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PHYSICAL REVIEW A VOLUME 1, NUMBER 3 MARCH 1970

Stimulated Electric Polarization and Photon Echoes*

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(Received 20 August 1969)

The change in the wave function of an atom or molecule which interacts with saturating incident radiation is shown to have a time evolution operator $U(t,0)=\sum |n)(n'|B(n\alpha)B^*(n'\alpha) \times e^{-i[\xi(n\alpha)+E(n)/\hbar]t}$, where the sum over n and n' is over both the $(2J_a+1)$ values of m_a and $(2J_b+1)$ values of m_b, and the sum over α is over the $(2J_a+1)+(2J_b+1)$ modes indicated by the index α. The eigenvalues $\xi(n\alpha)$ and their eigenvectors $B(n\alpha)$ depend on the intensity and polarization of the incident radiation, produce a modulation term in the electric polarization $\vec{P}$ of the molecule, and give rise to anomalous polarization in the stimulated radiation. This unitary operator is used to discuss the radiation stimulated by two pulses or photon echoes. Echoes from elliptic pulses are discussed for $J\leq 2$, and the linear-linear sequence is compared with the theory of Gordon, Wang, Patel, Slusher, and Tomlinson. Echoes from linear-circular and circular-linear pulse sequences are discussed in detail.

I. INTRODUCTION

The anomalous response of an atom or molecule to an intense radiation field is of considerable interest for the anomalous polarization which occurs in laser radiation,[1,2] the anomalous polarization which occurs in the cosmic OH emission,[3] and in the polarization phenomena which occurs in photon echoes.[4,5] The earlier calculations[1] for lasers were made using a perturbation approach and lead to the interesting conclusion that a laser operating between a $J_a=2 \leftrightarrow J_b=2$ transition prefers circular polarization, while a $J_a=2\leftrightarrow J_b=1$ laser prefers linear.[6] Photon echoes occur in the region in which the perturbation approach is no longer appropriate.

This paper uses an approach which permits the discussion of radiation fields which are so strong that a perturbation approach is not appropriate. The interaction of an atom or molecule with a single frequency time-dependent potential $V(t)$ for a finite time interval is considered. If this interaction is between energy levels with degeneracies of $(2J_a+1)$ and $(2J_b+1)$, the problem can be discussed in terms of $(2J_a+1)+(2J_b+1)$ modes. These modes have eigenvalues which depend on the nature and strength of the interaction. A time evolution operator is developed and tables of coefficients are given for all electric and magnetic dipole transitions for $J\leq 2$. The electric polarization of the atom or molecule, which is stimulated by the radiation field, is given in terms of the time evolution operator. This development permits the discussion of the intensity and elliptic polarization of photon echoes with arbitrary polarization for the first and second pulses. A detailed discussion is given for linear-linear, linear-circular, and circular-linear sequence of pulses for $J\leq 2$. The linear-linear effects are in agreement with the very elegant operator development of Gordon *et al.*[4] Linear-circular or circular-linear have distinctive features in the echo intensities and should yield an assignment of the J values of the transitions.

Since the development in this paper treats the interaction as the addition of a time-dependent term to the Hamiltonian or treats the radiation field in a classical manner, spontaneous emission cannot be included in the development. The finite lifetime of the energy levels cannot be included, and the theory is only applicable to problems for which the level lifetime is longer than the duration of the radiation pulses. The theory does suggest aspects which a complete quantum treatment must include for degenerate energy levels. Rabi, Ramsey, and Schwinger[7] used a doubly rotating coordinate system to discuss magnetic transitions between the magnetic sublevels of a given value of J (I for nuclei). Their method was well suited to strong radiation fields, and the second rotation was a measure of the nearness to resonance and the strength of the radiation field. An algebraic analysis of the type used in this paper would yield the matrix elements of their second rotation operator. An interaction stimulating transitions between energy levels with angular momenta J_a and J_b cannot be treated by their method, and an algebraic method is used. The result has the same usefulness for stimulated electric and magnetic dipole transitions between different energy levels of molecules as their method has for a given atomic sublevel. The limitations are also the same. The resulting expression for the electric polarization yields an explanation for most of the anomalous polarization effects for laser and echo experiments.

II. THEORY

A. Transitions between Two Degenerate Energy Levels

The Schrödinger equation for a system subjected to a time-varying perturbation is

$$i\hbar \frac{\partial\psi}{\partial t} = [H_0 + V(t)]\,\psi \,. \tag{1}$$

If $V(t)$ is a time-varying perturbation at a single frequency ω,

$$V(t) = V_0 e^{-i\omega t} + V_0^+ e^{i\omega t} \,, \tag{2}$$

and the interaction is between two degenerate sets of atomic levels $|a)$ and $|b)$; this equation has an interesting set of solutions. In the approximation in which terms at 2ω are omitted or the rotating wave approximation is used, a solution of this equation is of the form

$$\psi(t) = \sum_{n,\,\alpha} |n) D_{n\alpha}\, e^{-i(\xi_{n\alpha} + E_n/\hbar)t} \,, \tag{3}$$

$|n)$ is used to denote the allowed wave functions of H_0 with energy E_n. Direct substitution into Eq. (1) yields

$$\sum_\alpha \Big[D_{n'\alpha}\xi_{n'\alpha} - \sum_n \hbar^{-1}(n'|V_0|n)D_{n\alpha} \times e^{i(\xi_{n'\alpha} - \xi_{n\alpha} + \omega_{n'n} - \omega)t}\Big] e^{-i\xi_{n'\alpha}t} = 0 \,. \tag{4}$$

This equation can be satisfied for all t if

$$\xi_{n'\alpha} - \xi_{n\alpha} + \omega_{n'n} - \omega = 0 \,, \tag{5}$$

and
$$\xi_{n'\alpha} D_{n'\alpha} - \sum_n \hbar^{-1}(n'|V_0|n)D_{n\alpha} = 0 \,. \tag{6}$$

The discussion in this paper is limited to degenerate energy levels, but this condition is not essential. Let the upper atomic state be denoted by $|J_a m_a)$ and the lower atomic state by $|J_b m_b)$ with energy separation

$$E_a - E_b = \hbar\omega_{ab} \,. \tag{7}$$

For convenience let

$$\Delta = (\omega_{ab} - \omega) \,, \tag{8}$$

and then the condition which is given by Eq. (5) can be written

$$\xi(J_a\alpha) - \xi(J_b\alpha) = -\Delta \,. \tag{9a}$$

In much of the subsequent discussion, the notation

$$\xi(J_a\alpha) = \xi_\alpha \tag{9b}$$

is used. Whenever it is convenient $D(n\alpha)$, $\xi(n\alpha)$, etc., will be used for $D_{n\alpha}$, $\xi_{n\alpha}$, etc. With this notation, Eq. (6) can be written

$$\hbar\xi_\alpha D(m_a\alpha) - \sum_{m_b} (m_a|V_0|m_b)D(m_b\alpha) = 0 \,, \tag{10a}$$

$$\hbar(\xi_\alpha + \Delta)D(m_b\alpha) - \sum_{m_a} (m_a|V_0|m_b)^* D(m_a\alpha) = 0 \,. \tag{10b}$$

The coefficients of the D's form a Hermitian matrix, and this set of $(2J_a+1)+(2J_b+1)$ homogeneous linear equations has a nontrivial solution when the determinant of the coefficients is zero. This occurs for $(2J_a+1)+(2J_b+1)$ values of ξ_α, and the index α is used to denote these values. Substitution of these $\xi(J_a\alpha)$ or ξ_α for a given α yield a set of orthogonal unnormalized eigenvectors $D(n\alpha)$. These column vectors may be normalized, and then both the rows and columns are orthogonal. Denoting the normalized $D(n\alpha)$ by $B(n\alpha)$,

$$\sum_n B(n\alpha)B^*(n\alpha') = \delta_{\alpha\alpha'} \,, \tag{11a}$$

$$\sum_{\alpha} B(n\alpha)B^*(n'\alpha)=\delta_{nn'} , \tag{11b}$$

where n implies a sum over m_a and m_b. n denotes the row and α the column in the D or B matrices.

The solution of these equations is facilitated by noting that for $\xi_\alpha+\Delta\neq 0$, the $D(m_b\alpha)$ follow from the $D(m_a\alpha)$ and the roots ξ_α,

$$D(m_b\alpha)=(\xi_\alpha+\Delta)^{-1}\hbar^{-1}\sum_{m_a}(m_a|V_0|m_b)^*D(m_a\alpha). \tag{12}$$

Then, the $D(m_a\alpha)$ follow from the solution of the $(2J_a+1)$ homogeneous linear equations,

$$\hbar^2\xi_\alpha(\xi_\alpha+\Delta)D(m_a\alpha)-\sum_{m_a' m_b}(m_a|V_0|m_b)\times(m_a'|V_0|m_b)^*D(m_a'\alpha)=0. \tag{13}$$

Since the matrix element of V_0 is zero between m_b and m_b', the sum over m_b is equivalent to a sum over the complete set, and Eq. (13) reduces to

$$\hbar^2\xi_\alpha(\xi_\alpha+\Delta)D(m_a\alpha)-\sum_{m_a'}\times(m_a|V_0V_0^\dagger|m_a')D(m_a'\alpha)=0. \tag{14}$$

Again the coefficients form a Hermitian matrix. The allowed values of $\lambda=\xi_\alpha(\xi_\alpha+\Delta)$ follow from the requirement that the determinant of the coefficients be zero. This is probably the simplest procedure for finding the roots ξ_α in terms of the matrix elements of V. A similar equation for m_b follows by replacing m_a by m_b, m_a' by m_b', and $V_0V_0^\dagger$ by $V_0^\dagger V_0$, and letting $\xi_\alpha\neq 0$. Orthogonality occurs in many ways in this problem, and the columns of $D(m_a\lambda)$ with different values of $\lambda=\xi_\alpha(\xi_\alpha+\Delta)$ are orthogonal in the sense that

$$\sum_{m_a} D(m_a\lambda)D^*(m_a\lambda')=0 , \quad \lambda\neq\lambda' \tag{15a}$$

At $\Delta=0$ or resonance, this orthogonality condition can be used with the roots ξ_α, and orthogonality between column vectors occurs for the sum over either m_a or m_b separately. Thus, for $\Delta=0$ and $\lambda=\xi_\alpha{}^2$ the relationships

$$\sum_{m_a} B(m_a\lambda)B^*(m_a\lambda')=\tfrac{1}{2}\delta_{\lambda\lambda'}=\sum_{m_b} B(m_b\lambda)B^*(m_b\lambda') \tag{15b}$$

can be used with λ replaced by either $+|\xi_\alpha|$ or $-|\xi_\alpha|$.

B. Electric Dipole Interaction

For an electric dipole interaction, $V(t)$ is of the form

$$V(t)=-\vec{P}\cdot\vec{E}(t) , \tag{16}$$

where P is the electric dipole operator for the atom.

At a particular atomic or molecular site, the electric field $\vec{E}(t)$ is of the form

$$\vec{E}(t)=A\hat{u}e^{-i\omega t}+A^*\hat{u}^*e^{i\omega t} , \tag{17}$$

where A is the amplitude of the wave, and $\hat{u}$ is the complex polarization vector transverse to the direction of the wave, $\hat{k}\cdot\hat{u}=0$. For plane waves, the phase $e^{i\vec{k}\cdot\vec{r}_\alpha}$ can be included in A. For this perturbation, the matrix elements of interest are

$$-(J_am_a|V_0|J_bm_b)=(J_am_a|\vec{P}\cdot\hat{u}|J_bm_b)A=\frac{(J_a\|P\|J_b)}{(2J_a+1)^{1/2}}(J_b1m_bM|J_am_a)\times(\hat{e}_M^*\cdot\hat{u})A . \tag{18}$$

The notation of Messiah[8] is followed, and the Wigner-Eckart theorem is used to give the matrix element in terms of the Clebsch-Gordan coefficients, spherical basis vectors $\hat{e}_M$, and the reduced matrix element. For comparison with other data, it is noted that the reduced matrix element

$$|(J_a\|P\|J_b)|^2=(ea_0)^2S=0.72\times10^{-58}S . \tag{19}$$

The line strength S varies from 0.52 for the 0.632-μ line of Ne to 56 for the 3.39-μ transition.[9]

If the axis of quantization $\hat{z}$ is selected along the direction of the radiation $\hat{k}$, only $M=\pm1$ values occur. The set of equations implied by Eqs. (5), (10), etc. reduce to two independent sets with this axis of quantization. These independent sets are shown by solid and dashed lines in Figs. 1 and 2. Further simplification in notation occurs with

$$(2J_a+1)^{-1/2}(J_a\|P\|J_b)(\hat{e}_\mp^*\cdot\hat{u})A=\hbar u \quad\text{or}\quad \hbar v, \tag{20}$$

where the right-circular component u is used with $M=-1$ and the left-circular v with $M=+1$.

Interaction with the magnetic component $\vec{B}(t)$ of the radiation field is given by

$$V(t)=-\vec{M}\cdot\vec{B}(t),$$

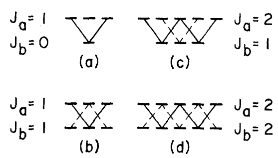

FIG. 1. States for integer J values discussed in text. Solid and dashed lines show the two independent sets of states connected by elliptically polarized radiation along the direction of the axis of quantization.

where $\vec{M}$ is the magnetic dipole operator for the atom and can be treated with a slight change of notation. The reduced matrix element in Eq. (18) becomes $(J_a \| M \| J_b)$, and the unit vector $\hat{u}$ and $\vec{B}$ are related by appropriate polarization conventions. Thus, the theory can be used for magnetic transitions between hyperfine levels or levels split by a crystalline field. Many such transitions occur in the microwave region.

For electric or magnetic dipole transitions, the form of Eq. (14) for $D(m_a\alpha)$ relates the coefficients $D(m_a\alpha)$, $D(m_a+2, \alpha)$, and $D(m_a-2, \alpha)$. For nonzero u and v, all values of $D(m_a\alpha)$ can be expressed in terms of either $D(m_a=J_a, \alpha)$ or $D(m_a=-J_a, \alpha)$ and the matrix elements of $(m_a|VV_0^{\dagger}|m_a')$. The $D(m_b\alpha)$ coefficients follow from Eq. (12), and the $B(n\alpha)$ follow by normalization of the column with index α. The role of m_a and m_b can be interchanged in the above discussion. Since the Clebsch-Gordan coefficients are tabulated for the higher values of J, the roots ξ_α and the $B(n\alpha)$ coefficients should be readily determined for a given set of values of Δ, u, and v by computer methods. Roots with $\xi_\alpha=0$ or $\xi_\alpha+\Delta=0$ do not contribute to the electric polarization, and these column vectors can be omitted in these calculations.

C. Time Evolution, Density Matrix, and Electric Polarization

The evolution in time of the wave function can now be described by the unitary matrix $U(t, t_0)$:

$$\psi(t)=U(t, t_0)\psi(t_0)\,, \tag{21}$$

where
$$U(t, t_0)=\sum_{n, n', \alpha} |n)e^{-iE_n\tau/\hbar} \times e^{-i\xi(n\alpha)\tau}B(n\alpha)B^*(n'\alpha)(n'|\,, \tag{22}$$

with $\tau=t-t_0\,.$

Again n and n' are summed over all values of m_a and m_b. The form of $U(t, t_0)$ follows from Eq. (3), with $D(n\alpha)=c_\alpha B(n\alpha)$, and c_α is determined at $t=t_0$. Although distinct modes with index α have appeared in the problem, it is not possible to discuss the probability of going from α to α'. Only the probability of going from state n' to n is a possible measurement for this molecule and is given by

$$|(n|U(t, t_0)|n')|^2 = |\sum_\alpha e^{-i\xi(n\alpha)\tau}B(n\alpha)B^*(n'\alpha)|^2\,. \tag{23}$$

The electric polarization $\vec{P}(t)$ of a molecule is given by

$$\vec{P}(t)=\mathrm{Tr}U^{\dagger}\vec{P}U\sigma(t_0)=\mathrm{Tr}\vec{P}U\sigma(t_0)U^{\dagger}=\mathrm{Tr}\vec{P}\sigma(t)\,. \tag{24}$$

$U^{\dagger}\vec{P}U$ expresses the evolution in time of the operator $\vec{P}$. The operator $\sigma(t)$ evolves in time as

$$\sigma(t)=|\psi(t))(\psi(t)|=U|\psi(t_0))(\psi(t_0)|U^{\dagger} = U\sigma(t_0)U^{\dagger}\,, \tag{25}$$

where $\sigma(t_0)$ describes the observable aspects for a molecule at t_0. An average of σ over an ensemble of molecules is regarded as the density matrix ρ. If the time evolution of all molecules is described by the same U, then $\sigma(t_0)$ can be replaced by $\rho(t_0)$. In greater detail, the electric polarization vector of a molecule stimulated by a time varying electric field is

$$\vec{P}(t)=\sum_{\substack{\alpha\alpha' \\ n, n', n'', n'''}} (n|\vec{P}|n')(n''|\sigma(t_0)|n''') \times B(n'\alpha)B^*(n''\alpha)B(n'''\alpha')B^*(n\alpha') \times \exp i[\omega_{nn'}\tau-\xi(n'\alpha)+\xi(n\alpha')]\tau\,. \tag{26}$$

Equations (23) and (26) imply different experiments. If the atom or molecule is definitely in the state $|n')$ at t_0, then a physical observation of this system at time t yields state $|n)$, with the probability given by Eq. (23). An observation has been made and the final state is known. The wave picture is more apparent in $\vec{P}$, and $\vec{P}$ can be related to the scattering amplitude of the molecule. For

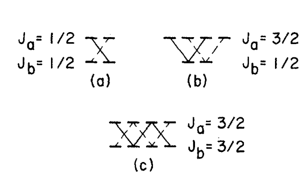

FIG. 2. States for half-integer J values discussed in text. Solid and dashed lines show the two independent sets of states connected by elliptically polarized radiation along the direction of the axis of quantization.

stimulated emission or absorption the stimulated amplitude and the phase are implied by $\vec{P}$. The sum of the stimulated amplitudes for a group of atoms yields the electric field at other locations. Of course, the unitary matrix implies the same information. In either stimulated emission or absorption, the spherical wave at distance r from the scattering molecule is proportional to

$$\vec{E}_s = -\vec{P}(\omega^2/c^2\epsilon_0)e^{ikr}/r, \tag{27}$$

where only $\hat{\theta}$ and $\hat{\varphi}$ components which are transverse to $\hat{r}$ are used for $\vec{P}$.

The $B(n\alpha)$ coefficients and perturbation theory can be related by noting that the evolution matrix element

$$(n|U(t,0)|n') = \sum_\alpha B(n\alpha)B^*(n'\alpha)e^{-i\xi(n\alpha)t}$$
$$\cong +\hbar^{-1}(n|V_0|n')[(e^{-i\Delta t}-1)/\Delta] \tag{28}$$

for nonzero Δ, small V_0, and $n \neq n'$. In this approximation, the electric polarization is proportional to the stimulating electric field. An expression for the time proportional transition probability follows by taking the absolute square of Eq. (28) and integrating over the frequency spread Δ, i.e., $(2\pi/\hbar^2)|(n|V_0|n')|^2 t$.

D. Saturation and Resonance

Saturation occurs for

$$|V_0| > \hbar\Delta \quad , \tag{29a}$$

and for radiation fields which meet this condition, the normalized $B(n\alpha)$ matrix becomes independent of the strength of the radiation field. This is quite apparent at resonance, $\Delta = 0$, and at resonance the $B(n\alpha)$ matrix is independent of the intensity and depends only on the polarization of the incident radiation. The strength of the radiation field occurs in the roots ξ_α. The ratio of these roots depends on the polarization of the incident radiation, and the amplitude of the roots depends on the intensity of the radiation. The unitary matrix describing the time evolution of a saturated system or a system at resonance is quite different from that described by Eq. (28) for a weak field away from resonance. Equation (28) describes a situation in which the electric polarization is proportional to the exciting electric field and corresponds to the region in which an electric susceptibility can be defined. This is no longer possible at saturation. The field strength appears only in the $e^{-i\xi(n\alpha)\tau}$ in Eq. (22), and the polarization of the field determines the $B(n\alpha)$ and the ratio of the $\xi(n\alpha)$.

Since the spectral distribution of a series of random pulses can be described to a certain extent by an electric field of the type given by Eq. (17) of short duration τ, the saturation condition for radiation with a bandwidth $\Delta\nu = \tau^{-1}$ is approximately

$$|V_0| > h\Delta\nu. \tag{29b}$$

When this condition applies, then the $\Delta = 0$ solution should provide a good approximation.

E. Photon Echoes

In photon-echo experiments a sequence of pulses is used, and Eq. (21) becomes for two pulses

$$\psi(t) = U_0(t,t_3)U(t_3,t_2)U_0(t_2,t_1)$$
$$\times U(t_1,0)\psi(0) = U(t,0)\psi(0), \tag{30}$$

where $U_0(t_2,t_1) = e^{-iH_0(t_2-t_1)/\hbar}$

is the evolution in time in the absence of the perturbation. The oscillating component of the polarization is given by Eq. (24). The molecules are assumed in state $|a)$ at $t=0$, and $\sigma(0)$ is a diagonal matrix in m_a. An electric polarization can now be expressed as

$$\vec{P}(t) = \sum_{\substack{m_a' m_b \\ n'', n'''}} e^{i\omega_{ab}(t-t_3)} e^{i(\omega_{n'''}-\omega_{n''})(t_2-t_1)}$$
$$\times (m_a'|\vec{P}|m_b)(m_b|U(t_3t_2)|n'')$$
$$\times (n''|U(t_10)|m_a)(m_a|\sigma(0)|m_a)$$
$$\times (m_a|U^+(t_10)|n''')(n'''|U^+(t_3t_2)|m_a') + \text{c. c.} \tag{31}$$

The sum over n'' and n''' implies a sum over m_a'', m_b'', m_a''', and m_b'''. In an ensemble of molecules interacting with a radiation field, a distribution of values will occur for ω_{ab}, and the sum of the stimulated components at a detector will have random phases. If $n'' = m_a''$ and $n''' = m_b'''$, a component of the polarization occurs with the time factor

$$e^{i\omega_{ab}(t-t_3-t_2+t_1)}.$$

When

$$t - t_3 = t_2 - t_1,$$

molecules with different ω_{ab} will be in phase for a time for the order of the reciprocal of the spread in ω_{ab}. At time $t = t_3 + t_2 - t_1$, an echo occurs. The strength of the echo depends on the matrix elements of U. In the nondegenerate two-level problem, the echo has maximum strength for a $\frac{1}{2}\pi$ pulse between 0 and t_1 and a π pulse between t_2 and t_3. Pulse experiments imply saturation, and in this approximation $\Delta = 0$.

A comparison of the development in this paper with the operator analysis of Gordon *et al.*[4] gives an insight into both methods. Their operator $e^{-iH_0\tau/\hbar}R$ is related to the unitary operator which is given by Eq. (22). The form of Eq. (22) permits the use of a unitary operator

$$U = e^{-iH_0\tau/\hbar} e^{i\Delta\tau} e^{-i\xi\tau},$$

where $(a|\Delta|a) = 0$, $(b|\Delta|b) = \Delta$. ξ is defined as $\xi = B\xi_\alpha B^\dagger$, where B is a unitary matrix, and ξ is a diagonal matrix with roots ξ_α. Since $\exp[i(B\xi_\alpha B^\dagger)\tau] = B(e^{i\xi_\alpha\tau})B^\dagger$, the B matrix is related to the B coefficients by $(n|B|\alpha) = (n|\alpha) = B(n\alpha)$. For $\Delta = 0$, the roots ξ_α occur as $\pm$ pairs and further simplification is possible. For linear polarization, the operator ξ takes on the simple form used in Ref. 4.

III. ROOTS ξ_α AND $B(n\alpha)$ MATRICES

The roots ξ_α and the $B(n\alpha)$ matrices are given in this section for the transitions between the energy levels shown in Figs. 1 and 2. In order to simplify the notation, the columns of $D(n\alpha)$ are given and $B(n\alpha)$ is obtained by normalizing these columns,

$$B(n\alpha) = N_\alpha D(n\alpha), \tag{32a}$$

where

$$\sum_n B^*(n\alpha)B(n\alpha) = (N_\alpha N_\alpha^*)\sum_n D^*(n\alpha)D(n\alpha) = 1. \tag{32b}$$

In any numerical problem, normalization is quite easy, but it is cumbersome in the algebraic form. In accord with Eq. (20), u is the strength of the right-circular component and v the strength of the left-circular component of the radiation.

A. $J_a = 1 - J_b = 0$

The allowed values of ξ_α are

$$\xi_1 = 0 = \xi_4, \quad \xi_2 = -\tfrac{1}{2}(\Delta + \delta), \quad \xi_3 = -\tfrac{1}{2}(\Delta - \delta), \tag{33a}$$

$$\text{where} \quad \delta^2 = \Delta^2 + 4(uu^* + vv^*). \tag{33b}$$

An unnormalized $D(n\alpha)$ matrix is given in Table I, and $B(n\alpha)$ can be obtained by normalizing each column. With the axis of quantization $\hat{z}$ selected along $\hat{k}$, the state 0_a or $m_a = 0$ is not affected by the perturbation.

TABLE I. $D(n\alpha)$ for $J_a = 1$, $J_b = 0$. ξ_α are defined by Eq. (33).

n \ α	1	2	3	4
-1_a	$-v^*$	$-u$	$-u$	0
$+1_a$	u^*	$-v$	$-v$	0
0_b	0	ξ_2	ξ_3	0
0_a	0	0	0	1

B. $J_a = 1 - J_b = 1$

Figure 1(b) emphasizes that the choice of the axis of quantization divides the problem into two independent parts. The roots for the group which is connected by solid lines are denoted by ξ_1, ξ_2, and ξ_3, and the group which is connected by dashed lines by ξ_4, ξ_5, and ξ_6. Then

$$\xi_1 = 0, \quad \xi_2 = -\tfrac{1}{2}(\Delta + \delta) = \xi_5, \quad \xi_3 = -\tfrac{1}{2}(\Delta - \delta) = \xi_6, \quad \xi_4 = -\Delta, \tag{34a}$$

$$\text{where} \quad \delta^2 = \Delta^2 + 2(uu^* + vv^*). \tag{34b}$$

An unnormalized $D(n\alpha)$ matrix is given in Table II. This matrix form illustrates the independence of the two groups. The second group follows from the first by changing $u \to -v^*$, $v \to -u^*$, and $\xi_\alpha \to \xi_\alpha + \Delta$.

C. $J_a = 2 - J_b = 1$

The eight values of ξ_α are given by

$$\xi_1 = 0 = \xi_6, \quad \xi_2 = \tfrac{1}{2}(-\Delta + \delta_+), \quad \xi_3 = \tfrac{1}{2}(-\Delta + \delta_-), \quad \xi_4 = -\tfrac{1}{2}(\Delta + \delta_+), \quad \xi_5 = -\tfrac{1}{2}(\Delta + \delta_-), \quad \xi_7 = \tfrac{1}{2}(-\Delta + \delta), \quad \xi_8 = -\tfrac{1}{2}(\Delta + \delta), \tag{35a}$$

$$\text{where} \quad \delta_\pm^2 = \Delta^2 + 4\lambda_\pm, \; \delta = [\Delta^2 + 2(uu^* + vv^*)]^{1/2} \tag{35b}$$

$$\text{and} \quad \lambda_\pm = \tfrac{7}{12}(uu^* + vv^*) \pm \tfrac{1}{12}[25(uu^* + vv^*)^2 - 96uu^*vv^*]^{1/2}. \tag{35c}$$

Table III gives the unnormalized $D(n\alpha)$. For this table f_α and g_α are

$$f_\alpha = -\xi_\alpha(\xi_\alpha + \Delta) + v^*v + \tfrac{1}{6}u^*u + \tfrac{1}{6}v^*u, \tag{36a}$$

$$g_\alpha = \xi_\alpha(\xi_\alpha + \Delta) - u^*u - \tfrac{1}{6}v^*v - \tfrac{1}{6}u^*v. \tag{36b}$$

TABLE II. $D(n\alpha)$ for $J_a = 1$, $J_b = 1$. ξ_α are defined by Eq. (34).

n \ α	1	2	3	4	5	6
-1_a	v^*	$-u$	$-u$	0	0	0
$+1_a$	u^*	v	v	0	0	0
0_b	0	$2^{1/2}\xi_2$	$2^{1/2}\xi_3$	0	0	0
-1_b	0	0	0	$-u$	v^*	v^*
$+1_b$	0	0	0	$-v$	$-u^*$	$-u^*$
0_a	0	0	0	0	$2^{1/2}(\xi_2+\Delta)$	$2^{1/2}(\xi_3+\Delta)$

For those roots which contain $\lambda_\pm$, it is convenient to note that $\xi_\alpha(\xi_\alpha+\Delta)=\lambda_\pm$, and this is suggested by Eq. (14).

D. $J_a = 2$ - $J_b = 2$

The 10 roots ξ_α are

$$\xi_1 = 0; \quad \xi_2 = \tfrac{1}{2}(-\Delta+\delta_+) = \xi_7;$$

$$\xi_3 = \tfrac{1}{2}(-\Delta+\delta_-) = \xi_8; \quad \xi_4 = -\tfrac{1}{2}(\Delta+\delta_+) = \xi_9;$$

$$\xi_5 = -\tfrac{1}{2}(\Delta+\delta_-) = \xi_{10}; \quad \xi_6 = -\Delta, \tag{37a}$$

where $\delta_\pm^2 = \Delta^2 + 4\lambda_\pm$, (37b)

and $\lambda_\pm = \frac{5}{12}(uu^* + vv^*)$

$$\pm\tfrac{1}{12}[(uu^*+vv^*)^2 + 32uu^*vv^*]^{1/2}. \tag{37c}$$

Table IV gives the unnormalized $D(n\alpha)$. For this table

$$f_\alpha = -\xi_\alpha(\xi_\alpha+\Delta) + \tfrac{1}{3}vv^* + \tfrac{1}{2}uu^* - \tfrac{1}{2}v^*u\,, \tag{38a}$$

$$g_\alpha = \xi_\alpha(\xi_\alpha+\Delta) - \tfrac{1}{3}uu^* - \tfrac{1}{2}vv^* + \tfrac{1}{2}vu^*. \tag{38b}$$

For those roots which contain $\lambda_\pm$, $\xi_\alpha(\xi_\alpha+\Delta)=\lambda_\pm$. Also, it may be noted from the basic Eq. (10) or from Table IV that the entries for the dashed lines in Fig. 1(d), $\alpha = 6$ through 10, may be obtained from the entries for the solid lines, $\alpha = 1$ through 5, by changing $u \to -v^*$, $v \to -u^*$, and $\xi_\alpha \to \xi_\alpha + \Delta$.

E. Half-Integer Values of J

Tables for the half-integer values $J_a = \frac{1}{2} \leftrightarrow J_b = \frac{1}{2}$, $J_a = \frac{3}{2} \leftrightarrow J_b = \frac{1}{2}$, $J_a = \frac{3}{2} \leftrightarrow J_b = \frac{3}{2}$ are given in Appendix B.

IV. POLARIZATION AND INTENSITY OF PHOTON ECHOES

Since photon-echo experiments are performed in the saturation region, the resonance or $\Delta = 0$ solution can be used for a discussion of the salient features of the experimental research. The $J_a = 2 - J_b = 1$ transition is used as an example of the development given in this paper. Only minor modifications of the $2-1$ transition are needed to discuss the remaining transition with $J \leq 2$. Equation (30) gives the polarization of an atom subjected to two pulses of given polarization and duration. Since the stimulating radiation sets the phase of all the atoms in the sample, the electric field of the echo at the detector is proportional to $\vec{P}$ and the detector signal can be analyzed in terms of $\vec{P}$. Equation (22) can be used to obtain the matrix elements of the unitary matrices, and Eqs. (35) and (36) and Table III can be used to obtain the roots ξ_α and the $B(n\alpha)$ matrices. For $\Delta = 0$, these roots and matrices simplify and

$$\xi_2 = -\xi_4, \quad \xi_3 = -\xi_5, \quad \xi_7 = -\xi_8, \tag{39a}$$

TABLE III. $D(n\alpha)$ for $J_a = 2$, $J_b = 1$. ξ_α are defined by Eq. (35) and f_α and g_α by Eq. (36).

n \ α	1	2	3	4	5	6	7	8
-2_a	$-v^*v^*$			$-uf_\alpha$		0	0	0
0_a	$6^{1/2}u^*v^*$			$-(vf_\alpha+ug_\alpha)/6^{1/2}$		0	0	0
$+2_a$	$-u^*u^*$			$-vg_\alpha$		0	0	0
-1_b	0			$\xi_\alpha f_\alpha$		0	0	0
$+1_b$	0			$\xi_\alpha g_\alpha$		0	0	0
-1_a	0	0	0	0	0	$-v^*$	$-u$	$-u$
$+1_a$	0	0	0	0	0	u^*	$-v$	$-v$
0_b	0	0	0	0	0	0	$2^{1/2}\xi_7$	$2^{1/2}\xi_8$

TABLE IV. $D(n\alpha)$ matrix for $J_a = 2 \longleftrightarrow J_b = 2$. ξ_α are given by Eq. (37) and f_α and g_α by Eq. (38). The entries 2–5 and 7–10 differ only by the index α.

η \ α	1	2 3 4 5	6	7 8 9 10
-2_a	v^*v^*	$-uf_\alpha/3^{1/2}$	0	0
0_a	$(\frac{2}{3})^{1/2}u^*v^*$	$(vf_\alpha - ug_\alpha)/2^{1/2}$	0	0
$+2_a$	u^*u^*	$vg_\alpha/3^{1/2}$	0	0
-1_b	0	$\xi_\alpha f_\alpha$	0	0
$+1_b$	0	$\xi_\alpha g_\alpha$	0	0
-2_b	0	0	uu	$v^*g_\alpha^*/3^{1/2}$
0_b	0	0	$(\frac{2}{3})^{1/2}uv$	$(-u^*g_\alpha^* + v^*f_\alpha^*)/2^{1/2}$
$+2_b$	0	0	vv	$-u^*f_\alpha^*/3^{1/2}$
-1_a	0	0	0	$(\xi_\alpha + \Delta)g_\alpha^*$
$+1_a$	0	0	0	$(\xi_\alpha + \Delta)f_\alpha^*$

$$B(m_a 2) = B(m_a 4), \qquad B(m_b 2) = -B(m_b 4), \tag{39b}$$

$$B(m_a 7) = -B(m_a 8), \qquad B(m_b 7) = B(m_b 8). \tag{39c}$$

Let $\tau = t_1 - 0$ denote the duration of the first pulse, ξ_α the roots for the first pulse, and $B_1(n\alpha)$ the corresponding matrix. For the second pulse let $\tau' = t_3 - t_2$ denote the duration, ξ'_α the roots, and $B_2(n\alpha)$ the corresponding matrix. In order to further simplify the notation, let

$$F(rs) = \sum_{m_a} B_2(m_a r) B_1^*(m_a s), \tag{40a}$$

$$G(rs) = \sum_{m_b} B_2^*(m_b r) B_1(m_b s). \tag{40b}$$

With this notation the electric polarization of the echo can be written

$$\begin{aligned}
\vec{P}(t) = -2ie^{-i\omega_{ab}(t - t_3 + t_2 - t_1)}(\sigma_a - \sigma_b) \sum_{m_a m_b} (m_a|\vec{P}|m_b)^* \\
\times (B_2^*(m_b 2)B_2(m_a 2)[F(22)G(22)\sin 2\xi_2\tau + F(23)G(23)\sin 2\xi_3\tau](1 - \cos 2\xi'_2\tau') \\
+ B_2^*(m_b 3)B_2(m_a 3)[F(32)G(32)\sin 2\xi_2\tau + F(33)G(33)\sin 2\xi_3\tau](1 - \cos 2\xi'_3\tau') \\
+ \{2B_2^*(m_b 2)B_2(m_a 3)[F(22)G(32)\sin 2\xi_2\tau + F(23)G(33)\sin 2\xi_3\tau] \\
+ 2B_2^*(m_b 3)B_2(m_a 2)[F(32)G(22)\sin 2\xi_2\tau + F(33)G(23)\sin 2\xi_3\tau]\} \sin\xi'_3\tau' \sin\xi'_2\tau' \\
+ B_2^*(m_b 7)B_2(m_a 7)[F(77)G(77)\sin 2\xi_7\tau](1 - \cos 2\xi'_7\tau')) + \text{c.c.}
\end{aligned} \tag{41}$$

Before discussing this equation in detail, it is convenient to note that the part of the above equation with index 7 describes the polarization of the echo for the $J_a = 1 \leftrightarrow J_b = 0$ transition.

A. $J_a = 1 \leftrightarrow J_b = 0$ and $J_a = 1 \leftrightarrow J_b = 1$

The echo signal for the $J_a = 1 \leftrightarrow J_b = 0$ transition is given by the fifth term in Eq. (41) by changing the index 7 to index 2 and then using Eq. (33) for ξ_2 and Table I for the $B(n2)$ coefficients. The simple form of $\sin 2\xi_2\tau(1 - \cos 2\xi'_2\tau')$ indicates that the echo intensity is a maximum for a first pulse of strength $\eta = 2\xi_2\tau = \frac{1}{2}\pi$ and a second pulse of strength $\eta' = 2\xi'_2\tau' = \pi$. For $\Delta = 0$ and the definition of elliptic polarization given in Appendix A, $B(-1_a 2) = -\frac{1}{2}e^{i\varphi}\sin\gamma$, $B(+1_a 2) = +\frac{1}{2}e^{-i\varphi}\cos\gamma$, and $B(0_b 2) = 2^{-1/2}$. The polarization of the echo is given by

$$(-\hat{e}_+ e^{-i\varphi'}\cos\gamma' + \hat{e}_- e^{i\varphi'}\sin\gamma')a, \tag{42}$$

and has the same elliptical polarization as the second pulse. For two elliptic pulses the amplitude is proportional to

$$a=(e^{i(\varphi'-\varphi)}\sin\gamma\sin\gamma' + e^{-i(\varphi'-\varphi)}\cos\gamma\cos\gamma')\sin\eta(1-\cos\eta'), \qquad (43)$$

and the intensity is proportional to aa^*. If the first pulse is right circular $\gamma=\frac{1}{2}\pi$ and the second pulse is left circular $\gamma'=\pi$, there is no echo, and this result is in accord with the intuitive conclusion that there is no interference between right- and left-circular polarization. If both pulses are right circular $\gamma=\gamma'=\frac{1}{2}\pi$, the echo is right circular and the intensity depends only on the pulse strengths η and η'. If the first pulse is linearly polarized along $\hat{\theta}$ making angle $\varphi=\psi$ with the $\hat{x}$ axis, and the second pulse is linearly polarized along $\hat{\theta}'=\hat{x}$ or $\varphi'=0$, the echo is polarized along $\hat{x}$ and the intensity is proportional to $\cos^2\psi$. These results are in accord with other developments for linear polarization.[4]

The $J_a=1 \leftrightarrow J_b=1$ transition is similar to the above discussion for the solid lines shown in Fig. 1(b), and the terms which must be added for the dashed lines yield an equal contribution. The echo has the same elliptic polarization as the second pulse, and the intensity is given by Eq. (43). The roots $\xi_2=\xi_5$ are given by Eq. (34a) and for the same intensity are smaller by $2^{-1/2}$ than the $1 \leftrightarrow 0$ transition.

B. $J_a=2 \leftrightarrow J_b=1$

Equation (41) and Table III permit a detailed analysis for elliptically polarized pulses. Only the limiting cases of circular and linear pulses are considered in detail in this paper, and Table V gives the necessary $B(n\alpha)$ coefficients for linear polarization $\hat{\theta}$ at angle φ to $\hat{x}$ and for right-circular polarization. Coefficients which depend on $\pm 2_a$, $\pm 1_a$, and 0_a should be multiplied by the same phase factor for a given pulse, but this phase is not included since the product of phase factors is the same for all terms in Eq. (41). This phase can be taken outside the sum and can be ignored in the calculations. Elliptic polarization and angles φ and γ are discussed in Appendix A.

Let the first pulse be linearly polarized along $\hat{\theta}$, which makes angle $\varphi=\psi$ with $\hat{x}$, and let the duration of the first pulse be given by

$$2\xi_2\tau=\eta, \quad 2\xi_3\tau=2\xi_7\tau=(\tfrac{3}{4})^{1/2}\eta=\zeta\,. \qquad (44a)$$

Then let the second pulse be linearly polarized along $\hat{\theta}=\hat{x}$ or $\varphi'=0$ and let the duration of the second pulse be

$$2\xi_2'\tau'=\eta', \quad 2\xi_3'\tau'=2\xi_7'\tau'=(\tfrac{3}{4})^{1/2}\eta'=\zeta'. \qquad (44b)$$

The electric polarization of the echo, which is given by Eq. (41), can be written

$$\vec{\mathrm{P}}(t)=(\mathrm{const})\,e^{-i\omega_{ab}t}(-\hat{e}_+e^{-i\varphi''}+\hat{e}_-e^{+i\varphi''})G+\mathrm{c.\,c.}, \qquad (45)$$

where $$G^2=(a_1+a_2+a_5)^2+(a_3+a_4)^2\,, \qquad (46a)$$

and $$\tan\varphi''=-(a_3+a_4)/(a_1+a_2+a_5)\,. \qquad (46b)$$

Thus, the echo is linearly polarized with $\hat{\theta}''$ making an angle φ'' with the $\hat{x}$ direction or the direction of the second pulse. The dependence of the amplitude of the pulse G and the phase angle φ'' on ψ, η, and η' is given by the a_i or

$$a_1=[3^{-1/2}(1+3\cos2\psi)\cos\psi\sin\eta + 2\sin\psi\sin2\psi\sin\zeta\,](1-\cos\eta'),$$

TABLE V. $B(n\alpha)$ coefficients for $J_a=2 \leftrightarrow J_b=1$ transitions for linear polarization $\gamma=\frac{1}{4}\pi$ and right-circular polarization $\gamma=\frac{1}{2}\pi$. $\hat{\theta}$ makes angle φ with $\hat{x}$.

	$\gamma=\frac{1}{4}\pi$			$\gamma=\frac{1}{2}\pi$		
α	2	3	7	2	3	7
-2_a	$(\frac{3}{16})^{1/2}e^{i2\varphi}$	$\frac{1}{2}e^{i2\varphi}$	0	$2^{-1/2}e^{i\varphi}$	0	0
0_a	$-(\frac{1}{8})^{1/2}$	0	0	0	$2^{-1/2}e^{i\varphi}$	0
$+2_a$	$(\frac{3}{16})^{1/2}e^{-i2\varphi}$	$-\frac{1}{2}e^{-i2\varphi}$	0	0	0	0
-1_b	$-\frac{1}{2}e^{i\varphi}$	$-\frac{1}{2}e^{i\varphi}$	0	$-2^{-1/2}$	0	0
$+1_b$	$\frac{1}{2}e^{-i\varphi}$	$-\frac{1}{2}e^{-i\varphi}$	0	0	$-2^{-1/2}$	0
-1_a	0	0	$-\frac{1}{2}e^{i\varphi}$	0	0	$-2^{-1/2}e^{i\varphi}$
$+1_a$	0	0	$\frac{1}{2}e^{-i\varphi}$	0	0	0
0_b	0	0	$2^{-1/2}$	0	0	$2^{-1/2}$

$$a_2 = (3^{1/2}\sin\psi\sin 2\psi\sin\eta + 2\cos\psi\cos 2\psi\sin\zeta)(1-\cos\zeta'),$$

$$a_3 = [-(1+3\cos 2\psi)\sin\psi\sin\eta + (12)^{1/2}\cos\psi\sin 2\psi\sin\zeta]\sin\tfrac{1}{2}\eta'\sin\tfrac{1}{2}\zeta' \qquad (46c)$$

$$a_4 = [+2\cos\psi\sin 2\psi\sin\eta - (\tfrac{16}{3})^{1/2}\sin\psi\cos 2\psi\sin\zeta]\sin\tfrac{1}{2}\eta'\sin\tfrac{1}{2}\zeta',$$

$$a_5 = 2\cos\psi\sin\zeta(1-\cos\zeta').$$

The a_i coefficients are the sums of the 5 products which are given in Eq. (41) and are in the sequence in which the products occur. The intensity of the echo is proportional to G^2 and the quantity

$$G^2(\psi,\eta,\eta')/G^2(0,\eta_0,\eta_0'), \qquad (47)$$

and the angle φ'' can be compared with the data given in Table II of Ref. 4. Equations (45)–(47) provide an analytic expression for entries given in their table. It should be noted that the angle φ'' of the linear polarization of the echo $\hat{\theta}''$ moves in the opposite sense of the linear polarization $\hat{\theta}$ of the first pulse at angle ψ, and when $\psi=+90°$, $\varphi''=-90°$.

If the second pulse is right circular, the electric polarization of the echo in Eq. (41) has the form

$$\vec{P}(t) = \text{const}\, e^{-i\omega_{ab}t}[-\hat{e}_+(a_3) + \hat{e}_-(a_1+a_2+a_5)] + \text{c.c.}, \qquad (48)$$

where the a_i are the sequence of five products occurring in this equation. Since the a_i are complex, the output is elliptic for a general elliptic first pulse. If the first pulse is left circular and the second pulse right circular, all $a_i=0$ and there is no echo. If the first pulse is right circular and the second pulse is right circular, the echo is right circular with amplitude

$$a_1 = \sin\eta(1-\cos\eta'),$$

$$a_2 = 6^{-1/2}\sin 6^{-1/2}\eta\,(1-\cos 6^{-1/2}\eta'),$$

$$a_5 = \sin 2^{-1/2}\eta\,(1-\cos 2^{-1/2}\eta'), \qquad (49a)$$

$$a_3 = 0.$$

From Eq. (35) the ratio of the roots ξ_α are known and

$$\eta' = 2\xi_2'\tau', \quad \xi_3/\xi_2 = 6^{-1/2}, \quad \xi_7/\xi_2 = 2^{-1/2}, \qquad (49b)$$

and η is the same without primes. The intensity of the echo is proportional to $(a_1+a_2+a_5)^2$, and is a function of η and η'. Since the strength of the Clebsch-Gordan coefficients for the transitions -2_a–-1_b, -1_a–0_b, and 0_a–$+1_b$ are in the ratio $1:2^{-1/2}:6^{-1/2}$, the echo depends on three pulse strengths with the same ratios.

If the first pulse is linear with $\hat{\theta}$ at angle $\varphi=\psi$ and the second pulse is right circular, the echo is elliptic with

$$a_1 = e^{i\psi}[(\tfrac{3}{16})^{1/2}\sin\eta + \tfrac{1}{2}\sin(\tfrac{3}{4})^{1/2}\eta](1-\cos\eta'),$$

$$a_2 = e^{i\psi}[(\tfrac{1}{48})^{1/2}\sin\eta](1-\cos 6^{-1/2}\eta'),$$

$$a_5 = e^{i\psi}[(\tfrac{1}{2})^{1/2}\sin(\tfrac{3}{4})^{1/2}\eta](1-\cos 2^{-1/2}\eta') \qquad (50)$$

$$a_3 = e^{-i\psi}[(\tfrac{1}{8})^{1/2}\sin\eta - (\tfrac{1}{6})^{1/2}\sin(\tfrac{3}{4})^{1/2}\eta] \times \sin(\tfrac{1}{24})^{1/2}\eta'\sin\tfrac{1}{2}\eta',$$

and η is given by Eq. (44a). The ellipticity can be determined from

$$\tan\gamma = (a_1+a_2+a_5)/a_3^*, \qquad (51)$$

with $\hat{\theta}''$ along $\hat{\theta}$. Over-all echo intensity is given by $(a_1+a_2+a_5)(a_1^*+a_2^*+a_5^*)+a_3a_3^*$. The intensity $a_3a_3^*$ of the left-circular component in the echo is quite sensitive to the strength of the first pulse and is proportional to

$$[\sin\eta - (\tfrac{4}{3})^{1/2}\sin(\tfrac{3}{4})^{1/2}\eta]^2. \qquad (52)$$

It is zero at a pulse strength of $\eta=0$, reaches its first maximum at 193° of $(0.48)^2$, next minimum at 276°, second maximum of $(0.94)^2$ at 386°, etc. The left-circular component also vanishes for a second pulse strength of $\tfrac{1}{2}\eta'$ or $(\tfrac{1}{24})^{1/2}\eta'$ equal to a multiple of π.

If the first pulse is right circular and the second pulse is linear with $\hat{\theta}'=\hat{x}$, the electric polarization is of the form

$$P(t) = \text{const}\, e^{-i\omega_{ab}t}[-\hat{e}_+(a_1+a_2+a_5-a_3-a_4) + \hat{e}_-(a_1+a_2+a_3+a_4+a_5)] + \text{c.c.}, \qquad (53)$$

where

$$a_1 = (\sin\eta + (\tfrac{2}{3})^{1/2}\sin 6^{-1/2}\eta)(1-\cos\eta'),$$

$$a_2 = (\sin\eta)(1-\cos(\tfrac{3}{4})^{1/2}\eta'),$$

$$a_5 = (2^{1/2}\sin\tfrac{1}{2}\eta)(1-\cos(\tfrac{3}{4})^{1/2}\eta'), \qquad (54)$$

$$a_3+a_4 = [(\tfrac{25}{3})^{1/2}\sin\eta - 2^{1/2}\sin 6^{-1/2}\eta]$$

$\times \sin\frac{1}{2}\eta' \sin(\frac{3}{16})^{1/2}\eta'$.

The ellipticity can be determined from

$$\tan\gamma'' = (a_1+a_2+a_3+a_4+a_5)/(a_1+a_2+a_3-a_4-a_5). \quad (55)$$

A distinctive feature is the absence of $\hat{y}$ polarization for $a_3+a_4=0$, and this occurs for a first-pulse strength of $\eta=154°$, $561°$, or for a second-pulse strength of $\frac{1}{2}\eta'$ or $(\frac{3}{16})^{1/2}\eta'$ equal to a multiple of π.

Although it does not seem important for experiments, the intensity can be brought to the same scale by multiplying Eq. (46c) by $\frac{1}{32}$, Eqs. (49a) and (50) by $\frac{1}{8}$, and Eq. (54) by $\frac{1}{4}$.

C. $J_a = 2 \leftrightarrow J_b = 2$

The electric polarization describing the echo for the $J_a = 2 - J_b = 2$ transition can be discussed in terms of Eq. (41). The first four terms in Eq. (41) describe the contribution of the solid lines in Fig. 1(d). The contribution of the dashed lines is given by adding four similar terms with with index 2 changed to 7 and index 3 changed to 8. The fifth term in Eq. (41) is omitted and a total of eight terms contribute to $\vec{P}$. Since the roots $\xi_2=-\xi_4=\xi_7=-\xi_9$ and $\xi_3=-\xi_5=\xi_8=-\xi_{10}$, only two pulse strengths are important. The ratio ξ_3/ξ_2 depends on the ellipticity of the pulse and is given in detail by Eq. (37).

If the first pulse is linear with $\hat{\theta}$ at angle $\varphi=\psi$, the second pulse is linear with $\hat{\theta}'=\hat{x}$, and the pulse strengths are written

$$2\xi_2\tau=\eta, \quad 2\xi_3\tau=\zeta=\tfrac{1}{2}\eta, \quad (56)$$

the electric polarization can be written in the form of Eq. (45). The amplitude G is given by

$$G^2=(a_1+a_2)^2+a_3^2, \quad (57a)$$

and the phase angle φ'' by

$$\tan\varphi''=a_3/(a_1+a_2), \quad (57b)$$

where
$$a_1=[(3+\cos2\psi)\cos\psi\sin\eta + 2\sin\psi\sin2\psi\sin\zeta](1-\cos\eta');$$
$$a_2=[\sin\psi\sin2\psi\sin\eta + 2\cos\psi\cos2\psi\sin\zeta](1-\cos\zeta'),$$
$$a_3=\{[3+\cos2\psi)\sin\psi + 2\cos\psi\sin2\psi]\sin\eta - 2[\cos\psi\sin2\psi + 2\sin\psi\cos2\psi]\sin\zeta\}\sin\tfrac{1}{2}\eta'\sin\tfrac{1}{2}\zeta', \quad (58)$$

a_1 includes the first and fifth terms in the sum, a_2 includes the second and sixth, and a_3 includes the third, fourth, seventh, and eighth terms. The echo is linearly polarized along $\hat{\theta}''$ at angle φ'' with $\hat{x}$. φ'' increases in the same sense as ψ, and at $\psi=90°$ the angle $\varphi''=+90$. Intensity is proportional to G^2. The analytic expression given above for intensity and echo polarization are in agreement with the numerical data of Ref. 4.

If the first pulse is right circular and the second pulse is linear along $\hat{x}$, then the echo polarization is of the form given by Eq. (53) with

$$a_1=(6^{1/2}\sin\eta+\sin\zeta)(1-\cos\eta'),$$
$$a_2=\sin\zeta(1-\cos\zeta'), \quad (59)$$
$$a_3=(6^{1/2}\sin\eta+\sin\zeta)\sin\tfrac{1}{2}\zeta'\sin\tfrac{1}{2}\eta'.$$

The pulse strengths for the right-circular pulse are

$$2\xi_2\tau=\eta, \quad 2\xi_3\tau=\zeta=(\tfrac{2}{3})^{1/2}\eta, \quad (60)$$

and the pulse strength for the linear pulse is given by Eq. (56). The output is elliptic with

$$\tan\gamma=(a_1+a_2+a_3)/(a_1+a_2-a_3). \quad (61)$$

At a pulse strength such that

$$6^{1/2}\sin\eta+\sin(\tfrac{2}{3})^{1/2}\eta=0, \quad (62)$$

the coefficients a_1 and a_3 are zero, and the echo pulse becomes linear along the second pulse.

If the first pulse is linear along $\hat{\theta}$ at angle $\varphi=\psi$ to $\hat{x}$ and the second pulse is right circular, then the echo polarization is of the form given by Eq. (48) with

$$a_1=e^{i\psi}\{(\tfrac{3}{4})^{1/2}\sin\eta\}(1-\cos\eta'),$$
$$a_2=e^{i\psi}\{(\tfrac{1}{12})^{1/2}\sin\eta+(\tfrac{1}{3})^{1/2}\sin\zeta\}(1-\cos\zeta'), \quad (63)$$
$$a_3=e^{-i\psi}\{(\tfrac{1}{2})^{1/2}\sin\eta-2^{1/2}\sin\zeta\}\sin\tfrac{1}{2}\zeta'\sin\tfrac{1}{2}\eta'.$$

η and ζ are given by Eq. (56) for linear polarization and η' and ζ' by Eq. (60) for circular polarization. The echo is elliptic with

$$\tan\gamma=(a_1+a_2)/a_3^*, \quad (64)$$

and with $\hat{\theta}''=\hat{\theta}$ along the linear polarization of the first pulse. The intensity of the right-circular component is proportional to $(a_1+a_2)(a_1^*+a_2^*)$, and the intensity of the left circular is proportional to $a_3a_3^*$. The intensity of the left-circular component of the echo varies with the pulse strength of the first pulse as

$$(\sin\eta-2\sin\tfrac{1}{2}\eta)^2, \quad (65)$$

and the intensity is zero for weak pulse strengths, reaches its first maximum of $(-2.6)^2$ at $\eta = 240°$, returns to zero at $\eta = 360$, and reaches its second maximum of $(+2.6)^2$ at $\eta = 480°$, etc. This variation in intensity with pulse strength should identify a $2-2$ transition. The output of the left-circular component is also zero for a second-pulse strength of $\frac{1}{2}\eta'$ or $(\frac{2}{3})^{1/2}\eta'$ equal to an integer times π.

A right-circular pulse followed by a right-circular pulse yields a right-circular echo. The amplitude of the echo is given by

$$a = 2^{-1/2}\sin\eta(1-\cos\eta') + 3^{-1/2}\sin\zeta(1-\cos\zeta'), \quad (66)$$

where η, η', ζ, and ζ' are given by Eq. (60).

The equations can all be brought to the same scale by multiplying Eqs. (58), (59), and (63) by $\frac{1}{8}$ and Eq. (66) by $\frac{1}{4}$.

D. $J_a = \frac{1}{2} \leftrightarrow J_b = \frac{1}{2}$

The echo electric polarization is given for arbitrary polarization of both pulses by

$$\vec{\mathrm{P}}(t) = \mathrm{const}\, e^{-i\omega_{ab}t}\{-\hat{e}_+ a_1 + \hat{e}_- a_2\} + \mathrm{c.c.} \quad (67)$$

where

$$a_1 = \pm e^{i\varphi}\sin\zeta(1-\cos\zeta'),$$
$$a_2 = \pm e^{i\varphi}\sin\eta\,(1-\cos\eta'), \quad (68)$$

and a_1 has the sign of $\cos\gamma$, and a_2 has the sign of $\sin\gamma$. The roots $\eta = 2\zeta_1\tau$ and $\zeta = 2\xi_3\tau$ are given by Eq. (B-1). The output is elliptical with $\hat{\theta}''$ at angle $\varphi'' = -\varphi$ with the $\hat{x}$ axis, and the eccentricity of the ellipse is

$$\tan\gamma'' = \pm[\sin\zeta(1-\cos\zeta')] \times[\sin\eta(1-\cos\eta')]^{-1}, \quad (69)$$

where the square brackets have the sign of $\tan\gamma$. This is a function of the pulse strength only and depends on both pulses. Two linear pulses, $\gamma = \gamma' = \frac{1}{4}\pi$ and $\zeta = \eta$, produce a linear echo along $\hat{\theta}''$ at angle $-\psi$ with $\hat{x}$. $\zeta = 0$ for a right-circular pulse, and the echo is right circular if either the first or second pulse is right circular.

E. $J_a = \frac{3}{2} \leftrightarrow J_a = \frac{1}{2}$

The echo with arbitrary polarizations of the two pulses is given by Eq. (67) with

$$a_1 = [\tfrac{1}{3}b_1\sin\eta(1-\cos\eta') + b_2\sin\zeta(1-\cos\zeta')]\cos\gamma',$$

$$a_2 = [b_1\sin\eta(1-\cos\eta') + \tfrac{1}{3}b_2\sin\zeta(1-\cos\zeta')]\sin\gamma', \quad (70)$$

and

$$b_1 = (e^{-i\varphi}\sin\gamma\sin\gamma' + \tfrac{1}{3}e^{i\varphi}\cos\gamma\cos\gamma') \times[(1-\tfrac{2}{3}\cos^2\gamma')(1-\tfrac{2}{3}\cos^2\gamma)^{1/2}]^{-1}, \quad (71)$$

$$b_2 = (e^{i\varphi}\cos\gamma\cos\gamma' + \tfrac{1}{3}e^{-i\varphi}\sin\gamma\sin\gamma') \times[(1-\tfrac{2}{3}\sin^2\gamma')(1-\tfrac{2}{3}\sin^2\gamma)^{1/2}]^{-1}.$$

$\eta = 2\xi_2\tau$, $\zeta = 2\xi_5\tau$, and ξ_2 and ξ_5 are given by Eq. (B2). The angle φ'' of $\hat{\theta}''$ with $\hat{x}$ and the eccentricity γ'' of the echo are given by

$$e^{-i\varphi''}\cos\gamma'' = a_1/(a_1a_1^* + a_2a_2^*) \quad (72a)$$

and

$$\tan\gamma'' = a_1/a_2^*, \quad (72b)$$

and intensity is given by $a_1a_1^* + a_2a_2^*$. If the first pulse is linearly polarized with $\hat{\theta}' = \hat{x}$, the angles $\gamma = \gamma' = \frac{1}{4}\pi$, and the pulse strength $\eta = \zeta$ and $\eta' = \zeta'$. $a_1 = a_2^*$, and the echo is linearly polarized along $\hat{\theta}''$ at angle φ'':

$$\tan\varphi'' = -\tfrac{1}{4}\tan\varphi. \quad (73)$$

This is in agreement with Ref. 4. If the second pulse is right circular $\gamma' = \frac{1}{2}\pi$, the echo is right circular. If the first pulse is right circular $\gamma = \frac{1}{2}\pi$ and the second pulse is linear $\gamma' = \frac{1}{4}\pi$, the echo polarization is proportional to

$$-\hat{e}_+(\sin\eta + 3^{1/2}\sin 3^{-1/2}\eta) + \hat{e}_-(3\sin\eta + 3^{-1/2}\sin 3^{-1/2}\eta), \quad (74)$$

and is elliptic with $\hat{\theta}'' = \hat{x}$. The eccentricity γ'' depends entirely on the strength of the first pulse. Varying the strength of the first pulse allows the polarization of the echo to be changed from right circular to left circular, etc. The echo radiation is entirely left circular at $\eta = 191.5°$ and entirely right circular at $\eta = 251°$.

F. $J_a = \frac{3}{2} \leftrightarrow J_b = \frac{3}{2}$

A linear pulse $\hat{\theta}$ at angle $\varphi = \psi$ followed by a linear pulse with $\varphi' = 0$ produces an echo polarization described by Eq. (45) with the echo linearly polarized along $\hat{\theta}''$ at an angle φ'':

$$\tan\varphi'' = -(a_3 + a_4)/(a_1 + a_2) \quad (75a)$$

and intensity

$$G^2 = (a_1 + a_2)^2 + (a_3 + a_4)^2. \quad (75b)$$

The a_i are given by

$$a_1 = [15\cos\psi + \cos 3\psi)\sin\eta + 6\sin\psi\sin 2\psi\sin\zeta](1-\cos\eta'),$$

$$a_2 = \tfrac{1}{3}[(6\sin\psi\sin 2\psi\,\sin\eta$$

$$+ (7\cos\psi + 9\cos 3\psi)\sin\zeta](1-\cos\zeta'), \qquad (76)$$

$$a_3 = -4[(5\sin\psi + \sin 3\psi)\sin\eta$$

$$+ (\sin\psi - 3\sin 3\psi)\sin\zeta]\sin\tfrac{1}{2}\eta'\sin\tfrac{1}{2}\zeta',$$

$$a_4 = \tfrac{2}{3}[(9\sin\psi - 3\sin 3\psi)\sin\eta$$

$$+ (5\sin\psi + 9\sin 3\psi)\sin\zeta](1-\cos\zeta'),$$

and $2\xi_1\tau = \eta$ and $2\xi_2\tau = \zeta = \frac{1}{3}\eta$. (77)

If the first pulse is right circular and the second pulse is right circular, the echo is right circular with amplitude

$$2\sin\zeta(1-\cos\zeta') + (\tfrac{4}{3})^{1/2}\sin\eta(1-\cos\eta'), \qquad (78)$$

where $2\xi_5\tau = \eta, \quad 2\xi_1\tau = \zeta = (\frac{3}{4})^{1/2}\eta$. (79)

There is no echo if a left-circular follows a right-circular pulse.

If the first pulse is linear with $\varphi = 0$ or $\hat{\theta}$ along $\hat{x}$ and the second pulse is right circular, the echo polarization is of the form of Eq. (67) with

$$a_1 = (-\sin\eta + 3\sin\zeta)(1-\cos\zeta'),$$

$$a_2 = -3(\sin\eta + \sin\zeta)(1-\cos\zeta') \qquad (80)$$

$$+(-3\sin\eta + \sin\zeta)(1-\cos\eta').$$

η and ζ are given by Eq. (77) and η' and ζ' by Eq. (79). The echo is right-circularly polarized for all second-pulse strengths for a first-pulse strength with η such that

$$-\sin\eta + 3\sin\tfrac{1}{3}\eta = 0, \qquad (81)$$

or for all first-pulse strengths where η is an integer times 270°. It is also right circular for a second-pulse strength of $\zeta' = 2\pi$, 4π, etc.

The echo for a right-circular first pulse followed by a linear second pulse with $\hat{\theta}' = \hat{x}$ is determined by Eq. (67) with

$$a_1 = (b_1 + b_2 - b_3 - 3b_4); \quad a_2 = (b_1 - b_2 + 3b_3 + b_4), \qquad (82)$$

and b_i coefficients of

$$b_1 = [3\sin\eta + (12)^{1/2}\sin\zeta](1-\cos\eta'),$$

$$b_2 = 4[\sin\eta + (\tfrac{4}{3})^{1/2}\sin\zeta]\sin\tfrac{1}{2}\zeta'\sin\tfrac{1}{2}\eta', \qquad (83)$$

$$b_3 = (\tfrac{4}{3})^{1/2}\sin\zeta(1-\cos\zeta'),$$

$$b_4 = \tfrac{1}{3}\sin\eta(1-\cos\zeta').$$

The roots η and ζ are given by Eq. (79) and η' and ζ' by Eq. (77). This pulse is elliptic with $\hat{\theta}'' = \hat{x}$ and an eccentricity given by $\tan\gamma'' = a_2/a_1$. It would appear very difficult to find pulse durations which would make the echo pure circular or pure linear.

V. ELECTRIC POLARIZATION STIMULATED BY SATURATING RADIATION

The electric polarization of an atom or molecule stimulated by radiation is given by Eq. (26). It is again assumed in this section that all $|a)$ states are equally probable, and $(m_a|\sigma|m_a) = \sigma_a$. Stimulated absorption and stimulated emission differ only by a negative sign, and the electric polarization is proportional to $\sigma_a - \sigma_b$ in a system of molecules experiencing the same stimulation. Thus, the electric polarization stimulated by Eq. (2) is

$$\vec{P}(t) = (\sigma_a - \sigma_b)e^{-i\omega t}\sum_{\substack{m_a m_a' \\ m_b}}\sum_{\alpha,\alpha'}(m_a|\vec{P}|m_b)^* e^{i(\xi_\alpha - \xi_{\alpha'})t}B^*(m_b\alpha)B(m_a'\alpha)B^*(m_a'\alpha')B(m_a\alpha') + \text{c. c.} \qquad (84)$$

If the root $\xi_\alpha = 0$ occurs, the $B(m_b\alpha)$ coefficients are zero. Since the sum over m_a' of $B^*(m_a'\alpha) \times B(m_a'0)$ is zero, this root can be omitted in the sum over α and α'. For $\xi_\alpha = -\Delta$, the $B(m_a\alpha)$ coefficients are zero and this root can also be omitted from the sum. This polarization is modulated at the various difference frequencies given by $\xi_\alpha - \xi_{\alpha'}$, and these appear as sidebands on each side of the stimulating frequency. It is interesting to note that with the axis of quantization $\hat{z}$ along the direction of the stimulating radiation $\hat{k}$ the electric polarization transverse to $\hat{k}$ is always of the form

$$\vec{P}(t) = \text{const}\, e^{-i\omega t}[-\hat{e}_+(vf) + \hat{e}_-(ug)] + \text{c. c.}, \qquad (85)$$

where f and g are functions of Δ, uu^*, and vv^*, and the various $e^{i(\xi_\alpha - \xi_{\alpha'})t}$. This indicates that even in the extreme saturation region the right-circular component of $\vec{P}(t)$ or the coefficient of $\hat{e}_-$ is proportional to the right-circular component of incident radiation u. g is modulated in time and is a function of the polarization and strength of the radiation field. This relationship implies that right-circular component stimulates only right-circular radiation and left-circular component stimulates only left-circular radiation. The degree of stimulation and the modulation induced by the stimulation depend on the intensity of both polarizations. In weak radiation fields this proportionality is expected. It is not intuitively obvious for strong radiation fields. The above conclusions are valid for $J \leq 2$, but it would seem that a general proof probably exists. Dienes[10] in his discussion of a damped system at resonance was able to show for arbitrary J that $P_r \propto E_r$ and $P_l \propto E_l$. This simple relationship is also true for the resolution of the polarization along $\hat{z}$ alone and $P_z \propto E_z$. The choice of the axis of quantization along direction of the radiation field and the resolution into right- and left-circular polarization seems to yield the simplest relationship between the electric polarization and the radiation for arbitrary polarization of the incident radiation. If other canonically conjugate states of polarization $\hat{u}_1$ and $\hat{u}_2$ are selected, and if the incident radiation has polarization $\hat{u}_1$, the electric polarization is composed of both $\hat{u}_1$ and $\hat{u}_2$ for saturating radiation.

The form of Eq. (85) clearly illustrates that the stimulated electric polarization of the molecule generates radiation of a different polarization than the stimulating radiation. The intensity of right-circular polarization in the stimulating radiation is proportional to uu^*, and the intensity in the stimulated radiation is proportional to $(uu^*)(gg^*)$. A measure of the degree of elliptic polarization of the incident radiation is

$$(uu^* - vv^*)/(uu^* + vv^*) \tag{86a}$$

and of the stimulated radiation is

$$(uu^*gg^* - vv^*ff^*)/(uu^*gg^* + vv^*ff^*) \quad . \tag{86b}$$

It is just this difference which introduces the anomalous polarization into laser or other molecular systems which are saturated. For weak radiation, $f = g$ and there is no difference in the degree of polarization. For $J = 1$, $f = g$ and the degree of polarization is the same in both weak and strong stimulating radiation. For $J > 1$, $g \neq f$, in general, and the degree of polarization of the stimulated radiation is different from the stimulating radiation.

The coefficients f and g in Eq. (85) are functions of Δ, uu^*, vv^*, and time, and the electric polarization $\vec{P}$ of the molecule is modulated. The stimulated radiation occurs at sideband frequencies, and such sidebands are expected for monochromatic radiation. Other processes occur which limit the lifetime of a state, and the sidebands have a finite duration and, therefore, have a spectral distribution. Fourier components of the stimulated polarization can occur at the radiation frequency ω. Other forward scattered components tend to change the spectral distribution of a pulse traveling through a medium.

Damping[11] has not been included in the calculation, but can be considered as another process limiting the lifetime of interaction with the incident radiation. A suitable average will again show many features of the polarization given by Eq. (84). The earlier calculations[1,2] used a damped density-matrix procedure and perturbation theory. Effects of saturation were apparent in the third-order term in the density matrix or in the third-order term in the unitary matrix. A similar expansion of the unitary matrix of this paper to third order or the $B(n\alpha)$ coefficients to third order would yield a combination of the coefficients of the same type as occurred in these earlier discussions. When perturbation theory is no longer appropriate, then the full discussion of this paper can be used. At resonance, $\Delta = 0$, the results will be similar to those of Dienes[10] for a phenomenologically damped atom, but will differ by the modulation or sidebands. For finite pulses the conclusions are quite similar. Dienes[10] also notes that for strong saturating radiation at resonance pure-circular or pure-linear radiation do not change their polarization during amplification. Furthermore, the addition of a small amount of the conjugate state of polarization in the incident radiation yields a lesser degree of this polarization in the stimulated radiation. This information is included in the coefficients f and g in this paper for $\Delta = 0$. It is not clear without a numerical calculation that these conclusions remain valid for $\Delta \neq 0$. Calculations for $\Delta \neq 0$ are being pursued but are not reported in this paper.

DISCUSSION

The direct algebraic solution of the time-dependent Schrödinger equation for a harmonic perturbation of finite duration leads to a set of modes and to a time-evolution operator which is a function of the eigenvalues $\xi(n\alpha)$ and eigenvectors $B(n\alpha)$ of these modes. For intense radiation or for radiation near resonance, these modes keep the time-evolution operator unitary. The stimulated electric polarization of a molecule which follows from this unitary time-evolution operator never has a magnitude which is greater than the matrix element of the electric-dipole operator. As the radiation field increases in intensity the stimulated electric polarization is modulated in time at frequencies which are a function of the eigenvalues of the modes. These eigenvalues depend on both the intensity and polarization of the incident radiation. Stimulated electric polarization generates a radiation field and can in a sense be regarded as a scattering amplitude. The amplitude or stimulated electric field is given by $\vec{P}$ and has a magnitude and polarization which depends on both the eigenvalues and eigenvectors.

Most of the anomalous features of radiation, which is stimulated by saturating radiation, are introduced by the unitary requirement. This is apparent in the rotation of the electric polarization in the two state problem considered by Feynman, Vernon, and Hellwarth.[12] Their approach keeps the magnitude of the electric-dipole constant. A similar argument could be made for the magnetization in the second rotation used in Ref. 7. From an entirely different point of view, Goldberger and Watson[13] in their analysis of the general theory of resonant scattering from long-lived virtual states find that the requirement that the S matrix be unitary and analytic has very significant effects on the scattering amplitude and cross section. In their discussion, an interaction described by the packet $\frac{1}{2}\Gamma \exp[-(\frac{1}{2}\Gamma - i\omega_0)t]$ has its maximum cross section for $\omega = \omega_0$, and the packet $\Gamma \exp[-(\frac{1}{2}\Gamma - i\omega_0)t]$ has a zero cross section at $\omega = \omega_0$. This latter packet has maximum in the cross section on each side of ω_0. Increasing the strength to $\frac{1}{2}r\Gamma$ causes the cross section to be a maximum at odd r and zero for even r at resonance or $\omega = \omega_0$. Although the authors note no examples of this deviation from the Breit-Wigner cross section occur, the oscillating character with increasing strength is similar to the oscillation caused by the single mode occurring in a two-state problem for a pulse whose duration is proportional to Γ.

Damping or the finite lifetime of the atomic or molecular states are not included in the theory. In a two-state problem, the phenomenological method introduced by Block[14] is most frequently used. For problems with a greater number of states, a truncated density matrix with phenomenological damping[1,2,15,16] has been quite convenient. Neither of these approaches are directly applicable to the results of this paper, but it would seem that the maximum amount of interference or wave aspects which can be stimulated should occur for the development given in this paper. Damping and other collision processes can only degrade these effects. Damping can be incorporated into the results of this paper as a perturbation. This perturbation must be formulated to form perturbed $B(n\alpha)$ eigenvectors in terms of the unperturbed $B(n\alpha)$.

No distinction appears in the development between stimulated emission and absorption, and the conclusions are equally valid for both cases. Phenomenon, such as the self-induced transparency of Hahn and McCall,[17] are inherent in the theory since the modulation of the polarization introduces the possibility of anomalous stimulated effects. For the higher values of $J > 1$, more than one mode occurs and it is no longer possible to generate a 2π pulse for all modes. Since the modes have different strengths, it would seem that the transparency should be between $\frac{1}{2}$ and 1 for $J > 1$. The research reported by Patel and Slusher[18] on self-induced transparency in SF_6 suggests that complete transparency is approached at high intensity. If J values greater than one are involved, this result is not in accord with the simple aspects which are suggested in this paper. Pulse shape, damping, and perturbing interactions may be sufficient to alter the character of the multiple modes to a single effective mode, but this is not obvious.[19]

Since photon-echo experiments maximize the wave aspect or interference aspect in the time domain, the processes are well described by the use of a sequence of time-evolution operators of the type given in this paper. Anomalous polarization effects in the stimulated radiation are suggested and it should be possible to experimentally observe these effects. Linear-circular or circular-linear may be more useful than the previously discussed[4] linear-linear sequence.

The development in this paper is suitable for stimulated effects with either $(\sigma_a - \sigma_b) \lesseqgtr 0$ and should apply to stimulated absorption as well as emission. Echo and selftransparency experiments are normally made for an absorbing medium and anomalous polarization in a lasing medium. For saturating radiation, the electric polarization differs only by a sign, and the stimulated aspects must be the same in either absorbing or lasing material. The two problems differ in the subsequent nonlinear problem for the growth or decay of the pulse.[20]

APPENDIX A

The polarization vector $\hat{u}$ of the radiation can be given in terms of unit vectors transverse to the direction of the radiation $\hat{k}$. The simplest of these are the linear polarization vectors $\hat{\theta}$ and $\hat{\varphi}$, which can be written in terms of the spherical basis vectors as

$$\hat{\theta} = 2^{-1/2}(-\hat{e}_+ e^{-i\varphi} + \hat{e}_- e^{i\varphi})\cos\theta - \hat{e}_0 \sin\theta, \quad \text{(A1)}$$

$$\hat{\varphi} = i2^{-1/2}(\hat{e}_+ e^{-i\varphi} + \hat{e}_- e^{i\varphi}) . \quad \text{(A2)}$$

θ, φ are the orientation angles of $\hat{k}$ relative to $\hat{z}$ in spherical polar coordinates. For $\theta = \varphi = 0$, $\hat{\theta} = \hat{x}$, $\hat{\varphi} = \hat{y}$. Right- and left-circular polarization are related to the spherical basis vectors by

$$\hat{u}_r = \tfrac{1}{2}[+\hat{e}_+(1 \mp \cos\theta)e^{-i\varphi} + \hat{e}_- (1 \pm \cos\theta)e^{i\varphi}]$$
$$\mp 2^{-1/2}\hat{e}_0 \sin\theta, \quad \text{(A3)}$$

and $\hat{u}_l$ is given by the lower signs.

A unit polarization vector $\hat{u}$ of the form

$$\hat{u} = \hat{\theta}\cos\tfrac{1}{2}\beta - i\varphi\sin\tfrac{1}{2}\beta$$

represents linear polarization along $\hat{\theta}$ with $\beta = 0$, right circular with $\beta = \frac{1}{2}\pi$, linear along $\hat{\varphi}$ with $\beta = \pi$, left circular with $\beta = \frac{3}{2}\pi$, etc.

For $\theta = 0$, this unit polarization vector is given in terms of the spherical basis vectors as

TABLE VI. $D(n\alpha)$ for $J_a=\frac{1}{2}\longleftrightarrow J_b=\frac{1}{2}$. ξ_α are defined by Eq. (B1).

$n \backslash \alpha$	1	2	3	4
$(-\frac{1}{2})_a$	$-(\frac{2}{3})^{1/2}u$	$-(\frac{2}{3})^{1/2}u$	0	0
$(+\frac{1}{2})_b$	ξ_1	ξ_2	0	0
$(+\frac{1}{2})_a$	0	0	$(\frac{2}{3})^{1/2}v$	$(\frac{2}{3})^{1/2}v$
$(-\frac{1}{2})_b$	0	0	ξ_3	ξ_4

$$\hat{u}=(\hat{e}_- e^{i\varphi}\sin\gamma-\hat{e}_+e^{-i\varphi}\cos\gamma), \tag{A4}$$

where $\gamma=\frac{1}{2}\beta+\frac{1}{4}\pi$. Equations (18) and (20) resolve the incident wave into spherical basis vectors, and for $\theta=0$ or $\hat{k}$ along $\hat{z}$, the amount of right- and left-circular components in the incident wave are

$$(\hat{e}_-^*\circ\hat{u})=e^{i\varphi}\sin\gamma=u/F, \tag{A5a}$$

$$(\hat{e}_+^*\circ\hat{u})=-e^{-i\varphi}\cos\gamma=v/F, \tag{A5b}$$

where F is a complex amplitude which is related to the matrix element and amplitude A in Eq. (20). With this notation, $\gamma=\frac{1}{4}\pi$ linear along $\hat{\theta}$, $\gamma=\frac{1}{2}\pi$ right circular, and $\gamma=\pi$ left circular.

APPENDIX B

1. $J_a=\frac{1}{2}\leftrightarrow J_b=\frac{1}{2}$

The $J_a=\frac{1}{2}\leftrightarrow J_b=\frac{1}{2}$ problem divides into the two independent parts shown in Fig. 2(a) by the solid and dashed lines. Roots ξ_α for the solid line are

$$\xi_1=\tfrac{1}{2}(-\Delta+\delta),\quad \xi_2=\tfrac{1}{2}(-\Delta-\delta), \tag{B1a}$$

$$\delta=(\Delta^2+\tfrac{8}{3}uu^*)^{1/2}, \tag{B1b}$$

and for the dashed line are

$$\xi_3=\tfrac{1}{2}(-\Delta+\delta'),\quad \xi_4=\tfrac{1}{2}(-\Delta-\delta'), \tag{B1c}$$

$$\delta'=(\Delta^2+\tfrac{8}{3}vv^*)^{1/2}. \tag{B1d}$$

An unnormalized $D(n\alpha)$ matrix is given in Table VI.

2. $J_a=\frac{3}{2}-J_b=\frac{1}{2}$

Roots ξ_α for the solid lines are

$$\xi_1=0,\quad \xi_2=\tfrac{1}{2}(-\Delta+\delta),\quad \xi_3=\tfrac{1}{2}(-\Delta-\delta), \tag{B2a}$$

$$\delta=(\Delta^2+4uu^*+\tfrac{8}{3}vv^*)^{1/2}, \tag{B2b}$$

and for the dashed lines are

$$\xi_4=0,\quad \xi_5=\tfrac{1}{2}(-\Delta+\delta'),\quad \xi_6=\tfrac{1}{2}(-\Delta-\delta'), \tag{B2c}$$

$$\delta'=\{\Delta^2+4vv^*+\tfrac{8}{3}uu^*\}^{1/2}. \tag{B2d}$$

An unnormalized $D(n\alpha)$ matrix is given in Table VII.

3. $J_a=\frac{3}{2}\leftrightarrow J_b=\frac{3}{2}$

Roots ξ_α for solid lines in Fig. 2(c) are

$$\begin{aligned}&\xi_1=\tfrac{1}{2}(-\Delta+\delta_+),\quad \xi_2=\tfrac{1}{2}(-\Delta+\delta_-),\\ &\xi_3=\tfrac{1}{2}(-\Delta-\delta_+),\quad \xi_4=\tfrac{1}{2}(-\Delta-\delta_-),\end{aligned} \tag{B3a}$$

and for dashed lines are

$$\begin{aligned}&\xi_5=\tfrac{1}{2}(-\Delta+\delta_+'),\quad \xi_6=\tfrac{1}{2}(-\Delta+\delta_-'),\\ &\xi_7=\tfrac{1}{2}(-\Delta-\delta_+'),\quad \xi_8=\tfrac{1}{2}(-\Delta-\delta_-'),\end{aligned} \tag{B3b}$$

where $$\delta_\pm=(\Delta^2+4\lambda_\pm)^{1/2}, \tag{B3c}$$

$$\lambda=\tfrac{2}{5}uu^*+\tfrac{4}{15}vv^*\pm\tfrac{4}{15}[(vv^*)^2+3vv^*uu^*]^{1/2}. \tag{B3d}$$

Primed quantities are given by changing $u\rightarrow-v^*$, $v\rightarrow u^*$. Also,

TABLE VII. $D(n\alpha)$ for $J_a=\frac{3}{2}\longleftrightarrow J_b=\frac{1}{2}$. ξ_α are defined by Eq. (B2).

$n \backslash \alpha$	1	2	3	4	5	6
$(-\frac{3}{2})_a$	$(\frac{1}{3})^{1/2}v^*$	$-u$	$-u$	0	0	0
$(+\frac{1}{2})_a$	$-u^*$	$-(\frac{1}{3})^{1/2}v$	$-(\frac{1}{3})^{1/2}v$	0	0	0
$(-\frac{1}{2})_b$	0	ξ_2	ξ_3	0	0	0
$(+\frac{3}{2})_a$	0	0	0	$(\frac{1}{3})^{1/2}u^*$	$-v$	$-v$
$(-\frac{1}{2})_a$	0	0	0	v^*	$-(\frac{1}{3})^{1/2}u$	$-(\frac{1}{3})^{1/2}u$
$(+\frac{1}{2})_b$	0	0	0	0	ξ_5	ξ_6

TABLE VIII. $D(n\alpha)$ for $J_a=\frac{3}{2}\longleftrightarrow J_b=\frac{3}{2}$. ξ_α are defined by Eqs. (B3a) and (B3b) and f_α and g_α by Eqs. (B3e) and (B3f).

$n \backslash \alpha$	1 2 3 4	5 6 7 8
$(-\frac{3}{2})_a$	$-(\frac{2}{5})^{1/2}uf_\alpha$	0
$(+\frac{1}{2})_a$	$(\frac{2}{5})^{1/2}[(\frac{4}{3})^{1/2}vf_\alpha-ug_\alpha]$	0
$(-\frac{1}{2})_b$	$\xi_\alpha f_\alpha$	0
$(+\frac{3}{2})_b$	$\xi_\alpha g_\alpha$	0
$(-\frac{3}{2})_b$	0	$+(\frac{2}{5})^{1/2}v^*f'_\alpha$
$(+\frac{1}{2})_b$	0	$-(\frac{2}{5})^{1/2}[(\frac{4}{3})^{1/2}u^*f'_\alpha-v^*g'_\alpha]$
$(-\frac{1}{2})_a$	0	$(\xi_\alpha+\Delta)f'_\alpha$
$(+\frac{3}{2})_a$	0	$(\xi_\alpha+\Delta)g'_\alpha$

$$f_\alpha=-\xi_\alpha(\xi_\alpha+\Delta)+\tfrac{2}{5}uu^*-(\tfrac{16}{75})^{1/2}uv^*, \qquad \text{(B3e)}$$

$$g_\alpha=\xi_\alpha(\xi_\alpha+\Delta)-\tfrac{2}{5}uu^*-\tfrac{8}{15}vv^*+(\tfrac{16}{75})^{1/2}u^*v. \qquad \text{(B3f)}$$

An unnormalized $D(n\alpha)$ matrix is given in Table VIII.

*Supported by U. S. Army Research Office, Durham, N.C.

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