Self-diffraction and phase conjugation in a gas of atoms with a hyperfine level structure

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A cubic susceptibility tensor describing all nonlinear optical phenomena within a one-photon absorption line is derived for a gas of atoms with a hyperfine level structure. This tensor is valid for either degenerate or nondegenerate four-wave mixing. It thus becomes possible to study the effect of the hyperfine structure and of the nuclear spin on self-diffraction, phase conjugation, and the self-effects of steady-state light waves in resonance with a common one-photon transition. Those amplitude properties and polarization properties of these nonlinear phenomena which stem exclusively from the hyperfine structure and the nuclear spin are determined. It thus becomes possible to experimentally identify transitions in terms of the electron angular momentum and the total angular momentum with allowance for the nuclear spin. It also becomes possible to select optimum conditions for optical image transmission and optical information processing. In several nonlinear phenomena involving degenerate four-wave mixing and the self-effects of light waves, a deviation from resonance with the hyperfine sublevels does not eliminate the effects of either the hyperfine structure itself or the nuclear spin, because of the resonance at zero frequency.

Experimental studies of self-diffraction and phase conjugation of steady-state light waves on resonant one-photon transitions have been carried out in monatomic gases[1–8] and also in mixed gaseous media containing active atoms along with a buffer gas.[9–13] Experiments on self-diffraction and phase conjugation in the steady state in molecular gases and solids are cited in reviews[14,15] and books.[16–18] The theoretical work on these nonlinear phenomena has often been carried out in the scalar version, without consideration of the vector nature of the electric field of the steady-state light waves and without consideration of the degeneracy of the resonant levels.[19–22,14,15] These factors have, on the other hand, been taken into account in several studies.[23–26] A model description of the resonant gas medium has been used in calculating polarization characteristics. The phase conjugation of steady-state light waves with linear and circular polarizations was studied theoretically in Ref. 27 by a quantum-mechanical approach. The very simple relaxation model used there introduces three characteristic constants in the equation for the density matrix of the moving gas atoms. The most comprehensive descriptions of these nonlinear phenomena, incorporating depolarizing elastic collisions and transitions of atoms to lower levels as a result of spontaneous emission in higher levels, were given in Refs. 28–30 and 7.

In all these papers, however, the theoretical discussion concerned gas atoms with a zero nuclear spin or a two-level model of an atom with a nonzero nuclear spin. For this reason, the results derived there are not suitable for describing those aspects of the behavior of self-diffraction and phase conjugation within a one-photon absorption line of the atom which stem exclusively from the hyperfine structure of the resonant levels. In contrast, the overwhelming majority of experiments on four-wave mixing have been carried out in gases of atoms with a nonzero nuclear spin. We thus see the need for a rigorous theory of self-diffraction and phase conjugation of steady-state light waves in which the hyperfine structure components of the resonant levels are taken into account.

In the present paper we analyze self-diffraction and phase conjugation of steady-state light waves of arbitrary polarization in a gas of atoms with a hyperfine level structure under some general assumptions. The degeneracy of the resonant levels, the thermal motion, the transition of atoms to a lower level as a result of radiative decay of the excited state, the linear absorption, and the changes caused in the refractive indices for the light waves by the presence of a buffer gas are all taken into account simultaneously. The intensities of the light waves are assumed to be low enough that saturation can be ignored, and we can use a perturbation theory. This theory is valid for both degenerate and nondegenerate four-wave mixing of light waves which are in resonance with the same one-photon transition of the atom.

To maintain a common approach in studying all the nonlinear phenomena which occur within a one-photon absorption line, we first calculate the cubic susceptibility tensor of a gas which includes resonant atoms with a hyperfine level structure as well as various impurities. The components of this tensor contain a triple resonance at frequencies of the incident waves and differences between these frequencies. A characteristic feature of this tensor is that it consists of two parts. One part stems from transitions of atoms to a lower level as a result of spontaneous decay of the excited state. This part of the tensor is substantially affected by the type of atomic transition and by the nuclear spin. In several cases, it dominates the nonlinear phenomena under consideration here.

Analysis of the cubic susceptibility tensor found here leads to several conclusions of general physical interest. The most important one is that the amplitude and polarization characteristics of self-diffraction, phase conjugation, and the nonlinear effects of light waves differ in their dependence on the hyperfine structure, the nuclear spin, and the degree of Doppler broadening. In several cases they are manifested

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in different ways in the cases of degenerate and nondegenerate four-wave mixing. When the Doppler width is small in comparison with the distance between components of the hyperfine structure in both resonant levels, only one hyperfine sublevel of the lower level and one of the upper level participate in each nonlinear phenomenon in the resonant situation. As a result, the calculations are simplified considerably, and the behavior emerges fairly clearly.

If the Doppler width is instead greater than the distance between the hyperfine components, in one or both levels, then several components of the hyperfine structure of one level participate in the physical process, even exactly in resonance at the frequency of the transition between two "hyperfine" sublevels. This situation seriously complicates the calculations, sometimes necessitating numerical methods. In the limiting case of large detuning from the resonances with hyperfine sublevels, either at the frequencies of the incident waves or at differences between them, the cubic susceptibility tensor derived here is the same as that for atoms with a zero nuclear spin. This assertion breaks down for several nonlinear phenomena which fall in the general category of degenerate four-wave mixing and self-effects of light waves, because of the resonance at zero frequency. It is impossible in principle to describe nonlinear phenomena of this sort within a one-photon absorption line on the basis of the model of a two-level atom without consideration of the hyperfine structure.

We will use the cubic susceptibility tensor derived here along with Maxwell's equations to calculate the interference electric field which describes self-diffraction, phase conjugation, and nonlinear effects when an arbitrary number of steady-state light waves are incident on a gas, for the case in which these waves are in resonance with the same one-photon transition. In the case of the self-diffraction of two light waves, we derive those amplitude and polarization characteristics which stem entirely from the hyperfine level structure and the nuclear spin. With this information, it becomes possible to identify the transitions $J_aF_a \to J_bF_b$ in terms of the electron angular momentum (J_a, J_b) and the total angular momentum (F_a, F_b), thereby distinguishing transitions $F_a = F \to F_b = F+1$ from transitions $F_a = F+1 \to F_b = F$ for all possible values of F, J_a, and J_b allowed by the selection rules. Corresponding relationships hold in the self-diffraction of three waves, when three or more light waves are incident on a gas. This information can be used to identify the transitions and also in optical information processing.

In the self-diffraction of counterpropagating waves, the case of most interest is phase conjugation in which the amplitude and polarization properties of the conjugate wave are governed exclusively by the hyperfine structure and the nuclear spin. Through a generalization of an experimental procedure used in phase conjugation,[24,25] we find a method for identifying the transitions $J_aF_a \to J_bF_b$ which yields the greatest amount of information for homogeneously broadened transitions, in the case in which transitions of atoms to a lower level as the result of the spontaneous decay of an excited state become a fundamental feature. It then becomes possible to experimentally distinguish transitions with a change in angular momentum $F_a = F \to F_b = F+1$ from transitions $F_a = F+1 \to F_b = F$. In several cases it also becomes possible to determine the ratio of relaxation constants. The distinctive amplitude and polarization properties which stem from the hyperfine structure and the nuclear spin are also seen in the case of the coupled waves which arise when four light waves, counterpropagating in pairs, are incident on a gas.

A general and important conclusion which is reached for all the nonlinear phenomena discussed here is that, despite the much greater complexity of the equations due to the hyperfine structure and the nuclear spin, there are still limiting cases, with strictly established values of the characteristic parameters, for which the amplitude and polarization properties of self-diffraction, phase conjugation, and self-effects of light waves can be described by simple equations which can easily be tested experimentally.

1. CUBIC SUSCEPTIBILITY TENSOR FOR ONE-PHOTON TRANSITIONS

We consider a gas of active atoms with nonzero nuclear spin. Each energy level of the atoms for $J \geqslant I$ $(J < I)$ is split into $2I+1$ $(2J+1)$ components with energies[31] $E_F = E + \hbar\Delta_F$, where $\hbar\Delta_F = (1/2)AC + BC(C+1)$, $C = F(F+1) - J(J+1) - I(I+1)$, and E is the energy of the atom if the nuclear spin is ignored. The quantity $\hbar\Delta_F$ is the hyperfine interaction energy; A and B are constants of the magnetic and quadrupole level splitting; and F, J, and I are the quantum numbers of the total angular momentum $(\mathbf{F} = \mathbf{J} + \mathbf{I})$, the electron angular momentum $\mathbf{J}$, and the nuclear spin $\mathbf{I}$. The state of an active atom in a hyperfine sublevel with an energy E_F is characterized by the quantum numbers J, I, and F and by the projection M_F of the total angular momentum $\mathbf{F}$ onto the quantization axis.

Some arbitrary number n_0 of steady-state plane waves is incident on this gas. The waves have frequencies ω_n, wave vectors $\mathbf{k}_n$, and complex amplitudes $\mathbf{a}_n$ with $n = 1,\ldots, n_0$. As a result of the nonlinear interaction of the incident waves with the active atoms, new waves form in the gas. The total electric field of all the light waves is

$$\mathbf{E}(\mathbf{r}, t) = \sum_n \mathbf{a}_n \exp[i(\mathbf{k}_n\mathbf{r} - \omega_n t)] + \text{c.c.}, \qquad (1)$$

where the terms with $n = 1,\ldots, n_0$ represent incident waves, while those with $n = n_0+1, n_0+2,\ldots,$ describe the new waves and nonlinear corrections to the incident waves. These corrections characterize nonlinear-effects of these waves. The frequencies of the waves in (1) may be the same, or they may take on different values near the frequency of the same one-photon transition of the active atom. The complex amplitudes $\mathbf{a}_n$ with $n = 1, 2,\ldots,$ either remain constant or vary slowly as functions of the coordinates.

The interaction of the active atoms with resonant waves (1) is described by a quantum-mechanical equation for the density matrix ρ in the FM_F representation in the resonant approximation:

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\nabla + i\omega_{F_bF_a} + \gamma_{ba}\right)\rho_{F_bM_{F_b}, F_aM_{F_a}}$$

$$= \frac{i}{\hbar}\left(\mathbf{E}\mathbf{d}_{F_bM_{F_b}, F_a'M_{F_a}'}\rho_{F_a'M_{F_a}', F_aM_{F_a}}\right.$$

$$\left. - \rho_{F_bM_{F_b}, F_b'M_{F_b}'}\mathbf{E}\mathbf{d}_{F_b'M_{F_b}', F_aM_{F_a}}\right), \qquad (2)$$

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla+i\omega_{F_bF_b'}+\gamma_b\right)\rho_{F_bM_{F_b},F_b'M_{F_b'}}$$

$$=\frac{\gamma_b N_b f(v)}{(2J_b+1)(2I+1)}\delta_{F_bF_b'}\delta_{M_{F_b}M_{F_b'}}$$

$$+\frac{i}{\hbar}(\mathbf{Ed}_{F_bM_{F_b},F_aM_{F_a}}\rho_{F_aM_{F_a},F_b'M_{F_b'}}$$

$$-\rho_{F_bM_{F_b},F_aM_{F_a}}\mathbf{Ed}_{F_aM_{F_a},F_b'M_{F_b'}}), \qquad (3)$$

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla+i\omega_{F_aF_a'}+\gamma_a\right)\rho_{F_aM_{F_a},F_a'M_{F_a'}}$$

$$=\frac{(\gamma_a N_a-\gamma N_b) f(v)}{(2J_a+1)(2I+1)}\delta_{F_aF_a'}\delta_{M_{F_a}M_{F_a'}}$$

$$+\frac{i}{\hbar}(\mathbf{Ed}_{F_aM_{F_a},F_bM_{F_b}}\rho_{F_bM_{F_b},F_a'M_{F_a'}}$$

$$-\rho_{F_aM_{F_a},F_bM_{F_b}}\mathbf{Ed}_{F_bM_{F_b},F_a'M_{F_a'}})$$

$$+\frac{\gamma(2J_b+1)}{|d_{ba}|^2}\mathbf{d}_{F_aM_{F_a},F_bM_{F_b}}\rho_{F_bM_{F_b},F_b'M_{F_b'}}\mathbf{d}_{F_b'M_{F_b'},F_a'M_{F_a'}}, \qquad (4)$$

$$\omega_{F_bF_a}=(E_{F_b}-E_{F_a})/\hbar=\omega_{ba}+\Delta_{F_b}-\Delta_{F_a}, \quad \omega_{ba}=(E_b-E_a)/\hbar,$$
$$\gamma_{ba}=(\gamma_b+\gamma_a)/2, \quad \gamma=4|d_{ba}|^2\omega_{ba}^3/3\hbar c^3(2J_b+1),$$
$$f(v)=(\pi^{1/2}u)^{-3}\exp(-v^2/u^2).$$

Here $\mathbf{d}_{F_bM_{F_b},F_aM_{F_a}}$ is the matrix element of the electric dipole moment operator $\mathbf{d}$; γ_{ba} is the half-width of the spectral line of the resonant transition; $\hbar\gamma_a$ and $\hbar\gamma_b$ are the homogeneous widths of the lower (E_a) and upper (E_b) resonant levels; γ is the probability for the spontaneous emission of a photon $\hbar\omega_{ba}$ by an isolated atom; d_{ba} is the reduced dipole moment of the resonant atomic transition $J_a \to J_b$; $f(v)$ is a Maxwell distribution; v is the velocity of the atom; u is the most probable velocity; and N_a (N_b) is the steady-state density of the atoms in level E_a (E_b) when all the hyperfine sublevels are taken into account, at $\mathbf{E}=0$. The quantities γ_a and γ_b stem from radiative decay and inelastic atomic collisions. The term containing γ in (4) describes transition of atoms to the lower level E_a as the result of spontaneous emission in the upper level E_b. The terms in (3) and (4) which contain N_a, N_b, and $f(v)$ incorporate a Boltzmann distribution of atoms among levels and a Maxwellian velocity distribution in statistical equilibrium, before the field (1) is turned on.

Equations (2)–(4) have been solved by perturbation theory in the cubic approximation in field (1). As a result, the vector

$$\mathbf{P}=\int \mathrm{Sp}(\rho\mathbf{d})d\mathbf{v}$$

which represents the dielectric polarization of the gas, can be written as the sum of a linear term $\mathbf{P}^L$ and a nonlinear term $\mathbf{P}^{NL}$:

$$\mathbf{P}(\mathbf{r},t)=\mathbf{P}^L(\mathbf{r},t)+\mathbf{P}^{NL}(\mathbf{r},t).$$

The linear term is

$$\mathbf{P}^L(\mathbf{r},t)=\sum_n \varkappa(\omega_n,\mathbf{k}_n)\mathbf{a}_n\exp[i(\mathbf{k}_n\mathbf{r}-\omega_n t)]+\text{c. c.},$$

where

$$\varkappa(\omega_n,\mathbf{k}_n)=-\sum_{F_aF_b}\frac{N_{ab}|d_{F_bF_a}|^2}{3\hbar(2I+1)}\int L_{F_bF_a}(\omega_n)f(v)d\mathbf{v}, \qquad (5)$$

and

$$N_{ab}=N_a/(2J_a+1)-N_b/(2J_b+1),$$
$$L_{F_bF_a}(\omega_n)=(\omega_n-\mathbf{k}_n\mathbf{v}-\omega_{F_bF_a}+i\gamma_{ba})^{-1},$$
$$d_{F_bF_a}=(-1)^{J_b+F_a+I+1}(2F_b+1)^{1/2}(2F_a+1)^{1/2}\begin{Bmatrix} J_b & F_b & I \\ F_a & J_a & 1 \end{Bmatrix} d_{ba}.$$

The nonlinear term is

$$P_i^{NL}(\mathbf{r},t)=\sum_{nmp}3\chi_{ijkl}(\omega_n+\omega_n-\omega_p;\omega_n,\omega_m,-\omega_p)a_{nj}a_{mk}a_{pl}^*$$
$$\times\exp\{i[(\mathbf{k}_n+\mathbf{k}_m-\mathbf{k}_p)\mathbf{r}-(\omega_n+\omega_m-\omega_p)t]\}+\text{c. c.} \qquad (6)$$

Here

$$\chi_{ijkl}(\omega_n+\omega_m-\omega_p;\omega_n,\omega_m,-\omega_p)$$
$$={}^1/_6(\chi^{(1)}_{nmp}\delta_{ij}\delta_{kl}+\chi^{(2)}_{nmp}\delta_{ik}\delta_{lj}+\chi^{(3)}_{nmp}\delta_{il}\delta_{jk}), \qquad (7)$$

and the coefficients are given by

$$\chi^{(1)}_{nmp}={}^1/_2(Q^{(1)}_{nmp}+Q^{(2)}_{nmp})+{}^1/_3(Q^{(0)}_{mnp}-Q^{(2)}_{mnp}),$$
$$\chi^{(2)}_{nmp}={}^1/_3(Q^{(0)}_{nmp}-Q^{(2)}_{nmp})+{}^1/_2(Q^{(1)}_{mnp}+Q^{(2)}_{mnp}),$$
$$\chi^{(3)}_{nmp}={}^1/_2(Q^{(2)}_{nmp}-Q^{(1)}_{nmp}+Q^{(2)}_{mnp}-Q^{(1)}_{mnp}).$$

Here

$$Q^{(\varkappa)}_{nmp}=N_{ab}\hbar^{-3}(2I+1)^{-1}\int d\mathbf{v}f(v)$$

$$\times\Bigg\{\sum_{F_aF_a'F_bF_b'}L_{F_bF_a}(\omega_n+\omega_m-\omega_p)$$

$$\times\Bigg[d^*_{F_a'F_b}d^*_{F_bF_a}d_{F_a'F_b'}d_{F_b'F_a}\begin{Bmatrix} 1 & \varkappa & 1 \\ F_a' & F_b & F_a \end{Bmatrix}$$

$$\times\begin{Bmatrix} 1 & \varkappa & 1 \\ F_a' & F_b' & F_a \end{Bmatrix}L_{F_a'F_a}(\omega_n-\omega_p)$$

$$\times(L^*_{F_b'F_a'}(\omega_p)-L_{F_b'F_a}(\omega_n))$$

$$+d^*_{F_bF_a}d^*_{F_aF_b'}d_{F_bF_a'}d_{F_a'F_b'}\begin{Bmatrix} 1 & \varkappa & 1 \\ F_b & F_a & F_b' \end{Bmatrix}$$

$$\times\begin{Bmatrix} 1 & \varkappa & 1 \\ F_b & F_a' & F_b' \end{Bmatrix}L_{F_bF_b'}(\omega_n-\omega_p)(L^*_{F_b'F_a'}(\omega_p)-L_{F_bF_a'}(\omega_n))\Bigg]$$

$$+\frac{i\gamma(2J_b+1)}{|d_{ba}|^2}\sum_{F_aF_a'F_a''F_bF_b'F_b''}d^*_{F_a'F_b}d^*_{F_bF_a}d_{F_b''F_a''}$$

$$\times d_{F_a''F_b}d^*_{F_b''F_a'}d_{F_b'F_a}$$

$$\times(-1)^{F_a+F_b''}\begin{Bmatrix} 1 & \varkappa & 1 \\ F_a' & F_b & F_a \end{Bmatrix}\begin{Bmatrix} 1 & \varkappa & 1 \\ F_b'' & F_a'' & F_b' \end{Bmatrix}$$

$$\times\begin{Bmatrix} F_a & \varkappa & F_a' \\ F_b'' & 1 & F_b' \end{Bmatrix}L_{F_bF_a}(\omega_n+\omega_m-\omega_p)$$

$$\times L_{F_a'F_a}(\omega_n-\omega_p)L_{F_b''F_b'}(\omega_n-\omega_p)$$

$$\times[L^*_{F_b'F_a''}(\omega_p)-L_{F_b''F_a''}(\omega_n)]\Bigg\}, \qquad (8)$$

$$L_{F_bF_a}(\omega_n+\omega_m-\omega_p)=[\omega_n+\omega_m-\omega_p$$
$$-(\mathbf{k}_n+\mathbf{k}_m-\mathbf{k}_p)\mathbf{v}-\omega_{F_bF_a}+i\gamma_{ba}]^{-1},$$
$$L_{F_aF_a'}(\omega_n-\omega_p)=[\omega_n-\omega_p-(\mathbf{k}_n-\mathbf{k}_p)\mathbf{v}-\omega_{F_aF_a'}+i\gamma_a]^{-1},$$
$$L_{F_bF_b'}(\omega_n-\omega_p)=[\omega_n-\omega_p-(\mathbf{k}_n-\mathbf{k}_p)\mathbf{v}-\omega_{F_bF_b'}+i\gamma_b]^{-1}.$$

Here $\varkappa = 0, 1, 2$; and a repeated vector index or a repeated tensor index (i,j,k,l) implies summation, in which these indices take on the values x, y, z, to characterize the vectors and tensors in the Cartesian coordinate system with axes x, y, z. The quantities a_{nj}, a_{mk}, and a_{pl} are projections of the vectors $\mathbf{a}_n$, $\mathbf{a}_m$, and $\mathbf{a}_p$ onto the Cartesian axes. The notation used for the $6j$ symbols and for the reduced dipole moment $(d_{F_bF_a})$ of the atomic transition $J_aF_a \to J_bF_b$ is the same as used in Ref. 32.

In general, the nonlinear phenomena of four-wave mixing and self-effects of waves can be described conveniently by means of a cubic susceptibility tensor.[33] This tensor is given by

$$P_i^{NL}(\mathbf{r},t)=\int_{-\infty}^{t}\chi_{ijkl}(t-t',t-t'',t-t''')E_j(\mathbf{r},t')$$
$$\times E_k(\mathbf{r},t'')E_l(\mathbf{r},t''')\,dt'\,dt''\,dt''', \tag{9}$$

where the tensor in an isotropic gas is symmetric under the interchange of any pair of the indices j, k, l when there is a simultaneous interchange of t', t'', and t'''. Substituting the electric field (1) into (9), and using the notation

$$\int_0^{\infty}\chi_{ijkl}(\tau_1,\tau_2,\tau_3)\exp[i(\omega_n\tau_1+\omega_m\tau_2+\omega_p\tau_3)]\,d\tau_1\,d\tau_2,\,d\tau_3$$
$$=\chi_{ijkl}(\omega_n+\omega_m+\omega_p;\ \omega_n,\omega_m,\omega_p). \tag{10}$$

for the cubic susceptibility tensor, we find a general expression which relates the nonlinear dielectric polarization of the gas (a vector) to the cubic susceptibility (a tensor) in the presence of steady-state plane waves:

$$P_i^{NL}(\mathbf{r},t)=\sum_{nmp}\{\chi_{ijkl}(\omega_n+\omega_m+\omega_p;\ \omega_n,\omega_m,\omega_p)a_{nj}a_{mk}a_{pl}$$
$$\times\exp[i((\mathbf{k}_n+\mathbf{k}_m+\mathbf{k}_p)\mathbf{r}-(\omega_n+\omega_m+\omega_p)t)]$$
$$+3\chi_{ijkl}(\omega_n+\omega_m-\omega_p;$$
$$\omega_n,\omega_m,-\omega_p)a_{nj}a_{mk}a_{pl}^*\exp[i((\mathbf{k}_n+\mathbf{k}_m-\mathbf{k}_p)\mathbf{r}$$
$$-(\omega_n+\omega_m-\omega_p)t)]\}+\text{c.c.} \tag{11}$$

The terms in (11) describe all possible nonlinear phenomena which fall in the category of four-wave mixing and self-effects. If the frequencies ω_n, ω_m, and ω_p, their sums, and their differences are not resonant, all the nonlinear phenomena embodied in (11) are equivalent. In a resonant situation, in contrast, the individual terms in (11) which contain the greatest number of resonances become the leading terms. They describe the specific nonlinear phenomenon which is characterized by this largest number of resonances. This specific nonlinear phenomenon emerges as a prominent phenomenon, in comparison with which the other phenomena appear weak. In this case the nonresonant terms and also the terms with smaller numbers of resonances create a background against which the main nonlinear phenomenon unfolds. In addition, all the terms of lower order contribute to interference effects. If we are interested in only the main nonlinear phenomenon, without interference effects, then we should omit the nonresonant terms from the general expression (11). We should also omit the terms with a number of resonances smaller than that in the leading terms.

In this problem, the frequencies ω_n, ω_m, and ω_p are in resonance with the one-photon transitions $J_aF_a \to J_bF_b$, with fixed J_a and J_b and with all possible values of F_a and F_b. The differences between these frequencies may vanish. In calculating the (vector) nonlinear dielectric polarization in (6), we thus retained only those terms which have a triple resonance. Among the three resonances, one occurs at a zero frequency, while the two others occur at frequencies from the set ω_n, ω_m, ω_p. The terms which have a triple resonance of this sort are contained in the second sum in the general expression (11). The first sum is a small correction in the problem at hand and can be omitted, as can the small corrections in the second sum in (11). It follows from a comparison of expression (6) with general expression (11) that the quantity in (7) is the part of the cubic susceptibility tensor [see (10)] which we are seeking: the part which describes four-wave mixing and nonlinear effects of waves in the case in which there is a triple resonance, at zero frequency and at frequencies which are in resonance with one-photon transitions $J_aF_a \to J_bF_b$ with various F_a's and F_b's. The tensor (7) without the terms containing γ can also be found from the cubic susceptibility tensor of Ref. 34, if depolarizing collisions are ignored in the latter, and if the terms with the specified triple resonance are retained.

Since some small corrections were omitted in the calculation of (6), the tensor (7) does not have the full interchange symmetry of the general cubic susceptibility tensor (10). It merely satisfies the relation

$$\chi_{ijkl}(\omega_n+\omega_m-\omega_p;\ \omega_n,\omega_m,-\omega_p)$$
$$=\chi_{ikjl}(\omega_m+\omega_n-\omega_p;\ \omega_m,\omega_n,\omega_p). \tag{12}$$

In the cubic susceptibility tensors (7) and (10)–(12), we have retained only the frequency arguments; for brevity, the other arguments ($\mathbf{k}_n$, $\mathbf{k}_m$, and $\mathbf{k}_p$) have been omitted. The dependence of the tensors (7) and (10)–(12) on the wave vectors results from the Doppler effect, as in the case of the linear dielectric susceptibility (5).

The terms in (6) which have identical indices $n = m = p$ describe the nonlinear effect of an incident wave. This effect was studied in Ref. 35 for atoms with a zero nuclear spin $(I = 0)$, for the case with depolarizing elastic collisions. To find a physical interpretation of the other terms in the sum in (6), we write this sum out explicitly:

$$\mathbf{P}^{NL}(\mathbf{r},t)=\sum_{nmp}\frac{1}{2}[\overset{(1)}{\chi}_{nmp}\mathbf{a}_n(\mathbf{a}_m\mathbf{a}_p^*)+\overset{(2)}{\chi}_{nmp}\mathbf{a}_m(\mathbf{a}_p^*\mathbf{a}_n)$$
$$+\overset{(3)}{\chi}_{nmp}\mathbf{a}_p^*(\mathbf{a}_n\mathbf{a}_m)]\exp\{i[(\mathbf{k}_n+\mathbf{k}_m-\mathbf{k}_p)\mathbf{r}$$
$$-(\omega_n+\omega_m-\omega_p)t]\}+\text{c.c.} \tag{13}$$

It follows that the nonlinear effect of two waves on the passage of a third reduces to the induction of diffraction gratings in the gas. The third wave passes through these gratings and is scattered (Refs. 16 and 36, for example). With $n = p$ or $m = p$, each of the incident waves undergoes independent diffractive scattering by the diffraction grating induced by the same two waves. This self-diffraction of light waves occurs both during copropagation of these waves and during counterpropagation, accompanied by phase conjugation. Consequently, the cubic susceptibility tensor in (7) describes the self-effects, self-diffraction, and phase conjugation of light waves in the cases of both degenerate $\omega_n = \omega_m = \omega_p$ and nondegenerate $\omega_n \neq \omega_m \neq \omega_p$ ($\omega_n = \omega_m \neq \omega_p$) four-wave mixing in a gas containing active atoms with a nonzero nuclear spin.

If the Doppler width $k_n u$ is small in comparison with the distance between hyperfine sublevels in the upper and lower levels, and if the frequencies ω_n with $n = 1, 2, \ldots$ of the light waves in (1) are close to the frequency $\omega_{F_bF_a}$ of the transition between two hyperfine sublevels, i.e., if

$$k_n u,\ |\omega_n - \omega_{F_bF_a}|,\ \gamma_{ba} \ll \min(|\Delta_{F_a} - \Delta_{F_a\pm1}|,\ |\Delta_{F_b} - \Delta_{F_b\pm1}|), \tag{14}$$

then only the two resonant hyperfine sublevels E_{F_a} and E_{F_b} participate in forming the nonlinear phenomena. For these sublevels, our basic expression, (8), becomes

$$Q^{(\varkappa)}_{nmp} = N_{ab}|d_{F_bF_a}|^4\hbar^{-3}(2I+1)^{-1}\int L_{F_bF_a}(\omega_n+\omega_m-\omega_p) \times[L^*_{F_bF_a}(\omega_p) - L_{F_bF_a}(\omega_n)]C^{(\varkappa)}_{F_aF_b}(\omega_n-\omega_p)f(v)d\mathbf{v}, \tag{15}$$

where

$$C^{(\varkappa)}_{F_aF_b}(\omega_n-\omega_p) = L_a(\omega_n-\omega_p)\begin{Bmatrix} 1 & \varkappa & 1 \\ F_a & F_b & F_a \end{Bmatrix}^2 + L_b(\omega_n-\omega_p)\begin{Bmatrix} 1 & \varkappa & 1 \\ F_b & F_a & F_b \end{Bmatrix}^2 + \frac{i\gamma(2J_b+1)|d_{F_bF_a}|^2}{|d_{ba}|^2}L_a(\omega_n-\omega_p)L_b(\omega_n-\omega_p) \times(-1)^{F_a+F_b}\begin{Bmatrix} 1 & \varkappa & 1 \\ F_a & F_b & F_a \end{Bmatrix}\begin{Bmatrix} 1 & \varkappa & 1 \\ F_b & F_a & F_b \end{Bmatrix}\begin{Bmatrix} F_a & \varkappa & F_a \\ F_b & 1 & F_b \end{Bmatrix},$$

$$L_a(\omega_n-\omega_p) = [\omega_n-\omega_p-(\mathbf{k}_n-\mathbf{k}_p)\mathbf{v}+i\gamma_a]^{-1},$$
$$L_b(\omega_n-\omega_p) = [\omega_n-\omega_p-(\mathbf{k}_n-\mathbf{k}_p)\mathbf{v}+i\gamma_b]^{-1}.$$

The dependence of $Q^{(\varkappa)}_{nmp}$ on the type of transition $J_aF_a \to J_bF_b$ between hyperfine sublevels is of fundamental importance, since for certain values of the total angular momenta F_a and F_b, corresponding to the selection rules, this dependence leads to the equality $\chi^{(3)}_{nmp} = 0$, for all possible values of J_a and J_b, as can be seen from the formula

$$\chi^{(3)}_{nmp} = {}^1/_2[(Q_{nmp}+Q_{mnp})(D^{(2)}-D^{(1)}) + \gamma(2J_b+1)|d_{F_bF_a}|^2(\gamma_a+\gamma_b)^{-1}|d_{ba}|^{-2}\cdot \times(\tilde{Q}_{nmp}+\tilde{Q}_{mnp})(\tilde{D}^{(2)}-\tilde{D}^{(1)})], \tag{16}$$

where

$$Q_{nmp} = N_{ab}|d_{F_bF_a}|^4\hbar^{-3}(2I+1)^{-1}\int L_{F_bF_a}(\omega_n+\omega_m-\omega_p) \times[L^*_{F_bF_a}(\omega_p) - L_{F_bF_a}(\omega_n)] \times[L_a(\omega_n-\omega_p)+L_b(\omega_n-\omega_p)]f(v)d\mathbf{v},$$

$$\tilde{Q}_{nmp} = iN_{ab}|d_{F_bF_a}|^4\hbar^{-3}(2I+1)^{-1}(\gamma_a+\gamma_b)\int L_{F_bF_a}(\omega_n+\omega_m-\omega_p) \times[L^*_{F_bF_a}(\omega_p) - L_{F_bF_a}(\omega_n)]L_a(\omega_n-\omega_p)L_b(\omega_n-\omega_p)f(v)d\mathbf{v},$$

$$D^{(\varkappa)} = \begin{Bmatrix} 1 & \varkappa & 1 \\ F_a & F_b & F_a \end{Bmatrix}^2,$$

$$\tilde{D}^{(\varkappa)} = (-1)^{F_a+F_b}\begin{Bmatrix} 1 & \varkappa & 1 \\ F_a & F_b & F_a \end{Bmatrix}\begin{Bmatrix} 1 & \varkappa & 1 \\ F_b & F_a & F_b \end{Bmatrix}\begin{Bmatrix} F_a & \varkappa & F_a \\ F_b & 1 & F_b \end{Bmatrix},$$

$\varkappa = 0, 1, 2.$

For the transitions $F_a = F \to F_b = F + 1$ and $F_a = F + 1 \to F_b = F$ we find

$$D^{(2)} - D^{(1)} = -4F(F+2)/15(F+1)(2F+1)(2F+3),$$
$$\tilde{D}^{(2)} - \tilde{D}^{(1)} = 2F(F+2)(4F^2+8F+5)/15(F+1)^2 \times(2F+1)^2(2F+3)^2.$$

For the transitions $F_a = F \to F_b = F$ we find

$$D^{(2)} - D^{(1)} = 2(F-1)(F+2)/15F(F+1)(2F+1),$$
$$\tilde{D}^{(2)} - \tilde{D}^{(1)} = -(F-1)(F+2)(2F^2+2F-1)/ 15F^2(F+1)^2(2F+1)^2.$$

For transitions $J_aF_a \to J_bF_b$ between hyperfine sublevels which involve a change in the total angular momentum, $F_a \to F_b$ (transitions of the types 1 → 0, 0 → 1, and 1 → 1), we thus have $\chi^{(3)}_{nmp} = 0$. In such cases we can, according to (13), choose the polarizations of the incident waves so that no diffraction gratings are induced, and no self-diffraction or phase conjugation occurs. Moreover, no nonlinear effects of the incident waves occur on these transitions. Transitions $J_aF_a \to J_bF_b$ which involve a change in the total angular momentum 1 → 0, 0 → 1, and 1 → 1 can occur for all electron angular momenta J_a and J_b allowed by the selection rules $J_a - J_b = 0, \pm 1$.

For atoms with zero nuclear spin, it was found in Refs. 37 and 27–30 that self-diffraction and phase conjugation do not occur for certain polarizations of the incident waves in the case of transitions $J_a \to J_b$ involving low electron angular momenta, 1 → 0, 0 → 1, and 1 → 1. The special role played by transitions accompanied by changes 1 → 0, 0 → 1, and 1 → 1 in the electron angular momentum, in the absence of hyperfine structure, has been pointed out in studies of the intensity and polarization of the output of gas lasers[38,39] and of nonlinear effects of light waves.[35] In this connection, expression (16) indicates that transitions $J_aF_a \to J_bF_b$ involving the changes 1 → 0, 0 → 1, and 1 → 1 in the total angular momentum play a special role for all possible values of J_a and J_b in the case of atoms which do have hyperfine structure.

If the Doppler width $k_n u$ is considerably smaller than the distance between hyperfine sublevels of the lower level

E_a, but greater than that of the upper level E_b, then two or more hyperfine sublevels of the upper level E_b will contribute to the formation of the nonlinear phenomenon, despite the strict resonance $\omega_n = \omega_{F_bF_a}$. The effect will be to greatly complicate a quantitative calculation of all the nonlinear effects described by (7) and (8). However, when the distances between the hyperfine sublevels in the lower level are much greater than those in the upper level, one can realize some special atomic-excitation regimes in which the frequencies ω_n $(n = 1,2,...,n_0)$ of the incident waves [see (1)] are in resonance with the frequency of the transition $J_a = J \rightarrow J_b = J + 1$ between the hyperfine sublevel E_{F_a} of the lower level with the smallest total angular momentum, $F_a = |J - I|$, and the upper level E_b:

$$k_n u,\ |\omega_n - \omega_{ba} + \Delta_{Fa}|,\ \gamma_{ba} \ll |\Delta_{Fa} - \Delta_{Fa\pm 1}|,\ \ |\Delta_{Fb}| \ll |\Delta_{Fa}|. \tag{17}$$

In this case, only one hyperfine sublevel E_{F_b} of the upper level—that with the smallest total angular momentum, $F_b = |J + 1 - I|$—participates in the formation of the nonlinear phenomena, because of the selection rules. Correspondingly, for the transition $J_a = J + 1 \rightarrow J_b = J$ and for the sublevel E_{F_a} with the maximum angular momentum, $F_a = J + 1 + I$, only the one sublevel E_{F_b} of the upper level which has the largest total angular momentum, $F_b = J + I$, participates in excitation regime (17), because of the selection rules. Consequently, for these minimum or maximum total angular momenta F_a and F_b, only the two sublevels E_{F_a} and E_{F_b} are involved in excitation regime (17). Equations (15) and (16) thus remain valid, regardless of the magnitude of the Doppler broadening in comparison with the distance between the hyperfine sublevels of the upper level.

We wish to stress that far from all three resonances at the hyperfine sublevels,

$$|\omega_n - \omega_{ba}| \gg |\Delta_{Fa} - \Delta_{Fb}|,\ \ |\omega_n - \omega_p| \gg |\Delta_{Fa}|,\ |\Delta_{Fb}|,$$

we can make the replacements $\omega_{F_bF_a} \rightarrow \omega_{ba}$, $\omega_{F_aF'_a} \rightarrow 0$, and $\omega_{F_bF'_b} \rightarrow 0$ in (8). We can then sum over all the hyperfine sublevels of the upper and lower levels, regardless of the nuclear spin I. As a result of this summation, expression (8) becomes

$$Q_{nmp}^{(\varkappa)} = N_{ab} |d_{ba}|^4 \hbar^{-3} \int L_{ba}(\omega_n + \omega_m - \omega_p)$$
$$\times [L_{ba}^*(\omega_p) - L_{ba}(\omega_n)] C_{ab}^{(\varkappa)}(\omega_n - \omega_p) f(v)\, d\mathbf{v}, \tag{18}$$

where

$$C_{ab}^{(\varkappa)}(\omega_n - \omega_p) = L_a(\omega_n - \omega_p) \begin{Bmatrix} 1 & \varkappa & 1 \\ J_a & J_b & J_a \end{Bmatrix}^2$$

$$+ L_b(\omega_n - \omega_p) \begin{Bmatrix} 1 & \varkappa & 1 \\ J_b & J_a & J_b \end{Bmatrix}^2$$

$$+ i\gamma (2J_b + 1) L_a(\omega_n - \omega_p) L_b(\omega_n - \omega_p) (-1)^{J_a + J_b} \begin{Bmatrix} 1 & \varkappa & 1 \\ J_a & J_b & J_a \end{Bmatrix}$$

$$\times \begin{Bmatrix} 1 & \varkappa & 1 \\ J_b & J_a & J_b \end{Bmatrix} \begin{Bmatrix} J_a & \varkappa & J_a \\ J_b & 1 & J_b \end{Bmatrix},$$

$$L_{ba}(\omega_n + \omega_m - \omega_p)$$
$$= [\omega_n + \omega_m - \omega_p - (\mathbf{k}_n + \mathbf{k}_m - \mathbf{k}_p)\mathbf{v} - \omega_{ba} + i\gamma_{ba}]^{-1}.$$

The tensor (7), combined with (18), describes the nonlinear phenomena without consideration of the nuclear spin I for transitions which are homogeneously and inhomogeneously broadened. This assertion does not apply to the nonlinear phenomena of degenerate four-wave mixing and nonlinear effects of the light waves, in which cases the inequalities $\max(|\omega_n - \omega_p|, |\mathbf{k}_n - \mathbf{k}_p| u) \gg |\Delta_{F_a}|, |\Delta_{F_b}|$ do not hold. In this case, no two-level model of the atom which ignores the hyperfine structure of the resonant levels is valid.

2. APPLICATION TO ATOMS WITH A ZERO NUCLEAR SPIN

Assuming the nuclear spin to be zero in our basic equation (8), we obtain (18). The cubic susceptibility tensor (7), with (18), therefore describes all the nonlinear phenomena which occur within the one-photon absorption line in the important case of atoms which lack a hyperfine structure, for the very simple model of irreversible relaxation, in (2)–(4), which uses the three constants γ_{ba}, γ_a, and γ_b.

To circumvent some mathematical difficulties in Eqs. (2)–(4), we omit the terms which stem from depolarizing elastic collisions. A generally accepted model for such collisions is described in Refs. 40 and 41. For atoms with a zero nuclear spin, however, we can retain the contribution of depolarizing elastic collisions in (2)–(4). As a result, we find expressions (7) and (18), with the substitutions

$$L_{ba}(\omega_n + \omega_m - \omega_p) \rightarrow L_{ba}^{(1)}(\omega_n + \omega_m - \omega_p) = [\omega_n + \omega_m - \omega_p$$
$$- (\mathbf{k}_n + \mathbf{k}_m - \mathbf{k}_p)\mathbf{v} - \omega_{ba} - \Delta_{ba}^{(1)} + i\gamma_{ba}^{(1)}]^{-1},$$
$$L_a(\omega_n - \omega_p) \rightarrow L_a^{(\varkappa)}(\omega_n - \omega_p) = [\omega_n - \omega_p - (\mathbf{k}_n - \mathbf{k}_p)\mathbf{v} + i\gamma_a^{(\varkappa)}]^{-1},$$
$$L_b(\omega_n - \omega_p) \rightarrow L_b^{(\varkappa)}(\omega_n - \omega_p) = [\omega_n - \omega_p - (\mathbf{k}_n - \mathbf{k}_p)\mathbf{v} + i\gamma_b^{(\varkappa)}]^{-1}. \tag{19}$$

Here $\gamma_{ba}^{(1)}$ and $\Delta_{ba}^{(1)}$ are the half-width and collisional shift of the spectral line, and $\gamma_a^{(\varkappa)}$ $(\gamma_b^{(\varkappa)})$ are the relaxation constants for the population $(\varkappa = 0)$, the orientation $(\varkappa = 1)$, and the alignment $(\varkappa = 2)$ of an atom in level E_a (E_b).

The cubic susceptibility tensor in (7), along with (18) and (19), generalizes the results of Ref. 30 to the case of nondegenerate four-wave mixing in a gas of atoms with a hyperfine structure. With $\mathbf{k}_n = \mathbf{k}_m = \mathbf{k}_p = \mathbf{k}$, this tensor becomes the tensor which was found in Ref. 35 to characterize nonlinear optical activity, if we symmetrize the latter tensor with respect to the indices j and l, and if we also introduce a factor of 3, in accordance with the definition of the cubic susceptibility tensor adopted here, (9). Moreover, without the term containing γ, the tensor in (7), with (18) and (19), can be found from the general expression for the cubic susceptibility tensor of atoms with a zero nuclear spin from Ref. 42, if in the latter expression we retain the terms with a triple resonance. Of the three resonances, one is at zero frequency, while the two others are at frequencies from the set ω_n, ω_m, and ω_p.

When depolarizing elastic collisions occur, the characteristic parameter $\chi_{nmp}^{(3)}$ of cubic susceptibility tensor (7), for atoms with a zero nuclear spin, is given by the following expression in the case of both degenerate and nondegenerate four-wave mixing:

$$\chi_{nmp}^{(3)}={}^1/_2(U_{nmp}+U_{mnp}), \tag{20}$$

where

$$U_{nmp}=N_{ab}|d_{ba}|^4\hbar^{-3}\int L_{ba}^{(1)}(\omega_n+\omega_m-\omega_p)$$

$$\times[L_{ba}^{(1)*}(\omega_p)-L_{ba}^{(1)}(\omega_n)]$$

$$\times[C_{ab}^{(2)}(\omega_n-\omega_p)-C_{ab}^{(1)}(\omega_n-\omega_p)]f(v)d\mathbf{v}.$$

For $C_{ab}^{(\varkappa)}(\omega_n-\omega_p)$ with $\varkappa=1$ and 2, we have made the substitutions (19). The dependence on the type of transition $J_a\rightarrow J_b$ is incorporated in the difference $C_{ab}^{(2)}(\omega_n-\omega_p)-C_{ab}^{(1)}(\omega_n-\omega_p)$. For transitions with small electron angular momenta, and for $J_a=0\rightarrow J_b=1$, this difference is

$$C_{ab}^{(2)}(\omega_n-\omega_p)-C_{ab}^{(1)}(\omega_n-\omega_p)$$

$$=(i/9)(\gamma_b^{(1)}-\gamma_b^{(2)})L_b^{(1)}(\omega_n-\omega_p)L_b^{(2)}(\omega_n-\omega_p)$$

for $J_a=1\rightarrow J_b=0$,

$$C_{ab}^{(2)}(\omega_n-\omega_p)-C_{ab}^{(1)}(\omega_n-\omega_p)$$

$$=(i/9)(\gamma_a^{(1)}-\gamma_a^{(2)})L_a^{(1)}(\omega_n-\omega_p)L_a^{(2)}(\omega_n-\omega_p)$$

for $J_a=1\rightarrow J_b=1$, and

$$C_{ab}^{(2)}(\omega_n-\omega_p)-C_{ab}^{(1)}(\omega_n-\omega_p)$$

$$=(i/36)[(\gamma_a^{(1)}-\gamma_a^{(2)})L_a^{(1)}(\omega_n-\omega_p)$$

$$\times L_a^{(2)}(\omega_n-\omega_p)+(\gamma_b^{(1)}-\gamma_b^{(2)})L_b^{(1)}(\omega_n-\omega_p)L_b^{(2)}(\omega_n-\omega_p)$$

$$+{}^1/_2\gamma(L_a^{(2)}(\omega_n-\omega_p)L_b^{(1)}(\omega_n-\omega_p)$$

$$-L_a^{(1)}(\omega_n-\omega_p)L_b^{(1)}(\omega_n-\omega_p))].$$

We see that, for transitions $J_a\rightarrow J_b$ involving a change in electron angular momentum 1 → 0, 0 → 1, or 1 → 1, the quantity (20) is nonzero only by virtue of depolarizing elastic collisions, which lead to the inequality $\gamma_a^{(1)}\neq\gamma_a^{(2)}$ $(\gamma_b^{(1)}\neq\gamma_b^{(2)})$. In such transitions (for certain polarizations of the incident waves), the diffraction gratings, the self-diffraction, and the phase conjugation are collision-induced phenomena, in accordance with the assertions in Refs. 37 and 28–30 for atoms with a zero nuclear spin. In this case the nonlinear effects of the light waves are also collision-induced.[35] In other transitions $J_a\rightarrow J_b$, there is no such strong dependence on $\gamma_a^{(1)}-\gamma_a^{(2)}$ $(\gamma_b^{(1)}-\gamma_b^{(2)})$. For $J_a=\frac{1}{2}\rightarrow J_b=\frac{1}{2}$, for example, we have

$$C_{ab}^{(2)}(\omega_n-\omega_p)-C_{ab}^{(1)}(\omega_n-\omega_p)$$

$$=-{}^1/_9[L_a^{(1)}(\omega_n-\omega_p)+L_b^{(1)}(\omega_n-\omega_p)$$

$$-{}^1/_3i\gamma L_a^{(1)}(\omega_n-\omega_p)L_b^{(1)}(\omega_n-\omega_p)].$$

3. INTERFERENCE FIELD IN THE CASE OF A ONE-PHOTON RESONANCE

The propagation of the light waves in (1) in a given resonant medium is described by means of Maxwell's equations

$$\operatorname{rot}\operatorname{rot}\mathbf{E}+\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\hat{\varepsilon}_0\mathbf{E}=-\frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\mathrm{P},\qquad \operatorname{div}(\hat{\varepsilon}_0\mathbf{E}+4\pi\mathbf{P})=0. \tag{21}$$

$$\hat{\varepsilon}_0\exp(-i\omega_nt)=\varepsilon_0(\omega_n)\exp(-i\omega_nt), \tag{22}$$

where $\varepsilon_0(\omega_n)$ is the dielectric constant, which corresponds to nonresonant levels of the active atoms and also of the impurity atoms of the buffer gas. The vector **P** incorporates the nonlinear effects of the incident waves and the excitation of new waves. The effect of the operator $\hat{\varepsilon}_0$ is defined in (22).

If the angles between the propagation directions of the incident waves are small, the interference field $\mathbf{E}=\mathbf{E}(\mathbf{r},t)$, represented previously as in (1), can conveniently be broken up into two parts. The first part, $\mathbf{E}^{(1)}(\mathbf{r},t)$, contains the same modes as at the entrance to the gas. The second part, $\mathbf{E}^{(2)}(\mathbf{r},t)$, includes new waves, which are a consequence of the interaction of the incident waves with the active atoms:

$$\mathbf{E}(\mathbf{r},t)=\mathbf{E}^{(1)}(\mathbf{r},t)+\mathbf{E}^{(2)}(\mathbf{r},t), \tag{23}$$

$$\mathbf{E}^{(1)}(\mathbf{r},t)=\sum_{n=1}^{n_0}\mathbf{b}_n\exp[i(\mathbf{k}_n\mathbf{r}-\omega_nt)]+\text{c.c.}, \tag{24}$$

$$\mathbf{E}^{(2)}(\mathbf{r},t)=\sum_f\mathbf{b}_f\exp[i(\mathbf{s}_f\mathbf{r}-\Omega_ft)]+\text{c.c.}, \tag{25}$$

where $\Omega_f=\omega_n+\omega_m-\omega_p$, and the subscript f takes on certain integer values, at which the subscript n, m, and p independently take on all possible values from the set of numbers $1,\ldots,n_0$, except in the case $m=p$ for arbitrary n or the case $n=p$ for arbitrary m, which are contained in (24). The amplitudes of the waves and the relationship between the frequencies and the wave vectors in (24) and (25) can be found by solving Maxwell's equations (21).

To simplify the equations, we choose the relationship between ω_n and $\mathbf{k}_n$ for the waves in (24) to be $\omega_n^2\varepsilon'(\omega_n,\mathbf{k}_n)=k_n^2c^2$. We also introduce some notation for the absorption coefficient: $\alpha_n=\omega^2\varepsilon''(\omega_n,\mathbf{k}_n)/2k_nc^2$. Here $\varepsilon'(\omega_n,\mathbf{k}_n)$ and $\varepsilon''(\omega_n,\mathbf{k}_n)$ are the real and imaginary parts of the dielectric constant:

$$\varepsilon(\omega_n,\mathbf{k}_n)=\varepsilon_0(\omega_n)+4\pi\varkappa(\omega_n,\mathbf{k}_n)=\varepsilon'(\omega_n,\mathbf{k}_n)+i\varepsilon''(\omega_n,\mathbf{k}_n),$$

where the resonant term $4\pi\varkappa(\omega_n,\mathbf{k}_n)$ is given in (5). We are not detracting from the generality of this discussion by taking this approach, since in this case the nonlinear corrections to the wave amplitude and phase are included in the complex amplitude $\mathbf{b}_n=\mathbf{b}_n(\mathbf{r})$. In addition, in solving Maxwell's equations, (21), we use the approximation of slowly varying amplitudes. Under this approximation we can ignore the second derivatives of these amplitudes with respect to the coordinates. From Maxwell's equations, (21), we then find a system of coupled equations for the amplitudes of the waves in (24), which are propagating in directions separated by a small angle:

$$\left(\frac{\mathbf{k}_n\nabla}{k_n}+\alpha_n\right)b_{ni}=\frac{6i\pi\omega_n^2}{k_nc^2}\Big[\chi_{ijkl}(\omega_n;\,\omega_n,\,\omega_n,\,-\omega_n)\,b_{nj}b_{nk}b_{nl}^*$$

$$+2\sum_{m=1}^{n_0}{}'\chi_{ijkl}(\omega_n;\,\omega_n,\,\omega_m,\,-\omega_m)\,b_{nj}b_{mk}b_{ml}^*\Big]. \tag{26}$$

Here $n = 1, 2,\ldots, n_0$. The prime on the summation sign means that there is no term with $m = n$. The first term in square brackets in (26) describes the self-effect of an individual incident wave. The other terms describe the nonlinear interaction and the exchange of energy between this incident wave and the other incident waves. The quantity $\mathbf{E}^{(1)}(\mathbf{r},t)$ thus describes the propagation of the incident waves, and all nonlinear corrections to these waves are taken into account.

In particular, when one wave, with a frequency ω_1 and a wave vector $\mathbf{k}_1$, is incident on the gas, Eqs. (26) become the single nonlinear equation

$$\left(\frac{\mathbf{k}_1\nabla}{k_1}+\alpha_1\right)\mathbf{b}_1=\frac{i\pi\omega_1^2}{k_1c^2}\Big\{\Big[Q_{111}^{(1)}+Q_{111}^{(2)}+\frac{2}{3}(Q_{111}^{(0)}-Q_{111}^{(2)})\Big]$$

$$\times|\mathbf{b}_1|^2\mathbf{b}_1+(Q_{111}^{(2)}-Q_{111}^{(1)})\mathbf{b}_1^2\mathbf{b}_1^*\Big\}, \tag{27}$$

where the quantities $Q_{111}^{(\varkappa)}$ with $\varkappa = 0$, 1, and 2 are given by (8), (15), and (18), depending on the conditions of the problem.

It follows from (27) that the hyperfine level structure has an important effect on the amplitude and nonlinear polarization effects of the light wave; an analysis of those effects, however, is rather complicated. In the particular case in which ω_1 is in resonance with the frequency $\omega_{F_bF_a}$ of a transition between two hyperfine sublevels, (14) or (17), we can apply the results of Ref. 35. Those results were derived for atoms with a zero nuclear spin. In order to make use of those results, we need to ignore depolarizing elastic collisions and make the substitutions

$$N_{ab}\to N_{ab}(2I+1)^{-1},\quad d_{ba}\to d_{F_bF_a},\quad \gamma\to\gamma|d_{F_bF_a}/d_{ba}|^2,$$

$$\omega_{ba}\to\omega_{F_bF_a},\quad \Delta_{ba}^{(1)}\to 0,\quad \gamma_{ba}^{(1)}\to\gamma_{ba},\quad \gamma_a^{(\varkappa)}\ (\gamma_b^{(\varkappa)})\to\gamma_a(\gamma_b),$$

$$J_a\to F_a,\quad J_b\to F_b,\quad C_\varkappa\to iC_{F_aF_b}^{(\varkappa)},\quad \varkappa=0,1,2,$$

thereby incorporating the presence of a nuclear spin.

To determine the amplitudes $\mathbf{b}_f$ and the wave vectors $\mathbf{s}_f$ of the new waves in (25), we solve Maxwell's equations (21) by perturbation theory, assuming that we can ignore the nonlinear depletion (or intensification) of the incident waves. In other words, we assume that incident waves with frequencies ω_n, wave vectors $\mathbf{k}_n$, and complex amplitudes $\mathbf{a}_n$ with $n = 1, \ldots, n_0$ are propagating linearly through the gas, with a dispersion law $\omega_n^2\varepsilon'(\omega_n,\mathbf{k}_n) = k_n^2c^2$ and an absorption coefficient $\alpha_n = \omega_n^2\varepsilon''(\omega_n,\mathbf{k}_n)/2k_nc^2$. We also assume that the angle between the propagation directions of the incident waves is on the order of a milliradian, so we can ignore the term with $\mathbf{s}_f\mathbf{P}^{NL}$. From Eqs. (21) we then find the following equation for the unknown quantities:

$$\left(\frac{\mathbf{s}_f\nabla}{s_f}+\alpha_f\right)b_{fi}=\frac{12i\pi\Omega_f^2}{s_fc^2}\chi_{ijkl}(\Omega_f;\,\omega_n,\,\omega_m,\,-\omega_p)$$

$$\times a_{nj}a_{mk}a_{pl}^*\exp[i(\mathbf{k}_n+\mathbf{k}_m-\mathbf{k}_p-\mathbf{s}_f)\mathbf{r}], \tag{28}$$

where

$$\Omega_f^2\varepsilon'(\Omega_f,\,\mathbf{s}_f)=s_f^2c^2,\quad \alpha_f=\Omega_f^2\varepsilon''(\Omega_f,\,\mathbf{s}_f)/2s_jc^2,$$

and where each value of the index f corresponds to a specific set of values of n, m, and p.

Equation (28) describes the excitation of new waves as the result of a four-wave mixing of the incident waves with the active atoms within the one-photon absorption line. The projection b_{fi} of the amplitude $\mathbf{b}_f$ of a new wave onto the Cartesian axis i at the point $\mathbf{r} = \mathbf{s}_fL_f/s_f$ at the exit from the gas becomes

$$b_{fi}=6G(\Omega_f,\,\mathbf{s}_f)\,a_n(0)\,a_m(0)\,a_p^*(0)$$

$$\times\chi_{ijkl}(\Omega_f;\,\omega_n,\,\omega_m,\,-\omega_p)\,l_{nj}l_{mk}l_{pl}^*, \tag{29}$$

where

$$G(\Omega_f,\mathbf{s}_f)=\frac{2i\pi\Omega_f^2\{\exp[-(q_f+i\Delta_f)L_f]-\exp(-\alpha_fL_f)\}}{s_fc^2(\alpha_f-q_f-i\Delta_f)},$$

$$q_f=(\alpha_n\mathbf{k}_n/k_n+\alpha_m\mathbf{k}_m/k_m+\alpha_p\mathbf{k}_p/k_p)\mathbf{s}_f/s_f,$$
$$\Delta_f=(\mathbf{s}_f-\mathbf{K}_f)\mathbf{s}_f/s_f,\quad \mathbf{K}_f=\mathbf{k}_n+\mathbf{k}_m-\mathbf{k}_p,$$
$$\mathbf{a}_n(0)=a_n(0)\mathbf{l}_n,\quad \mathbf{l}_n\mathbf{l}_n^*=1.$$

Here $a_n(0)$ is the amplitude of the incident wave at the boundary point $\mathbf{r} = 0$ at the entrance to the gas, $\mathbf{l}_n$ is the complex unit polarization vector of the incident wave, and L_f is the optical path length, which is the absolute value of the vector $\mathbf{L}_f = \mathbf{s}_fL_f/s_f$. The vector $\mathbf{l}_n$ does not vary inside the gas; it retains the value with which it entered the gas. The quantity q_f contains the absorption coefficients only for the incident waves. By virtue of the factor $G(\Omega_f,\mathbf{s}_f)$, the intensity of the new wave in (29) reaches a maximum when this wave is propagating in the direction corresponding to the phase-matching condition $\mathbf{s}_f = \mathbf{K}_f$.

The new waves in (29) may be thought of as the result of Raman scattering of the light or as the result of diffractive scattering by the induced gratings described by the terms in sum (13). In the latter case, the new waves in (29) represent the waves of the first order of diffraction.

4. SELF-DIFFRACTION OF NEARLY PARALLEL INCIDENT WAVES

We assume that two light waves as in (1), with $n_0 = 2$, are incident on a gas. The interference field in (23) then contains four waves. Of them, the two with the frequencies ω_1 and ω_2, the wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$, and the complex amplitudes $\mathbf{b}_1$ and $\mathbf{b}_2$ describe the propagation of the incident waves, (24), in the active zone of the gas. The nonlinear effects of the waves and the interaction between them are taken into account. Two other waves described by (25) with $f = 3$ and 4 are the waves of the first order of diffraction, with frequencies Ω_3 and Ω_4, wave vectors $\mathbf{s}_3$ and $\mathbf{s}_4$, and amplitudes $\mathbf{b}_3$ and $\mathbf{b}_4$. For the latter we find, using (7) and (29),

$$\mathbf{b}_3=G(\Omega_3,\mathbf{s}_3)a_1^2(0)a_2^*(0)[(^2/_3Q_{112}^{(0)}+Q_{112}^{(1)}+^1/_3Q_{112}^{(2)})\mathbf{l}_1(\mathbf{l}_1\mathbf{l}_2^*)+(Q_{112}^{(2)}-Q_{112}^{(1)})\mathbf{l}_2^*\mathbf{l}_1^2], \tag{30}$$

$$\mathbf{b}_4=G(\Omega_4,\mathbf{s}_4)a_1^*(0)a_2^2(0)[(^2/_3Q_{221}^{(0)}+Q_{221}^{(1)}+^1/_3Q_{221}^{(2)})\mathbf{l}_2(\mathbf{l}_2\mathbf{l}_1^*)+(Q_{221}^{(2)}-Q_{221}^{(1)})\mathbf{l}_1^*\mathbf{l}_2^2], \tag{31}$$

where

$$\Omega_3=2\omega_1-\omega_2,\quad \Omega_4=2\omega_2-\omega_1,\quad \mathbf{b}_3=b_3\mathbf{l}_3,\quad \mathbf{b}_4=b_4\mathbf{l}_4.$$

Here $\mathbf{l}_3$ and $\mathbf{l}_4$ are complex unit polarization vectors of the diffracted waves. The quantities which appear in $G(\Omega_3,\mathbf{s}_3)$ and $G(\Omega_4,\mathbf{s}_4)$ are

$$q_3=(2\alpha_1\mathbf{k}_1/k_1+\alpha_2\mathbf{k}_2/k_2)\mathbf{s}_3/s_3,$$
$$q_4=(\alpha_1\mathbf{k}_1/k_1+2\alpha_2\mathbf{k}_2/k_2)\mathbf{s}_4/s_4,$$
$$\mathbf{K}_3=2\mathbf{k}_1-\mathbf{k}_2,\quad \mathbf{K}_4=2\mathbf{k}_2-\mathbf{k}_1.$$

The waves of the first order of diffraction in a gas have been studied in many places (e.g., Refs. 14–18), where they have been used primarily for optical image transmission and optical signal processing. The effect of the hyperfine level structure on the properties of these waves either has not been studied in detail or has been ignored altogether. We will accordingly take a look at certain effects in which the influence of the hyperfine structure can be seen particularly clearly.

If the first incident wave is polarized linearly and the second circularly, the diffracted wave with amplitude (30) is elliptically polarized, while the other wave, with amplitude (31), is circularly polarized, with the same rotation direction for the electric vector as in the incident circularly polarized wave. If, on the other hand, the second incident wave is linearly polarized, and the first circularly, the diffracted wave with amplitude (31) is elliptically polarized, while the polarization of the wave (30) is the same as that of the first incident wave. The ratio of the axes of the polarization ellipse and the direction in which the electric vector rotates are strong functions of the hyperfine structure of the resonant levels in this case.

We direct the z axis along the vector $\mathbf{k}_2$, and we assume that the first incident wave is linearly polarized ($\mathbf{l}_1=\mathbf{l}_x$). We assume that the second wave is circularly polarized with a right-hand polarization: $\mathbf{l}_2=(\mathbf{l}_x+i\mathbf{l}_y)/2^{1/2}$, where $\mathbf{l}_x$ and $\mathbf{l}_y$ are unit vectors along the Cartesian axes x and y. Under the inequality (14) or (17), along with the conditions

$$|\omega_1-\omega_2|,\quad |\mathbf{k}_1-\mathbf{k}_2|u\ll\gamma_a,\ \gamma_b \tag{32}$$

the ratio of the projections (b_{3y} and b_{3x}) of amplitude (30) onto the Cartesian axes y and x is, for both homogeneously and inhomogeneously broadened transitions,

$$b_{3y}/b_{3x}=-iC_0, \tag{33}$$

where

$$C_0=3(C^{(2)}-C^{(1)})/2(C^{(0)}+2C^{(2)}),$$
$$C^{(\varkappa)}=D^{(\varkappa)}+\Gamma(F_a,J_a;F_b,J_b)\tilde{D}^{(\varkappa)},\quad \varkappa=0,\ 1,\ 2.$$

The coefficient Γ is

$$\Gamma(F_a,J_a;F_b,J_b)=\frac{\gamma}{\gamma_a+\gamma_b}(2F_a+1)(2F_b+1)\times(2J_b+1)\begin{Bmatrix} I & F_a & J_a\\ 1 & J_b & F_b\end{Bmatrix}^2. \tag{34}$$

For transitions $J_aF_a\to J_bF_b$ involving a change in the total angular momentum $F_a=F\to F_b=F+1$ and $F_a=F+1\to F_b=F$, for all possible changes in the electron angular momentum, $J_a=J\to J_b=J+1$, $J_a=J+1\to J_b=J$, and $J_a=J\to J_b=J$, we find

$$C_0=\frac{F(F+2)[-2(F+1)(2F+1)(2F+3)+\Gamma(F_a,J_a;F_b,J_b)(4F^2+8F+5)]}{(F+1)(2F+1)(2F+3)(4F^2+8F+5)-\Gamma(F_a,J_a;F_b,J_b)(8F^4+32F^3+42F^2+20F+5)},$$

$$\Gamma(F,J;F+1,J+1)=\frac{\gamma(F+J+I+2)(F+J+I+3)(F+J-I+1)(F+J-I+2)}{4(\gamma_a+\gamma_b)(F+1)(J+1)(2J+1)},$$

$$\Gamma(F+1,J+1;F,J)=\Gamma(F,J;F+1,J+1)(2J+1)/(2J+3),$$

$$\Gamma(F,J+1;F+1,J)=\frac{\gamma(F-J+I)(F-J+I+1)(J-F+I)(J-F+I+1)}{4(\gamma_a+\gamma_b)(F+1)(J+1)(2J+3)},$$

$$\Gamma(F+1,J;F,J+1)=\Gamma(F,J+1;F+1,J)(2J+3)/(2J+1),$$

$$\Gamma(F,J;F+1,J)=\Gamma(F+1,J;F,J)=\frac{\gamma(F+J+I+2)(F+J-I+1)(F-J+I+1)(J-F+I)}{4(\gamma_a+\gamma_b)(F+1)J(J+1)}.$$

For transitions $F_a=F\to F_b=F$, for all possible $J_a=J\to J_b=J+1$, $J_a=J+1\to J_b=J$, and $J_a=J\to J_b=J$, these quantities take a different form:

$$C_0=\frac{^1/_2(F-1)(F+2)[2F(F+1)(2F+1)-\Gamma(F,J_a;F,J_b)(2F^2+2F-1)]}{F(F+1)(2F+1)(3F^2+3F-1)-\Gamma(F,J_a;F,J_b)(3F^4+6F^3-2F^2-5F+3)},$$

$$\Gamma(F,J;F,J+1)=\frac{\gamma(2F+1)(F-J+I)(J-F+I+1)(F+J-I+1)(F+J+I+2)}{4(\gamma_a+\gamma_b)F(F+1)(J+1)(2J+1)},$$

$$\Gamma(F,J+1;F,J)=\Gamma(F,J;F,J+1)(2J+1)/(2J+3),$$
$$\Gamma(F,J;F,J)=\frac{\gamma(2F+1)[F(F+1)+J(J+1)-I(I+1)]^2}{4(\gamma_a+\gamma_b)F(F+1)J(J+1)}.$$

According to (33), the elliptically polarized wave (30) has right-hand polarization in the case $C_0<0$ and left-hand polarization in the case $C_0>0$. The ratio of the axes of the polarization ellipse is $|C_0|$. By virtue of (34), the deformation of the polarization ellipse and the rotation direction of the electric vector of the diffracted wave depend strongly on the type of transition $J_aF_a\to J_bF_b$ and also on the nuclear spin, except in the case of transitions involving a change in total angular momentum $F_a\to F_b$ of the types $0\to 1$, $1\to 0$, and $1\to 1$, in which case the polarization ellipse degenerates into a line segment along the x axis. In a dense gas, with $\gamma\ll\gamma_a+\gamma_b$, we can ignore the quantity in (34) in the expression for $C^{(\varkappa)}$ with $\varkappa=0$, 1, and 2. For transitions $F_a-F_b=\pm 1$ $(F_a+F_b>1)$ and $F_a=F_b=\frac{1}{2}$, the elliptical polarization is then right-handed, while for $F_a-F_b=0$ $(F_a>1)$ it is left-handed. In a low-density gas, with $\gamma\lesssim\gamma_a+\gamma_b$, the nature of the elliptical polarization is determined by the quantity in (34).

Assume that the incident waves are linearly polarized and that their polarization vectors $\mathbf{l}_1$ and $\mathbf{l}_2$ form an arbitrary angle ψ. The diffracted waves (30) and (31) are then also linearized polarized, and their polarization vectors $\mathbf{l}_3$ and $\mathbf{l}_4$ form angles ψ_3 and ψ_4, respectively, with the vector $\mathbf{l}_1$. The positive direction for all angles is clockwise, if we look along the vector $\mathbf{k}_1$. In this case, when inequality (14) or (17) holds, along with (32), the angles ψ, ψ_3, and ψ_4 between the polarization planes of the waves involved in the self-diffraction, are related by

$$\operatorname{tg}\psi_3=C_0\operatorname{tg}\psi, \tag{35}$$

$$\operatorname{ctg}\psi_4=\operatorname{ctg}\psi+3(C^{(2)}-C^{(1)})/(2C^{(0)}+3C^{(1)}+C^{(2)})\sin\psi\cos\psi. \tag{36}$$

For transitions $J_aF_a\to J_bF_b$ involving a change in the total angular momentum $0\to 1$, $1\to 0$, or $1\to 1$, we have $\psi_3=0$ and $\psi_4=\psi$. For other transitions, the polarization behavior is less obvious, except in the case $\gamma\ll\gamma_a+\gamma_b$, in which the angle ψ_3 is negative for $F_a-F_b=\pm 1$ $(F_a+F_b>1)$ and $F_a=F_b=\frac{1}{2}$, while it is positive for $F_a-F_b=0$ $(F_a>1)$. If $\gamma\lesssim\gamma_a+\gamma_b$, then we must use (34) in calculating ψ_3 and ψ_4.

As an example we consider a gas containing a vapor of copper atoms with a nuclear spin $I=3/2$ and a resonant transition $2P^0_{3/2}\to 2D_{5/2}$, for which we have $J_a=\frac{3}{2}$ and $J_b=\frac{5}{2}$. For simplicity we choose $\psi=\pi/4$. If we set $\gamma/(\gamma_a+\gamma_b)=0$, then the angle ψ_3 for transitions $J_aF_a\to J_bF_b$ involving a change in total angular momentum $1\to 2$ $(2\to 1)$, $2\to 3$ $(3\to 2)$, or $3\to 4$ is negative and equal to −19°26′, −23°23′, and −24°46′, respectively. For transitions which do not involve a change in the total angular momentum, $2\to 2$ and $3\to 3$, the angle ψ_3 is positive and equal to 13°14′ and 15°57′. When the parameter $\gamma/(\gamma_a+\gamma_b)$ reaches its maximum value of unity, as the total angular momentum changes $F_a=1\to F_b=2$, $F_a=2\to F_b=3$, and $F_a=3\to F_b=4$ the angle ψ_3 becomes −6°23′, −2°22′, and −45°, respectively. If there is no change in total angular momentum ($2\to 2$ and $3\to 3$), we find the angle ψ_3 to be 12°29′ and 15°42′. For $F_a=2\to F_b=1$ and $F_a=3\to F_b=2$, the angle ψ_3 takes on different values: −19°14′ and −23°20′. It thus becomes possible to experimentally distinguish the transitions $F_a=1\to F_b=2$ and $F_a=2\to F_b=3$ from $F_a=2\to F_b=1$ and $F_a=3\to F_b=2$. The strong dependence of ψ_3 on $\gamma/(\gamma_a+\gamma_b)$ shows that in a low-density gas with $\gamma\sim\gamma_a+\gamma_b$ the influx of atoms to the lower level due to the spontaneous decay of the excited state is extremely important.

Using (33), (35), and (36), we can experimentally identify transitions $J_aF_a\to J_bF_b$ when I and $\gamma/(\gamma_a+\gamma_b)$ are known. If J_a, J_b, F_a, F_b, and I are known instead, these expressions are useful for determining $\gamma/(\gamma_a+\gamma_b)$.

When three light waves of the form (1), with $n_0=3$, are incident on a gas, self-diffraction of the three waves occurs. Self-diffraction of each pair of waves of the set of three incident waves also occurs, independently. The diffracted waves which arise in the process are spatially separated, because they are propagating in different directions. Spatial separation of these waves would also be promoted by the introduction of a suitable buffer gas, to change the refractive indices and to simultaneously satisfy the dispersion law $\Omega_f^2\varepsilon'(\Omega_f,\mathbf{s}_f)=s_f^2c^2$ and the phase-matching condition $\mathbf{s}_f=\mathbf{K}_f$ for one selected diffracted wave. As a result, this wave is amplified, while the other diffracted waves are suppressed.[36] This circumstance allows us to treat the self-diffraction of two and three waves separately when a set of a large number of waves of the form (1) is incident on a gas.

The amplitudes of interference field (25) in the case of the self-diffraction of three waves is given by general formula (29). In expanded form, these amplitudes are

$$\mathbf{b}_1=G(\Omega_1,\mathbf{s}_1)a_1^*(0)a_2(0)a_3(0)[\chi^{(1)}_{231}\mathbf{l}_2(\mathbf{l}_3\mathbf{l}_1^*)$$
$$+\chi^{(2)}_{231}\mathbf{l}_3(\mathbf{l}_1^*\mathbf{l}_2)+\chi^{(3)}_{231}\mathbf{l}_1^*(\mathbf{l}_2\mathbf{l}_3)], \tag{37}$$

$$\mathbf{b}_2=G(\Omega_2,\mathbf{s}_2)a_1(0)a_2^*(0)a_3(0)[\chi^{(1)}_{312}\mathbf{l}_3(\mathbf{l}_1\mathbf{l}_2^*)$$
$$+\chi^{(2)}_{312}\mathbf{l}_1(\mathbf{l}_2^*\mathbf{l}_3)+\chi^{(3)}_{312}\mathbf{l}_2^*(\mathbf{l}_3\mathbf{l}_1)], \tag{38}$$

$$\mathbf{b}_3=G(\Omega_3,\mathbf{s}_3)a_1(0)a_2(0)a_3^*(0)[\chi^{(1)}_{123}\mathbf{l}_1(\mathbf{l}_2\mathbf{l}_3^*)$$
$$+\chi^{(2)}_{123}\mathbf{l}_2(\mathbf{l}_3^*\mathbf{l}_1)+\chi^{(3)}_{123}\mathbf{l}_3^*(\mathbf{l}_1\mathbf{l}_2)], \tag{39}$$

where

$$\Omega_1=-\omega_1+\omega_2+\omega_3, \quad \Omega_2=\omega_1-\omega_2+\omega_3, \quad \Omega_3=\omega_1+\omega_2-\omega_3.$$

The quantities $G(\Omega_f,\mathbf{s}_f)$ with $f=1, 2, 3$ contain q_f and $\mathbf{K}_f$ in the form

$$q_f=\sum_{n=1}^{3}\alpha_n(\mathbf{k}_n\mathbf{s}_f)/k_n s_f,$$
$$\mathbf{K}_1=-\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3, \quad \mathbf{K}_2=\mathbf{k}_1-\mathbf{k}_2+\mathbf{k}_3, \quad \mathbf{K}_3=\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_3.$$

Polarization spectroscopy using the diffracted waves (37)–(39) is more versatile than a self-diffraction of two waves, because of the greater diversity of behavior. From this behavior we single out the most characteristic features on which the hyperfine structure of the resonant levels has a strong effect. For example, assume that all three incident waves are linearly polarized, and that the polarization vectors $\mathbf{l}_2$ and $\mathbf{l}_3$ are orthogonal to $\mathbf{l}_1$. We thus have $\mathbf{l}_1\mathbf{l}_2=\mathbf{l}_1\mathbf{l}_3=0$ and $\mathbf{l}_2\mathbf{l}_3\neq 0$. The diffracted wave with amplitude (37) is then polarized in the same plane as the first incident wave, and its intensity $I(\mathbf{s}_1)$ at the exit from the gas is

$$I(\mathbf{s}_1)=(2\pi/c)^2|G(\Omega_1,\mathbf{s}_1)\chi_{231}^{(3)}(\mathbf{l}_2\mathbf{l}_3)|^2 I_1 I_2 I_3,$$
$$I_n=c|a_n(0)|^2/2\pi, \quad n=1,\ 2,\ 3,$$

where I_n is the intensity of the incident wave at the entrance to the gas. By virtue of $\chi_{231}^{(3)}$, the hyperfine structure of the resonant levels and the nuclear spin strongly influence the diffraction efficiency in the transmission of optical images and optical information, particularly under conditions (14) or (17), when (16) holds.

Another characteristic aspect of the behavior arises when the angles at which the incident waves converge are small, under the inequality

$$|\omega_2-\omega_3|, \quad |\mathbf{k}_2-\mathbf{k}_3|u\ll\gamma_a,\ \gamma_b.$$

In this case we can set $\chi_{231}^{(1)}=\chi_{231}^{(2)}$ in (37) for homogeneously and inhomogeneously broadened transitions. If the second and third incident waves are assumed to be circularly polarized with polarization vectors $\mathbf{l}_2=\mathbf{l}_3^*$ in this case, we find the following expression for $\mathbf{b}_1$ in (37):

$$\mathbf{b}_1=G(\Omega_1,\mathbf{s}_1)a_1^*(0)a_2(0)a_3(0)(\chi_{231}^{(1)}+\chi_{231}^{(3)}]\mathbf{l}_1^*.$$

The polarization vector $\mathbf{l}_1$ here is arbitrary, while the amplitude $\mathbf{b}_1$ may vary significantly, depending on the type of transition $J_aF_a\rightarrow J_bF_b$, because of the term $\chi_{231}^{(3)}$. Other diffracted waves behave analogously when the polarizations of the incident waves are chosen correspondingly.

5. SELF-DIFFRACTION OF COUNTERPROPAGATING LIGHT WAVES; PHASE CONJUGATION

Let us consider the self-diffraction of three incident waves of the form (1) with $n_0=3$, for the case in which two reference waves, with $\mathbf{k}_1$ and $\mathbf{k}_2$ are propagating in opposite directions, at an angle slightly different from π, while the wave vector $\mathbf{k}_3$ of the signal wave makes a small angle, on the order of a milliradian, with $\mathbf{k}_2$ (Refs. 14–18 and 36). We assume that the first, second, and third waves enter the gas at the boundary points $\mathbf{r}=0$, $\mathbf{r}=\mathbf{r}_2$, and $\mathbf{r}=\mathbf{r}_3$. The diffracted waves are then described by (37)–(39), in which the quantities $a_2(0)$ and $a_3(0)$ are replaced in accordance with

$$a_2(0)\rightarrow a_2(\mathbf{r}_2)\exp(-\alpha_2L_2), \quad a_3(0)\rightarrow a_3(\mathbf{r}_3)\exp(-\alpha_3L_3) \tag{40}$$

(with similar replacements for the complex-conjugate quantities). Here α_2 (α_3) is the absorption coefficient, and L_2 (L_3) is the optical path length of the incident wave in the active zone of the gas.

Of the three diffracted waves, we will study in detail the one which is propagating opposite the signal wave. Its electric field $\mathbf{E}^{NL}(\mathbf{r},t)$, at the point $\mathbf{r}=\mathbf{s}_3L/s_3$, at the exit from the gas, can be written as follows, according to (39) and (40):

$$\mathbf{E}^{NL}(\mathbf{r},\ t)=\mathbf{S}R\exp[i(\mathbf{s}_3\mathbf{r}-\Omega_3t)]+\text{c. c.}, \tag{41}$$

where

$$\mathbf{S}=\chi_{123}^{(1)}\mathbf{l}_1(\mathbf{l}_2\mathbf{l}_3^*)+\chi_{123}^{(2)}\mathbf{l}_2(\mathbf{l}_3^*\mathbf{l}_1)+\chi_{123}^{(3)}\mathbf{l}_3^*(\mathbf{l}_1\mathbf{l}_2),$$
$$R=G(\Omega_3,\ \mathbf{s}_3)a_1(0)a_2(\mathbf{r}_2)a_3^*(\mathbf{r}_3)\exp(-\alpha_2L_2-\alpha_3L_3),$$

and L is the optical path length of wave (41) in the gas.

We restrict the discussion below to degenerate four-wave mixing $\omega_1=\omega_2=\omega_3=\omega$ under the equalities $\mathbf{k}_1+\mathbf{k}_2=0$ and $\Omega_3=\omega$. In this case the diffracted wave in (41), which is propagating in the optimum direction, $\mathbf{s}_3=-\mathbf{k}_3$, becomes the conjugate of the signal wave, as in other gaseous and solid media.[14–18] In this case, however, the coefficient η for reflection into the conjugate wave depends strongly on the type of transition between the hyperfine sublevels and on the polarizations of the incident waves, as follows from the relations

$$I^{NL}=\eta I_3,$$
$$\eta=(2\pi^2\omega^2/\alpha k_3c^3)^2I_1I_2|\mathbf{S}|^2[1-\exp(-2\alpha L)]^2\exp(-2\alpha L),$$
$$\alpha=\omega^2\varepsilon''(\omega,\ \mathbf{k}_1)/2k_1c^2, \quad \alpha_1=\alpha_2=\alpha_3=\alpha, \quad L_2=L_3=L,$$

where I_1 (I_2), I_3, and I^{NL} are the intensities of the reference wave, the signal wave, and the conjugate wave.

For linearly polarized waves in the case $\mathbf{l}_1=\mathbf{l}_2=\mathbf{l}_3$ or $\mathbf{l}_1\mathbf{l}_3=\mathbf{l}_2\mathbf{l}_3=0$ and $\mathbf{l}_1\mathbf{l}_2\neq 0$, the diffracted wave in (41) is the exact conjugate of the wavefront of the signal wave, in accordance with the results of Ref. 17, which were derived for phase conjugation without an analysis of the contribution of the hyperfine level structure. The latter structure affects only the intensity of this conjugate wave.

For homogeneously broadened transitions,

$$|\mathbf{k}_1-\mathbf{k}_3|u\ll\gamma_a,\ \gamma_b, \quad k_nu\ll\gamma_{ba}, \quad n=1,\ 2,\ 3, \tag{42}$$

and for circularly polarized reference waves, $\mathbf{l}_1=\mathbf{l}_2^*$, we find, by virtue of the equality $\chi_{123}^{(1)}=\chi_{123}^{(2)}$ in (41),

$$\mathbf{S}=(\chi_{123}^{(1)}+\chi_{123}^{(3)})\mathbf{l}_3^*.$$

In this case, therefore, complete phase conjugation, in the spatial and polarization senses, of a signal wave of arbitrary polarization $\mathbf{l}_3$ is thus achieved, as in Refs. 43, 44, and 47. Those previous studies ignored the hyperfine structure and the homogeneously broadened nature of the resonant transition.

An important dependence of the conjugate wave (41) on the hyperfine structure of the resonant levels arises for linearly polarized incident waves under resonance conditions (14) or (17). Following the experiments of Ref. 24, we choose the polarization planes for the incident waves in such a way that the polarization vectors $\mathbf{l}_1$ and $\mathbf{l}_2$ of the reference waves make an angle of 2θ, while the angle between the polarization vector $\mathbf{l}_3$, of the signal wave and the bisector of the angle between the vectors $\mathbf{l}_1$ and $\mathbf{l}_2$ has an arbitrary value θ_c:

$$\mathbf{l}_1=\mathbf{l}_x\cos\theta-\mathbf{l}_y\sin\theta,\quad \mathbf{l}_2=\mathbf{l}_x\cos\theta+\mathbf{l}_y\sin\theta,$$

$$\mathbf{l}_3=\mathbf{l}_x\cos\theta_c+\mathbf{l}_y\sin\theta_c.$$

The Cartesian axis z is parallel to $\mathbf{k}_1$, while the x axis is directed along the bisector of the angle between the vectors $\mathbf{l}_1$ and $\mathbf{l}_2$. The angles are positive when they are measured from the x axis toward the y axis.

Under the assumptions which we have adopted here, the intensity I corresponding to the projection $\mathbf{l}_3\mathbf{E}^{NL}$ of the electric field of the conjugate wave (41) onto the polarization vector $\mathbf{l}_3$, of the signal wave is a function of these angles. It is given by

$$I(\theta,\ \theta_c)=c|\mathbf{l}_3\mathbf{S}R|^2/2\pi,$$

where

$$\begin{aligned}\mathbf{l}_3\mathbf{S}=\{&(Q_{123}+Q_{213})({}^1/{}_3D^{(0)}+{}^1/{}_2D^{(1)}+{}^1/{}_6D^{(2)})\\&+[\gamma(2J_b+1)|d_{FbFa}|^2/(\gamma_a+\gamma_b)|d_{ba}|^2](\tilde{Q}_{123}+\tilde{Q}_{213})({}^1/{}_3\tilde{D}^{(0)}\\&+{}^1/{}_2\tilde{D}^{(1)}+{}^1/{}_6\tilde{D}^{(2)})\}\cos(\theta+\theta_c)\cos(\theta-\theta_c)\\&+{}^1/{}_2\{(Q_{123}+Q_{213})(D^{(2)}-D^{(1)})\\&+[\gamma(2J_b+1)|d_{FbFa}|^2/(\gamma_a+\gamma_b)|d_{ba}|^2](\tilde{Q}_{123}+\tilde{Q}_{213})\\&\times(\tilde{D}^{(2)}-\tilde{D}^{(1)})\}\cos 2\theta.\end{aligned}\qquad(43)$$

In a dense gas, with $\gamma\ll\gamma_a+\gamma_b$, the terms in (43) which contain γ can be ignored. Using (43), we can easily determine that value $\theta_c^{(0)}$ of the angle θ_c for which the intensity $I(\theta,\theta_c^{(0)})$ vanishes $[I(0,\theta_c^{(0)})=0]$ at $\theta=0$, for both homogeneously and inhomogeneously broadened transitions. As a result we find that the equality $I(0,\theta_c^{(0)})=0$ holds for only atomic transitions $J_aF_a\to J_bF_b$ which satisfy the inequality $D^{(2)}-D^{(1)}\leqslant 0$, and the angle $\theta_c^{(0)}$ is given by

$$\cos^2\theta_c^{(0)}=2F(F+2)/(6F^2+12F+5),\qquad(44)$$

for $F_a=F\to F_b=F+1$, $F_a=F+1\to F_b=F$ and

$$\cos^2\theta_c^{(0)}={}^1/{}_2\qquad(45)$$

for $F_a=1/2\to F_b=1/2$.

Experimentally, it is useful to study, along with (44) and (45), the ratio $I(0,\theta_c)/I(0,\pi/2)$ as a function of the angle θ_c:

$$\frac{I(0,\theta_c)}{I(0,\pi/2)}=\left(\frac{6F^2+12F+5}{4F^2+8F+5}\right)^2$$

$$\times\left(\frac{2F(F+2)}{6F^2+12F+5}-\cos^2\theta_c\right)^2,\qquad F_a-F_b=\pm1,$$

$$I(0,\theta_c)/I(0,\pi/2)=4({}^1/{}_2-\cos^2\theta_c)^2,\qquad F_a=F_b={}^1/{}_2.$$

For transitions $J_aF_a\to J_bF_b$, for which a different inequality holds, $D^{(2)}-D^{(1)}\geqslant 0$, we should choose that value $\theta^{(0)}$ of the angle θ for which the specified intensity $I(\theta^{(0)},\theta_c)$ vanishes $[I(\theta^{(0)},0)=0]$ at $\theta_c=0$. This case occurs on transitions $F_a=F\to F_b=F$ $(F>\frac{1}{2})$ under a different condition, because of the different arrangement of polarization planes:

$$\cos^2\theta^{(0)}=(F-1)(F+2)/(2F-1)(2F+3),$$

$$\frac{I(\theta,0)}{I(0,\pi/2)}=\frac{(2F-1)^2(2F+3)^2}{(3F^2+3F-1)^2}\left(\frac{(F-1)(F+2)}{(2F-1)(2F+3)}-\cos^2\theta\right)^2.$$

(46)

Expressions (44)–(46) can be used to experimentally identify transitions $J_aF_a\to J_bF_b$, on the basis of the change in the total angular momentum, $F_a\to F_b$. The reason is that there is a mutually one-to-one correspondence between F and $\theta_c^{(0)}$ (and also between F and $\theta^{(0)}$). For example, corresponding to transitions with a change in total angular momentum $0\to1$ $(1\to0)$, $1\to2$ $(2\to1)$, $2\to3$ $(3\to2)$, and $3\to4$ $(4\to3)$ are angles $\theta_c^{(0)}$ equal to 90°, 59°17′, 56°40′, and 55°49′. Correspondingly, angles $\theta^{(0)}$ of 90°, 64°07′, 61°52′, and 61°05′ correspond to transitions without a change in the total angular momentum ($1\to1$, $2\to2$, $3\to3$, and $4\to4$). For half-integer values of the total angular momenta, the transitions $\frac{1}{2}\to\frac{1}{2}$, $\frac{1}{2}\to\frac{3}{2}$ $(\frac{3}{2}\to\frac{1}{2})$, $\frac{3}{2}\to\frac{5}{2}$ $(\frac{5}{2}\to\frac{3}{2})$, and $\frac{5}{2}\to\frac{7}{2}$ $(\frac{7}{2}\to\frac{5}{2})$ correspond to angles $\theta_c^{(0)}$ of 45°, 63°26′, 57°34′, and 56°09′, while transitions $\frac{3}{2}\to\frac{3}{2}$, $\frac{5}{2}\to\frac{5}{2}$, and $\frac{7}{2}\to\frac{7}{2}$ correspond to angles $\theta^{(0)}$ of 67°33′, 62°40′, and 61°24′.

In a low-density gas, with $\gamma\lesssim\gamma_a+\gamma_b$, the terms in (43) with a factor of γ become important, and the expressions for $\theta_c^{(0)}$, $\theta^{(0)}$, $I(0,\theta_c)/I(0,\pi/2)$, and $I(\theta,0)/I(0,\pi/2)$ take a simple form for homogeneously broadened transitions, (42). For such transitions, we have

$$Q_{123}=Q_{213}=\tilde{Q}_{123}=\tilde{Q}_{213}$$

$$=\frac{2N_{ab}|d_{F_bF_a}|^4\gamma_{ba}(\gamma_a+\gamma_b)(\omega-\omega_{F_bF_a}-i\gamma_{ba})}{\hbar^3(2I+1)\gamma_a\gamma_b[(\omega-\omega_{F_bF_a})^2+\gamma_{ba}^2]^2}.\qquad(47)$$

By virtue of (43) and (47), we can replace (44) and (45) by

$$\cos^2\theta_c^{(0)}=-3(C^{(2)}-C^{(1)})/(2C^{(0)}+3C^{(1)}+C^{(2)}).\qquad(48)$$

If the right side of this equation is negative, then we should use

$$\cos^2\theta^{(0)}=3(C^{(2)}-C^{(1)})/(2C^{(0)}-3C^{(1)}+7C^{(2)}),\qquad(49)$$

according to (43) and (47).

For transitions $J_aF_a\to J_bF_b$ with any possible change in the electron angular momentum ($J_a=J\to J_b=J+1$, $J_a=J+1\to J_b=J$, and $J_a=J\to J_b=J$), relation (48) with $F_a=F\to F_b=F+1$ and $F_a=F+1\to F_b=F$ becomes

$$\cos^2\theta_c^{(0)}=F(F+2)[2(F+1)(2F+1)(2F+3) -\Gamma(F_a,J_a;F_b,J_b)(4F^2 +8F+5)][(F+1)(2F+1)(2F+3)(6F^2+12F+5) -\Gamma(F_a,J_a;F_b,J_b)(12F^4 +48F^3+63F^2+30F+5)]^{-1}, \qquad (50)$$

Relation (49) takes the following form in the case $F_a=F\rightarrow F_b=F$ $(F>\frac{1}{2})$:

$$\cos^2\theta^{(0)}=(F-1)(F+2)[2F(F+1)(2F+1) -\Gamma(F,J_a;F,J_b)(2F^2 +2F-1)][2F(F+1)(2F+1)(2F+3)(2F-1) -\Gamma(F,J_a;F,J_b)(8F^4 +16F^3-7F^2-15F+8]^{-1}. \qquad (51)$$

According to (48), the transition $F_a=\frac{1}{2}\rightarrow F_b=\frac{1}{2}$ with $J_a=J\rightarrow J_b=J+1$, $J_a=J+1\rightarrow J_b=J$, and $J_a=J\rightarrow J_b=J$ can be written in a special way:

$$\cos^2\theta_c^{(0)}=[6-\Gamma({}^1/_2,J_a;{}^1/_2,J_b)]/4[3-\Gamma({}^1/_2,J_a;{}^1/_2,J_b)].$$

In the region $3<\Gamma(\frac{1}{2},J_a;\frac{1}{2},J_b)<6$ we find from (49)

$$\cos^2\theta^{(0)}=[6-\Gamma({}^1/_2,J_a;{}^1/_2,J_b)]/2\Gamma({}^1/_2,J_a;{}^1/_2,J_b), \qquad (52)$$

The last equation can hold only if the arrival of atoms in the lower level E_a due to spontaneous emission from the upper level E_b dominates.

Let us apply (48)–(52) to the homogeneously broadened resonant transition $2P^{(0)}_{3/2}\rightarrow 2D_{5/2}$ of copper atoms with $I=J_a=\frac{3}{2}$ and $J_b=\frac{5}{2}$ in an extremely low-density gas, with $\gamma/(\gamma_a+\gamma_b)=1$. Corresponding to transitions $F_a\rightarrow F_b$ with a change in total angular momentum 0 → 1, 1 → 2, 2 → 3, and 3 → 4 are angles $\theta_c^{(0)}$ of 90°, 71°30′, 78°30′, and 45°, respectively. Corresponding to transitions 2 → 1 and 3 → 2, on the other hand, are the angles $\theta_c^{(0)}=59°26'$ and $\theta_c^{(0)}=56°42'$. Corresponding to transitions without a change in the total angular momentum (1 → 1, 2 → 2, and 3 → 3) are angles $\theta^{(0)}$ of 90°, 64°48′, and 62°4′.

The experiments of Ref. 24 used copper vapor at a temperature $T=1800$ K and light waves with a wavelength $\lambda=510.5$ nm in resonance with the transition $2P^0_{3/2}\rightarrow 2D_{5/2}$. The measurements yielded $\theta_c^{(0)}=53°$. According to (48), the value $\theta_c^{(0)}=53°$ is reached on the homogeneously broadened transition $F_a=3\rightarrow F_b=4$ at $\gamma/(\gamma_a+\gamma_b)\approx\frac{1}{2}$. In the experiments of Ref. 24, however, inhomogeneously broadened transitions were studied. Consequently, to calculate $\theta_c^{(0)}$ we need to know not only the value of $\gamma/(\gamma_a+\gamma_b)$ but also the number of hyperfine sublevels in the lower and upper levels which lie within the Doppler width. That information was not given in Ref. 24.

6. EQUATIONS FOR COUPLED WAVES

The hyperfine level structure and the nuclear spin also have strong effects on the self-diffraction of four incident waves of the type in (1), with $n_0=4$, if these waves have identical frequencies $\omega_1=\omega_2=\omega_3=\omega_4=\omega$ $(k_1=k_2=k_3=k_4=k)$ and are propagating in opposite directions in pairs:

$$\mathbf{k}_1+\mathbf{k}_2=0,\quad \mathbf{k}_3+\mathbf{k}_4=0, \qquad (53)$$

where the waves with $\mathbf{k}_1$ and $\mathbf{k}_2$ serve as reference waves, while those with $\mathbf{k}_3$ and $\mathbf{k}_4$ are the signal waves. This spectroscopic scheme is one of the most promising for holography, as has been pointed out in numerous studies, which are covered in reviews[14,15] and books.[17,18] In particular, studies of this sort have been carried out in the steady state in a gas containing sodium vapor.[1,2] In those studies, the optical image transmission, energy transfer, and other questions were examined without consideration of a hyperfine structure. We will therefore point out some new possibilities which arise when the hyperfine structure is taken into account.

Under conditions (53), the nonlinear interaction of the first, second, and third incident waves generates a new wave, which propagates in the $\mathbf{k}_4$ direction. The nonlinear interaction of the first, second, and fourth incident waves gives rise to a new wave, with $\mathbf{k}_3$. If we assume that the intensities of the first and second waves are high in comparison with those of the third and fourth, then the amplitudes of the weak waves are described by the following system of coupled equations, according to Maxwell's equations, (21):

$$(\mathbf{k}_3\nabla/k+\alpha_3)b_{3i}=(12i\pi\omega^2/kc^2)\chi_{ijkl}(\omega;\omega,\omega,-\omega)b_{1j}b_{2k}b_{4l}^*, \qquad (54)$$

$$(\mathbf{k}_4\nabla/k+\alpha_4)b_{4i}=(12i\pi\omega^2/kc^2)\chi_{ijkl}(\omega;\omega,\omega,-\omega)b_{1j}b_{2k}b_{3l}^*. \qquad (55)$$

Here the amplitudes $\mathbf{b}_1$ and $\mathbf{b}_2$ of the strong reference waves are given quantities, and the degree of coupling of the weak signal waves depends on the type of resonant transition and on the polarizations of the incident waves.

The contribution of the hyperfine structure of the resonant levels to the propagation of the waves with the amplitudes $\mathbf{b}_3$ and $\mathbf{b}_4$ is optimum for linearly polarized incident waves with polarization vectors $\mathbf{l}_1=\mathbf{l}_2$, $\mathbf{l}_3=\mathbf{l}_4$, and $\mathbf{l}_1\mathbf{l}_3=0$, for which Eqs. (54) and (55) become

$$(\mathbf{k}_3\nabla/k+\dot{\alpha}_3)\mathbf{b}_3=(2i\pi\omega^2/kc^2)\chi^{(3)}_{124}(\mathbf{b}_1\mathbf{b}_2)\mathbf{b}_4^*, \qquad (56)$$

$$(\mathbf{k}_4\nabla/k+\alpha_4)\mathbf{b}_4=(2i\pi\omega^2/kc^2)\chi^{(3)}_{123}(\mathbf{b}_1\mathbf{b}_2)\mathbf{b}_3^*. \qquad (57)$$

Let us assume that the frequency ω of the incident waves is in resonance with a transition between two hyperfine sublevels, (14) or (17). According to (16), the third and fourth waves then propagate independently when the total angular momentum 1 → 0, 0 → 1, and 1 → 1 for the resonant transitions $J_aF_a\rightarrow J_bF_b$ changes. For other transitions, the propagation of one of these signal waves is coupled with the propagation of the other, and in each case the propagation depends strongly on the type of transition and on the nuclear spin. For homogeneously broadened transitions (42), for example, we have

$$\chi^{(3)}_{123}=\chi^{(3)}_{124}=Q_{123}(C^{(2)}-C^{(1)}),$$

where the dependence on the type of transition $J_aF_a\rightarrow J_bF_b$

and on the nuclear spin I is incorporated in the difference $C^{(2)} - C^{(1)}$, as can be easily verified experimentally with the help of (34).

If all four waves are polarized in a common plane, then Eqs. (56) and (57) with the replacements

$$\chi^{(3)}_{124} \to \sum_{\varkappa=1}^{3} \chi^{(\varkappa)}_{124}, \quad \chi^{(3)}_{123} \to \sum_{\varkappa=1}^{3} \chi^{(\varkappa)}_{123} \tag{58}$$

follow from (54) and (55).

Finally, for circularly polarized waves with $\mathbf{l}_1 = \mathbf{l}_2^*$ and $\mathbf{l}_3 = \mathbf{l}_4^*$, in the case of homogeneously broadened transitions (42), we again find Eqs. (56) and (57) from (54) and (55), with the replacements

$$\chi^{(3)}_{124} \to \chi^{(1)}_{124} + \chi^{(3)}_{124}, \quad \chi^{(3)}_{123} \to \chi^{(1)}_{123} + \chi^{(3)}_{123}. \tag{59}$$

In the last two cases, i.e., (58) and (59), the effect of the hyperfine level structure is quite different from that in (56) and (57), because of the terms added to $\chi^{(3)}_{124}$ and $\chi^{(3)}_{123}$.

7. DISCUSSION

When four or more light waves are incident on a gas, various types of four-wave mixing occur, involving one new wave and any three of the incident waves. Certain types of four-wave mixing involve an incident wave twice and constitute a self-diffraction of two waves. Corresponding to the various types of four-wave mixing there are various types of self-diffraction of either two or three waves. Each process proceeds independently. The diffracted waves which arise are spatially separated by virtue of the phase matching and are described by Eqs. (30), (31), and (37)-(39), with the appropriate changes in indices. If, among a large number of incident waves, there are two which are propagating in opposite directions, then along with the self-diffraction of waves making small angles with each other there are independent self-diffraction processes of various types, accompanied by the formation of conjugate waves. The latter are described by (41), with altered indices of the incident waves.

Several features of the effect of the hyperfine level structure and of the nuclear spin on the self-diffraction and phase conjugation within the one-photon absorption line fundamentally distinguish these processes from the self-diffraction and phase conjugation which involve forbidden transitions between hyperfine sublevels of two atomic levels. For example, in the case of coherent anti-Stokes scattering of light,[17,45] two pump waves with frequencies ω_1 and ω_2 ($\omega_1 > \omega_2$) satisfy the condition for Raman resonance, $\omega_1 - \omega_2 \approx \omega_{cb}$, on a forbidden transition with a frequency ω_{cb}. They induce a diffraction grating in the medium; the wave with frequency ω_1 is scattered by this grating. Raman spectroscopy[17] uses, along with two pump waves with frequencies ω_1 and ω_2 ($\omega_1 - \omega_2 \approx \omega_{cb}$), a third test wave at a nonresonant frequency ω_3. This third wave undergoes diffractive scattering by the periodic structure induced by the two pump waves. Several experiments[46] have revealed a self-diffraction of waves with frequencies ω_1 and ω_2 at resonance with the frequencies ω_{ca} and ω_{ba} of adjacent transitions $E_a \to E_c$ and $E_a \to E_b$ having a common lower level E_a and two different upper levels, E_b and E_c ($E_c > E_b$), which form a forbidden transition with a frequency $\omega_{cb} \approx \omega_1 - \omega_2$. An example of phase conjugation using a forbidden transition is Doppler-free coherent spectroscopy,[33] in which two pump waves, with the same frequency $\omega_1 = \omega_2 = \omega$, are propagating in opposite directions ($\mathbf{k}_1 + \mathbf{k}_2 = 0$) and satisfy the two-photon resonance $\omega_1 + \omega_2 \approx \omega_{cb}$, while the test wave, with $\omega_3 = \omega$, is propagating at a small angle from the wave vector $\mathbf{k}_2$. In none of these cases of self-diffraction and phase conjugation involving a forbidden transition is the resonance at the zero frequency involved. In no case does the arrival of atoms to the lower level due to the spontaneous decay of the excited state contribute. As was shown in Ref. 34, the hyperfine structure is reflected in the amplitude and polarization behavior in a completely different way in these cases. The effect of this hyperfine structure always fades away when the detuning from the resonance involving the hyperfine sublevels becomes sufficiently large, in contrast with the cases of self-diffraction and phase conjugation within a one-photon absorption line, in which this effect often persists.

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