Theory of the inverse Faraday effect in view of ultrafast magnetization experiments

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We supplement the theory of the inverse Faraday effect, which was developed in the 1960s, to the conditions used today in ultrafast magnetization experiments. We show that assumptions used to derive the effective Hamiltonian and magnetization are not valid under these conditions. We extended the approach to be applicable to describe magnetization dynamics at femtosecond time scales. We show that after the action of an ultrafast laser pulse the system is brought with a certain probability to a state, the magnetic signature of which is different from before the excitation.

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I. INTRODUCTION

Ultrafast optical control of the magnetic state of a medium is presently a subject of intense research. It is of importance for the development of alternative concepts for high-speed magnetic recording and information processing. A series of experiments has revealed direct optical control of magnetization via the inverse Faraday effect (IFE),[1,2] i.e.,the generation of an effective magnetic field by light. In these experiments, magnetization reversal on subpicosecond time scales induced by high-intensity laser pulses was demonstrated. The results motivated intense experimental[3–13] and theoretical[14–21] investigations in the field of ultrafast magnetization, but the mechanisms of magnetization reversal on femtosecond time scales and the origin of the IFE are still poorly understood.

The IFE was predicted theoretically by Pitaevskii[22] in 1960 from a pure phenomenological ansatz on the basis of a thermodynamic potential describing the internal energy of a system. Van der Ziel *et al.*[23] observed the effect in 1966 and Pershan *et al.*[24] provided a detailed explanation of the IFE from the quantum-mechanical point of view. The equation $\mathbf{M}(t) = -\gamma \mathbf{E}^*(t) \times \mathbf{E}(t)$, connecting the magnetic field $\mathbf{M}(t)$ to the generating electric field $\mathbf{E}(t)$ of the light pulse, was derived,[22,24] which has been used until today to describe the IFE. According to it, the change of magnetization is proportional to the intensity of the pulse and vanishes after the action of the pulse. However, in all modern measurements of the IFE, the magnetization remains altered and takes some time to stabilize.

With the advent of ultrafast pump-probe experiments, the experimental conditions today are quite different from the ones realized in the past. The duration of the laser pulse used in the experiment of van der Ziel *et al.*[23] was 30 ns. In the experiments carried out nowadays, the duration of the laser pulse is equal to or even shorter than 100 fs. That is about six orders of magnitude shorter than in the 1960s and the laser fluence used today is much higher. In the experiment of Kimel *et al.*[1] the fluence was about 10^{11} W/cm^2, which is four orders of magnitude higher than in Ref. 23. Another essential difference lies in the observation of the magnetization dynamics. Ziel *et al.*[23] measured the magnetization during the time the pulse was present and the variation of magnetization was zero after the action of the pulse. Nowadays, the magnetization dynamics after the action of the laser field is of interest and requires an interpretation, which is opposite the essence of the studies in 1960s. Thus there should be different mechanisms that are responsible for the stationary and ultrafast IFE.

The understanding of these ultrafast mechanisms is essential for the investigation of spin precessions, which arise after the action of circularly polarized light pulse on a system. A proper set of equations that would describe the full picture of the magnetic vector oscillations has been sought in several publications.[8,9,18,19] The problem that the authors encounter is the inclusion of the correct time dependence of the induced magnetization $\mathbf{M}(t)$ in such equations. According to the standard expression $\mathbf{M}(t) = -\gamma \mathbf{E}^*(t) \times \mathbf{E}(t)$, there is no magnetic field after the action of the laser light, which apparently is not the case since the spin precessions are observed for much longer times than the pulse duration.[4–10] In Refs. 8,17, and 19 the induced magnetic field was introduced as a δ function. It is a convenient however not quite exact approximation when it is applied to derive the spin oscillations since their period is typically on the order or just one order of magnitude higher than a pulse duration. Another useful approach to connect the laser magnetic action to spin oscillations has been suggested by Galkin and Ivanov,[18] but also under the assumption that the system is under "the action of a short magnetic field pulse of high amplitude and width δt much smaller than the problem characteristic time." The primary motivation of our work is to find the correct way to introduce the time dependence of the induced magnetization. We derive our approach without any approximations concerning the pulse length. We obtain that the induced magnetization does not remain zero after the action of an ultrashort laser pulse.

Reid *et al.*[10] have experimentally shown that "magnetism on the subpicosecond time scale cannot be adequately described by the thermodynamic model of the inverse Faraday effect." They compared the initial amplitudes of the observed oscillations, excited by the 50-fs-long light pulse, with static measurements of the materials Verdet constant, which is proportional to γ, over a range of temperatures and found that the two have very different temperature dependences. They also obtained that the frequency of the oscillations are 30 times higher than expected for magnetization precessing in the external field. The second question that we try to answer is where these disagreements with the classical theories[22,24] come from.

We want to update and extend the approach developed by Pershan *et al.*,[24] which is the commonly accepted theoretical foundation of the IFE, to the experimental conditions today characterized by the availability of ultrafast lasers. Although the process that is studied by Pershan *et al.*and in the present work is the same, the mechanism of the stationary and ultrafast IFE is shown to be different. We explain why the thermodynamic approach that works in the nanosecond region is not valid for the description of the magnetization dynamics on subpicosecond time scales.

II. EFFECTIVE HAMILTONIAN APPROACH

The stimulated Raman-like scattering process was suggested to be responsible for the magnetization reversal by light.[7,24] In this process, the laser pulse excites the transition of an electron from the initial state to a virtual intermediate one and then to the final state. Through the intermediate state, the spins or magnetic moments of the system are influenced and at the end the system is in a different magnetic state.

This process is described by the time-dependent Schrödinger equation. The wave function of the system due to the action of the electric field $\mathbf{E}(t)$ is found by the solution of

$$i\frac{\partial\Psi(t)}{\partial t} = [\mathcal{H}_0 + V(t)]\Psi(t) \tag{1}$$

using the Volterra iteration method, where $V(t)$ is the perturbation due to the action of a field and $\mathcal{H}_0$ is the unperturbed Hamiltonian. It includes all electron interactions, particularly for the effects on the spin of the electrons, such as spin-orbit and Zeeman interactions. The solution is the expansion

$$\begin{aligned}\Psi(t) &= e^{-iH_0t}[\Psi_0 + \Psi_1(t) + \Psi_2(t) + \cdots] \\ &= e^{-iH_0t}\left[1 - \frac{i}{\hbar}\int_{-\infty}^{t}\bar{V}(t')dt'\right. \\ &\quad \left. -\frac{1}{\hbar^2}\int_{-\infty}^{t}\bar{V}(t')dt'\int_{-\infty}^{t'}\bar{V}(t'')dt'' + \cdots\right]\Psi_0, \end{aligned} \tag{2}$$

with $\bar{V}(t) = e^{iH_0t}V(t)e^{-iH_0t}$.

The Raman-like process is of second order in the inverse speed of light $1/c$. The perturbation is of first order $1/c$: $V(t) = -\mathbf{d}\cdot\mathbf{E} = \frac{1}{c}\mathbf{d}\cdot\dot{\mathbf{A}}$, where $\mathbf{d}$ is the dipole moment of the system and $\mathbf{A}$ is the vector potential. Therefore, the third term of the expansion in Eq. (2) is of interest here. Pershan *et al.*[24] introduced an effective Hamiltonian $\mathcal{H}_{\text{eff}}$ of the system defined by transition amplitudes between initial states i and final states f,

$$\begin{aligned}&\langle f|-\frac{i}{\hbar}\int_{-\infty}^{t}\mathcal{H}_{\text{eff}}(t')dt'|i\rangle \\ &\quad = \langle f|-\frac{1}{\hbar^2}\int_{-\infty}^{t}\bar{V}(t')dt'\int_{-\infty}^{t'}\bar{V}(t'')dt''|i\rangle,\end{aligned} \tag{3}$$

which leads to

$$\langle f|\mathcal{H}_{\text{eff}}(t)|i\rangle = -\frac{i}{\hbar}\sum_j\langle f|\bar{V}(t)|j\rangle\int_{-\infty}^{t}\langle j|\bar{V}(t')|i\rangle dt', \tag{4}$$

with j being the intermediate states. Taking the most general form of the perturbation $V(t) = v(t)e^{i\omega t} + v^*(t)e^{-i\omega t}$ and assuming that the amplitude of the perturbation $v(t)$ varies on a characteristic time scale T that is much larger than $1/(\omega\pm\omega_{ij})$, the approximation

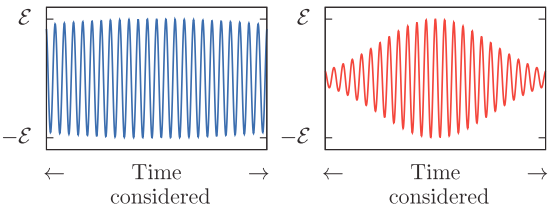

FIG. 1. (Color online) Pulse with amplitude that does not change noticeably in time (left) and Gaussian-shaped pulse (right). Time T characterizing the pulse duration is a factor 20 times shorter for the right pulse as compared to the left pulse.

$$\int_{-\infty}^{t}v(t')e^{i(\omega_{ij}\pm\omega)t'}dt' \approx v(t)\frac{e^{i(\omega_{ij}\pm\omega)t}}{i(\omega_{ij}\pm\omega)} \tag{5}$$

becomes valid except for resonant transitions $\pm\omega\approx\omega_{ij}$, where $\omega_{ij} = \frac{\epsilon_j-\epsilon_i}{\hbar}$ with $\epsilon_{i\,(j)}$ the energy of the state i (j). Under these conditions one obtains

$$\int_{-\infty}^{t}e^{i\omega_{ij}t'}\bar{V}(t')dt' = v(t)\frac{e^{i(\omega_{ij}+\omega)t}}{i(\omega_{ij}+\omega)} + v^*(t)\frac{e^{i(\omega_{ij}-\omega)t}}{i(\omega_{ij}-\omega)}. \tag{6}$$

Then the effective Hamiltonian is found as

$$\langle f|\mathcal{H}_{\text{eff}}(t)|i\rangle = -\frac{i}{\hbar}\sum_j\left[\frac{v_{ij}(t)v_{jf}^*(t)}{\omega_{ij}+\omega} + \frac{v_{jf}(t)v_{ij}^*(t)}{\omega_{ij}-\omega}\right]e^{i\omega_{if}t}. \tag{7}$$

The terms $v_{ij}v_{jf}e^{i(\pm2\omega t+\omega_{ij})}$ correspond to a second harmonic process. They connect the initial state to final states, which are energetically widely separated from the initial state and need not be considered here.

The ultrafast magnetization experiments are carried out with pulses several tens of fs long ($T\sim1/|\omega_{ij}-\omega|$). Therefore, the change of the pulse amplitude in time cannot be be neglected anymore. For example, if we choose the perturbation to be a circularly polarized Gaussian-shaped pulse $v(t) = -\mathbf{d}\cdot\boldsymbol{\mathcal{E}}e^{-t^2/T^2}$ (where $\boldsymbol{\mathcal{E}}$ is the amplitude of the electric field) the approximation in Eq. (5) would be valid only under the assumption that the laser field could be considered almost stationary, i.e., if $T\to\infty$, which was actually the condition considered by Pershan *et al.*[24] The differences between the pulses are illustrated in Fig. 1. The left-hand plot exhibits the shape of a pulse for which the approximation in Eq. (5) holds. During the time considered, the amplitude of $v(t)$ does not change significantly and the time integral over the field is determined by the periodic function $e^{\pm i\omega t}$. In the right-hand plot the constant T that characterizes the pulse width is 20 times shorter and the variation of $v(t)$ is important. When integrating over the pulse, the factor e^{-t^2/T^2} cannot be omitted.

The exact solution of the integral in Eq. (6) for the Gaussian-shaped laser pulse is given by

$$\begin{aligned}\int_{-\infty}^{t}e^{i\omega_{ij}t'}\bar{V}(t')dt' &= -\mathbf{d}\cdot\boldsymbol{\mathcal{E}}\,\frac{T\sqrt{\pi}}{2} \\ &\times\left\{e^{-[T(\omega_{ij}+\omega)]^2/4}\left[1+\text{erf}\left(\frac{t}{T}-\frac{i}{2}T(\omega_{ij}+\omega)\right)\right]\right. \\ &\left.+e^{-[T(\omega_{ij}-\omega)]^2/4}\left[1+\text{erf}\left(\frac{t}{T}-\frac{i}{2}T(\omega_{ij}-\omega)\right)\right]\right\}.\end{aligned} \tag{8}$$

Thus the effective Hamiltonian [Eq. (4)] for this pulse is

$$\begin{aligned}\langle f|\mathcal{H}_{\rm eff}(t)|i\rangle &= -\frac{i\sqrt{\pi}}{\hbar}\mathcal{E}^2 T\sum_j d_{ij}d_{jf}e^{-t^2/T^2}e^{i\omega_{jf}t}\cos\omega t \\ &\times\left[e^{-[T(\omega_{ij}+\omega)]^2/4}\mathrm{erfc}\left(\frac{i}{2}T\left(\omega_{ij}+\omega\right)-\frac{t}{T}\right)\right. \\ &\left.+\ e^{-[T(\omega_{ij}-\omega)]^2/4}\mathrm{erfc}\left(\frac{i}{2}T\left(\omega_{ij}-\omega\right)-\frac{t}{T}\right)\right]. \quad (9)\end{aligned}$$

For large complex arguments $z = |z|\,e^{i\theta}$, $|z| \to \infty$, and the polar angle $|\theta| < 3\pi/4$, the function $\mathrm{erfc}(z)$ approaches asymptotically $\frac{e^{-z^2}}{\sqrt{\pi}z}$.[25] Substituting this asymptote into Eq. (9), one obtains exactly Eq. (7). The range of validity of Eq. (7) may be determined precisely. From the condition $|\theta| < 3\pi/4$ it follows that $T|\omega_{ij} \pm \omega| > 2t/T$ and the condition $T|\omega_{ij} \pm \omega| \gg 1$ is necessary for $|z| \to \infty$. Thus Eq. (7) is certainly applicable to the experiments of Ziel *et al.*[23] with pulse durations on the order of nanoseconds; however, it is not valid for pulses several tens of fs long and times larger than the pulse duration.

In order to obtain the effective Hamiltonian for a system, the transition amplitudes between the initial, intermediate (which would mix orbital momentum and spin), and final states with different magnetic quantum numbers from the initial ones should be calculated and summed. Thus the simplest possible example to demonstrate the discrepancies between the two different approaches in describing the laser excitation to obtain the effective Hamiltonian is a three-level system as depicted in Fig. 2. However, the results of such a comparison are general because the temporal behavior of the functions presented later would be similar in many-level systems. We considered the excitation of a Gaussian-shaped laser pulse that is 100 fs long ($T = 10^{-13}$ s) and calculated the time evolutions of the amplitudes of the effective Hamiltonians in Eqs. (7) and (9). The results are plotted in Figs. 3(a) and 3(b), respectively, in units of energy $\xi = \mathcal{E}^2 d_{ij}d_{jf}T/\hbar$. We can estimate the amplitude by making the following reasonable assumptions: If the dipole matrix elements are of the order of 1 a.u. (≈53 pm) and the electric-field amplitude is about 10^7 V/m, which is a typical value for laser fluences of 10^{11} W/cm^2, then $\xi \approx 10^{-4}$ eV.

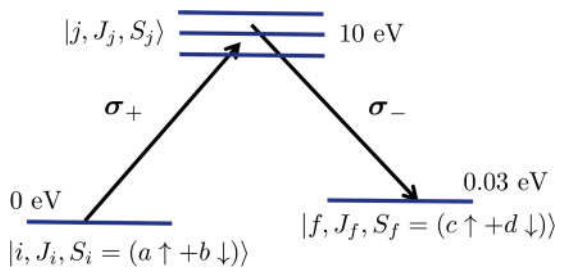

FIG. 2. (Color online) The three-level system investigated. The laser pulse causes transitions from the initial state $|i\rangle$ to the intermediate $|j\rangle$ and then to the final one $|f\rangle$, with a magnetic state different from the one of the initial state. The spin is influenced by the spin-orbit coupling of $|j\rangle$.

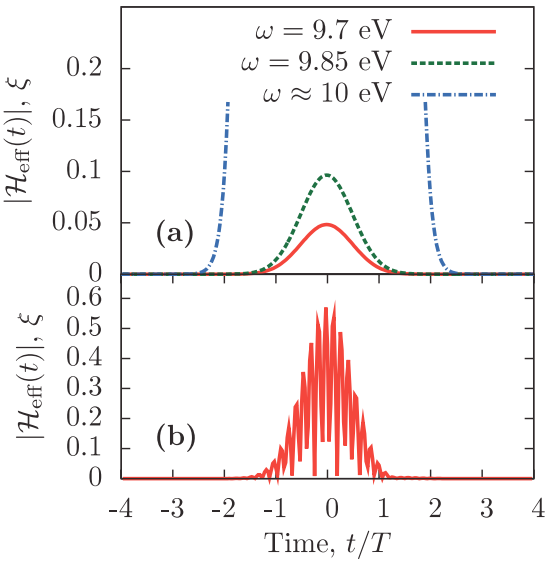

FIG. 3. (Color online) Time evolution of the amplitude of the effective Hamiltonian (a) in Eq. (7) at different laser frequencies and (b) in Eq. (9) at the laser frequency $\omega = 9.7$ eV.

Figure 3(a) shows that the amplitudes of the function in Eq. (7) reproduce the typical behavior of transition amplitudes when the excitation frequency is off resonance. The maximum increases when the excitation frequency is closer to resonance. At resonance the function in Eq. (7) simply diverges. It is indicated in Fig. 3(a) by a curve taken with a small detuning off resonance: $\omega - \omega_{ij} = 10^{-6}$ eV. This divergence is a manifestation of the importance of the assumption of Pershan *et al.*[24] that the excitation frequency must be significantly far from any resonances $|\omega \pm \omega_{ij}| \gg 1/T$.

In Fig. 3(b) we depict the action of the effective Hamiltonian in Eq. (9) for only one excitation frequency, namely, $\omega =$ 9.7 eV, because plots of close-by frequencies overlap in a way that cannot be graphically resolved. The functions at the frequencies $\omega = 9.85$ and 10 eV are very similar: The height of the maximum is almost the same; only the positions of the local maxima are different. The amplitudes of Eq. (9) are oscillating functions, which is consistent with the presence of the term $\cos\omega t$.

From the plots one could see that at the frequency $\omega =$ 9.7 eV the amplitude of the function in Eq. (7) is one order of magnitude smaller than the one in Eq. (9). Furthermore, the former function is smooth, while the latter is oscillating. The completely different behavior of both functions arises from the fact that the validity condition of the approximation in Eq. (5) is not satisfied since $T(\omega_{ij} - \omega) \approx 10$. Therefore, the maximum of the function in Eq. (9) is still proportional to T, while the one in Eq. (7) is proportional to $\frac{2\omega}{|\omega_{ij}^2 - \omega^2|}$, thus explaining the factor of 10 difference between the amplitudes.

Though both functions differ significantly under the chosen conditions, they approach each other with an increase of T. Figure 4 shows both functions for $\omega = 9.7$ eV, when T is one order of magnitude larger. The oscillations of the function in Eq. (9) still remain because the terms $v_{ij}v_{jf}e^{i(\pm 2\omega t + \omega_{ij})}$ were not eliminated in Eq. (9).

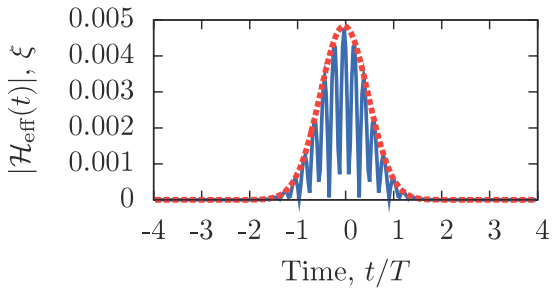

FIG. 4. (Color online) Time evolutions of the amplitude of both effective Hamiltonians in Eqs. (7) [dashed (red) line] and (9) [solid (blue) line] at $T = 10^{-12}$ s and at the laser frequency $\omega = 9.7$ eV. The unit ξ is rescaled according to the increased T.

In order to derive the magnetization dynamics from the effective Hamiltonian, Pershan *et al.*[24] defined a potential function as

$$F(t) = \sum_{if} \mathcal{H}^{if}_{\rm eff}(t)\rho_{if}(t), \tag{10}$$

where ρ_{if} is the density matrix for the atomic system. They obtained the time dependence of the magnetization as derivative of the potential function with respect to an external magnetic field H, $M(t) = -\partial F(t)/\partial H|_{H=0}$, under the assumption that $v_{ij}(t)v^*_{jf}(t)$ changes slowly compared to thermal relaxation times of the system.

If we apply this relation to our system, we would be able to describe the magnetization dynamics only during the pulse, i.e., when $\mathcal{H}^{if}_{\rm eff}(t)$ is nonzero. According to this relation, the functional and, consequently, the magnetization would be zero after the action of the pulse. This is certainly not the case in the experiments of Kimel *et al.*,[1] where the magnetization dynamics is observed after the pulse. This prescription on the basis of thermodynamical potentials fails due to the steady-state conditions [Eq. (2.11) in Ref. 26] and the thermodynamical equilibrium underlying the derivation, which simply cannot be considered in the ultrafast magnetization experiments, where intensities are very high and the time scales are shorter than any relaxation time of the system.

III. SECOND ORDER WAVE-FUNCTION

In order to study the time dependence of the magnetization after the action of a fast laser pulse we suggest that the second-order wave function Ψ_2 introduced in Eq. (2) should be calculated, which gives the probability of the transitions, leading to the change of the magnetic state of the system. It is related to the effective Hamiltonian by the integral

$$\Psi_2(t) = -\frac{i}{\hbar}\int_{-\infty}^{t} \mathcal{H}_{\rm eff}(t')dt'. \tag{11}$$

The second-order wave function Ψ_2 was calculated for the three-level system using the Hamiltonians in Eqs. (7) and (9), respectively. The solution resulting from the Hamiltonian in Eq. (7) is denoted by $\bar{\Psi}_2$. The time evolutions of $|\bar{\Psi}_2|$ and $|\Psi_2|$ are plotted in Figs. 5(a) and 5(b), respectively, in the dimensionless units $w = \xi T/\hbar$, with $T = 100$ fs.

For the calculation of $\bar{\Psi}_2$ under the action of the Hamiltonian $\mathcal{H}_{\rm eff}(t)$ in Eq. (7) we applied the approximation in Eq. (5),

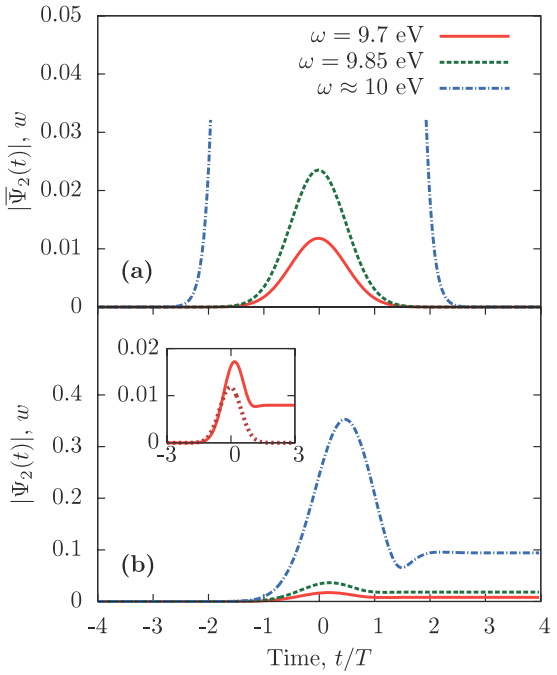

FIG. 5. (Color online) Time evolution of the second-order wave function (a) $|\bar{\Psi}_2(t)|$ applying the approximation in Eq. (5) and (b) $|\Psi_2(t)|$ according to the effective Hamiltonian in Eq. (9). The inset shows $|\Psi_2(t)|$ [solid (red) line] and $|\bar{\Psi}_2(t)|$ [dashed (brown) line] at $\omega = 9.7$ eV.

which is consistent with the derivation of the Hamiltonian in Eq. (7). We find that the evolution of $\bar{\Psi}_2$ is proportional to the effective Hamiltonian [Fig. 5(a)] and thus it has the same functional dependence as the potential function in Eq. (10). This confirms that the functional F can be used to calculate the magnetization under the condition that Eq. (5) is applicable. Compatible with Eq. (10), the magnetization goes to zero after the excitation is completed according to the time evolution of $\bar{\Psi}_2$.

Except at resonance, the functions $|\bar{\Psi}_2|$ and $|\Psi_2|$ exhibit the same behavior during the first half of the pulse [see the inset of Fig. 5(b)], but the key difference is that the function $|\Psi_2|$ is nonzero after the action of the pulse. This means that the system remains in an altered state, i.e., with an altered magnetization, after the laser pulse has faded away. Therefore, $\Psi_2(t)$ is able to describe the magnetization dynamics after the excitation in the ultrafast magnetization experiments.

For example, the simplest mechanism of magnetization changes via the Raman-like scattering process is depicted in Fig. 2. Due to some internal or external magnetic field, all spins in the system are aligned in one direction. The light is circularly polarized and propagating in a different direction, which is chosen as the axis of quantization. Therefore, the ground state is a mixture of spin-up and -down states. The role of the spin-orbit coupling is to split the excited states with different combinations of $|M_L + 1, \uparrow\rangle$ and $|M_L + 1, \downarrow\rangle$ (where M_L is the projection of the orbital moment of the initial state). Thus selection rules and dipole matrix elements for the transition with circularly polarized light to the excited state are different for each component.[24] Thereby, the spin of the electron is influenced by the virtual state. After the emission of

a photon, the electron arrives at a state with spin components that are different from the initial ones.

Calculating the second-order function $\Psi_2(t)$, one would obtain the probabilities for the new state to be spin up or spin down. In order to calculate this function correctly in a solid state, the wave functions, which describe all transitions over excited levels j and final states f, which would effect the spin, should be summarized $\Psi_2(t) = \sum_{jf} \Psi_2^{jf}(t)$. Therefore, the simplified picture presented here does not influence the conclusions on the temporal behavior, which is the main concern of this paper. However, the role of the excited states should be accurately studied in a real material.

The induced magnetization $M_\alpha(t)$ can be derived from this function with the help of the momentum operators $\hat{\mathbf{j}}_\alpha$ (α stays for x,y,z) as follows. If the wave function of an atom is Ψ', then its magnetization is $M_\alpha = -\mu_B g_J \langle \Psi' | \hat{\mathbf{j}}_\alpha | \Psi' \rangle / |\Psi'|^2$. We substitute $\Psi' = \Psi_0 + \Psi_2$ for our system because we have to take into account the total influence of the unchanged ground state i and the state f, which becomes occupied due to the action of light. Subtracting the initial magnetization, we obtain

$$M_\alpha = -\mu_B g_J \left(\frac{\langle \Psi_0 + \Psi_2 | \hat{\mathbf{j}}_\alpha | \Psi_0 + \Psi_2 \rangle}{|\Psi_0 + \Psi_2|^2} - \frac{\langle \Psi_0 | \hat{\mathbf{j}}_\alpha | \Psi_0 \rangle}{|\Psi_0|^2} \right)$$
$$\approx -\mu_B g_J \left(\langle \Psi_0 | \hat{\mathbf{j}}_\alpha | \Psi_2 \rangle + \langle \Psi_2 | \hat{\mathbf{j}}_\alpha | \Psi_0 \rangle + \langle \Psi_2 | \hat{\mathbf{j}}_\alpha | \Psi_2 \rangle \right). \quad (12)$$

The first two terms in small parentheses are proportional to $w = \mathcal{E}^2 d_{ij} d_{jf} T^2/\hbar^2$ and the last term in small parentheses is proportional to w^2; thus it is negligible and can be ignored. The equation becomes

$$M_\alpha(t) \approx -\mu_B g_J [\langle \Psi_0 | \hat{\mathbf{j}}_\alpha | \Psi_2(t) \rangle + \langle \Psi_2(t) | \hat{\mathbf{j}}_\alpha | \Psi_0 \rangle]. \quad (13)$$

According to it, the induced magnetization $M_\alpha(t)$ is proportional to w, which is proportional to the light fluence. The same dependence has been seen in the experiments.[1,5,6] Another result is that the time dependence of the magnetization is determined by the function $\Psi_2(t)$ and has similar behavior, as depicted in Fig. 5. It is completely different from the expression $-\gamma \mathbf{E}^*(t) \times \mathbf{E}(t)$ in the ultrafast regime, but approaches it with increasing T [see the discussion of Eq. (9)]. The function $\Psi_2(t)$ and, consequently, the induced magnetization depend greatly on the ultrashort laser pulse properties (such as shape or frequency). This statement is supported by the observation in Ref. 8 that the initial phase and amplitude of the oscillation of the polarization of the probe pulse depend on the pump wavelength. This results in many opportunities for tuning spin dynamics by adjusting the laser properties.

IV. CONCLUSIONS

In the case of ultrashort laser pulses ($T \sim 1/\Delta\omega$), the effective Hamiltonian and Ψ_2 are no longer proportional and there is nothing like the functional in Eq. (10) to obtain magnetization. Under these circumstances, the effective Hamiltonian must be integrated over time to obtain the wave function describing the transitions that cause the change of the magnetic state. Thus we conclude that the change of magnetization after the action of an ultrashort laser pulse can be obtained when the time-dependent Schrödinger equation is solved without approximations. We suggest that the magnetic action of the light should be considered as the magnetization of the sample changes from its ground-state value to some nonequilibrium one after the excitation. This description would be helpful to derive the correct expression for the spin oscillations.

In summary, we extended the theory of Pershan *et al.*,[24] developed to describe the inverse Faraday effect, to the regime of ultrafast magnetization dynamics, which is the focus of current research. We showed that the approximations used at that time cannot be applied to ultrafast pump-probe-type laser experiments. The exact solution of the time-dependent Schrödinger equation up to the second order of $1/c$ explains why ultrashort laser pulses can cause a change of magnetization. A laser pulse excites two transitions in the system: from the initial to the intermediate state and from the intermediate to the final state, which is in a *different* magnetic state from the initial one, in which the system remains with a certain probability. Magnetization due to the action of the pulse is related to this probability, but not to a thermodynamical functional, which is derived from an effective Hamiltonian. The same considerations can be applied to other magneto-optical effects, showing that a subpicosecond magnetization dynamics should be treated differently from that in a nanosecond region. The formalism outlined to describe the action of ultrashort laser pulses is general and may also be applicable to other optical experiments.

ACKNOWLEDGMENTS

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