# SELF-SQUEEZING OF ELLIPTICALLY POLARIZED LIGHT PROPAGATING IN A KERR-LIKE OPTICALLY ACTIVE MEDIUM

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## CONTENTS



## I. INTRODUCTION

Quantum and stochastic properties of light fields can, in most cases, be described in terms of coherent states that are quantum field states being as close as possible to classical fields with well-defined amplitude and phase [1–3]. The well-defined diagonal Glauber-Sudarshan quasidistribution $P(\alpha)$ allows for calculations of all relevant mean values of fields that as we say "have classical counterparts." However, there are optical fields that "have no classical counterparts," that is, fields for which the quasidistribution $P(\alpha)$ does not exist as a well-defined, positive definite dis-

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tribution function. Such fields have quantum properties that cannot be explained in the language of classical stochastic quantities. They require fully quantum description, and generation and detection of such field states have been the subject of numerous, both theoretical and experimental, efforts since the mid 1970s. To this day, many nonlinear processes have been analyzed as candidates for producing nonclassical states of light, which include parametric down conversion [4–19], resonance fluorescence [20–27], four-wave mixing [28–35], harmonics generation [36–48], anharmonic oscillator [49–70], light propagation in Kerr media from the point of view of photon statistics [71–73] and squeezing [74–91], and multiphoton absorption and other multiphoton processes [92–189]. The nonclassical properties of light are already the subject of review articles [190–197] and books [198–200], in which the basic information and extensive literature can be found.

Nonclassical effects, such as photon antibunching, sub-Poissonian photon statistics, and squeezing, are a result of nonlinear interaction of quantum light with a nonlinear medium; thus, the nonlinear interaction of light with matter is a crucial element in generation fields with nonclassical properties. The earliest observations of photon antibunching are due to Kimble et al. [23] in resonance fluorescence, confirming the theoretical predictions of Carmichael and Walls [20] and Kimble and Mandel [21]. Sub-Poissonian photon distribution was measured by Short and Mandel [123], and the first observation of squeezing was due to Slusher et al. [33]. Later on a number of successful experiments were performed producing light with nonclassical properties, [10, 11, 27, 32, 33, 169].

Unlike photon antibunching, squeezing is an effect that is sensitive to the phase of the field, the fluctuations of which can essentially reduce its value and even destroy it altogether. The detection of squeezing requires rather sophisticated techniques, such as balanced homodyne detection [201, 202], allowing for the elimination of the local oscillator noise. Despite the differences between photon anticorrelation and squeezing, both processes have one important common feature: Their nature is purely quantum and fields exhibiting such properties have no analogs in classical optics. The two effects can coexist in the same nonlinear process, their areas of existence can be separated, or only one of them can appear in a given process. Especially interesting, in our opinion, is the process of propagation of strong light in a nonlinear Kerr-like medium. Some time ago Tanaś and Kielich [74, 75] showed that almost perfect squeezing can be obtained in such a process, while at the same time photon statistics remain untouched. This process was referred to as self-squeezing, because the squeezing of the quantum field fluctuations is caused by the self-interaction of light via the nonlinear medium. The one-mode version of the process, which is applicable for circularly polarized light propagating in an isotropic Kerr medium, was considered by Tanaś [49] in terms of an anharmonic oscillator model. The model, which allows for exact solutions, became very popular later, and many properties of the field states generated in the model have been revealed and studied [50–70].

Classically, a strong laser field with elliptical polarization is known to rotate its polarization ellipse when propagating through a Kerr medium, an effect observed by Maker et al. [203]. To explain this effect there is no need for field quantization (see, for example, [198]). Here, however, we are interested in effects that are quantum in nature and cannot be explained with the field being classical.

To describe properly the effects associated with the propagation of elliptically polarized light in a Kerr medium, the two-mode description of the field is needed. Such a description was used in the early studies [71–75] of the quantum field effects that appear during propagation. In those studies, the Heisenberg equations of motion for the field operators were solved and their solutions used to calculate appropriate quantities revealing sub-Poissonian photon statistics or squeezing. Recently, Agarwal and Puri [82] reexamined the problem of propagation of elliptically polarized light through a Kerr medium. They discussed not only the Heisenberg equations of motion for the field operators, but also the evolution of the field states themselves. The polarization state of the field propagating in a Kerr medium can be described by the Stokes parameters, which are the expectation values of the corresponding Stokes operators when the quantum description of the field is used. Quantum fluctuations of the Stokes parameters of light propagating in a Kerr medium have recently been discussed by Tanaś and Kielich [204].

In this chapter, we consider propagation of strong light through a macroscopically isotropic, nonlinear medium, taking into account not only electric-dipole contributions to the interaction Hamiltonian, but also contributions from the electric–magnetic dipole and electric–dipole–quadrupole linear and nonlinear susceptibilities of the medium. We introduce general expressions for the effective Hamiltonians of the second and fourth order in the field strength using the circular polarization basis for the field. Such effective Hamiltonians lead to the Heisenberg equations of motion for the field operators that have exact solutions in the form of the translation operator. This means that the field propagating in the nonlinear medium undergoes a nonlinear change in phase (or self-phase modulation), which for quantum fields means essential changes of the quantum state of the field leading to self-squeezing of light. These additional contributions that we take into account mean that our results are valid for media with nonlinear optical activity. It is our aim to calculate the field expectation values describing photon antibunching and squeezing of the field propagating in such a medium.

## II. THE EFFECTIVE INTERACTION HAMILTONIAN

We consider $N$ microsystems (atoms, molecules, or elementary cells in a crystal) confined in a volume $V$ and subjected to the electromagnetic field of a light beam with the electric field vector $\mathbf{E}(\mathbf{r}, t)$ and the magnetic field vector $\mathbf{B}(\mathbf{r}, t)$ in the point $\mathbf{r}$ at time $t$. The total Hamiltonian of such a system has the form

$$H = H_{\mathrm{N}} + H_{\mathrm{F}} + H_{\mathrm{I}} \tag{1}$$

where $H_{\mathrm{N}}$ is the Hamiltonian of the system of $N$ microsystems, and $H_{\mathrm{F}}$ is the Hamiltonian of the free field.

We are interested in the explicit form of the Hamiltonian $H_{\mathrm{I}}$ describing the interaction of the system with the electromagnetic field. This interaction is in general nonlinear and contains all multipolar transitions both electric and magnetic [205–207]. In nonlinear optics we use, for simplicity and convenience, effective interaction Hamiltonians [208], in which it is sufficient to include terms up to the fourth order with respect to the electric and magnetic field strengths [206, 207, 209, 210].

In this chapter we take into account only contributions to the interaction Hamiltonian with even powers of the field strengths

$$H_{\mathrm{I}} = H_{\mathrm{I}}^{(2)} + H_{\mathrm{I}}^{(4)} + \cdots = \sum_{n=1}^{\infty} H_{\mathrm{I}}^{(2n)} \tag{2}$$

Restricting our considerations to the case of weak spatial dispersion (which means that we neglect higher multipoles [209, 210]), we can write for $N$ uncorrelated molecules [211]

$$\begin{aligned} H_{\mathrm{I}}^{(2)} = -\frac{N}{2}\Big\{\alpha_{ij}E_iE_j + \tfrac{1}{3}\big[\eta_{i(jk)}E_i\nabla_k E_j + \eta_{(ik)j}(\nabla_k E_i)E_j\big] \\ +\rho_{ij}E_iB_j + \lambda_{ij}B_iE_j + \text{h.c.}\Big\} \end{aligned} \tag{3}$$

where, according to the Einstein summation convention, the summation over the repeated indices is understood in (3).

In Eq. (3) the second-rank tensor $\alpha_{ij}$ describes the linear electric–electric polarizability of the molecule coming from the electric-dipole–electric-dipole transitions. Similarly, the second-rank pseudotensors $\rho_{ij}$

and $\lambda_{ij}$ denote the polarizabilities: electric–magnetic resulting from the quantum transitions electric dipole–magnetic dipole, and magnetic–electric resulting from the transitions magnetic dipole–electric dipole, respectively. The third-rank tensor $\eta_{i(jk)}$ denotes the linear electric–electric polarizability resulting from the transitions electric dipole–electric quadrupole [209, 210], while the tensor $\eta_{(ij)k}$ comes from the same transitions in reversed order.

In the same multipolar approximation the fourth-order Hamiltonian has the form [206, 207, 209, 210]

$$\begin{aligned} H_{\rm I}^{(4)} = -\frac{N}{24}\Big\{ &\gamma_{ijkl} E_i E_j E_k E_l \\ &+ \tfrac{1}{3}\big[ \eta_{ijk(lm)} E_i E_j E_k \nabla_m E_l + \eta_{ij(km)l} E_i E_j (\nabla_m E_k) E_l \\ &\quad + \eta_{i(jm)kl} E_i (\nabla_m E_j) E_k E_l + \eta_{(im)jkl} (\nabla_m E_i) E_j E_k E_l \big] \\ &+ \kappa_{ijkl} E_i E_j E_k B_l + \rho_{ijkl} E_i E_j B_k E_l \\ &+ \sigma_{ijkl} E_i B_j E_k E_l + \lambda_{ijkl} B_i E_j E_k E_l + \text{h.c.} \Big\} \end{aligned} \tag{4}$$

where the fourth-rank tensor $\gamma_{ijkl}$ denotes the nonlinear polarizability resulting from the four electric-dipole transitions, the fourth-rank pseudotensors $\kappa_{ijkl}$, $\rho_{ijkl}$, and $\sigma_{ijkl}$ denote the nonlinear polarizabilities: electric–magnetic resulting from the transitions electric dipole–magnetic dipole and two electric dipoles, while $\lambda_{ijkl}$ the magnetic–electric polarizability associated with the transitions magnetic dipole–electric dipole and two electric dipoles. The fifth-rank tensor $\eta_{(im)jkl}$ defines the electric quadrupole polarizability associated with the transitions electric quadrupole–electric dipole and two electric dipoles [207, 210]. The remaining tensors $\eta_{i(jm)kl}$, $\eta_{ij(km)l}$, and $\eta_{ijk(lm)}$ differ from the first by the permutation of the position of the electric–quadrupole transition, which is labeled by the indices in parentheses.

For classical fields the electric field vector can be split into two complex conjugate parts [1]:

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}^{(+)}(\mathbf{r}, t) + \mathbf{E}^{(-)}(\mathbf{r}, t) \tag{5}$$

where the components $\mathbf{E}^{(+)}(\mathbf{r}, t)$ and $\mathbf{E}^{(-)}(\mathbf{r}, t)$ are related to the time dependences $\exp(-i\omega t)$ (positive frequency part) and $\exp(+i\omega t)$ (negative frequency part), respectively. The transversal electric field can be expressed as a superposition of plane waves:

$$\mathbf{E}(\mathbf{r},t) = \sum_k \{\mathbf{E}^{(+)}(\mathbf{k})\exp[\mathrm{i}(\mathbf{k}\cdot\mathbf{r} - \omega_k t)] + \mathbf{E}^{(-)}(\mathbf{k})\exp[-\mathrm{i}(\mathbf{k}\cdot\mathbf{r} - \omega_k t)]\} \tag{6}$$

The same decomposition can be performed for the magnetic field vector $\mathbf{B}(\mathbf{r},t)$, where we have the relation (in SI units)

$$B_i^{(+)} = \frac{1}{\omega}\varepsilon_{ijk}k_j E_k^{(+)} \tag{7}$$

where $\varepsilon_{ijk}$ is the Levi-Cività antisymmetric tensor.

As usual, we assume that the light wave is propagating along the $z$ axis of the Cartesian coordinate system $\{x, y, z\}$. It will be convenient later on to use the circular basis associated with the unit vectors of the form (the angular momentum convention is used here)

$$\mathbf{e}_{\pm} = \frac{1}{\sqrt{2}}(\mathbf{x} \pm \mathrm{i}\mathbf{y}) \tag{8}$$

where $\mathbf{e}_+$ describes right and $\mathbf{e}_-$ left polarization of the field. The vectors $\mathbf{x}$ and $\mathbf{y}$ are the unit vectors along the $x$ and $y$ of the Cartesian reference frame, and i is the imaginary unit ($\mathrm{i} = \sqrt{-1}$).

Assuming the microsystems to be freely oriented, the Hamiltonians (3) and (4) have to be averaged over all possible orientations. As a result of such averaging only the rotational invariants of the polarizability tensors appearing in these Hamiltonians will remain [198], which have the form

$$\begin{aligned}\langle\alpha_{ij}\rangle_\Omega &= \alpha\delta_{ij}\\ \langle\eta_{i(jm)}\rangle_\Omega &= \langle\eta_{(im)j}\rangle_\Omega = 0\\ \langle\rho_{ij}\rangle_\Omega &= \rho\delta_{ij} \qquad \langle\lambda_{ij}\rangle_\Omega = \lambda\delta_{ij}\end{aligned} \tag{9}$$

where $\alpha = \alpha_{\alpha\alpha}/3$, $\rho = \rho_{\alpha\alpha}/3$, and $\lambda = \lambda_{\alpha\alpha}/3$ are the mean polarizabilities of the molecules.

In the nonlinear case we have [198]

$$\langle\gamma_{ijkl}\rangle_\Omega = \gamma_1\delta_{ij}\delta_{kl} + \gamma_2\delta_{ik}\delta_{jl} + \gamma_3\delta_{il}\delta_{jk} \tag{10}$$

where

$$\begin{aligned}\gamma_1 &= \tfrac{1}{30}[4\gamma_{\alpha\alpha\beta\beta} - \gamma_{\alpha\beta\alpha\beta} - \gamma_{\alpha\beta\beta\alpha}]\\ \gamma_2 &= \tfrac{1}{30}[-\gamma_{\alpha\alpha\beta\beta} + 4\gamma_{\alpha\beta\alpha\beta} - \gamma_{\alpha\beta\beta\alpha}]\\ \gamma_3 &= \tfrac{1}{30}[-\gamma_{\alpha\alpha\beta\beta} - \gamma_{\alpha\beta\alpha\beta} + 4\gamma_{\alpha\beta\beta\alpha}]\end{aligned} \tag{11}$$

and similar relations hold for the pseudotensors $\kappa_{ijkl}$, $\rho_{ijkl}$, $\sigma_{ijkl}$, and $\lambda_{ijkl}$.

For the nonlinear dipole–quadrupole polarizability we have [198]

$$\langle \eta_{ijk(lm)} \rangle_\Omega = \eta_1 \delta_{ij} \varepsilon_{klm} + \eta_2 \delta_{ik} \varepsilon_{jlm} + \eta_3 \delta_{il} \varepsilon_{jkm} + \eta_4 \delta_{jk} \varepsilon_{ilm} + \eta_5 \delta_{jl} \varepsilon_{ikm} + \eta_6 \delta_{kl} \varepsilon_{ijm} \tag{12}$$

where $\varepsilon_{ijk}$ is the Levi-Cività antisymmetric tensor, while the constants $\eta_1, \eta_2, \ldots, \eta_6$ are defined by the following matrix equation:

$$\begin{pmatrix} \eta_1 \\ \eta_2 \\ \eta_3 \\ \eta_4 \\ \eta_5 \\ \eta_6 \end{pmatrix} = \frac{\eta_{\alpha\beta\gamma(\delta\phi)}}{30} \begin{pmatrix} 3 & -1 & 1 & -1 & 1 & 0 \\ -1 & 3 & -1 & -1 & 0 & 1 \\ 1 & -1 & 3 & 0 & -1 & 1 \\ -1 & -1 & 0 & 3 & -1 & -1 \\ 1 & 0 & -1 & -1 & 3 & -1 \\ 0 & 1 & 1 & -1 & -1 & 3 \end{pmatrix} \begin{pmatrix} \delta_{\alpha\beta}\varepsilon_{\gamma\delta\phi} \\ \delta_{\alpha\gamma}\varepsilon_{\beta\delta\phi} \\ \delta_{\alpha\delta}\varepsilon_{\beta\gamma\phi} \\ \delta_{\beta\gamma}\varepsilon_{\alpha\delta\phi} \\ \delta_{\beta\delta}\varepsilon_{\alpha\gamma\phi} \\ \delta_{\gamma\delta}\varepsilon_{\alpha\beta\phi} \end{pmatrix} \tag{13}$$

The Hamiltonian (6) describing the interaction of $N$ microsystems with the electromagnetic field propagating in a definite direction can be simplified because the summation in (6) is restricted to definite $k$ only. Having this restriction in mind and applying the circular polarization basis (8), we can write the Hamiltonian (6), up to the fourth order in the field strength, in the following form (see Appendix A):

$$\begin{aligned} H_{\mathrm{I}}^{(2)} = & -\tfrac{1}{2}\chi_R^L [E_+^- E_+^+ + E_-^- E_-^+ + E_+^+ E_+^- + E_-^+ E_-^-] \\ & -\tfrac{\mathrm{i}}{2}\chi_A^L [E_+^- E_+^+ - E_-^- E_-^+ + E_+^+ E_+^- - E_-^+ E_-^-] \end{aligned} \tag{14}$$

$$\begin{aligned} H_{\mathrm{I}}^{(4)} = & -\tfrac{1}{12}\chi_R^{NL} \Big[ (E_+^-)^2 (E_+^+)^2 + (E_-^-)^2 (E_-^+)^2 + (E_+^- E_+^+)^2 \\ & \quad + (E_-^- E_-^+)^2 + E_+^- (E_+^+)^2 E_+^- + E_-^- (E_-^+)^2 E_-^- \\ & \quad + \text{terms with reversed superscripts} \Big] \\ & - \tfrac{1}{24}\kappa_R^{NL} [4E_+^- E_-^- E_+^+ E_-^+ + E_+^- E_+^+ E_-^- E_-^+ + E_+^- E_-^+ E_+^+ E_-^- \\ & \quad + E_-^- E_-^+ E_+^- E_+^+ + E_+^- E_-^+ E_-^- E_+^+ + E_-^- E_+^+ E_+^- E_-^+ \\ & \quad + E_+^- E_+^+ E_-^+ E_-^- + E_-^- E_-^+ E_+^+ E_+^- + E_-^- E_+^+ E_-^+ E_+^- \\ & \quad + \text{terms with reversed superscripts}] \\ & - \tfrac{\mathrm{i}}{12}\chi_A^{NL} \Big[ (E_+^-)^2 (E_+^+)^2 - (E_-^-)^2 (E_-^+)^2 + (E_+^- E_+^+)^2 \\ & \quad - (E_-^- E_-^+)^2 + E_+^- (E_+^+)^2 E_+^- - E_-^- (E_-^+)^2 E_-^- \\ & \quad + \text{terms with reversed superscripts} \Big] \end{aligned} \tag{15}$$

where we have introduced the following linear and nonlinear molecular parameters (we neglect local field corrections):

$$\chi_R^L = \frac{N}{3}\operatorname{Re}\alpha_{\alpha\alpha} \tag{16}$$

$$\chi_A^L = -\frac{\mathrm{i}Nk_z}{3\omega}\operatorname{Im}\rho_{\alpha\alpha} \tag{17}$$

$$\chi_R^{NL} = \frac{N}{15}\operatorname{Re}\left[-\gamma_{\alpha\alpha\beta\beta} + 3\gamma_{\alpha\beta\alpha\beta}\right] \tag{18}$$

$$\kappa_R^{NL} = \frac{N}{15}\operatorname{Re}\left[3\gamma_{\alpha\alpha\beta\beta} + \gamma_{\alpha\beta\alpha\beta}\right] \tag{19}$$

$$\chi_A^{NL} = -\frac{\mathrm{i}4Nk_z}{15}\left\{\frac{1}{\omega}\operatorname{Im}\left[\sigma_{\alpha\alpha\beta\beta} - 3\sigma_{\alpha\beta\alpha\beta}\right] - \frac{1}{3}\operatorname{Re}\eta_{\alpha(\beta\gamma)\beta\delta}\varepsilon_{\alpha\gamma\delta}\right\} \tag{20}$$

The real and imaginary parts of the linear and nonlinear polarizabilities are given in the case when the ground state of the molecule is nondegenerate by (A.38); in the case of even degeneracy by (A.42), (A.44), and (A.48); and in the case of odd degeneracy by (A.46), (A.47), and (A.49). The nonzero and independent components of the nonlinear polarizability tensors $\operatorname{Re}\gamma_{\alpha\beta\gamma\delta}$, $\operatorname{Im}\sigma_{\alpha\beta\gamma\delta}$, and $\operatorname{Re}\eta_{\alpha(\beta\gamma)\delta\phi}$ symmetrical with respect to the time reversal are collected in Tables I–III for 102 magnetic point groups of symmetry, whereas in Table IV the linear $\chi_R^L$, $\chi_A^L$ and nonlinear $\chi_R^{NL}$, $\kappa_R^{NL}$, $\chi_A^{NL}$ molecular parameters are collected for 102 magnetic point symmetry groups (Appendix B).

## III. THE SOLUTION OF THE EQUATIONS OF MOTION FOR THE FIELD OPERATORS

In quantum electrodynamics the field vectors (6) and (7) become operators in the Hilbert space, and we have

$$\mathbf{E}^{(+)}(\mathbf{k}) = \mathrm{i}\sum_{\lambda} c(\omega_k)\mathbf{e}^{(\lambda)}(\mathbf{k})\hat{a}_{\mathbf{k}\lambda} \tag{21}$$

where $c(\omega_k)$ is the normalization factor, which, depending on the unit system, has the form

$$c(\omega_k) = \begin{cases} \sqrt{\dfrac{2\pi\hbar\omega_k}{V}} & \text{in CGS} \\ \sqrt{\dfrac{\hbar\omega_k}{2\varepsilon_0 V}} & \text{in SI} \end{cases} \tag{22}$$

where $V$ is the quantization volume.

In Eq. (21) $\hat{a}_{\mathbf{k}\lambda}$ is the annihilation operator of a photon with the momentum $\hbar\mathbf{k}$ and the polarization $\lambda$ defined by the unit vector $\mathbf{e}^{(\lambda)}(\mathbf{k})$. The photon annihilation and creation operators $\hat{a}_{\mathbf{k}\lambda}$ and $\hat{a}^+_{\mathbf{k}\lambda}$ satisfy the boson commutation rules

$$\begin{aligned} \left[\hat{a}_{\mathbf{k}\lambda}, \hat{a}^+_{\mathbf{k}\lambda}\right] &= \delta_{\mathbf{k}\mathbf{k}'}\delta_{\lambda\lambda'} \\ \left[\hat{a}_{\mathbf{k}\lambda}, \hat{a}_{\mathbf{k}\lambda}\right] &= \left[\hat{a}^+_{\mathbf{k}\lambda}, \hat{a}^+_{\mathbf{k}\lambda}\right] = 0 \end{aligned} \tag{23}$$

The unit vectors describing the polarization state of the field are, in general, complex quantities and satisfy the orthonormality conditions

$$e^{(\lambda)*}_{k\sigma} e^{(\lambda')}_{k\tau} = \delta_{\sigma\tau}\delta_{\lambda\lambda'} \qquad e^{(\lambda)}_{k\sigma} k_\sigma = 0 \tag{24}$$

For a quasimonochromatic wave of frequency $\omega$ propagating along the $z$ axis of the laboratory reference frame one can discard the summation over $k$ in Eq. (6) and, in view of (21), write

$$E^{(+)}_\sigma(z,t) = \mathrm{i}c(\omega)\exp[-\mathrm{i}(\omega t - kz)] \sum_{\lambda=1,2} e^{(\lambda)}_\sigma \hat{a}_\lambda \tag{25}$$

where $k = \omega/c$ is the value of the wave vector $\mathbf{k}$.

The field (25) represents, in fact, a two-mode field, when it is a coherent superposition of two modes with orthogonal polarizations. Usually, such two modes can be replaced by a one mode of the field with elliptical polarization

$$e_\sigma \hat{a} = e^{(1)}_\sigma \hat{a}_1 + e^{(2)}_\sigma \hat{a}_2 \tag{26}$$

where $e^{(1)}_\sigma$ and $e^{(2)}_\sigma$ denote $\sigma$ components of the orthogonal unit polarization vectors $\mathbf{e}^{(1)}$ and $\mathbf{e}^{(2)}$ associated with the modes $\hat{a}_1$ and $\hat{a}_2$, and similarly $e_\sigma$ denotes $\sigma$ component of the polarization vector of the mode $\hat{a}$.

The transformation (26) can be interpreted as a decomposition of the initially elliptically polarized light into two orthogonal modes. Taking into account the normalization conditions (24), we get from (26)

$$\hat{a} = e_1^* \hat{a}_1 + e_2^* \hat{a}_2 \tag{27}$$

were

$$e_1^* = e_\sigma^* e_\sigma^{(1)} \qquad e_2^* = e_\sigma^* e_\sigma^{(2)} \tag{28}$$

Assuming the two modes as linearly polarized along $x$ and $y$ we have from (27)

$$\hat{a} = e_x^* \hat{a}_x + e_y^* \hat{a}_y \tag{29}$$

where [212]

$$\begin{aligned} e_x &= \cos\eta \cos\theta - \mathrm{i}\sin\eta \sin\theta \\ e_y &= \cos\eta \sin\theta + \mathrm{i}\sin\eta \cos\theta \end{aligned} \tag{30}$$

with $\theta$ and $\eta$ denoting the azimuth and ellipticity of the polarization ellipse of the incoming light.

Analogously to the circular representation (8) of the polarization vector we can introduce, according to (26) and (29), the circular basis for the field operators:

$$\begin{aligned} \hat{a}_1 &= \hat{a}_+ = \frac{1}{\sqrt{2}}\left(\hat{a}_x - \mathrm{i}\hat{a}_y\right) \\ \hat{a}_2 &= \hat{a}_- = \frac{1}{\sqrt{2}}\left(\hat{a}_x + \mathrm{i}\hat{a}_y\right) \end{aligned} \tag{31}$$

Both representations can be used to describe the interaction of the elliptically polarized light with the medium. However, as has been shown previously [73–75] the circular representation has a clear advantage over the Cartesian representation because it allows for the simple operator solution of the equations of motion in the propagator form.

The time evolution of the field operators is described by the Heisenberg equations of motion:

$$\frac{\partial \mathbf{E}^{(\pm)}(\mathbf{r}, t)}{\partial t} = \frac{1}{\mathrm{i}\hbar}\left[\mathbf{E}^{(\pm)}(\mathbf{r}, t), H\right] \tag{32}$$

For the free field the Hamiltonian $H$ is the free Hamiltonian $H_F$, and the solution to Eq. (32) is given by Eq. (6) describing the free (fast) evolution of the field. When the interaction of the field with the medium comes into play, the Hamiltonian $H$ in (32) contains, beside the free part $H_F$, also the interaction part $H_I$. In this case the solution to (32) is no longer the free field (6), but an additional (slow) time dependence appears. This additional time dependence, which is due to the interaction $H_I$, reveals itself in the fact that the amplitudes $E^{\pm}(k)$ given by Eq. (21) become time dependent.

Usually, one considers the time evolution of a field that is confined in a cavity of volume $V$. In our case, we deal instead with a field propagating in a medium of a certain length $z$. So, instead of the time dependence, we consider the length dependence of the propagating field. However, for plane waves the transition from the cavity problem to the propagation problem can be performed by replacing the time $t$ by $z/c$, where $c$ is the speed of light [208]. Recently, Blow et al. [213] have shown that the correct treatment of the propagation processes requires the continuous-mode description, instead of the discrete-mode description used here. Blow et al. [214] have also shown that the exact solution of the quantum self-phase modulation problem can be obtained within the continuous-mode formalism. This new formalism allows us to avoid an anomalous dependence on the size of the cavity that appears when the discrete-mode formalism is applied to describe propagation effects. However, the essential features of the quantum propagation problem, such as the emergence of photon antibunching and squeezing, can be revealed with the discrete-mode formalism, and we keep using it here.

After the replacement $t \to z/c$, the Heisenberg equations of motion (32) become equations describing the dependence of the operators for the $k$th mode on $z$:

$$\frac{\partial E^{(\pm)}(k;z)}{\partial z} = \frac{1}{\mathrm{i}\hbar c}\left[E^{(\pm)}(k;z), H\right] \tag{33}$$

The next essential step is to write down the quantum form of the effective interaction Hamiltonian, which for classical fields is given by Eqs. (14) and (15). We obtain the effective interaction Hamiltonian by inserting the quantum form (25) of the field into (14) and (15) and taking the normal order of the field operators in all terms. This leads us to the following expressions for the interaction Hamiltonian:

$$H_I^{(2)} = -\tilde{\chi}_R^L\left(\hat{a}_+^+\hat{a}_+ + \hat{a}_-^+\hat{a}_-\right) - \mathrm{i}\tilde{\chi}_A^L\left(\hat{a}_+^+\hat{a}_+ - \hat{a}_-^+\hat{a}_-\right) \tag{34}$$

$$\begin{aligned} H_I^{(4)} = &-\tfrac{1}{2}\tilde{\chi}_R^{NL}\left(\hat{a}_+^{+2}\hat{a}_+^2 + \hat{a}_-^{+2}\hat{a}_-^2\right) - \tilde{\kappa}_R^{NL}\hat{a}_+^+\hat{a}_-^+\hat{a}_-\hat{a}_+ \\ &-\frac{\mathrm{i}}{2}\tilde{\chi}_A^{NL}\left(\hat{a}_+^{+2}\hat{a}_+^2 - \hat{a}_-^{+2}\hat{a}_-^2\right) \end{aligned} \tag{35}$$

where we have introduced the notation

$$\tilde{\chi}_R^L = c(\omega)^2 \chi_R^L \qquad \tilde{\chi}_A^L = c(\omega)^2 \chi_A^L \tag{36}$$

$$\begin{aligned} \tilde{\chi}_R^{NL} &= c(\omega)^4 \chi_R^{NL} \qquad \tilde{\kappa}_R^{NL} = c(\omega)^4 \kappa_R^{NL} \\ \tilde{\chi}_A^{NL} &= c(\omega)^4 \chi_A^{NL} \end{aligned} \tag{37}$$

According to (33)–(35) we get the equation of motion for the field annihilation operators (free evolution has been eliminated):

$$\begin{aligned} \frac{\mathrm{d}}{\mathrm{d}z}\hat{a}_\pm(z) = \frac{\mathrm{i}}{\hbar c}\Big\{\tilde{\chi}_R^L \pm \mathrm{i}\tilde{\chi}_A^L + \left[\tilde{\chi}_R^{NL} \pm \mathrm{i}\tilde{\chi}_A^{NL}\right]\hat{a}_\pm^+(z)\hat{a}_\pm(z) \\ + \tilde{\kappa}_R^{NL}\hat{a}_\mp^+(z)\hat{a}_\mp(z)\Big\}\hat{a}_\pm(z) \end{aligned} \tag{38}$$

Since $\hat{a}_+^+\hat{a}_+$ and $\hat{a}_-^+\hat{a}_-$ are constants of motion, Eq. (38) has an exact solution in the form of the translation operator

$$\begin{aligned} \hat{a}_\pm(z) = \exp\{\mathrm{i}[\varphi_\pm(z) + \varepsilon_\pm(z)\hat{a}_\pm^+(0)\hat{a}_\pm(0) \\ + \delta(z)\hat{a}_\mp^+(0)\hat{a}_\mp(0)]\}\hat{a}_\pm(0) \end{aligned} \tag{39}$$

where the notation is the following:

$$\begin{aligned} \varphi_\pm(z) &= \frac{z}{\hbar c}\left(\tilde{\chi}_R^L \pm \mathrm{i}\tilde{\chi}_A^L\right) \\ \varepsilon_\pm(z) &= \frac{z}{\hbar c}\left(\tilde{\chi}_R^{NL} \pm \mathrm{i}\tilde{\chi}_A^{NL}\right) \\ \delta(z) &= \frac{z}{\hbar c}\tilde{\kappa}_R^{NL} \end{aligned} \tag{40}$$

Taking into account the fact that the component of the dipole polarization is by definition given by [211]

$$\hat{P}^+ = -\frac{\partial H_\mathrm{I}}{\partial E^-} \tag{41}$$

and that in the circular basis

$$\left(n_\pm^2 - 1\right)E_\pm^+ = 4\pi P_\pm^+ \tag{42}$$

we get, in view of (34) and (35), for the refractive indices of the right and left circularly polarized waves

$$n_{\pm}^2 - 1 = \frac{4\pi}{c(\omega)^2}\left\{\tilde{\chi}_R^L \pm i\tilde{\chi}_A^L + \left[\tilde{\chi}_R^{NL} \pm i\tilde{\chi}_A^{NL}\right]\hat{a}_{\pm}^{+}\hat{a}_{\pm} + \tilde{\kappa}_R^{NL}\hat{a}_{\mp}^{+}\hat{a}_{\mp}\right\} \quad (43)$$

This gives us for the circular optical birefringence in the presence of strong light the formula

$$n_{+}^2 - n_{-}^2 = \frac{4\pi}{c(\omega)^2}\left\{2i\tilde{\chi}_A^L + \left[\tilde{\chi}_R^{NL} - \tilde{\kappa}_R^{NL}\right]\left(\hat{a}_{+}^{+}\hat{a}_{+} - \hat{a}_{-}^{+}\hat{a}_{-}\right) + i\tilde{\chi}_A^{NL}\left(\hat{a}_{+}^{+}\hat{a}_{+} + \hat{a}_{-}^{+}\hat{a}_{-}\right)\right\} \quad (44)$$

Formula (44) is the quantum counterpart of the earlier obtained [215] classical formula in which the first term denotes the natural optical activity of the medium, the second term denotes the rotation of the polarization ellipse induced by the strong field [203], and the third term denotes the nonlinear change in optical activity caused by the strong field [198, 212, 216]. Taking into account (43) and (44), we rewrite the field (39) in the form known from ellipsometry:

$$\hat{a}_{\pm}(z) = \exp(i\alpha \pm i\phi)\hat{a}_{\pm}(0) \quad (45)$$

where we have by definition

$$\alpha = \frac{1}{2}\frac{\omega}{c}(n_{+} + n_{-})z = \alpha_0 + \delta\alpha \quad (46)$$

$$\phi = \frac{1}{2}\frac{\omega}{c}(n_{+} - n_{-}) = \phi_0 + \delta\phi \quad (47)$$

where $\phi$ is the angle of rotation of the polarization ellipse after the field passed the path $z$ in the medium. In the absence of a strong field we have

$$a_0 = \frac{z}{\hbar c}\tilde{\chi}_R^L \quad (48)$$

$$\phi_0 = \frac{z}{\hbar c}\tilde{\chi}_A^L \quad (49)$$

while the changes due to the strong field are given by

$$\delta\alpha = \frac{z}{2\hbar c}\left\{\left[\tilde{\chi}_R^{NL} + \tilde{\kappa}_R^{NL}\right]\left(\hat{a}_+^+\hat{a}_+ + \hat{a}_-^+\hat{a}_-\right) + \mathrm{i}\tilde{\chi}_A^{NL}\left(\hat{a}_+^+\hat{a}_+ - \hat{a}_-^+\hat{a}_-\right)\right\} \quad (50)$$

$$\delta\phi = \frac{z}{2\hbar c}\left\{\left[\tilde{\chi}_R^{NL} - \tilde{\kappa}_R^{NL}\right]\left(\hat{a}_+^+\hat{a}_+ - \hat{a}_-^+\hat{a}_-\right) + \mathrm{i}\tilde{\chi}_A^{NL}\left(\hat{a}_+^+\hat{a}_+ + \hat{a}_-^+\hat{a}_-\right)\right\} \quad (51)$$

If in particular the light wave is linearly polarized, we have ($a_+ = a_- = a/\sqrt{2}$)

$$\delta\alpha = \frac{z}{2\hbar c}\left[\tilde{\chi}_R^{NL} + \tilde{\kappa}_R^{NL}\right]\hat{a}^+\hat{a} \quad (52)$$

$$\delta\phi = \frac{\mathrm{i}z}{2\hbar c}\tilde{\chi}_A^{NL}\hat{a}^+\hat{a} \quad (53)$$

and we see that $\delta\phi$ appears only for media with nonlinear optical activity.

A word of caution should be added here. Since the quantum counterparts to the classical formulas describing nonlinear changes of the refractive index of the medium are extracted from the operator solution for the field operators, and since the mean value of the product of two operators is not the product of their mean values, these formulas cannot be treated too seriously as the quantum expressions for the nonlinear refractive index. They simply allow for the identification of particular contributions, as in classical description, but in fact the complete solutions for the field operators enter the experimentally measurable quantities, such as light intensity, photon correlation functions, and field variances. This will become clear in the next sections.

## IV. PHOTON STATISTICS

Since in the isotropic medium described by the Hamiltonian given in (34) and (35) the photon number operators $\hat{a}_+^+\hat{a}_+$ and $\hat{a}_-^+\hat{a}_-$ are constants of motion (they commute with the Hamiltonian), any function of these operators is also a constant of motion and, as a result, photon statistics of the circular components of the field do not change in the course of propagation. If the component before entering the nonlinear medium is, say, in a coherent state with the Poissonian photon distribution, the photon distribution of the beam outgoing from the medium will remain Poissonian despite the fact that, as we show in the next section, the state of the field is no longer the coherent state. So, if there are no sub-Poissonian photon statistics of the incoming beam, there will be no sub-Poissonian photon statistics in either circular component of the outgoing beam.

However, one can easily check [74, 75] that the linear polarization is not preserved during the propagation of quantum light through the nonlinear isotropic medium and this leads to the sub-Poissonian photon statistics, which is our subject in this section.

Let us take, for example, the component of the linear polarization along the $x$ axis. The photon number operator $\hat{a}_x^+\hat{a}_x$ of this component does not commute with the interaction Hamiltonian, which means that the photon statistics of this component can change due to the interaction with the medium. Knowing the solutions (39) for the field operators $\hat{a}_+(z)$ and $\hat{a}_-(z)$, for the circular components, we can use relation (31) to write down corresponding solutions for the operators $\hat{a}_x(z)$ and $\hat{a}_y(z)$. These solutions allow us to find any characteristic for the polarization component $x$ or $y$ outgoing from the medium if we know the state of the field at the input. To choose one we can place a polarizer after the medium. Thus, assuming that the incoming field is in the coherent state with elliptical polarization defined by the azimuth $\theta$ and the ellipticity $\eta$, we get, for the mean number of photons with the polarization $x$ after the path $z$ passed by the light in the medium, the following expression:

$$\begin{aligned}\langle \hat{a}_x^+(z)\hat{a}_x(z)\rangle &= \tfrac{1}{2}\langle[\hat{a}_+^+(z) + \hat{a}_-^+(z)][\hat{a}_+(z) + \hat{a}_-(z)]\rangle \\ &= \tfrac{1}{2}\left(|\alpha_+|^2 + |\alpha_-|^2\right) + \mathrm{Re}\left\{\alpha_+^*\alpha_- \exp\left[-\mathrm{i}(\varphi_+ - \varphi_-)\right.\right. \\ &\qquad \left.\left. + (\mathrm{e}^{-\mathrm{i}(\varepsilon_+ - \delta)} - 1)|\alpha_+|^2 + (\mathrm{e}^{-\mathrm{i}(\varepsilon_- - \delta)} - 1)|\alpha_-|^2\right]\right\}\end{aligned} \tag{54}$$

where

$$\begin{aligned}\alpha_+ &= \frac{\alpha}{\sqrt{2}}(\cos\eta + \sin\eta)\,\mathrm{e}^{-\mathrm{i}\theta} \\ \alpha_- &= \frac{\alpha}{\sqrt{2}}(\cos\eta - \sin\eta)\,\mathrm{e}^{\mathrm{i}\theta}\end{aligned} \tag{55}$$

and $\alpha$ is the eigenvalue of the annihilation operator of the incoming light that we assume as being in the coherent state:

$$\hat{a}(0)\ |\alpha\rangle = \alpha\ |\alpha\rangle \tag{56}$$

Thus, $|\alpha|^2 = |\alpha_+|^2 + |\alpha_-|^2$ is the mean number of photons of the incoming field. The quantities $\varphi_\pm = \varphi_\pm(z)$, $\varepsilon_\pm = \varepsilon_\pm(z)$, and $\delta = \delta(z)$ are given by Eq. (40). To shorten the notation we shall omit the argument $z$.

In view of (55), expression (54) can be rewritten in a slightly different form:

$$\langle \hat{a}_x^+(z)\hat{a}_x(z)\rangle = \frac{|\alpha|^2}{2}[1 + \cos 2\eta \exp B \cos(2\theta + C)] \tag{57}$$

where

$$\begin{aligned} B = {} & \frac{|\alpha|^2}{2}(1 + \sin 2\eta)[\cos(\varepsilon_+ - \delta) - 1] \\ & + \frac{|\alpha|^2}{2}(1 - \sin 2\eta)[\cos(\varepsilon_- - \delta) - 1] \end{aligned} \tag{58}$$

$$\begin{aligned} C = {} & -(\varphi_+ - \varphi_-) - \frac{|\alpha|^2}{2}(1 + \sin 2\eta)\sin(\varepsilon_+ - \delta) \\ & + \frac{|\alpha|^2}{2}(1 - \sin 2\eta)\sin(\varepsilon_- - \delta) \end{aligned} \tag{59}$$

Formula (57) has been obtained with the effective use of the commutation relations (23); that is, to obtain it we have taken into account the quantum properties of the field. Just for reference, it is worth remembering that the corresponding formula for classical fields reads

$$\begin{aligned} \langle \hat{a}_x^+(z)\hat{a}_x(z)\rangle_{\text{class}} & \\ = {} & \frac{1}{2}\left(|\alpha_+|^2 + |\alpha_-|^2\right) + \mathrm{Re}\Big\{\alpha_+^*\alpha_- \exp\big[-\mathrm{i}(\varphi_+ - \varphi_-) \\ & \qquad -\mathrm{i}(\varepsilon_+ - \delta)|\alpha_+|^2 + \mathrm{i}(\varepsilon_- - \delta)|\alpha_-|^2\big]\Big\} \\ = {} & \frac{|\alpha|^2}{2}\Bigg\{1 + \cos 2\eta \cos\Bigg[2\theta - (\varphi_+ - \varphi_-) \\ & \qquad - \frac{|\alpha|^2}{2}(1 + \sin 2\eta)(\varepsilon_+ - \delta) + \frac{|\alpha|^2}{2}(1 - \sin 2\eta)(\varepsilon_- - \delta)\Bigg]\Bigg\} \\ = {} & \frac{|\alpha|^2}{2}\Bigg\{1 + \cos 2\eta \cos\Bigg[2\theta - (\varphi_+ - \varphi_-) \\ & \qquad - \frac{|\alpha|^2}{2}\sin 2\eta(\varepsilon_+ + \varepsilon_- - 2\delta) - \frac{|\alpha|^2}{2}(\varepsilon_+ - \varepsilon_-)\Bigg]\Bigg\} \end{aligned} \tag{60}$$

In formula (60), as in formula (44) one can identify particular effects related to the propagation of light in the nonlinear medium. Namely,

$\varphi_+ - \varphi_-$ describes the natural optical activity, $\varepsilon_+ + \varepsilon_- - 2\delta$ describes the rotation of the polarization ellipse induced by the strong light (because of the $\sin 2\eta$ factor appearing in this term, the elliptical polarization of the field is necessary to observe this effect), and $\varepsilon_+ - \varepsilon_-$ describes the nonlinear change in optical activity.

To make the difference between the quantum formula (57) and its classical counterpart (60) more explicit, let us assume that the medium is composed of optically inactive molecules; then, we have $\varphi_+ - \varphi_- = 0$ and $\varepsilon_+ = \varepsilon_- = \varepsilon$. Moreover, assume that the incoming field is linearly polarized ($\eta = 0$) with the azimuth $\theta = \pi/2$, that is, perpendicularly to the observed polarization component. In this case the classical formula (60) gives zero, whereas the quantum formula (57) is different from zero because of the exponential function appearing in it. This means that for quantum fields during the propagation in the nonlinear isotropic medium, photons with the polarization orthogonal to the polarization of the incoming field will appear. In other words, in the nonlinear medium the linear polarization of the field is not preserved, an effect already discussed by Ritze [73]. The quantum effects in the polarization of light propagating in a Kerr medium have been recently discussed in more detail by Tanaś and Gantsog [217].

Now, we come back to the main topic of this section, that is, the problem of sub-Poissonian photon statistics. To convince ourselves whether the field outgoing from the nonlinear medium exhibits sub-Poissonian photon statistics, we have to calculate the second-order correlation function $\langle \hat{a}_x^{+2}(z)\hat{a}_x^2(z)\rangle$. Applying the solutions (39), and assuming that the incoming beam is in the coherent state $|\alpha\rangle$, we arrive at

$$\begin{aligned}
\langle \hat{a}_x^{+2}(z)\hat{a}_x^2(z)\rangle &= \tfrac{1}{4}\left\langle [\hat{a}_+^+(z) + \hat{a}_-^+(z)]^2[\hat{a}_+(z) + \hat{a}_-(z)]^2\right\rangle \\
&= \tfrac{1}{4}\left(|\alpha_+|^4 + |\alpha_-|^4 + 4|\alpha_+|^2|\alpha_-|^2\right) \\
&\quad + \tfrac{1}{2}\mathrm{Re}\Big\{\alpha_+^{*2}\alpha_-^2 \exp\big[-2\mathrm{i}(\varphi_+ - \varphi_-) - \mathrm{i}(\varepsilon_+ - \varepsilon_-) \\
&\qquad + (e^{-2\mathrm{i}(\varepsilon_+ - \delta)} - 1)|\alpha_+|^2 + (e^{2\mathrm{i}(\varepsilon_- - \delta)} - 1)|\alpha_-|^2\big] \\
&\qquad + 2|\alpha_+|^2\alpha_+^*\alpha_- \exp\big[-\mathrm{i}(\varphi_+ - \varphi_-) - \mathrm{i}(\varepsilon_+ - \delta) \\
&\qquad + (e^{-\mathrm{i}(\varepsilon_+ - \delta)} - 1)|\alpha_+|^2 + (e^{\mathrm{i}(\varepsilon_- - \delta)} - 1)|\alpha_-|^2\big] \\
&\qquad + 2|\alpha_-|^2\alpha_-^*\alpha_+ \exp\big[+\mathrm{i}(\varphi_+ - \varphi_-) - \mathrm{i}(\varepsilon_- - \delta) \\
&\qquad + (e^{\mathrm{i}(\varepsilon_+ - \delta)} - 1)|\alpha_+|^2 + (e^{-\mathrm{i}(\varepsilon_- - \delta)} - 1)|\alpha_-|^2\big]\Big\}
\end{aligned} \tag{61}$$

where $\alpha_+$ and $\alpha_-$ are given by (55).

Light is said to exhibit sub-Poissonian photon statistics if

$$\langle \hat{a}_x^{+2}(z)\hat{a}_x^2(z)\rangle - \langle \hat{a}_x^+(z)\hat{a}_x(z)\rangle^2 < 0 \tag{62}$$

Expression (61) is quite complicated and it is not easy to say without numerical analysis whether condition (62) can be satisfied. Usually, the normalized second-order correlation function is considered; it is defined by the relation

$$g^{(2)}(z) = \frac{\langle \hat{a}_x^{+2}(z)\hat{a}_x^2(z)\rangle}{\langle \hat{a}_x^+(z)\hat{a}_x(z)\rangle^2} \tag{63}$$

and condition (62) can then be written as

$$g^{(2)}(z) - 1 < 0 \tag{64}$$

Another measure of the sub-Poissonian photon statistics is the $q$ parameter introduced by Mandel [41] and defined as

$$q = \frac{\langle(\Delta\hat{n})^2\rangle}{\langle\hat{n}\rangle} - 1 = \left[g^{(2)}(z) - 1\right]\langle \hat{a}_x^+\hat{a}_x\rangle \tag{65}$$

Negative values of the parameter $q$ denote sub-Poissonian photon statistics, and the limit $q = -1$ is reached for number states without photon number fluctuations.

In Fig. 1 we plot both $g^{(2)}(z) - 1$ and $q(z)$ against $(\varepsilon - \delta)|\alpha|^2$ for a medium containing optically inactive molecules, and for the elliptical polarization of the incoming light with $\eta = \pi/8$ and $\theta = -\pi/4$. Both functions show oscillatory behavior with both negative and positive values. Negative values of these functions mean the sub-Poissonian photon statistics of the $x$ component of the outgoing field. For optically nonactive molecules they depend only on the molecular parameter $\tilde{\gamma}_1(\omega)$,

$$\varepsilon(z) - \delta(z) = -\frac{2Nz}{\hbar c}\tilde{\gamma}_1(\omega) \tag{66}$$

which in Fig. 1 we have assumed as equal to $1 \times 10^{-6}$, according to the estimation made by Ritze and Bandilla [71]. To get values of $(\varepsilon - \delta)|\alpha|^2$ of the order of unity, a field is needed with the mean number of photons $|\alpha|^2 \approx 10^6$. From Fig. 1 it is seen that the values of $g^{(2)}(z) - 1$ obtained in this process are rather small—of the order $\varepsilon - \delta$. However, the $q$ parameter reaches the value $-0.63$, which means considerable narrowing of the

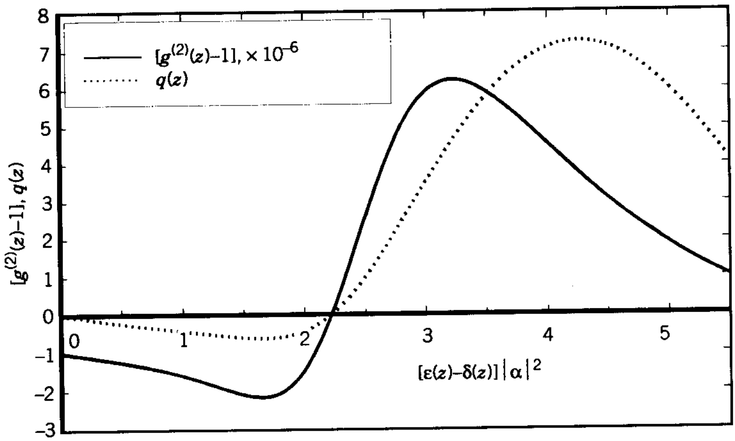


**Figure 1.** Plots of $g^{(2)}(z) - 1$ (scaled in units $1 \times 10^{-6}$) and $q(z)$ against the scaled intensity of light $[\varepsilon(z) - \delta(z)]|\alpha|^2$, assuming that $\varepsilon(z) - \delta(z) = 1 \times 10^{-6}$, $\theta = -\pi/4$, and $\eta = \pi/8$.

photon number distribution for strong fields. If the value $q = -1$ means 100% reduction of the photon number fluctuations, a 63% reduction can be obtained in the propagation process considered here. The possibility to get sub-Poissonian photon statistics in this process was predicted by Tanaś and Kielich [72] with the perturbative method, and confirmed by Ritze [73], who obtained exact solutions to the problem.

For optically active molecules, $\varphi_+ - \varphi_+ \neq 0$ and $\varepsilon_+ - \varepsilon_- \neq 0$, the general form of the solutions given by (57) and (61) must be used. If, however, $\tilde{\gamma}_1(\omega) = 0$, then the solutions simplify considerably, because

$$\varepsilon_+ - \delta = -(\varepsilon_- - \delta) = \mathrm{i}\frac{Nz}{\hbar c}\left[\tilde{\sigma}_2(\omega) + \tilde{\sigma}_3(\omega)\right] = \sigma \tag{67}$$

We obtain, in this case,

$$\begin{aligned}\langle \hat{a}_x^+(z)\hat{a}_x(z)\rangle = \frac{|\alpha|^2}{2}\Big\{1 + \cos 2\eta \exp\left[|\alpha|^2(\cos\sigma - 1)\right] \\ \times\cos\left[2\theta - (\varphi_+ - \varphi_-) - |\alpha|^2 \sin\sigma\right]\Big\}\end{aligned} \tag{68}$$

$$\begin{aligned}\langle \hat{a}_x^{+2}(z)\hat{a}_x^2(z)\rangle = \frac{|\alpha|^4}{4}\Big\{1 + \frac{1}{2}\cos^2 2\eta + \frac{1}{2}\cos^2 2\eta \exp D \cos E \\ + 2\cos 2\eta \exp F \cos G\Big\}\end{aligned} \tag{69}$$

where

$$\begin{aligned} D &= |\alpha|^2(\cos 2\sigma - 1) \\ E &= 4\theta - 2(\varphi_+ - \varphi_-) - 2\sigma - |\alpha|^2 \sin 2\sigma \\ F &= |\alpha|^2(\cos \sigma - 1) \\ G &= 2\theta - (\varphi_+ - \varphi_-) - \sigma - |\alpha|^2 \sin \sigma \end{aligned} \tag{70}$$

Expressions (68)–(70) depend upon two molecular parameters:

$$\varphi_+ - \varphi_- = \frac{\mathrm{i}Nz}{\hbar c}\tilde{\rho} \quad \text{and} \quad \sigma = \frac{\mathrm{i}Nz}{\hbar c}\left[\tilde{\sigma}_2(\omega) + \tilde{\sigma}_3(\omega)\right]$$

describing the natural and nonlinear optical activity.

All the expressions derived here are valid for arbitrary polarization of the incoming beam defined by the parameters $\theta$ and $\eta$. It is interesting to note that for the circular polarization of entering light ($\eta = \pm\pi/4$) we have

$$\begin{aligned} \langle \hat{a}_x^+(z)\hat{a}_x(z)\rangle &= \frac{|\alpha|^2}{2} \\ \langle \hat{a}_x^{+2}(z)\hat{a}_x^2(z)\rangle &= \frac{|\alpha|^4}{4} \end{aligned} \tag{71}$$

which means $g^2(z) - 1 = 0$. This result is not surprising in view of our earlier discussion concerning the photon statistics of circular components of light propagating in a Kerr medium. In fact, it confirms our statement that the photon statistics of such light do not change during the propagation. The polarizer choosing the component $x$ only reduces the intensity of the beam to one-half of the incoming intensity, but its statistics remain unchanged. Any deviation from the circular polarization of the incoming light will cause, as is easy to check, changes in the photon statistics of light propagating in the medium.

## V. SQUEEZING

The fact that the photon statistics of elliptically polarized light do not change when the light propagates through the isotropic nonlinear medium does not mean that the state of the field does not change during such interaction. It turns out that the field can become squeezed as a result of such interaction; that is, it can be in a squeezed state, which has no classical analog and requires quantum interpretation. To show this, we introduce two Hermitian field operators $\hat{Q}_\sigma$ and $\hat{P}_\sigma$ defined as [41, 193]

$$\hat{Q}_\sigma = \hat{a}_\sigma + \hat{a}_\sigma^+ \qquad \hat{P}_\sigma = -\mathrm{i}\left(\hat{a}_\sigma - \hat{a}_\sigma^+\right) \tag{72}$$

where $\sigma$ denotes $+(-)$ in the circular basis or $x(y)$ in the Cartesian basis. The operators $\hat{Q}_\sigma$ and $\hat{P}_\sigma$ satisfy the commutation rules

$$\left[\hat{Q}_\sigma, \hat{P}_{\sigma'}\right] = 2\mathrm{i}\delta_{\sigma\sigma'} \tag{73}$$

A squeezed state of the electromagnetic field is defined [195] as a state of the field in which the variance of $\hat{Q}_\sigma$ or $\hat{P}_\sigma$ is smaller than unity

$$\left\langle\left(\Delta\hat{Q}_\sigma\right)^2\right\rangle < 1 \quad \text{or} \quad \left\langle\left(\Delta\hat{P}_\sigma\right)^2\right\rangle < 1 \tag{74}$$

where $\Delta\hat{Q}_\sigma = \hat{Q}_\sigma - \langle\hat{Q}_\sigma\rangle$. On introducing the normal order of the creation and annihilation operators, definition (74) can be rewritten in the form [41]

$$\left\langle:\left(\Delta\hat{Q}_\sigma\right)^2:\right\rangle < 0 \quad \text{or} \quad \left\langle:\left(\Delta\hat{P}_\sigma\right)^2:\right\rangle < 0 \tag{75}$$

To calculate the quantities occurring in definition (75) for the process of light propagation in the nonlinear medium considered here, it suffices to insert into (75) the operator solutions (39) and next calculate the expectation value in the initial state of the field, which we assume to be the coherent state $|\alpha\rangle$ defined by (56). If one of the normally ordered variances appears to be negative, then the corresponding component of the field is in a squeezed state, which has no classical analog. Our calculations give for the normally ordered variances of the resulting field the following expressions:

$$\begin{aligned}
&\left\langle:\left[\Delta\hat{Q}_\pm(z)\right]^2:\right\rangle \\
&\quad= \left\langle:\left[\hat{a}_\pm(z) + \hat{a}_\pm^+(z)\right]^2:\right\rangle - \left\langle\hat{a}_\pm(z) + \hat{a}_\pm^+(z)\right\rangle^2 \\
&\quad= 2\,\mathrm{Re}\Big\{\alpha_\pm^2 \exp\left[2\mathrm{i}\varphi_\pm + \varepsilon_\pm + \left(\mathrm{e}^{2\mathrm{i}\varepsilon_\pm} - 1\right)|\alpha_\pm|^2 + \left(\mathrm{e}^{2\mathrm{i}\delta} - 1\right)|\alpha_\mp|^2\right. \\
&\qquad\qquad \left. - \alpha_\pm^2 \exp\left[2\mathrm{i}\varphi_\pm + 2\left(\mathrm{e}^{\mathrm{i}\varepsilon_\pm} - 1\right)|\alpha_\pm|^2 + 2\left(\mathrm{e}^{\mathrm{i}\delta} - 1\right)|\alpha_\mp|^2\right]\right\} \\
&\qquad + 2|\alpha_\pm|^2\left\{1 - \exp\left[2(\cos\varepsilon_\pm - 1)|\alpha_\pm|^2 + 2(\cos\delta - 1)|\alpha_\mp|^2\right]\right\}
\end{aligned} \tag{76}$$

where $\alpha_{\pm}$ are given by (55), while $\varphi_{\pm} = \varphi_{\pm}(z)$, $\varepsilon = \varepsilon(z)$, and $\delta = \delta(z)$ are given by (40). For the operators $\hat{P}_{\pm}$ we get

$$\left\langle :\left[\Delta \hat{P}_{\pm}(z)\right]^2: \right\rangle = -2\,\mathrm{Re}\{\cdots\} + 2|\alpha_{\pm}|^2\{\cdots\} \tag{77}$$

where the expressions in the braces are the same as in (76).

Especially interesting is the case of circularly polarized incoming field, because photon statistics of such a field do not change. Let us assume that the incoming beam is circularly polarized with $\eta = \pi/4$ and $\theta = 0$. Then $|\alpha_+|^2 = |\alpha|^2$, $|\alpha_-|^2 = 0$, and formula (76) takes the much simpler form

$$\begin{aligned} \left\langle :\left[\Delta \hat{Q}_{+}(z)\right]^2: \right\rangle = 2|\alpha|^2 \Big\{ &\exp\left[|\alpha|^2(\cos 2\varepsilon_+ - 1)\right]\cos\left(\varphi_+ + \varepsilon_+ + |\alpha|^2 \sin 2\varepsilon_+\right) \\ &- \exp\left[2|\alpha|^2(\cos\varepsilon_+ - 1)\right]\cos\left(\varphi_+ + 2|\alpha|^2 \sin\varepsilon_+\right)\Big\} \\ &+ 2|\alpha|^2\Big\{1 - \exp\left[2|\alpha|^2(\cos\varepsilon_+ - 1)\right]\Big\} \end{aligned} \tag{78}$$

Similarly (77) goes over into

$$\left\langle :\left[\Delta \hat{P}_{+}(z)\right]^2: \right\rangle = -2|\alpha|^2\{\cdots\} + 2|\alpha|^2\{\cdots\} \tag{79}$$

with the contents of the braces the same as in (78).

Assuming the initial phase $\varphi_0$ so that $\varphi_+ + \varphi_0 = 0$, and assuming the value $\varepsilon_+(z) = 1 \times 10^{-6}$, similarly as for the photon statistics case, we have plotted in Fig. 2 expressions (78) and (79) as functions of $\varepsilon_+ |\alpha|^2$. It is seen that the normally ordered variances (78) and (79) exhibit oscillatory behavior on $\varepsilon_+ |\alpha|^2$, taking both positive and negative values. Whenever one of the variances takes negative values, the corresponding component of the field $\hat{Q}_+(z)$ or $\hat{P}_+(z)$ is said to be squeezed. This means that despite the Poissonian photon statistics, the field can be in a squeezed state. It is also worth noting that the values of squeezing possible in the propagation process are quite large. With our definition of the operators $\hat{Q}_+$ and $\hat{P}_+$ the value allowed by quantum mechanics for (78) or (79) is minus unity, which means no quantum fluctuations in the corresponding component of the field. It is seen from Fig. 2 that the first minimum of $\langle :[\Delta \hat{Q}_+(z)]^2: \rangle$ has a value of $-0.66$, while the second minimum already has a value of $-0.97$, which means 97% of the value allowed by quantum mechanics. This result can even be improved by tuning the initial phase $\varphi_0$ [75]; this means a reduction of the quantum fluctuations in the field by two orders of magnitude with respect to the fluctuations in the vacuum (or a coherent state). The first minimum of $\langle :[\Delta \hat{P}_+(z)]^2: \rangle$ has a value of $-0.92$, which also means a considerable reduction of quantum fluctuations. Expressions (78) and (79) after corresponding changes of the variables become identical to the results obtained for the anharmonic oscillator [49].

For the $x$ component of the polarization of the incoming beam, we get for the normally ordered variances the following formulas:

$$\begin{aligned}
&\left\langle :[\Delta\hat{Q}_x(z)]^2: \right\rangle \\
&= \left\langle :[\hat{a}_x(z) + \hat{a}_x^+(z)]^2: \right\rangle - \langle \hat{a}_x(z) + \hat{a}_x^+(z)\rangle^2 \\
&= \tfrac{1}{2}\Big\{\left\langle :[\hat{a}_+(z) + \hat{a}_-(z) + \hat{a}_+^+(z) + \hat{a}_-^+(z)]^2: \right\rangle \\
&\qquad - \langle \hat{a}_+(z) + \hat{a}_-(z) + \hat{a}_+^+(z) + \hat{a}_-^+(z)\rangle^2\Big\} \\
&= \mathrm{Re}\Big\{\alpha_+^2 \exp\left[2\mathrm{i}\varphi_+ + \mathrm{i}\varepsilon_+ + (\mathrm{e}^{2\mathrm{i}\varepsilon_+} - 1)|\alpha_+|^2 + (\mathrm{e}^{2\mathrm{i}\delta} - 1)|\alpha_-|^2\right] \\
&\qquad - \alpha_+^2 \exp\left[2\mathrm{i}\varphi_+ + 2(\mathrm{e}^{\mathrm{i}\varepsilon_+} - 1)|\alpha_+|^2 + 2(\mathrm{e}^{\mathrm{i}\delta} - 1)|\alpha_-|^2\right] \\
&\qquad + \alpha_-^2 \exp\left[2\mathrm{i}\varphi_- + \mathrm{i}\varepsilon_- + (\mathrm{e}^{2\mathrm{i}\varepsilon_-} - 1)|\alpha_-|^2 + (\mathrm{e}^{2\mathrm{i}\delta} - 1)|\alpha_+|^2\right] \\
&\qquad - \alpha_-^2 \exp\left[2\mathrm{i}\varphi_- + 2(\mathrm{e}^{\mathrm{i}\varepsilon_-} - 1)|\alpha_-|^2 + 2(\mathrm{e}^{\mathrm{i}\delta} - 1)|\alpha_+|^2\right] \\
&\qquad + 2\alpha_+\alpha_- \exp\left[\mathrm{i}(\varphi_+ + \varphi_- + \delta) + (\mathrm{e}^{\mathrm{i}(\varepsilon_+ + \delta)} - 1)|\alpha_+|^2 \right. \\
&\qquad\qquad \left. + (\mathrm{e}^{\mathrm{i}(\varepsilon_- - \delta)} - 1)|\alpha_-|^2\right] \qquad (80) \\
&\qquad - 2\alpha_+\alpha_- \exp\left[\mathrm{i}(\varphi_+ + \varphi_-) + (\mathrm{e}^{\mathrm{i}\varepsilon_+} + \mathrm{e}^{\mathrm{i}\delta} - 2)|\alpha_+|^2 \right. \\
&\qquad\qquad \left. + (\mathrm{e}^{\mathrm{i}\varepsilon_-} + \mathrm{e}^{\mathrm{i}\delta} - 2)|\alpha_-|^2\right]\Big\} \\
&\quad + \Big\{|\alpha_+|^2 - |\alpha_+|^2 \exp\left[2(\cos\varepsilon_+ - 1)|\alpha_+|^2 + 2(\cos\delta - 1)|\alpha_-|^2\right] \\
&\qquad + |\alpha_-|^2 - |\alpha_-|^2 \exp\left[2(\cos\varepsilon_- - 1)|\alpha_-|^2 + 2(\cos\delta - 1)|\alpha_+|^2\right] \\
&\quad + 2\,\mathrm{Re}\Big[\alpha_+^*\alpha_- \exp\left[-\mathrm{i}(\varphi_+ - \varphi_-) + (\mathrm{e}^{-\mathrm{i}(\varepsilon_+ - \delta)} - 1)|\alpha_+|^2 \right. \\
&\qquad\qquad \left. + (\mathrm{e}^{-\mathrm{i}(\varepsilon_- - \delta)} - 1)|\alpha_-|^2\right] \\
&\qquad - \alpha_+^*\alpha_- \exp\left[-\mathrm{i}(\varphi_+ - \varphi_-) + (\mathrm{e}^{-\mathrm{i}\varepsilon_+} + \mathrm{e}^{\mathrm{i}\delta} - 2)|\alpha_+|^2 \right. \\
&\qquad\qquad \left. + (\mathrm{e}^{\mathrm{i}\varepsilon_-} + \mathrm{e}^{-\mathrm{i}\delta} - 2)|\alpha_-|^2\right]\Big]\Big\}
\end{aligned}$$

and for the other component we have

$$\left\langle :[\Delta\hat{P}_x(z)]^2: \right\rangle = -\mathrm{Re}\{\cdots\} + \{\cdots\} \qquad (81)$$

with the contents of the braces the same as in (80).

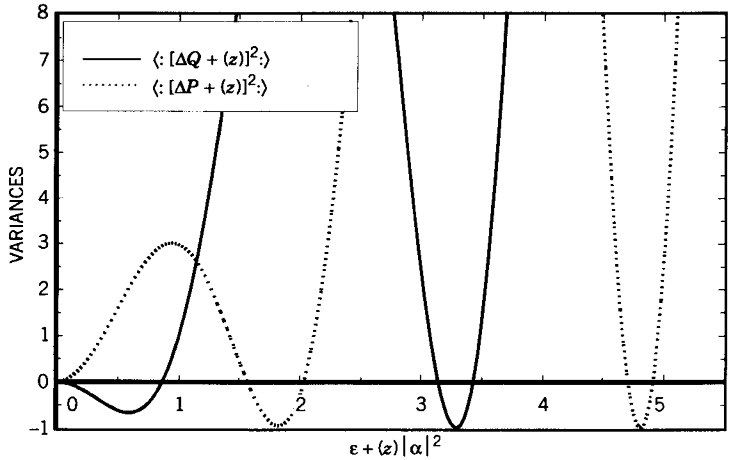


**Figure 2.** Plots of the normally ordered field variances $\langle:[\Delta\hat{Q}_+(z)]^2:\rangle$ and $\langle:[\Delta\hat{P}_+(z)]^2:\rangle$ against the scaled intensity of light $\varepsilon_+(z)|\alpha|^2$, assuming that $\varepsilon_+(z) = 1 \times 10^{-6}$, $\theta = 0$, $\varphi_+ + \varphi_0 = 0$, and $\eta = \pi/4$.

Expressions (80) and (81) are exact. They are, however, very complicated and only numerical analysis allows us to give a definite answer to whether the field is squeezed or not. They can be considerably simplified under certain assumptions concerning the polarization of the field and the character of the medium.

For optically inactive molecules detailed analysis of (80) and (81) has been carried out [74, 75], showing that the components $\hat{Q}_x(z)$ and $\hat{P}_x(z)$ can also become squeezed after passing through the nonlinear medium. The maximum values of squeezing for these components are the same as for $\hat{Q}_+$ and $\hat{P}_+$, although the minima can appear for different values of the field intensity. It is also interesting that for linear polarization of the incoming field perpendicular to the measured polarization ($\theta = \pi/2$), the outgoing field, which is completely quantum in nature, can also show squeezing.

For optically active molecules, expressions (80) and (81) in their extended form must be used. However, as a rule the tensors describing the nonlinear optical activity have orders of magnitude smaller values than the tensors $\gamma_{ijkl}(\omega)$, and in practice $\varepsilon_+ \approx \varepsilon_-$. Thus, in most cases one can neglect contributions from the nonlinear optical activity, which would be essential only if the result were a function of the difference $\varepsilon_+ - \varepsilon_-$.

The mechanism of producing squeezed states described in this chapter is universal in the sense that it takes place for any molecules or atoms including those with spherical symmetry.

## VI. CONCLUSIONS

The subject of this chapter was the problem of producing quantum fields that have no classical analogs in the process of propagation of strong light in a nonlinear Kerr-like medium. The possibility of appearance of two nonclassical effects such as sub-Poissonian photon statistics and squeezing was discussed in detail. We showed that both effects can be produced in corresponding components of the field. Thus, the process of propagation of light in isotropic media can be a source of nonclassical fields. Despite the small values of $\varepsilon(z) - \delta(z)$ for real physical situations, the value of the $q$ parameter measuring the sub-Poissonian character of the photon number distribution can be reduced to $-0.63$ for strong fields which is 63% of the limit allowed by quantum mechanics.

The process discussed turns out to be even more effective in producing squeezed states of the field. In this way one can get more than 97% of squeezing. The effect of squeezing also occurs for circularly polarized light, for which there is no change in photon statistics. This means that the squeezed states can exist with Poissonian photon statistics, which is characteristic for coherent states of the field. Our considerations show explicitly the difference between sub-Poissonian photon statistics and squeezing. The form of solutions (39) for the field operators, which is as a matter of fact nonlinear phase modulation, lead to squeezing of the field states, while the number of photons $\hat{a}_+^+\hat{a}_+(\hat{a}_-^+\hat{a}_-)$, which does not depend on the phase, does not change. To get sub-Poissonian photon statistics, a nonlinear change in the number of photons is needed. It can be achieved for the $x$ and $y$ components of the field for which the number of photons $\hat{a}_x^+\hat{a}_x(\hat{a}_y^+\hat{a}_y)$ does change due to the interaction.

We would like to emphasize the fact that our solutions are exact analytical solutions, which is rather exceptional for this type of problems, and this fact is worthy of attention on its own right. We have referred to the process of squeezing that occurs during the propagation of light in a nonlinear medium as self-squeezing [74, 75]. This is the field itself that causes squeezing of its own quantum fluctuations. The effect of self-squeezing accompanies to a certain degree all other nonlinear processes. In recent years experimental and theoretical studies of nonlinear optical activity have been developed [76, 162, 215, 218].

Squeezing is an effect that depends on the phase of the field, and it is interesting to study the quantum phase properties of the field. Quite recently, since Pegg and Barnett [219] introduced the Hermitian phase formalism, considerable progress has been achieved in studies of phase properties of optical fields. For fields propagating in a Kerr medium such results have been reported by Gantsog and Tanaś [220, 221]. It turned out also that the problem of propagation has exact analytical solutions even when the linear dissipation is included [222]; thus, the quantum phase properties of light propagating in a Kerr medium with dissipation have also been studied [223, 224]. The quantum phase properties of optical fields, however, are beyond the scope of this work, and require separate treatment.

## APPENDIX A

Restricting our considerations to weak spatial dispersion and omitting in (6) summation over $k$, the linear $H_{\mathrm{I}}^{(2)}/N$ and nonlinear $H_{\mathrm{I}}^{(4)}/N$ interaction Hamiltonians of the microsystem (atom, molecule) with the electromagnetic field can be written as (in SI units)

$$H_{\mathrm{I}}^{(2)}/N = -\left[H(-\omega;\omega) + H(\omega;-\omega)\right] \tag{A.1}$$

$$\begin{aligned} H_{\mathrm{I}}^{(4)}/N = -[&H(-\omega;-\omega,\omega,\omega) + H(-\omega;\omega,-\omega,\omega) \\ &+H(-\omega;\omega,\omega,-\omega) + H(\omega;\omega,-\omega,-\omega) \\ &+H(\omega;-\omega,\omega,-\omega) + H(\omega;-\omega,-\omega,\omega)] \end{aligned} \tag{A.2}$$

where

$$\begin{aligned} H(-\omega;\omega) = \tfrac{1}{2}\Big\{&\alpha_{ij}(-\omega;\omega)E_i^-E_j^+ \\ &+\tfrac{1}{3}\left[\eta_{i(jk)}(-\omega;\omega)E_i^-\nabla_k E_j^+ + \eta_{(ik)j}(-\omega;\omega)(\nabla_k E_i^-)E_j^+\right] \\ &+\rho_{ij}(-\omega;\omega)E_i^-B_j^+ + \lambda_{ij}(-\omega;\omega)B_i^-E_j^+\Big\} + \text{h.c.} \end{aligned} \tag{A.3}$$

$$\begin{aligned} H(-\omega;-\omega,\omega,\omega) = \tfrac{1}{24}\Big\{&\gamma_{ijkl}(-\omega;-\omega,\omega,\omega)E_i^-E_j^-E_k^+E_l^+ \\ &+\tfrac{1}{3}\big[\eta_{ijk(lm)}(-\omega;-\omega,\omega,\omega)E_i^-E_j^-E_k^+\nabla_m E_l^+ \\ &+\eta_{ij(km)l}(-\omega;-\omega,\omega,\omega)E_i^-E_j^-(\nabla_m E_k^+)E_l^+ \\ &+\eta_{i(jm)kl}(-\omega;-\omega,\omega,\omega)E_i^-(\nabla_m E_j^-)E_k^+E_l^+ \\ &+\eta_{(im)jkl}(-\omega;-\omega,\omega,\omega)(\nabla_m E_i^-)E_j^-E_k^+E_l^+\big] \\ &+\rho_{ijkl}(-\omega;-\omega,\omega,\omega)E_i^-E_j^-E_k^+B_l^+ \\ &+\kappa_{ijkl}(-\omega;-\omega,\omega,\omega)E_i^-E_j^-B_k^+E_l^+ \\ &+\sigma_{ijkl}(-\omega;-\omega,\omega,\omega)E_i^-B_j^-E_k^+E_l^+ \\ &+\lambda_{ijkl}(-\omega;-\omega,\omega,\omega)B_i^-E_j^-E_k^+E_l^+\Big\} + \text{h.c.} \end{aligned} \tag{A.4}$$

with

$$\mathbf{E}^{\pm} = \mathbf{E}^{\pm}(\mathbf{r},t) \qquad \mathbf{B}^{\pm} = \mathbf{B}^{\pm}(\mathbf{r},t) \tag{A.5}$$

and

$$\mathbf{E}^{\pm}(\mathbf{r},t) = \mathbf{E}^{\pm}(k)\exp[\pm \mathrm{i}(\mathbf{k}\cdot\mathbf{r} - \omega t)] \tag{A.6}$$

The last relation is the result of restricting the summations over $k$ in (6) to one term with definite $k$; that is, we consider a one-mode field.

A characteristic feature of the linear and nonlinear response of the medium to a force oscillating with the frequency $\omega$ is its dependence on $\omega$, which is referred to as time dispersion. Accordingly, in the case of an optical force the Hamiltonians (3) and (4) should depend on the time dispersion. This dependence enters the Hamiltonians via the coupling constants, i.e., the linear and nonlinear and electric and magnetic polarizabilities of the molecules. The linear and nonlinear and electric and magnetic polarizabilities for an individual molecule obtained according to quantum mechanical formulas can be found in Refs. 206, 207, and 225 and are given by the formulas

$$
{}^{(a)}_{A}\chi^{(b)}_{B}(-\omega;\omega) = \frac{\rho_{\Psi\Psi}}{\hbar}\sum_{\Phi f\neq\Psi p}\left\{\frac{\langle\Psi p|\mathbf{M}^{(a)}_{A}|\Phi f\rangle\langle\Phi f|\mathbf{M}^{(b)}_{B}|\Psi p\rangle}{\omega+\omega_{\Phi\Psi}} + \frac{\langle\Psi p|\mathbf{M}^{(b)}_{B}|\Phi f\rangle\langle\Phi f|\mathbf{M}^{(a)}_{A}|\Psi p\rangle}{-\omega+\omega_{\Phi\Psi}}\right\} \tag{A.7}
$$

$$
{}^{(a)}_{A}\chi^{(b,c,d)}_{BCD}(\omega) = \frac{S\{[\mathbf{M}^{(a)}_{A},\mathbf{M}^{(b)}_{B}],[\mathbf{M}^{(c)}_{C},\mathbf{M}^{(d)}_{D}]\}\rho_{\Psi\Psi}}{3!\hbar} \times \sum_{\Phi f,\Lambda l,\Upsilon u\neq\Psi p}\langle\Psi p|\mathbf{F}_{\Phi f\Lambda l\Upsilon u}|\Psi p\rangle
$$

$$
\begin{aligned}
\mathbf{F}_{\Phi f\Lambda l\Upsilon u} = {} & \frac{\mathbf{M}^{(a)}_{A}|\Phi f\rangle\langle\Phi f|\mathbf{M}^{(b)}_{B}|\Lambda l\rangle\langle\Lambda l|\mathbf{M}^{(c)}_{C}|\Upsilon u\rangle\langle\Upsilon u|\mathbf{M}^{(d)}_{D}}{(\omega+\omega_{\Phi\Psi})(2\omega+\omega_{\Lambda\Psi})(\omega+\omega_{\Upsilon\Psi})} \\
& + \frac{\mathbf{M}^{(a)}_{A}|\Phi f\rangle\langle\Phi f|\mathbf{M}^{(c)}_{C}|\Lambda l\rangle\langle\Lambda l|\mathbf{M}^{(b)}_{B}|\Upsilon u\rangle\langle\Upsilon u|\mathbf{M}^{(d)}_{D}}{(\omega+\omega_{\Phi\Psi})\omega_{\Lambda\Psi}(\omega+\omega_{\Upsilon\Psi})} \\
& + \frac{\mathbf{M}^{(b)}_{B}|\Phi f\rangle\langle\Phi f|\mathbf{M}^{(c)}_{C}|\Lambda l\rangle\langle\Lambda l|\mathbf{M}^{(d)}_{D}|\Upsilon u\rangle\langle\Upsilon u|\mathbf{M}^{(a)}_{A}}{(\omega+\omega_{\Phi\Psi})\omega_{\Lambda\Psi}(-\omega+\omega_{\Upsilon\Phi})} \\
& + \frac{\mathbf{M}^{(d)}_{D}|\Phi f\rangle\langle\Phi f|\mathbf{M}^{(a)}_{A}|\Lambda l\rangle\langle\Lambda l|\mathbf{M}^{(b)}_{B}|\Upsilon u\rangle\langle\Upsilon u|\mathbf{M}^{(c)}_{C}}{(-\omega+\omega_{\Phi\Psi})\omega_{\Lambda\Psi}(\omega+\omega_{\Upsilon\Psi})}
\end{aligned} \tag{A.8}
$$

$$
\begin{aligned}
& + \frac{\mathbf{M}^{(c)}_{C}|\Phi f\rangle\langle\Phi f|\mathbf{M}^{(b)}_{B}|\Lambda l\rangle\langle\Lambda l|\mathbf{M}^{(d)}_{D}|\Upsilon u\rangle\langle\Upsilon u|\mathbf{M}^{(a)}_{A}}{(-\omega+\omega_{\Phi\Psi})\omega_{\Lambda\Psi}(-\omega+\omega_{\Upsilon\Psi})} \\
& + \frac{\mathbf{M}^{(c)}_{C}|\Phi f\rangle\langle\Phi f|\mathbf{M}^{(d)}_{D}|\Lambda l\rangle\langle\Lambda l|\mathbf{M}^{(a)}_{A}|\Upsilon u\rangle\langle\Upsilon u|\mathbf{M}^{(b)}_{B}}{(-\omega+\omega_{\Phi\Psi})(-2\omega+\omega_{\Lambda\Psi})(-\omega+\omega_{\Upsilon\Psi})}
\end{aligned}
$$

where $S\{[M_A^{(a)}, \mathbf{M}_B^{(b)}], [\mathbf{M}_C^{(c)}, \mathbf{M}_D^{(d)}]\}$ is the operator denoting summation over the permutations of the elements contained in the square brackets, and we have for the linear polarizabilities

$$
\begin{aligned}
\alpha_{ij}(-\omega;\omega) &= {}^{(1)}_{e}\chi^{(1)}_{e\,ij}(-\omega;\omega)\\
\rho_{ij}(-\omega;\omega) &= {}^{(1)}_{e}\chi^{(1)}_{m\,ij}(-\omega;\omega)\\
\lambda_{ij}(-\omega;\omega) &= {}^{(1)}_{m}\chi^{(1)}_{e\,ij}(-\omega;\omega)\\
\eta_{i(jk)}(-\omega;\omega) &= {}^{(1)}_{e}\chi^{(2)}_{e\,i(jk)}(-\omega;\omega)\\
\eta_{(ik)j}(-\omega;\omega) &= {}^{(2)}_{e}\chi^{(1)}_{e\,(ik)j}(-\omega;\omega)
\end{aligned}
\tag{A.9}
$$

and for the nonlinear polarizabilities

$$
\begin{aligned}
\gamma_{ijkl}(-\omega;-\omega,\omega,\omega) &= {}^{(1)}_{e}\chi^{(1,1,1)}_{eee\,ijkl}(-\omega;-\omega,\omega,\omega)\\
\rho_{ijkl}(-\omega;-\omega,\omega,\omega) &= {}^{(1)}_{e}\chi^{(1,1,1)}_{eem\,ijkl}(-\omega;-\omega,\omega,\omega)\\
\kappa_{ijkl}(-\omega;-\omega,\omega,\omega) &= {}^{(1)}_{e}\chi^{(1,1,1)}_{eme\,ijkl}(-\omega;-\omega,\omega,\omega)\\
\sigma_{ijkl}(-\omega;-\omega,\omega,\omega) &= {}^{(1)}_{e}\chi^{(1,1,1)}_{mee\,ijkl}(-\omega;-\omega,\omega,\omega)\\
\lambda_{ijkl}(-\omega;-\omega,\omega,\omega) &= {}^{(1)}_{m}\chi^{(1,1,1)}_{eee\,ijkl}(-\omega;-\omega,\omega,\omega)\\
\eta_{ijk(lm)}(-\omega;-\omega,\omega,\omega) &= {}^{(1)}_{e}\chi^{(1,1,2)}_{eee\,ijk(lm)}(-\omega;-\omega,\omega,\omega)\\
\eta_{ij(km)l}(-\omega;-\omega,\omega,\omega) &= {}^{(1)}_{e}\chi^{(1,2,1)}_{eee\,ij(km)l}(-\omega;-\omega,\omega,\omega)\\
\eta_{i(jm)kl}(-\omega;-\omega,\omega,\omega) &= {}^{(1)}_{e}\chi^{(2,1,1)}_{eee\,i(jm)kl}(-\omega;-\omega,\omega,\omega)\\
\eta_{(im)jkl}(-\omega;-\omega,\omega,\omega) &= {}^{(2)}_{e}\chi^{(1,1,1)}_{eee\,(im)jkl}(-\omega;-\omega,\omega,\omega)
\end{aligned}
\tag{A.10}
$$

Expressions (A.7) and (A.8) are the quantum mechanical formulas for the linear and nonlinear polarizabilities of the microsystem being in one of the stationary states of the $f$-fold degenerate energy level with the energy $\hbar\omega_\Psi$. This state, $|\Psi f\rangle$, is defined by the quantum numbers $\Psi$ labeling the energy levels of the microsystem and the quantum numbers $f$ labeling the states belonging to the level $\Psi$. Moreover, $\rho_{\Psi\Psi}$ denotes the expectation value of the unperturbed density matrix in the state $|\Psi\rangle$ (the probability that the microsystem is in the stationary state $|\Psi\rangle$), $\omega_{\phi\Psi} = \omega_\phi - \omega_\Psi$ is the transition frequency between the levels $\phi$ and $\Psi$, and $M_A^{(a)}$ is the operator of the multipolar electric ($A = e$) or magnetic ($A = m$) moment of the order $a$.

From definitions (A.7) and (A.8) one can easily show that

$$\begin{aligned} {}^{(a)}_{A}\chi^{(b)}_{B}(-\omega;\omega)^* &= {}^{(a)}_{A}\chi^{(b)}_{B}(\omega;-\omega) \\ {}^{(a)}_{A}\chi^{(b,c,d)}_{BCD}(-\omega;-\omega,\omega,\omega)^* &= {}^{(a)}_{A}\chi^{(b,c,d)}_{BCD}(\omega;\omega,-\omega,-\omega) \end{aligned} \tag{A.11}$$

It is also easy to check that the polarizabilities are invariant with respect to the following permutations:

$$\begin{aligned} {}^{(a)}_{A}\chi^{(b,c,d)}_{BCD}(-\omega;-\omega,\omega,\omega) &= {}^{(a)}_{A}\chi^{(c,b,d)}_{CBD}(-\omega;-\omega,\omega,\omega) \\ &= {}^{(a)}_{A}\chi^{(d,c,b)}_{DCB}(-\omega;\omega,\omega,-\omega) \\ &= {}^{(a)}_{A}\chi^{(b,d,c)}_{BDC}(-\omega;-\omega,\omega,\omega) \\ &= {}^{(b)}_{B}\chi^{(a,c,d)}_{ACD}(-\omega;-\omega,\omega,\omega) \\ &= {}^{(b)}_{B}\chi^{(a,d,c)}_{ADC}(-\omega;-\omega,\omega,\omega) \end{aligned} \tag{A.12}$$

which allows us to replace the polarizabilities ${}^{(a)}_{A}\chi^{(b,c,d)}_{BCD}(-\omega;\omega,-\omega,\omega)$ occurring in $H(-\omega,\omega,-\omega,\omega)$, and the polarizabilities ${}^{(a)}_{A}\chi^{(b,c,d)}_{BCD}(-\omega;\omega,\omega-\omega)$ occurring in $H(-\omega;\omega,\omega,-\omega)$ with ${}^{(a)}_{A}\chi^{(c,b,d)}_{CBD}(-\omega;-\omega,\omega,\omega)$ and ${}^{(a)}_{A}\chi^{(d,c,b)}_{DCB}(-\omega;-\omega,\omega,\omega)$, respectively. This means the same frequency dependence, that is, $(-\omega;-\omega,\omega,\omega)$, as in $H(-\omega;-\omega,\omega,\omega)$. Similarly, the polarizabilities occurring in $H(\omega;-\omega,\omega,-\omega)$ and $H(\omega;-\omega,-\omega,\omega)$ can be replaced with those occurring in $H(\omega;\omega,-\omega,-\omega)$.

The symmetry properties (A.12) allow for the reduction of the number of the coupling constants occurring in (A.2) from six to two types, and (A.11) allows us to replace the coupling constants of the type ${}^{(a)}_{A}\chi^{(b,c,d)}_{BCD}(\omega;\omega,-\omega,-\omega)$ with ${}^{(a)}_{A}\chi^{(b,c,d)}_{BCD}(-\omega;-\omega,\omega,\omega)$, which reduces the problem to one type of the coupling only.

Generally, the linear and nonlinear polarizabilities are complex quantities and can be written in the form

$$ {}^{(a)}_{A}\chi^{(b)}_{B}(-\omega;\omega) = {}^{(a)}_{A}\chi^{(b)\prime}_{B}(-\omega;\omega) + \mathrm{i}\,{}^{(a)}_{A}\chi^{(b)\prime\prime}(-\omega;\omega) \quad \text{(A.13)} $$

$$ {}^{(a)}_{A}\chi^{(b,c,d)}_{BCD}(-\omega;-\omega,\omega,\omega) = {}^{(a)}_{A}\chi^{(b,c,d)\prime}_{BCD}(-\omega;-\omega,\omega,\omega) + \mathrm{i}\,{}^{(a)}_{A}\chi^{(b,c,d)\prime\prime}_{BCD}(-\omega;-\omega,\omega,\omega) \quad \text{(A.14)} $$

where

$$ {}^{(a)}_{A}\chi^{(b)\prime}_{B}(-\omega;\omega) = \mathrm{Re}\,{}^{(a)}_{A}\chi^{(b)}_{B}(-\omega;\omega) \quad \text{(A.15)} $$

$$ {}^{(a)}_{A}\chi^{(b)\prime\prime}_{B}(-\omega;\omega) = \mathrm{Im}\,{}^{(a)}_{A}\chi^{(b)}_{B}(-\omega;\omega) \quad \text{(A.16)} $$

and similarly for ${}^{(a)}_{(A}\chi^{(b,c,d)\prime}_{BCD}(-\omega;-\omega,\omega,\omega)$ and ${}^{(a)}_{(A}\chi^{(b,c,d)\prime\prime}_{BCD}(-\omega;-\omega,\omega,\omega)$.

Applying (A.7) and (A.8) we can obtain quantum mechanical expressions for the real and imaginary parts of the linear and nonlinear polarizabilities, which allows us to check the following additional permutation relations [226–228]:

$$ {}^{(a)}_{A}\chi^{(b)\prime}_{B\,i_1\cdots i_a j_1\cdots j_b}(-\omega;\omega) = {}^{(b)}_{B}\chi^{(a)\prime}_{A\,j_1\cdots j_b i_1\cdots i_a}(-\omega;\omega) \quad \text{(A.17)} $$

$$ {}^{(a)}_{A}\chi^{(b)\prime\prime}_{B\,i_1\cdots i_a j_1\cdots j_b}(-\omega;\omega) = -{}^{(b)}_{B}\chi^{(a)\prime\prime}_{A\,j_1\cdots j_b i_1\cdots i_a}(-\omega;\omega) \quad \text{(A.18)} $$

$$ \begin{aligned} &{}^{(a)}_{A}\chi^{(b,c,d)\prime}_{BCD\,i_1\cdots i_a j_1\cdots j_b k_1\cdots k_c l_1\cdots l_d}(-\omega;-\omega,\omega,\omega) \\ &\quad = {}^{(c)}_{C}\chi^{(d,a,b)\prime}_{DAB\,k_1\cdots k_c l_1\cdots l_d i_1\cdots i_a j_1\cdots j_b}(-\omega;-\omega,\omega,\omega) \end{aligned} \quad \text{(A.19)} $$

$$ \begin{aligned} &{}^{(a)}_{A}\chi^{(b,c,d)\prime\prime}_{BCD\,i_1\cdots i_a j_1\cdots j_b k_1\cdots k_c l_1\cdots l_d}(-\omega;-\omega,\omega,\omega) \\ &\quad = -{}^{(c)}_{C}\chi^{(d,a,b)\prime\prime}_{DAB\,k_1\cdots k_c l_1\cdots l_d i_1\cdots i_a j_1\cdots j_b}(-\omega;-\omega,\omega,\omega) \end{aligned} \quad \text{(A.20)} $$

Above, the indices $i_1 \cdots i_a$ label the components of the operator $\mathbf{M}^{(a)}_{A}$ of the electric ($A = e$) or magnetic ($A = m$) multipole moment of order $a$, which is a tensor of rank $a$ having the following form in the Cartesian frame $(z, y, z)$:

$$ \mathbf{M}^{(a)}_{A} = \mathrm{e}_{i_1} \cdots \mathrm{e}_{i_a} M^{(a)}_{A\,i_1\cdots i_a} \quad \text{(A.21)} $$

where the indices $i_p$ can take the values $x$, $y$, and $z$ for $p = 1, \ldots, a$, whereas $e_{i_p}$ represents the unit vector along the $i_p$ axis of the frame $(z, y, z)$. If $A = e$ then $\mathbf{M}_e^{(a)}$ is a polar tensor, which is symmetric with respect to the $a!$ permutations of the indices $i \ldots i_a$; if $A = m$ then $\mathbf{M}_m^{(a)}$ is an axial tensor symmetric with respect to the $(a - 1)!$ permutations of the indices $i_2 \ldots i_a$. The indices with respect to which the tensor is invariant are separated in (A.3) and (A.4) by the parentheses.

Acting with the time reversal operator $R$ on the quantum mechanical expressions defining the real and imaginary parts of the linear as well as nonlinear polarizability, it is easy to check that

$$
\begin{array}{ll}
\mathrm{Re}\, \alpha_{ij}(-\omega;\omega) & \mathrm{Im}\, \rho_{ij}(-\omega;\omega) \\
\mathrm{Im}\, \lambda_{ij}(-\omega;\omega) & \\
\mathrm{Re}\, \eta_{i(jk)}(-\omega;\omega) & \mathrm{Re}\, \eta_{(ij)k}(-\omega;\omega) \\
\mathrm{Re}\, \gamma_{ijkl}(-\omega;-\omega,\omega,\omega) & \\
\mathrm{Re}\, \eta_{ijk(lm)}(-\omega;-\omega,\omega,\omega) & \mathrm{Re}\, \eta_{ij(km)l}(-\omega;-\omega,\omega,\omega) \\
\mathrm{Re}\, \eta_{i(jm)kl}(-\omega;-\omega,\omega,\omega) & \mathrm{Re}\, \eta_{(im)jkl}(-\omega;-\omega,\omega,\omega) \\
\mathrm{Im}\, \rho_{ijkl}(-\omega;-\omega,\omega,\omega) & \mathrm{Im}\, \kappa_{ijkl}(-\omega;-\omega,\omega,\omega) \\
\mathrm{Im}\, \sigma_{ijkl}(-\omega;-\omega,\omega,\omega) & \mathrm{Im}\, \lambda_{ijkl}(-\omega;-\omega,\omega,\omega)
\end{array}
\qquad (\mathrm{A.22})
$$

are invariant with respect to the time reversal [229], and the remaining expressions change their sign under time reversal.

One of the postulates of quantum mechanics is the invariance of the Schrödinger equation with respect to the time-reversal operation $R$ [229]. This postulate implies that if the function $\Psi$ is a solution of the Schrödinger equation, then the function $\Psi' = R\Psi$ is also a solution of this equation with the same energy $E_\Psi = E_{R\Psi}$. A physical quantity $O$ is invariant with respect to the time reversal if its value is the same in $\Psi$ and $R\Psi$, and it is antisymmetrical if the two values differ in sign. Of course, the relation $R\Psi = \Psi$ rules out the quantities antisymmetrical with respect to the time reversal.

Assuming that the microsystems are freely oriented, we can average (A.1) and (A.2) over all orientations of the microsystems. Performing such averaging according to (9)–(12) and applying (7) as well as the permutation relations (A.11)–(A.12), the linear $H_I^{(2)}/N$ and nonlinear $H_I^{(4)}/N$ interaction Hamiltonians describing the interaction of the microsystem with the electromagnetic field take, in the circular polarization basis (8), the following forms:

$$
\begin{aligned}
\left\langle H_{\mathrm{I}}^{(2)}/N \right\rangle_{\Omega} = &-\tfrac{1}{2}\underline{\chi}_{R}^{L}\left[E_{-}^{+}E_{+}^{+} + E_{-}^{-}E_{-}^{+} + E_{+}^{+}E_{-}^{+} + E_{-}^{+}E_{-}^{-}\right] \\
&-\frac{\mathrm{i}}{2}\underline{\chi}_{A}^{L}\left[E_{+}^{-}E_{+}^{+} - E_{-}^{-}E_{-}^{+} + E_{+}^{+}E_{+}^{-} - E_{-}^{+}E_{-}^{-}\right]
\end{aligned}
\tag{A.23}
$$

$$
\begin{aligned}
\left\langle H_{\mathrm{I}}^{(4)}/N \right\rangle_{\Omega} = &-\tfrac{1}{12}\underline{\chi}_{R}^{NL}\Big[(E_{+}^{-})^{2}(E_{+}^{+})^{2} + (E_{-}^{-})^{2}(E_{-}^{+})^{2} \\
&+(E_{+}^{+}E_{-}^{+})^{2} + (E_{-}^{-}E_{-}^{+})^{2} \\
&+\text{terms with reversed superscripts}\Big] \\
&-\tfrac{1}{12}\underline{\tilde{\chi}}_{R}^{NL}\Big[E_{+}^{-}(E_{+}^{+})^{2}E_{+}^{-} + E_{-}^{-}(E_{-}^{+})^{2}E_{-}^{-}\Big] \\
&-\tfrac{1}{12}\underline{\tilde{\chi}}_{R}^{NL*}\Big[E_{+}^{+}(E_{+}^{-})^{2}E_{+}^{+} + E_{-}^{+}(E_{-}^{-})^{2}E_{-}^{+}\Big] \\
&-\tfrac{1}{6}\underline{\kappa}_{R}^{NL}\Big[E_{+}^{-}E_{-}^{-}E_{+}^{+}E_{-}^{+} + \tfrac{1}{4}(E_{+}^{-}E_{+}^{+}E_{-}^{+}E_{-}^{+} + E_{-}^{-}E_{-}^{+}E_{+}^{-}E_{+}^{+} \\
&+E_{+}^{-}E_{-}^{+}E_{-}^{-}E_{+}^{+} + E_{-}^{-}E_{+}^{+}E_{+}^{-}E_{-}^{+}) \\
&+\text{terms with reversed superscripts}\Big] \\
&-\tfrac{1}{24}\underline{\tilde{\kappa}}_{R}^{NL}\left[E_{+}^{-}E_{+}^{+}E_{-}^{+}E_{-}^{-} + E_{-}^{-}E_{-}^{+}E_{+}^{+}E_{+}^{-}\right. \\
&\left.+E_{+}^{-}E_{-}^{+}E_{+}^{+}E_{-}^{-} + E_{-}^{-}E_{+}^{+}E_{-}^{+}E_{+}^{-}\right] \\
&-\tfrac{1}{24}\underline{\tilde{\kappa}}_{R}^{NL*}\left[E_{+}^{+}E_{+}^{-}E_{-}^{-}E_{-}^{+} + E_{-}^{+}E_{-}^{-}E_{+}^{-}E_{+}^{+}\right. \\
&\left.+E_{+}^{+}E_{-}^{-}E_{+}^{-}E_{-}^{+} + E_{-}^{+}E_{+}^{-}E_{-}^{-}E_{+}^{+}\right] \\
&-\tfrac{1}{12}\underline{\chi}_{R}^{NL}\Big[(E_{+}^{-})^{2}(E_{+}^{+})^{2} - (E_{-}^{-})^{2}(E_{-}^{+})^{2} \\
&+(E_{-}^{-}E_{+}^{+})^{2} - (E_{-}^{-}E_{-}^{+})^{2} \\
&+E_{+}^{-}(E_{+}^{+})^{2}E_{+}^{-} - E_{-}^{-}(E_{-}^{+})^{2}E_{-}^{-} \\
&+\text{terms with reversed superscripts}\Big]
\end{aligned}
\tag{A.24}
$$

where

$$\underline{\chi}_R^L = \tfrac{1}{3}\mathrm{Re}\,\alpha_{\alpha\alpha}(-\omega;\omega) \tag{A.25}$$

$$\bar{\chi}_A^L = -\frac{\mathrm{i}k_z}{3\omega}\mathrm{Im}\,\rho_{\alpha\alpha}(-\omega;\omega) \tag{A.26}$$

$$\underline{\chi}_R^{NL} = \tfrac{1}{15}\mathrm{Re}\big[-\gamma_{\alpha\alpha\beta\beta}(-\omega;-\omega,\omega,\omega) + 3\gamma_{\alpha\beta\alpha\beta}(-\omega;-\omega,\omega,\omega)\big] \tag{A.27}$$

$$\underline{\tilde{\chi}}_R^{NL} = \tfrac{1}{15}\big[-\gamma_{\alpha\alpha\beta\beta}(-\omega;-\omega,\omega,\omega) + 3\gamma_{\alpha\beta\alpha\beta}(-\omega;-\omega,\omega,\omega)\big] \tag{A.28}$$

$$\underline{\kappa}_R^{NL} = \tfrac{1}{15}\mathrm{Re}\big[3\gamma_{\alpha\alpha\beta\beta}(-\omega;-\omega,\omega,\omega) + \gamma_{\alpha\beta\alpha\beta}(-\omega;-\omega,\omega,\omega)\big] \tag{A.29}$$

$$\underline{\tilde{\kappa}}_R^{NL} = \tfrac{1}{15}\big[3\gamma_{\alpha\alpha\beta\beta}(-\omega;-\omega,\omega,\omega) + \gamma_{\alpha\beta\alpha\beta}(-\omega;-\omega,\omega,\omega)\big] \tag{A.30}$$

$$\underline{\chi}_A^{NL} = -\frac{\mathrm{i}4k_z}{15}\bigg\{\frac{1}{\omega}\mathrm{Im}\big[\sigma_{\alpha\alpha\beta\beta}(-\omega;-\omega,\omega,\omega) - 3\sigma_{\alpha\beta\alpha\beta}(-\omega;-\omega,\omega,\omega)\big] - \tfrac{1}{3}\mathrm{Re}\,\eta_{\alpha(\beta\gamma)\beta\delta}(-\omega;-\omega,\omega,\omega)\varepsilon_{\alpha\gamma\delta}\bigg\} \tag{A.31}$$

and

$$E = E(k) \tag{A.32}$$

The total interaction Hamiltonian describing the interaction of the optical field with an ensemble of $N$ microsystems confined in the unit volume is, according to (1) and (2), equal to

$$H_{\mathrm{I}} = H_{\mathrm{I}}^{(2)} + H_{\mathrm{I}}^{(4)} + \cdots = \sum_{n=1}^{\infty} H_{\mathrm{I}}^{(2n)} \tag{A.33}$$

where

$$H_{\mathrm{I}}^{(2n)} = \sum_{p=1}^{N} \left\langle H_{\mathrm{I}}^{(2n)}/N \right\rangle_{\Omega} \tag{A.34}$$

and the summation runs over all microsystems.

Let us assume that the microsystems are identical. In this case the summation over $p$ simplifies considerably, and it becomes trivial if the ground state of the microsystem is nondegenerate. For the nondegenerate case, we have

$$R\Psi = \Psi \tag{A.35}$$

which implies vanishing of the polarizabilities antisymmetrical with respect to the time reversal, and we have

$$\underline{\tilde{\chi}}_R^{NL} = \underline{\tilde{\chi}}_R^{NL*} = \underline{\chi}_R^{NL} \tag{A.36}$$

$$\underline{\tilde{\kappa}}_R^{NL} = \underline{\tilde{\kappa}}_R^{NL*} = \underline{\kappa}_R^{NL} \tag{A.37}$$

where the summation over $p$ reduces to the multiplication of the parameters (A.25)–(A.31) over $N$, which leads to (14)–(20) with the molecular constants:

$$\begin{aligned} \operatorname{Re} \alpha_{\alpha\alpha} &= \operatorname{Re} \alpha_{\alpha\alpha}(-\omega;\omega) \\ \operatorname{Im} \rho_{\alpha\alpha} &= \operatorname{Im} \rho_{\alpha\alpha}(-\omega;\omega) \\ \operatorname{Re} \gamma_{\alpha\beta\gamma\delta} &= \operatorname{Re} \gamma_{\alpha\beta\gamma\delta}(-\omega;-\omega,\omega,\omega) \\ \operatorname{Im} \sigma_{\alpha\beta\gamma\delta} &= \operatorname{Im} \sigma_{\alpha\beta\gamma\delta}(-\omega;-\omega,\omega,\omega) \\ \operatorname{Re} \eta_{\alpha(\beta\gamma)\beta\delta} &= \operatorname{Re} \eta_{\alpha(\beta\gamma)\beta\delta}(-\omega;-\omega,\omega,\omega) \end{aligned} \tag{A.38}$$

If the ground state $\Psi$ of the microsystem is $2u$-fold degenerate and the states are labeled by the indices $f = 1, 2, \ldots, u$ associated with the particular wave functions in such a way that

$$R\Psi f = \Psi(2f) \tag{A.39}$$

then, according to quantum mechanics, the probability of finding a microsystem in any state of the $s$-fold degenerate level $\Psi$ is the same for all states and equal to $\rho_{\Psi\Psi}$. This means that for a large number $N$ of identical microsystems in the unit volume, in each state of the $s$-fold degenerate level $\Psi$ will be the same number of microsystems, equal to $(N/s)\rho_{\Psi\Psi}$. In our case this number will be equal to $N/2u$ because we have already incorporated $\rho_{\Psi\Psi}$ into (A.7 and (A.8) defining the linear and nonlinear polarizabilities. As a result, the summation over $p$ can be replaced by the summation over the states of the $2u$-fold degenerate level $\Psi$

$$\sum_{p=1}^{N} \gamma_{\alpha\beta\gamma\delta}(-\omega;-\omega,\omega,\omega) = \frac{N}{2u}\sum_{f=1}^{u}\left[\gamma_{\alpha\beta\gamma\delta}(-\omega;-\omega,\omega,\omega)_{\Psi f} + \gamma_{\alpha\beta\gamma\delta}(-\omega;-\omega,\omega,\omega)_{\Psi(2f)}\right] \tag{A.40}$$

where the first term is the nonlinear polarizability in the state $\Psi f$, and the second term is in the state $\Psi(2f)$; the polarizabilities are complex quantities. Keeping in mind the fact that the polarizabilities symmetrical with respect to the time reversal are the same in the states $\Psi f$ and $\Psi(2f) = R\Psi f$, while antisymmetrical differ in sign, in view of (A.40), (A.10), (A.14), and (A.22), we have

$$\sum_{p=1}^{N} \gamma_{\alpha\beta\gamma\delta}(-\omega;-\omega,\omega,\omega) = N \operatorname{Re} \gamma_{\alpha\beta\gamma\delta} \tag{A.41}$$

where the mean polarizability is given by

$$\operatorname{Re} \gamma_{\alpha\beta\gamma\delta} = \frac{1}{u} \sum_{f=1}^{u} \operatorname{Re} \gamma_{\alpha\beta\gamma\delta}(-\omega;-\omega,\omega,\omega)_{\Psi f} \tag{A.42}$$

Similarly, we get

$$\sum_{p=1}^{N} \sigma_{\alpha\beta\gamma\delta}(-\omega;-\omega,\omega,\omega) = \mathrm{i}\, N \operatorname{Im} \sigma_{\alpha\beta\gamma\delta} \tag{A.43}$$

where

$$\operatorname{Im} \sigma_{\alpha\beta\gamma\delta} = \frac{1}{u} \sum_{f=1}^{u} \operatorname{Im} \sigma_{\alpha\beta\gamma\delta}(-\omega;-\omega,\omega,\omega)_{\Psi f} \tag{A.44}$$

In the case of odd, $(2u + 1)$-fold, degeneracy of the level $\Psi$, we assume additionally that for at least one state, say, $\Psi 0$, the following relation is satisfied:

$$R\Psi 0 = \Psi 0 \tag{A.45}$$

and for the remaining states (A.39) holds. As a result of (A.45) the polarizability antisymmetrical with respect to the time reversal vanishes in the state $\Psi$. In a similar way as before, in place of (A.42) and (A.44), we get

$$\operatorname{Re}\gamma_{\alpha\beta\gamma\delta} = \frac{1}{2u+1}\left[\operatorname{Re}\gamma_{\alpha\beta\gamma\delta}(-\omega;-\omega,\omega,\omega)_{\Psi 0} + 2\sum_{f=1}^{u}\operatorname{Re}\gamma_{\alpha\beta\gamma\delta}(-\omega;-\omega,\omega,\omega)_{\Psi f}\right] \tag{A.46}$$

$$\operatorname{Im}\sigma_{\alpha\beta\gamma\delta} = \frac{1}{2u+1}\left[\operatorname{Im}\sigma_{\alpha\beta\gamma\delta}(-\omega;-\omega,\omega,\omega)_{\Psi 0} + 2\sum_{f=1}^{u}\operatorname{Im}\sigma_{\alpha\beta\gamma\delta}(-\omega;-\omega,\omega,gu)_{\Psi f}\right] \tag{A.47}$$

Using (A.41)–(A.47), we can perform the summation over $p$ in (A.34). For both even and odd degeneracies, $H_{\mathrm{I}}^{(2)}$ and $H_{\mathrm{I}}^{(4)}$ are still described by (14) and (15) with the molecular parameters (16)–(20), which for even degeneracies are given by

$$\begin{aligned}
\operatorname{Re}\alpha_{\alpha\alpha} &= \frac{1}{u}\sum_{f=1}^{u}\operatorname{Re}\alpha_{\alpha\alpha}(-\omega;\omega)_{\Psi f}\\
\operatorname{Im}\rho_{\alpha\alpha} &= \frac{1}{u}\sum_{f=1}^{u}\operatorname{Im}\rho_{\alpha\alpha}(-\omega;\omega\omega)_{\Psi f}\\
\operatorname{Re}\eta_{\alpha(\beta\gamma)\beta\delta} &= \frac{1}{u}\sum_{f=1}^{u}\operatorname{Re}\eta_{\alpha(\beta\gamma)\beta\delta}(-\omega;-\omega,\omega,\omega)_{\Psi f}
\end{aligned} \tag{A.48}$$

whereas $\operatorname{Re}\gamma_{\alpha\alpha\beta\beta}$ and $\operatorname{Re}\gamma_{\alpha\beta\alpha\beta}$ are defined by (A.42), and $\operatorname{Im}\sigma_{\alpha\alpha\beta\beta}$ and $\operatorname{Im}\sigma_{\alpha\beta\alpha\beta}$ are defined by (A.44). For odd degeneracies the parameters are given by

$$\operatorname{Re}\alpha_{\alpha\alpha} = \frac{1}{2u+1}\left[\operatorname{Re}\alpha_{\alpha\alpha}(-\omega;\omega)_{\Psi 0} + 2\sum_{f=1}^{u}\operatorname{Re}\alpha_{\alpha\alpha}(-\omega;\omega)_{\Psi f}\right]$$

$$\text{Im}\,\rho_{\alpha\alpha} = \frac{1}{2u+1}\left[\text{Im}\,\rho_{\alpha\alpha}(-\omega;\omega)_{\Psi 0} + 2\sum_{f=1}^{u}\text{Im}\,\rho_{\alpha\alpha}(-\omega;\omega)_{\Psi f}\right] \tag{A.49}$$

$$\text{Re}\,\eta_{\alpha(\beta\gamma)\beta\delta} = \frac{1}{2u+1}\left[\text{Re}\,\eta_{\alpha(\beta\gamma)\beta\delta}(-\omega;-\omega,\omega,\omega)_{\Psi 0} + 2\sum_{f=1}^{u}\text{Re}\,\eta_{\alpha(\beta\gamma)\beta\delta}(-\omega;-\omega,\omega,\omega)_{\Psi f}\right]$$

while $\text{Re}\,\gamma_{\alpha\alpha\beta\beta}$ and $\text{Re}\,\gamma_{\alpha\beta\alpha\beta}$ are defined by (A.46), and $\text{Im}\,\sigma_{\alpha\alpha\beta\beta}$ and $\text{Im}\,\sigma_{\alpha\beta\alpha\beta}$ are define by (A.47).

Taking into account (A.38), (A.42), (A.44), and (A.46)–(A.49) defining the nonlinear molecular polarizabilities for both the nondegenerate and degenerate electronic states and (A.10) and (A.14)–(A.16), one can check that $\text{Re}\,\gamma_{\alpha\beta\gamma\delta}$, $\text{Im}\,\sigma_{\alpha\beta\gamma\delta}$, and $\text{Re}\,\eta_{\alpha(\beta\gamma)\delta\Phi}$ have the same permutation symmetry as ${}^{(1)}_{e}\chi^{(1,1,1)\prime}_{eee\,\alpha\beta\gamma\delta}(-\omega;-\omega,\omega,\omega)$, ${}^{(1)}_{e}\chi^{(1,1,1)\prime\prime}_{mee\,\alpha\beta\gamma\delta}(-\omega;-\omega,\omega,\omega)$, and ${}^{(1)}_{e}\chi^{(2,1,1)\prime}_{eee\,\alpha(\beta\gamma)\delta\Phi}(-\omega;-\omega,\omega,\omega)$, respectively. This symmetry can be found from (A.12), (A.19), and (A.20).

## APPENDIX B

In Appendix A we showed that the tensor $\text{Re}\,\gamma_{\alpha\beta\gamma\delta}$ is invariant with respect to the permutations of the indices $\alpha$ and $\beta$ as well as $\gamma$ and $\delta$, and also with respect to the permutations of the pairs of indices $\alpha\beta$ and $\gamma\delta$. The tensor $\text{Im}\,\sigma_{\alpha\beta\gamma\delta}$ defining the nonlinear molecular parameter $\chi_A^{NL}$ is invariant under the permutation $\gamma$ and $\delta$, and the tensor $\text{Re}\,\eta_{\alpha(\beta\gamma)\delta\phi}$ is symmetric with respect to the permutations $\delta$ and $\phi$ as well as $\beta$ and $\gamma$ ($\beta$ and $\gamma$ are associated with the electric quadrupole operator).

To find the molecular parameters $\chi_R^L$, $\chi_A^L$, $\chi_R^{NL}$, $\kappa_R^{NL}$, and $\chi_A^{NL}$ for the molecules with a definite molecular symmetry one has to know the explicit form of the polarizability tensors $\text{Re}\,\gamma_{\alpha\beta\gamma\delta}$, $\text{Im}\,\sigma_{\alpha\beta\gamma\delta}$, and $\text{Re}\,\eta_{\alpha(\beta\gamma)\delta\phi}$. Applying the group theory methods [230, 228] the components of the tensors have been found for 102 magnetic point groups, and the results are presented in Tables I–III. The molecular parameters $\chi_R^L$, $\chi_A^L$, $\chi_R^{NL}$, $\kappa_R^{NL}$, and $\chi_A^{NL}$ for all these symmetry groups are collected in Table IV.

TABLE I
Fourth-Rank Polar i-tensor Re $\gamma_{\alpha\beta\gamma\delta}$ for 102 Magnetic Point Groups

| Magnetic Point Group | N | I | Form of the i-tensor Re $\gamma_{\alpha\beta\gamma\delta}$ |
|---|---|---|---|
| $1, \bar{1}, \underline{\bar{1}}$ | 81 | 21 | $a_1 \equiv$ 1111, 2222, 3333,<br>1122 = 2211, 1133 = 3311, 2233 = 3322,<br>1212 = 1221 = 2121 = 2112,1313 = 1331 = 3131 = 3113,<br>2323 = 2332 = 3232 = 3223<br>$b_1 \equiv$ 1112 = 1121 = 1211 = 2111, 2221 = 2212 = 2122 = 1222,<br>1233 = 2133 = 3312 = 3321,<br>1323 = 1332 = 3123 = 3132 = 2313 = 2331 = 3213 = 3231<br>$c_1 \equiv$ 1113 = 1131 = 1311 = 3111, 3331 = 3313 = 3133 = 1333,<br>1322 = 3122 = 2213 = 2231,<br>1232 = 1223 = 2132 = 2123 = 3212 = 2312 = 3221 = 2321,<br>2223 = 2232 = 2322 = 3222, 3332 = 3323 = 3233 = 2333,<br>2311 = 3211 = 1123 = 1132,<br>2131 = 2113 = 1231 = 1213 = 3121 = 1321 = 3112 = 1312 |
| $2, \underline{2}, m, \underline{m}, 2/m, \underline{2}/m, 2/\underline{m}, \underline{2}/\underline{m}$ | 41 | 13 | $a_1, b_1$ |
| $222, \underline{22}2, mm2, \underline{mm}2, \underline{2m}m, mmm, \underline{mm}m, \underline{mmm}, mm\underline{m}$ | 21 | 9 | $a_1$ |
| $4, \underline{4}, \bar{4}, \underline{\bar{4}}, 4/m, \underline{4}/m, 4/\underline{m}, \underline{4}/\underline{m}$ | 29 | 7 | $d_1 \equiv$ 1111 = 2222, 3333, 1122 = 2211,<br>1133 = 3311 = 2233 = 3322, 1212 = 1221 = 2112 = 2121,<br>2323 = 2332 = 3223 = 3232 = 1313 = 1331 = 3113 = 3131<br>$e_1 \equiv$ 1112 = −2221 = 1121 = −2212 =<br>1211 = −2122 = 2111 = −1222 |
| $422, \underline{4}2\underline{2}, 4\underline{22}, 4mm, \underline{4}m\underline{m}, 4\underline{mm}, \bar{4}2m, \underline{\bar{4}}2\underline{m}, \underline{\bar{4}}m\underline{2}, \bar{4}\underline{2m}, 4/mmm, \underline{4}/mm\underline{m}, 4/m\underline{mm}, 4/\underline{mmm}, 4/\underline{m}mm, \underline{4}/\underline{mmm}$ | 21 | 6 | $d_1$ |

TABLE I *(Continued)*

| Magnetic Point Group | N | I | Form of the i - tensor Re $\gamma_{\alpha\beta\gamma\delta}$ |
|---|---|---|---|
| $3, \bar{3}, \underline{\bar{3}}$ | 53 | 7 | $h_I \equiv 1111 = 2222 = 1122 + 2(1212), 3333, 1122 = 2211,$<br>$1212 = 1221 = 2112 = 2121, 1133 = 3311 = 2233 = 3322,$<br>$1313 = 1331 = 3113 = 3131 = 2323 = 2332 = 3223 = 3232$<br>$j_I \equiv 1113 = -1232 = -1223 = -2132 =$<br>$1131 = -2123 = -3221 = -2312 =$<br>$1311 = -3212 = -2321 = -1322 =$<br>$3111 = -3122 = -2213 = -2331,$<br>$k_I \equiv 2223 = -2131 = -2113 = -1231 =$<br>$2232 = -1213 = -3112 = -1321 =$<br>$2322 = -3121 = -1312 = -2311 =$<br>$3222 = -3211 = -1123 = -1132$ |
| $32, 3\underline{2}, 3m, \bar{3}m, 3\underline{m},$<br>$\bar{3}\underline{m}, \underline{\bar{3}m}, \underline{\bar{3}}m$ | 37 | 6 | $h_I, j_I$ |
| $6, \underline{6}, \bar{6}, \underline{\bar{6}}, 6/m, \underline{6}/m,$<br>$6/\underline{m}, \underline{6}/\underline{m}, 622, 6\underline{2}\underline{2},$<br>$6\underline{22}, 6mm, \underline{6}m\underline{m}, 6\underline{mm},$<br>$\bar{6}m2, \underline{\bar{6}}m\underline{2}, \underline{\bar{6}}\underline{2}\underline{m}, \bar{6}\underline{m2},$<br>$6/mmm, \underline{6}/\underline{m}m\underline{m},$<br>$6/m\underline{mm}, 6/\underline{mmm},$<br>$6/\underline{m}mm, \underline{6}/m\underline{m}m,$<br>$\infty, \infty/m, \infty m, \infty/\underline{m},$<br>$\infty\underline{m}, \infty/mm, \infty/\underline{m}m,$<br>$\infty/m\underline{m}$ | 21 | 5 | $h_I$ |
| $23, m\underline{3}, \underline{m}3, 432, 4\underline{3}\underline{2},$<br>$\bar{4}3m, \underline{\bar{4}}3\underline{m}, m3m,$<br>$\underline{m}3m, m3\underline{m}, \underline{m}3\underline{m},$ | 21 | 3 | $m_I \equiv 1111 = 2222 = 3333,$<br>$1122 = 2233 = 3311 = 2211 = 3322 = 1133,$<br>$1212 = 2323 = 3131 = 1221 = 2332 = 3113 =$<br>$2121 = 3232 = 1313 = 1331 = 2112 = 3223 = 1331$ |
| $Y, Y_h, K, K_h$ | 21 | 2 | $m_I$ and $1111 = 2222 = 3333 = 1122 + 2(1212)$ |

*Note.* The components of the nonlinear polarizability tensor Re $\gamma_{\alpha\beta\gamma\delta}$ are denoted by the subscripts $\alpha\beta\gamma\delta$, taking values 1, 2, 3 in the molecular reference frame. N and I denote the number of nonzero and independent components, respectively. Sets of components recurring in various point groups are denoted by lowercase letters.

TABLE II
Fourth-Rank Axial i-tensor Im $\sigma_{\alpha\beta\gamma\delta}$ for 102 Magnetic Point Groups

| Magnetic Point Group | N | I | Form of the i-tensor Im $\sigma_{\alpha\beta\gamma\delta}$ |
|---|---|---|---|
| 1 | 81 | 54 | $a_2 \equiv$ 1111, 2222, 3333,<br>1122, 2211, 1133, 3311, 2233, 3322,<br>1212 = 1221, 1313 = 1331, 2323 = 2332,<br>2121 = 2112, 3131 = 3113, 3232 = 3223<br>$b_2 \equiv$ 1112 = 1121, 1211, 2111, 2221 = 2212, 1222, 2122,<br>1233, 2133, 3312 = 3321, 1323 = 1332,<br>3123 = 3132, 2313 = 2331, 3213 = 3231<br>$c_2 \equiv$ 1113 = 1131, 1311, 3111, 3331 = 3313, 1333, 3133,<br>1322, 3122, 2213 = 2231, 1232 = 1223,<br>2132 = 2123, 3212 = 3221, 2312 = 2321,<br>2223 = 2232, 2322, 3222, 3332 = 3323, 2333, 3233,<br>2311, 3211, 1123 = 1132, 2131 = 2113,<br>1231 = 1213, 3121 = 3112, 1321 = 1312 |
| $2, \underline{2}$ | 41 | 28 | $a_2, b_2$ |
| $m, \underline{m}$ | 40 | 26 | $c_2$ |
| $222, \underline{22}2$ | 21 | 15 | $a_2$ |
| $mm2, \underline{mm}2, \underline{2m}m$ | 20 | 13 | $b_2$ |
| $4, \underline{4}$ | 39 | 14 | $d_2 \equiv$ 1111 = 2222, 3333,<br>1122 = 2211, 1212 = 1221 = 2121 = 2112,<br>1133 = 2233, 1313 = 1331 = 2323 = 2332,<br>3311 = 3322, 3131 = 3113 = 3232 = 3223<br>$e_2 \equiv$ 1112 = −2221 = 1121 = −2212, 1211 = −2122,<br>2111 = −1222,<br>1233 = −2133, 1323 = −2313 = 1332 = −2331,<br>3123 = −3213 = 3132 = −3231 |
| $\bar{4}, \underline{\bar{4}}$ | 40 | 14 | $f_2 \equiv$ 1111 = −2222,<br>1122 = −2211, 1212 = 1221 = −2121 = −2112,<br>1133 = −2233, 1313 = 1331 = −2323 = −2332,<br>3311 = −3322, 3131 = 3113 = −3232 = −3223<br>$g_2 \equiv$ 1112 = 2221 = 1121 = 2212, 1211 = 2122,<br>2111 = 1222,<br>1233 = 2133, 1323 = 2313 = 1332 = 2331,<br>3312 = 3321, 3123 = 3213 = 3132 = 3231 |
| $422, \underline{4}2\underline{2}, 4\underline{22}$ | 21 | 8 | $d_2$ |
| $4mm, \underline{4}m\underline{m}, 4\underline{mm}$ | 18 | 6 | $e_2$ |
| $\bar{4}2m, \underline{\bar{4}}2\underline{m}, \underline{\bar{4}}m\underline{2}, \bar{4}\underline{2m}$ | 20 | 7 | $f_2$ |
| 3 | 71 | 18 | $h_2 \equiv$ 1111 = 2222 = 1122 + 2(1212), 3333,<br>1212 = 1221 = 2112 = 2121, 1122 = 2211,<br>1313 = 1331 = 2323 = 2332, 1133 = 2233,<br>3131 = 3113 = 3232 = 3223, 3311 = 3322<br>$i_2 \equiv$ 1112 = −2221 = 1121 = −2212 = $-\frac{1}{2}$(1211 + 2111),<br>2111 = −1222, 1211 = −2122,<br>1233 = −2133, 1323 = −2313 = 1332 = −2331,<br>3123 = −3213 = 3132 = −3231 |

TABLE II *(Continued)*

| Magnetic Point Group | N | I | Form of the i - tensor Re $g_{\alpha\beta\gamma\delta}$ |
|---|---|---|---|
| | | | $j_2 \equiv$ 1113 = −1223 = −1232 = −2123 =<br>1131 = −2132 = −2213 = −2231,<br>1311 = −1322 = −2312 = −2321,<br>3111 = −3122 = −3212 = −3221<br>$k_2 \equiv$ 2223 = −2113 = −2131 = −1213 =<br>2232 = −1231 = −1123 = −1132,<br>2322 = −2311 = −1321 = −1312,<br>3222 = −3211 = −3121 = −3112 |
| $32, 3\underline{2}$ | 37 | 10 | $h_2, j_2$ |
| $3m, 3\underline{m}$ | 34 | 8 | $i_2, k_2$ |
| $6, \underline{6}, \infty$ | 39 | 12 | $h_2, i_2$ |
| $\bar{6}, \underline{\bar{6}}$ | 32 | 6 | $j_2, k_2$ |
| $622, \underline{6}2\underline{2}, 6\underline{22}$ | 21 | 7 | $h_2$ |
| $6mm, \underline{6}m\underline{m}, 6\underline{mm}, \infty m,$<br>$\infty\underline{m}$ | 18 | 5 | $i_2$ |
| $\bar{6}m2, \underline{\bar{6}}m\underline{2}, \underline{\bar{6}}2\underline{m}, \bar{6}\underline{m2}$ | 16 | 3 | $k_2$ |
| 23 | 21 | 5 | $l \equiv$ 1111 = 2222 = 3333.<br>1122 = 2233 = 3311, 2211 = 1133 = 3322,<br>1212 = 2323 = 3131 = 1221 = 2332 = 3113,<br>2121 = 3232 = 1313 = 2112 = 3223 = 1331 |
| $432, \underline{4}3\underline{2}$ | 21 | 3 | $m_2 \equiv$1111 = 2222 = 3333,<br>1122 = 2233 = 3311 = 2211 = 1133 = 3322,<br>1212 = 2323 = 3131 = 1221 = 2332 = 3113 =<br>2121 = 3232 = 1313 = 2112 = 3223 = 1331 |
| $\bar{4}3m, \underline{\bar{4}}3\underline{m}$ | 18 | 2 | $0_2 \equiv$ 1122 = 2233 = 3311 = −2211 = −1133 = −3322,<br>1212 = 2323 = 3131 = −2121 = −3232 = −1313 =<br>1221 = 2332 = 3113 = −2112 = −3223 = −1331 |
| $Y, K$ | 21 | 2 | $m_2$ and 1111 = 2222 = 3333 = 1122 + 2(1212) |

In the remaining groups:

$\bar{1}, \underline{\bar{1}}, 2/m, \underline{2}/m, 2/\underline{m}, \underline{2}/\underline{m}, mmm, \underline{mm}m, \underline{mmm}, mm\underline{m}, 4/m, \underline{4}/m, 4/\underline{m}, \underline{4}/\underline{m}, 4/mmm,$
$\underline{4}/mm\underline{m}, 4/m\underline{mm}, 4/\underline{mmm}, 4/\underline{m}mm, \underline{4}/\underline{mm}m, \bar{3}, \underline{\bar{3}}, \bar{3}m, \bar{3}\underline{m}, \underline{\bar{3}m}, \bar{3}m, 6/m, \underline{6}/m, 6/\underline{m}, \underline{6}/\underline{m},$
$6/mmm, \underline{6}/\underline{m}m\underline{m}, 6/m\underline{mm}, 6/\underline{mmm}, 6/\underline{m}mm, \underline{6}/m\underline{m}m, \infty/m, \infty/\underline{m}, \infty/mm, \infty/\underline{mm}, \infty/m\underline{m},$
$m3, \underline{m}3, m3m, \underline{m}3m, m3\underline{m}, \underline{m}3\underline{m}, Y_h$ and $K_h$

all components vanish

*Note*. The components of the nonlinear polarizability tensor Im $\sigma_{\alpha\beta\gamma\delta}$ are denoted by the subscripts $\alpha\beta\gamma\delta$, taking values 1, 2, 3 in the molecular reference frame. N and I denote the number of nonzero and independent components, respectively. Sets of components recurring in various point groups are denoted by lowercase letters.

TABLE III
Fifth-Rank Polar i-tensor Re $\eta_{\alpha(\beta\gamma)\delta\phi}$ for 102 Magnetic Point Groups

| Magnetic Point Group | N | I | Form of the i-tensor Re $\eta_{\alpha(\beta\gamma)\delta\phi}$ |
|---|---|---|---|
| 1<br>$4m2$, | 243 | 108 | $a_3 \equiv$ 11123 = 11132, 11312 = 11321 = 13112 = 13121,<br>31112 = 31121, 11213 = 11231 = 12113 = 12131,<br>12311 = 13211, 31211 = 32111, 21113 = 21131,<br>21311 = 23111,<br>22213 = 22231, 22321 = 22312 = 23221 = 23212,<br>32221 = 32212, 22123 = 22132 = 21223 = 21232,<br>21322 = 23122, 32122 = 31222, 12223 = 12232,<br>12322 = 13222,<br>33312 = 33321, 33123 = 33132 = 31332 = 31323,<br>31233 = 32133, 32331 = 32313 = 33231 = 33213,<br>13332 = 13323, 13233 = 12333, 23331 = 23313,<br>23133 = 21333<br>$b_3 \equiv$ 33333,<br>11113 = 11131, 11311 = 13111, 31111,<br>22223 = 22232, 22322 = 23222, 32222,<br>11223 = 11232 = 12123 = 12132, 11322 = 13122,<br>31122, 32211, 22113 = 22131 = 21213 = 21231,<br>22311 = 23211, 12312 = 12321 = 13212 = 13221,<br>12213 = 12231, 31212 = 31221 = 32112 = 32121,<br>21123 = 21132, 23121 = 23112 = 21321 = 21312,<br>11333 = 13133, 13313 = 13331, 33311, 31133,<br>31313 = 31331 = 33113 = 33131,<br>22333 = 23233, 23323 = 23332, 33322, 32233,<br>32323 = 32332 = 33223 = 33232<br>$c_3 \equiv$ 11111, 22222,<br>11112 = 11121, 11211 = 12111, 21111,<br>22221 = 22212, 22122 = 21222, 12222,<br>33331 = 33313, 33133 = 31333, 13333,<br>33332 = 33323, 33233 = 32333, 23333,<br>11332 = 11323 = 13132 = 13123, 11233 = 12133,<br>13213 = 13231 = 12313 = 12331, 32311 = 33211,<br>21313 = 21331 = 23113 = 23131, 21133, 23311,<br>31312 = 31321 = 33112 = 33121, 31132 = 31123,<br>31231 = 31213 = 32131 = 32113, 13312 = 13321,<br>22331 = 22313 = 23231 = 23213, 22133 = 21233,<br>23123 = 23132 = 21323 = 21332, 31322 = 33122,<br>12323 = 12332 = 13223 = 13232, 12233, 13322,<br>32321 = 32312 = 33221 = 33212, 32231 = 32213,<br>32132 = 32123 = 31232 = 31223, 23321 = 23312,<br>11222 = 12122, 12212 = 12221, 21122,<br>21221 = 21212 = 22121 = 22112, 22211,<br>22111 = 21211, 21121 = 21112, 12211,<br>12112 = 12121 = 11212 = 11221, 11122,<br>33111 = 31311, 31131 = 31113, 13311,<br>13113 = 13131 = 11313 = 11331, 11133,<br>33222 = 32322, 32232 = 32223, 23322,<br>23223 = 23232 = 22323 = 22332, 22233 |

TABLE III *(Continued)*

| Magnetic Point Group | N | I | Form of the i-tensor Re $\eta_{\alpha(\beta\gamma)\delta\phi}$ |
|---|---|---|---|
| $2, \underline{\bar{2}}$ | 121 | 52 | $a_3, b_3$ |
| $m, \underline{m}$ | 122 | 56 | $c_3$ |
| $222, \underline{22}2$ | 60 | 24 | $a_3$ |
| $mm2, \underline{mm}2, \underline{2m}m$ | 61 | 28 | $b_3$ |
| $4, \underline{4}$ | 117 | 26 | $d_3 \equiv$ 11123 = 11132 = −22213 = −22231,<br>31112 = 31121 = −32221 = −32212,<br>11312 = 11321 = −22321 = −22312 =<br>13112 = 13121 = −23221 = −23212,<br>11213 = 11231 = −22123 = −22132 =<br>12113 = 12131 = −21223 = −21232,<br>12311 = 13211 = −21322 = −23122,<br>31211 = 32111 = −32122 = −31222,<br>21113 = 21131 = −12223 = −12232,<br>21311 = 23111 = −12322 = −13222,<br>12333 = 13233 = −21333 = −23133,<br>13323 = 13332 = −23313 = −23331,<br>31323 = 31332 = −32313 = −32331 =<br>33123 = 33132 = −33213 = −33231<br>$e_3 \equiv$ 33333, 11113 = 11131 = 22223 = 22232,<br>11311 = 13111 = 22322 = 23222, 31111 = 32222,<br>11322 = 13122 = 22311 = 23211,<br>12123 = 12132 = 11223 = 11232 = 21213 = 21231 =<br>22113 = 22131, 12312 = 12321 = 13212 = 13221 =<br>21321 = 21312 = 23121 = 23112,<br>31212 = 31221 = 32112 = 32121,<br>12213 = 12231 = 21123 = 21132, 31122 = 32211,<br>11333 = 13133 = 22333 = 23233, 31133 = 32233,<br>13313 = 13331 = 23323 = 23332, 33311 = 33322,<br>31313 = 31331 = 33113 = 33131 =<br>32323 = 32332 = 33223 = 33232 |
| $\bar{4}, \underline{\bar{4}}$ | 116 | 26 | $f_3 \equiv$ 11123 = 11132 = 22213 = 22231,<br>31112 = 31121 = 32221 = 32212,<br>11312 = 11321 = 22321 = 22312 =<br>13112 = 13121 = 23221 = 23212,<br>11213 = 11231 = 22123 = 22132 =<br>12113 = 12131 = 21223 = 21232,<br>12311 = 13211 = 21322 = 23122,<br>31211 = 32111 = 32122 = 31222,<br>21113 = 21131 = 12223 = 12232,<br>21311 = 23111 = 12322 = 13222,<br>12333 = 13233 = 21333 = 23133,<br>13323 = 13332 = 23313 = 23331, 31233 = 32133,<br>31323 = 31332 = 32313 = 32331 =<br>33123 = 33132 = 33213 = 33231, 33312 = 33321<br>$g_3 \equiv$ 11113 = 11131 = −22223 = −22232,<br>11311 = 13111 = −22322 = −23222, 31111 = −32222, |

TABLE III *(Continued)*

| Magnetic Point Group | N | I | Form of the i-tensor Re $\eta_{\alpha(\beta\gamma)\delta\phi}$ |
|---|---|---|---|
| | | | 11322 = 13122 = −22311 = −23211,<br>12123 = 12132 = −21213 = −21231 =<br>11223 = 11232 = −22113 = −22131,<br>12312 = 12321 = −21321 = −21312 =<br>13212 = 13221 = −23121 = 23112,<br>12213 = 12231 = −21123 = −21132, 31122 = −32211,<br>11333 = 13133 = −22333 = −23233, 31133 = −32233,<br>13313 = 13331 = −23323 = −23332, 33311 = −33322,<br>31313 = 31331 = −32323 = −32332 =<br>33113 = 33131 = −33223 = −33232 |
| $422, \underline{4}2\underline{2}, 4\underline{22}$, | 56 | 11 | $d_3$ |
| $4mm, \underline{4}m\underline{m}, 4\underline{mm}$ | 61 | 15 | $e_3$ |
| $\bar{4}2m, \underline{\bar{4}}\,\underline{2m}, \underline{\bar{4}}m\underline{2}, \bar{4}\underline{2m}$ | 60 | 13 | $f_3$ |
| 3 | 229 | 36 | $h_3 \equiv$ 11123 = 11132 = −22213 = −22231 =<br>−2(11213) − 21113,<br>31112 = 31121 = −32221 = −32212 =<br>32122 = 31222 = −31211 = −32111,<br>11312 = 11321 = −22321 = −22312 =<br>13112 = 13121 = −23221 = −23212 =<br>−(1/2)[12311 + 21311],<br>11213 = 11231 = −22123 = −22132 =<br>12113 = 12131 = −21223 = −21232,<br>12311 = 13211 = −21322 = −23122,<br>21113 = 21131 = −12223 = −12232,<br>21311 = 23111 = −12322 = −13222,<br>12333 = 13233 = −21333 = −23133,<br>13323 = 13332 = −23313 = −23331,<br>31323 = 31332 = −32313 = −32331 =<br>33123 = 33132 = −33213 = −33231<br>$i_3 \equiv$ 11133 = −12233 = −21233 = −22133,<br>13311 = −13322 = −23312 = −23321,<br>11313 = −12323 = −21323 = −22313 =<br>11331 = −12332 = −21332 = −22331 =<br>13113 = −13223 = −23123 = −23213 =<br>13131 = −13232 = −23123 = −23231,<br>31131 = −31232 = −32132 = −32231 =<br>31113 = −32123 = −32213 = −31223,<br>31311 = −31322 = −32312 = −32321 =<br>33111 = −33122 = −33212 = −33221,<br>11111 = −(2/3)[22221 + 22122 + (1/2)12222],<br>11122 = (2/3)[2(22221) − 22122 − (1/2)12222],<br>11212 = 11221 = 12112 = 12121 =<br>(1/3)[22221 + 22122 − 12222],<br>12211 = (1/3)[−2(22221) + 4(22122) − 12222],<br>21112 = 21121 = (2/3)[(1/2)22221 − 22122 + 12222],<br>21211 = 22111 = (2/3)[−22221 + (1/2)22122 + 12222],<br>12222, 22221 = 22212, 22122 = 21222 |

TABLE III *(Continued)*

| Magnetic Point Group | N | I | Form of the i-tensor Re $\eta_{\alpha(\beta\gamma)\delta\phi}$ |
|---|---|---|---|
| | | | $j_3 \equiv$ 33333,<br>11333 = 13133 = 22333 = 23233, 31133 = 32233,<br>13313 = 13331 = 23323 = 23332, 33311 = 33322,<br>31313 = 31331 = 33113 = 33131 =<br>32323 = 32332 = 33223 = 33232,<br>11113 = 11131 = 22223 = 22232 = 2(11223) + 12213,<br>11311 = 13111 = 22322 = 23222 = 11322 + 2(12312),<br>31111 = 32222 = 31122 + 2(31212),<br>11223 = 11232 = 12123 = 12132 =<br>22113 = 22131 = 21213 = 21231,<br>12213 = 12231 = 21123 = 21132,<br>11322 = 13122 = 22311 = 23211,<br>12312 = 12321 = 13212 = 13221 =<br>21321 = 21312 = 23121 = 23112,<br>31212 = 31221 = 32112 = 32121, 31122 = 32211 |
| | | | $k_3 \equiv$ 22233 = −21133 = −12133 = −11233,<br>23322 = −23311 = −13321 = −13312,<br>22332 = −21331 = −12331 = −11332 =<br>22323 = −21313 = −12313 = −11323 =<br>23232 = −23131 = −13231 = −13132 =<br>23223 = −23113 = −13213 = −13123,<br>32232 = −32131 = −31231 = −31132 =<br>32223 = −32113 = −31213 = −31123,<br>33222 = −33121 = −33112 = −32311 =<br>32322 = −31321 = −31312 = −33211,<br>22222 = −(2/3)[11112 + 11211 + (1/2)21111],<br>22211 = (1/3)[4(11112) − 2(11211) − 21111],<br>22121 = 22112 = 21221 = 21212 =<br>(1/3)[11112 + 11211 − 21111],<br>21122 = (2/3)[−11112 + 2(11211) − (1/2)21111],<br>12221 = 12212 = (1/3)[11112 − 2(11211) − 2(21111)],<br>12122 = 11222 = (2/3)[−11112 + (1/2)11211 + 21111],<br>11112 = 11121, 11211 = 12111, 21111 |
| $32, 3\underline{2}$ | 112 | 16 | $h_3, i_3$ |
| $3m, 3\underline{m}$ | 117 | 20 | $j_3, k_3$ |
| $6, \infty, \underline{6}$ | 117 | 20 | $h_3, j_3$ |
| $\bar{6}, \underline{\bar{6}}$ | 112 | 16 | $i_3, k_3$ |
| $622, \underline{6}2\underline{2}, 6\underline{22}$ | 56 | 8 | $h_3$ |
| $6mm, \underline{6}m\underline{m}, 6\underline{mm}$,<br>$\infty m, \infty\underline{m}$ | 61 | 12 | $j_3$ |
| $\bar{6}m2, \underline{\bar{6}}\,\underline{2m}, \underline{\bar{6}}m\underline{2}, \bar{6}\underline{m2}$ | 56 | 8 | $i_3$ |
| 23 | 60 | 8 | $l_3 \equiv$ 11123 = 11132 = 22231 = 33312 = 22213 = 33321<br>11312 = 22123 = 33231 = 11321 = 22132 = 33213 =<br>13112 = 21223 = 32331 = 13121 = 21232 = 32313,<br>11213 = 22321 = 33132 = 11231 = 22312 = 33123 = |

TABLE III *(Continued)*

| Magnetic Point Group | N | I | Form of the i-tensor Re $\eta_{\alpha(\beta\gamma)\delta\phi}$ |
|---|---|---|---|
| | | | 12113 = 23221 = 31332 = 12131 = 23212 = 31323,<br>21113 = 32221 = 13332 = 21131 = 32212 = 13323,<br>21311 = 32122 = 13233 = 23111 = 31222 = 12333,<br>31112 = 12223 = 23331 = 31121 = 12232 = 23313,<br>31211 = 12322 = 23133 = 32111 = 13222 = 21333,<br>13211 = 21322 = 32133 = 12311 = 23122 = 31233 |
| $432, \underline{4}3\underline{2}$ | 48 | 3 | $m_3$ ≡11312 = 22123 = 33231 = 11321 = 22132 = 33213 =<br>13112 = 21223 = 32331 = 13121 = 21232 = 32313 =<br>−11213 = −22321 = −33132 = −11231 = −22312 =<br>−33123 = −12113 = −23221 = −31332 = −12131 =<br>−23212 = −31323,<br>21113 = 32221 = 13332 = 21131 = 32212 = 13323 =<br>−31112 = −12223 = −23331 = −31121 = −12232 =<br>−23313,<br>21311 = 32122 = 13233 = 23111 = 31222 = 12333 =<br>−31211 = −12322 = −23133 = −32111 = −13222 =<br>−21333 |
| $\bar{4}3m, \underline{\bar{4}}3\underline{m}$ | 60 | 5 | $0_3$ ≡ 11123 = 22231 = 33312 = 11132 = 22213 = 33321,<br>13211 = 21322 = 32133 = 12311 = 23122 = 31233,<br>11312 = 22123 = 33231 = 11321 = 22132 = 33213 =<br>13112 = 21223 = 32331 = 13121 = 21232 = 32313 =<br>11213 = 22321 = 33132 = 11231 = 22312 = 33123 =<br>12113 = 23221 = 31332 = 12131 = 23212 = 31323,<br>21113 = 32221 = 13332 = 21131 = 32212 = 13323 =<br>31112 = 12223 = 23331 = 31121 = 12232 = 23313,<br>21311 = 32122 = 13233 = 23111 = 31222 = 12333 =<br>31211 = 12322 = 23133 = 13222 = 21333 = 32111 |
| $Y, K$ | 48 | 1 | 11213 = 22321 = 33132 = 11231 = 22312 = 33123 =<br>12113 = 23221 = 31332 = 12131 = 23212 = 31323 =<br>−11312 = −22123 = −33231 = −11321 = −22132 =<br>−33213 = −13112 = −21223 = −32331 = −13121 =<br>−21232 = −32313 = (1/2)21311,<br>21311 = 32122 = 13233 = 23111 = 31222 = 12333 =<br>31112 = 12223 = 23331 = 31121 = 12232 = 23313 =<br>−31211 = −12322 = −23133 = −32111 = −13222 =<br>−21333 = −21113 = −32221 = −13332 = −21131 =<br>−32212 = −13323 |

In the remaining magnetic point groups:
$\bar{1}$, $\underline{\bar{1}}$, $2/m$, $\underline{2}/m$, $2/\underline{m}$, $\underline{2}/\underline{m}$, $mmm$, $\underline{mm}m$, $\underline{mmm}$, $mm\underline{m}$, $4/m$, $\underline{4}/m$, $4/\underline{m}$, $\underline{4}/\underline{m}$, $4/mmm$, $\underline{4}/mm\underline{m}$, $4/m\underline{mm}$, $4/\underline{mmm}$, $4/\underline{m}mm$, $\underline{4}/\underline{mm}m$, $\bar{3}$, $\underline{\bar{3}}$, $\bar{3}m$, $\bar{3}\underline{m}$, $\underline{\bar{3}m}$, $\underline{\bar{3}}m$, $6/m$, $\underline{6}/m$, $6/\underline{m}$, $\underline{6}/\underline{m}$, $6/mmm$, $\underline{6}/\underline{m}m\underline{m}$, $6/\underline{mmm}$, $6/\underline{mmm}$, $6/m\underline{m}m$, $\underline{6}/mm\underline{m}$, $\infty/m$, $\infty/\underline{m}$, $\infty/mm$, $\infty/\underline{m}m$, $\infty/m\underline{m}$, $m3$, $\underline{m}3$, $m3m$, $\underline{m}3m$, $m3\underline{m}$, $\underline{m3m}$, $Y_h$ and $K_h$ all components vanish

*Note.* The components of the nonlinear polarizability tensor Re $\eta_{\alpha(\beta\gamma)\delta\phi}$ are denoted by the subscripts $\alpha\beta\gamma\delta\phi$, taking values 1, 2, 3 in the molecular reference frame. N and I denote the number of nonzero and independent components, respectively. Sets of components recurring in various point groups are denoted by lowercase letters.

TABLE IV
Linear $\chi_R^L, \chi_A^L$ and Nonlinear $\chi_R^{NL}, \kappa_R^{NL}, \chi_A^{NL}$ Molecular Parameters for 102 Magnetic Point Groups

| | Hamiltonian $H_I$ | | | | |
|---|---|---|---|---|---|
| | $H_I^{(2)}$ | | $H_I^{(4)}$ | | |
| Magnetic Point Group | $\chi_R^L$ | $\chi_A^L$ | $\chi_R^{NL}$ | $\kappa_R^{NL}$ | $\chi_A^{NL}$ |
| $1, 2, \underline{2}, 222, 2\underline{22}$ | $\chi_R^L$ | $\chi_A^L$ | $\chi_R^{NL}$ | $\kappa_R^{NL}$ | $\chi_A^{NL}$ |
| $\bar{1}, \underline{\bar{1}}, m, \underline{m}, 2/m, \underline{2}/m, 2/\underline{m}, \underline{2}/\underline{m}, mm2, \underline{mm}2,$ | $\chi_R^L$ | 0 | $\chi_R^{NL}$ | $\kappa_R^{NL}$ | 0 |
| $\underline{2m}m, mmm, \underline{mm}m, \underline{mmm}, mm\underline{m}$ | | | | | |
| $4, \underline{4}, 422, \underline{4}2\underline{2}, 4\underline{22}$ | $a_1$ | $b_1$ | $c_1$ | $d_1$ | $e_1$ |
| $\bar{4}, \underline{\bar{4}}, 4/m, 4/\underline{m}, \underline{4}/\underline{m}, 4mm, \underline{4}m\underline{m}, 4/\underline{mm}, \bar{4}2m, \underline{\bar{4}}2\underline{m},$ | $a_1$ | 0 | $c_1$ | $d_1$ | 0 |
| $\underline{\bar{4}}m\underline{2}, 4\underline{2m}, 4/mmm, \underline{4}/mm\underline{m}, 4/m\underline{mm}, 4/\underline{mmm}, 4/\underline{m}mm,$ | | | | | |
| $\underline{4}/\underline{mm}m$ | | | | | |
| $3, 32, 3\underline{2}, 6, \underline{6}, \infty, 622, \underline{6}2\underline{2}, 6\underline{22}$ | $a_1$ | $b_1$ | $c_2$ | $d_2$ | $e_2$ |
| $\bar{3}, \underline{\bar{3}}, 3m, 3\underline{m}, \bar{3}m, \bar{3}\underline{m}, \underline{\bar{3}m}, \underline{\bar{3}}m, \bar{6}, \underline{\bar{6}}, 6/m, \underline{6}/m, 6/\underline{m}, \underline{6}/\underline{m},$ | $a_1$ | 0 | $c_2$ | $d_2$ | 0 |
| $\infty/m, \infty/\underline{m}, 6mm, \underline{6}m\underline{m}, 6/\underline{mm}, \infty m, \infty\underline{m}, \bar{6}m2, \underline{\bar{6}}2\underline{m},$ | | | | | |
| $\underline{\bar{6}}m\underline{2}, \bar{6}\underline{m2}, 6/mmm, \underline{6}/\underline{m}m\underline{m}, \underline{6}/m\underline{mm}, 6/\underline{mmm}, 6/\underline{m}mm,$ | | | | | |
| $\underline{6}/m\underline{m}m, \infty/mm, \infty/\underline{m}m, \infty/m\underline{m}$ | | | | | |
| $23$ | $a_2$ | $b_2$ | $c_3$ | $d_3$ | $e_3$ |
| $432, \underline{4}3\underline{2}$ | $a_2$ | $b_2$ | $c_3$ | $d_3$ | $e_4$ |
| $m3, \underline{m}3, \bar{4}3m, \underline{\bar{4}}3\underline{m}, m3m, \underline{m}3m, m3\underline{m}, \underline{m}3\underline{m}$ | $a_2$ | 0 | $c_3$ | $d_3$ | 0 |
| $Y, K$ | $a_2$ | $b_2$ | $c_4$ | $d_4$ | $e_5$ |
| $Y_h, K_h$ | $a_2$ | 0 | $c_4$ | $d_4$ | 0 |

*Note.* where we used the notation

$$\chi_R^L = \frac{N}{3}\operatorname{Re}(\alpha_{11} + \alpha_{22} + \alpha_{33}) \qquad \chi_A^L = -\frac{\mathrm{i}Nk_z}{3\omega}\operatorname{Im}(\rho_{11} + \rho_{22} + \rho_{33})$$

$$a_1 = \frac{N}{3}\operatorname{Re}(2\alpha_{11} + \alpha_{33}) \qquad b_1 = -\frac{\mathrm{i}Nk_z}{3\omega}\operatorname{Im}(2\rho_{11} + \rho_{33})$$

$$a_2 = N\operatorname{Re}\alpha_{11} \qquad b_2 = -\frac{\mathrm{i}Nk_z}{\omega}\operatorname{Im}\rho_{11}$$

$$\chi_R^{NL} = \frac{2N}{15}\operatorname{Re}\{\gamma_{1111} + \gamma_{2222} + \gamma_{3333} - \gamma_{1122} - \gamma_{1133} - \gamma_{2233} + 3[\gamma_{1212} + \gamma_{1313} + \gamma_{2323}]\}$$

$$c_1 = \frac{2N}{15}\operatorname{Re}\{2\gamma_{1111} + \gamma_{3333} - \gamma_{1122} - 2\gamma_{1133} + 3[\gamma_{1212} + 2\gamma_{1313}]\}$$

$$c_2 = \frac{2N}{15}\operatorname{Re}\{\gamma_{3333} + \gamma_{1122} - 2\gamma_{1133} + 7\gamma_{1212} + 6\gamma_{1313}\}$$

$$c_3 = \frac{2N}{5}\operatorname{Re}\{\gamma_{1111} - \gamma_{1122} + 3\gamma_{1212}\} \quad c_4 = 2N\operatorname{Re}\gamma_{1212}$$

TABLE IV *(Continued)*

$$\kappa_R^{NL} = \frac{2N}{15}\mathrm{Re}\{2(\gamma_{1111}+\gamma_{2222}+\gamma_{3333})+3(\gamma_{1122}+\gamma_{1133}+\gamma_{2233}) +\gamma_{1212}+\gamma_{1313}+\gamma_{2323}\}$$

$$d_1 = \frac{2N}{15}\mathrm{Re}\{2(2\gamma_{1111}+\gamma_{3333})+3(\gamma_{1122}+2\gamma_{1133})+\gamma_{1212}+2\gamma_{1313}\}$$

$$d_2 = \frac{2N}{15}\mathrm{Re}\{2\gamma_{3333}+7\gamma_{1122}+6\gamma_{1133}+9\gamma_{1212}+2\gamma_{1313}\}$$

$$d_3 = \frac{2N}{5}\mathrm{Re}\{2\gamma_{1111}+3\gamma_{1122}+\gamma_{1212}\} \quad d_4 = 2N\,\mathrm{Re}\{\gamma_{1122}+\gamma_{1212}\}$$

$$\chi_A^{NL} = -\frac{i4Nk_z}{15}\left\{\frac{1}{\omega}\mathrm{Im}[-2(\sigma_{1111}+\sigma_{2222}+\sigma_{3333})+\sigma_{1122}+\sigma_{1133} +\sigma_{2233}+\sigma_{2211}+\sigma_{3311}+\sigma_{3322}-3(\sigma_{1212} +\sigma_{1313}+\sigma_{2323}+\sigma_{2121}+\sigma_{3131}+\sigma_{3232})] -\frac{1}{3}\mathrm{Re}[\eta_{1(23)33}-\eta_{1(32)22}+\eta_{2(31)11}-\eta_{2(13)33}+\eta_{3(12)22}-\eta_{3(21)11} +\eta_{3(31)23}-\eta_{2(21)23}+\eta_{1(12)31}-\eta_{3(32)31}+\eta_{2(23)12}-\eta_{1(13)12} -\eta_{1(13)32}+\eta_{1(22)23}-\eta_{2(11)13}+\eta_{2(33)31}-\eta_{3(22)21}+\eta_{3(11)12}]\right\}$$

$$e_1 = -\frac{i8Nk_z}{15}\left\{\frac{1}{\omega}\mathrm{Im}[-2\sigma_{1111}-\sigma_{3333}+\sigma_{1122}+\sigma_{1133}+\sigma_{3311} -3(\sigma_{1212}+\sigma_{1313}+\sigma_{3131})] -\frac{1}{3}\mathrm{Re}[\eta_{1(23)33}+\eta_{2(31)11}+\eta_{3(12)22}+\eta_{3(31)23} +\eta_{1(12)31)}+\eta_{2(23)12}+\eta_{1(22)23}+\eta_{2(33)31}+\eta_{3(11)12}]\right\}$$

$$e_2 = -\frac{i8Nk_z}{15}\left\{\frac{1}{\omega}\mathrm{Im}[-\sigma_{3333}-\sigma_{1122}+\sigma_{1133}+\sigma_{3311}-7\sigma_{1212}-3(\sigma_{1313}+\sigma_{3131})] -\frac{1}{3}\mathrm{Re}[\eta_{1(23)33}+\eta_{2(31)11}+2\eta_{3(12)22}+\eta_{3(31)23} +\eta_{1(12)31}+\eta_{2(23)12}+\eta_{1(22)23}+\eta_{2(33)31}]\right\}$$

$$e_3 = -\frac{i4Nk_z}{5}\left\{\frac{1}{\omega}\mathrm{Im}[-2\sigma_{1111}+\sigma_{1122}+\sigma_{2211}-3(\sigma_{1212}+\sigma_{2121})] -\frac{1}{3}\mathrm{Re}[\eta_{1(23)33}-\eta_{1(32)22}+\eta_{3(31)23}-\eta_{2(21)23}-\eta_{1(33)32}+\eta_{1(22)23}]\right\}$$

$$e_4 = -\frac{i8Nk_z}{5}\left\{\frac{1}{\omega}\mathrm{Im}[-\sigma_{1111}+\sigma_{1122}-3\sigma_{1212}]-\frac{1}{3}\mathrm{Re}[\eta_{1(23)33}-\eta_{3(31)23}+\eta_{1(22)23}]\right\}$$

$$e_5 = i8Nk_z\left\{\frac{1}{\omega}\mathrm{Im}\,\sigma_{1212}+\frac{1}{6}\mathrm{Re}\,\eta_{1(23)33}\right\}$$

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