Nonlinear optics of optomagnetics: Quantum and classical treatments

A. Hamed Majedi [*]
Department of Electrical and Computer Engineering and Department of Physics and Astronomy, Waterloo Institute for Nanotechnology, University of Waterloo, Waterloo, Ontario, N2L 3G1, Canada

Brahim Lounis
Laboratoire Photonique, Numérique et Nanosciences (LP2N), Institut d'Optique Graduate School CNRS and Université de Bordeaux LP2N - Institut d'Optique d'Aquitaine, Rue Francois Mitterand, F-33400 Talence Cedex, France

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Optomagnetics emerges as a growing field of research cross-linking optics, magnetism, and material science. Here we provide a microscopic quantum-mechanical and a macroscopic classical model to describe an optomagnetic medium, i.e., optical gyration coefficient from a nonlinear optics point of view. Our self-consistent quantum-mechanical formulation considers all orders of perturbing field and results not only in finding generalized Pitaevskii's relationship, where photoinduced magnetization can be expanded in terms of light power, but also provides compact and analytical expressions for optical gyration vector coefficients. Classical treatment is then developed based on the anharmonic Drude-Lorentz model showing that the photoinduced DC magnetization is proportional to odd harmonics of the light power. The difference in quantum and classical results is revealed and discussed. Having a pump-probe setup in mind, we describe how a probe light signal can propagate down an optomagnetic medium, i.e., a medium that is magnetized by intense circularly polarized pump light, via its permittivity tensor and find light propagation characteristics. Inverse Faraday and Cotton-Mouton effects are discussed as a result of circular and linear birefringences and their Verdet constants have been analytically found.

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I. INTRODUCTION

Nonlinear optics continues to play a central role not only in the advancement of optical sciences and photonic technologies but also to provide a powerful tool to probe the structure and properties of materials. Initiated with the series of experiments, direct optical generation and control of magnetization revives the less-explored field of optomagnetics and recreates an exciting synergy between photonic and magnetism communities [1–5]. The origin of optomagnetics stems from a nonlinear interaction of not-linearly polarized intensive light with the orbital and spin moments of electronic structure where light angular momentum and gyration rules the generation, control, processing, and detection of magnetization in matter. The optomagnetic effect is mostly understood and explored in light of the inverse Faraday effect (IFE), i.e., the generation of static magnetization by circularly polarized light. IFE was first predicted by Pitaevskii in 1960 based on a phenomenological ansatz on a ground of a generalized Maxwell-Abraham stress tensor in a transparent dispersive medium [6]. He predicted that the static magnetization is related to the optical field intensity through an optical gyration coefficient γ in the form of

$$\mathbf{M}_{DC} = \gamma \mathbf{E} \times \mathbf{E}^*, \tag{1}$$

where $\mathbf{E}$ is the complex electric field intensity. The first experimental observation of the so-called Pitaevskii relationship was carried out by van der Ziel, Pershan, and Malmstrom, and the term inverse Faraday effect (IFE) was coined by them in 1965 [7]. They provided the quantum-mechanical model of IFE based on the effective Hamiltonian method at the low-frequency limit and justified Pitaevskii's relationship [8]. After a renewed interest in ultrafast optical control of magnetization [9], a new theoretical attempt has been initiated for quantum modeling of optomagnetics. The effective Hamiltonian method is further considered in the time domain for the Gaussian-shaped laser pulse to study transient magnetization by Popova *et al.* [10]. Taguchi and Tatara introduced the quantum-mechanical Green's function formalism to include spin and spin-orbit contributions to the photoinduced magnetization in THz frequencies. Their work explicitly revealed the equal contribution of orbital and spin magnetization in Pitaevskii's relationship [11,12]. Based on the perturbative solution of the Liouville–von Neumann equation for a generalized nonlinear light-matter interaction, Battiato *et al.* provided an exact solution of photoinduced static magnetization up to the second order in the electric field intensity [13,14]. Their density matrix formulation highlights the various physical effects arising from diagonal and off-diagonal elements due to coherence between different levels and state occupation while the dephasing is phenomenologically considered.

This paper articulates the theory of optomagnetism by focusing on its fundamental physics, finding the

[*]Also at Perimeter Institute for Theoretical Physics, Waterloo, Ontario, Canada; ahmajedi@uwaterloo.ca

generalized Pitaevskii's relationship and the light propagation in optomagnetic media. Both quantum-mechanical and classical treatments are presented in detail. We reveal the difference between quantum and classical treatments, where classical prediction gives an incomplete description from the perspective of the generalized Pitaevskii relationship. We then focus on how a weak electromagnetic field, i.e., probe signal, is copropagating down a photomagnetic medium, i.e., the medium that is magnetized by intensive circularly polarized light. Our formulation provides the permittivity tensor for the optomagnetic medium and attenuation and propagation constants for the probe signal, leading to definition of the Verdet constant of IFE and rotatory power of the inverse Cotton-Mouton effect.

II. QUANTUM-MECHANICAL THEORY OF OPTOMAGNETISM

The underlying physics of optomagnetism for an atomic system can be captured by solving the Schrödinger equation under the influence of a circularly polarized electromagnetic field. The optical field $\mathbf{E}_p(t)$ as a discrete sum of positive and negative frequency components of the pump frequency, i.e., ω_p, is considered in the form of

$$\mathbf{E}_p(t) = \mathbf{E}_p(\omega_p)e^{i\omega_p t} + \text{c.c.} \tag{2}$$

$$= \tfrac{1}{2}E_o(\omega_p)(\mathbf{x} + i\mathbf{y})e^{i\omega_p t} + \text{c.c.}, \tag{3}$$

where $\mathbf{x}, \mathbf{y}, \mathbf{z}$ are the Cartesian unit vectors and $E_o(\omega_p)$ is the real amplitude of the electric field. The atomic spinor wave function $\Psi(\mathbf{r}, t)$ is the solution to the following time-dependent Schrödinger equation:

$$i\hbar\frac{\partial}{\partial t}\Psi(\mathbf{r}, t) = \hat{H}_o\Psi(\mathbf{r}, t) - \hat{\mu}\mathbf{E}_p(t)\Psi(\mathbf{r}, t)$$

$$= \hat{H}_o\Psi(\mathbf{r}, t) - e\hat{\mathbf{r}}\mathbf{E}_p(t)\Psi(\mathbf{r}, t), \tag{4}$$

which is written in terms of the sum of the Hamiltonian $\hat{H}_o$ for a free atomic system and the dipole interaction Hamiltonian, where e is the electron charge, and $\hat{\mu}$ and $\hat{\mathbf{r}} = \mathbf{x}\hat{x} + \mathbf{y}\hat{y} + \mathbf{z}\hat{z}$ are the dipole moment and position vector operators, respectively. We seek the general solution to Eq. (4) using perturbation theory, namely, the Rayleigh-Schrödinger method, where the atomic spinor wave function can be written in the following expansion:

$$\Psi(\mathbf{r}, t) = \sum_{N=0}\lambda^N\Psi^{(N)}(\mathbf{r}, t), \tag{5}$$

and λ is the perturbation parameter set in the Hamiltonian in the form of $\hat{H}_o - \lambda\hat{\mu}\mathbf{E}_p(t)$ [15]. Assuming that the atomic system rests initially in its nondegenerate ground state, i.e., $\Psi^{(0)}(\mathbf{r}, t)$, with the energy $E_g = \hbar\omega_g$ given by

$$\Psi^{(0)}(\mathbf{r}, t) = u_g(\mathbf{r})e^{i\omega_g t}, \tag{6}$$

the remaining terms in the perturbation expansion (5) obey the following expression:

$$i\hbar\frac{\partial}{\partial t}\Psi^{(N)}(\mathbf{r}, t) = \hat{H}_o\Psi^{(N)}(\mathbf{r}, t) - e\hat{\mathbf{r}}\mathbf{E}_p(t)\Psi^{(N-1)}(\mathbf{r}, t), \tag{7}$$

where N is an integer number. Note that $u_g(\mathbf{r})$ represents the stationary ground-state spinor of the atomic system in the absence of any electromagnetic interaction. The solution to Eq. (7) can be written as the summation of the atomics' eigenfunction spinors, i.e., $u_l(\mathbf{r})$,

$$\Psi^{(N)}(\mathbf{r}, t) = \sum_l a_l^{(N)}(t)u_l(\mathbf{r})e^{i\omega_l t}, \tag{8}$$

with the time-dependent probability amplitude $a^{(N)}(t)$ as

$$a_m^{(N)}(t) = \frac{1}{i\hbar}\int_{-\infty}^{t}\sum_l a_l^{(N-1)}(t')u_l(\mathbf{r})V_{ml}(t')e^{-i\omega_{ml}t}dt', \tag{9}$$

where the interaction Hamiltonian for the circularly polarized light and two-dimensional cross section of the atomic system is

$$V_{ml}(t) = -\frac{e}{2}E_o(\omega_p)e^{i\omega_p t}\langle u_m(\mathbf{r})|\hat{x} + i\hat{y}|u_l(\mathbf{r})\rangle + \text{c.c.}$$

$$= -\frac{e}{2}E_o(\omega_p)r_{ml}e^{i\omega_p t} + \text{c.c.} \tag{10}$$

To obtain the DC magnetization based on the perturbed eigenfunction spinors, one can use the magnetization operator $\hat{\mathbf{M}}$ as

$$\langle\hat{\mathbf{M}}\rangle_{DC} = \sum_N\left\langle\Psi^{(N)}\middle|\hat{\mathbf{M}}\middle|\Psi^{(N)}\right\rangle$$

$$= \sum_{Nl}\left|a_l^{(N)*}(t)a_l^{(N)}(t)\right|_{DC}\int u_l^*(\mathbf{r})\hat{\mathbf{M}}u_l(\mathbf{r})d^3\mathbf{r}, \tag{11}$$

where $|a_l^{(N)*}(t)a_l^{(N)}(t)|_{DC}$ is the DC term in the probability density, i.e., $a_l^{(N)*}(t)a_l^{(N)}(t)$, and

$$\hat{\mathbf{M}} = \frac{Ne}{2m}(\hat{\mathbf{L}} + g_s\hat{\mathbf{S}}) \tag{12}$$

is considered as the summation of the angular momentum $\hat{\mathbf{L}}$ and spin $\hat{\mathbf{S}}$ operators [16]. In expression (12), N is the electron's number density that is exposed to the light, m is the mass of the electron, and g_s is the electron spin g factor. In order to explicitly express the photoinduced DC magnetization in terms of the optical field via the interaction Hamiltonian, i.e., Eq. (10), Eq. (11) can be written as

$$\langle\hat{\mathbf{M}}\rangle_{DC} = \mathbf{m}^{(0)} + \mathbf{m}^{(1)}|E_o(\omega_p)|^2 + \mathbf{m}^{(2)}|E_o(\omega_p)|^2|E_o(\omega_q)|^2$$

$$+ \mathbf{m}^{(3)}|E_o(\omega_p)|^2|E_o(\omega_q)|^2|E_o(\omega_r)|^2 + \cdots, \tag{13}$$

where ω_p, ω_q, ω_r, ... are the pump frequencies, and $\mathbf{m}^{(i)}$ are introduced as the ith-order optical gyration vectors. This equation explicitly shows that the photoinduced DC magnetization can be expanded as a power series of pumping intensity. The optical gyration vectors crucially depend on the detail of the dipole moment vector, pump frequencies, and the detail of eigenenergy spinors through Eq. (11). Considering the circularly polarized light, i.e., Eq. (2), propagating in the z direction and the transition dipole moment in the x-y plane, i.e., Eq. (10), the expansion of the magnetization and spin operators in the Cartesian coordinate system dictates the direction of optical gyration vectors in $\pm z$ direction. $\mathbf{m}^{(0)}$ is related to the collective orbital magnetic moment of the atomic system in its ground state in the absence of any interaction as

$$\mathbf{m}^{(0)} = \frac{Ne}{2m}\langle u_g|(\hat{\mathbf{L}} + g_s\hat{\mathbf{S}})|u_g\rangle$$

$$= \frac{Ne}{2m}\int u_g^*(\mathbf{r})(\hat{\mathbf{L}} + g_s\hat{\mathbf{S}})u_g(\mathbf{r})d^3\mathbf{r}. \tag{14}$$

The first-order optical gyration vector $\mathbf{m}^{(1)}$ is the result of the optical field interaction with pump frequency ω_p that excites the atomic system from its ground-state energy $E_g = \hbar\omega_g$ to the mth eigenenergy state $E_m = \hbar\omega_{mg} = \hbar(\omega_m - \omega_g)$ with the probability amplitude $a_m^{(1)}(t)$ and can be written as

$$\mathbf{m}^{(1)} = \frac{Ne}{2m}\left(\frac{e}{2\hbar}\right)^2 \sum_m \langle u_m|(\hat{\mathbf{L}} + g_s\hat{\mathbf{S}})|u_m\rangle |r_{mg}|^2 [D^{-1}(\omega_p - \omega_{mg}) + D^{-1}(\omega_p + \omega_{mg})], \tag{15}$$

where D is defined as a quantum-mechanical dispersion relation for positive and negative pumping frequency ω as

$$D(\omega \pm \omega_{ij}) \triangleq |(\omega \pm \omega_{ij})(\omega \pm \omega_{ij}^*)| = (\omega \pm \omega_{ij})^2 + \Gamma_{ij}^2, \tag{16}$$

and ω_{ij} is crudely updated to incorporate the damping phenomena for the transition probability between two energy bands as $\omega_{ij} = \omega_i - \omega_j - i\Gamma_{ij}$. Γ_{ij} is related to the population decay rate of the upper level i and does not represent the dephasing process or the cascaded population among the excited states. Note that $\mathbf{m}^{(1)}$ consists of the second-harmonic generation magnetization oscillating at $2\omega_p$ as well. The second-order correction to the probability amplitude can yield the second-order gyration vector $\mathbf{m}^{(2)}$ under the influence of the nondegenerate pumping frequency ω_q as

$$\begin{aligned}\mathbf{m}^{(2)} = \frac{Ne}{2m}\left(\frac{e}{2\hbar}\right)^4 \sum_n \langle u_n|(\hat{\mathbf{L}} + g_s\hat{\mathbf{S}})|u_n\rangle |r_{mg}r_{nm}|^2 \\ \times \big[D^{-1}(\omega_p - \omega_{mg})D^{-1}(\omega_p + \omega_q - \omega_{ng}) \\ + D^{-1}(\omega_p - \omega_{mg})D^{-1}(\omega_p - \omega_q - \omega_{ng}) \\ + D^{-1}(\omega_p + \omega_{mg})D^{-1}(\omega_p + \omega_q + \omega_{ng}) \\ + D^{-1}(\omega_p + \omega_{mg})D^{-1}(\omega_p - \omega_q + \omega_{ng})\big].\end{aligned} \tag{17}$$

The third-order optical gyration vector is given in volts. This procedure can be systematically applied to the Nth-order optical gyration vector. The higher-order terms in the photoinduced DC magnetization, i.e., $\langle\hat{\mathbf{M}}\rangle_{DC}$, are proportional to $(\frac{e|E_o|}{2\hbar})^{2N}$. Although the optical gyration vectors depend on the detail of materials' atomic spectra and band structures, they do not possess any symmetry restrictions. The photoinduced magnetization can thus be allowed in any materials, regardless of their electrical, magnetic, and optical properties. Knowing the fact that the intensity of circularly polarized light, expressed by Eq. (2), is proportional to its helicity, i.e., $\mathbf{z}|E_o(\omega_p)|^2 = 2i\mathbf{E}_p \times \mathbf{E}_p^*$, Eq. (13) represents the quantum-mechanical version of a generalized Pitaevskii relationship and in the case of degenerate pumping field can be written as

$$\begin{aligned}\langle\hat{\mathbf{M}}\rangle_{DC} &= \sum_{N=0} \mathbf{m}^{(N)}|(2i\mathbf{E}_p \times \mathbf{E}_p^*)|^N \\ &= \mathbf{m}^{(0)} + \mathbf{m}^{(1)}|2i\mathbf{E}_p \times \mathbf{E}_p^*| + \mathbf{m}^{(2)}|2i\mathbf{E}_p \times \mathbf{E}_p^*|^2 \\ &\quad + \mathbf{m}^{(3)}|2i\mathbf{E}_p \times \mathbf{E}_p^*|^3 + \cdots .\end{aligned} \tag{18}$$

The generalized Pitaevskii relationship in this case consists of the ground-state magnetization in the absence of any electromagnetic radiation, and the second term represents the Pitaevskii relation, i.e., Eq. (1). This equation predicts that the optomagnetic effect should be more pronounced in the materials that do not possess ground-state magnetization.

Note that the photoinduced DC magnetization crucially depends on the interaction of not-linearly-polarized light through dipole interaction r_{ml}, i.e., Eq. (10) in the case of circular polarization, with the expectation value of the magnetization operator acting on spinor eigenfunctions, $\langle u_l|(\hat{\mathbf{L}} + g_s\hat{\mathbf{S}})|u_l\rangle$. Obviously, a linearly polarized electric field does not induce any magnetization, as evidenced by the interaction Hamiltonian, i.e., Eq. (10). The photoinduced static magnetization presented in Eq. (18) can be also generalized to consider time-varying cases where the magnetization is expressed in terms of pump frequencies $\omega_p, \omega_q, \ldots$ and their harmonics. This can be done by finding the expectation value of the magnetization operators between various spinor eigenfunctions where their energy differences correspond to the harmonics of light frequencies. Similar to application of nonlinear optical susceptibility, the photomagnetic effect can be employed not only for optical processes such as harmonic generation, up/down conversion, switching, and mixing but also for probing magnetic properties of materials.

III. CLASSICAL THEORY OF OPTOMAGNETISM

The first attempt to classically treat the IFE dates back to 1975 when Zon and Kupershmidt used the Drude-Lorentz model to justify the Pitaevskii relationship [17]. This method is further considered for the free-electron gas by Hertel and Fähnle [18,19] and reused to find the Verdet constant associated with IFE by Battiato *et al.* [14]. Hereby we use the nonlinear Drude-Lorentz model based on anharmonic oscillator model to go beyond the Pitaevskii relationship and make a comparison with our quantum-mechanical treatment presented in Sec. II.

To model the photomagnetic effect, the Drude-Lorentz model is adopted in a nonlinear regime under the influence of high-intensity circularly polarized light. The local electric field will cause the average position of an electron distribution, i.e., $\mathbf{r}(t)$, to be displaced from its equilibrium. For high-intensity light, a large deviation from the average position is expected, and the electrons experience anharmonic potential in the form of $U(\mathbf{r}) = \frac{1}{2}m\omega_o^2\mathbf{r}^2 + \frac{1}{3}ma\mathbf{r}^3 + \frac{1}{4}mb\mathbf{r}^4$, where m is the mass of the electron, ω_o is the resonant frequency of the oscillator corresponding to the main observed atomic spectral line, and a, b characterize the strength of the anharmonicity [15,20]. For materials exhibiting centrosymmetric and noncentrosymmetric inversion symmetry, $a = 0$ and $b = 0$, respectively [16]. The equation of motion of the electron position can take the form

$$m\ddot{\mathbf{r}}(t) + m\Gamma\dot{\mathbf{r}}(t) + m\omega_o^2\mathbf{r}(t) + ma\mathbf{r}^2(t) + mb\mathbf{r}^3(t) = \mathbf{F}(t), \tag{19}$$

where e is the electron charge, Γ is the friction term representing the energy loss associated with the material absorption process, $\mathbf{F}(t) = e\mathbf{E}(t)$ is acting force, and $\mathbf{E}(t)$ is the vector electric field associated with the light pump in the form of

$$\mathbf{E}(t) = \mathrm{Re}\{\mathbf{E}e^{i\omega_p t}\} = \mathrm{Re}\{E_o(\mathbf{x} + i\mathbf{y})e^{i\omega_p t}\}, \tag{20}$$

where Re{.} denotes the real part of a complex function. The intensity of the circularly polarized light creates a helicity

of the wave in the plane perpendicular to its direction of propagation given by

$$\mathbf{z}|E_o|^2 = \tfrac{1}{2}i\mathbf{E}\times\mathbf{E}^*, \tag{21}$$

which is enforcing a gyrating motion on the electrons. This light-induced localized current density in the region compared to the wavelength of light leads to a magnetic moment density or magnetization as [21]

$$\mathbf{M}(t) = \frac{Ne}{2m}\mathbf{L}(t) = N\frac{e}{2}\mathbf{r}(t)\times\mathbf{v}(t), \tag{22}$$

where N is the number density of electrons exposed to light, $\mathbf{L}$ is the angular momentum, and $\mathbf{v}(t) = \dot{\mathbf{r}}$ is the average electron velocity. Note that the intrinsic angular momentum of electrons that is proportional to their spin cannot be considered in such a classical treatment.

To find the magnetization, one needs to solve the nonlinear equation (19) under the influence of the electric field based on the perturbation method analogous to that presented in Sec. II. Using expression (20), the equation of motion (19) has a solution in the form of

$$\mathbf{r}(t) = \mathrm{Re}\left\{\sum_{n=1}^{n}\mathbf{r}e^{in\omega_p t}\right\} = \mathrm{Re}\left\{\sum_{n=1}^{n}\zeta^n\mathbf{r}_e^{(n)}E_o^n e^{in\omega_p t}\right\}, \tag{23}$$

where ζ is the perturbation parameter, $\mathbf{r}_e^{(n)} = \mathbf{x}x_e^{(n)} + \mathbf{y}y_e^{(n)}$ is the nth-order solution in the frequency domain, and $y_e^{(n)} = i^n x_e^{(n)}$ due to circular polarization of the incident light. The magnetization in Eq. (22) can be written in the frequency domain

$$\begin{aligned}\mathbf{M} = \mathbf{z}\frac{Ne\omega}{4}\mathrm{Re}\Bigg\{&\sum_{n=1}^{n}x_e^{(n)}E_o^n e^{in\omega_p t}\left[\sum_{n=1}^{n}ni^{n+1}x_e^{(n)}E_o^n e^{in\omega_p t} + \sum_{n=1}^{n}n(-1)^{n+1}i^{n+1}x_e^{*(n)}E_o^{*n}e^{-in\omega_p t}\right]\\ &-\sum_{n=1}^{n}nix_e^{(n)}E_o^n e^{in\omega_p t}\left[\sum_{n=1}^{n}i^n x_e^{(n)}E_o^n e^{in\omega_p t} + \sum_{n=1}^{n}(-1)^n i^n x_e^{*(n)}E_o^{*n}e^{-in\omega_p t}\right]\Bigg\}.\end{aligned} \tag{24}$$

Equation (24) evidently shows that the photomagnetic effect is a purely nonlinear phenomenon with respect to the electric field intensity, as the DC term is proportional to the light intensity E_o^2 and its odd harmonics.

Using Eq. (21), the DC component of the magnetization can be generally written as

$$\begin{aligned}\mathbf{M}_{DC} &= \sum_{k=0}^{n}\gamma^{(2k+1)}\left(\frac{i}{2}\mathbf{E}\times\mathbf{E}^*\right)\left|\frac{i}{2}(\mathbf{E}\times\mathbf{E}^*)\right|^{2k}\\ &= \gamma^{(1)}\left(\frac{i}{2}\mathbf{E}\times\mathbf{E}^*\right) + \gamma^{(3)}\left(\frac{i}{2}\mathbf{E}\times\mathbf{E}^*\right)\left|\left(\frac{i}{2}\mathbf{E}\times\mathbf{E}^*\right)\right|^2\\ &\quad + \cdots,\end{aligned} \tag{25}$$

where the coefficients $\gamma^{(2k+1)}$ take the form

$$\gamma^{(2k+1)} = -\omega_p(2k+1)\mathrm{Re}\left\{i^{2k}\left|x_e^{(2k+1)}\right|^2\right\} \tag{26}$$

and represent the optical gyration coefficients, similar to magnetogyration coefficients [22,23]. Equation (25) clearly shows that the DC magnetization depends on the odd power of the light intensity or helicity vector irrespective of any symmetry in the structure of the material, a prediction that is an incomplete based on quantum-mechanical treatment. It is interesting to compare the optical gyration coefficients with the linear and nonlinear susceptibilities, i.e. $\chi^{(n)}$, based on the power series expansion of the electrical field for polarization, i.e. $P(t) = \epsilon_o(\chi^{(1)}E(t) + \chi^{(2)}E^2(t) + \chi^{(3)}E^3(t) + \cdots)$. We noted that the optical gyration coefficients can be expressed based on susceptibilities as follows:

$$\left|\gamma^{(2k+1)}\right| = \frac{(2k+1)\omega_p}{Ne}\left(\frac{\epsilon_o}{2^{k+1}}\right)^2\left|\chi^{(2k+1)}\right|^2. \tag{27}$$

A similar prediction as the third-order nonlinearity induced by IFE has been made in the context of magnetoplasmonic structures [24]. This classical prediction is also seen from our quantum-mechanical treatment where the optical gyration coefficients are proportional to the square of electrical dipole transition moment, i.e., Eq. (15). Equation (27) shows that materials with large linear and nonlinear susceptibilities should exhibit large optical gyration coefficients while they scale linearly with pump optical frequency and inversely with density number of electrons in the material. This fact is corroborated with the quantum-mechanical treatment where the optical gyration vectors depend on transition dipole moments similar to nonlinear $\chi^{(2)}$ and $\chi^{(3)}$ materials. Our model then predicts the optical gyration coefficients in two-dimensional materials should be larger than their bulk counterparts.

To fully consider the effect of anharmonicity of the electron's potential, i.e., nonlinear parameters a and b, the first three orders of the solution (23) can be expressed as

$$y_e^{(1)} = ix_e^{(1)} = i\frac{e}{m}\frac{1}{D(\omega_p)}, \tag{28}$$

$$y_e^{(2)} = -x_e^{(2)} = a\left(\frac{e}{m}\right)^2\frac{1}{D^2(\omega_p)D(2\omega_p)}, \tag{29}$$

and

$$y_e^{(3)} = -ix_e^{(3)} = -i\left(\frac{e}{m}\right)^3\left(\frac{2a^2}{D(2\omega_p)} - b\right)\frac{1}{D^3(\omega_p)D(3\omega_p)}, \tag{30}$$

where

$$D(\omega_p) \triangleq \omega_o^2 - \omega_p^2 + i\omega_p\Gamma \tag{31}$$

is the dispersion function of a damped harmonic oscillator. The first two orders of the optical gyration coefficients are

then

$$\gamma^{(1)} = -\left(\frac{e}{2m}\right)\epsilon_o\omega_{pl}^2\frac{\omega_p}{|D(\omega_p)|^2} \tag{32}$$

$$\gamma^{(3)} = \left(\frac{e}{2m}\right)\epsilon_o\omega_{pl}^2\left(\frac{e}{m}\right)^4\frac{3\omega_p}{|D^3(\omega_p)|^2|D(3\omega_p)|^2} \times\left|\left(\frac{2a^2}{D(2\omega_p)}-b\right)\right|^2, \tag{33}$$

where $\omega_{pl}\triangleq\sqrt{\frac{Ne^2}{m\epsilon_o}}$ is the plasma frequency of the material. Equation (32) is independent of the nonlinear coefficients a and b and is in agreement with the results in [14,18]. The classical model shows that each optical gyration coefficient is a collective response of N orbiting electrons represented by the plasma frequency ω_{pl} and the gyromagnetic ratio of electron, i.e., $\frac{e}{2m}$, which is dictated by the helicity of the pumping light and is modified by the frequency response of a classical atom as an anharmonic oscillator to the odd harmonics of pumping light intensity. This is the consequence of angular momentum conservation between light and N noninteracting electrons in the presence of an anharmonic oscillator representing atomic structure. Evidently, Eq. (25) is partially inconsistent with its quantum-mechanical counterpart, i.e., Eq. (18). The photoinduced magnetization is proportional to all harmonics of light intensity, in Eq. (18), but the even powers are absent in the classical equation (25). This fact stems from the fundamental difference in how the state of the electron is considered quantum mechanically by the wave functions and operators acting on it through momentum and angular momentum, and classically, by its position vector and its temporal derivative.

It is straightforward to justify that the magnetic field associated with the circularly polarized light, i.e., $\mathbf{B}=\mathbf{z}B_o(-i\mathbf{x}+\mathbf{y})e^{i\omega_p t}$, has no contribution to the DC magnetization in the context of our quantum-mechanical and classical treatments.

IV. LIGHT PROPAGATION IN OPTOMAGNETIC MEDIA

We are considering the propagation of a weak linearly polarized optical signal with a frequency of ω_s in an optomagnetic material where a DC magnetization is induced by the copropagation of strong, circularly polarized pump light in z direction. The photoinduced magnetic field by the pump light, $B_o=\mu_o M_{DC}$, where M_{DC} is governed by Eq. (25), breaks the directional symmetry of the linear dielectric constant for the optical signal similar to magneto-optic material, leading to linear birefringence or the Cotton-Mouton effect and circular birefringence or Faraday effect. The dependence of the imaginary part of the permittivity, i.e., ϵ_{ij}'', on B_o leads to circular birefringence or the Faraday effect, while dependence of the real part of the permittivity, i.e., ϵ_{ij}', on B_o leads to linear birefringence or the Cotton-Mouton effect [25]. These effects can be described by exploiting the Drude-Lorentz model for weak optical signals where the Lorentz force acting on the bound electrons is due to the electric field of a light signal and DC magnetic field produced by circularly polarized light. The solution of a linear version of Eq. (19), i.e., $a=b=0$, where $F(t)=e(\mathbf{x}E_x+\mathbf{y}E_y)+ev(t)\times\mathbf{z}B_o$, leads to an anisotropic relative dielectric constant that can be cast into the following tensor form:

$$\bar{\bar{\epsilon}}_r(\omega_s)=\begin{pmatrix}\epsilon_{xx}'-i\epsilon_{xx}'' & \epsilon_{xy}'-i\epsilon_{xy}'' & 0\\ \epsilon_{yx}'-i\epsilon_{yx}'' & \epsilon_{yy}'-i\epsilon_{yy}'' & 0\\ 0 & 0 & \epsilon_{zz}'-i\epsilon_{zz}''\end{pmatrix} =\begin{pmatrix}1+\frac{\omega_{pl}^2}{D_F^2(\omega_s)}D(\omega_s) & -i\omega_{pl}^2\omega_c\frac{\omega_s}{D_F^2(\omega_s)} & 0\\ i\omega_{pl}^2\omega_c\frac{\omega_s}{D_F^2(\omega_s)} & 1+\frac{\omega_{pl}^2}{D_F^2(\omega_s)}D(\omega_s) & 0\\ 0 & 0 & 1+\frac{\omega_{pl}^2}{D(\omega_s)}\end{pmatrix}, \tag{34}$$

where

$$D_F^2(\omega_s)\triangleq\omega_o^2-\left(1+\omega_c^2\right)\omega_s^2+i\omega_s\Gamma=D^2(\omega_s)-\omega_c^2\omega_s^2 \tag{35}$$

is the modified dispersion function due to the presence of magnetic field, and $\omega_c=\frac{eB_o}{m}=\frac{\mu_o e}{m}M_{DC}$ is the cyclotron frequency that is induced by the circularly polarized pump light. Note that the z axis is not affected by the photoinduced magnetic field. By expanding the complex permittivity elements in series with respect to the photoinduced magnetic field B_o, we obtain the real and complex parts of the permittivity elements

$$\epsilon_r'=\epsilon_{xx}'=\epsilon_{yy}'\approx 1+\frac{\omega_{pl}^2}{|D(\omega_s)|^2}\left(\omega_o^2-\omega_s^2\right) \tag{36}$$

$$\epsilon_{xx}''=\epsilon_{yy}''\approx\frac{\omega_{pl}^2\Gamma}{|D(\omega_s)|^2}\omega_s \tag{37}$$

$$\epsilon_{xy}'=-\epsilon_{yx}'\approx-2\omega_{pl}^2\Gamma\frac{\omega_s^2\left(\omega_o^2-\omega_s^2\right)}{|D(\omega_s)|^4}\omega_c \tag{38}$$

$$\epsilon_{xy}''=-\epsilon_{yx}''\approx\omega_{pl}^2\frac{\left(\omega_o^2-\omega_s^2\right)^2-\omega_s^2\Gamma^2}{|D(\omega_s)|^4}\omega_s\omega_c \tag{39}$$

$$\epsilon_{zz}'=1+\frac{\omega_{pl}^2}{|D(\omega_s)|^2}\left(\omega_o^2-\omega_s^2\right) \tag{40}$$

$$\epsilon_{zz}''=\frac{\omega_{pl}^2\Gamma}{|D(\omega_s)|^2}\omega_s. \tag{41}$$

It is worth noting that the Onsager symmetry of the permittivity, i.e., $\epsilon_{ij}'(\omega_s,B_o)=\epsilon_{ji}'(\omega_s,-B_o)$ and $\epsilon_{ij}''(\omega,B_o)=-\epsilon_{ji}'(\omega,-B_o)$, holds, but Hermiticity of the dielectric constant, i.e., $\epsilon_{ij}(\omega_s,B_o)=\epsilon_{ji}^*(\omega_s,B_o)$, is valid where the damping factor Γ or absorption is absent. Any media described by the anisotropic permittivity tensor, i.e., Eq. (34), has two normal propagation modes with relative permittivities $(\epsilon_{xx}'\pm\epsilon_{xy}'')-i(\epsilon_{xx}''\mp\epsilon_{xy}')$. The permittivity tensor is then diagonalized in the coordinate systems with orthogonal unit vectors $\mathbf{e}_\pm=\frac{1}{\sqrt{2}}(\mathbf{x}\pm i\mathbf{y})$ and $\mathbf{z}$ represented by uniaxial optical symmetry. The complex propagation constants, $\alpha_{1,2}+i\beta_{1,2}$, for an optical signal are

$$\alpha_{1,2}+i\beta_{1,2}\approx\frac{k_o}{2\sqrt{\epsilon_r'}}(\epsilon_{xx}''\pm\epsilon_{xy}')\left(1\pm\frac{\epsilon_{xy}''}{2\epsilon_r'}\right) +ik_o\sqrt{\epsilon_r'}\left(1\mp\frac{\epsilon_{xy}''}{2\epsilon_r'}\right), \tag{42}$$

where $k_o=\frac{2\pi}{\lambda_o}$ is the free-space wave number in terms of wavelength λ_o. The ability of the optomagnetic medium to

rotate the linear polarization of an optical signal that leads to circular birefringence is commonly referred to as the inverse Faraday effect and can be found by its rotatory power ρ_{xy}, similar to the magneto-optic media, which is defined by the rotation angle per unit length as

$$\rho_{xy} = \frac{\beta_1 - \beta_2}{2} \approx -\frac{\pi}{\lambda_o}\frac{\epsilon''_{xy}}{\sqrt{\epsilon'_r}}. \tag{43}$$

The rotatory power is a linear function of the photoinduced magnetic field B_o. The rotatory power of the optomagnetic medium can be defined based on the intensity of the pump light and in the first order can be expressed as a function of light pump light intensity E_o^2 through the Verdet constant of the inverse Faraday effect V_{IFE} as

$$V_{IFE}(\omega_p, \omega_s) \triangleq \frac{\rho_{xy}}{E_o^2} = -\frac{\pi}{2\lambda_o}\frac{e^2}{m^2}\frac{\omega_{pl}^4\omega_s}{c^2}\frac{\omega_p}{|D(\omega_p)|^2} \times \frac{\left(\omega_o^2 - \omega_s^2\right)^2 - \Gamma^2\omega_s^2}{|D(\omega_s)|^4}\frac{1}{\sqrt{1 + \frac{\omega_{pl}^2(\omega_o^2-\omega_s^2)}{|D(\omega_s)|^2}}}. \tag{44}$$

Note that the Verdet constant of the inverse Faraday effect is defined as a real quantity that is related to the polarization rotatory power as a function of both light pump and light signal frequencies and is different than the Verdet constant of the Faraday effect.

The change in the optical refractive index due to the presence of a static magnetic field was originally discovered by Voigt in 1902 in gases [26] and by Cotton and Mouton in 1907 for liquids [27]. Investigation of Eqs. (34) and (36) reveals that the real part of the diagonal permittivity elements is also altered by the photoinduced magnetic field, leading to linear birefringence in both x-z and y-z planes that can be called the inverse Voigt or Cotton-Mouton effect. The difference in the real part of the permittivity scales with M_{DC}^2, as

$$\epsilon'_{xx} - \epsilon'_{zz} \approx \mu_o^2\frac{e^2}{m^2}\frac{\omega_{pl}^2\omega_s^2\left(\omega_o^2 - \omega_s^2\right)}{|D(\omega_s)|^4}|\mathbf{M}_{DC}|^2 = \frac{e^4}{4m^4}\frac{\omega_{pl}^6}{c^4}\frac{\omega_s^2\left(\omega_o^2 - \omega_s^2\right)}{|D(\omega_s)|^4}\frac{\omega_p^2}{|D(\omega_p)|^4}E_o^4. \tag{45}$$

The rotatory power of the linear birefringence can be then calculated as

$$\rho_{xz} = -\rho_{yz} = \frac{\rho_{xy}}{2} = -\frac{\pi}{2\lambda_o}\frac{\epsilon''_{xy}}{\sqrt{\epsilon'_r}}. \tag{46}$$

The inverse Cotton-Mouton effect is weaker than the inverse Faraday effect, and its rotatory power is half of that produced by the inverse Faraday effect.

Both the inverse Faraday and Cotton-Mouton effects can be used in free space and integrated photonic systems for all-optical signal processing and nonreciprocal polarization devices without incorporating magnetic devices and characterization setup.

V. CONCLUSIONS

Optomagnetics have developed into an expanding research area with a potential of new discoveries in ultrafast magnetism and optics, novel applications in high-speed magnetic recording, information processing, and spintronics, as well as probing quantum and 2D materials.

We have provided a unified and generalized theoretical framework for optomagnetics, both quantum and classical treatments, through a prism of IFE effects. First, we start quantum-mechanical treatment to not only obtain a clear relationship between the photoinduced magnetization and transition dipole moments but also a generalized Pitaevskii's relationship. Using a perturbative method to solve the Schrödinger equation in the presence of a circularly polarized wave, our method explicitly and compactly finds the optical gyration vectors due to both orbital and spin magnetic moments. The effect of the damping phenomena is incorporated into the theory by introducing the excited-state population decay rate. Our formulation can be easily employed for quantum confined structures, i.e., quantum wells, wires, and dots.

Secondly, we employ the anharmonic Drude-Lorentz model to find the generalized Pitaevskii's relationship and its associated optical gyration coefficients. Comparison between quantum and classical treatments reveals the incompleteness of the classical treatment while it can lay down the basics for description of optomagnetic medium.

Lastly, the propagation of linearly polarized light signal through an optomagnetic medium, which is described by its first-order gyration coefficient, is analyzed through a typical pump-probe setup. Our formalism explicitly provides the analytical expressions of Verdet's constants for IFE and the inverse Cotton-Mouton effect through the permittivity tensor of the optomagnetic medium.

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APPENDIX

Here we report the first three order corrections to the probability amplitude, i.e., Eq. (9), under the influence of circularly polarized light with pump frequencies ω_p, ω_q, ω_r and the interaction Hamiltonian, i.e., Eq. (10). They read as

$$a_m^{(1)}(t) = \frac{e}{2\hbar}E_o(\omega_p)\left[\frac{r_{mg}e^{i(\omega_p-\omega_{mg})t}}{\omega_p - \omega_{mg}} - \frac{r_{mg}^* e^{-i(\omega_p+\omega_{mg})t}}{\omega_p + \omega_{mg}}\right], \tag{A1}$$

$$a_n^{(2)}(t) = \left(\frac{e}{2\hbar}\right)^2 E_o(\omega_p)E_o(\omega_q)\sum_m \left[\frac{r_{mg}r_{nm}e^{i(\omega_p+\omega_q-\omega_{ng})t}}{(\omega_p-\omega_{mg})(\omega_p+\omega_q-\omega_{ng})} - \frac{r_{mg}r_{nm}^* e^{i(\omega_p-\omega_q-\omega_{ng})t}}{(\omega_p-\omega_{mg})(\omega_p-\omega_q-\omega_{ng})}\right.$$
$$\left. - \frac{r_{mg}^* r_{nm} e^{-i(\omega_p+\omega_q+\omega_{ng})t}}{(\omega_p+\omega_{mg})(\omega_p+\omega_q+\omega_{ng})} + \frac{r_{mg}^* r_{nm}^* e^{-i(\omega_p-\omega_q+\omega_{ng})t}}{(\omega_p+\omega_{mg})(\omega_p-\omega_q-\omega_{ng})}\right], \tag{A2}$$

$$a_v^{(3)}(t) = \left(\frac{e}{2\hbar}\right)^3 E_o(\omega_p)E_o(\omega_q)E_o(\omega_r)\sum_{mn}\left[\frac{-r_{mg}r_{nm}r_{vn}e^{i(\omega_p+\omega_q+\omega_r-\omega_{vg})t}}{(\omega_p-\omega_{mg})(\omega_p+\omega_q-\omega_{ng})((\omega_p+\omega_q+\omega_r-\omega_{vg}))}\right.$$
$$+ \frac{r_{mg}r_{nm}^* r_{vn}e^{i(\omega_p-\omega_q+\omega_r-\omega_{vg})t}}{(\omega_p-\omega_{mg})(\omega_p-\omega_q-\omega_{ng})((\omega_p-\omega_q+\omega_r-\omega_{vg}))} + \frac{r_{mg}^* r_{nm}r_{vn}e^{-i(\omega_p+\omega_q-\omega_r+\omega_{vg})t}}{(\omega_p+\omega_{mg})(\omega_p+\omega_q+\omega_{ng})((\omega_p+\omega_q-\omega_r+\omega_{vg}))}$$
$$- \frac{r_{mg}^* r_{nm}^* r_{vn}e^{-i(\omega_p-\omega_q-\omega_r+\omega_{vg})t}}{(\omega_p+\omega_{mg})(\omega_p-\omega_q+\omega_{ng})((\omega_p-\omega_q-\omega_r+\omega_{vg}))} - \frac{r_{mg}r_{nm}r_{vn}^* e^{i(\omega_p+\omega_q-\omega_r-\omega_{vg})t}}{(\omega_p-\omega_{mg})(\omega_p+\omega_q-\omega_{ng})((\omega_p+\omega_q-\omega_r-\omega_{vg}))}$$
$$+ \frac{r_{mg}r_{nm}^* r_{vn}^* e^{i(\omega_p-\omega_q-\omega_r-\omega_{vg})t}}{(\omega_p-\omega_{mg})(\omega_p-\omega_q-\omega_{ng})((\omega_p-\omega_q-\omega_r-\omega_{vg}))} + \frac{r_{mg}^* r_{nm}r_{vn}^* e^{-i(\omega_p+\omega_q+\omega_r+\omega_{vg})t}}{(\omega_p+\omega_{mg})(\omega_p+\omega_q+\omega_{ng})((\omega_p+\omega_q+\omega_r+\omega_{vg}))}$$
$$\left. - \frac{r_{mg}^* r_{nm}^* r_{vn}^* e^{-i(\omega_p-\omega_q+\omega_r+\omega_{vg})t}}{(\omega_p+\omega_{mg})(\omega_p-\omega_q+\omega_{ng})((\omega_p-\omega_q+\omega_r+\omega_{vg}))}\right]. \tag{A3}$$

The third-order optical gyration vector can be written as

$$\mathbf{m}^{(3)} = \frac{Ne}{2m}\left(\frac{e}{2\hbar}\right)^6 \sum_{mnv}\langle u_v|(\hat{\mathbf{L}}+g_s\hat{\mathbf{S}})|u_v\rangle|r_{mg}r_{nm}r_{vn}|^2\big[D^{-1}(\omega_p-\omega_{mg})D^{-1}(\omega_p+\omega_q-\omega_{ng})D^{-1}(\omega_p+\omega_q+\omega_r-\omega_{vg})$$
$$+ D^{-1}(\omega_p-\omega_{mg})D^{-1}(\omega_p-\omega_q-\omega_{ng})D^{-1}(\omega_p-\omega_q+\omega_r-\omega_{vg})$$
$$+ D^{-1}(\omega_p+\omega_{mg})D^{-1}(\omega_p+\omega_q+\omega_{ng})D^{-1}(\omega_p+\omega_q-\omega_r+\omega_{vg})$$
$$+ D^{-1}(\omega_p+\omega_{mg})D^{-1}(\omega_p-\omega_q+\omega_{ng})D^{-1}(\omega_p-\omega_q-\omega_r+\omega_{vg})$$
$$+ D^{-1}(\omega_p-\omega_{mg})D^{-1}(\omega_p+\omega_q-\omega_{ng})D^{-1}(\omega_p+\omega_q-\omega_r-\omega_{vg})$$
$$+ D^{-1}(\omega_p-\omega_{mg})D^{-1}(\omega_p-\omega_q-\omega_{ng})D^{-1}(\omega_p-\omega_q-\omega_r-\omega_{vg})$$
$$+ D^{-1}(\omega_p+\omega_{mg})D^{-1}(\omega_p+\omega_q+\omega_{ng})D^{-1}(\omega_p+\omega_q+\omega_r+\omega_{vg})$$
$$+ D^{-1}(\omega_p+\omega_{mg})D^{-1}(\omega_p-\omega_q+\omega_{ng})D^{-1}(\omega_p-\omega_q+\omega_r+\omega_{vg})\big]. \tag{A4}$$

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