Multiple-time-scale perturbation theory applied to laser excitation of atoms and molecules*

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The multiple-linear-time-scale method is used to construct a perturbation theory for N-level quantum systems subjected to time-dependent perturbations. The secular and small-denominator terms which plague conventional time-dependent perturbation theory are avoided; consequently, the theory is useful for treating long-term behavior and resonant interactions. The introduction of a self-energy operator allows the shifts in energy levels to be displayed explicitly. When the perturbation is the dipole interaction with an electromagnetic field, the successive approximations yield the well-known rotating-wave approximation, corrections due to counter rotating terms, and the Bloch-Siegert frequency shift. The formalism is applicable to arbitrary pulse shapes, provided that $\| H_1 \| \ll \hbar\bar{\omega}$, where $\| H_1 \|$ represents the interaction strength and $\bar{\omega}$ is a characteristic frequency of the field. Rabi oscillations induced by multiphoton resonances are automatically included, and this effect is demonstrated in the case of a square pulse with frequency approximately one half the resonance frequency for a transition between two vibrational levels of a molecule. These calculations are compared to an exact numerical solution in order to find the limits of validity of the approximation. Even at high intensities ($I \sim 10^{14}$ W/cm^2) the shifted resonance frequency is quite accurate; however, the approximate and exact solutions are slightly out of phase, so that the approximate solution can only be trusted for a few Rabi cycles. For much lower intensities ($I \sim 10^{10}$ W/cm^2), the approximate solution is valid for many thousands of Rabi cycles.

I. INTRODUCTION

The advent of high-power lasers capable of stable operation over relatively long periods of time has made possible the experimental investigation of coherent resonant excitation (Rabi oscillations) of atoms and molecules by optical and infrared light; consequently it is desirable to develop approximation schemes suitable for the theoretical description of such experiments. One such technique, the rotating-wave approximation,[1] has been in existence for a long time, and the purpose of this paper is to present a scheme which justifies and extends this method. We will show that the rotating-wave approximation appears in the leading order of a systematic approximation scheme which automatically generates the Bloch-Siegert shifts in the energy levels, describes Rabi oscillations caused by multiphoton resonances, and gives the corrections to the wave function caused by the high-frequency (counterrotating) terms in the Hamiltonian.

In order to motivate our method we first briefly review the difficulties encountered in using conventional time-dependent perturbation theory to describe the resonant interactions and long-term behavior of quantum systems.

Consider a system governed by the Hamiltonian $H_0+\lambda H_1(t)$, where H_0 is the unperturbed Hamiltonian, $H_1(t)$ represents the effect of a time-dependent perturbation, and λ is a formal expansion parameter. The usual perturbation theory describes the effects of H_1 in terms of transitions between eigenstates of H_0. It is well known that this description becomes valid only after a time sufficiently long so that the uncertainty principle allows a sharp definition of the final-state energy. It is also known, but not usually emphasized, that for sufficiently long times conventional perturbation theory fails no matter how weak the perturbation. Thus if we expand the wave function in the basis of the eigenstates of H_0,

$$\psi(t) = \sum_\alpha C_\alpha(t) e^{-iE_\alpha t}\phi_\alpha ,$$

and then expand C_α in powers of λ,

$$C_\alpha = \sum_{n=0}^{\infty} C_\alpha^{(n)} \lambda^n ,$$

we will find that some of the coefficients contain terms linear in t. This always happens for $C^{(2)}$ even if the external field is not resonant with any transition of the system. Consequently after a time $t \sim O(\lambda^{-2})$ we would find $|\lambda^2 C^{(2)}(t)| \approx |C^{(0)}(t)|$. Terms of this sort are called secular because of a similar problem which occurs in the perturbation theory for planetary orbits. Near a resonance a closely related difficulty arises which is due to the appearance of small energy denominators. For example, if the external field is purely sinusoidal with frequency ω and the system is initially in its ground state we have the familiar result

$$C_\alpha^{(1)} = \frac{1}{2\hbar}\left(\frac{e^{i(E_\alpha - E_0 - \omega)t} - 1}{E_\alpha - E_0 - \omega} + \frac{e^{i(E_\alpha - E_0 + \omega)t} - 1}{E_\alpha - E_0 + \omega}\right)(H_1)_{\alpha 0}.$$

From this expression we see that a near-resonance [i.e., $E_\alpha - E_0 - \omega = O(\lambda)$] yields $|\lambda C_\alpha^{(1)}| \approx 1$ for times $t \gtrsim O(\lambda^{-1})$. These small denominator terms will be referred to as quasisecular.

Difficulties of the sort just mentioned occur in several areas such as planetary orbit theory,[2] nonlinear mechanics,[3] and the derivation of kinetic equations from statistical mechanics.[4,5] Of the various techniques developed to deal with these problems we have chosen to use the method of multiple linear time scales which has been employed in derivations of the kinetic equations.[4] It is interesting to note that the idea of a many-body wave function depending on many time variables was introduced by Dirac[6] in his relativistically covariant version of the Schrödinger equation. This theory was later generalized by Tomonaga[7] in his work on quantum field theory. Dirac's many-time quantum mechanics formally resembles the method described below. A version of the multiple-time-scale method closely related to ours has been applied to the problem of coherent spontaneous emission from many-body systems.[7a]

The method of multiple linear time scales is based on the existence of several time or frequency scales related by a small parameter. The physical time t is then replaced by (or extended to) a multicomponent variable $(t_0, t_1, \ldots)$ in which the t_k's represent the distinct time scales involved. By using the extra degrees of freedom thus introduced we can construct a solution, in the form of an expansion in the small parameter, which is free of secular and quasisecular terms. In our problem the presence of different time scales follows from the assumption that the response frequency of the system is small compared to the characteristic frequency of the external perturbation. Let $\|H_1\|$ be a suitable measure of the strength of H_1; then the response frequency is roughly $\|H_1\|/\hbar$. If $\bar{\omega}$ is the average frequency contained in the external field, the small parameter of the theory is $\|H_1\|/\hbar\omega$. The estimate presented in Sec. V shows that the smallness of this parameter does not impose severe restrictions on the laser intensity.

The plan of the paper is as follows: In Sec. II we develop the multiple-time-scale method, give a simple example, and finally give the general prescription for finding the solution. We show, in Sec. III, that unitarity (conservation of probability) is guaranteed to each order uniformly in time. In Sec. IV we use the method to generalize the Bloch-Siegert theory of field-dependent frequency shifts. In Sec. V we demonstrate a two-photon coherent resonance, and we investigate the limits of validity of the theory by comparing the approximate results for the two-photon problem with an exact numerical solution. Finally in Sec. VI we summarize and discuss the results.

II. MULTIPLE TIME SCALES

In order to have something definite in mind, let us think of an atom or molecule interacting with an electromagnetic field in the dipole approximation. We will not consider ionization or dissociation processes; consequently the continuum levels will be ignored and we only have to deal with N-level quantum systems. The interaction Hamiltonian has the form

$$H_1(t) = -\vec{\mathcal{E}}(t)\cdot\vec{d},$$

where $\vec{d}$ is the dipole operator and $\vec{\mathcal{E}}(t)$ is the electric field. Let H_0 be the Hamiltonian for the unperturbed system, and let M be a Hermitian operator that commutes with H_0. We write the Schrödinger equation as

$$i\frac{\partial\psi}{\partial t} = (H_0 + M)\psi + (\lambda H_1 - M)\psi.$$

The operator M is not required for the elimination of secularities and quasisecularities; however, we will see that a judicious choice for M allows us to exhibit very clearly the shifts in the unperturbed energy levels caused by the external field. These shifts may be regarded as self-energy effects, so we will call M a self-energy operator. Since M must vanish in the absence of an external field, we assume for it an expansion with no constant term,

$$M = \sum_{k=1}^{\infty} \lambda^k M^{(k)},$$

where each $M^{(k)}$ commutes with H_0.

For our purposes it is convenient to work in the basis provided by the eigenfunctions $\{\phi_\alpha\}$ of H_0; they are simultaneously eigenfunctions of M and of the combination $K = H_0 + M$. Thus we have

$$H_0\phi_\alpha = E_\alpha^{(0)}\phi_\alpha, \quad M^{(k)}\phi_\alpha = E_\alpha^{(k)}\phi_\alpha, \quad K\phi_\alpha = E_\alpha\phi_\alpha,$$

and

$$E_\alpha = \sum_{n=0}^{\infty} \lambda^n E_\alpha^{(n)}.$$

If we expand the exact time-dependent wave function ψ as

$$\psi = \sum_\alpha C_\alpha(t) e^{-iE_\alpha t}\phi_\alpha,$$

then the Schrödinger equation leads to a set of equations for C_α which we write in matrix notation

as

$$i\frac{\partial C}{\partial t}=[\lambda H^{(1)}(t)-M]C\,, \tag{2.1}$$

where

$$H^{(1)}_{\alpha\beta}\equiv e^{iE_{\alpha\beta}t}(\phi_\alpha,H_1(t)\phi_\beta)$$

and

$$E_{\alpha\beta}\equiv E_\alpha-E_\beta\,.$$

We now seek a solution of (2.1) having the form

$$C(t)=\sum_{n=0}^{\infty}\lambda^n C^{(n)}(\lambda,t,\lambda t,\lambda^2 t,\ldots)\,,$$

where the dependence on the combinations $\lambda^k t$ represents the effects of the various time scales. The idea of the multiple-time-scale method is to replace t by a multicomponent variable $(t_0,t_1,t_2,\ldots)$. In this extended space the physical time axis is represented by the line $t_k=\lambda^k t$ for $k\geq 0$, which we call the physical line, and the operator $\partial/\partial t$ is represented by the directional derivative along this line,

$$\frac{\partial}{\partial t}\to\sum_{n=0}^{\infty}\lambda^n\frac{\partial}{\partial t_n}\,.$$

As a shorthand notation we put $\vec{t}_k=(t_k,t_{k+1},\ldots)$, and denote the evaluation of $F(\vec{t}_0)$ on the physical line by $F(\vec{t}_0)|_{\mathrm{PL}}$. The Schrödinger equation is extended to

$$\sum_{n=0}^{\infty}\lambda^n\frac{\partial}{\partial t_n}C(\vec{t}_0)=\sum_{n=1}^{\infty}\lambda^n[Q^{(n)}(\lambda,\vec{t}_0)-M^{(n)}]C(\vec{t}_0)\,, \tag{2.2}$$

where the $Q^{(n)}$'s satisfy

$$\begin{aligned}&Q^{(1)}(\lambda,\vec{t}_0)|_{\mathrm{PL}}=H^{(1)}(t)\,,\\ &Q^{(k)}(\lambda,\vec{t}_0)|_{\mathrm{PL}}=0\,,\quad k\geq 2\,.\end{aligned} \tag{2.3}$$

The definition of the physical line and the properties of $Q^{(n)}$ guarantee that every solution $C(\vec{t}_0)$ of (2.2) generates a solution $C(t)=C(\vec{t}_0)|_{\mathrm{PL}}$ of the physical Schrödinger equation. Our task is to use the extra freedom obtained by the extension to obtain a secularity-free solution in the form

$$C(\vec{t}_0)=\sum_{n=0}^{\infty}\lambda^n C^{(n)}(\lambda,\vec{t}_0)\,.$$

Note that the "expansion" coefficients must be allowed to depend on λ; this is necessary because the $Q^{(k)}$'s must depend on λ in order to satisfy the conditions (2.3). This complication could be avoided by setting $Q^{(1)}=H^{(1)}(t_0)$ and $Q^{(k)}=0$, $k\geq 2$ but we will see later that the $Q^{(k)}$'s are needed for the elimination of quasisecular behavior. Substituting the expansion for C into (2.2) and equating coefficients of equal powers of the explicit λ's, we obtain an infinite set of equations for the $C^{(n)}$'s. If the expansion converges, these $C^{(n)}$'s determine a solution to (2.2) which leads to a solution of the Schrödinger equation by restriction to the physical line. We assume the convergence of the expansion, provided only that there are no secularities or quasisecularities in the coefficients $C^{(n)}$. The equations are

$$\sum_{m=0}^{n}i\partial_m C^{(n-m)}=\sum_{l=1}^{n}(Q^{(l)}-M^{(l)})C^{(n-l)}\,, \tag{2.4}$$

where $\partial_m\equiv\partial/\partial t_m$ and $C^{(k)}=0$ for $k<0$.

Before giving the general prescription for determining a suitable solution to (2.4), it may be useful to illustrate the avoidance of secular behavior by an example which, although grossly oversimplified, has the virtue that it can be solved exactly. Consider a "one-level" quantum system; i.e., H_1 is a number which we take to be time independent. The Schrödinger equation has the solution

$$C(t)=e^{-i\lambda H^{(1)}t}C(0)\,,$$

and conventional perturbation theory gives

$$C=[1-i\lambda H^{(1)}t+(1/2!)(-i\lambda H^{(1)}t)^2+\ldots]C(0)\,,$$

which plainly displays the secularity problem. In this case no quasisecular behavior can arise and we are not interested in the level shift so we can take $Q^{(1)}=H^{(1)}$, $Q^{(k)}=0$, $k\geq 2$ and $M=0$. The first two equations of the set (2.4) are then

$$\begin{aligned}&i\partial_0 C^{(0)}=0\,,\\ &i\partial_0 C^{(1)}+i\partial_1 C^{(0)}=H^{(1)}C^{(0)}\,.\end{aligned}$$

The first equation simply tells us that $C^{(0)}$ is independent of t_0; we denote this by $C^{(0)}=C^{(0;1)}$, where in general $C^{(n;m)}$ is a part of the nth-order coefficient that depends on $\vec{t}_m$ only. Integration of the second equation with respect to t_0 yields

$$C^{(1)}=(1/i)t_0(H^{(1)}-i\partial_1)C^{(0;1)}+C^{(1;1)}\,.$$

Thus $C^{(1)}$ exhibits secularity in t_0; this must be prevented by imposing the auxiliary condition

$$(H^{(1)}-i\partial_1)C^{(0;1)}=0\,,$$

with the solution

$$C^{(0;1)}=e^{-iH^{(1)}t_1}C^{(0;2)}\,.$$

At this stage the function $C^{(1;1)}$ is unknown; it is determined by the next equation

$$i\partial_0 C^{(2)}+i\partial_1 C^{(1)}+i\partial_2 C^{(0)}=H^{(1)}C^{(1)}\,.$$

Inserting the solutions for $C^{(0)}$ and $C^{(1)}$ and integrating with respect to t_0 yields

$$C^{(2)}=-it_0[(H^{(1)}-i\partial_1)C^{(1;1)}-e^{-iH^{(1)}t_1}i\partial_2 C^{(0;2)}]+C^{(2;1)}\,.$$

To prevent the t_0 secularity in $C^{(2)}$ we set the coefficient of t_0 equal to zero; this yields

$$i\partial_1(e^{iH^{(1)}t_1}C^{(1;1)}) + i\partial_2 C^{(0;2)} = 0.$$

This equation produces a t_1 secularity in $C^{(1;1)}$ which is eliminated by the condition

$$\partial_2 C^{(0;2)} = 0.$$

At this stage we have, on the physical line,

$$C^{(0)} = e^{-i\lambda H^{(1)}t}C^{(0;3)},$$

$$C^{(1)} = e^{-i\lambda H^{(1)}t}C^{(1;2)},$$

$$C^{(2)} = C^{(2;1)}.$$

The unknown functions $C^{(1;2)}$ and $C^{(2;1)}$ will be determined by the next equation in the series; however, if we are willing to neglect the $\vec{t}_2$ dependence and corrections to the amplitudes of $O(\lambda^2)$, then we can set $C^{(1;2)} = 0$ and $C^{(0;3)} = C(0)$ to get

$$C(t) = e^{-i\lambda H^{(1)}t}C(0) + O(\lambda^2).$$

In this simple example the multiple-time-scale method yields an essentially exact solution. In a more complicated problem we hope to get a good approximation to the exact solution.

We now turn to the general problem of obtaining solutions to (2.4) which are free of secular and quasisecular terms. At the same time we will choose the quantities $E_\alpha^{(k)}$ so as to extract the level shifts. The basic idea is to choose the quantities $Q^{(n)}$ so that all quasisecular terms are turned into secular terms which are then removed by the mechanism outlined in the example. For this purpose we need to isolate the terms that are independent of t_k and lead to t_k secularities as well as the terms that contain frequencies of $O(\lambda)$ and lead to quasisecularities. We can conveniently pick out the t_k-independent terms by using the t_k average, defined by

$$\langle A\rangle_k \equiv \lim_{T\to\infty}\frac{1}{T}\int_0^T dt_k A(t_k),$$

and we denote the nonconstant part by $A'(t_k) = A(t_k) - \langle A\rangle_k$. The part of A that leads to quasisecular behavior, after t_k integration, is denoted by $\mathcal{P}A$; it is defined by the following procedure:

(1) Decompose A' into high- and low-frequency parts,

$$A'(t_k) = \int\frac{d\omega}{2\pi}\theta(\omega^2 - \lambda^2\bar{\omega}^2)\hat{A}'(\omega)e^{-i\omega t_k} + \int\frac{d\omega}{2\pi}\theta(\lambda^2\bar{\omega}^2 - \omega^2)\hat{A}'(\omega)e^{-i\omega t_k},$$

where $\hat{A}'$ is the Fourier transform with respect to t_k, θ is the unit step function, and $\bar{\omega}$ is a suitable unit of frequency.

(2) For $|\omega| \leq \lambda\bar{\omega}$ write $\hat{A}'$ as

$$\hat{A}' = \hat{A}_0' + \hat{A}_1',$$

where $\hat{A}_0'$ is $O(1)$ and $\hat{A}_1'$ is $O(\lambda)$ (or smaller). Note that this is an order-of-magnitude analysis rather than a formal expansion in powers of λ.

(3) Define $\mathcal{P}A$ as

$$\mathcal{P}A = \langle A\rangle_k + \int\frac{d\omega}{2\pi}\theta(\lambda^2\bar{\omega}^2 - \omega^2)\hat{A}_0'(\omega)e^{-i\omega t_k}.$$

This procedure discards the high-frequency part of A, which obviously cannot lead to quasisecularities, and it also discards those terms in the low-frequency part that are sufficiently small to cancel the small denominators produced by t_k integration; therefore, $\mathcal{P}A$ contains all terms that can lead to quasisecular behavior. A special class of operators satisfying $\mathcal{P}A = 0$ is defined by

$$A = \frac{\partial}{\partial t_k}B, \tag{2.5}$$

where $\lambda|B| \ll 1$ or, equivalently, by

$$\hat{A} = \omega\hat{B}, \tag{2.6}$$

where $\hat{B}$ satisfies

$$\lambda\int\frac{d\omega}{2\pi}|\hat{B}(\omega)| \ll 1.$$

Any operator of the form (2.5) has $\langle A\rangle_k = 0$, and the equivalent definition (2.6) guarantees that $\hat{A}_0' = 0$; therefore $\mathcal{P}A = 0$. Operators of this form occur in the general discussion and in the applications discussed in Secs. IV and V. It also follows from the definition of $\mathcal{P}A$ that $\langle(1-\mathcal{P})A\rangle_k = 0$ for any A.

The application of these ideas to Eqs. (2.4) can begin with the $n=1$ equation since the $n=0$ equation simply gives $C^{(0)} = C^{(0;1)}$. We will carry out the procedure through $n=2$ in order to illustrate the problems involved in the general case. The $n=1$ equation can be written as

$$i\partial_0 C^{(1)} + i\partial_1 C^{(0;1)} = (H^{(1)} - M^{(1)} + R^{(1)})C^{(0;1)}, \tag{2.7}$$

where we have satisfied (2.3) by setting $Q^{(1)} = H^{(1)} + R^{(1)}$, with

$$R^{(1)}|_{\mathrm{PL}} = 0. \tag{2.8}$$

A sufficient condition for the absence of t_0 secularities in $C^{(1)}$ is $\langle\partial_0 C^{(1)}\rangle_0 = 0$; therefore the auxiliary condition for eliminating t_0 secularity from $C^{(1)}$ is obtained by imposing this condition on the t_0 average of (2.7) to find

$$i\partial_1 C^{(0;1)} = (\langle H^{(1)}\rangle_0 - M^{(1)} + \langle R^{(1)}\rangle_0)C^{(0;1)}. \tag{2.9}$$

The proper choice for $M^{(1)}$ becomes evident if we use $M_{\alpha\beta}^{(1)} = E_\alpha^{(1)}\delta_{\alpha\beta}$ and write out (2.9) in com-

ponent notation,

$$i\partial_1 C_\alpha^{(0,1)} = (\langle H_{\alpha\alpha}^{(1)}\rangle_0 - E_\alpha^{(1)})C_\alpha^{(0;1)} + \sum_{\beta\neq\alpha}\langle H_{\alpha\beta}^{(1)}\rangle_0 C_\beta^{(0;1)} + \sum_\beta \langle R_{\alpha\beta}^{(1)}\rangle_0 C_\beta^{(0;1)}.$$

If $E_\alpha^{(1)}$ and the off-diagonal coupling terms are temporarily suppressed, it is clear that the constant $\langle H_{\alpha\alpha}^{(1)}\rangle_0$ plays the role of an energy eigenvalue associated with ∂_1. There is another diagonal term $\langle R_{\alpha\alpha}^{(1)}\rangle_0$, but it cannot be considered an eigenvalue since it necessarily depends on some of the variables $t_1, t_2, \ldots$. This follows from (2.8), which can only be satisfied by functions depending on at least two of the variables $t_0, t_1, \ldots$. By the choice

$$E_\alpha^{(1)} = \langle H_{\alpha\alpha}^{(1)}\rangle_0,$$

we remove the t_1 eigenvalue from the perturbation and include it in the zeroth-order Hamiltonian k. From the definition (2.1) of $H_{\alpha\beta}^{(1)}$ we have

$$E_\alpha^{(1)} = -(\phi_\alpha, \vec{d}\phi_\alpha)\cdot\langle\vec{\mathcal{E}}(t_0)\rangle_0;$$

therefore $E_\alpha^{(1)} = 0$ for fields that have no static component. This is true for the fields of interest to us, and $M^{(1)} = 0$ will be assumed from now on.

We must now choose $R^{(1)}$ so that quasisecular behavior in $C^{(1)}$ is prohibited. The equation for $C^{(1)}$ is, after substituting (2.9) into (2.7),

$$i\partial_0 C^{(1)} = (H^{(1)} + R^{(1)} - \langle H^{(1)} + R^{(1)}\rangle_0)C^{(0;1)}.$$

The troublesome terms are in $H^{(1)}$ so we choose $R^{(1)}$ as

$$R^{(1)} = \mathcal{P}H^{(1)}(\lambda^{-1}t_1) - \mathcal{P}H^{(1)}(t_0),$$

which satisfies (2.8) and has the effect of substituting $\lambda^{-1}t_1$ for t_0 in the low-frequency part of $H^{(1)}$. Thus

$$H^{(1)} + R^{(1)} = (1-\mathcal{P})H^{(1)}(t_0) + \mathcal{P}H^{(1)}(\lambda^{-1}t_1),$$

and

$$\langle H^{(1)} + R^{(1)}\rangle_0 = \mathcal{P}H^{(1)}(\lambda^{-1}t_1).$$

This choice for $R^{(1)}$ replaces the low-frequency part of $H^{(1)}$, which is the source of quasisecular behavior, by an operator independent of t_0. The t_0 quasisecularities are thereby turned into t_0 secularities which are automatically eliminated.

With the notation

$$H^{(1;0)}(t_0) = (1-\mathcal{P})H^{(1)}(t_0),$$

$$H^{(1;1)}(t_1) = \mathcal{P}H^{(1)}(\lambda^{-1}t_1), \tag{2.10}$$

the equations for $C^{(1)}$ and $C^{(0)}$ become

$$i\partial_1 C^{(0;1)} = H^{(1;1)}C^{(0;1)}, \tag{2.11}$$

$$i\partial_0 C^{(1)} = H^{(1;0)}C^{(0;1)}, \tag{2.12}$$

where we have used $M^{(1)} = 0$. The Schrödinger equation (2.11) with effective Hamiltonian $H^{(1;1)}$ is the same as the equation obtained by neglecting the counterrotating terms in the language of the conventional rotating-wave approximation. The solution of (2.11) is conveniently described by the usual unitary time-evolution operator $U^{(1)}(t_1)$:

$$C^{(0;1)} = U^{(1)}C^{(0;2)}, \quad i\partial_1 U^{(1)} = H^{(1;1)}U^{(1)}, \quad U^{(1)}(0) = 1. \tag{2.13}$$

Thus the t_1 dependence is entirely contained in $U^{(1)}$ since $C^{(0;2)}$ depends only on $\vec{t}_2$. We now substitute this result in (2.12) and solve for $C^{(1)}$:

$$C^{(1)} = C^{(1;1)} + \frac{1}{i}\int_0^{t_0} dt_0' H^{(1;0)}(t_0')C^{(0;1)}. \tag{2.14}$$

Integrals of this form will occur repeatedly in the later stages of the solution so it is useful to introduce a general notation at this point. In solving the rth equation we will construct operators $H^{(r;s)}$ $(0 \leq s \leq r)$ analogous to $H^{(1;1)}$ and $H^{(1;0)}$; for each of these we define the operators

$$I^{(r;s)} \equiv \frac{1}{i}\int_0^{t_s} dt_s' H^{(r;s)}(t_s'),$$

$$J^{(r;s)} \equiv \int \frac{d\omega}{2\pi}\frac{1}{\omega}\hat{H}^{(r;s)}(\omega)e^{-i\omega t_s},$$

which are related by

$$I^{(r;s)} = J^{(r;s)} - J_0^{(r;s)}, \quad J_0^{(r;s)} = J^{(r;s)}(0).$$

We should emphasize that $H^{(r;s)}$ will be constructed so that it is independent of t_n $(n \neq s)$. Now consider the $n = 2$ equation of (2.4) which determines the unknown functions $C^{(0;2)}$ and $C^{(1;1)}$ appearing in the solution of the $n = 1$ equation:

$$i\partial_0 C^{(2)} + i\partial_1 C^{(1)} + i\partial_2 C^{(0;1)} = (Q^{(1)} - M^{(1)})C^{(1)} + (Q^{(2)} - M^{(2)})C^{(0)}.$$

Substitution of the previous results into this equation yields

$$i\partial_0 C^{(2)} + (i\partial_1 - H^{(1;0)} - H^{(1;1)})C^{(1;1)} + i\partial_2 C^{(0;1)} = (W^{(2)} - M^{(2)} + Q^{(2)})C^{(0;1)}, \tag{2.15}$$

where

$$W^{(2)} = H^{(1;0)}I^{(1;0)} + [H^{(1;1)}, I^{(1;0)}] = H^{(1;0)}J^{(1;0)} - H^{(1;0)}J_0^{(1;0)} + [H^{(1;1)}, J^{(1;0)}] - [H^{(1;1)}, J_0^{(1;0)}]. \tag{2.16}$$

In this expression only the first term can be a source of t_0 quasisecularity, since the second and

third terms contain no low frequencies and the fourth term is independent of t_0. The quasisecular behavior of the first term is eliminated by choosing

$$Q^{(2)} = \mathcal{P}(H^{(1;0)}J^{(1;0)})(\lambda^{-1}t_1) - \mathcal{P}(H^{(1;0)}J^{(1;0)})(t_0) + R^{(2)},$$

where $R^{(2)}$ satisfies (2.3); this yields

$$H^{(1;0)}J^{(1;0)} + Q^{(2)} = (1-\mathcal{P})(H^{(1;0)}J^{(1;0)})(t_0) + \mathcal{P}(H^{(1;0)}J^{(1;0)})(\lambda^{-1}t_1) + R^{(2)}.$$

The dependence of the third term in (2.16) on both t_1 and t_0, would cause some difficulties later. To prevent this we put

$$R^{(2)} = [H^{(1;1)}(\lambda t_0), J^{(1;0)}(t_0)] - [H^{(1;1)}(t_1), J^{(1;0)}(t_0)] + S^{(2)},$$

where $S^{(2)}$ satisfies (2.3). Thus the t_1 dependence of the third term is changed back to a t_0 dependence. In order to see that this procedure does not produce any new quasisecularities, we write out the resulting term:

$$[H^{(1;1)}(\lambda t_0), J^{(1;0)}(t_0)] = \int \frac{d\omega'}{2\pi} \int \frac{d\omega}{2\pi} \frac{1}{\omega} [\hat{H}^{(1;1)}(\omega'), \hat{H}^{(1;0)}(\omega)] e^{-i(\lambda\omega' + \omega)t_0}.$$

The definitions, (2.10), of $H^{(1;1)}$ and $H^{(1;0)}$ guarantee that the integral is constrained to a region with $\omega' \leq \bar{\omega}$ and $\omega \gg \lambda\bar{\omega}$; consequently no small denominator can arise from t_0 integration. At this point we have

$$\begin{aligned} W^{(2)} + Q^{(2)} &= H^{(2;0)} + X^{(2)} + S^{(2)}, \\ H^{(2;0)} &= (1-\mathcal{P})(H^{(1;0)}J^{(1;0)}) - H^{(1;0)}J_0^{(1;0)} + [H^{(1;1)}(\lambda t_0), J^{(1;0)}], \\ X^{(2)} &= \mathcal{P}(H^{(1;0)}J^{(1;0)})(\lambda^{-1}t_1) - [H^{(1;1)}, J_0^{(1;0)}]. \end{aligned} \tag{2.17}$$

Note that $X^{(2)}$ when regarded as a function of the physical time t has no Fourier components for frequencies exceeding $\lambda\bar{\omega}$. Substitution of these results into (2.15) gives us

$$i\partial_0 C^{(2)} + (i\partial_1 - H^{(1;0)} - H^{(1;1)})C^{(1;1)} + i\partial_2 C^{(0;1)} = (H^{(2;0)} + X^{(2)} - M^{(2)} + S^{(2)})C^{(0;1)}. \tag{2.18}$$

Since $H^{(2;0)}$ has no static part, the static part of $X^{(2)}_{\alpha\alpha}$ plays the same role for the $n=2$ equation that the static part of $H^{(1)}_{\alpha\alpha}$ played for the $n=1$ equation; therefore we define $M^{(2)}$ by

$$E^{(2)}_\alpha = \langle X^{(2)}_{\alpha\alpha} \rangle_1. \tag{2.19}$$

We will show later that $E^{(2)}_\alpha$ is real. With this choice for $M^{(2)}$ we impose $\langle \partial_0 C^{(2)} \rangle_0 = 0$ on the t_0 average of (2.18) in order to eliminate t_0 secularity; this yields

$$(i\partial_1 - H^{(1;1)})C^{(1;1)} + i\partial_2 C^{(0;1)} = (X^{(2)} - M^{(2)} + S^{(2)})C^{(0;1)}, \tag{2.20}$$

where we have used $\langle H^{(2;0)} \rangle_0 = \langle H^{(1;0)} \rangle_0 = 0$, together with the assumption that $S^{(2)}$ is independent of t_0. A further simplification results from introducing

$$D^{(1;1)} = U^{(1)\dagger} C^{(1;1)},$$

using (2.13), and then rewriting (2.20) as

$$i\partial_1 D^{(1;1)} + i\partial_2 C^{(0;2)} = U^{(1)\dagger}(X^{(2)} - M^{(2)} + S^{(2)})U^{(1)} C^{(0;2)}. \tag{2.21}$$

At this stage we are faced with possible t_1 quasisecularities which are eliminated by the final choice

$$S^{(2)} = U^{(1)}\{\mathcal{P}[U^{(1)\dagger}(X^{(2)} - M^{(2)})U^{(1)}](\lambda^{-1}t_2) - \mathcal{P}[U^{(1)\dagger}(X^{(2)} - M^{(2)})U^{(1)}](t_1)\} U^{(1)\dagger},$$

which allows (2.21) to be cast in the form

$$i\partial_1 D^{(1;1)} + i\partial_2 C^{(0;2)} = (H^{(2;1)} + H^{(2;2)})C^{(0;2)}, \tag{2.22}$$

where

$$H^{(2;1)} = (1-\mathcal{P})[U^{(1)\dagger}(X^{(2)} - M^{(2)})U^{(1)}](t_1), \tag{2.23}$$

$$H^{(2;2)} = \mathcal{P}[U^{(1)\dagger}(X^{(2)} - M^{(2)})U^{(1)}](\lambda^{-1}t_2). \tag{2.24}$$

The operator $H^{(2;2)}$ when regarded as a function of the physical time t has no Fourier components for frequencies exceeding $\lambda^2\bar{\omega}$.

The condition which prohibits t_1 secularities comes from the t_1 average of (2.22) together with $\langle H^{(2;1)} \rangle_1 = 0$; it is

$$i\partial_2 C^{(0;2)} = H^{(2;2)} C^{(0;2)}. \tag{2.25}$$

Thus $H^{(2;2)}$ is an effective Hamiltonian determining the t_2 dependence of $C^{(0)}$. This interpretation requires that $H^{(2;2)}$ be Hermitian; unfortunately this is not obvious from an inspection of the definition, in contrast to the case of $H^{(1;1)}$. From (2.24) we see that $H^{(2;2)}$ is Hermitian whenever $X^{(2)}$ is, and from (2.17) together with the fact that $J^{(1;0)}$ is anti-Hermitian it is easily seen that the anti-Hermitian part of $X^{(2)}$ comes entirely from the first term:

$$X^{(2)}_A = X^{(2)} - X^{(2)\dagger} = \mathcal{P}(H^{(1;0)}J^{(1;0)})_A(\lambda^{-1}t_1).$$

We proceed by showing that $(H^{(1;0)}J^{(1;0)})_A$ is annihilated by the $\mathcal{P}$ operation. Again using anti-Hermiticity of $J^{(1;0)}$ we have

$$(H^{(1;0)}J^{(1;0)})_A = H^{(1;0)}J^{(1;0)} + J^{(1;0)}H^{(1;0)}$$

$$= \int \frac{d\omega}{2\pi} \int \frac{d\omega'}{2\pi} \left(\hat{H}^{(1;0)}(\omega) \frac{\hat{H}^{(1;0)}(\omega')}{\omega'} + \frac{\hat{H}^{(1;0)}(\omega')}{\omega'} \hat{H}^{(1;0)}(\omega) \right) e^{-i(\omega+\omega')t_0}$$

$$= \int \frac{d\omega}{2\pi} \int \frac{d\omega'}{2\pi} \frac{\hat{H}^{(1;0)}(\omega)}{\omega} \frac{\hat{H}^{(1;0)}(\omega')}{\omega'} (\omega + \omega') e^{-i(\omega+\omega')t_0}$$

$$= \int \frac{d\omega}{2\pi} \omega \left(\int \frac{d\omega'}{2\pi} \frac{\hat{H}^{(1;0)}(\omega - \omega')}{\omega - \omega'} \frac{\hat{H}^{(1;0)}(\omega')}{\omega'} \right) e^{-i\omega t_0}.$$

The last equation shows that the Fourier transform of $(H^{(1;0)}J^{(1;0)})_A$ is of the form (2.6); therefore, $\mathcal{P}(H^{(1;0)}J^{(1;0)})_A = 0$. The Hermiticity of $X^{(2)}$ guarantees the reality of $E_\alpha^{(2)}$ and the Hermiticity of $H^{(2;2)}$. This allows us to express the solution of (2.25) by

$$C^{(0;2)} = U^{(2)}C^{(0;3)}, \quad i\partial_2 U^{(2)} = H^{(2;2)}U^{(2)},$$
$$U^{(2)}(0) = 1. \tag{2.26}$$

Returning to (2.21) we solve for $D^{(1;1)}$ and get

$$C^{(1;1)} = U^{(1)}(D^{(1;2)} + I^{(2;1)}C^{(0;2)}).$$

The last step in the solution of the $n = 2$ equation is to determine the t_0 dependence of $C^{(2)}$ by integrating (2.18), after substitution of the various results obtained above. The solution is

$$C^{(2)} = C^{(2;1)} + (I^{(2;0)} + I^{(1;0)}U^{(1)}I^{(2;1)}U^{(1)\dagger})C^{(0;1)} + I^{(1;0)}U^{(1)}D^{(1;2)},$$

where the functions $C^{(0;3)}$, $C^{(2;1)}$, and $D^{(1;2)}$ are still undetermined.

We have carried out this rather elaborate argument in order to illustrate in detail the mechanisms involved in the elimination of secularities and quasisecularities and also the proper choice of the level shifts; having done so, we can now state the prescription for determining the solution to any required accuracy. In the first place, the only purpose served by the $Q^{(n)}$'s is to switch dependence from one time variable to another; therefore we can replace the explicit choices for the $Q^{(n)}$'s by the following rule:

Substitution rule: In any of the operators occurring in the Eqs. (2.4) a dependence on t_k may be replaced by a dependence on $\lambda^{k-l}t_l$.

Now suppose we want the solution correct to $O(\lambda^k)$ with the time dependence correct through t_l [we will call this the (λ^k, t_l) order]; the procedure is as follows:

(1) Truncate (2.4) at $n = k + l$ and impose the boundary conditions

$$C^{(m;j)} = 0, \quad 1 \leq m \leq k, \quad j \geq l+1$$
$$C^{(0;j)} = C(0), \quad j \geq l+1.$$

(2) Solve the set of $n+1$ equations recursively: (a) Assuming that equations 0, 1, ..., $r-1$ have been solved, write their solutions in the form

$$C^{(s)} = C^{(s;r-s)} + F^{(s)}C^{(0;r)}, \quad 1 \leq s \leq r-1$$
$$C^{(0)} = U^{(r-1)} \cdots U^{(1)}C^{(0;r)},$$

and substitute into equation r to get

$$\partial_0 C^{(r)} + \sum_{j=1}^{r-1} \partial_{r-j} C^{(j;r-j)} + U^{(n-1)} \cdots U^{(1)} \partial_n C^{(0)}$$
$$= (W^{(n)} - M^{(n)})C^{(0)} + (H^{(1)} - M^{(1)})C^{(r-1)} - \sum_{j=2}^{r-1} M^{(j)}C^{(r-j;j)}.$$

(b) Choose $M^{(n)}$ as the static diagonal part of $W^{(n)}$. (c) Successively eliminate secularities and quasisecularities in t_0, t_1, ..., t_{r-1} from the rth equation. (i) Assuming this has been done for t_0, ..., t_{l-1} we are left with an equation having ∂_l as the lowest-order time derivative. Use the substitution rule to eliminate t_l quasisecularities and terms depending jointly on t_l and $\vec{\mathcal{E}}_{l+1}$. (ii) Impose the auxiliary condition to eliminate t_l secularities. (iii) Iterate (i) and (ii) until the final equation

$$\partial_r C^{(0;r)} = H^{(r;r)}C^{(0;r)}$$

is obtained. (d) Iterate (a), (b), and (c) up to $r = k + l$.

Finally we would like to point out that the rather complicated appearance of the multiple-time-scale method is largely due to the fact that we have dealt with the most general case. In particular, we have made no assumptions regarding the shape of the laser pulse; i.e., the form of the function $\vec{\mathcal{E}}(t)$. In the specific examples discussed in Secs. IV and V, where a simple square pulse is assumed, we will see that this technique is really straightforward in practice.

III. CONSERVATION OF PROBABILITY

In this section we will show that the inner product between two solutions of (2.2), both calculated to order λ^n, is conserved in time with an error of order λ^{n+1}. Let B and C be solutions of (2.2), then

$$i\frac{\partial}{\partial t}(B,C)=\sum_{n=0}^{\infty}\lambda^n i\partial_n(B,C)\,|_{\mathrm{PL}}=\sum_{n=0}^{\infty}\lambda^n[(B,i\partial_n C)-(i\partial_n B,C)]_{\mathrm{PL}}$$

$$=\Big[\sum_{k=0}^{\infty}\lambda^k\Big(B,\sum_{m=0}^{k}i\partial_{k-m}C^{(m)}\Big)-\sum_{k=0}^{\infty}\lambda^k\Big(\sum_{m=0}^{k}i\partial_{k-m}B^{(m)},C\Big)\Big]_{\mathrm{PL}}.$$

Now we can use (2.4) to get

$$i\frac{\partial}{\partial t}(B,C)=\Big[\sum_{k=0}^{\infty}\lambda^k\Big(B,\sum_{m=0}^{k}(Q^{(k-m)}-M^{(k-m)})C^{(m)}\Big)-\sum_{k=0}^{\infty}\lambda^k\Big(\sum_{m=0}^{k}(Q^{(k-m)}-M^{(k-m)})B^{(m)},C\Big)\Big]_{\mathrm{PL}}.$$

After substituting the expansion for B in the first term, for C in the second, and suitably renaming the summation variables we find

$$i\frac{\partial}{\partial t}(B,C)=\sum_{n=1}^{\infty}\lambda^n(B,[Q^{(n)}-Q^{(n)\dagger}]C)_{\mathrm{PL}}=0,$$

where the last equality follows from the conditions (2.3) and the Hermiticity of $M^{(n)}$. Thus we have established that

$$\sum_{n=0}^{\infty}\lambda^n\sum_{m=0}^{n}(B^{(n-m)},C^{(m)})=(B(0),C(0)).$$

Since the zeroth-order coefficients $B^{(0)}$ and $C^{(0)}$ develop in time by a product of unitary operators $U^{(1)}U^{(2)}\ldots$, the $n=0$ term on the left-hand side cancels the initial value on the right-hand side. However, we must recall that the coefficients $C^{(m)}$ depend on λ; therefore we cannot set to zero the coefficient of each λ^n for $n\geq 1$. Fortunately we can do something just as satisfactory. We have a condition of the form

$$\sum_{n=1}^{\infty}\lambda^n f^{(n)}(\lambda)=0, \tag{3.1}$$

where

$$f^{(n)}(\lambda)\equiv\sum_{m=0}^{n}(B^{(n-m)},C^{(m)}). \tag{3.2}$$

The method used to generate the solutions guarantees that the amplitudes $C^{(m)}$ (and $B^{(m)}$) are bounded by

$$|C_\alpha^{(m)}|\leq G,$$

where G is a constant satisfying $\lambda G\ll 1$; therefore $f^{(n)}$ is bounded by

$$|f^{(n)}|\leq NG^2(n+1), \tag{3.3}$$

where N is the number of components of C_α. From (3.1) we have

$$\sum_{k=1}^{n}\lambda^k f^{(k)}(\lambda)=-\sum_{k=n+1}^{\infty}\lambda^k f^{(k)}(\lambda), \tag{3.4}$$

and by (3.3) the sum on the right-hand side is bounded by

$$NG^2\sum_{k=n+1}^{\infty}(k+1)\lambda^k=NG^2\Big(\frac{(n+2)}{1-\lambda}+\frac{\lambda}{(1-\lambda)^2}\Big)\lambda^{n+1}$$

$$=O(\lambda^{n+1}). \tag{3.5}$$

Combining (3.4) with (3.5) and the definition of $f^{(k)}$ we arrive at the conclusion stated at the beginning of this section:

$$\Big(\sum_{k=0}^{n}\lambda^k B^{(k)},\sum_{l=0}^{n}\lambda^l C^{(l)}\Big)=(B(0),C(0))+O(\lambda^{n+1}).$$

IV. BLOCH-SIEGERT THEORY

We now use the method presented in Sec. II to construct a generalization of the Bloch-Siegert theory of the Rabi oscillations and resonance shift for a two-level system. Since our method is applicable to any N-level system, we do not make use of the isomorphism between spin-$\frac{1}{2}$ and two-level systems which is required for the application of the original Bloch-Siegert theory.[1] Consequently, the contributions of nonresonant levels to the frequency shifts are automatically included. A treatment of the two-level problem using a technique closely related to ours has been given by Shirley.[8]

Consider an N-level system interacting with the field $\vec{\mathcal{E}}(t)=\vec{\mathcal{E}}_0\cos\omega t$ through the Hamiltonian

$$H_{\alpha\beta}^{(1)}=V_{\alpha\beta}e^{iE_{\alpha\beta}t}\cos\omega t,$$
$$V_{\alpha\beta}=(\phi_\alpha,-\vec{\mathcal{E}}_0\cdot\vec{d}\phi_\beta). \tag{4.1}$$

We assume that ω satisfies

$$|E_{10}^{(0)}-\omega|=O(\lambda)\omega$$

and (4.2)

$$|E_{\alpha\beta}^{(0)}-n\omega|\gg\lambda\omega,$$

for all other combinations $\alpha\beta$ and $n\geq 1$. In other words the only resonance is the single photon resonance with the $0\rightarrow 1$ transition. We will solve this problem in the (λ^0,t_2) order; i.e., we neglect $C^{(k)}$ for $k\geq 1$ and determine $C^{(0)}$ as a function of t_0, t_1, and t_2. The t_2 dependence is required for the correct description of the Bloch-Siegert level shifts. The first step is to split $H^{(1)}$ into high- and low-

frequency parts and thereby calculate $H^{(1;1)}$ and $H^{(1;0)}$. Since $\vec{\mathcal{E}}(t)$ has a vanishing time average we have $E_\alpha^{(1)}=0$; consequently,

$$E_{\alpha\beta}-\omega=E_{\alpha\beta}^{(0)}-\omega+\lambda^2E_{\alpha\beta}^{(2)}+\ldots,$$

so that the conditions (4.2) are also satisfied when $E_{\alpha\beta}^{(0)}$ is replaced by the exact $E_{\alpha\beta}$. Thus we can pick out the low-frequency terms despite the fact that $E_\alpha^{(2)}$ is not yet determined. From (4.1) and (4.2) with $E_{\alpha\beta}^{(0)}\to E_{\alpha\beta}$ we see that the low-frequency part $H^{(1;1)}$, defined by (2.10), has the matrix elements

$$H_{10}^{(1;1)}(t_1)=\tfrac{1}{2}V_{10}e^{i\nu t_1}=[H_{01}^{(1;1)}]^*,$$
$$H_{\alpha\beta}^{(1;1)}=0,\quad \{\alpha,\beta\}\neq\{0,1\},\tag{4.3}$$

where

$$\nu\equiv\lambda^{-1}(E_{10}-\omega).$$

The remaining high-frequency terms give the operator $H^{(1;0)}$, defined by (2.10), as

$$H_{10}^{(1;0)}=\tfrac{1}{2}V_{10}e^{-i(\omega+E_{10})t_0}=[H_{01}^{(1;0)}]^*,$$
$$H_{\alpha\beta}^{(1;0)}=H_{\alpha\beta}^{(1)},\quad \{\alpha,\beta\}\neq\{0,1\}.\tag{4.4}$$

The Schrödinger equation (2.11) with this $H^{(1;1)}$ reduces the problem to a pure two-level system described in the rotating-wave approximation. The nonresonant levels and the shifts in the energies of the resonant levels are neglected in this approximation. Since the unitary operator $U^{(1)}$ is the identity except for the subspace spanned by ϕ_0 and ϕ_1, it is convenient to represent the nontrivial part of $U^{(1)}$ in terms of Pauli matrices,

$$U^{(1)}=e^{-i(\nu/2)t_1\sigma_3}[\cos(\eta t_1)-i\sin(\eta t_1)F],$$
$$F=(-\nu/2\eta)\sigma_3+(V_{10}/2\eta)\sigma_1,\tag{4.5}$$
$$2\eta=(|V_{10}|^2+\nu^2)^{1/2},$$

where $\sigma_1,\sigma_2,\sigma_3$ are the standard Pauli matrices. In deriving this result we have made the simplifying, but not essential, assumption that the matrix elements $V_{\alpha\beta}$ are real.

In order to obtain the Bloch-Siegert shift in the resonance frequency we have to find the t_2 dependence of $C^{(0)}$ from (2.26). The second-order shift $E_\alpha^{(2)}$ and the effective Hamiltonian $H^{(2;2)}$ are both determined by the quantity $X^{(2)}$, defined by (2.17); this in turn involves $J^{(1;0)}$ which, in the present case, is given by

$$J_{10}^{(1;0)}=\frac{1}{2}V_{10}\frac{e^{-i(\omega+E_{10})t_0}}{\omega+E_{10}}=-[J_{01}^{(1;0)}]^*,$$

and

$$J_{\alpha\beta}^{(1;0)}=-\frac{1}{2}V_{\alpha\beta}\left(\frac{e^{i(E_{\alpha\beta}+\omega)t_0}}{E_{\alpha\beta}+\omega}+\frac{e^{i(E_{\alpha\beta}-\omega)t_0}}{E_{\alpha\beta}-\omega}\right),\tag{4.6}$$

for all other $\alpha\beta$. Since $H^{(1;0)}$ and $J^{(1;0)}$ both have nonvanishing matrix elements between resonant and nonresonant states, the nonresonant states can contribute to the low-frequency part of the product. From (4.4) and (4.6) it is easy to verify that only the diagonal elements of the product can have low-frequency behavior; in fact we find

$$\mathcal{P}(H^{(1;0)}J^{(1;0)})=\tfrac{1}{2}(\Delta_0+\Delta_1)+\tfrac{1}{2}(\Delta_0-\Delta_1-2\xi)\sigma_3$$

for the (00) and (11) elements, and

$$\mathcal{P}(H^{(1;0)}J^{(1;0)})_{\alpha\beta}=\delta_{\alpha\beta}\Delta_\beta$$

for all other $\alpha\beta$, where

$$\xi=\frac{|\frac{1}{2}V_{10}|^2}{\omega+E_{10}},$$

$$\Delta_\alpha=\frac{1}{2}{\sum_\beta}'\frac{E_{\alpha\beta}|V_{\alpha\beta}|^2}{(E_{\alpha\beta})^2-\omega^2},$$

and $\sum'$ means to omit any term with a resonant denominator. Since the commutator term in (2.17) contains $H^{(1;1)}$ it has no matrix elements except for those connecting the resonant levels. The relevant part of $J_0^{(1;0)}$ is given by

$$J_0^{(1;0)}=\frac{1}{2}\frac{iV_{10}}{\omega+E_{10}}\sigma_2,$$

while $H^{(1;1)}$ can be expressed as

$$H^{(1;1)}=\tfrac{1}{2}V_{10}[\cos(\nu t_1)\sigma_1+\sin(\nu t_1)\sigma_2],\tag{4.7}$$

so that

$$[H^{(1;1)},J_0^{(1;0)}]=-2\xi\cos(\nu t_1)\sigma_3,$$
$$X^{(2)}=\tfrac{1}{2}(\Delta_0+\Delta_1)+\tfrac{1}{2}(\Delta_0-\Delta_1-2\xi)\sigma_3+2\xi\cos(\nu t_1)\sigma_3\tag{4.8}$$

for $\{\alpha,\beta\}=\{0,1\}$, and $X_{\alpha\beta}^{(2)}=\delta_{\alpha\beta}\Delta_\alpha$ for all other $\alpha\beta$. Combining (4.8) and (2.19) we find the second-order level shift

$$E_0^{(2)}=\Delta_0-\xi,\quad E_1^{(2)}=\Delta_1+\xi,\quad E_\alpha^{(2)}=\Delta_\alpha,\quad \alpha\geq 2.$$

In order to get $H^{(2;2)}$ from (2.24) we first need $X^{(2)}-M^{(2)}$, which is given by

$$X^{(2)}-M^{(2)}=2\xi\cos(\nu t_1)\sigma_3,$$

with all other matrix elements vanishing. Using (4.5) for $U^{(1)}$ we then have

$$U^{(1)\dagger}(X^{(2)}-M^{(2)})U^{(1)}=2\xi\left[2\left(\frac{\nu}{2\eta}\right)\sigma_3-\frac{V_{10}}{2\eta}\sigma_1\right]\frac{\nu}{2\eta}\cos(\nu t_1)+\xi\frac{V_{10}}{\eta}\sigma_2\cos(\nu t_1)\sin(2\eta t_1)$$
$$+2\xi\left[1-2\left(\frac{\nu}{2\eta}\right)^2\sigma_3+\frac{\nu}{2\eta}\frac{V_{10}}{2\eta}\sigma_1\right]\cos(\nu t_1)\cos(2\eta t_1).$$

The first term satisfies (2.6) and is discarded by the $\mathcal{P}$ operation. The other terms could contribute only if $(2\eta)^2 \cong \nu^2$, but this is impossible since $(2\eta)^2 - \nu^2 = |V_{10}|^2$. Thus we find that $H^{(2;2)}$ vanishes, and $U^{(2)}$ is the identity.

This completes the calculation in the (λ^0, t_2) order; the solution is

$$C^{(0;1)} = U^{(1)} U^{(2)} C(0),$$

where $U^{(1)}$ is given by (4.5) and $U^{(2)} = 1$. With the assumption that the system is initially in the ϕ_0 state we can easily calculate the occupation probability $n_1 = |C_1^{(0;1)}|^2$. This expression must be evaluated on the physical line by setting $t_1 = \lambda t$; the strength parameter λ can then be recombined with V_{10} to give

$$n_1 = \frac{|V_{10}|^2}{2\eta} \sin^2 \eta t, \quad 2\eta = [\,|V_{10}|^2 + (E_{10} - \omega)^2\,]^{1/2}, \qquad E_{10} = E_{10}^{(0)} + E_{10}^{(2)}. \tag{4.9}$$

In this approximation the population oscillates between levels 0 and 1 with frequency 2η. The amplitude for this oscillation has its maximum when $\omega = E_{10}$; however, E_{10} is itself a function of ω, so this is really a dispersion relation for ω, which can be written as

$$\begin{aligned}\omega &= E_{10}^{(0)} + 2\xi + \Delta_1 - \Delta_0, \\ &= E_{10}^{(0)} + \frac{1}{2}\frac{|V_{10}|^2}{E_{10} + \omega} + \frac{1}{2}\sum_{\beta}{}' \frac{E_{1\beta}|V_{1\beta}|^2}{(E_{1\beta})^2 - \omega^2} \\ &\quad - \frac{1}{2}\sum_{\beta}{}' \frac{E_{0\beta}|V_{0\beta}|^2}{(E_{0\beta})^2 - \omega^2}.\end{aligned}$$

The solution will be denoted by ω^*. The lowest-order correction to the bare resonance frequency $E_{10}^{(0)}$ is obtained by putting $E_{\alpha\beta} = E_{\alpha\beta}^{(0)}$ and $\omega = E_{10}^{(0)}$ on the right-hand side:

$$\delta\omega = \omega^* - E_{10}^{(0)} \cong \frac{|V_{10}|^2}{4E_{10}^{(0)}} + \frac{1}{2}\sum_{\beta}{}' \frac{E_{1\beta}^{(0)}|V_{1\beta}|^2}{(E_{1\beta}^{(0)})^2 - (E_{10}^{(0)})^2} - \frac{1}{2}\sum_{\beta}{}' \frac{E_{0\beta}^{(0)}|V_{0\beta}|^2}{(E_{0\beta}^{(0)})^2 - (E_{10}^{(0)})^2}.$$

The first term is the standard Bloch-Siegert shift, and the other terms give the contributions due to the nonresonant levels. Corrections to the amplitude due to the counterrotating terms could be calculated by evaluating (2.14) to get $C^{(1)}$ but we will not pursue this matter here.

V. COHERENT TWO-PHOTON RESONANCES

The coherent resonant excitation, or Rabi oscillation, discussed in Sec. IV is associated with the microscopic processes of resonant emission and absorption of single photons. In this section the Rabi oscillation associated with resonant two-photon[9] emission and absorption will be calculated in the (λ^0, t_2) order. In addition to demonstrating the effect we will use this calculation as a test case by comparing the results to an exact numerical computation.

The notation of Sec. IV will be used, but the condition (4.2) is replaced by

$$|E_{10}^{(0)} - 2\omega| = O(\lambda^2)\omega, \tag{5.1}$$

which is the two-photon resonance condition. To simplify matters we assume that only this resonance is present. In this case it is easy to see that the t_1 Hamiltonian $H^{(1;1)}$ must vanish so that $U^{(1)}$ is the identity. Consequently $H^{(1;0)} = H^{(1)}$ and $J^{(1;0)}$ is given by (4.6), which is now valid for all $\alpha\beta$. We proceed to the calculation of $U^{(2)}$. According to (2.24) we have

$$H^{(2;2)} = \mathcal{P}(X^{(2)} - M^{(2)})(\lambda^{-1}t_2),$$

and by (2.17)

$$X^{(2)} = \mathcal{P}(H^{(1)}J^{(1;0)})(\lambda^{-1}t_1).$$

By using the explicit expressions for $H^{(1)}$ and $J^{(1;0)}$ we get

$$(H^{(1)}J^{(1;0)})_{\alpha\beta} = -\frac{1}{4}\sum_{\gamma}\left(\frac{2E_{\gamma\beta}}{(E_{\gamma\beta})^2 - \omega^2} e^{iE_{\alpha\beta}t_0} + \frac{e^{i(E_{\alpha\beta}+2\omega)t_0}}{E_{\gamma\beta} + \omega} + \frac{e^{i(E_{\alpha\beta}-2\omega)t_0}}{E_{\gamma\beta} - \omega}\right) V_{\alpha\gamma} V_{\gamma\beta}.$$

According to the general result proved in Sec. II we can replace this expression by its Hermitian part before applying the $\mathcal{P}$ operation. The result is

$$\begin{aligned}X_{\alpha\alpha}^{(2)} &= \Delta_\alpha, \\ X_{10}^{(2)} &= \frac{1}{8}\sum_{\beta}\frac{(2E_\beta - E_1 - E_0)V_{1\beta}V_{\beta 0}}{(\omega + E_{\beta 1})(\omega - E_{\beta 0})} e^{i[(E_{10}-2\omega)/\lambda]t_1} \\ &= [X_{01}^{(2)}]^*,\end{aligned}$$

with all other matrix elements vanishing. The level shifts are given by

$$E_\alpha^{(2)} = \langle X_{\alpha\alpha}^{(2)} \rangle_1 = \Delta_\alpha;$$

consequently only the (01) and (10) elements of $X^{(2)} - M^{(2)}$ are nonvanishing. By virtue of the resonance condition (5.1), we have $(E_{10} - 2\omega)/\lambda = O(\lambda)\omega$; therefore the $\mathcal{P}$ operation does nothing to $X^{(2)}$, and $H^{(2;2)}$ can be written as

$$H^{(2;2)} = \tfrac{1}{2}\Gamma[\cos(\rho t_2)\sigma_1 + \sin(\rho t_2)\sigma_2],$$

with

$$\Gamma \equiv \frac{1}{4}\sum_{\beta}\frac{(2E_\beta - E_1 - E_0)V_{1\beta}V_{\beta 0}}{(\omega + E_{\beta 1})(\omega - E_{\beta 0})}, \tag{5.2}$$

$$\rho \equiv (E_{10} - 2\omega)/\lambda^2.$$

It should be emphasized that the resonance condition (5.1) for the two-photon case is more stringent than the corresponding condition (4.2) for the one-photon case. If we were to use the weaker condition $E_{10}^{(0)} - 2\omega = O(\lambda)$ [so that $(E_{10}^{(0)} - 2\omega)/\lambda = O(1)$] there would be no low-frequency term in $X^{(2)}(t_1)$. This would in turn give $H^{(2;2)} = 0$; consequently $C^{(0)}$ would be independent of time. In this case the oscillations between levels 0 and 1 would first occur in $C^{(1)}$; therefore the amplitude of n_1 would be $O(\lambda^2)$, rather than $O(1)$.

The two-photon problem has now been cast in the same form as the one-photon problem; in fact $H^{(2;2)}$ is obtained from Eq. (4.7) for $H^{(1;1)}$ by $\nu \to \rho$ and $V_{10} \to \Gamma$. This transformation applied to (4.9) gives the solution for $n_1(t)$:

$$n_1 = |\Gamma/2\eta|^2 \sin^2\eta t\,, \quad 2\eta = [\Gamma^2 + (E_{10} - 2\omega)^2]^{1/2}\,,$$
$$E_{10} = E_{10}^{(0)} + \Delta_1 - \Delta_0\,. \tag{5.3}$$

The resonance frequency ω^* is defined as the solution of $\omega = \frac{1}{2}E_{10}(\omega)$; thus the lowest-order shift in the resonance is given by

$$\delta\omega^* = \omega^* - \tfrac{1}{2}E_{10}^{(0)}$$
$$\cong \frac{1}{2}\sum_{\beta} \frac{E_{1\beta}^{(0)}|V_{1\beta}|^2}{(E_{1\beta}^{(0)})^2 - (\frac{1}{2}E_{10}^{(0)})^2} - \frac{1}{2}\sum_{\beta} \frac{E_{0\beta}^{(0)}|V_{0\beta}|^2}{(E_{0\beta}^{(0)})^2 - (\frac{1}{2}E_{10}^{(0)})^2}$$
$$= \frac{4}{3}\frac{|V_{10}|^2}{E_{10}^{(0)}} + \Delta_1' - \Delta_0'\,, \tag{5.4}$$

where Δ_0', Δ_1' are obtained from Δ_0, Δ_1 by dropping the 0 and 1 terms from the summations and putting $\omega = \frac{1}{2}E_{10}^{(0)}$. The first term in (5.4) evidently represents the resonance shift for a pure two-level system, and the remaining terms give the corrections due to the nonresonant levels. In connection with the nonresonant levels, it is interesting to observe that a two-photon Rabi oscillation can occur even in the absence of nonresonant intermediate states. If the nonresonant terms are omitted from Γ we find

$$\Gamma = \frac{1}{4}\frac{E_{10}^{(0)}(V_{11} - V_{00})V_{10}}{\omega(\omega - E_{10}^{(0)})}\,, \tag{5.5}$$

which is nonzero if $V_{11} \neq V_{00}$. Nonvanishing diagonal elements $V_{\alpha\alpha}$ are possible for molecules and for atoms exposed to a static electric or magnetic field.

In order to get an idea of the size and importance of these effects, we have applied the theory to a simplified model of the HF molecule. This molecule was chosen because its large dipole moment gives a strong interaction with the field, and because of the availability of a computed electronic ground-state potential and nuclear dipole moment $d(r)$.[10] The simplifications used are the neglect of excited electronic states and the rotational substructure of the vibrational spectrum. Approximate wave functions ϕ_α are obtained by fitting the potential with a Poeschl-Teller[11] potential and $d(r)$ by the expression $d_0(1+y^2)^{-\sigma}$, where $y = \sinh\alpha(r - r_0)$ is the Poeschl-Teller variable, $d_0 = 1.94$ D, and $\sigma = 1.11$. The small parameter is taken to be

$$\lambda = \mathcal{E}_0\bar{d}/\hbar\omega\,,$$

where $\bar{d}$ is the average of the absolute values of the transition matrix elements $d_{\alpha\beta}$ ($\alpha \neq \beta$), and ω is the laser frequency. The resonance condition (5.1) requires $\omega \cong \frac{1}{2}E_{10}^{(0)} = 2046.97$ cm^{-1}, so we use this value in computing λ:

$$\lambda = 1.368\times10^{-8}\sqrt{I} \tag{5.6}$$

where I is measured in W/cm^2. Thus $\lambda \cong 1$ corresponds to $I \cong 10^{16}$ W/cm^2. We conclude that the condition $\lambda \ll 1$ does not impose a serious constraint on the laser intensity.

In evaluating the resonance shift $\delta\omega^*$, and the Rabi frequency 2η, it was found that only the first five levels made significant contributions to the sums; this allows us to treat the molecule as a five-level system. Since only two of these levels are resonantly coupled, it might be thought that the model could be reduced to a two-level system. We investigate this possibility by evaluating (5.4) to obtain $\delta\omega^*/\omega^* = -0.178\lambda^2$. In the two-level model only the first term in (5.4) would remain; this yields $\delta\omega^*/\omega^* = 0.504\lambda^2$. Thus we see that the nonresonant levels make a significant contribution to the resonance shift. The quantity Γ which enters into the Rabi frequency is not so sensitive to the nonresonant levels. Evaluating Γ by (5.2) gives $\Gamma/\omega^* = 0.796\lambda^2$, and the two-level formula (5.5) gives $\Gamma/\omega^* = 0.815\lambda^2$. However, the correct calculation of Γ is important since it directly determines two essential experimental parameters. The first is the rise time, the time between minimum and maximum values of $n_1(t)$; for $\omega = \omega^*$ this is given by $\tau = \pi/\Gamma$. By using (5.6) we can express τ in terms of the intensity I (W/cm^2): $\tau = 5.464\times10^{10}I^{-1}$ nsec. This gives a lower bound for the pulse length to be used in the investigation of the two-photon Rabi oscillation. The second property determined by Γ is the line shape, which is given by the maximum probability $\bar{n}_1(\omega)$. According to (5.3) this can be written as

$$\bar{n}_1(\omega) = \frac{1}{4}\frac{\Gamma^2}{(\omega - \omega^*)^2 + (\Gamma/2)^2}\,;$$

therefore Γ is the full width at half-maximum. The fractional linewidth is very small for reasonable intensities [$\Gamma/\omega^* = 1.49\times10^{-16}I$(W/cm^2)]; conse-

quently the experimental observation of the two-photon Rabi oscillation will require either high intensities ($I \gg 10^{10}$ W/cm^2) or a very monochromatic laser ($\delta\omega/\omega \sim 10^{-6}$). In considering the first option it is important to remember that the processes of ionization and dissociation which are neglected in our model will become important at high intensities. However, if these rates are not too large and if the laser can be tuned sufficiently accurately to the resonance frequency, then the resonant oscillation may still be observable. Finally we should point out that the choice of the HF molecule was made without regard for the existence of a laser with the correct frequency. However we do not expect that a different choice will change the order of magnitude of the results given above for the resonance shift, the rise time, and the linewidth.

As a test of the limits of validity of the multiple-time-scale technique, we have carried out a numerical solution of the differential equations (2.1) for the five-level system considered above. In an exact solution, the effects of M must cancel out; therefore we set $M=0$ in solving (2.1). Since we are trying to determine when the theory fails, we use values of λ corresponding to extremely high intensities ($I \sim 10^{13}$ W/cm^2). A comparison of the approximate and exact results for $n_1(t)$ is shown in the figures. The numerical solution exhibits small-amplitude, high-frequency structrue which is neglected in the (λ^0, t_2) approximation. Since it is not possible to show these rapid oscillations in a small scale graph, we have instead represented the numerical solution by the envelope of the high-frequency part, which is shown as a lightly shaded band. The approximate analytical solution is shown as a solid curve. In Fig. 1 we see the results for the case $\omega=\omega^*$, $\lambda=0.045$. In the second half-period of the oscillation, not shown on the graph, the numerical solution lies above rather than below the solid curve. This feature is clearly visible in Fig. 2, which represents the case $\omega=\omega^*$, $\lambda=0.090$. In Fig. 2 we can also see that both numerical and analytical solutions have a maximum amplitude of 1. This is to be expected when the system is driven at the correct resonance frequency; consequently, the second-order calculation of ω^* is quite adequate. It is interesting to observe that the width of the band in Fig. 2 is twice that in Fig. 1. This is predicted by the theory from the fact that the value of λ for Fig. 2 is twice that for Fig. 1. An improved calculation of C_1 would have the form $C_1 = C_1^{(0)} + \lambda C_1^{(1)} + O(\lambda^2)$, with a corresponding occupation probability $n_1 = |C_1^{(0)}|^2 + 2\lambda\,\mathrm{Re} C_1^{(0)*} C_1^{(1)} + O(\lambda^2)$; therefore the correction to the (λ^0, t_2) approximation is linear in λ.

In Fig. 3 we exhibit an off-resonance calculation with $\omega = \frac{1}{2}E_{10}^{(0)}$, $\lambda=0.09$. The departure from exact resonance, $\delta\omega^*/\omega^* = 1.45\times10^{-3}$, prevents $n_1(t)$ from reaching unity; this is to be expected since the width is $\Gamma/\omega^* = 6.5\times10^{-3}$. The dashed curve in Fig. 3 shows the result of an analytical calculation in which the nonresonant levels were neglected. The large errors in amplitude and Rabi frequency which occur in this case can be traced to the large

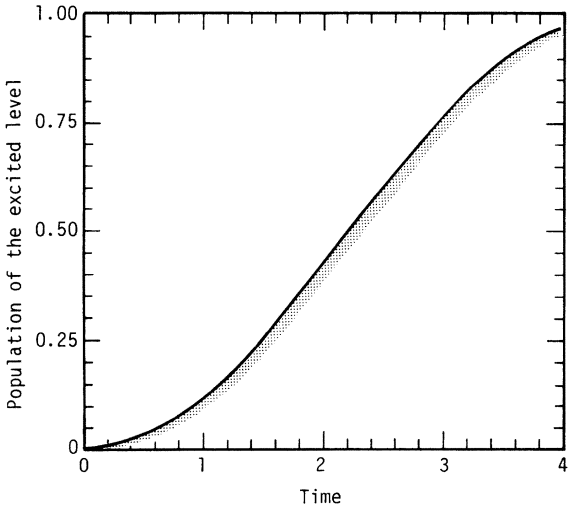

FIG. 1. $n_1(t)$ vs t. The unit of time is $10^4\hbar/D = 433/\omega_0 = 1.123\times10^{-12}$ sec, where $D=5.86$ eV is the dissociation energy and $\omega_0 = 3.86\times10^{14}$ sec^{-1} is the bare resonance frequency for HF. The solid curve is the approximate solution and the band is the envelope of the numerical solution for $\omega=\omega^*$ (the shifted resonance frequency) and $\lambda=0.045$ ($I=08\times10^{13}$ W/cm^2).

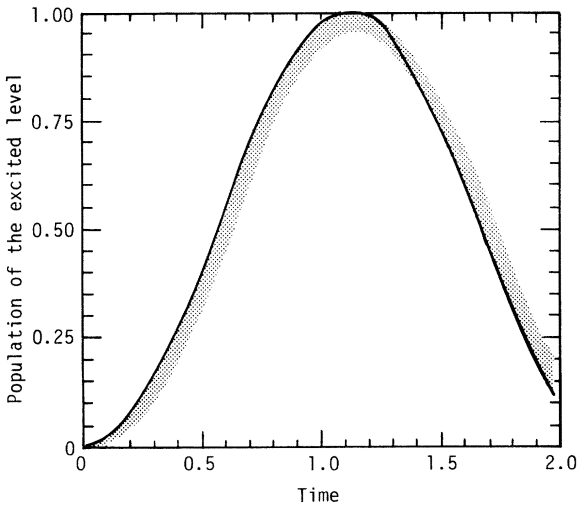

FIG. 2. $n_1(t)$ versus t. The units are the same as in Fig. 1. The solid curve is the approximate solution and the band is the envelope of the numerical solution for $\omega=\omega^*$ and $\lambda=0.09$ ($I=4.32\times10^{13}$ W/cm^2).

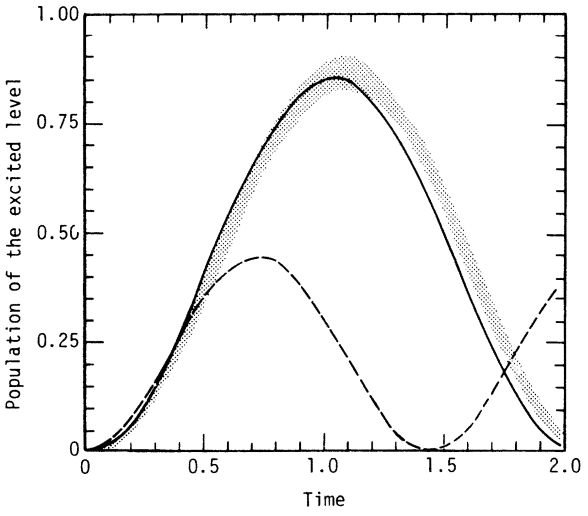

FIG. 3. $n_1(t)$ versus t. The units are the same as in Fig. 1. The solid curve is the approximate solution including nonresonant levels and the dashed curve is the approximate solution obtained by neglecting the nonresonant levels. The band is the numerical solution for $\omega=\omega_0$ (the bare resonance frequency) and $\lambda=0.09$ ($I=4.32\times10^{13}$ W/cm^2).

error in computing ω^* from the two-level formula. This emphasizes the importance of retaining a sufficient number of nonresonant levels in the calculation.

The results shown in the figures cover at most one complete cycle of the Rabi oscillation. We have also studied the long-term behavior of $n_1(t)$ by following the numerical solution shown in Fig. 2 through many cycles. The low-frequency structure, indicated by the band in the graph, turns out to be quite accurately periodic; but this period τ' differs from the theoretical period τ by $\tau'-\tau \cong 0.02\tau$. This is consistent with the estimate $\tau'-\tau=O(\lambda^2)\tau$ which can be derived from the theory. For this high intensity ($\lambda=0.09$, $I\sim10^{13}$ W/cm^2) the two curves will be out of step by 0.1τ after only five Rabi cycles. For much smaller intensities ($\lambda\sim10^{-3}$, $I\sim10^{10}$ W/cm^2) the two curves would stay in step for many thousands of Rabi cycles.

As a final check on the accuracy of the analytical solution we observe that in the (λ^0, t_2) approximation only the resonant levels 0 and 1 are populated. On the other hand, the numerical solution exhibits nonzero populations for the nonresonant levels. For the case shown in Fig. 2 we find $\bar{n}_2 \cong 0.03$, with negligible populations for the other nonresonant levels. This value of $\bar{n}_2$ is consistent with the estimate $\bar{n}_2=O(\lambda^2)$ which follows from the theory. For larger values of the coupling constant the numerical solution gives substantial populations in the nonresonant levels; in this case it would be necessary to compute higher-order corrections $C_\alpha^{(1)}, C_\alpha^{(2)}, \ldots$ to the amplitudes in order to achieve agreement between the theory and the exact solution.

VI. SUMMARY AND DISCUSSION

Our objective in this paper has been to show that the method of multiple time scales, when applied to the Schrödinger equation for an N-level system interacting with a time-dependent external field, provides a systematic and natural approximation scheme which incorporates the virtues of the rotating-wave approximation. The validity of the general formalism worked out in Sec. II depends on the assumption that the characteristic frequency $\bar{\omega}$ of the external field $\mathcal{E}(t)$ is large compared to the response frequency $\|H^{(1)}\|/\hbar$, but the formalism does not require any further assumptions about the form of $\mathcal{E}(t)$. Thus the method can be used to describe the response of an N-level system to laser pulses of arbitrary shape. In a separate publication we will present some applications of the formalism to realistic laser pulse shapes, in particular, chirped pulses.

The method used to generate the solution as a modified power series in the coupling constant λ eliminates the truly secular terms which appear in perturbation theory; and it also avoids the appearance of quasisecular terms associated with energy denominators of $O(\lambda)$. The order-by-order definition of the self-energy operator allows the field-dependent level shifts to be extracted in a systematic and explicit way. As a result of this construction the rotating-wave approximation appears in the lowest nontrivial order. More precisely, if we compute the wave function correct to $O(\lambda^0)$ and include the time dependences on t_0 and t_1 [this is the (λ^0, t_1) order], then we obtain the conventional rotating-wave approximation for a Rabi oscillation driven by a one-photon resonance. The (λ^1, t_1) approximation would incorporate corrections due to the counterrotating terms. The (λ^0, t_2) approximation yields a generalization of the Bloch-Siegert theory of intensity-dependent level shifts which includes the effects of nonresonant levels.

The theory automatically includes multiphoton effects as seen in Sec. V, where a Rabi oscillation due to a two-photon resonance is demonstrated. In this case the nonresonant levels play a vital role in determining the shifted resonance frequency. For the model of HF considered here the fractional line width $[\Gamma/\omega^*\sim10^{-16}\ I\ (\mathrm{W/cm^2})]$ is quite small for moderate intensities, but the rise time ($\tau\sim10^{10}I^{-1}$ nsec) is reasonably short. At higher intensities it would be necessary to estimate the

rates of ionization and dissociation with a more complicated model. Thus it would probably be best to attempt the fine tuning of the laser required for experiments at $I \sim 10^{10}$ W/cm^2.

The comparison of the (λ^0, t_2) approximation with an exact numerical solution shows that the calculation of the resonance frequency correct to $O(\lambda^2)$ is quite adequate even for rather large values of λ. However, at these values of λ the approximate time dependence of $n_1(t)$ is reliable for only a few Rabi periods. At moderate intensities ($\lambda \sim 10^{-3}$) this is not a problem.

ACKNOWLEDGMENTS

The numerical differential equation solver used in Sec. V is a version of the program written by W. A. Gear and modified by Alan Hindmarsh.

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[1]F. Bloch and A. Siegert, Phys. Rev. 57, 522 (1940).

[2]H. Poincarê, *Les Méthodes Nouvelles de la Mecanique Cêleste* (Dover, New York, 1957), Vol. 1, Chap. 3.

[3]N. Krylov and N. Bogoliubov, *Introduction to Non-Linear Mechanics* (Princeton U.P., Princeton, N. J., 1947).

[4]G. Sandri, Ann. Phys. (N. Y.) 24, 332 (1963); 24, 380 (1963); G. V. Ramanathan and G. Sandri, J. Math. Phys. 10, 1763 (1969).

[5]E. A. Frieman, J. Math, Phys. 4, 410 (1963); E. A. Frieman and R. Goldman, *ibid.* 7, 2153 (1966).

[6]P. A. M. Dirac, Proc. R. Soc. Lond. 136, 453 (1932).

[7]S. Tomonaga, Prog. Theor. Phys. 1, 27 (1946).

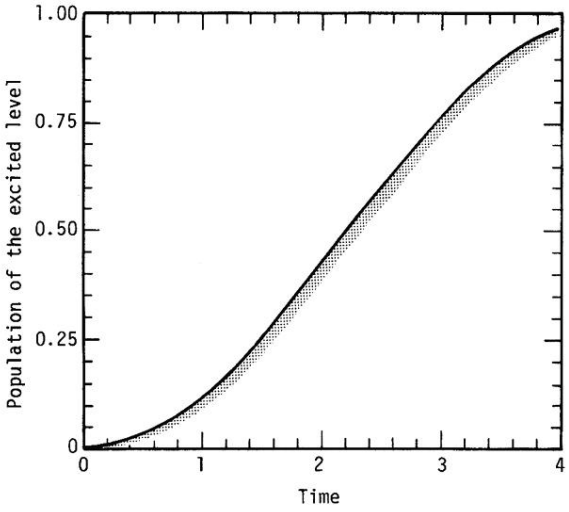
[7(a)]P. S. Lee, Y. C. Lee, and C. T. Chang, Phys. Rev. A 8, 1722 (1973); P. S. Lee and Y. C. Lee, *ibid.* 8, 1727 (1973).

[8]J. H. Shirley, J. Appl. Phys. 34, 783 (1963).

[9]J. H. Shirley, Phys. Rev. 138, B979 (1965). This paper presents a method, based on the Floquet theorem for interactions periodic in time, which can be used to describe multiphoton Rabi oscillations induced by a monochromatic field, or possibly a finite superposition of fields with different frequencies.

[10]A. M. Karo and L. Allen, J. Chem. Phys. 31, 968 (1959).

[11]G. Poeschl and E. Teller, Z. Phys. 83, 143 (1933). The potential parameters used are $\alpha = 1.0476$, $r_0 = 0.72831$, $\mu = 52.012$, $\nu = 2.5570$.



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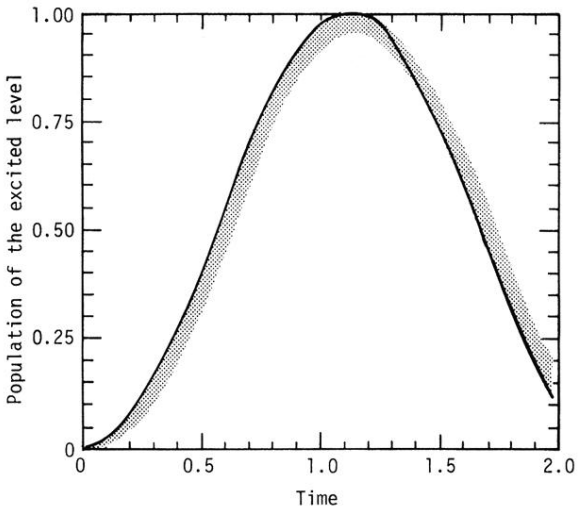

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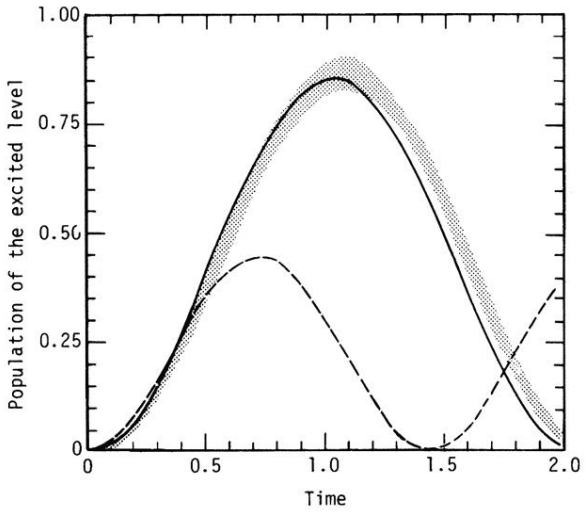

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