PHYSICAL REVIEW A VOLUME 36, NUMBER 12 DECEMBER 15, 1987

Intensity-dependent Faraday effect as a tool for controlling the process of light self-squeezing

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(Received 13 April 1987)

A phenomenological (classical) and quantum treatment of the laser light-intensity-induced Faraday effect in isotropic media and crystals is given. The effect, being universal (occurring in all matter), is shown to act as a stimulator controlling the self-squeezing of light: depending on the helicity of circular polarization of the light wave and on the sense of the dc magnetic field, the sign of the pseudoscalar parameter of nonlinear coupling changes leading to an enhancement or weakening of self-squeezing. Moreover, by varying the state of polarization of the light wave or the magnetic field strength, control of the efficiency of light-squeezing generation can be achieved.

I. INTRODUCTION

Maker *et al.*[1] have observed that intense elliptically polarized laser light exhibits rotation of its polarization plane on traversal of an isotropic medium. If, additionally, a dc magnetic field is applied, intensity-dependent magneto-optical rotation appears in Faraday or, respectively, Voigt configuration according to whether the light beam propagates parallel or perpendicularly to the magnetic field.[2] Laser intensity-dependent Faraday rotation has been observed in indium antimonate.[3] The nonlinear Faraday effect plays a considerable role in light self-focusing and parametric wave interaction.[4] The laser beam intensity-dependent Faraday effect has been analyzed theoretically for molecular fluids[5,6] and crystals,[2,7] as well as atomic gases.[8]

The optical nonlinearities of bodies play a decisive part in the generation of squeezed states of the electromagnetic field. The theory of these states is now developing rapidly[9–11] and the first experiments have already been carried out.[12–15]

In the present paper we show that the intensity-dependent Faraday effect leads to squeezed states of the field, just as is the case for free nonlinear propagation of light.[16] There is, however, an essential difference: in the case under consideration by us, the parameters of nonlinear coupling (nonlinear gyration) are linearly dependent on the strength of the externally applied dc magnetic field and, as a result, change their sign according to the sense of the magnetic field (according to whether it is applied parallel or antiparallel to the propagation direction of the wave). Obviously, the same can be achieved by a change in helicity of the circular polarization of the light wave. This permits the enhancement, or weakening, of the efficiency of the light self-squeezing generated.

The experiment with nonlinear self-interaction of a single laser beam at Faraday configuration can be extended to two beams,[5,6] with the probe beam at Faraday or Voigt configuration[2] and the laser beam stimulating the optical nonlinearity propagating at an arbitrary angle to the dc magnetic field. Recently, Lakshmi and Agarwal[17] have discussed the squeezing characteristic of radiation from the Hanle effect, in which the scattering radiation exhibits changes due to the magnetic levels (transitions).

II. PHENOMENOLOGICAL TREATMENT

Consider an arbitrary nondissipative medium in a homogeneous dc magnetic field with induction $\mathbf{B}$ on which a light wave with electric vector

$$\mathbf{E}(t)=\mathbf{E}^{+}(\omega)e^{-i\omega t}+\mathbf{E}^{-}(\omega)e^{i\omega t}$$

and circular frequency ω is incident.

The phenomenological approach to nonlinear optical processes starts from the time-average free energy F (Ref. 18) determining the interaction between the medium and the field of radiation. In our case F has the following form (we apply the Einstein convention):

$$\begin{aligned}F=-\tfrac{1}{2}\{&\chi_{ij}(\omega,\mathbf{B})E_i^-(\omega)E_j^+(\omega)+\chi_{ij}^*(\omega,\mathbf{B})E_i^+(\omega)E_j^-(\omega)\\&+\chi_{ijkl}(\omega,\mathbf{B})[E_i^-(\omega)E_j^-(\omega)E_k^+(\omega)E_l^+(\omega)+E_i^-(\omega)E_k^+(\omega)E_j^-(\omega)E_l^+(\omega)+E_i^-(\omega)E_k^+(\omega)E_l^+(\omega)E_j^-(\omega)]\\&+\chi_{ijkl}^*(\omega,\mathbf{B})[E_i^+(\omega)E_j^+(\omega)E_k^-(\omega)E_l^-(\omega)+E_i^+(\omega)E_k^-(\omega)E_j^+(\omega)E_l^-(\omega)+E_i^+(\omega)E_k^-(\omega)E_l^-(\omega)E_j^+(\omega)]+\text{H.c.}\}\ ,\end{aligned} \quad (1)$$

whence, in a linear approximation in $\mathbf{B}$, we obtain the expansion

$$\begin{aligned}\chi_{ij}(\omega,\mathbf{B})&=\chi_{ij}(-\omega;\omega)+\chi_{ijk}(-\omega;\omega,0)B_k+\cdots\ ,\\ \chi_{ijkl}(\omega,\mathbf{B})&=\chi_{ijkl}(-\omega;-\omega,\omega,\omega)\\&\quad+\chi_{ijklm}(-\omega;-\omega,\omega,\omega,0)B_m+\cdots\ .\end{aligned} \quad (2)$$

Above, we have omitted terms in the successive harmonics, as well as higher powers of the field $\mathbf{B}$, considered by Manakov *et al.*[19]

The second-rank tensor $\chi_{ij}(-\omega;\omega)$ determines the linear electric dipole susceptibility. The axial tensor of third rank (of linear gyration) $\chi_{ijk}(-\omega;\omega,0)$ describes the magnetoelectric susceptibility of second order intervening in the usual Faraday effect.[2,19] The tensor of fourth rank $\chi_{ijkl}(-\omega;-\omega,\omega,\omega)$ describes the nonlinear susceptibility of third order determining the light-intensity-dependent refractive index.[1] The axial tensor of fifth rank (of nonlinear gyration) $\chi_{ijklm}(-\omega;-\omega,\omega,\omega,0)$ describes a nonlinear magnetoelectric susceptibility of fourth order present in the intensity-dependent Faraday effect.[2] The quantum-mechanical form of the gyration tensors $\chi_{ijk}(-\omega;\omega,0)$ and $\chi_{ijklm}(-\omega;-\omega,\omega,\omega,0)$ have been given by us[2,7] together with their tabulated nonzero and mutually independent components for all crystallographical classes.[2]

We assume that the light beam propagates in a macroscopically isotropic medium composed of molecules that are not optically active (see Appendix). The propagation direction, denoted as the z direction, is assumed to be parallel to the static magnetic field **B** (Faraday configuration). Applying the circular basis for the optical fields (angular momentum convention) for right and left helicity of polarization,

$$E^{\pm}_{\pm}(\omega)=\frac{1}{\sqrt{2}}[E_x^{+}(\omega)\mp iE_y^{+}(\omega)] ,$$

we can write the time-averaged free energy (1) for a body without intrinsic gyration (for the general case, see Appendix):

$$F=-\chi_G^l(B_z)\{E^{-}_{+}(\omega)E^{+}_{+}(\omega)-E^{-}_{-}(\omega)E^{+}_{-}(\omega)\}-\tfrac{1}{2}\chi_R^{\rm nl}\{[E^{-}_{+}(\omega)]^2[E^{+}_{+}(\omega)]^2+[E^{-}_{-}(\omega)]^2[E^{+}_{-}(\omega)]^2\}$$
$$-\mathcal{H}_R^{\rm nl}E^{-}_{+}(\omega)E^{-}_{-}(\omega)E^{+}_{-}(\omega)E^{+}_{+}(\omega)-\tfrac{1}{2}\chi_G^{\rm nl}(B_z)\{[E^{-}_{+}(\omega)]^2[E^{+}_{+}(\omega)]^2-[E^{-}_{-}(\omega)]^2[E^{+}_{-}(\omega)]^2\} , \qquad (3)$$

where we have introduced the linear (Faraday) magnetogyration parameter

$$\chi_G^l(B_z)=i[\chi_{xyz}(\omega)-\chi_{yxz}(\omega)]B_z , \qquad (4)$$

the nonlinear coupling parameters

$$\chi_R^{\rm nl}=12[\chi_{xyxy}(\omega)+\chi_{xyyx}(\omega)] ,$$
$$\mathcal{H}_R^{\rm nl}=24\chi_{xxyy}(\omega)+\chi_R^{\rm nl} , \qquad (5)$$

and nonlinear magnetogyration parameter

$$\chi_G^{\rm nl}(B_z)=12i[\chi_{xxxyz}(\omega)-\chi_{yyyxz}(\omega)]B_z . \qquad (6)$$

In (4)–(6) we have used the shortened notation $\chi_{ijk}(\omega)=\chi_{ijk}(-\omega;\omega,0)$, $\chi_{ijkl}(\omega)=\chi_{ijkl}(-\omega;-\omega,\omega,\omega)$, and $\chi_{ijklm}(\omega)=\chi_{ijklm}(-\omega;-\omega,\omega,\omega,0)$.

On defining the right and left circular components of the electric polarization vector **P** as[18]

$$P^{+}_{+}(\omega)=-\frac{\partial F}{\partial E^{-}_{+}(\omega)},$$

$$P^{+}_{-}(\omega)=-\frac{\partial F}{\partial E^{-}_{-}(\omega)} ,$$

we obtain, by (3), the following expressions:

$$P^{+}_{\pm}(\omega)=\{\pm\chi_G^l(B_z)+[\chi_R^{\rm nl}\pm\chi_G^{\rm nl}(B_z)]\,|E^{+}_{\pm}(\omega)|^2+\mathcal{H}_R^{\rm nl}\,|E^{+}_{\mp}(\omega)|^2\}E^{+}_{\pm}(\omega) . \qquad (7)$$

Taking into account the relation $(n_{\pm}^2-n^2)E_{\pm}(\omega)=4\pi P_{\pm}(\omega)$ between the polarization and the refractive index of the medium, we have from (7)

$$n_{\pm}-n=\frac{2\pi}{n}\{\pm\chi_G^l(B_z)+\mathcal{H}_R^{\rm nl}\,|E^{+}_{\mp}(\omega)|^2+[\chi_R^{\rm nl}\pm\chi_G^{\rm nl}(B_z)]\,|E^{+}_{\pm}(\omega)|^2\} , \qquad (8)$$

or, for the difference of the refractive indices describing the optical rotation,

$$n_{+}-n_{-}=\frac{2\pi}{n}\{2\chi_G^l(B_z)+\chi_G^{\rm nl}(B_z)[\,|E^{+}_{+}(\omega)|^2+|E^{+}_{-}(\omega)|^2]+(\mathcal{H}_R^{\rm nl}-\chi_R^{\rm nl})[\,|E^{+}_{-}(\omega)|^2-|E^{+}_{+}(\omega)|^2]\}. \qquad (9)$$

The first term on the right-hand side of (9) describes the usual Faraday effect related to the linear gyration constant $\chi_G^l(B_z)$. The second term describes the light-intensity-dependent Faraday effect[5] related to the nonlinear gyration constant $\chi_G^{\rm nl}(B_z)$, and the last term describes the light-intensity-dependent rotation of the polarization ellipse.[1]

There is, however, a very essential difference between the two last-named nonlinear effects: Whereas the effect of rotation of the ellipse (or, rather, elliptical birefringence[20]) takes place only in the case of elliptically polarized light,[1] the nonlinear Faraday effect occurs for arbitrary light polarization—linear ($E_{+}=E_{-}=E/\sqrt{2}$) and circular (E_{+} or E_{-}) as well as elliptical ($E_{+}\neq E_{-}$). Thus the two effects can be separated by appropriately choosing the state of light polarization.

On inserting the polarization (7) into the Maxwell equations, we obtain for slowly varying amplitudes of the electromagnetic field the following equations:

$$\frac{dE^{+}_{\pm}(\omega,z)}{dz}=\frac{i2\pi\omega}{cn}\{\pm\chi_G^l(B_z)+[\chi_R^{\rm nl}\pm\chi_G^{\rm nl}(B_z)]\,|E^{+}_{\pm}(\omega,z)|^2+\mathcal{H}_R^{\rm nl}\,|E^{+}_{\mp}(\omega,z)|^2\}E^{+}_{\pm}(\omega,z) . \qquad (10)$$

Since $|E^{+}_{\pm}(\omega,z)|^2$ does not depend on z, i.e., $(d/dz)|E^{+}_{\pm}(\omega,z)|^2=0$, Eq. (10) has a simple exponential solution,[21] which in our case has the form

$$E_{\pm}^{\pm}(\omega,z)=\exp(i\Phi_{\pm}z)E_{\pm}^{\pm}(\omega,0)\ , \tag{11}$$

where

$$\Phi_{\pm}=\frac{2\pi\omega}{cn}\{\pm\chi_G^l(B_z)+[\chi_R^{\rm nl}\pm\chi_G^{\rm nl}(B_z)]\,|E_{\pm}^{\pm}(\omega)|^2+\mathcal{H}_R^{\rm nl}\,|E_{\mp}^{\pm}(\omega)|^2\} \tag{12}$$

determines the light-intensity phase shifts of the field amplitude in the presence of a dc magnetic field B_z (in the absence of natural gyration, considered by us[22]).

III. QUANTUM TREATMENT

In conformity with the classical equation (10), we go over from the Heisenberg equation of motion for the time evolution of the field operators to the equations of propagation of the wave[23] through an active medium in which it traverses a path of well defined length z:

$$\frac{\partial E(\omega,z)}{\partial z}=\frac{n}{i\hbar c}[E(\omega,z),H_I]\ . \tag{13}$$

In the quantum approach, the operators of the electric field are expressed as follows in terms of annihilation operators (in cgs units):

$$E_{\pm}^{+}(\omega)=i\left[\frac{2\pi\hbar\omega}{Vn^2}\right]^{1/2}a_{\pm}\ , \tag{14}$$

where the annihilation operators $a_{\pm}$ in circular basis fulfill the commutation relations

$$[a_i,a_j^{+}]=\delta_{ij}\quad (i,j=+\sigma r-)\ , \tag{15}$$

where V is the field quantization volume.

The next essential step resides in finding the analytical form of the Hamiltonian H_I of interaction between the material system and the field of radiation. On the microscopic level, H_I is correctly expressed by way of the Fermi operators of the optically active bond electrons in the medium and the Bose operators of the light field modes.[24,25] In general the problem is a highly involved one[26] and has been solved for a specific atomic model and nonlinear process. To circumvent these difficulties in more general cases of arbitrary bodies we have recourse to effective Hamiltonians of interaction.[18,24–27] In practice, such Hamiltonians are constructed on the basis of the time-averaged free energy F in which the field vectors are replaced by boson operators in accordance with the relation (14). Thus, we have[24,27] $H_I\rightarrow\int F\,d^3{\rm r}$ or rather, as in the present case of a homogeneous medium, $H_I=VF$, and by (3) and (14) we obtain the following effective interaction Hamiltonian:

$$\begin{aligned}H_I=&-\hbar\tilde{\chi}_G^l(B_z)(a_+^{\dagger}a_+-a_-^{\dagger}a_-)\\&-\frac{\hbar}{2}\tilde{\chi}_R^{\rm nl}[(a_+^{\dagger})^2(a_+)^2+(a_-^{\dagger})^2(a_-)^2]\\&-\hbar\tilde{\mathcal{H}}_R^{\rm nl}a_+^{\dagger}a_-^{\dagger}a_-a_+\\&-\frac{\hbar}{2}\tilde{\chi}_G^{\rm nl}(B_z)[(a_+^{\dagger})^2(a_+)^2-(a_-^{\dagger})^2(a_-)^2]\ ,\end{aligned} \tag{16}$$

where the coupling constants are now given by

$$\begin{aligned}&\tilde{\chi}_G^l(B_z)=\frac{V}{\hbar}\left[\frac{2\pi\hbar\omega}{Vn^2}\right]\chi_G^l(B_z)\ ,\\&\tilde{\chi}_R^{\rm nl}=\frac{V}{\hbar}\left[\frac{2\pi\hbar\omega}{Vn^2}\right]^2\chi_R^{\rm nl},\quad \tilde{\mathcal{H}}_R^{\rm nl}=\frac{V}{\hbar}\left[\frac{2\pi\hbar\omega}{Vn^2}\right]^2\mathcal{H}_R^{\rm nl}\ ,\\&\tilde{\chi}_G^{\rm nl}(B_z)=\frac{V}{\hbar}\left[\frac{2\pi\hbar\omega}{Vn^2}\right]^2\chi_G^{\rm nl}(B_z)\ .\end{aligned} \tag{17}$$

If there is a need for expressing the preceding phenomenological susceptibilities in quantum-mechanical form, this can be done in the well-known manner.[2,8,28]

With regard to (13), (15), and (16), we obtain for the field operators in circular basis:

$$\frac{da_{\pm}(z)}{dz}=\frac{in}{c}\{\pm\tilde{\chi}_G^l(B_z)+\tilde{\mathcal{H}}_R^{\rm nl}a_{\mp}^{\dagger}(z)a_{\mp}(z)+[\tilde{\chi}_R^{\rm nl}\pm\tilde{\chi}_G^{\rm nl}(B_z)]a_{\pm}^{\dagger}(z)a_{\pm}(z)\}a_{\pm}(z)\ . \tag{18}$$

Since the photon number operators $a_+^{\dagger}a_+$ and $a_-^{\dagger}a_-$ are constants of motion, they commute with the Hamiltonian (16); the equations (18) have a formal solution in the form of the translation operator

$$a_{\pm}(z)=\exp\{iz[\Phi_{\pm}^0(B_z)+\epsilon_{\pm}(B_z)a_{\pm}^{\dagger}(0)a_{\pm}(0)+\delta a_{\mp}^{\dagger}(0)a_{\mp}(0)]\}a_{\pm}(0)\ , \tag{19}$$

where we have introduced the following notation:

$$\begin{aligned}&\Phi_{\pm}^0(B_z)=\pm\frac{n}{c}\tilde{\chi}_G^l(B_z)\ ,\\&\epsilon_{\pm}(B_z)=\frac{n}{c}[\tilde{\chi}_R^{\rm nl}\pm\tilde{\chi}_G^{\rm nl}(B_z)]\ ,\\&\delta=\frac{n}{c}\tilde{\mathcal{H}}_R^{\rm nl}\ .\end{aligned} \tag{20}$$

The solution of (19) in the absence of a magnetic field is identical with that for self-squeezing[16] and resembles that of Milburn *et al.*[23] for squeezing in degenerate four-wave mixing in the linearly polarized case. As was predictable, there is correct correspondence between the classical (11) and quantum (19) fields.

We now write the quantum field operators (19) in a manner to extract, from the phase, the part due to rotation of the plane of polarization:

$$a_{\pm}(z)=\exp[i(\phi\pm\alpha)]a_{\pm}(0)\ , \tag{21}$$

where, by definition, we have

$$\phi=\frac{1}{2}\frac{\omega}{c}(n_++n_-)z=\frac{nz}{2c}[(\tilde{\chi}_R^{\rm nl}+\tilde{\mathcal{H}}_R^{\rm nl})(a_+^{\dagger}a_++a_-^{\dagger}a_-)-\tilde{\chi}_G^{\rm nl}(B_z)(a_-^{\dagger}a_--a_+^{\dagger}a_+)]\ , \tag{22}$$

$$\alpha=\frac{1}{2}\frac{\omega}{c}(n_+-n_-)z$$
$$=\frac{nz}{2c}[2\tilde{\chi}^{l}_{G}(B_z)+(\tilde{\mathcal{H}}^{\rm nl}_{R}-\tilde{\chi}^{\rm nl}_{R})(a^\dagger_-a_--a^\dagger_+a_+)$$
$$+\tilde{\chi}^{\rm nl}_{G}(B_z)(a^\dagger_+a_++a^\dagger_-a_-)] , \qquad (23)$$

with α the angle by which the polarization plane is rotated on traversal of the light wave through the medium of length z. Obviously, the quantum expression (23) corresponds to the classical magneto-optical rotation (9).

The formulas (21)–(23) hold for elliptical polarization, when $a_+\neq a_-$. In the particular case of linearly polarized light ($a_+=a_-=a/\sqrt{2}$), the phases (22) and (23) take the form

$$\phi_l=\frac{nz}{2c}(\tilde{\chi}^{\rm nl}_{R}+\tilde{\mathcal{H}}^{\rm nl}_{R})a^\dagger a , \qquad (22')$$

$$\alpha_l=\frac{nz}{2c}[2\tilde{\chi}^{l}_{G}(B_z)+\tilde{\chi}^{\rm nl}_{G}(B_z)a^\dagger a] . \qquad (23')$$

We note that for the case of linear light polarization the rotation of the plane of polarization (23′) is due only to linear and nonlinear magnetogyration. Obviously, this is a specific feature of Faraday's effect. In a medium with natural optical activity[22] a role similar to that of magneto-optical gyration is played by linear and nonlinear gyration, the numerical value of which is, however, much smaller[4] and can sometimes be neglected by comparison with the Verdet constants $\tilde{\chi}^{l}_{G}/B_z$ and $\tilde{\chi}^{\rm nl}_{G}/B_z$ taken into account above.

IV. SQUEEZING

On defining the Hermitian operators describing the in-phase and out-of-phase quadrature components of the electromagnetic field as follows:

$$Q_\pm=a_\pm+a^\dagger_\pm,\quad P_\pm=-i(a_\pm-a^\dagger_\pm) , \qquad (24)$$

squeezed states of light are defined[9] as states in which one of the normally ordered variances becomes negative:

$$\langle:(\Delta Q_\pm)^2:\rangle<0 \quad\text{or}\quad \langle:(\Delta P_\pm)^2:\rangle<0 . \qquad (25)$$

Using the solution (19) and the commutation relations (15), one easily calculates the normally ordered variances (25). The result is as follows:

$$\langle:[\Delta Q_\pm(z)]^2:\rangle=2\,{\rm Re}\{\alpha^2_\pm\exp[2i\Phi^0_\pm(B_z)z+i\epsilon_\pm(B_z)z+(e^{2i\epsilon_\pm(B_z)z}-1)|\alpha_\pm|^2+(e^{2i\delta z}-1)|\alpha_\mp|^2]$$
$$-\alpha^2_\pm\exp[2i\Phi^0_\pm(B_z)z+2(e^{i\epsilon_\pm(B_z)z}-1)|\alpha_\pm|^2+2(e^{i\delta z}-1)|\alpha_\mp|^2]\}$$
$$+2|\alpha_\pm|^2[1-\exp(2\{\cos[\epsilon_\pm(B_z)z]-1\}|\alpha_\pm|^2+2[\cos(\delta z)-1]|\alpha_\mp|^2)] , \qquad (26)$$

where $\Phi^0_\pm$, $\epsilon_\pm$, and δ are defined by (20). In (26), the incoming beam has been assumed as being in a coherent state $|\alpha\rangle$ with complex amplitudes $\alpha_\pm$ given by[16]

$$\alpha_\pm=\frac{\alpha}{\sqrt{2}}(\cos\eta\pm\sin\eta)e^{\mp i\Theta} , \qquad (27)$$

where Θ and η are the azimuth and ellipticity of its polarization ellipse. For a linearly polarized beam $\eta=0$, whereas for a circularly polarized beam $\eta=\pm\pi/4$.

To obtain the solution for $\langle:[\Delta P_\pm(z)]^2:\rangle$ it suffices to change the sign of the first Re term in (26). The solution (26) is rather complicated. Without performing a numerical analysis, one cannot say whether it can be negative or not. We have performed such an analysis[16] for the variances in the absence of the static magnetic field proving the possibility of self-squeezing of light propagating through isotropic media, and have shown that variances of the type (26) exhibit oscillatory behavior with minima that dip to more than 90% of squeezing. In order to obtain such strong squeezing, however, one would have to tune precisely to the minimum by properly adjusting the intensity of the incoming beam. Since the minima appear for rather high intensities, it may prove difficult to adjust the intensity so as to obtain exactly a minimum of fluctuations. As we have shown in this paper, there is an extra physical factor—the magnetic field—that can be used to tune precisely to the minimum fluctuations. As it is seen from (20), both $\Phi^0_\pm(B_z)$ and $\epsilon_\pm(B_z)$ vary continuously with variations of the magnetic field strength. Thus both the linear phase $\Phi^0_\pm(B_z)$ of the optical field and the nonlinear coupling constant $\epsilon_\pm(B_z)$ are changed simultaneously under the influence of the static magnetic field. The ordinary Faraday variation in linear phase can be compensated by changing the initial phase of the incoming optical field, and the magnetic field dependence of the nonlinear coupling constant $\epsilon_\pm(B_z)$ can be used for precise tuning to the minimum fluctuations. Thus the intensity-dependent Faraday effect can be used to increase the efficiency of self-squeezing.

The nonlinear coupling parameters are rather small and we have $\epsilon_\pm z\ll\epsilon_\pm z|\alpha_\pm|^2$ and $\delta z\ll\delta z|\alpha_\pm|^2$. This allows us to obtain much simpler approximate formulas for the variances (26), which read

$$\langle:[\Delta Q_\pm(z)]^2:\rangle$$
$$\approx-2\{\epsilon_\pm(B_z)|\alpha_\pm|^2z\sin\Theta_\pm+[\epsilon^2_+(B_z)|\alpha_\pm|^4$$
$$+\delta^2|\alpha_+|^2|\alpha_-|^2]z^2$$
$$\times(\cos\Theta_\pm-1)\} , \qquad (28)$$

where

$$\Theta_\pm=-2\Theta+2z[\Phi^0_\pm(B_z)+\epsilon_\pm(B_z)|\alpha_\pm|^2+\delta|\alpha_\mp|^2] . \qquad (29)$$

The formulas (28) are much simpler to interpret than the exact formulas (26) and reproduce their exact results quite satisfactorily. The oscillatory behavior of the variances is clearly visible, and with regard to (27) their dependence on the polarization state of the incoming beam is obvious. The dependence of the variances on the static magnetic field is through $\Phi^0_{\pm}(B_z)$ and $\epsilon_{\pm}(B_z)$, and our preceding discussion is equally valid for the approximate formulas (28) as for the exact formulas (26). Numerical evaluations of the simpler formulas (28), however, are quite easy to perform even using a pocket calculator.

V. CONCLUSIONS

We have discussed the intensity-dependent Faraday effect as a means to enhance the efficiency of self-squeezing of light propagating through isotropic nonlinear media. Although the normally ordered variances of the quadrature components of the optical field as derived above in a static magnetic field have the same form as in the absence of a magnetic field[16] and, for a circularly polarized incoming beam are the same as for an anharmonic oscillator,[29] the magnetic field dependence of the linear phase $\Phi^0_{\pm}(B_z)$ and nonlinear coupling constant $\epsilon_{\pm}(B_z)$ introduce a novel factor of practical importance. This dependence will permit the precise tuning to minimum fluctuations by varying the strength of the static magnetic field.

In contradistinction to other mechanisms leading to the generation of squeezed states of the electromagnetic field,[9–11] the nonlinear Faraday effect considered in the present work permits the control of the efficiency of squeezing by way of external factors, such as the state of polarization of the incoming wave, the properties of the nonlinear medium, and, essentially, the strength of the dc or ac externally applied magnetic field—in particular, its sense.

APPENDIX: EFFECTIVE INTERACTION HAMILTONIAN FOR ARBITRARY NONLINEAR MEDIUM

In the expansions (2) we had restricted ourselves to frequency dispersion. We shall now moreover take spatial dispersion into account, so that the susceptibilities will be dependent on the wave vector **k** of the field of radiation.[30,31] In other words, multipolar electric and magnetic contributions will be included in the interaction Hamiltonian.[32,33]

At weak spatial dispersion, the linear and nonlinear electric susceptibilities in the absence as well as in the presence of the field **B** can be written in the form of the electric and magnetic multipolar susceptibility tensors[30,31]

$$\chi_{ij}(-\omega;\omega;\mathbf{k};-\mathbf{k})={}^{(1)}_{e}\chi^{(1)}_{e\,ij}+\frac{i\omega n}{3c}\left({}^{(1)}_{e}\chi^{(2)}_{e\,i(jp)}-{}^{(2)}_{e}\chi^{(1)}_{e\,(ip)j}\right)s_p+\frac{n}{c}\left({}^{(1)}_{e}\chi^{(1)}_{m\,iu}\delta_{upj}+{}^{(1)}_{m}\chi^{(1)}_{e\,uj}\delta_{upi}\right)s_p+\cdots\,, \tag{A1}$$

$$\chi_{ijl}(-\omega;\omega,0;\mathbf{k};-\mathbf{k},0)={}^{(1)}_{e}\chi^{(1)m}_{e\,ijl}+\frac{i\omega n}{3c}\left({}^{(1)}_{e}\chi^{(2)m}_{e\,i(jp)l}-{}^{(2)}_{e}\chi^{(1)m}_{e\,(ip)jl}\right)s_p+\frac{n}{c}\left({}^{(1)}_{e}\chi^{(1)m}_{m\,iul}\delta_{upj}+{}^{(1)}_{m}\chi^{(1)m}_{e\,ujl}\delta_{upi}\right)s_p+\cdots\,, \tag{A2}$$

$$\begin{aligned}\chi_{ijkl}(-\omega;-\omega,\omega,\omega;\mathbf{k};\mathbf{k},-\mathbf{k},-\mathbf{k})={}&{}^{(1)}_{e}\chi^{(1,1,1)}_{eee\,ijkl}+\frac{i\omega n}{3c}\left({}^{(1)}_{e}\chi^{(1,1,2)}_{eee\,ijk(lp)}+{}^{(1)}_{e}\chi^{(1,2,1)}_{eee\,ij(kp)l}-{}^{(1)}_{e}\chi^{(2,1,1)}_{eee\,i(jp)kl}-{}^{(2)}_{e}\chi^{(1,1,1)}_{eee\,(ip)jkl}\right)s_p\\&+\frac{n}{c}\left({}^{(1)}_{e}\chi^{(1,1,1)}_{eem\,ijku}\delta_{upl}+{}^{(1)}_{e}\chi^{(1,1,1)}_{eme\,ijul}\delta_{upk}+{}^{(1)}_{e}\chi^{(1,1,1)}_{mee\,iukl}\delta_{upj}+{}^{(1)}_{m}\chi^{(1,1,1)}_{eee\,ujkl}\delta_{upi}\right)s_p+\cdots\,,\end{aligned} \tag{A3}$$

$$\begin{aligned}\chi_{ijklr}(-\omega;-\omega,\omega,\omega,0;\mathbf{k};\mathbf{k},-\mathbf{k},-\mathbf{k},0)={}&{}^{(1)}_{e}\chi^{(1,1,1)m}_{eee\,ijklr}+\frac{i\omega n}{3c}\left({}^{(1)}_{e}\chi^{(1,1,2)m}_{eee\,ijk(lp)r}+{}^{(1)}_{e}\chi^{(1,2,1)m}_{eee\,ij(kp)lr}-{}^{(1)}_{e}\chi^{(2,1,1)m}_{eee\,i(jp)klr}-{}^{(2)}_{e}\chi^{(1,1,1)m}_{eee\,(ip)jklr}\right)s_p\\&+\frac{n}{c}\left({}^{(1)}_{e}\chi^{(1,1,1)m}_{eem\,ijkur}\delta_{upl}+{}^{(1)}_{e}\chi^{(1,1,1)m}_{eme\,ijulr}\delta_{upk}\right.\\&\left.+{}^{(1)}_{e}\chi^{(1,1,1)m}_{mee\,iuklr}\delta_{upj}+{}^{(1)}_{m}\chi^{(1,1,1)m}_{eee\,ujklr}\delta_{upi}\right)s_p+\cdots\,,\end{aligned} \tag{A4}$$

where s is a unit vector in the propagation direction of the wave $\mathbf{k}=(\omega n/c)\mathbf{s}$, and δ_{upi} is the Levi-Cività tensor, whereas n and c denote, respectively, the refractive index of the light wave in the medium and its velocity in vacuum.

Above ${}^{(p)}_{A}\chi^{(q)}_{D}={}^{(p)}_{A}\chi^{(q)}_{D}(-\omega;\omega)$ is the linear multipolar electric ($A=e$) or, respectively, magnetic ($A=m$) susceptibility of the pth order related with a multipolar electric ($D=e$) or, respectively, magnetic ($D=m$) transition of the qth order, whereas ${}^{(p)}_{A}\chi^{(q)m}_{D}={}^{(p)}_{A}\chi^{(q)m}_{D}(-\omega;\omega,0)$ represents the linear (in the first approximation of quantum-mechanical perturbation calculus) magnetically induced variation in the susceptibility ${}^{(p)}_{A}\chi^{(q)}_{D}$.

Similarly, ${}^{(p)}_{A}\chi^{(q,r,s)}_{DFG}={}^{(p)}_{A}\chi^{(q,r,s)}_{DFG}(-\omega;-\omega,\omega,\omega)$ is the nonlinear multipolar electric ($A=e$) or, respectively, magnetic ($A=m$) susceptibility of the pth order due to three multipolar electric or magnetic transitions of order q, r, and s, respectively. The linear magnetic variation in the susceptibility ${}^{(p)}_{A}\chi^{(q,r,s)}_{DFG}$ is given by ${}^{(p)}_{A}\chi^{(q,r,s)m}_{DFG}={}^{(p)}_{A}\chi^{(q,r,s)m}_{DFG}(-\omega;-\omega,\omega,\omega,0)$.

The Hamiltonian (1) is the time-averaged free energy; when deriving it, the following general relation, fulfilled by the linear and nonlinear multipole susceptibility, was taken into account:[28,34]

$$[{}^{(p)}_{A}\chi^{(q)}_{B}(-\omega;\omega)]^* = {}^{(p)}_{A}\chi^{(q)}_{B}(\omega;-\omega) \,,$$
$$[{}^{(p)}_{A}\chi^{(q,r,s)}_{DFG}(-\omega;-\omega,\omega,\omega)]^* = {}^{(p)}_{A}\chi^{(q,r,s)}_{DFG}(\omega;\omega,-\omega,-\omega) \quad \text{(A5)}$$

(the asterisk stands for the complex conjugate) as well as the invariancy of the nonlinear multipole susceptibilities ${}^{(p)}_{A}\chi^{(q,r,s)}_{DFG}(-\omega;\omega_1,\omega_2,\omega_3)$ with respect to the 3! space of ternary combinations $q\omega_1 D$, $r\omega_2 F$, and $s\omega_3 G$. The condition (A5) ensures that the linear and nonlinear electric ($A=e$) and magnetic ($A=m$) polarizations are real for real electric and magnetic fields. The above relations hold as well in the presence of a dc field **B**.

From the well-known Bloembergen permutation relations[28,34] it results immediately that the nonlinear multipole susceptibilities ${}^{(p)}_{A}\chi^{(q,r,s)}_{DFG}(-\omega;-\omega,\omega,\omega)$ as well as their linear magnetic variations are invariant with respect to pairwise permutations of pA and qD, and of rF and sG:

$$\begin{aligned}{}^{(p)}_{A}\chi^{(q,r,s)}_{DFG}(-\omega;-\omega,\omega,\omega) &= {}^{(q)}_{D}\chi^{(p,r,s)}_{AFG}(-\omega;-\omega,\omega,\omega)\\ &= {}^{(p)}_{A}\chi^{(q,s,r)}_{DGF}(-\omega;-\omega,\omega,\omega)\\ &= {}^{(q)}_{D}\chi^{(p,s,r)}_{AGF}(-\omega;-\omega,\omega,\omega) \,. \end{aligned} \quad \text{(A6)}$$

This relation permits a reduction from 4 to 2 in the number of nonlinear multipole susceptibilities of Eqs. (A3) and (A4) involving a quadrupolar electric or a dipolar magnetic transition.

With regard to the quantum-mechanical definition of linear and nonlinear multipolar susceptibilities,[31,32] it can be shown that even if the widths of the energy levels are neglected (transparent medium) the ${}^{(p)}_{A}\chi^{(q)}_{B}(-\omega;\omega)$ and ${}^{(p)}_{A}\chi^{(q,r,s)}_{DFG}(-\omega;-\omega,\omega,\omega)$ as well as their magnetically induced variations are complex for complex wave functions of the stationary states (degeneracy of the energy levels), i.e.,

$${}^{(p)}_{A}\chi^{(q)}_{B}(-\omega;\omega) = {}^{(p)}_{A}\eta^{(q)}_{B}(-\omega;\omega) + i\,{}^{(p)}_{A}\zeta^{(q)}_{B}(-\omega;\omega) \,, \quad \text{(A7)}$$

$${}^{(p)}_{A}\chi^{(q,r,s)}_{DFG}(-\omega;-\omega,\omega,\omega) = {}^{(p)}_{A}\eta^{(q,r,s)}_{DFG}(-\omega;-\omega,\omega,\omega) + i\,{}^{(p)}_{A}\zeta^{(q,r,s)}_{DFG}(-\omega;-\omega,\omega,\omega) \,, \quad \text{(A8)}$$

with their real and imaginary parts fulfilling the following relations:

$${}^{(p)}_{A}\eta^{(q)}_{B}(-\omega;\omega) = {}^{(q)}_{B}\eta^{(p)}_{A}(-\omega;\omega) \,, \quad \text{(A9)}$$

$${}^{(p)}_{A}\zeta^{(q)}_{B}(-\omega;\omega) = -{}^{(q)}_{B}\zeta^{(p)}_{A}(-\omega;\omega) \,, \quad \text{(A10)}$$

$${}^{(p)}_{A}\eta^{(q,r,s)}_{DFG}(-\omega;-\omega,\omega,\omega) = {}^{(r)}_{F}\eta^{(s,p,q)}_{GAD}(-\omega;-\omega,\omega,\omega) \,, \quad \text{(A11)}$$

$${}^{(p)}_{A}\zeta^{(q,r,s)}_{DFG}(-\omega;-\omega,\omega,\omega) = -{}^{(r)}_{F}\zeta^{(s,p,q)}_{GAD}(-\omega;-\omega,\omega,\omega) \,. \quad \text{(A12)}$$

These relations reduce from 2 to 1 the number of linear and nonlinear multipolar susceptibilities of (A1)–(A4) involving a quadrupolar electric or dipolar magnetic transition.

Acting with the time-inversion operator R (Ref. 35) on the quantum-mechanical expression for the real and imaginary parts of the linear and nonlinear multipolar susceptibilities in the presence and in the absence of the field **B**, it can be shown that the quantities

$$\begin{aligned}&{}^{(p)}_{e}\eta^{(q)}_{e},\; {}^{(p)}_{e}\zeta^{(q)}_{m},\; {}^{(p)}_{e}\eta^{(q,r,s)}_{eee},\; {}^{(p)}_{e}\zeta^{(q,r,s)}_{eem}\,,\\ &{}^{(p)}_{e}\zeta^{(q)m}_{e},\; {}^{(p)}_{e}\eta^{(q)m}_{m},\; {}^{(p)}_{e}\zeta^{(q,r,s)m}_{eee},\; {}^{(p)}_{e}\eta^{(q,r,s)m}_{eem}\end{aligned} \quad \text{(A13)}$$

are invariant with respect to time inversion. After Birss,[36] we refer to them as i tensors. The other quantities [see Eq. (A13′)]

$$\begin{aligned}&{}^{(p)}_{e}\zeta^{(q)}_{e},\; {}^{(p)}_{e}\eta^{(q)}_{m},\; {}^{(p)}_{e}\zeta^{(q,r,s)}_{eee},\; {}^{(p)}_{e}\eta^{(q,r,s)}_{eem}\,,\\ &{}^{(p)}_{e}\eta^{(q)m}_{e},\; {}^{(p)}_{e}\zeta^{(q)m}_{m},\; {}^{(p)}_{e}\eta^{(q,r,s)m}_{eee},\; {}^{(p)}_{e}\zeta^{(q,r,s)m}_{eem}\end{aligned} \quad \text{(A13′)}$$

undergo a change in sign (c tensors). According to Neumann's principle,[36] c tensors can exist solely in magnetic media (the operation of time inversion does not occur as an independent element of symmetry[36]), whereas i tensors can exist as well in nonmagnetic media.

By having recourse to the permutation relations (A6)–(A12) and to tables giving the shape of i and c tensors of the second, third, fourth, fifth, and sixth rank for all 90 magnetic classes[2,22,36] and the classes Y, Y_h (icosahedral) and K, K_h (complete group of rotations), we find that for crystals with the symmetries $\bar{3}m$, $\underline{\bar{3}m}$, $\bar{6}m2$, $\underline{\bar{6}}m\underline{2}$, $\bar{6}\underline{2}\underline{m}$, $6/mmm$, $6/\underline{m}mm$, $\underline{6}/mm\underline{m}$, $6/\underline{mmm}$, $\underline{6}/m\underline{m}m$, Y_h, and K_h the Hamiltonian (1) takes the form (3) in circular basis and on going over to normal ordering, and that the coupling constants for the magnetic classes $\bar{6}m\underline{2}$ and $6/\underline{m}mm$ take the form

$$\begin{aligned}&\chi^l_R = \underline{b} + d, \quad \chi^l_G(B_z) = \underline{g} + j \,,\\ &\chi^{\rm nl}_R = \underline{A} + D, \quad \mathcal{H}^{\rm nl}_R = \underline{A}' + D' \,,\\ &\chi^{\rm nl}_G(B_z) = \underline{G} + I \,,\end{aligned} \quad \text{(A14)}$$

whereas for the classes $\bar{3}m$, $\underline{\bar{3}m}$, $\bar{6}m2$, $\bar{6}\underline{2m}$, $6/mmm$, $\underline{6}/\underline{m}mm$, $6/\underline{mmm}$, $\underline{6}/m\underline{m}m$, Y_h, K_h and isotropic media with centrosymmetric molecules only the constants underlined in (A14) are nonzero. Above, we have used the following notation:

$$\underline{b} = 2\,{}^{(1)}_{e}\eta^{(1)}_{e\,xx} \,,$$

$$d = \frac{4n}{c}\left[-\frac{\omega}{3}\,{}^{(1)}_{e}\zeta^{(2)}_{e\,x(xz)} + {}^{(1)}_{e}\eta^{(1)}_{m\,xy}\right]s_z \,,$$

$$\underline{g} = -2\,{}^{(1)}_{e}\zeta^{(1)m}_{e\,xyz}B_z \,,$$

$$j = \frac{4n}{c}\left[-\frac{\omega}{3}\,{}^{(1)}_{e}\eta^{(2)m}_{e\,x(yz)z} + {}^{(1)}_{e}\zeta^{(1)m}_{m\,xxz}\right]s_z B_z \,,$$

$$\underline{A} = 24\,{}^{(1)}_{e}\eta^{(1,1,1)}_{eee\,xyxy} \,,$$

$$D = -\frac{96n}{c}\left[\frac{\omega}{3}\,{}^{(1)}_{e}\zeta^{(1,1,2)}_{eee\,xyx(yz)} + {}^{(1)}_{e}\eta^{(1,1,1)}_{eem\,yxxx}\right]s_z \,,$$

$$\underline{A}' = 12({}^{(1)}_{e}\eta^{(1,1,1)}_{eee\,xxxx} + {}^{(1)}_{e}\eta^{(1,1,1)}_{eee\,xxyy}) \,,$$

$$D' = \frac{48n}{c}\left[-\frac{\omega}{3}\left({}_e^{(1)}\zeta_{eee\,xxx(xz)}^{(1,1,2)} + {}_e^{(1)}\zeta_{eee\,xxy(yz)}^{(1,1,2)}\right) + {}_e^{(1)}\eta_{eem\,xxxy}^{(1,1,1)} - {}_e^{(1)}\eta_{eem\,xxyx}^{(1,1,1)}\right]s_z\ ,$$

$$\underline{G} = -24\,{}_e^{(1)}\zeta_{eee\,xxxyz}^{(1,1,1)m}B_z\ ,$$

$$I = \frac{96n}{c}\left[\frac{\omega}{3}\,{}_e^{(1)}\eta_{eee\,xxx(yz)z}^{(1,1,2)m} + {}_e^{(1)}\zeta_{eem\,xyxyz}^{(1,1,1)m}\right]s_z B_z\ .$$

On perusal of (A13) and (A13′) one readily checks that the underlined constants contain i tensors whereas those not underlined contain c tensors. For numerical values of the respective susceptibility tensor components for the various crystal symmetries, we refer to the literature.[37,38]

In the case of crystals with other symmetries the interaction Hamiltonian (3) is of a more highly complicated form since, in addition to the underlined constants which exist for all crystals irrespective of their symmetry, new constants appear, leading moreover to other, new combinations of fields.

ACKNOWLEDGMENT

This work was supported by Polish Academy of Sciences Research Project No. CPBP-01-07.

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