Floquet theory of bound-continuum transitions due to a periodic interaction

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The calculation of transition rates from bound to continuum states of a Hamiltonian under the influence of a time-periodic perturbation is formulated in terms of the Floquet theory by constructing quasiperiodic states on a restriction of the underlying composite Hilbert space. The required restriction is related to the circumstance that a steady rate for the excitation of a continuum level can only be defined in the limit of the imaginary part of the level shift tending to zero. Model numerical calculations are presented which demonstrate that the amplitudes thus obtained agree with perturbation theory where the latter is expected to be applicable. Near an intermediate resonance, where the standard perturbation theory diverges, the Floquet theory leads to a typical resonant structure, which is also shown to result from regularizing the perturbation theory, thereby demonstrating that the Floquet theory automatically takes account of the relevant higher-order terms in the perturbation expansion.

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I. INTRODUCTION

It has recently been shown [1] that, if, in the presence of a classical electromagnetic field, a quantum system possesses normalizable Floquet states, the amplitudes for transitions between the unperturbed states, which correspond to the absorption of an integral number of photons, can be defined unambiguously and calculated nonperturbatively. When a bound state is coupled to a continuum, however, the eigenenergy becomes complex, necessitating a modification of the conventional Hilbert space in which Hermitian operators have real eigenvalues [2–4]. Since the Floquet theory relies on the existence of normalizable quasiperiodic states of the interacting system in the composite Hilbert space of square-integrable and time-periodic functions [5], whose eigenenergies are real, application of this theory to calculate transition rates to a continuum can only be expected to be possible in the limit $(E_I/E_R)\rightarrow 0$, where E_R and E_I denote, respectively, the real and imaginary parts of the energy. But this is just the condition under which a transition rate can be defined in any case [3,6], and, therefore, the feasibility of treating bound-continuum transitions within the framework of the Floquet theory merits serious consideration, especially since the S-matrix elements, which are on the energy shell, can then be calculated nonperturbatively. The Friedrichs model [7] in which a single discrete state is coupled to a continuum through interaction with a quantized (time-independent) field, ignoring continuum-continuum couplings, has served as a paradigm in the investigations of Prigogine and co-workers [2,8]. The Floquet theory being semiclassical in nature, we shall use a model in which the effect of the field is represented by a time-dependent, periodic, interaction. We shall also generalize the model to include more than one bound state and allow for continuum-continuum couplings. At the same time, to minimize the complexity of the problem without distorting its essential features, we shall assume the continuum to be labeled by the energy alone.

As in the case of a system having only bound states [1], we first formulate the problem as the solution of an eigenvalue equation for an infinite Hermitian matrix. For this matrix to represent a self-adjoint operator in a Hilbert space, its Hermitian form must be real [9,10], in which case one can expect an eigenvalue calculation in practice using a truncated matrix, which would always yield a real value, to converge to the correct result as the dimension is increased. On the basis of known results from the theory of the ac Stark effect [11], we then argue that by restricting the expansion in terms of the time-periodic functions appropriately, the imaginary part of the energy can be made to vanish, so that, in accordance with the discussion above, a transition rate can be properly defined. This restriction is also shown to be readily understandable from the point of view of perturbation theory. Numerical evidence via a model calculation is presented to illustrate the necessity and sufficiency of this criterion. Nonperturbative calculation of transition amplitudes using the Floquet theory in the above limit is illustrated for various models. For low coupling strengths, the results thus obtained agree with lowest-order perturbation theory (LOPT), except in the vicinity of a resonance between two bound states, where the perturbation theory has to be suitably modified [12] to avoid encountering a spurious divergence.

II. FORMALISM

Let the Hamiltonian of the unperturbed system H_0 have N_B bound states (discrete eigenvalues) and a continuum specified by energy ε,

$$0\leq\varepsilon\leq\infty\ ,$$
$$H_0=\sum_{\mu=k}^{N_B}\varepsilon_\mu|\mu\rangle\langle\mu|+\int_0^\infty\varepsilon\,d\varepsilon|\varepsilon\rangle\langle\varepsilon|\ , \tag{1}$$
$$\langle\mu|\nu\rangle=\delta_{\mu\nu},\quad\langle\varepsilon|\varepsilon'\rangle=\delta(\varepsilon-\varepsilon')\ .$$

For simplicity, let the periodic interaction causing transitions be given by $2V\cos\omega t$, with V real. As in Ref. [1], a switching factor $\exp(-\eta|t|)$ is understood to be associated with V. Let the bound-bound, bound-continuum, and continuum-continuum matrix elements be denoted, respectively, by $V_{\mu\nu}$, $V_{\mu\varepsilon}$, and $V_{\varepsilon\varepsilon'}$. For reference (initial) state $|\phi_a\rangle$, we seek a solution of the Schroedinger equation

$$i\frac{\partial|\psi_a\rangle}{\partial t}=(H_0+V)|\psi_a\rangle \tag{2}$$

of the form [1]

$$|\psi_a\rangle=\exp(-\varepsilon_a+\Delta_a e^{-\eta|t|})t[|a\rangle + \{|F_a(t)\rangle-|a\rangle\}e^{-\eta|t|}] , \tag{3}$$

$$|F_a(t)\rangle=\sum_n e^{-in\omega t}|\chi_n^a\rangle , \tag{4}$$

with $|\chi_n^a\rangle$ independent of time. Let us expand $|\chi_n^a\rangle$ in the Hilbert space of H_0 as

$$|\chi_n^a\rangle=\sum_{\mu=1}^{N_B}\alpha_{n\mu}^a|\mu\rangle+\int_0^\infty A_n^a(\varepsilon)|\varepsilon\rangle d\varepsilon . \tag{5}$$

We now expand $A_n^a(\varepsilon)$ in terms of Laguerre polynomials as

$$\begin{aligned} A_n^a(\varepsilon)&=\sum_{l=0}^\infty C_{nl}^a L_l(\varepsilon)e^{-\varepsilon/2} ,\\ C_{nl}^a&=\int_0^\infty A_n^a(\varepsilon)L_l(\varepsilon)e^{-\varepsilon/2}d\varepsilon . \end{aligned} \tag{6}$$

Similarly, $V_{\mu\varepsilon}$ and $V_{\varepsilon\varepsilon'}$ may also be expanded as

$$V_{\mu\varepsilon}=\sum_{l=0}^\infty v_{l\mu}L_l(\varepsilon)e^{-\varepsilon/2} , \tag{7}$$

and

$$V_{\varepsilon\varepsilon'}=\sum_{l=0}^\infty\sum_{m=0}^\infty g_{lm}L_l(\varepsilon)e^{-\varepsilon/2}L_m(\varepsilon')e^{-\varepsilon'/2} . \tag{8}$$

Note that for a general Hermitian V, $v_{l\mu}=v_{\mu l}^*$ and $g_{lm}=g_{ml}^*$. The normalization condition [5],

$$\frac{\omega}{2\pi}\int_0^{2\pi/\omega}\langle\psi_a|\psi_a\rangle dt=1 \quad (\eta\to 0) ,$$

then takes the form

$$\sum_n\left[\sum_{\mu=1}^{N_B}|\alpha_{n\mu}^a|^2+\sum_l|C_{nl}^a|^2\right]=1 . \tag{9}$$

The amplitude for a transition from $|i\rangle$ to $|\varepsilon\rangle$ may now be calculated as in Ref. [1], provided the system of equations obtained on using Eqs. (5)–(8) in Eq. (2), (and letting $\eta\to 0$), can be solved with an initial condition corresponding to $|i\rangle$ as $V\to 0$. The equations to be solved are

$$\begin{aligned}(\varepsilon_a+\Delta_a+n\omega-\varepsilon_\mu)\alpha_{n\mu}^a&=\sum_{\nu=1}^{N_B}(\alpha_{n-1,\nu}^a+\alpha_{n+1,\nu}^a)V_{\mu\nu}\\ &\quad+\sum_l v_{l\mu}(C_{n-1,l}^a+C_{n+1,l}^a)\end{aligned} \tag{10}$$

and

$$(\varepsilon_a+\Delta_a+n\omega-\varepsilon)\sum_l C_{nl}^a L_l(\varepsilon)e^{-\varepsilon/2}=\sum_{\nu=1}^{N_B}(\alpha_{n-1,\nu}^a+\alpha_{n+1,\nu}^a)\sum_l v_{l\nu}L_l(\varepsilon)e^{-\varepsilon/2}+\sum_{lm}g_{lm}L_m(\varepsilon)e^{-\varepsilon/2}(C_{n-1,l}^a+C_{n+1,l}^a) , \tag{11}$$

$$\mu=1,N_B,\quad n=-\infty,\ldots,-1,0,1,\ldots,\infty .$$

The initial condition is

$$\alpha_{n\mu}^a\xrightarrow[V\to 0]{}\delta_{n0}\delta_{a\mu} . \tag{11a}$$

When the reference state $|a\rangle$ is a bound state, Eq. (11) may be recast as follows. Multiplying Eq. (11) by $L_l(\varepsilon)e^{-\varepsilon/2}$ and integrating over ε, we get

$$\begin{aligned}(\varepsilon_a+\Delta_a+n\omega)C_{nl}^a-\sum_m\gamma_{lm}C_{nm}^a&=\sum_{\nu=1}^{N_B}(\alpha_{n-1,\nu}^a+\alpha_{n+1,\nu}^a)v_{l\nu}\\ &\quad+\sum_m g_{ml}(C_{n-1,m}^a+C_{n+1,m}^a) ,\end{aligned} \tag{12}$$

where

$$\gamma_{lm}=\gamma_{ml}=\int_0^\infty\varepsilon L_l(\varepsilon)L_m(\varepsilon)e^{-\epsilon}d\varepsilon . \tag{13}$$

From the properties of the Laguerre polynomials, it readily follows that

$$\begin{aligned}\gamma_{lm}&=0,|1-m|\rangle 1 ,\\ \gamma_{l,1+1}&=-(1+1) ,\\ \gamma_{l,l}&=2l+1 .\end{aligned} \tag{14}$$

Equations (10) and (12) may be cast as an eigenvalue equation for Δ_a by defining a column vector $\mathbf{X}^a$ constructed from $\alpha_{n\mu}^a$, $n=-\infty,\ldots,\infty$, $\mu=1,N_B$, followed by C_{nl}^a, $n=-\infty,\ldots,\infty$, $l=0,\ldots,\infty$, with [cf. Eq. (9)]

$$(\mathbf{X}^a)^\dagger(\mathbf{X}^a)=1 .$$

Similarly, a coefficient matrix $\mathbf{A}$ can be formed from $v_{l\mu}$, γ_{lm}, and g_{lm} such that Eqs. (10) and (12) are equivalent to the matrix equation

$$\mathbf{A}\mathbf{X}^a=\Delta_a\mathbf{X}^a . \tag{15}$$

Owing to the symmetry properties of the elements constituting $\mathbf{A}$, [see below Eq. (8)], it is a Hermitian matrix.

Therefore, Δ_a is a Hermitian form [10], given by

$$\Delta_a=(\mathbf{X}^a)^\dagger \mathbf{A}(\mathbf{X}^a) . \tag{16}$$

From the properties of Hilbert matrices [10], we know that Δ_a, if it exists, must be real. However, for the ac Stark problem, where $V=eE_0x$, E_0 being the amplitude of the electric field, Yajima [11] has shown that Δ_a is actually complex, with Im $(\Delta_a)=O(E_0^{2N_a})$, where N_a is the smallest integer such that $(\varepsilon_a+N_a\omega)>0$. In the quantized picture, N_a denotes the minimum number of photons required to be absorbed in order to make a real transition to the continuum. Now, in the Floquet theory [1], this process is made possible because of the presence of the periodic function $\exp(-iN_a\omega t)$. Therefore, by restricting n in Eq. (4) through $n<N_a$, in the *continuum sector* of the expansion of $|\chi_n^a\rangle$ [Eq. (5)], we may expect the resultant Δ_a to be real and hence the Hermitian form, Eq. (16), to exist. This is, of course, an approximation, since the underlying function space is no longer complete. However, as explained in the Introduction, for bound-continuum transitions, a probability per unit time is indeed an approximate concept [6]. Thus we rewrite Eqs. (4) and (5) as

$$|F_a(t)\rangle=\sum_{n=-\infty}^{\infty} e^{-in\omega t}\sum_{\mu=1}^{N_B}\alpha_{n\mu}^a|\mu\rangle + \sum_{n=-\infty}^{n_{\max}^c} e^{-in\omega t}\int_0^\infty A_n^a(\varepsilon)|\varepsilon\rangle d\varepsilon ,$$

$$n_{\max}^c=[-\varepsilon_a/\omega] , \tag{17}$$

where $[x]$ denotes the largest integer less than x. The vector $\mathbf{X}^a$ and matrix $\mathbf{A}$ can be modified to conform to this new expansion in an obvious manner and Eq. (15) solved numerically with l less than some value, say, $l_{\max}$, which may then be increased until the final results are converged.

It must be stated here that apart from the argument given above and that based on perturbation theory below, no rigorous proof of the existence of a convergent solution of Eq. (15) under the condition $n<N_a$ has been obtained, though some numerical evidence is presented later on. The necessity of the condition $n<N_a$ for the reality of Δ_a can be understood on the basis of perturbation theory by noting that once a real (energy-conserving) transition to the continuum is made, no further transitions are permitted, according to this condition. In perturbation theory, this is equivalent to dropping all terms with vanishing denominators in the *continuum sector,* which are precisely the terms that contribute to the imaginary part of the level shift through the well-known operator relation

$$\frac{1}{x\pm i\varepsilon}=\mathbf{P}\frac{1}{x}\mp i\pi\delta(x) .$$

Note that in principle there need be no restriction on n due to real transitions between discrete energy states since the δ function does not contribute. In practice, though, perturbation theory has to be modified to handle intermediate resonances [12], whereas the Floquet theory, being nonperturbative in nature, needs no special attention. It is also worth stressing that the condition $n<N_a$ does not imply LOPT. First of all, n is not restricted from below. Second, n indicates the *net* number of quanta absorbed; any number of virtual transitions without violating this condition at any stage are permitted. Finally, as mentioned before, there is no restriction on n in the bound-state sector. Therefore, the transition amplitudes calculated as described below represent the summation of those terms of the conventional perturbation series that do not involve any vanishing denominators in the continuum sector.

Calculation of the S-matrix element connecting a bound state $|i\rangle$ and a continuum $|\varepsilon_f\rangle$ may now proceed as in Ref. [1]. Before that however, the question of wave-function renormalization and level shift of the final state must be addressed. Since the level shift now is a function of the continuous variable ε, Δ_a in Eqs. (10) and (11) would also have to be discretized just as $A_n^a(\varepsilon)$, which would obviously lead to a much more complicated set of equations. In practice, however, it is often a good approximation to set $\Delta(\varepsilon_f)=0$ and $A_n^{\varepsilon_f}(\varepsilon)=\delta_{n0}\delta(\varepsilon-\varepsilon_f)$. This can be seen by considering the case when H_0 is just the kinetic energy operator. For a continuum reference state specified by momentum $\mathbf{k}_f$, $|\psi_{\mathbf{k}_f}\rangle$ is then given by [13]

$$|\psi_{\mathbf{k}_f}\rangle=|\mathbf{k}_f\rangle e^{-iEt}\sum_{n=-\infty}^{\infty} J_n(\mathbf{k}_f\cdot\boldsymbol{\alpha}_0)e^{in\omega t} ,$$

where E is the unperturbed energy $(=k_f^2/2)$ and α_0 is proportional to the field amplitude. Thus $\Delta(k_f)=0$ and $A_n^{\mathbf{k}_f}(\mathbf{k})=J_n(\mathbf{k}_f\cdot\boldsymbol{\alpha}_0)\delta(\mathbf{k}-\mathbf{k}_f)$. Therefore the wave-function renormalization factor [14], appropriate for this continuum state, is just $J_0(\mathbf{k}_f\cdot\boldsymbol{\alpha}_0)$, which is close to unity for small field amplitudes. For illustrative model calculations, it is, therefore, sufficient to calculate the S-matrix element from

$$S_{fi}=-i\lim_{\eta\to 0}\int_{-\infty}^{\infty} e^{i\varepsilon_f t}\langle\varepsilon_f|V|\psi_i\rangle dt .$$

Using Eqs. (3)–(8) and evaluating the integral as in Ref. [1] finally leads to

$$S_{fi}=-2\pi i T_{fi}^{(n)}\delta(\varepsilon_f-\varepsilon_i-n\omega) , \tag{18}$$

with the T-matrix elements given by

$$T_{fi}^{(n)}=\sum_{\mu=1}^{N_B}(\alpha_{n-1,\mu}^i+\alpha_{n+1,\mu}^i)V_{\varepsilon_f\mu} + \sum_{lm} g_{lm}(C_{n-1,l}^i+C_{n+1,l}^i)e^{-\varepsilon_f/2}L_m(\varepsilon_f) . \tag{19}$$

The transition probability per unit time to a final state satisfying the energy conservation condition implied by the δ function in Eq. (18) is then given by (e.g., Ref. [13], Chap. 6)

$$W_{fi}=2\pi|T_{fi}^{(n)}|^2\delta(\varepsilon_f-\varepsilon_i-n\omega) .$$

Actually, beyond the first accessible continuum level, i.e., $n>n_{\max}^c+1$, the second sum on the right-hand side of

Eq. (19) does not contribute anything, while for $n=n^c_{\max}$, $C^i_{n+1,l}\equiv 0$. $T^{(n)}_{fi}$ is the amplitude for a transition to the continuum corresponding to the net absorption of n quanta of the field represented semiclassically by the periodic interaction $2V\cos\omega t$. For the sake of convenience, we shall often refer to it as the n-photon transition amplitude. Also, since Eqs. (9), (10), and (12) determine $\alpha^i_{n\mu}$ and $C^i_{n,l}$ only to within an overall constant phase, Eq. (19) only fixes $|T^{(n)}_{fi}|$, which, however, completely determines the transition probability. The numerical results discussed below, therefore, refer to $|T^{(n)}_{fi}|$.

III. MODEL CALCULATIONS

In this section, specific numerical calculations are described, which illustrate the significance of the condition $n<N_a$ and the relationship of the nonperturbative transition amplitude as given by Eq. (19) to its perturbative approximation.

A. One bound state and no continuum-continuum coupling

This is the Friedrichs model and, in the notation of the preceding section, corresponds to the choice $N_B=1$ and $V_{\varepsilon\varepsilon'}\equiv 0$. Also, the interaction is assumed to have no diagonal elements, i.e., $V_{11}=0$. For numerical work, the remaining quantities are specified as $\varepsilon_1=-0.5$ and $V_{1\varepsilon}=E_0e^{-\varepsilon/2}$, with a variable coupling strength E_0. From Eqs. (7) and (8) we then have $V_{l1}=E_0\delta_{l0}$ and $g_{lm}\equiv 0$. For any given ω, $n^c_{\max}$ is predetermined as [Eq. (17)] $n^c_{\max}=[0.5/\omega]$. In general, for the bound-state sector, an upper limit n_B for n is fixed to start with, which is then increased to check for convergence.

The calculations presented here were carried out for $\omega=0.3$, the corresponding $n^c_{\max}$ being 1. In this particular model, since the only bound state is fed by the continuum [cf. Eq. (10)] alone, fixing $n^c_{\max}=1$ also fixes the maximum value of n_B required as two. Thus, in this case, only one has to check for convergence in $l_{\max}$. Also n can only take *odd* values in the continuum sector, owing to the absence of continuum-continuum coupling. The first continuum level that can be excited, therefore, corresponds to the absorption of three quanta. Further, the next higher value of $n^c_{\max}$ is really three, for which no convergent results are to be expected. Finally, for small E_0, one would expect the nonperturbative (NPT) Floquet results to agree with LOPT calculations. Δ_1 and $T^{(3)}$ in the latter approximation can be calculated from standard expressions [1] as follows:

$$\Delta_1=-\int_0^\infty \frac{2|V_{1\varepsilon}|^2(\varepsilon-\varepsilon_1)^2}{(\varepsilon-\varepsilon_1)^2-\omega^2}d\varepsilon\ .$$

Substituting for $V_{1\varepsilon}$ and carrying out the integral, we get

$$\Delta_1=I(\omega)+I(-\omega)\ , \tag{20}$$

where

$$I(\omega)=-E_0^2e^{-(\varepsilon_1+\omega)}E_1(-\varepsilon_1-\omega)\ , \tag{21}$$

E_1 being the exponential integral. Similarly,

$$T^{(3)}=\int_0^\infty \frac{V_{\varepsilon_f 1}V_{1\varepsilon}V_{\varepsilon 1}}{-2\omega(\varepsilon-\varepsilon_1-\omega)}d\varepsilon,\quad \varepsilon_f=\varepsilon_1+3\omega\ ,$$

which reduces to

$$T^{(3)}=\frac{-E_0^3}{2\omega}e^{-(\varepsilon_1+\omega)}e^{-\varepsilon_f/2}E_1(-\varepsilon_1-\omega)\ . \tag{22}$$

The convergence of Δ_1 and $|T^{(3)}|$ with respect to $l_{\max}$ is illustrated in Table I for $n^c_{\max}=1$ and $n^c_{\max}=3$. In the former case, results for $E_0=0.001$ and 0.1 are presented, both of which converge smoothly with increasing $l_{\max}$. In contrast, for $n^c_{\max}=3$ and $E_0=0.1$, the results are seen to oscillate, thereby verifying the need for the limitation on n in the continuum sector. (These values are converged with respect to n in the bound sector, with $n_B=4$.) Also, LOPT agrees very well with NPT for $E_0=0.001$, but not at $E_0=0.1$.

Nonconvergence of the eigenvalues when $n^c_{\max}$ exceeded $[-\varepsilon_1/\omega]$ was also verified for the more general models considered below. For further discussion, only the results that have converged in n_B and $l_{\max}$ (for the appropriate $n^c_{\max}$) are presented in the following sections.

TABLE I. Convergence of level shift and transition amplitude with $l_{\max}$. One bound state of energy -0.5 and no continuum-continuum coupling, $\omega=0.3$.

	$n^c_{\max}=1, n_B=2$				$n^c_{\max}=3, n_B=4$	
	$\lvert T^{(3)}\rvert$		Δ_1		$E_0=0.1$	
$l_{\max}$	$E_0=0.001$	$E_0=0.1$	$E_0=0.001$	$E_0=0.1$	$\lvert T^{(3)}\rvert$	Δ_1
5	1.942×10^{-9}	1.860×10^{-3}	-2.113×10^{-6}	-2.024×10^{-2}	1.979×10^{-3}	-2.022×10^{-2}
10	2.016×10^{-9}	1.917×10^{-3}	-2.168×10^{-6}	-2.067×10^{-2}	1.895×10^{-3}	-2.067×10^{-2}
20	2.035×10^{-9}	1.930×10^{-3}	-2.183×10^{-6}	-2.077×10^{-2}	2.698×10^{-3}	-2.068×10^{-2}
30	2.037×10^{-9}	1.931×10^{-3}	-2.184×10^{-6}	-2.078×10^{-2}	1.951×10^{-3}	-2.078×10^{-2}
40	2.038×10^{-9}	1.931×10^{-3}	-2.185×10^{-6}	-2.078×10^{-2}	1.820×10^{-3}	-2.079×10^{-2}
50	2.038×10^{-9}	1.931×10^{-3}	-2.185×10^{-6}	-2.078×10^{-2}	2.724×10^{-3}	-2.069×10^{-2}
80	2.038×10^{-9}	1.931×10^{-3}	-2.185×10^{-6}	-2.078×10^{-2}	1.809×10^{-3}	-2.079×10^{-2}
100	2.038×10^{-9}	1.931×10^{-3}	-2.185×10^{-6}	-2.078×10^{-2}	2.093×10^{-3}	-2.076×10^{-2}
LOPT	2.038×10^{-9}	2.038×10^{-3}	-2.185×10^{-6}	-2.185×10^{-2}	2.038×10^{-3}	-2.185×10^{-2}

B. One bound state with continuum-continuum coupling

Allowing for continuum-continuum coupling opens up new pathways and permits calculation of transition amplitudes for the excitation of more than one final state, both in the Floquet theory and LOPT. Thus, for $\omega=0.2$, $n^c_{\max}$ is 2, but now both $\varepsilon_f=\varepsilon_1+3\omega$ and $\varepsilon_f=\varepsilon_1+4\omega$ are possible final-state energies. In the quantized picture, these correspond to sequential absorptions along the paths (i) $|1\rangle\rightarrow|\varepsilon\rangle\rightarrow|1\rangle\rightarrow|\varepsilon_f\rangle+|1\rangle\rightarrow|\varepsilon\rangle\rightarrow|\varepsilon'\rangle\rightarrow|\varepsilon_f\rangle$ and (ii) $|1\rangle\rightarrow|\varepsilon\rangle\rightarrow|\varepsilon'\rangle\rightarrow|1\rangle\rightarrow|\varepsilon_f\rangle$, respectively. In (i), the amplitude for the first path is again given by Eq. (22), while the new alternative provides an extra amplitude given by

$$T^{(3)}(\text{extra})=\int_0^\infty\int_0^\infty \frac{V_{\varepsilon_f\varepsilon'}V_{\varepsilon'\varepsilon}V_{\varepsilon 1}}{(\varepsilon'-\varepsilon_1-2\omega)(\varepsilon-\varepsilon_1-\omega)} \times d\varepsilon\, d\varepsilon',\ \varepsilon_f=\varepsilon_1+3\omega\ . \tag{23}$$

Choosing for $V'_{\varepsilon\varepsilon}$ the simple Hermitian form

$$V'_{\varepsilon\varepsilon}=0.1E_0e^{-\varepsilon/2}e^{-\varepsilon'/2}\ ,$$

the additional contribution is readily evaluated as

$$T^{(3)}(\text{extra})=0.01E_0^3e^{-(2\varepsilon_1+3\omega)}e^{-\varepsilon_f/2} \times E_1(-\varepsilon_1-\omega)E_1(-\varepsilon_1-2\omega)\ . \tag{24}$$

Similarly, $T^{(4)}$ may also be shown to be given by

$$T^{(4)}=-(0.1E_0^4/3\omega)e^{-(2\varepsilon_1+3\omega)}e^{-\varepsilon_f/2} \times E_1(-\varepsilon_1-\omega)E_1(-\varepsilon_1-2\omega)\ ,$$
$$\varepsilon_f=\varepsilon_i+4\omega\ . \tag{25}$$

$|T^{(3)}|$ and $|T^{(4)}|$ according to the Floquet theory (solid lines) and LOPT (dotted lines) are shown in Fig. 1 as a function of E_0. Again, for low E_0 both theories are in excellent agreement and, as expected, nonperturbative effects begin to show up earlier for the higher-order transition. In both cases, these model calculations have been confined to values of E_0 sufficiently high to illustrate the onset of nonperturbative behavior.

C. Two bound states and a continuum

This is the simplest model that permits transitions to all energetically allowed continuum levels, without violating the condition $n<N_a$. As we have seen, with only one bound state, not more than two levels (separated by ω) can be excited. This is, of course, because of the fact that the sole bound state is coupled only to the continuum and, therefore, the effective maximum value of n_B is determined by $n^c_{\max}$. However, when there are two or more bound states, there is no such limit on n_B, since these states can feed one another without the participation of the continuum. More importantly, this model permits the occurrence of an intermediate resonance, around $\omega=\varepsilon_2-\varepsilon_1$, where ordinary perturbation theory completely breaks down. As has been shown [12], perturbation theory can be regularized to deal with this type of situation. Let us consider the case $N_1=2$, which corresponds to two-photon transition from the ground state to the continuum. In LOPT, we have

$$T^{(2)}_{f1}(\text{LOPT})=\frac{V_{\varepsilon_f 2}V_{21}}{(\varepsilon_2-\varepsilon_1-\omega)}+\int_0^\infty\frac{V_{\varepsilon_f\varepsilon}V_{\varepsilon 1}}{(\varepsilon-\varepsilon_1-\omega)}d\varepsilon\ ,$$
$$\varepsilon_f=\varepsilon_1+2\omega\ . \tag{26}$$

Choosing the additional matrix elements in this model as $V_{12}=0.75E_0$ (same as the dipole matrix element connecting the $1s$ and $2p$ levels of hydrogen) and $V_{2\varepsilon}=2V_{1\varepsilon}$, Eq. (26) becomes

$$T^{(2)}_{f1}(\text{LOPT})=\frac{1.5E_0^2e^{-\varepsilon_f/2}}{(\varepsilon_2-\varepsilon_1-\omega)}+0.1E_0^2e^{-(\varepsilon_1+\omega)} \times e^{-\varepsilon_f/2}E_1(-\varepsilon_1-\omega)\ . \tag{27}$$

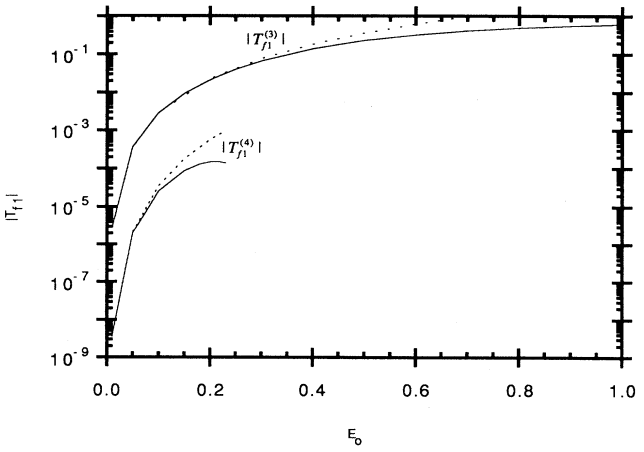

FIG. 1. Three- and four-photon transition amplitudes (a.u.) for the model described in Sec. III B, as a function of the field amplitude E_0 (a.u.).

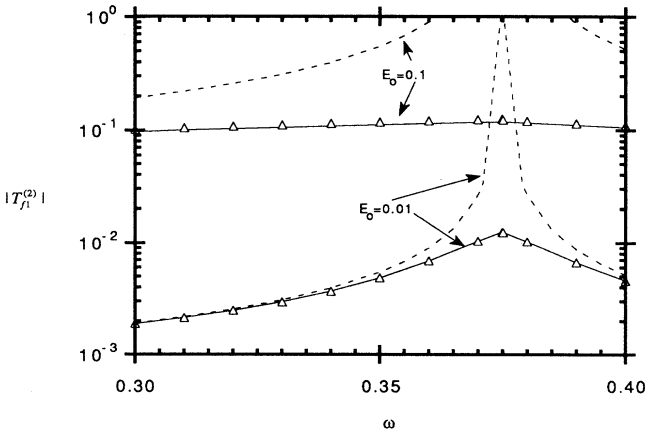

FIG. 2. Intermediate resonance in transitions to the continuum through the net absorption of two quanta. Model parameters are as in Sec. III C. Solid line-NPT; open triangles-MPT; dotted line-LOPT. All quantities are in atomic units.

The second term on the right-hand side, representing transitions in which the intermediate state is in the continuum, is well behaved for $(\varepsilon_1+\omega)<0$, whereas the first term has a singularity at $\omega=\varepsilon_2-\varepsilon_1$. This can be removed by first treating the coupling between the two bound states nonperturbatively and then using perturbation theory to describe transitions to the continuum. Since a detailed account of this procedure is presented in Ref. [12], only an outline is given here. Standard perturbation theory [15] starts from the expansion

$$|\psi_1\rangle=\sum_{r=1}^{\infty} a_r(t)e^{-i\varepsilon_r t}|r\rangle \tag{28}$$

(where the summation also includes an integration over the continuum), and determines a_r perturbatively, assuming the zeroth-order approximation to be $a_r^0=\delta_{r0}$. To allow for a resonance between states $|1\rangle$ and $|2\rangle$, we first solve the two-level atom model involving only these states, in the rotating-wave approximation (RWA), to get

$$a_1=\alpha e^{-i\Delta_1 t},\quad a_2=\beta e^{-i(\varepsilon_1+\Delta_1+\omega-\varepsilon_2)t}$$

where

$$\alpha=\left[\frac{\Omega-\epsilon}{2\Omega}\right]^{1/2},\quad \beta=-\left[\frac{\epsilon+\Omega}{2\Omega}\right]^{1/2},\quad \Delta_1=-\frac{\epsilon+\Omega}{2},$$

$$\epsilon<0,$$

$$\alpha=\left[\frac{\Omega+\epsilon}{2\Omega}\right]^{1/2},\quad \beta=\left[\frac{\Omega-\epsilon}{2\Omega}\right]^{1/2},\quad \Delta_1=\frac{\Omega-\epsilon}{2},$$

$$\epsilon>0,$$

and

$$\varepsilon=\omega-(\epsilon_2-\epsilon_1),\quad \Omega=\sqrt{\epsilon^2+4|V_{12}|^2}\,.$$

Using the above expressions for a_1 and a_2 as the zeroth-order approximation, the amplitude a_f corresponding to the continuum level $|\epsilon_f\rangle$ may now be calculated by perturbation theory. This procedure modifies Eq. (27) as

$$T_{f1}^{(2)}(\mathrm{MPT})=2E_0e^{-\varepsilon_f/2}\beta+0.1\alpha E_0^2e^{-(\varepsilon_1+\omega+\Delta_1)}e^{-\varepsilon_f/2}\times E_1(-\varepsilon_1-\omega-\Delta_1)\,. \tag{29}$$

It may be verified that $|T_{f1}^{(2)}(\mathrm{MPT})|\to|T_{f1}^{(2)}(\mathrm{LOPT})|$ as $V_{12}/|\varepsilon|\to 0$.

For numerical calculations, ε_2 was chosen to be equal to -0.125, so that the two bound-state energies were the same as those of the first two levels of hydrogen. The variation of $|T^{(2)}|$ with ω in the range 0.3–0.4 (where $n^c_{\max}=1$) is shown in Fig. 2, for $E_0=0.01$ and 0.1, according to the Floquet theory (NPT) as well as the two versions of perturbation theory. The modified perturbation theory (MPT) is seen to be in excellent agreement with NPT, while the applicability of LOPT is limited to low E_0 and to values of ω sufficiently removed from the resonance frequency. The variation of $|T^{(2)}|$ with the field amplitude is depicted in Fig. 3 (off resonance) and Fig. 4 (near resonance). In the former case, LOPT fares not too badly, as expected, while MPT begins to deviate from NPT for $E_0\geq 0.17$, presumably because of the RWA which ignores the counterrotating terms (which are fully accounted for in NPT). At $\omega=0.3479$, LOPT, of course, breaks down completely, whereas the validity of MPT extends to higher field strengths, again because RWA gets better as exact resonance is approached.

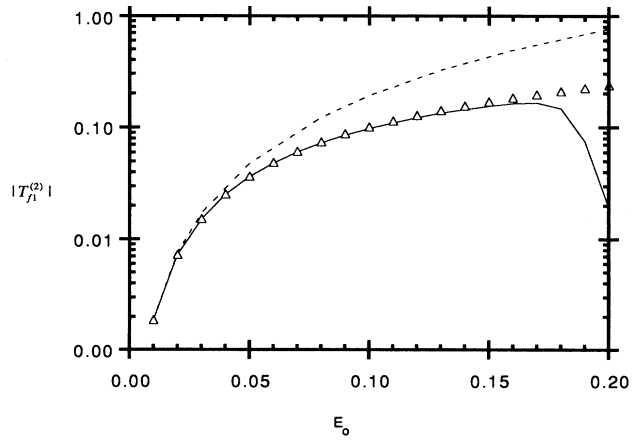

FIG. 3. Variation of the two-photon transition amplitude for the model described in Sec. III C in the nonresonance region, $\omega=0.3$, with E_0. Solid line-NPT; open triangles-MPT; dotted line-LOPT. All quantities are in atomic units.

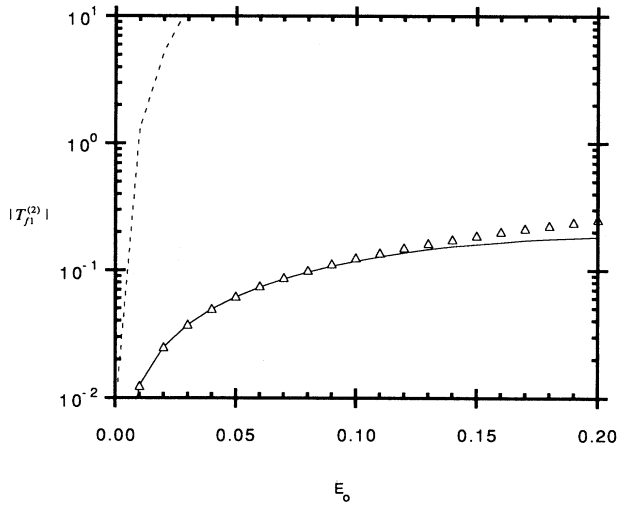

FIG. 4. Same as Fig. 3, but close to resonance, $\omega=0.3479$ (a.u.).

IV. SUMMARY AND CONCLUSIONS

In the foregoing, we have examined the conditions under which transitions from a bound to a continuum state of a quantum system, due to a time-periodic interaction, can be described using the Floquet theory, which rests on the possibility of representing the fully interacting wave function as a normalizable quasiperiodic state. This turns out to be the case, provided an appropriately truncated function space composed of the eigenfunctions of

the unperturbed Hamiltonian and the set of time-periodic functions of the form $\exp(in\omega t)$, is employed in constructing the wave function. The approximation involved in this procedure corresponds to the condition under which a transition rate to a given final state can be defined according to the general theory of damping phenomena [6], namely, the neglect of the imaginary part of the energy shift, in comparison with its real part. The numerical calculations described in Sec. III illustrate the application of the present formalism to specific models and bring out *inter alia* the special efficacy of this approach in dealing with intermediate resonances. In conclusion, to the extent that a transition rate can be defined at all, the Floquet theory provides a viable means of performing nonperturbative calculations for both bound-bound and bound-continuum transitions under the influence of a time-periodic perturbation.

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