Approximate Perturbation Theory for High-Order Electromagnetic Transitions

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An approximation method is developed which yields very simple and general analytical results for electromagnetic bound-bound or bound-free transitions for any arbitrarily high order of interaction with the electromagnetic field. The approximation is perturbative in the sense that transition probabilities are proportional to the field intensity raised to the power N, where N is the order of the interaction. Limiting values of electromagnetic field strength are given within which the approximation is valid. These limits depend upon the binding potential which produces the bound states. A feature of the results is that they show the onset of high-order dominance effects which can occur at high-field intensities.

I. INTRODUCTION

The method to be described below applies to the description of transitions in bound quantum systems subjected to relatively intense electromagnetic fields. As used here, "relatively intense" refers to fields sufficiently intense that high-order multiphoton processes can occur to measurable extent, but not so intense that perturbation theory fails. The domain of intensities thus delineated is the same as that appropriate to high-order perturbation calculations[1] of electromagnetically caused transitions. The method given here is nonperturbative in origin.

We shall find results which can be called "perturbative" in the sense that for a process of order N (i.e., one which proceeds with N photons), the transition probability is proportional to the electromagnetic field intensity raised to the power N. This is a fundamental feature of perturbation theory. However, the results do not exactly duplicate perturbation theory. They are approximations to perturbation theory with a validity which improves as N increases. The method given here is of extremely simple analytical form in contrast to the very great difficulty posed by high-order perturbation theory.[1] The present method is also simpler in application than the momentum-translation method,[2-4] from which it follows as a limiting case.

In Sec. II below, the mathematical formalism will be developed for the case when only an intense field is present, as well as for the case when an additional weak electromagnetic field also exists. Section III is devoted to an examination of the limits of validity of the approximation, using three-dimensional Coulomb and harmonic-oscillator problems as models. A qualitative discussion of the results is given in Sec. IV with emphasis on the prediction that lowest-order processes cease to be dominant when the electromagnetic field becomes intense.

II. DEVELOPMENT OF APPROXIMATION

We shall employ as a starting point the momentum-translation approximation,[2-4] which is an analytical method valid for transitions caused in bound systems by an external plane-wave electromagnetic field, subject to the conditions

$$\omega/E \ll 1, \quad eaa_0(\omega/E) \ll 1, \tag{1}$$

where ω is the frequency of the electromagnetic field, E is a characteristic energy of the bound system, a is the amplitude of the vector potential $\vec{A}$ of the electromagnetic field, a_0 is the "size" parameter of the bound system, and units with $\hbar = c = 1$ are employed. Equation (1) imposes only a weak limitation on the intensity of the field. Of most immediate interest for present purposes is the fact that the inequalities of Eq. (1) can be satisfied for high-order multiphoton processes.

The T matrix for a transition of order N caused by the electromagnetic field is given[4] by

$$T_{fi}^{(N)} = i^N (E_i - E_f)(\phi_f, J_N(ea\vec{x}\cdot\vec{\epsilon})\,\phi_i), \tag{2}$$

where ϕ_i and ϕ_f are the stationary-state wave functions for the initial and final states (of energies E_i, E_f) in the absence of the electromagnetic field, $J_N(z)$ is the Bessel function of the first kind of order N, $\vec{\epsilon}$ is the polarization vector of the electromagnetic field, and $\vec{x}$ is a radius vector. When the transition involves not only the absorption or emission of N photons of energy ω, but also the absorption or emission of a single photon of energy ω' from a field of low intensity, then the T matrix is given[4] by

$$T_{fi}^{(N,1)} = \tfrac{1}{2}\, i^{N+1} (E_i - E_f)\, e^{\pm i\alpha}(ea') \times(\phi_f, \vec{x}\cdot\vec{\epsilon}'\, J_N(ea\vec{x}\cdot\vec{\epsilon})\,\phi_i). \tag{3}$$

In this expression, $\vec{\epsilon}'$ is the polarization vector and a' is the amplitude of the vector potential $\vec{A}'$ for the weak field. The quantity α is a phase displacement between the $\vec{A}$ and $\vec{A}'$ fields.

The matrix elements in Eqs. (2) and (3) can be simplified by the approximation

$$J_N(z) \approx (N!)^{-1} (\tfrac{1}{2}z)^N . \tag{4}$$

Equation (4) is both a small-argument approximation for the Bessel function and an asymptotic result when $N \to \infty$ for fixed argument. Both points of view will be considered when the conditions under which Eq. (4) is valid are examined in Sec. III.

A. Problem with Intense Field Only

We shall presume the system described by the wave functions ϕ_i and ϕ_f is a central-force system, so that the matrix element in Eq. (2) can be written

$$(\phi_f, J_N(ea\vec{\mathrm{x}}\cdot\vec{\epsilon})\phi_i)$$

$$\approx (1/N!)(\tfrac{1}{2}ea)^N \int_0^\infty r^2\, dr R_f^*(r) R_i(r)$$

$$\times \int d\Omega\, Y_{l_f}^{m_f *}(\theta,\varphi)\, Y_{l_i}^{m_i}(\theta,\varphi)(\vec{\mathrm{x}}\cdot\vec{\epsilon})^N , \tag{5}$$

with the help of Eq. (4), and where $R(r)$ and $Y_l^m(\theta,\varphi)$ are the radial and angular parts of the wave function ϕ. It is convenient to choose the axis of the spherical coordinates along $\vec{\epsilon}$, so that

$$(\vec{\mathrm{x}}\cdot\vec{\epsilon})^N = r^N \cos^N\theta .$$

The angular part of this can be expressed in terms of spherical harmonics by the expansion

$$\cos^N\theta = \sum_{j=0}^{N} b_j P_j(\cos\theta)$$

$$= \sum_{j=0}^{N} b_j \left(\frac{4\pi}{2j+1}\right)^{1/2} Y_j^0(\theta,\varphi) . \tag{6}$$

The angular integral in Eq. (5) now contains three spherical-harmonic factors. The solid-angle integral of the product of three spherical harmonics is well known,[5] and leads to the result

$$\int d\Omega\, Y_{l_f}^{m_f *}(\theta,\varphi)\, Y_{l_i}^{m_i}(\theta,\varphi)\cos^N\theta$$

$$= \sum_{j=0}^{N} b_j (-)^{m_f} (2l_i+1)^{1/2}(2l_f+1)^{1/2}$$

$$\times \begin{pmatrix} l_i & l_f & j \\ m_i & -m_f & 0 \end{pmatrix} \begin{pmatrix} l_i & l_f & j \\ 0 & 0 & 0 \end{pmatrix} . \tag{7}$$

Equation (7) contains within it the requirement that $m_i = m_f$. The result for Eq. (5) is given by

$$(\phi_f, J_N(ea\vec{\mathrm{x}}\cdot\vec{\epsilon})\phi_i)$$

$$\approx \frac{1}{N!}(\tfrac{1}{2}ea)^N \sum_{j=0}^{N} b_j (-)^{m_i} (2l_i+1)^{1/2}(2l_f+1)^{1/2}$$

$$\times \begin{pmatrix} l_i & l_f & j \\ m_i & -m_i & 0 \end{pmatrix} \begin{pmatrix} l_i & l_f & j \\ 0 & 0 & 0 \end{pmatrix} \int_0^\infty r^{N+2}\, dr\, R_f^*(r) R_i(r) . \tag{8}$$

A particularly simple special case arises if either the initial or final state is an s state, since the sum in Eq. (8) then reduces to a single term. Suppose we set $l_i = 0$, which, in turn, implies $m_i = 0$. Then both of the Wigner 3-j coefficients in Eq. (8) are the same, and each has the value

$$\begin{pmatrix} 0 & l_f & j \\ 0 & 0 & 0 \end{pmatrix} = \frac{(-)^j}{(2j+1)^{1/2}}\, \delta_{j l_f} .$$

The matrix element for this case is

$$(\phi_f, J_N(ea\vec{\mathrm{x}}\cdot\vec{\epsilon})\phi_i)_{l_i=0} \approx \frac{1}{N!}\left(\frac{ea}{2}\right)^N b_{l_f} \frac{1}{(2l_f+1)^{1/2}}$$

$$\times \int_0^\infty r^{N+2}\, dr R_f^*(r) R_i(r) . \tag{9}$$

Had the case $l_f = 0$ rather than $l_i = 0$ been considered, the result is given by Eq. (9) with l_i in place of l_f. If both $l_i = 0$ and $l_f = 0$, then the matrix element is

$$(\phi_f, J_N(ea\vec{\mathrm{x}}\cdot\vec{\epsilon})\phi_i)_{l_i=l_f=0} \approx \frac{1}{(N+1)!}\left(\frac{ea}{2}\right)^N$$

$$\times \int_0^\infty r^{N+2}\, dr\, R_f^*(r) R_i(r) , \tag{10}$$

where we have used the fact that $b_0 = (N+1)^{-1}$,[6] and note that N must be even.

B. Combined Intense and Weak Fields

In most physical problems an integer number of photons from the intense field will not be resonant with the transition energy of the bound system, so a photon of appropriate energy to conserve overall energy conservation must either be emitted or be present initially so that it can be absorbed. The relevant T matrix is given by Eq. (3).

We again use the approximation given in Eq. (4), which leads to

$$(\phi_f, \vec{\mathrm{x}}\cdot\vec{\epsilon}\,' J_N(ea\vec{\mathrm{x}}\cdot\vec{\epsilon})\phi_i)$$

$$\approx \frac{1}{N!}\left(\frac{ea}{2}\right)^N \int_0^\infty r^2\, dr R_f^*(r) R_i(r)$$

$$\times \int d\Omega\, Y_{l_f}^{m_f *}(\theta,\varphi)\, Y_{l_i}^{m_i}(\theta,\varphi)\,(\vec{\mathrm{x}}\cdot\vec{\epsilon}\,')(\vec{\mathrm{x}}\cdot\vec{\epsilon})^N . \tag{11}$$

The term $(\vec{\mathrm{x}}\cdot\vec{\epsilon})^N$ leads again to Eq. (6), but now $\vec{\mathrm{x}}\cdot\vec{\epsilon}\,' = r\cos\theta'$ must also be considered. The addition theorem can be used to relate $\cos\theta'$ to the θ, φ coordinates. With β and γ as the θ and φ coordinates of ϵ' with respect to ϵ we have

$$\cos\theta' = \tfrac{4}{3}\pi \sum_{m=-1}^{1} Y_1^{m *}(\beta,\gamma)\, Y_1^m(\theta,\varphi) .$$

The angular integration of Eq. (11) now contains four spherical-harmonic factors. The number of spherical-harmonic factors can be reduced by

using the expression

$$Y_{l_f}^{m_f*} Y_{l_i}^{m_i} = (-)^{m_f} \sum_{L,M} \left(\frac{(2l_i+1)(2l_f+1)(2L+1)}{4\pi}\right)^{1/2} \times \begin{pmatrix} l_i & l_f & L \\ m_i & -m_f & M \end{pmatrix} \begin{pmatrix} l_i & l_f & L \\ 0 & 0 & 0 \end{pmatrix} Y_L^{M*} .$$

The final result for the angular integral in Eq. (11) is then

$$\int d\Omega\, Y_{l_f}^{m_f*}(\theta,\varphi)\, Y_{l_i}^{m_i}(\theta,\varphi)\,(\vec{x}\cdot\vec{\epsilon}\,')(\vec{x}\cdot\vec{\epsilon})^N = r^{N+1}\left(\tfrac{4}{3}\pi\right)^{1/2} \sum_{m=-1}^{1} Y_1^{m*}(\beta,\gamma) \times \sum_{j=0}^{N} b_j (-)^{m_f+m} \sum_{L=|j-1|}^{j+1} (2L+1)[(2l_i+1)(2l_f+1)]^{1/2} \begin{pmatrix} l_i & l_f & L \\ m_i & -m_f & m \end{pmatrix} \begin{pmatrix} l_i & l_f & L \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} j & 1 & L \\ 0 & m & -m \end{pmatrix} \begin{pmatrix} j & 1 & L \\ 0 & 0 & 0 \end{pmatrix} . \quad (12)$$

Equation (12), when substituted into Eq. (11), gives the general result for the two-fields case for a central potential.

As in the single-field problem, the result simplifies considerably when either l_i or l_f is zero. As before, the result is symmetrical with respect to these two special cases. This time, the $l_f = 0$ case will be presented explicitly. The 3-j coefficients in Eq. (12) take on the values

$$\begin{pmatrix} l_i & l_f & L \\ m_i & -m_f & m \end{pmatrix} \to \begin{pmatrix} l_i & 0 & L \\ m_i & 0 & m \end{pmatrix} = (-)^{l_i+m_i}(2l_i+1)^{-1/2}\,\delta_{l_iL}\delta_{m,-m_i}$$

and

$$\begin{pmatrix} l_i & l_f & L \\ 0 & 0 & 0 \end{pmatrix} \to \begin{pmatrix} l_i & 0 & L \\ 0 & 0 & 0 \end{pmatrix} = (-)^{l_i}(2l_i+1)^{-1/2}\,\delta_{l_iL} .$$

The matrix element in this case is

$$(\phi_f, \vec{x}\cdot\vec{\epsilon}\,' J_N(ea\vec{x}\cdot\vec{\epsilon})\,\phi_i)_{l_f=0} \approx \frac{1}{N!}\left(\frac{ea}{2}\right)^N \left(\tfrac{4}{3}\pi\right)^{1/2}(-)^{m_i} \times Y_1^{m_i*}(\beta,\gamma)(2l_i+1)^{1/2} \sum_{j=0}^{N} b_j \begin{pmatrix} j & 1 & l_i \\ 0 & -m_i & m_i \end{pmatrix} \begin{pmatrix} j & 1 & l_i \\ 0 & 0 & 0 \end{pmatrix} \times \int_0^\infty r^{N+3}\, dr\, R_f^*(r) R_i(r) . \quad (13)$$

The 3-j coefficients remaining in Eq. (13) imply the constraints

$$l_i + 1 \geq j \geq |l_i - 1| , \quad |m_i| \leq 1 ,$$

which means that Eq. (13) is really much simpler than it appears to be.

A further specialization occurs if l_i is set equal to zero as well as l_f. The 3-j coefficients in Eq. (13) imply that $j = 1$ and $m_i = 0$, and each 3-j coefficient has the value $-3^{-1/2}$. The final result is then

$$(\phi_f, \vec{x}\cdot\vec{\epsilon}\,' J_N(ea\vec{x}\cdot\vec{\epsilon})\,\phi_i)_{l_i=l_f=0} \approx \frac{1}{N!\,(N+2)}\left(\frac{ea}{2}\right)^N \times \vec{\epsilon}\cdot\vec{\epsilon}\,' \int_0^\infty r^{N+3}\, dr\, R_f^*(r) R_i(r) , \quad (14)$$

where we have used $b_1 = 3(N+2)^{-1}$, [6] and N must be odd. This matrix element can be checked against a sample calculation in Ref. 4 for transitions between the 1s and 2s states in hydrogen. When hydrogenic 1s and 2s radial wave functions are substituted in Eq. (14), the result is the same as that given by Eq. (32) of Ref. 4, which is a low-intensity limiting case of a more general expression.

III. LIMITS ON VALIDITY OF APPROXIMATION

The validity of the above work depends upon the accuracy of the approximation of Eq. (4). The conditions for validity of the approximation will be estimated first for Eq. (4) as an asymptotic limit for $N \to \infty$, and then for the small-argument limit $z \to 0$. The conditions for validity depend upon the radial wave functions. Two cases will be discussed which have somewhat different properties. One is the problem of bound-bound transitions in a Coulomb potential, and the other is the harmonic-oscillator problem.

A. High-Order Limit

The basic conditions for this case are twofold:

$$N \gg 1 \quad (15)$$

and

$$|ea\vec{x}\cdot\vec{\epsilon}\,| < N . \quad (16)$$

The difficulty with the condition (16) is that the magnitude $r = |\vec{x}|$ enters into a volume integration, so that all values of r must be considered, and Eq. (16) appears to be meaningless. However, bound-state wave functions have the property that they fall off sharply for large r, and little error occurs if the radial integral is truncated beyond some value of r. This truncation will be illustrated by a specific example.

The radial eigenfunctions for hydrogenlike atoms can be written as [7]

$$R(r) = B_{nl}\, e^{-\rho/2}\rho^l L_{n-l-1}^{2l+1}(\rho) , \quad (17)$$

where B_{nl} is a normalization factor, n is the principal quantum number, l is the angular momentum quantum number, and

$$\rho = 2Zr/na_0 ,$$

with Ze the nuclear charge. When a solution of the type of Eq. (17) is inserted into the radial integral which occurs in Eq. (8),

$$\int_0^\infty dr\, r^{N+2} R_f^*(r) R_i(r) \equiv \int_0^\infty dr\, I(r) , \tag{18}$$

the integrand $I(r)$ goes to zero at $r=0$ and ∞. There are, therefore, extrema encompassed between the limits of integration. Equation (17) is a finite sum of terms of the type $e^{-\rho/2}\rho^p$, each of which has zeros at $\rho=0$ and ∞ and a maximum at $\rho=2p$. The largest value of r which will be considered to make a significant contribution to the integral in Eq. (18) is that value corresponding to the last extremum which can occur in the terms constituting $I(r)$. This last extremum arises from the term of highest power in $L_{n-l-1}^{2l+1}(\rho)$, namely, ρ^{n-l-1}. Consider the case of bound-bound transitions in a Coulomb potential, that is, the case where $R_f(r)$ and $R_i(r)$ are both of the form of Eq. (17). The term containing the highest power of r in the $I(r)$ of Eq. (18) is thus

$$\bar{I}(r) \sim r^{N+2} e^{-\rho_f/2} e^{-\rho_i/2} \rho_f^{l_f} \rho_i^{l_i} \rho_f^{n_f-l_f-1} \rho_i^{n_i-l_i-1} ,$$

where $\rho_f = 2Zr/n_f a_0$, $\rho_i = 2Zr/n_i a_0$. When written directly in terms of r, this is

$$\bar{I}(r) \sim \exp\left[-\frac{Zr}{a_0}\left(\frac{1}{n_f}+\frac{1}{n_i}\right)\right] r^{N+n_f+n_i} . \tag{19}$$

The extremum of Eq. (19) is at

$$\bar{r} = \frac{a_0}{Z}(N+n_f+n_i)\frac{n_f n_i}{n_f+n_i} . \tag{20}$$

Beyond this value of r, exponential damping of $I(r)$ occurs, and little contribution to the radial integral is to be expected. The condition (16) will be interpreted as $ea\bar{r} < N$, with $\bar{r}$ given by Eq. (20). This yields directly the intensity-parameter limitation

$$eaa_0 < \frac{ZN}{N+n_f+n_i}\left(\frac{1}{n_f}+\frac{1}{n_i}\right) . \tag{21}$$

If the above considerations are applied to the radial integral of Eq. (13) instead of Eq. (8), substantially the same results are obtained. It is found that Eq. (21) is to be replaced by

$$eaa_0 < \frac{ZN}{N+n_f+n_i+1}\left(\frac{1}{n_f}+\frac{1}{n_i}\right) , \tag{22}$$

which is a trivial change in view of the $N \gg 1$ condition. If, in addition to $N \gg 1$, it is also true that $N \gg n_f$ and $N \gg n_i$, then Eqs. (21) and (22) yield the N-independent result

$$eaa_0 < Z\left(\frac{1}{n_f}+\frac{1}{n_i}\right) . \tag{23}$$

More conservative conditions would be obtained if it were required that $ea\kappa\bar{r} < N$, with κ some constant greater than unity.

Another example with somewhat different properties is provided by the three-dimensional harmonic oscillator. The radial wave functions for this problem may be written

$$R(r) = C_{nl} e^{-\rho^2/2} \rho^l L_n^{l+(1/2)}(\rho^2) , \tag{24}$$

where

$$\rho = (m\omega_0)^{1/2} r$$

and the energy eigenvalues associated with the n and l quantum numbers are

$$E = (2n+l+\tfrac{3}{2})\omega_0 .$$

Now the term containing the highest power of r in the integrand of Eq. (18) is

$$\bar{I}(r) \sim e^{-m\omega_0 r^2} r^{N+2n_f+2n_i+l_f+l_i+2} ,$$

which has an extremum at

$$\bar{r}^2 = (2m\omega_0)^{-1}(N+2n_f+2n_i+l_f+l_i+2) .$$

The condition $ea\bar{r} < N$ now leads to

$$ear_0 < 2^{1/2} N (N+2n_f+2n_i+l_f+l_i+2)^{-1/2} , \tag{25}$$

where r_0 is the radius characteristic of the harmonic-oscillator problem,

$$r_0 = (m\omega_0)^{-1/2} .$$

From the radial integral of the two-fields problem of Eq. (13), the trivially modified result

$$ear_0 < 2^{1/2} N (N+2n_f+2n_i+l_f+l_i+3)^{-1/2} \tag{26}$$

is obtained. Both Eqs. (25) and (26) reduce to

$$ear_0 < (2N)^{1/2} , \tag{27}$$

when N is much greater than all the n and l quantum numbers. Note that Eq. (27) is not independent of N as was Eq. (23), and that Eq. (27) taken together with $N \gg 1$ implies a broad domain of applicability for the Bessel-function approximation [Eq. (4)].

B. Low-Intensity Limit

The requirement that $N \gg 1$ employed in the preceding section is a natural one to be consistent with $\omega/E \ll 1$ expressed in Eq. (1). When we deal with the single-field or resonant case, $N \gg 1$ is an inescapable consequence of $\omega/E \ll 1$; but in the nonresonant case, when a second field participates in the transition in addition to the intense field, then it is possible to have $\omega/E \ll 1$ and also have N be 1, 2, or any other integer. The results following from Eq. (4) can be sustained without requiring $N \gg 1$, if it is required instead that

$$|ea\vec{x}\cdot\vec{\epsilon}| \ll 1 . \tag{28}$$

The condition (28) now takes the place of the two conditions (15) and (16) to provide a justification of Eq. (4).

The results of the preceding section may be employed directly if the N which appears in Eq. (16) is replaced by unity, and the symbol $<$ is replaced by $\ll$. That is, Eq. (22) for the Coulomb case becomes

$$eaa_0 \ll \frac{Z}{N+n_f+n_i+1}\left(\frac{1}{n_f}+\frac{1}{n_i}\right), \tag{29}$$

and the harmonic-oscillator result [Eq. (26)] takes the form

$$ear_0 \ll 2^{1/2}(N+2n_f+2n_i+l_f+l_i+3)^{-1/2}. \tag{30}$$

An explicit example in which these conditions may be applied is provided by the case of a transition with $n_i = 2$, $n_f = 1$, and $l_i = l_f = 0$ in hydrogen ($Z = 1$). The results for this problem with $N = 1$ are presented in Fig. 1 of Ref. 3, which shows a departure of perturbation theory from momentum-translation approximation at values of $y \geq 0.2$ (where y is defined as $\frac{2}{3}eaa_0$) or $eaa_0 \geq 0.3$. The relevant condition to be applied here is Eq. (29) which, with the appropriate parameters substituted, yields $eaa_0 \ll 0.3$ as a condition for validity of the low-intensity approximation. Other comparisons for larger N values also verify the reliability of the conditions given above.

The differences between the high-order and the low-intensity case are twofold. The high-order case leads to a significantly larger range of intensity over which the approximation is valid, as can be seen immediately by comparing the high-order conditions (21), (22), (26), and (27) with the low-intensity conditions (29) and (30). However, the constraint (15) on the multiplicity of the process is absent in the low-intensity case.

IV. DISCUSSION

The great advantages of the methods presented here are simplicity and generality. For example, the matrix element given in Eq. (8) requires only a knowledge of radial wave functions. If these wave functions are known analytically or numerically, the final result requires only a single integration and the performance of a finite sum. Equation (11) when combined with Eq. (12) requires only a single integration and three finite sums, two of which can contain no more than three terms each. The results thus obtained are approximations to perturbation theory of arbitrarily high order. High-order perturbation theory when done directly presents truly formidable analytical difficulties, in contrast to the very simple results presented here.

The possibility of obtaining closed-form analytical results for high-order processes provides an opportunity to examine nontrivial general features of high-order processes. For instance, if Eq. (14) is applied to the problem of multiphoton $1s$-$2s$ transitions in hydrogen, the result obtained for the T matrix is (see also Ref. 4)

$$T_{fi}^{(N,1)} = \pm e^{\pm i\alpha} i^{N+1} \frac{1}{2^{1/2}3^3} \frac{\vec{\epsilon}\cdot\vec{\epsilon}\,' y'}{ma_0^2}\left(\frac{y}{2}\right)^N \times (N+3)(N+1)^2 \tag{31}$$

in the notation of Refs. 3 and 4, where $y = \frac{2}{3}eaa_0$ and $y' = \frac{2}{3}ea'a_0$. As expected this contains a factor $(eaa_0)^N$, which means that the transition probability depends upon the intensity of the field raised to the power N —the multiphoton order of the process. Since $\frac{1}{2}y$ is constrained by the intensity limiting condition Eq. (29), it must be less than unity, and $(\frac{1}{2}y)^N$ is a factor which declines as N increases. For any "ordinary" intensity, the transition amplitude is so strongly damped by $(\frac{1}{2}y)^N$ that only low-order processes need be considered. However, since Eq. (31) is a closed-form result, another feature appears which is not obvious. When N becomes large, the transition amplitude is enhanced by a factor which increases as N^3. Equation (31) depends upon N as

$$T(N) = Cx^N(N+3)(N+1)^2, \tag{32}$$

where it is understood that $0 \leq x < 1$, and C is independent of N (except for the trivial phase factor i^N). Equation (32) has an extremum when

$$N = -2 + \tfrac{3}{2}(-\ln x)^{-1} + [1 + (-\ln x)^{-1} + \tfrac{9}{4}(-\ln x)^{-2}]^{1/2}.$$

This extremum will occur at $N = 1$ when $y = 2x = 0.57$, and at $N = 3$ when $y = 2x = 1.03$. These intensity values lie outside the region of validity of the present approximation method, and are not acceptable as correct predictions. The correct locations for the extrema come from the complete momentum-translation approximation results given in Ref. 4, which exhibit an $N = 1$ extremum at $y = 0.34$ and an $N = 3$ extremum at $y = 0.78$. Nevertheless, Eq. (31) has the general character of showing an enhancement of large-N effects, so that low-order processes do not dominate high-order processes in such an overwhelming way when y is relatively large [i.e., when y approaches the limits imposed by Eq. (22) or Eq. (29)]. The polynomial in N, which multiplies the $(\frac{1}{2}y)^N$ factor in Eq. (31), may be regarded as arising from the net effect of all the "diagrams" which can occur. Such "diagrams" can be very great in number, and Eq. (31) gives (at least approximately) the sum of all such diagrams including cancellation effects.

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Analytic Study of Pulse Chirping in Self-Induced Transparency*

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We analyze the phase modulation, or chirping, of coherent lossless light pulses propagating without distortion through resonant absorbers. In order to do this, we generalize the pioneering work of McCall and Hahn in two different directions. In the first place, of course, we abandon their assumption that the phase of the pulses has no temporal dependence. We prove that for slowly varying single pulses, chirping is not possible. However, we describe many multiple-pulse trains which are necessarily chirped, even under the slowly varying envelope restriction, and also describe the envelope modulations which produce large chirps. We show that certain zero-π chirped pulse trains are contained as special cases of our general results. Our second generalization of the McCall-Hahn work concerns the background material in which the two-level resonant atoms are suspended. We allow the host medium to possess significant nonresonant nonlinearities. We find that undistorted lossless single pulses are possible in such a medium and that they are necessarily chirped.

I. INTRODUCTION

The discovery of self-induced transparency (SIT) by McCall and Hahn[1] has focused attention again on the very old problem of light propagation in dielectrics. Even before 1920 the classical investigations of Sommerfeld and Brillouin,[2] among others, were sufficiently complete and in accord with experimental observation to discourage further serious study. However, the Lorentz linear model of dielectrics, the model used in those early studies, is adequate only if the light intensity is low, or if the light frequency is far from any of the atomic resonances of the dielectric medium.

It is only in the past decade that intense and practically monochromatic laser light has been available as a strong probe of optically resonant systems. The response of such systems, when strongly probed at resonance, is not well described by the Lorentz model of harmonically oscillating charges. Important nonlinearities arise in the light-dielectric interaction, and these are instrumental in producing a wide range of nonclassical effects, such as SIT,[1] photon echoes, optical nutation, and others.[3] The very recent achievement of continuously operating and continuously tunable dye lasers[4] promises to add further impetus to modern experimental studies of light propagation in resonant dielectrics.

One of the most interesting of the new phenomena observed in high-intensity coherent light propagation is frequency modulation of the electric field. By analogy with similar phenomena which are common at much lower frequencies, a frequency-modulated electromagnetic wave is said to be "chirped," and we will use this terminology frequently.

In this paper we analyze situations in which chirped optical waves may occur in SIT. In addition to a time-varying field phase we also allow nonresonant nonlinearities of the host medium in which the resonant atoms are imbedded.

We imply several restrictions on our work by the words chosen to describe it. By SIT we mean the propagation *without distortion* of an electric wave's envelope and phase through a medium containing resonant atoms. Because we will always work close to resonance, we assume, along with McCall and Hahn, that the resonant atoms may be treated as if they had only two energy levels separated by a transition frequency almost equal to the field-carrier frequency. In the most general case, these atoms are embedded in a host medium