $\omega \gg \omega_M$. This assumption is valid as the ratio ω_M/ω is typically 10^{-4} or less. Then the equation (1) can be solved with the use of the Kapitza pendulum method [15]. The general approach to this problem is described below.

Let us consider a system of differential equations for an arbitrary number of coordinates A_i with a rapidly oscillating external source:

$$\frac{dA_i}{dt} = f_i(\mathbf{A}) + g_i(\mathbf{A})\cos\omega t + h_i(\mathbf{A})\sin\omega t, \tag{2}$$

where we take into account the arbitrary phase of the source in the right-hand part of the equation by introducing the two sets of real functions g_i and h_i, and $\mathbf{A}$ is the coordinate vector. One may determine the eigenfrequencies of the system (2) as

$$\Omega_{f\,ij} = \frac{\partial f_i}{\partial A_j}, \quad \Omega_{g\,ij} = \frac{\partial g_i}{\partial A_j}, \quad \Omega_{h\,ij} = \frac{\partial h_i}{\partial A_j}, \tag{3}$$

and suppose that they all are much smaller than that of the external source, i.e. $\Omega_{f,g,h\,ij} \ll \omega$. We also suppose that $\Omega_{f\,ij} \sim \Omega_{g\,ij} \sim \Omega_{h\,ij} \sim \Omega$, where Ω is the characteristic frequency of system motion. This is governed by the fact that the oscillation of the external source, which is explicitly written in (2) in terms of g_i and h_i, is much faster than all the characteristic times of the system (including the time at which the magnitude of the source changes). Such a supposition corresponds to a wide range of physical systems, including the system under consideration (1) in which an electromagnetic wave of optical frequency acts on magnetization of a ferromagnet.

Then we may seek the solution of the system (2) in the form

$$\mathbf{A} = \mathbf{U} + \mathbf{a}, \tag{4}$$

supposing that $\mathbf{U}$ is a "slow" part of the solution with the typical frequency Ω, and $\mathbf{a}$ is the "fast"oscillating part with the characteristic frequency ω. It is then straightforward to split the functions f_i, g_i, h_i into the series

$$f_i(\mathbf{A}) \approx f_i(\mathbf{U}) + \sum_j \left.\frac{\partial f_i}{\partial A_j}\right|_{\mathbf{U}} a_j + \frac{1}{2}\sum_{jk} \left.\frac{\partial^2 f_i}{\partial A_j \partial A_k}\right|_{\mathbf{U}} a_j a_k, \tag{5}$$

$$g_i(\mathbf{A}) \approx g_i(\mathbf{U}) + \sum_j \left.\frac{\partial g_i}{\partial A_j}\right|_{\mathbf{U}} a_j, \tag{6}$$

$$h_i(\mathbf{A}) \approx h_i(\mathbf{U}) + \sum_j \left.\frac{\partial h_i}{\partial A_j}\right|_{\mathbf{U}} a_j. \tag{7}$$

Here we provide terms up to $(\Omega/\omega)^2$ for a general solution (see below). However, we need only terms linear in Ω/ω to solve (1) in the framework of the current paper.

By substituting (5)–(7) into (2) and averaging over a small time period corresponding to the frequency ω, one may obtain the equation for the "slow" part $\mathbf{U}$:

$$\begin{aligned}\dot{U}_i = f_i &- \frac{1}{2\omega}\sum_j \left(\frac{\partial g_i}{\partial A_j} h_j - \frac{\partial h_i}{\partial A_j} g_j\right) \\ &- \frac{1}{2\omega^2}\sum_{jk}\left(\frac{\partial g_i}{\partial A_j}\frac{\partial f_i}{\partial A_k} g_k + \frac{\partial h_i}{\partial A_j}\frac{\partial f_i}{\partial A_k} h_k \right.\\ &\left. - \frac{1}{2}\frac{\partial^2 f_i}{\partial A_j \partial A_k}(g_j g_k + h_j h_k)\right),\end{aligned} \tag{8}$$

where we restrict ourselves by the second order in Ω/ω and consider the functions f_i, g_i, h_i, and their derivatives are taken at the $\mathbf{U}$ coordinate. Note that we suppose that the second derivative of f_i gives a term proportional to Ω^2, e.g., $\frac{\partial^2 f_i}{\partial A_j \partial A_k} g_j \propto \Omega^2$. It is obvious from (8) that an arbitrary shift of the phase of the oscillating source would lead to change of g_i and h_i while keeping $\mathbf{U}$ intact. The equation of motion for a classic pendulum with vibrating suspension [15] is obtained from the third term of the right-hand part of (8) ($\propto 1/\omega^2$).

Usually Eq. (8) is then used to calculate the dynamics of the system averaged over "fast" oscillations of the source at the coordinate vector $\mathbf{U}$. This is done for the dynamics of the magnetized medium in [16,17] and recently in [18,19]. However in order to find the sources of the first and second harmonics of the fast oscillating terms at the ω frequency, we need to consider the "fast" part of the solution. This can be made by using the perturbation theory after substituting Eqs. (5)–(7) into (2) and taking into account Eq. (8). The "fast" part oscillating at the source frequency ω is then integrated in the form

$$\begin{aligned}a_i^\omega = &\left(\frac{g_i}{\omega}\sin\omega t - \frac{h_i}{\omega}\cos\omega t\right) \\ &- \left(\sum_j \frac{\partial f_i}{\partial A_j}\frac{h_j}{\omega^2}\sin\omega t + \sum_j \frac{\partial f_i}{\partial A_j}\frac{g_j}{\omega^2}\cos\omega t\right),\end{aligned} \tag{9}$$

where we suppose again that the functions f_i, g_i, h_i and their derivatives are taken at the $\mathbf{U}$ point. It is clear from (9) that the expression in the second bracket in the right-hand part is smaller than the first one as Ω/ω; we neglect all smaller

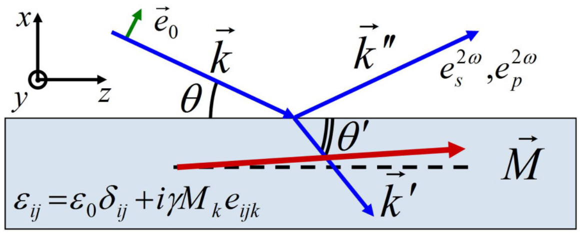

FIG. 1. Geometry of the system under consideration. A p-polarized optical wave is incident at the surface of a ferromagnet magnetized in the longitudinal geometry. Oscillations of magnetization are schematically shown as a shift of $\mathbf{M}$ from the initial z direction.

terms in the solution. We may substitute the solution (9) into series (5)–(7) in order to obtain the condition of applicability of this series expansion. Substituting the first bracket of the right-hand part of (9) gives the term $\sim\frac{\Omega}{\omega}g_j$, $\sim\frac{\Omega}{\omega}h_j$ in the first order of the Taylor series, and the term $\sim(\frac{\Omega}{\omega})^2 g_j$, $\sim(\frac{\Omega}{\omega})^2 h_j$ in the second order of the series, etc. Accordingly, substituting the second bracket of the right-hand part of (9) gives a term proportional to square of the ratio of frequencies in the first order of the Taylor series already. Taking for simplicity that f_i, g_i, and h_i are of the same order of value, we get the expected result that the small parameter for the series expansion of (5)–(7) is Ω/ω.

One may also find the "fast" part of the solution oscillating at the double frequency 2ω. In the lowest order of the perturbation theory it takes the form

$$a_i^{2\omega} = \frac{1}{4\omega^2}\left(\sum_j \left(\frac{\partial h_i}{\partial A_j}h_j - \frac{\partial g_i}{\partial A_j}g_j\right)\cos 2\omega t - \sum_j \left(\frac{\partial g_i}{\partial A_j}h_j + \frac{\partial h_i}{\partial A_j}g_j\right)\sin 2\omega t\right), \tag{10}$$

which is proportional to $(1/\omega)^2$.

We can now apply the general solution described above to the Landau-Lifshitz-Gilbert equation (1). In order to do this, we take into account that the magnetization vector can be written through the two angles, φ and β, with the amplitude $|\mathbf{M}| = M_s$, as

$$\mathbf{M} = M_s(\cos\varphi \sin\beta, \sin\varphi \sin\beta, \cos\beta) \tag{11}$$

in Cartesian coordinate system, as shown in Fig. 1. According to (9), the part of magnetization oscillating at the frequency ω has a term linear in $\Omega/\omega \equiv \omega_M/\omega$. If one takes the Cartesian coordinate system in such a way that the equilibrium magnetization is parallel to the z axis and the magnetic field of the wave is parallel to the y axis ($\mathbf{h}' = -e_0'\mathbf{e}_y \cos\omega t$, where e_0' is the wave electric field magnitude, $\mathbf{e}_y$ is the unit vector in the y direction; see Fig. 1), the magnetization has the following form:

$$\mathbf{M} = \mathbf{M}_0 + \mathbf{m} = \left(\frac{\omega_M}{\omega}h_0' \sin(\omega t - \mathbf{k}'\mathbf{r}), -\alpha\frac{\omega_M}{\omega}h_0' \sin(\omega t - \mathbf{k}'\mathbf{r}), M_s\right) \tag{12}$$

up to the first order in ω_M/ω, where $\mathbf{M}_0$ is the static and $\mathbf{m}$ is the oscillating part of magnetization. This oscillating part $\mathbf{m}$ of magnetization gives rise to the second harmonic generation, as we show below.

The Maxwell's equations are solved when considering the magnetization oscillations as a perturbation. We write the dielectric permittivity of the medium in the usual form:

$$\varepsilon_{ij} = \varepsilon_0\delta_{ij} + i\gamma M_k e_{ijk}, \tag{13}$$

where δ_{ij}, e_{ijk} are the Kronecker delta and the antisymmetric Levi-Civita tensor, respectively. The real unperturbed electric field $\mathbf{e}'$ is found by solving the Maxwell's equations with the magnetization $\mathbf{M}_0$. After that, the linear in $\mathbf{m}$ correction $\delta\mathbf{e}'$ is found as a solution of the equation

$$\nabla \times (\nabla \times \delta\mathbf{e}') + \frac{\varepsilon_0}{c^2}\delta\ddot{\mathbf{e}}' = -i\frac{\gamma}{c^2}(\ddot{\mathbf{e}}' \times \mathbf{m} + 2\dot{\mathbf{e}}' \times \dot{\mathbf{m}} + \mathbf{e}' \times \ddot{\mathbf{m}}), \tag{14}$$

which follows from the Maxwell's equations in the linear order in the gyrotropic component γ of the dielectric permittivity, c being the light velocity. The right-hand part of the equation (14) acts as a source of the electric field $\delta\mathbf{e}'$ and is proportional to the square of unperturbed field in accordance with (12), therefore it leads to the generation of the second-harmonic field. Note that Eq. (14) is written for the second derivatives of $\delta\mathbf{e}'$, hence it does not describe the rectification effect (or the zero-frequency field). This problem is discussed below.

Let us consider the p-polarized electromagnetic wave with the electric field $\mathbf{e} = \mathbf{e_0}\cos(\omega t - \mathbf{kr})$, $\mathbf{e}_0 = \mathbf{e}_x\cos\theta + \mathbf{e}_z\sin\theta$ incident at the surface of a ferromagnet as shown in Fig. 1. Here θ is the incident sliding angle, $\mathbf{k} = k(\cos\theta\mathbf{e}_z - \sin\theta\mathbf{e}_x)$ is the wave vector, and $\mathbf{e}_i$ are the unit vectors of the Cartesian coordinate system. There are two eigenmodes inside the medium, which have a different refractive index and structure [20] (approximate electric field structure for the modes is written out below). Since these modes have elliptical polarization in the general case, both of them are excited by the p-polarized incident wave. The wave vectors of these modes inside a medium are determined from the boundary conditions at the magnetic interface as

$$k'_\pm \approx k_0\sqrt{\varepsilon_0}\left(1 \pm \frac{\gamma M_s}{2\varepsilon_0^{3/2}}\cos\theta\right), \tag{15}$$

where $k_0 = \frac{\omega}{c}$. Corresponding sliding angles $\theta'_\pm$ inside the magnetic medium are equal to

$$\cos\theta'_\pm \approx \frac{\cos\theta}{\sqrt{\varepsilon_0}}\left(1 \mp \frac{\gamma M_s}{2\varepsilon_0^{3/2}}\cos\theta\right). \tag{16}$$

The Cartesian components of the electric field of the optical wave inside the medium may also be easily found by satisfying the boundary conditions of continuity of the tangential component of the electric field strength vector, the normal component of the electric field induction vector, and the tangential component of the magnetic field strength vector:

$$e'_{x+} = e'_{x-} = 2e_0 \frac{\cos(\omega t - \mathbf{k}'\mathbf{r}) \sin\theta \cos\theta}{\varepsilon_0 \sin\theta + \sqrt{\varepsilon_0 - \cos^2\theta}}, \tag{17}$$

$$e'_{y\pm} = 2e_0 \sin(\omega t - \mathbf{k}'\mathbf{r}) \frac{\pm\sqrt{\varepsilon_0}\sin\theta + \frac{\gamma M_s}{2}\tan\theta}{\varepsilon_0 \sin\theta + \sqrt{\varepsilon_0 - \cos^2\theta}}, \tag{18}$$

$$e'_{z\pm} = 2e_0 \cos(\omega t - \mathbf{k}'\mathbf{r}) \times \frac{\sqrt{\varepsilon_0 - \cos^2\theta}\sin\theta \pm \frac{\gamma M_s}{2\sqrt{\varepsilon_0}} \frac{2\varepsilon_0 - \cos^2\theta}{\sqrt{\varepsilon_0 - \cos^2\theta}} \tan\theta}{\varepsilon_0 \sin\theta + \sqrt{\varepsilon_0 - \cos^2\theta}}. \tag{19}$$

Note that this solution is an approximate one and is applicable only when the terms proportional to γM_s are small. Solution (12) contains the magnitude of the unperturbed magnetic field wave inside the medium, which is determined from the boundary conditions as

$$h'_0 = e_0 \frac{4\varepsilon_0 \sin\theta}{\varepsilon_0 \sin\theta + \sqrt{\varepsilon_0 - \cos^2\theta}}. \tag{20}$$

Substituting (17)–(19) and (12) into Eq. (14), we then solve this equation and finally find the double-frequency electric field outside the medium from the boundary conditions. The s and p component of its magnitude have the form

$$e_s^{2\omega} = \frac{e_0^2}{M_s} \frac{\omega_M}{\omega} \frac{\gamma M_s}{2} \frac{\varepsilon_0 \sqrt{\varepsilon_0 - \cos^2\theta}\tan^2\theta}{(\varepsilon_0 \sin\theta + \sqrt{\varepsilon_0 - \cos^2\theta})^2}, \tag{21}$$

$$e_p^{2\omega} = \alpha \frac{e_0^2}{M_s} \frac{\omega_M}{\omega} \frac{\gamma M_s}{2} \frac{(\varepsilon_0 - \cos^2\theta)\sin\theta\cos\theta}{\varepsilon_0(\varepsilon_0 \sin\theta + \sqrt{\varepsilon_0 - \cos^2\theta})^2}. \tag{22}$$

Second harmonic field determined by its p and s components, (21) and (22), appears due to oscillations of magnetization of the ferromagnet in the magnetic field of the light wave. This is the main result of current paper; it is discussed in Sec. III.

As we have mentioned above, the static (zero-frequency) electric field is not described by Eq. (14). However, second-order nonlinear optical effects such as SHG and rectification typically coexist. In order to show that the rectified signal appears in our case as well, we provide a simple model in which the electron motion is described by Newton's law. This method is very similar to one used by Gaponov and Miller in order to calculate the ponderomotive force that acts on a charged particle in an electromagnetic field of high frequency [21]. We suppose that "free" conduction electrons are in charge of the optical response of the system. Their motion is described as

$$\ddot{\mathbf{r}} = -\frac{e}{m_e}\mathbf{e}' - \frac{\lambda}{m_e}\dot{\mathbf{r}} \times \mathbf{M}(t), \tag{23}$$

where $\mathbf{r}$ is the electron coordinate, e is its absolute charge, m_e is its mass, and λ is the constant of Lorentz-like force induced by the magnetization, which leads, e.g., to anomalous Hall effect and has spin-orbit roots [22]. Supposing that λ is small, we first solve the equation (23) neglecting the Lorentz-like force. At the next step we substitute the obtained solution into this force in order and find the corresponding correction to $\mathbf{r}(t)$. Averaging this force over the time period of wave with (12) gives an effective electric field that acts on the electrons:

$$\mathbf{E}_{\text{eff}} = -\gamma \frac{\omega}{\omega_p^2} \langle \dot{\mathbf{e}}' \times \mathbf{m} \rangle_t, \tag{24}$$

TABLE I. Contributions to SHG appearing in different combinations of the polarizations of the exciting and SHG light. "+" or "−" represent existence or absence of SHG light, respectively. SHG discovered in current paper is marked after slash where applicable.

Polarization	M_x	M_y	M_z	Nonmagnetic
$P_{\text{in}}P_{\text{out}}$	−	+	−/+	+
$P_{\text{in}}S_{\text{out}}$	+	−	+/+	−
$S_{\text{in}}P_{\text{out}}$	−	+	−	+
$S_{\text{in}}S_{\text{out}}$	−	−	+	−

where the Lorentz-like force constant λ is expressed through the medium constant of gyrotropy γ, ω_p is the electron plasma frequency, and $\langle \ldots \rangle_t$ stands for averaging over time. After averaging with $\mathbf{m}$ determined by (12) and $\mathbf{e}'$ defined as (17)–(19), we obtain

$$E_{\text{eff}\,x} = -\alpha E_{\text{eff}\,y} = -\alpha \frac{e_0^2}{M_s} \frac{\omega\omega_M}{\omega_p^2} \gamma M_s \frac{4\varepsilon_0 \sin^2\theta \sqrt{\varepsilon_0 - \cos^2\theta}}{(\varepsilon_0 \sin\theta + \sqrt{\varepsilon_0 - \cos^2\theta})^2}, \tag{25}$$

$$E_{\text{eff}\,z} = -\alpha \frac{e_0^2}{M_s} \frac{\omega\omega_M}{\omega_p^2} \gamma M_s \frac{4\varepsilon_0 \sin^2\theta \cos\theta}{(\varepsilon_0 \sin\theta + \sqrt{\varepsilon_0 - \cos^2\theta})^2}. \tag{26}$$

Thus, we have the rectification effect due to magnetization oscillations inside a medium. This is the second main result of the current paper.

III. RESULTS AND DISCUSSION

The main results of our consideration presented in Sec. II are the equations for the s- and p-polarized components of the electric field of the SHG wave (21), (22) and the rectified field (25), (26) driven by oscillations of the magnetic moment in the medium under the influence of the p-polarized incident wave in the longitudinal geometry. We analyze these equations below.

A. Second harmonic generation

It is known [23] that for the p-polarized light incident at the surface of an isotropic ferromagnet there is only a p-polarized nonmagnetic SHG response, while only the transversal component of magnetization may give rise to the p-polarized magnetic SHG signal (see Table I). There is only s-polarized magnetization-induced SHG for both polar and longitudinal geometries of the experiment. This is governed by the symmetry of the surface of a ferromagnet at which the inversion symmetry is broken, as the SHG polarization $\mathbf{P}^{2\omega}$ can be fully described by the following expression:

$$\mathbf{P}^{2\omega} \propto \mathbf{n}\mathbf{e}^2 + \mathbf{e}(\mathbf{n}\cdot\mathbf{e}) + \mathbf{n}\times\mathbf{e}\,(\mathbf{M}\cdot\mathbf{e}) + \mathbf{M}\times\mathbf{e}\,(\mathbf{n}\cdot\mathbf{e}) + \mathbf{n}\times\mathbf{M}\,\mathbf{e}^2, \tag{27}$$

where $\mathbf{n}$ is the surface normal vector and $\mathbf{e}$ is the magnitude of the electric field of the incident wave. The first two terms in the right-hand part of (27) stand for the nonmagnetic response, while the last three are linear in $\mathbf{M}$. It is then straightforward

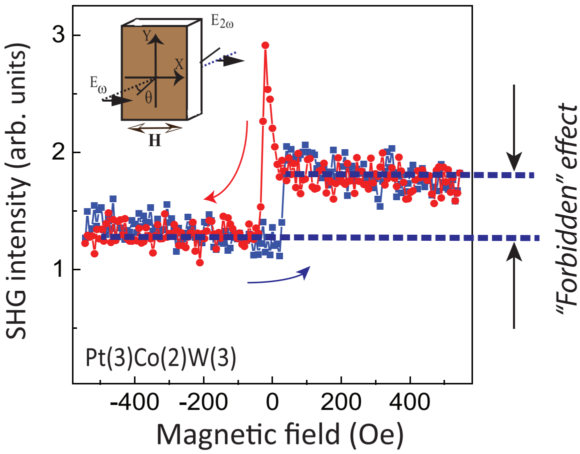

FIG. 2. Dependence of the SHG intensity for the $P_{\text{in}}P_{\text{out}}$ polarizaton combination on longitudinal magnetic field for the sliding angle $\theta = 70^\circ$ for Pt(3 nm)/Co(3 nm)/W(3 nm) film. The picture is taken from [10]. (The Cartesian coordinate x here corresponds to the coordinate z in the current paper.)

to obtain the results summarized in Table I. One can see that the p-polarized SHG is "forbidden" for $M_y = 0$.

The second harmonic generation (21), (22) in this paper is of different nature. It is governed by the oscillation of magnetization of the magnetic field $\mathbf{h}'$ inside the medium together with the nonlinearity of the material equation for the electric induction; it contains the vector product of the electric field $\mathbf{e}'$ and the oscillating magnetization. This mechanism is not related to the break of the inversion symmetry at the interface, instead it utilizes the break of this symmetry by the wave vector $\mathbf{k}' \propto \mathbf{e}' \times \mathbf{h}'$. Therefore it removes the symmetry restriction on magnetization-induced effects in SHG for the $P_{\text{in}}P_{\text{out}}$ combination of polarizations illustrated by Table I. We suppose that the same would apply to the $S_{\text{in}}P_{\text{out}}$ polarization combination.

The "forbidden" effect was earlier observed in [10]; the main result of this experiment is shown in Fig. 2. Here the SHG intensity hysteresis in the longitudinal geometry measured for the $P_{\text{in}}P_{\text{out}}$ polarizations' combination shows a clear difference in the SHG signal for the positive and negative saturating magnetic field. This "forbidden" effect may be explained by the mechanism discussed in the current paper.

This is supported by the following estimations. In the experiment, the pulsed laser radiation at the 820 nm wavelength with 30 fs pulse duration is used. The peak pulse power is about 70 kW and the beam diameter is 30 μm, which gives the intensity of $\sim 10^{10}\,\frac{\text{W}}{\text{cm}^2}$ and the electric field of $2.7 \cdot 10^6\,\frac{\text{V}}{\text{cm}}$. As the saturation magnetization of cobalt is 1400 G, we get the ratio $^{h_0}/_{M_s} \approx 6.5$. We can estimate the gyrotropic term of the dielectric permittivity from the MOKE polarization rotation angle, which is about $5 \cdot 10^{-3}$ rad for Co films. By taking the approximate value $\varepsilon_0 \sim 10$ by the order of value, we obtain $\gamma M_s \approx 0.05$, which is a small parameter indeed.

The frequency ratio, which is the main small parameter that determines the magnetization oscillation magnitude, is $^{\omega_M}/_{\omega} \approx 0.6 \cdot 10^{-4}$, and the Gilbert damping constant is approximately $\alpha \sim 0.1$ by the order of value for a Co/Pt system (see below). Using Eq. (22) and neglecting the angular dependence, we arrive at the estimation $e_p^{2\omega} \approx 10^{-8} e_0$. Typical SHG efficiency for a ferromagnetic surface is $I_s^{2\omega} \approx 10^{-14} I^\omega$, hence the electric field $e^{2\omega} \approx 10^{-7} e_0$ [2]. Thus, the interference of the discovered magnetic SHG with the nonmagnetic SHG response from the surface gives $I^{2\omega} \approx 10^{-15} I^\omega$, which is only an order of magnitude smaller than that for the nonmagnetic signal: $^{I^{2\omega}}/_{I_s^{2\omega}} \approx 0.1$. The experimentally observed "forbidden" effect that can be compared to our estimations is determined as $\frac{I^{2\omega}(+H)-I^{2\omega}(-H)}{I^{2\omega}(+H)+I^{2\omega}(-H)}$ and is approximately 0.17. Thus, the suggested mechanism gives the value of the same order of magnitude as observed in the experiment.

TABLE II. Typical parameters for different ferromagnetic materials and estimations for the "forbidden" $P_{\text{in}}P_{\text{out}}$ SHG effect.

Material	M_s, G	γM_s	α	$^{I^{2\omega}}/_{I_s^{2\omega}}$	Ref.
YIG	200	$5 \cdot 10^{-4}$	$2.3 \cdot 10^{-4}$	$2.3 \cdot 10^{-6}$	[24]
$Ni_{80}Fe_{20}$	800	0.02	0.01	$4 \cdot 10^{-3}$	[25,26]
CoFeB	1200	0.04	0.015	$1.2 \cdot 10^{-2}$	[26]
Co	1400	0.05	0.02	$2 \cdot 10^{-2}$	[27]
Thin Co/Pt	1400	0.05	$0.04 - 0.22$	$0.04 - 0.22$	[13]

According to [10], the observed "forbidden" magnetization-induced SHG intensity effect decreases as the Co layer thickness grows in a Co/Pt or Co/W bilayer film. This is consistent with the fact that the discussed SHG effect is proportional to the Gilbert damping constant α, which is enhanced in an FM/HM system due to the spin current flow at the ferromagnet/heavy metal interface [13]. As this is a surface effect, it decreases as the cobalt thickness grows. Accordingly, the "forbidden" SHG effect in this paper decreases.

Table II summarizes the results of rough estimations of the "forbidden" SHG effect for different materials. One can see that the increase of saturation magnetization and of the Gilbert damping constant leads to the increase of the effect. So the best choice for its observation is a thin Co/Pt multilayer system, which stays in agreement with the mentioned experiments.

The dependences of the calculated SHG fields (21), (22) on the sliding angle θ and the dielectric permittivity ε_0 of the medium are shown in Fig. 3. The field of the s-polarized SHG wave grows as the sliding angle increases. It diverges at $\theta = \frac{\pi}{2}$ when the solution (21) is incorrect for $\tan\theta \to \infty$. The electric field of the p-polarized SHG wave reveals a maximum at the $\theta \approx 15^\circ$ sliding angle. This field has a maximum at quite small dielectric permittivity ($\varepsilon_0 \approx 1.2$) and, contrary to the field of the s-polarized SHG wave, decreases as $^1/_{\varepsilon_0}$ for $\varepsilon_0 \gg 1$.

The most powerful method to experimentally reveal the discussed "forbidden" SHG effect may be based on its dependence on the Gilbert damping constant. It is possible to manipulate this constant, e.g., by varying the magnetic material or its thickness, the material or thickness of a neighboring layer in a ferromagnet/heavy metal system. Also, the properties of a thin polycrystalline ferromagnetic film may be changed by different methods such as irradiating it with ions [28] or fabricating the sample under special conditions [29], or even by adding a very small amount of impurity (for instance,

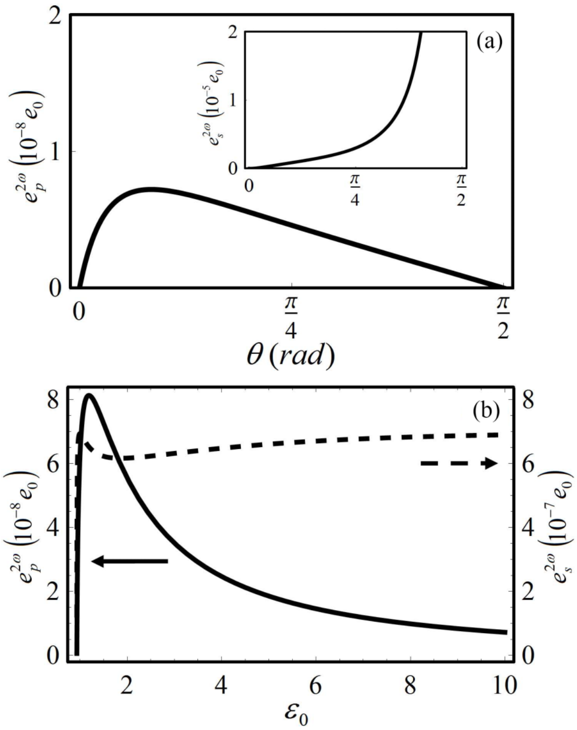

FIG. 3. (a) Dependence of electric field of the "forbidden" p-polarized second-harmonic wave on the sliding angle for $\varepsilon_0 = 10$ (the inset shows the same for the "allowed" s-polarized wave obtained in the current paper). (b) Dependence of electric field of the p-polarized (solid line) and s-polarized (dashed line) second-harmonic wave on dielectric permittivity for $\theta = 15^\circ$. Other parameters are the same as mentioned in the text for thin Co/Pt system.

it is known that the conductivity of nickel is sufficiently changed by adding only 3% of copper [30]). However there is no regular method of precise control of the Gilbert damping constant. Therefore such technological and experimental work requires a lot of investigations and is beyond the scope of the current paper. The angular dependence of the "forbidden" effect may also be checked in an experiment: it should reach its maximum at a certain angle and tend to zero both for sliding and normal incidence of the exciting wave.

B. Rectification effect

The effective rectified field that appears due to magnetization oscillations under the subjection of the magnetic field of the optical wave is determined by (25) and (26). We may estimate this field for realistic parameters of a femtosecond laser pulse described above. The plasma frequency of a metal with the electron concentration of 10^{22} cm^{-3} is $\omega_p = 5.6 \cdot 10^{15} s^{-1}$. The angle function in (25) reaches its maximum at $\theta \to \pi/2$, which corresponds to normal incidence of the light wave. Substituting all parameters into (25) and (26) we obtain $E_{\mathrm{eff}\,y} \approx 10^{-6} e_0 \approx 3.7 \frac{\mathrm{V}}{\mathrm{cm}}$. This effect is relatively weak, while this value can be increased by lowering the plasma frequency, e.g., by taking diluted magnetic semiconductors.

We note that the z component of the effective field is caused by the x component of the light wave. Therefore it should be zero due to screening effects that are not taken into account in (25) and (26). On the other hand, the x component of the effective field would be canceled due to the same screening effect. Therefore the effective field has only the y component, which is larger than the other components since it does not contain the small damping factor α. Taking this into account, from a symmetry point of view the effective field may be written as

$$\mathbf{E}_{\mathrm{eff}} \sim \mathbf{M} \times \mathbf{n}. \tag{28}$$

The electric current caused by this effective field may be determined from Ohm's law $\mathbf{j} = \sigma \mathbf{E}_{\mathrm{eff}}$. For the thickness of 30 nm and the width of the the current flow area equal to the beam diameter of 30 μm, we estimate the constant electric current as $I_e \approx 4$ mA. For the 80 MHz pulse repetition rate this gives the average current $\langle I_e \rangle \approx 1$ nA, which can hardly be detected in real systems. However, this current appears at an electromagnetic wave envelope time which is usually 30–50 ps and thus should emit the THz radiation with the characteristic frequency of 20–33 THz. In real systems, characteristic time of the electric current relaxation is determined by the electron-phonon interaction and is of the order of 100 fs–1 ps. Thus the electromagnetic wave generation is usually restricted to several THz. The polarization of a THz wave is determined by (28), and is the same as the conventional one for the spintronic THz emitters [8]. However, this additional mechanism does not depend on the constant of Gilbert damping and therefore should exist as well for a single ferromagnetic layer. Contrary to the mechanism that provides THz generation in ferromagnetic/nonmagnetic systems, the effect discovered here should give the signal growing with the thickness of the FM layer.

IV. CONCLUSION

In conclusion, we theoretically investigate the nonlinear optical effects that appear due to magnetization oscillations under the influence of the optical wave on a ferromagnet surface. Based on the Kapitza pendulum approach we show that the light-induced magnetization dynamics in a ferromagnet can provide a mechanism for the second harmonic generation. Although the magnitude of the magnetization oscillations is small, laser-induced magnetization dynamics can provide the SHG response comparable to the nonmagnetic one, as well as to the THz generation through the rectification effect.

The SHG effect is often used as a powerful tool for the diagnostics of surfaces and buried interfaces of centrosymmetric media [2]. It is widely applied for studies of surfaces of ferromagnets, which may reveal specific magnetic properties different from those of bulky materials [31]. The SHG mechanism suggested in this paper is supported by the recent observation of the intensity magnetooptical SHG effect under the application of the longitudinal magnetic field for the $P_{\mathrm{in}}P_{\mathrm{out}}$ combination of polarizations of the incident and SHG waves, which is symmetry forbidden in a ferromagnetic medium [10–12]. Therefore this effect expands the number of mechanisms involved in the interaction of light with magnetic materials and should be definitely taken into consideration for the correct interpretation of the second harmonic generation in magnets.

We have shown that such a "forbidden" effect is a consequence of damping of magnetization oscillations in a magnetic system. Therefore it may be identified in the experiment by its dependence on the Gilbert damping constant; besides, a way to study damping in thin ferromagnetic films by SHG experiments is revealed. The THz generation mechanism suggested in the current paper may be used for spintronic terahertz emitters, which have been intensively developed in the last decade [8].

ACKNOWLEDGMENT

This work is supported by the Russian Science Foundation, Grant No. 23-22-00295.

[1] Y. R. Shen, *The Principles of Nonlinear Optics* (John Wiley & Sons, Inc., Hoboken, New Jersey, 2003).

[2] O. A. Aktsipetrov, Surface nonlinear optics and nonlinear magneto- optics at moscow state university, J. Opt. Soc. Am. B **28**, A27 (2011).

[3] K. Y. Kim, A. J. Taylor, J. H. Glownia, and G. Rodrigues, Coherent control of terahertz supercontinuum generation in ultrafast laser–gas interactions, Nat. Photon. **2**, 605 (2008).

[4] V. B. Gildenburg and N. V. Vvedenskii, Optical-to-thz wave conversion via excitation of plasma oscillations in the tunneling-ionization process, Phys. Rev. Lett. **98**, 245002 (2007).

[5] N. V. Vvedenskii, A. A. I. Korytin, V. A. Kostin, A. A. Murzanev, A. A. Silaev, and A. N. Stepanov, Optical-to-thz wave conversion via two-color laser-plasma generation of terahertz radiation using a frequency-tunable half harmonic of a femtosecond pulse, Phys. Rev. Lett. **112**, 055004 (2014).

[6] T. Kampfrath, M. Battiato, P. Maldonado, G. Eilers, J. Nötzold, S. Mährlein, V. Zbarsky, F. Freimuth, Y. Mokrousov, S. Blügel, M. Wolf, I. Radu, P. M. Oppeneer, and M. Münzenberg, Terahertz spin current pulses controlled by magnetic heterostructures, Nat. Nanotechnol. **8**, 256 (2013).

[7] T. Seifert, S. Jaiswal, U. Martens, J. Hannegan, L. Braun, P. Maldonado, F. Freimuth, A. Kronenberg, J. Henrizi, I. Radu, E. Beaurepaire, Y. Mokrousov, P. M. Oppeneer, M. Jourdan, G. Jakob, D. Turchinovich, L. M. Hayden, M. Wolf, M. Münzenberg, M. Kläui, and T. Kampfrath, Efficient metallic spintronic emitters of ultrabroadband terahertz radiation, Nat. Photon. **10**, 483 (2016).

[8] C. Bull, S. M. Hewett, R. Ji, C.-H. Lin, T. Thomson, D. M. Graham, and P. W. Nutter, Spintronic terahertz emitters: Status and prospects from a materials perspective, APL Mater. **9**, 090701 (2021).

[9] R.-P. Pan, H. D. Wei, and Y. R. Shen, Optical second-harmonic generation from magnetized surfaces, Phys. Rev. B **39**, 1229 (1989).

[10] T. V. Murzina, V. V. Radovskaya, I. Y. Pashen'kin, N. S. Gusev, A. I. Maydykovskiy, and E. A. Mamonov, Effect of inhomogeneous magnetization in optical second harmonic generation from layered nanostructures, Opt. Express **29**, 2106 (2021).

[11] I. A. Kolmychek, V. V. Radovskaya, K. A. Lazareva, E. E. Shalygina, N. S. Gusev, A. I. Maidykovskii, and T. V. Murzina, Magnetic-field-induced optical second-harmonic generation study of Co/Pt and Co/Ta interfaces, J. Exp. Theor. Phys. **130**, 555 (2020).

[12] T. V. Murzina, I. A. Kolmychek, N. S. Gusev, and A. I. Maidykovskii, Giant magnetic field induced effects in the second-harmonic generation in a planar anisotropic ta/co/pt structure, JETP Lett. **111**, 333 (2020).

[13] Y. Tserkovnyak, A. Brataas, and G. E. W. Bauer, Enhanced gilbert damping in thin ferromagnetic films, Phys. Rev. Lett. **88**, 117601 (2002).

[14] Y. Tserkovnyak, A. Brataas, G. E. W. Bauer, and B. I. Halperin, Nonlocal magnetization dynamics in ferromagnetic heterostructures, Rev. Mod. Phys. **77**, 1375 (2005).

[15] P. L. Kapitza, Pendulum with a vibrating suspension, Usp. Fiz. Nauk **44**, 7 (1951).

[16] A. I. Akhiezer and S. V. Peletminskii, Behavior of ferrodielectrics and antiferromagnets in a rapidly oscillating magnetic field, Sov. Phys. Solid State **10**, 2609 (1968).

[17] A. K. Zvezdin and V. G. Red'ko, Domain structure of a ferromagnet in a rapidly oscillating magnetic field, ZhETF Pis. Red. **21**, 445 (1975) [JETP Lett. **21**, 203 (1975)].

[18] Y. I. Dzhezherya, K. O. Demishev, and V. N. Korenivskii, Kapitza problem for the magnetic moments of synthetic antiferromagnetic syste, JETP Lett. **115**, 284 (2012).

[19] K. V. Kulikov, D. V. Anghel, A. T. Preda, M. Nashaat, M. Sameh, and Y. M. Shukrinov, Kapitza pendulum effects in a josephson junction coupled to a nanomagnet under external periodic drive, Phys. Rev. B **105**, 094421 (2022).

[20] L. D. Landau and E. M. Lifshitz, *Course of Theoretical Physics, Vol. 8: Electrodynamics of Continuous Media* (Butterworth–Heinemann, Oxford, 1984).

[21] A. V. Gaponov and M. A. Miller, Potential wells for charged particles in a high-frequency electromagnetic field, J. Exptl. Theoret. Phys. (U.S.S.R.) **34**, 242 (1958).

[22] J. Sinova, S. O. Valenzuela, J. Wunderlich, C. H. Back, and T. Jungwirth, Spin hall effects, Rev. Mod. Phys. **87**, 1213 (2015).

[23] A. A. Rzhevsky, B. B. Krichevtsov, D. E. Bürgler, and C. M. Schneider, Interfacial magnetization in exchange-coupled Fe/Cr/Fe structures investigated by second harmonic generation, Phys. Rev. B **75**, 144416 (2007).

[24] H. Yu, O. d'Allivy Kelly, V. Cros, R. Bernard, P. Bortolotti, A. Anane, F. Brandl, R. Huber, I. Stasinopoulos, and D. Grundler, Magnetic thin-film insulator with ultra-low spin wave damping for coherent nanomagnonics, Sci. Rep. **4**, 6848 (2014).

[25] D. Zhang, J. J. Yue, Z. X. Kou, L. Lin, Y. Zhai, and H. R. Zhai, The investigation of ferromagnetic resonance linewidth in $Ni_{80}Fe_{20}$ films with submicron rectangular elements, AIP Adv. **6**, 056125 (2016).

[26] D. O. Krivulin, I. Y. Pashen'kin, R. V. Gorev, P. A. Yunin, M. V. Sapozhnikov, A. V. Grunin, S. A. Zaharova, and V. N. Leont'yev, Influence of radiation exposure on the magnetic properties of feromagnetic/Irmn films with exchange bias, Techn. Phys. **68**, 843 (2023).

[27] E. V. Skorokhodov, E. S. Demidov, S. N. Vdovichev, and A. A. Fraerman, Ferromagnetic resonance in a system of magnetic

films with different curie temperatures, J. Exp. Theor. Phys. **124**, 617 (2017).

[28] S. A. Gusev, M. N. Drozdov, O. L. Ermolaeva, A. A. Fraerman, N. S. Gusev, V. Y. Mikhailovskii, Y. V. Petrov, M. V. Sapozhnikov, and S. N. Vdovichev, The modification of the structure of multilayer Co/Pt films by the irradiation with a focused helium ion beam, AIP Conf. Proc. **1748**, 030002 (2016).

[29] O. Guillon, C. Elsässer, O. Gutfleisch, J. Janek, S. Korte-Kerzel, D. Raabe, and C. A. Volkert, Manipulation of matter by electric and magnetic fields: Toward novel synthesis and processing routes of inorganic materials, Mater. Today **21**, 527 (2018).

[30] Y. Onose, Y. Shiomi, and Y. Tokura, Lorenz number determination of the dissipationless nature of the anomalous hall effect in itinerant ferromagnets, Phys. Rev. Lett. **100**, 016601 (2008).

[31] I. A. Kolmychek, V. B. Novikov, N. S. Gusev, I. Y. Pashen'kin, E. A. Karashtin, and T. V. Murzina, Second harmonic generation versus linear magnetooptical response studies of laser-induced switching of pinning effects in antiferromagnetic/ferromagnetic films, Photonics **10**, 1303 (2013).