PHYSICAL REVIEW B **89**, 014413 (2014)

Quantum theory of the inverse Faraday effect

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(Received 21 August 2013; revised manuscript received 2 December 2013; published 14 January 2014)

We provide a quantum theoretical description of the magnetic polarization induced by intense circularly polarized light in a material. Such effect—commonly referred to as the inverse Faraday effect—is treated using beyond-linear response theory, considering the applied electromagnetic field as external perturbation. An analytical time-dependent solution of the Liouville–von Neumann equation to second order is obtained for the density matrix and used to derive expressions for the optomagnetic polarization. Two distinct cases are treated, the long-time adiabatic limit of polarization imparted by continuous wave irradiation, and the full temporal shape of the transient magnetic polarization induced by a short laser pulse. We further derive expressions for the Verdet constants for the inverse, optomagnetic Faraday effect and for the conventional, magneto-optical Faraday effect and show that they are in general different. Additionally, we derive expressions for the Faraday and inverse Faraday effects within the Drude-Lorentz theory and demonstrate that their equality does not hold in general, but only for dissipationless media. As an example, we perform initial quantum mechanical calculations of the two Verdet constants for a hydrogenlike atom and we extract the trends. We observe that one reason for a large inverse Faraday effect in heavy atoms is the spatial extension of the wave functions rather than the spin-orbit interaction, which nonetheless contributes positively.

DOI: 10.1103/PhysRevB.89.014413 PACS number(s): 78.20.Ls, 78.20.Bh, 78.47.J−

I. INTRODUCTION

In magneto-optics one studies the influence of the magnetization of a sample on the light traversing the sample. This influence gives, e.g., rise to the Faraday effect (FE), which is usually observed as a change in the light's polarization caused by a magnetically polarized material (see, e.g., Refs. [1] and [2]). Optomagnetics [3], conversely, is concerned with a different action of intensive light on a material: circularly polarized light may induce a magnetic polarization in the material. Such "inverse" action was first observed in the 1960s through the discovery of the inverse Faraday effect (IFE) [4,5].

The IFE has been a popular topic in physics for a long time. Its application in the field of plasma physics is established [6–8]. However, in recent years it has gained a renewed importance in the field of solid-state physics. A key discovery was that of the all-optically induced magnetization which was imparted by femtosecond laser pulses in a magnetic material [9]. Since then the induced optomagnetic polarization has been considered as a main possible driver of the ultrafast magnetization response [3,10–16]. Its popularity increased even more after it was recognized that it could offer a route to ultrafast, all-optical magnetization reversal [11–13]. Helicity-dependent all-optical switching of the magnetization has meanwhile been demonstrated in several rare-earth transition-metal alloys [17–19]. Optically induced magnetizations have been also detected in other classes of materials [20], and they have been proposed to be responsible for spin oscillations in antiferromagnets [21–23].

In spite of the significant technological applications, the origin of the optomagnetic polarization and thus of the IFE have remained unclear from a theoretical point of view. Several semiclassical approaches [24–27] to describe it have been attempted. Although these do provide very helpful physical insight on the effect, they are not materials specific and lack the ability to make numerical predictions on the *ab initio* level. A first quantum-mechanical treatment of the effect was proposed by Pershan *et al.* and dates back to the 1960s [5]. More recently, owing to the renewed interest in the field more modern approaches were proposed [28–33], but these are still limited to model systems and cannot address the question of providing a materials-specific framework for the calculation of the optomagnetic polarization.

Here we aim to derive a quantum-mechanically exact expression for the IFE that can be implemented in an *ab initio* code to perform materials-specific calculations. We use a newly developed theoretical framework [34] to give the full explicit expressions for the induced optomagnetic polarization. This we do (in Sec. III) for the case of a constant magnetization imparted by a continuous electromagnetic field in the adiabatic limit. As a second result, given the current importance of the IFE driven by femtosecond laser pulses, we provide in Sec. IV a more specific expression for the optomagnetic response to a fs light pulse. We furthermore derive expressions for the Verdet constants of the Faraday and the inverse Faraday effect in paramagnetic materials in Sec. VI. We stress that the provided expressions are quantum mechanical and exact, since they are derived by the very definition of the two effects—however, with the restriction that the electric dipole approximation is used for the interaction of the electrons with light, which holds very well for the description of linear magneto-optical effects at optical frequencies (see Ref. [2]). Moreover, these expressions as well as those for the optically induced polarization can be implemented in a density-functional theory (DFT) band-structure code to describe the response of real materials.

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1098-0121/2014/89(1)/014413(9)

In relation to the origin of the IFE, we are in addition able to point out that there exists a difference between the FE and IFE Verdet constants. Earlier theoretical and experimental works suggested that these two constants would be of equal strength. First, the two appearing constants are often (but not always) experimentally similar in amplitude [4]. Second, theoretical results [5,24] seemed to confirm this equality. The classical treatment in Ref. [24] was unfortunately based on a fundamental assumption, that of nondissipative materials. This is not valid in a generic material and cannot be assumed from the outset in a quantum treatment. Conversely, the quantum treatment proposed in Ref. [5] made use of a very crude approximation that hid the difference between the two effects and could give only an order-of-magnitude estimation of the process. The nonequivalence of the IFE and FE became highlighted through recent experimental investigations [35,36], which revealed significant differences between the two effects [35], and a deviation of the IFE from the classical expression for an absorbing medium [36].

To address the question of the relation between the FE and IFE also on the classical level we show in Sec. II how even a treatment based on the Drude-Lorentz model (see, e.g., Ref. [25]) reveals that the two Verdet constants are different, and can provide physical intuition as to what their difference has to be ascribed to.

Lastly, in Sec. V we perform numerical calculations of the IFE in hydrogenlike atoms. We perform an initial investigation of how the IFE is influenced by the shape of the electronic orbitals. These investigation suggest that the extent of the wave function could play a role: more delocalized electrons contribute more to the IFE, which is found, for a hydrogen-type atom, to grow quadratically with the radius of the electronic orbit. We also exemplify how the spin-orbit interaction (SO) favors the IFE.

II. CLASSICAL TREATMENT OF FARADAY AND INVERSE FARADAY EFFECTS

The Drude-Lorentz model has been very successful in providing a general physical understanding of the processes involved in conduction in metals. Here, as a premise to the quantum treatment, we use similar concepts to show how the Faraday and the inverse Faraday effect can be described by the same constant only in nondissipative media, and to provide physical insight in the origin of their difference.

The equation of motion of the electron in the Drude-Lorentz approximation is (see, e.g., Ref. [25])

$$\frac{d^2\mathbf{r}(t)}{dt^2}+\gamma\frac{d\mathbf{r}(t)}{dt}+\omega_0^2\mathbf{r}(t)=\frac{\mathbf{F}(t)}{m},\qquad \mathbf{r}(0)=\mathbf{r}_0;\quad \frac{d\mathbf{r}(0)}{dt}=\mathbf{v}_0, \tag{1}$$

where m is the electron effective mass, $\mathbf{r}$ represents its position, $\mathbf{F}$ is the external force acting on it, and $\mathbf{v}_0$ and $\mathbf{r}_0$ are its initial velocity and position. The term $\gamma d\mathbf{r}(t)/dt$ and the one $\omega_0^2\mathbf{r}(t)$ represent phenomenologically, respectively, the randomization with average 0 of the linear momentum coming from the scatterings and the presence of a resonance in the system.

Before computing the Verdet constant for the FE it needs to be recalled that in the literature there are two different effects called by the same name of FE. The first one is the magneto-optical Faraday rotation or ellipticity generated in ferromagnetic materials. In this case, for an isotropic material, where ϵ^0_{xx} denotes the diagonal element of the dielectric tensor and ϵ^0_{xy} the small off-diagonal term induced by the presence of a magnetic moment oriented along the z axis, the generalized Faraday rotation is given by

$$\theta_{\rm FE}+i\,\varepsilon_{\rm FE}\approx\frac{i\pi L}{\lambda_v}\frac{\epsilon^0_{xy}}{\sqrt{\epsilon^0_{xx}}}, \tag{2}$$

where $\theta_{\rm FE}$ is the rotation of the polarization plane of the light traversing the material, $\varepsilon_{\rm FE}$ is the induced ellipticity, L is the distance the light travels inside the material, and λ_v is the wavelength of the light in vacuum. Traditionally, no Verdet constant is defined for this type of Faraday effect.

The common definition of the Verdet constant $\mathcal{V}_{\rm FE}$ applies to the second related case: the Faraday rotation of linearly polarized laser light traversing a material subjected to an externally applied magnetic field. The Verdet constant $\mathcal{V}_{\rm FE}$ for the FE is defined as the rotation of the polarization axis of linearly polarized light after traversing a unit depth of material subject to a unit of magnetic field. In the case of an isotropic material the Faraday rotation angle induced by a magnetic field along the z axis on linearly polarized light propagating along z can be computed from the dielectric tensor of the material. Again, if ϵ^0 is the unperturbed dielectric tensor of the material and $\Delta\epsilon^B_{xy}$ is the small change of the off-diagonal term of the dielectric tensor induced by the presence of the external magnetic field $\mathbf{B}=B\hat{z}$, then the generalized Faraday rotation is given by

$$\theta_{\rm FE}+i\,\varepsilon_{\rm FE}=\mathcal{V}_{\rm FE}\,L\,B\approx\frac{i\pi L}{\lambda_v}\frac{\Delta\epsilon^B_{xy}}{\sqrt{\epsilon^0_{xx}}}. \tag{3}$$

The commonly used Verdet constant is the real part of the complex Verdet constant $\mathcal{V}_{\rm FE}$ defined above. Note that Eq. (3) reduces to the more ordinarily used expression [4,5] for nearly transparent materials.

To compute the FE we compute the dielectric tensor and how it is modified by an external magnetic field. We therefore assume as external force

$$\mathbf{F}(t)=e\mathbf{E}_0\sin(\omega t)+e(d\mathbf{r}(t)/dt)\times\mathbf{B}, \tag{4}$$

where e is the electron charge, $\mathbf{E}_0$ the amplitude of the linearly polarized electric field oscillating with frequency ω, and $\mathbf{B}$ a static magnetic field. Without loss of generality we use $\mathbf{B}=B\hat{z}$. Of the solution of Eq. (1) one is interested only in the asymptotic behavior for $t\to\infty$. One can thus obtain the component of $\mathbf{r}(t)$ that oscillates with frequency ω and calculate as a function of $\mathbf{E}_0$ the electric dipole moment, using $\mathbf{P}=eN\mathbf{r}$ with N the electron density. This gives the dielectric tensor ϵ of the system. Expanding the susceptibility in series with respect to the external magnetic field we obtain for the diagonal elements

$$\epsilon_{xx}=1+\frac{Ne^2}{m\left(\omega_0^2+i\gamma\omega-\omega^2\right)}+\mathcal{O}(B^2)=\epsilon^0_{xx}+\mathcal{O}(B^2), \tag{5}$$

while the off-diagonal term ϵ_{xy} is given by

$$\epsilon_{xy} = \frac{iNe^3\omega}{m^2\left(\omega_0^2 + i\gamma\omega - \omega^2\right)^2} B + \mathcal{O}(B^3) = \Delta\epsilon_{xy}^B + \mathcal{O}(B^3). \tag{6}$$

The complex Verdet constant can then be written using Eq. (3) as

$$\mathcal{V}_{\mathrm{FE}} \approx -\frac{\pi Ne^3}{m^2\lambda_v n_0} \frac{\omega}{\left(\omega_0^2 + i\gamma\omega - \omega^2\right)^2}, \tag{7}$$

where we used $n_0 = \sqrt{\epsilon_{xx}^0(\omega)}$.

The IFE on the other hand is obtained by using in Eq. (1) circularly polarized light as external electric force (in SI units)

$$\mathbf{F}(t) = eE_0[\hat{x}\sin(\omega t) + \hat{y}\cos(\omega t)]. \tag{8}$$

Again one looks for the asymptotic response and can compute the magnetization simply as

$$\mathbf{M} = \frac{e}{2}\mathbf{r}(t) \times \frac{d\mathbf{r}(t)}{dt}. \tag{9}$$

Asymptotically, for $t \to \infty$, the electron moves on an elliptic trajectory. The constant component of the asymptotic behavior of $\mathbf{M}$ gives the amplitude of the magnetization induced by the circularly polarized light as

$$\mathbf{M}_{\mathrm{IFE}} = -\frac{Ne^3}{2m^2} \frac{\omega}{\left(\omega^2 - \omega_0^2\right)^2 + \gamma^2\omega^2} E_0^2. \tag{10}$$

It is now possible to provide the expression for the Verdet constant of the IFE by using its definition

$$\mathbf{M}_{\mathrm{IFE}} = \frac{\lambda_v n_0}{2\pi} \mathcal{V}_{\mathrm{IFE}} E_0^2, \tag{11}$$

where $n_0 = \sqrt{\epsilon_{xx}^0(\omega)}$ is the refractive index in the absence of the external field. This leads to the expression for the Verdet constant of the IFE

$$\mathcal{V}_{\mathrm{IFE}} = -\frac{\pi Ne^3}{m^2\lambda_v n_0} \frac{\omega}{\left(\omega^2 - \omega_0^2\right)^2 + \gamma^2\omega^2}. \tag{12}$$

Taking the difference of the two complex constants one obtains

$$\frac{\mathcal{V}_{\mathrm{IFE}} - \mathcal{V}_{\mathrm{FE}}}{\mathcal{V}_{\mathrm{IFE}} + \mathcal{V}_{\mathrm{FE}}} = -\frac{i\gamma\omega}{\omega^2 - \omega_0^2}, \tag{13}$$

which is proportional to γ and hence appears only if the dissipative term is included in the treatment. Note that, even though the normalized ratio is purely imaginary, the two constants differ both in the real and the imaginary part since their sum appearing in the denominator is not purely real. As the earlier derivation of Pitaevskii [24] was based on the assumption of a dissipationless medium ($\gamma = 0$), the equivalence of the two Verdet constants was concluded. Our derivation highlights on the one hand the fundamental difference between the FE and IFE and on the other hand pinpoints that, in realistic dissipative materials, they are in general not equivalent.

Expression (10) for the induced magnetization provides additional insight in connection to recent experiments. A previous derivation [26] of the IFE for a conductive plasma predicted that the induced magnetization would scale as ω^{-3}; however, recent experiments [37] could not confirm this. Equation (10) exemplifies that in the absence of a resonance ($\omega_0 = 0$) and of dissipation ($\gamma = 0$) one obtains $\mathbf{M}_{\mathrm{IFE}} \propto \omega^{-3}$. However, such conditions are rarely met in lossy materials with plentiful interband transitions. In particular, as Eq. (13) illustrates, the difference between the FE and IFE is most pronounced near a resonance $\omega \approx \omega_0$.

III. THE OPTOMAGNETIC POLARIZATION OF THE INVERSE FARADAY EFFECT

The quantum-mechanical response to an external field interacting with a material leads to a change of related physical quantities such as electrical polarization, magnetic polarization, and population of a given level. The frequency of the response, its order in the amplitude of the perturbing field, and of course the physical observable under consideration are used to classify the phenomenon of interest. The IFE is defined as the static magnetic polarization induced in the material by the interaction with circularly polarized light and being proportional to its intensity (i.e., second order in the electric field). Following the approach introduced in Ref. [34] we will decompose the response of the system into orders of the perturbation and collect for the static term of the induced magnetic moment all contributions at the second order in the electric field of the light. The system's Hamiltonian, $\hat{\mathrm{H}}_0$, is supposed to be known and diagonalizable. The external perturbation is assumed to be due to a continuous wave that is switched on at $t = 0$,

$$\hat{V}(t) = eE_0\left[\hat{x}\cos(\omega t) + \hat{y}\sin(\omega t)\right]\Theta(t), \tag{14}$$

where e is the electronic charge, $\hat{x}$ and $\hat{y}$ the components of the position operator, E_0 the amplitude of the electric field, ω the angular frequency of the light, and $\Theta(t)$ the Heaviside function. The Laplace transform of the temporal shape $\hat{V}(t)$ of the external perturbation corresponding to circularly polarized light is

$$\hat{V}(\mathcal{E}) = -\frac{i\hbar e\,\hat{\mathbf{r}}\cdot\mathbf{E}_+}{\mathcal{E} + \hbar\omega} - \frac{i\hbar e\,\hat{\mathbf{r}}\cdot\mathbf{E}_-}{\mathcal{E} - \hbar\omega}, \tag{15}$$

where we introduced $\mathbf{E}_+ = \mathbf{E}_-^* = E_0(\hat{x} - i\hat{y})/2$ and $\mathcal{E}$ is the Laplace transform variable. The (bilateral) Laplace transform is defined through $\tilde{f}(\varepsilon) = \int_{-\infty}^{+\infty} f(t)\exp[i\frac{\varepsilon}{\hbar}t]dt$, with $\varepsilon = E + i\epsilon$, and E and ϵ real. The second-order correction to the density matrix of the system is denoted as $\hat{\rho}^{[2]}$ (t). Hence, the Laplace transformed induced magnetization is the expectation value of the magnetization operator $\hat{\mathbf{M}}$,

$$\langle\mathbf{M}^{[2]}(\mathcal{E})\rangle = \mathrm{Tr}\{\hat{\mathbf{M}}\,\hat{\rho}^{[2]}(\mathcal{E})\}, \tag{16}$$

where $\hat{\mathbf{M}} = \mu_B(g_s\hat{\mathbf{S}} + \hat{\mathbf{L}})$ is the operator for the total moment, i.e., due to the spin operator $\hat{\mathbf{S}}$ and orbital angular momentum operator, $\hat{\mathbf{L}}$. In a first step we will neglect the transient behavior, which will be treated in Sec. IV. To obtain the corresponding expression for the imparted polarization, we have to isolate the expectation value of the magnetic moment operator on the second-order static correction to the asymptotic value of the density matrix.

As discussed in Ref. [34], one can extract the static part of the second-order response, i.e., the zeroth harmonic generation (ZHG), which consists of three terms, $\hat{\rho}^{[2]}_{\mathrm{ZHG}}(\mathcal{E}) = (\hat{\rho}^{[2]}_{\mathrm{ZHG,\,o}} + \hat{\rho}^{[2]}_{\mathrm{ZHG,\,dA}} + \hat{\rho}^{[2]}_{\mathrm{ZHG,\,dB}} + \mathrm{H.c.})\frac{i\hbar}{\mathcal{E}}$. This consequently leads to three contributions for the optically induced static magnetization, $\mathbf{M}^{\mathrm{IFE}} = (\mathbf{M}^{\mathrm{IFE}}_{\mathrm{o}} + \mathbf{M}^{\mathrm{IFE}}_{\mathrm{dA}} + \mathbf{M}^{\mathrm{IFE}}_{\mathrm{dB}} + \mathrm{c.c.})E_0^2$, with

$$\mathbf{M}^{\mathrm{IFE}}_{\mathrm{o}} = e^2 \sum_{n\neq m} \langle m|\hat{\mathbf{M}}|n\rangle\langle n| \frac{{}^{\langle 2\rangle}\left[\hat{r}^+, \frac{{}^{\langle 2'\rangle}[\hat{r}^-,\hat{\rho}_0]}{\hbar^{\langle 1',3'\rangle}\check{\Omega}-\hbar\omega}\right]}{\hbar^{\langle 1,3\rangle}\check{\Omega}} |m\rangle,$$

$$\mathbf{M}^{\mathrm{IFE}}_{\mathrm{dA}} = -\,e^2 \sum_{n} \langle n|\hat{\mathbf{M}}|n\rangle\langle n| \left[\hat{r}^+, \frac{{}^{\langle 2\rangle}[\hat{r}^-,\hat{\rho}_0]}{\left(\hbar^{\langle 1,3\rangle}\check{\Omega}-\hbar\omega\right)^2}\right] |n\rangle, \tag{17}$$

$$\mathbf{M}^{\mathrm{IFE}}_{\mathrm{dB}} = -\,e^2 \sum_{n} \langle n|\hat{\mathbf{M}}|n\rangle\langle n| \frac{\left[\hat{r}^+, \frac{{}^{\langle 2\rangle}[\hat{r}^+,\hat{\rho}_0]}{\hbar^{\langle 1,3\rangle}\check{\Omega}-\hbar\omega}\right]}{2\hbar\omega} |n\rangle,$$

where $|m\rangle$ and $|n\rangle$ are eigenstates of $\hat{\mathrm{H}}_0$. Since we assumed for simplicity that the direction of propagation of the circularly polarized light is along the z axis the optical transition operators are defined as $\hat{r}^+ = (\hat{x} + i\hat{y})/2$ and $\hat{r}^- = \hat{r}^{+\dagger}$. The order notation (denoted here by superscripts $\langle\cdots\rangle$) gives the position of the operator in a product of operators; see Ref. [34]. The superoperator $\check{\Omega}$ in the formulas above is defined in Eq. (33) of Ref. [34].

The three terms above underline different physical effects. The term $\mathbf{M}^{\mathrm{IFE}}_{\mathrm{o}}$ arises from the appearance of off-diagonal terms in the perturbed density matrix due to the coherence between different levels which has been induced by the circularly polarized light. The second term $\mathbf{M}^{\mathrm{IFE}}_{\mathrm{dA}}$ is a diagonal contribution to the magnetic moment stemming from the occurring population of empty levels due to the circularly polarized light. The third term $\mathbf{M}^{\mathrm{IFE}}_{\mathrm{dB}}$ originates as well from a repopulation of levels, but it is a static diagonal contribution coming from the part of the response that leads to the second harmonic generation. Note how this term diverges for $\omega \to 0$, i.e., when the frequency of the second harmonic 2ω coincides with the field frequency ω.

We remark that the above analysis underlines that the origin of the IFE is equivalent to that of the optical rectification [38], in which an intense laser field induces an electric polarization P in a material. Both the IFE and optical rectification (cf. Ref. [34]) are second-order processes in the applied electromagnetic field, and can be described by the static contribution in the second-order density matrix, $\hat{\rho}^{[2]}_{\mathrm{ZHG}}$.

It is now possible to provide the quantum expression for the Verdet constant of the IFE by using its definition in Eq. (11). The frequency-dependent Verdet constant $\mathcal{V}_{\mathrm{IFE}}$ of the IFE is hence given by an expression identical to Eq. (17), but with the prefactor $2\pi/(\lambda_v\sqrt{\epsilon^0_{xx}})$ where the diagonal element of the dielectric tensor without the external field ϵ^0_{xx} can be explicitly computed using conventional first-order response theory [2,34] but, for sake of shortness, it will only be left indicated. The expression in Eq. (17) can be rewritten using the matrix element notation,

$$\mathbf{M}^{\mathrm{IFE}}_{\mathrm{o}} = e^2 \sum_{n\neq m;\,l} \mathbf{M}_{mn} \frac{\frac{r^+_{nl}r^-_{lm}(\rho^0_{mm}-\rho^0_{ll})}{E_l-E_m+i\hbar\Gamma_{lm}-\hbar\omega} - \frac{r^-_{nl}r^+_{lm}(\rho^0_{ll}-\rho^0_{nn})}{E_n-E_l+i\hbar\Gamma_{nl}-\hbar\omega}}{E_n - E_m + i\hbar\Gamma_{nm}},$$

$$\mathbf{M}^{\mathrm{IFE}}_{\mathrm{dA}} = e^2 \sum_{n,l} \mathbf{M}_{nn} \left(\frac{r^+_{nl}r^-_{ln}\left(\rho^0_{ll} - \rho^0_{nn}\right)}{(E_l - E_n + i\hbar\Gamma_{ln} - \hbar\omega)^2} + \frac{r^-_{nl}r^+_{ln}\left(\rho^0_{ll} - \rho^0_{nn}\right)}{(E_n - E_l + i\hbar\Gamma_{nl} - \hbar\omega)^2} \right), \tag{18}$$

$$\mathbf{M}^{\mathrm{IFE}}_{\mathrm{dB}} = e^2 \sum_{n,l} \frac{\mathbf{M}_{nn}r^+_{nl}r^+_{ln}\left(\rho^0_{nn} - \rho^0_{ll}\right)(i\hbar\Gamma_{ln} - \hbar\omega)}{\hbar\omega[(E_l - E_n)^2 + (\hbar\Gamma_{ln} + i\hbar\omega)^2]},$$

where the matrix elements are defined as $r^{\pm}_{nm} = \langle n|\hat{r}^{\pm}|m\rangle$. The matrix Γ in the formulas above is defined in Eq. (29) of Ref. [34]; it takes into account the dephasing due to the open nature of the quantum system. Its matrix elements Γ_{nm} are positive parameters which are usually determined phenomenologically and often approximated to be state independent.

Expression (18) is suitable for implementation in *ab initio* band-structure calculations. For comparison with previous work, we mention that a classical treatment for a nonabsorbing medium gave [24]

$$\mathbf{M}_{\mathrm{IFE}} = \frac{\mathrm{Im}\left[\epsilon^0_{xy}\right]}{4\pi} E_0^2, \tag{19}$$

with $\epsilon^0_{xy}(\omega)$ the off-diagonal dielectric tensor component responsible for the magneto-optical Faraday effect and where Im[ ] stands for the imaginary part. For both the classical nonabsorbing expression (19) and the general quantum case (18) the laser-induced optomagnetic field, which has been used in recent spin-dynamics simulations [14,16], can be obtained from $\mathbf{H} = \mathbf{M}/\chi$, with χ the material's static magnetic susceptibility.

IV. THE INVERSE FARADAY EFFECT TRIGGERED BY FEMTOSECOND LASER PULSES

In the above formulation we exploited the static part of the second-order density matrix to compute the adiabatically induced magnetic polarization. An extension of this formulation to the transient magnetic polarization induced by a fs laser pulse can be obtain by employing the time-dependent second-order density matrix. We have already shown in Ref. [34] how it is possible to obtain an explicit expression for the temporal evolution of the system as a response at the second order. In that treatment, the modeling of the laser pulse was somewhat simplified, as it was assumed that the pulse resulted from a stepwise on-switching of the electromagnetic field, and a stepwise off-switching of the same after a time T. Here we aim to present a more sophisticated approach which will lead to better results without increasing the computational effort. To this end we use a more physically meaningful temporal shape of the pump pulse. In spite of the fact that this will lead to an apparently more complex expression, the same number of matrix elements as for the simpler approach have to be numerically computed while the time evolution is derived completely analytically.

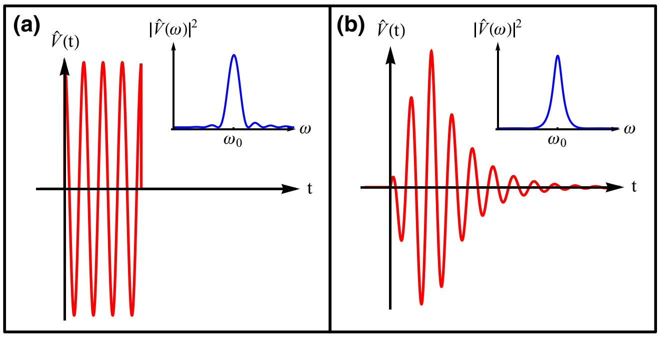

FIG. 1. (Color online) Temporal shapes of the perturbing femtosecond laser pulses. The shape used in Ref. [34] for a block-shaped perturbative pulse is shown in (a). The inset in (a) shows its Fourier transform. In (b) the temporal shape of the pulse as given by Eq. (20) is shown and the inset shows its Fourier transform.

The temporal shape for a perturbation pulse used in Ref. [34] is shown in Fig. 1(a): the perturbation is abruptly switched on at time 0 and switched off at time T. The advantage is that the Laplace transform has a very simple shape and leads to simple analytical formula for the response. The disadvantage is that the spectrum is unphysically broad: in the inset in Fig. 1(a) the Fourier transform of the temporal shape of the perturbing potential shows finite spectral weight at frequencies farther from the central frequency ω_0. At the second order, interference between these frequencies and the central wavelength of the light arises. To avoid this unphysical effect we propose a better approximation for the temporal shape of the perturbing pulse:

$$\hat{V}\,(\mathcal{E}) = \sum_{j=\pm 1} \frac{\hbar^2\gamma\big(e^{\frac{i(\mathcal{E}+j\hbar\omega_0)T}{\hbar}} - 1\big)}{(\mathcal{E}+j\hbar\omega_0)\,(\mathcal{E}+j\hbar\omega_0+i\gamma\hbar)}\hat{V}_j, \tag{20}$$

where $\hat{V}_j = -e\hat{\mathbf{r}}\cdot E_0(\hat{x} - j\,i\hat{y})/2$, ω_0 is the central frequency of the laser, T is the length of the pulse, and γ gives how fast the time profile reaches the maximum after the switching on and controls the bandwidth. This temporal shape is shown in Fig. 1(b), and its Fourier spectrum is shown in the inset.

Explicit expressions for the response of the system can be now analytically derived. The first-order correction to the density matrix induced by the laser pulse in Eq. (20) is

$$\hat{\rho}_P^{[1]}(\mathcal{E}) = \sum_{j=\pm 1} \frac{{}^{\langle 2\rangle}[\hat{V}_j,\hat{\rho}_0]}{\mathcal{E} - \hbar^{\langle 1,3\rangle}\check{\Omega}}\,\frac{\hbar^2\gamma\big(e^{\frac{i(\mathcal{E}+j\hbar\omega)T}{\hbar}} - 1\big)}{(\mathcal{E}+j\hbar\omega)\,(\mathcal{E}+j\hbar\omega+i\hbar\gamma)}. \tag{21}$$

The second order, in spite of being a lengthier expression, can still be explicitly written; this response is given in Eq. (A1) of the Appendix. It is then possible to compute the expectation value of the magnetization operator $\hat{\mathbf{M}}$. Note that the coefficients in Eq. (A1) can be computed numerically, but no numerical integration of the time dependence is required since the complete time evolution has already been obtained analytically. This gives a double advantage: (1) it is possible to have an arbitrary time resolution without an increase in computational cost and, more importantly, (2) the computational cost is equivalent to the one of a static second-order effect, which would make these calculations feasible for real materials and not only model systems. In Fig. 2 we report a typical behavior of the second-order optomagnetic polarization induced by circularly polarized light, and computed, as explained in the following paragraph, for a hydrogenlike atom.

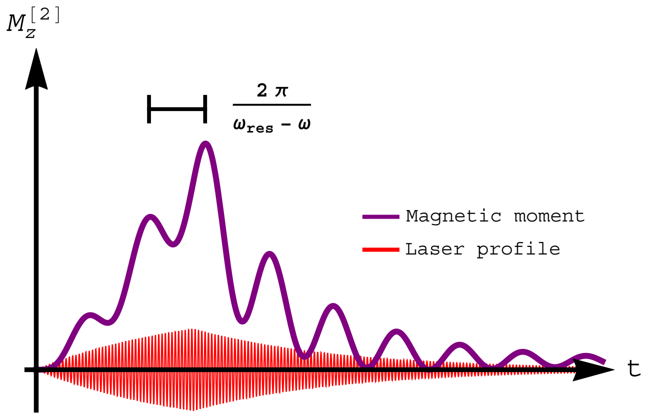

FIG. 2. (Color online) Calculated shape of the optomagnetic polarization response to a pulse in arbitrary units.

The amplitude of the peak increases the closer the frequency of the laser pulse is to the resonance [as expressed in Eq. (22) below]. Conversely, the oscillations have a frequency given by the detuning $\omega_{\rm res} - \omega$ (see Fig. 2) and an amplitude that increases with increasing oscillation frequency $\omega_{\rm res} - \omega$.

V. THE INVERSE FARADAY EFFECT FOR HYDROGENLIKE ATOMS

We report here actual calculations for the IFE in the simplest real system: an atom with hydrogenlike wave functions. Because of their elemental structure, they have been chosen as first examples of numerical application of the expressions derived so far. Obviously, this represents a very simplistic system and the study of real materials is required to make material-dependent predictions. However, this case can provide a basic physical picture to extract trends, albeit with limitations. We study here the case of the valence electron being in different hydrogenlike levels.

The behavior of the IFE with the frequency ω of the circularly polarized laser can be extracted immediately. If $\hbar\omega_{\rm res}$ is the energy difference between the energy of the valence state and the first conduction state, then

$$\mathcal{V}_{\rm IFE} \propto \frac{1}{(\omega - \omega_{\rm res})^2}, \tag{22}$$

which means that even though the IFE is enhanced by being close to resonance, it does not vanish away from the resonance.

Similarly, optical absorption can be present even at frequencies that are off resonance. The energy window where absorption is still present is proportional to the dephasing factor appearing in the superoperator $\check{\Omega}$ as defined in Eq. (33) of Ref. [34]. Note that the mentioned dephasing factor is the same broadening as conventionally used in the Kubo formula for the optical conductivity [2]. Conversely, the decay of the IFE away from resonance is not dependent on the dephasing

TABLE I. Amplitude (in arbitrary units) of the IFE for atoms with hydrogenlike valence electron depending on the orbital character of the wave function of the last occupied state. The left-hand columns give the amplitude of the effect for the case in which no spin-orbit (SO) interaction is taken into account. The right-hand columns give the values for the case in which SO is included in the wave function of the valence states.

	$\mathcal{V}_{\mathrm{IFE}}\,(\omega-\omega_{\mathrm{res}})^2$					
	no SO			with SO		
	$l=0$	$l=1$	$l=2$	$l=0$	$l=1$	$l=2$
$n=1$	1.00			1.19		
$n=2$	5.65	9.16		6.74	9.29	
$n=3$	17.90	24.10	38.06	21.42	24.39	38.06

times. By choosing a material with a small broadening at the lowest absorption energy, one could try to tune the frequency close enough to resonance to have a significant amplification of the IFE, but far enough to have negligible absorption.

In Table I we provide the value of the Verdet constant $\mathcal{V}_{\mathrm{IFE}}$ multiplied by $(\omega-\omega_{\mathrm{res}})^2$ to exclude the already treated behavior in Eq. (22) and normalized to the value obtained for the case that the only occupied state is the 1s orbital. The two parts of the table refer to calculations done with hydrogen wave functions without SO and with hydrogen wave functions modified by the inclusion of SO. Each row gives the principal quantum number n of the orbital occupied by the valence electrons, while columns pertain to the different orbital quantum number l. In the calculations all the states at higher n's are considered empty and allowed for the transitions.

To extract trends, the behavior of the IFE coefficient has been studied for modified hydrogenlike wave functions. Using the Bohr radius a_0 as a parameter in the analytical expression of the hydrogen wave functions, we obtain wave functions having the same angular character and same radial dependence, but that are either contracted or expanded (since for a given orbital the average radius is $\langle \hat{r}_{e-n}\rangle \propto a_0$). This can be used as a first approximation of the influence of the contraction of valence states for heavy elements. The analytical expression (18) of the IFE suggests a quadratic dependence on the Bohr radius used as a parameter; i.e., the wider the orbit is the more effective the IFE becomes.

Considering various n and l quantum numbers in the calculations we find that the IFE Verdet constant shows a slightly more than linear dependence on the average electron radius $\langle \hat{r}_{e-n}\rangle$ [with $\langle \hat{r}_{e-n}\rangle \propto 3n^2 - l(l+1)$], instead of the expected quadratic one. This suggests that the presence of nodes in the wave function reduces the efficiency of the IFE. Needless to say, the here-indicated trend requires further investigation in future studies of periodic solids.

A final remark can be made on the effect of SO. Its inclusion always enhances the IFE even if, in the case of hydrogenlike states, its effect is small compared to other contributions. The increase of the Verdet constant $\mathcal{V}_{\mathrm{IFE}}$ due to SO is proportional to the SO splitting of the levels. It is thus small and nonnegligible only when the first empty states are very close to the nucleus. This is expected since we are studying hydrogenlike atoms. In more complex materials the SO coupling is expected to play a larger role. As is well known, this small coupling of spin and orbital momentum gives rise to the magneto-optical FE. In the case of the IFE of real materials future *ab initio* calculations will be required to shed light on its precise dependence on the SO interaction.

VI. COMPARISON OF THE FARADAY AND INVERSE FARADAY EFFECTS

It is instructive to consider the FE with the aim of comparing its Verdet constant $\mathcal{V}_{\mathrm{FE}}$ with the one of the IFE, $\mathcal{V}_{\mathrm{IFE}}$. We recall that the Verdet constant is defined for the Faraday rotation induced to the light's polarization plane upon traversing a material to which an external magnetic field has been applied. In the following we derive ϵ^0_{xx} and $\Delta\epsilon^B_{xy}$ within the theoretical framework of Ref. [34] and compute $\mathcal{V}_{\mathrm{FE}}$ from Eq. (3).

We have to compute the response of a system with Hamiltonian $\hat{\mathrm{H}}_0$ to the perturbation $\hat{V}\,(t)$ coming from the simultaneous application of an external static magnetic field and a linearly polarized laser field. The Laplace transform of $\hat{V}\,(t)$ is

$$\hat{V}\,(\mathcal{E}) = \hat{V}_M \frac{i\hbar}{\mathcal{E}} + \hat{V}_L\left(\frac{i\hbar}{\mathcal{E}+\hbar\omega} + \frac{i\hbar}{\mathcal{E}-\hbar\omega}\right), \tag{23}$$

where $\hat{V}_M = -\hat{\mathbf{M}}\cdot\mathbf{B}$, with $\mathbf{B}$ the external magnetic field, and $\hat{V}_L = -\mathrm{e}\hat{\mathbf{r}}\cdot\mathbf{E}/2$.

Next, we compute the time evolution of the system at the first order. Transient terms of the expansion are neglected since the Verdet constant does not include effects coming from the non-adiabatic switching on of the magnetic field or the probing laser. At the first order the asymptotic responses to the laser and to the external magnetic field do not interfere and give two contributions to the response that simply add up:

$$\hat{\rho}^{[1]}_{\mathrm{ASY}}\,(\mathcal{E}) = \hat{\rho}^{[1]}_{\mathrm{Zeeman}} \frac{i\hbar}{\mathcal{E}} + \hat{\rho}^{[1]}_{\mathrm{FHG}}\left(\frac{i\hbar}{\mathcal{E}+\hbar\omega} + \frac{i\hbar}{\mathcal{E}-\hbar\omega}\right), \tag{24}$$

where the Zeeman effect is given by

$$\hat{\rho}^{[1]}_{\mathrm{Zeeman}}\,(\mathcal{E}) = -\frac{{}^{\langle 2\rangle}[\hat{V}_M,\hat{\rho}_0]}{\hbar\,{}^{\langle 1,3\rangle}\check{\Omega}}\,\frac{i\hbar}{\mathcal{E}}, \tag{25}$$

and the first harmonic generation (FHG) of the laser,

$$\hat{\rho}^{[1]}_{\mathrm{FHG}}\,(\mathcal{E}) = -\sum_{j=\pm 1} \frac{{}^{\langle 2\rangle}[\hat{V}_L,\hat{\rho}_0]}{\hbar\,{}^{\langle 1,3\rangle}\check{\Omega} + j\hbar\omega}\,\frac{i\hbar}{\mathcal{E}+j\hbar\omega}, \tag{26}$$

is the part of the response used to compute the dielectric tensor without the perturbation coming from the external magnetic field (see Ref. [34] for details).

At the second order the electric and the magnetic parts of the perturbation can interfere with each other:

$$\begin{aligned}\hat{\rho}^{[2]}_{\mathrm{ASY}}\,(\mathcal{E}) = {} & \hat{\rho}^{[2]}_{\mathrm{Zeeman}^2}\frac{i\hbar}{\mathcal{E}} + \hat{\rho}^{[2]}_{\mathrm{ZHG}}\frac{i\hbar}{\mathcal{E}} + \hat{\rho}^{[2]}_{\mathrm{ABS}}\left(\frac{i\hbar}{\mathcal{E}}\right)^2 \\ & + \hat{\rho}^{[2]}_{\mathrm{SHG}}\frac{i\hbar}{\mathcal{E}+2\hbar\omega} + \hat{\rho}^{[2]\dagger}_{\mathrm{SHG}}\frac{i\hbar}{\mathcal{E}-2\hbar\omega} \\ & + \hat{\rho}^{[2]}_{\mathrm{INT}}\frac{i\hbar}{\mathcal{E}+\hbar\omega} + \hat{\rho}^{[2]\dagger}_{\mathrm{INT}}\frac{i\hbar}{\mathcal{E}-\hbar\omega}.\end{aligned} \tag{27}$$

We can hence distinguish second-order responses to the magnetic field or to the light alone and an interference term. To second order in the perturbation $\hat{V}_M$ we obtain the second-order Zeeman effect:

$$\hat{\rho}^{[2]}_{\mathrm{Zeeman}^2}(\mathcal{E}) = \frac{{}^{\langle 2\rangle}\left[\hat{V}_M, \frac{{}^{\langle 2'\rangle}[\hat{V}_M,\hat{\rho}_0]}{\hbar^{\langle 1',3'\rangle}\check{\Omega}}\right]}{\hbar^{\langle 1,3\rangle}\check{\Omega}} \frac{i\hbar}{\mathcal{E}}. \tag{28}$$

At the second order in $\hat{V}_L$ we find the second harmonic generation (SHG) with poles in $2\hbar\omega$ and $-2\hbar\omega$, the zero-th harmonic generation with pole in 0, and the absorption with a double pole in 0. All these contributions to the response are identical to the case without the external magnetic field [34]. The remaining second-order asymptotic term (first order in the perturbation $\hat{V}_M$ and first in $\hat{V}_L$) has poles in $\hbar\omega$ and $-\hbar\omega$ and is given by

$$\hat{\rho}^{[2]}_{\mathrm{INT}} = \frac{{}^{\langle 2\rangle}\left[-\hat{\mathbf{M}}\cdot\mathbf{B}, \frac{{}^{\langle 2'\rangle}[-e\frac{\hat{\mathbf{r}}\cdot\mathbf{E}}{2},\hat{\rho}_0]}{\hbar^{\langle 1',3'\rangle}\check{\Omega}+\hbar\omega}\right]}{\hbar^{\langle 1,3\rangle}\check{\Omega}} + \frac{{}^{\langle 2\rangle}\left[-e\frac{\hat{\mathbf{r}}\cdot\mathbf{E}}{2}, \frac{{}^{\langle 2'\rangle}[-\hat{\mathbf{M}}\cdot\mathbf{B},\hat{\rho}_0]}{\hbar^{\langle 1',3'\rangle}\check{\Omega}}\right]}{\hbar^{\langle 1,3\rangle}\check{\Omega}+\hbar\omega}. \tag{29}$$

We now recall that the dielectric tensor represents the proportionality constant between the oscillating electric field and the electric polarization induced in the material and oscillating with the same frequency. Since $\hat{\rho}^{[2]}_{\mathrm{INT}}$ oscillates at the same frequency of the laser it gives a contribution to the dielectric tensor. One can thus write

$$\begin{aligned}\epsilon_{\alpha\beta} = \epsilon^0_{\alpha\beta} + \Delta\epsilon^B_{\alpha\beta} &= \mathrm{Tr}\big\{-e\,\hat{r}_\alpha\,\hat{\rho}^{[1]}_{\mathrm{FHG}}(E_\beta)\big\}/E_\beta \\ &+ \mathrm{Tr}\big\{-e\,\hat{r}_\alpha\,\hat{\rho}^{[2]}_{\mathrm{INT}}(E_\beta,\mathbf{B})\big\}/E_\beta,\end{aligned} \tag{30}$$

where $\alpha,\beta = x,y$, or z stand for Cartesian components of the matrix or vector. The change in the dielectric tensor induced by the magnetic field is

$$\begin{aligned}\Delta\epsilon^B_{\alpha\beta} = -\frac{e^2}{2}\Bigg(&\mathrm{Tr}\left\{\hat{r}_\alpha \frac{{}^{\langle 2\rangle}\left[\hat{r}_\beta, \frac{{}^{\langle 2'\rangle}[\hat{\mathbf{M}}\cdot\mathbf{B},\hat{\rho}^{[0]}_{th}]}{\hbar^{\langle 1',3'\rangle}\check{\Omega}}\right]}{\hbar^{\langle 1,3\rangle}\check{\Omega}+\hbar\omega}\right\} \\ &+ \mathrm{Tr}\left\{\hat{r}_\alpha \frac{{}^{\langle 2\rangle}\left[\hat{\mathbf{M}}\cdot\mathbf{B}, \frac{{}^{\langle 2'\rangle}[\hat{r}_\beta,\hat{\rho}^{[0]}_{th}]}{\hbar^{\langle 1',3'\rangle}\check{\Omega}+\hbar\omega}\right]}{\hbar^{\langle 1,3\rangle}\check{\Omega}}\right\}\Bigg).\end{aligned} \tag{31}$$

It is now possible to compute the complex Verdet constant (for simplicity we assume again that the external magnetic field is along z, the linear polarization of the light is along x, and the measured Faraday rotation is in the x-y plane):

$$\begin{aligned}\mathcal{V}_{\mathrm{FE}} = \frac{i\pi e^2\mu_B\hbar^2}{2m^2\lambda_v\sqrt{\epsilon^0_{xx}}}\sum_{n,m,l}\Bigg[&\frac{x_{mn}\left(\frac{y_{nl}M^z_{lm}(\rho^0_m-\rho^0_l)}{E_l-E_m+i\hbar\Gamma_{lm}} - \frac{y_{lm}M^z_{nl}(\rho^0_l-\rho^0_n)}{E_n-E_l+i\hbar\Gamma_{nl}}\right)}{E_n-E_m+i\hbar\Gamma_{nm}+\hbar\omega} \\ &+ \frac{x_{mn}\left(\frac{y_{lm}M^z_{nl}(\rho^0_m-\rho^0_l)}{E_l-E_m+i\hbar\Gamma_{lm}+\hbar\omega} - \frac{y_{nl}M^z_{lm}(\rho^0_l-\rho^0_n)}{E_n-E_l+i\hbar\Gamma_{nl}+\hbar\omega}\right)}{E_n-E_m+i\hbar\Gamma_{nm}}\Bigg],\end{aligned} \tag{32}$$

where we have rewritten the formula in the usual notation. For *ab initio* calculations it can be rewritten in terms of the canonical momenta operators using the relation $\hat{\Pi}_\alpha = \frac{m}{i\hbar}[\hat{H}_0,\hat{r}_\alpha]$.

One should note how the expressions in Eqs. (17)—together with Eq. (11)—and (32) are different. This emphasizes that a quantum treatment shows that there exists a fundamental difference between the Verdet constants of the IFE and of the FE. To obtain detailed information on what the values of the two Verdet constants for a specific material would be, *ab initio* calculations are required which are beyond the scope of the present work. Nonetheless, insight in the origin of their difference could already be provided on the classical level of the Drude-Lorentz theory. As shown in Sec. II, already at the level of the Drude-Lorentz theory the difference of the two constants is evident, and can be ascribed to the presence of dissipation within the material. The inequality of the Verdet constants in the presence of dissipation is consistent with a recent experiment [35], in which differences between the IFE and FE were observed for absorbing materials. Also, it has been noted recently that the classical expression for the IFE did not match well to helicity-dependent measurements [36].

VII. CONCLUSIONS AND OUTLOOK

We have investigated the quantum origin of the optomagnetic polarization that is induced by circularly polarized light in a material. Using perturbation theory to second order in the radiation field, we have evaluated second-order corrections to the density matrix, from which analytical expressions for the optomagnetic polarization have been derived. This has been done for two cases of interest, the optically induced magnetization due to continuous radiation, and the transient magnetization induced by a short laser pulse. We have further derived the most general quantum mechanical expressions for the Verdet constants of the IFE and of the FE. Comparing these expressions we have observed that the two effects are not equal. Their difference is further exemplified by a classical derivation within the Drude-Lorentz theory, which predicts equivalence of the the two effects only in the absence of dissipation.

The derived expressions for the optomagnetic polarization Eq. (18), which is proportional to the Verdet constant of the IFE, and the Verdet constant of the FE of a paramagnetic material, Eq. (32), have a wide applicability range to real materials. In a similar way as the Kubo linear-response formula for the frequency-dependent dielectric tensor or the FE these quantities can be computed using *ab initio* methods. The obtained expressions are lengthy, but their implementation in a DFT-based band-structure code appears straightforward. In particular, the transient optomagnetic polarization is provided without requiring the numerical integration of the full time dependence of the electronic system. Finally, we have shown how to analytically derive the time response to a perturbation pulse, making the analysis of fs response straightforwardly, since the time evolution is already explicitly given and only the materials-dependent coefficients and poles need to be computed numerically.

ACKNOWLEDGMENTS

We thank K. Carva, G. P. Brivio, G. Benedek, and G. Fratesi for valuable discussions. This work has been supported by the European Community's Seventh Framework Programme (FP7/2007-2013) under Grant Agreement No. 214810, "FANTOMAS," and No. 281043, "FemtoSpin," and the Swedish National Infrastructure for Computing (SNIC). G.B. acknowledges support from the C. M. Lerici Foundation.

APPENDIX: SECOND-ORDER RESPONSE TO LASER PULSE

We report here the explicit expression of the second-order response to a fs laser pulse:

$$
\begin{aligned}
\hat{\rho}_P^{[2]}(\mathcal{E}) = i\hbar \sum_{\substack{j=\pm 1\\ l=\pm 1}} \frac{1}{\mathcal{E}-\hbar^{\langle 1,3\rangle}\check{\Omega}} \Bigg(& i\hbar\gamma\, {}^{\langle 2\rangle}\!\left[\hat{V}_l, \frac{{}^{\langle 2'\rangle}[\hat{V}_j,\hat{\rho}_0]\big(1-e^{\frac{i(\mathcal{E}+l\hbar\omega-\hbar^{\langle 1',3'\rangle}\check{\Omega})}{\hbar}T}\big)}{(j\hbar\omega+\hbar^{\langle 1',3'\rangle}\check{\Omega})(j\hbar\omega+i\hbar\gamma+\hbar^{\langle 1',3'\rangle}\check{\Omega})(\mathcal{E}+l\hbar\omega-\hbar^{\langle 1',3'\rangle}\check{\Omega})}\right] \\
& - i\hbar\gamma\, {}^{\langle 2\rangle}\!\left[\hat{V}_l, \frac{{}^{\langle 2'\rangle}[\hat{V}_j,\hat{\rho}_0]\big(1-e^{\frac{i(\mathcal{E}+l\hbar\omega-\hbar^{\langle 1',3'\rangle}\check{\Omega})}{\hbar}T}-e^{\frac{i(\mathcal{E}+(j+l)\hbar\omega+i\hbar\gamma)}{\hbar}T}+e^{\frac{i(\mathcal{E}+(j+l)\hbar\omega)}{\hbar}T}\big)}{(j\hbar\omega+\hbar^{\langle 1',3'\rangle}\check{\Omega})(j\hbar\omega+i\hbar\gamma+\hbar^{\langle 1',3'\rangle}\check{\Omega})(\mathcal{E}+l\hbar\omega+i\hbar\gamma-\hbar^{\langle 1',3'\rangle}\check{\Omega})}\right] \\
& - {}^{\langle 2\rangle}\!\left[\hat{V}_l, \frac{{}^{\langle 2'\rangle}[\hat{V}_j,\hat{\rho}_0]}{(j\hbar\omega+\hbar^{\langle 1',3'\rangle}\check{\Omega})}\right]\left(\frac{1-e^{\frac{i(\mathcal{E}+(j+l)\hbar\omega)}{\hbar}T}}{\mathcal{E}+(j+l)\hbar\omega}-\frac{1-e^{\frac{i(\mathcal{E}+(j+l)\hbar\omega+i\hbar\gamma)}{\hbar}T}}{\mathcal{E}+(j+l)\hbar\omega+i\hbar\gamma}\right) \\
& + {}^{\langle 2\rangle}\!\left[\hat{V}_l, \frac{{}^{\langle 2'\rangle}[\hat{V}_j,\hat{\rho}_0]}{(j\hbar\omega+i\hbar\gamma+\hbar^{\langle 1',3'\rangle}\check{\Omega})}\right]\left(\frac{1-e^{\frac{i(\mathcal{E}+(j+l)\hbar+i\hbar\gamma\omega)}{\hbar}T}}{\mathcal{E}+(j+l)\hbar\omega+i\hbar\gamma}-\frac{1-2e^{\frac{i(\mathcal{E}+(j+l)\hbar\omega+i\hbar\gamma)}{\hbar}T}+e^{\frac{i(\mathcal{E}+(j+l)\hbar\omega)}{\hbar}T}}{\mathcal{E}+(j+l)\hbar\omega+2i\hbar\gamma}\right)\Bigg).
\end{aligned}
\tag{A1}
$$

The expression is complicated because the shape of the starting pulse is in itself complicated. Nonetheless the reader should appreciate that the whole time response is explicitly given (since the inverse Laplace transform of simple poles is straightforward) and there is no need to do any numerical time integration. The computation of the coefficients has the cost of multiplication of three matrices.

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