Semiclassical Floquet theory of the S matrix for electromagnetic interactions

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The calculation of the S matrix for a system acted upon by an external electromagnetic field, when the fully interacting system can be represented by normalized Floquet states, is reformulated to include adiabatic switching and wave-function renormalization. This procedure yields, in general, nonvanishing matrix elements between states whose energies differ by an integral multiple of the photon energy. The induced level shifts do not disturb this condition; however, a *relative* level shift between the initial (in) and final (out) states is essential for a transition to occur. A numerical scheme to facilitate use of this formalism in practice has been devised and applied to a model calculation. The present nonperturbative results agree with lowest-order perturbation theory for weak fields, but deviate towards lower values at higher field strengths. Representative data for the simultaneous excitation of more than one level are presented to show that transition amplitudes are well defined in the presence of intermediate resonances. Gauge dependence of the level shifts and transition amplitudes is also studied, both theoretically and numerically.

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I. INTRODUCTION

In the calculation of transition probabilities due to the interaction of an intense external electromagnetic field with a quantum system, the fact that it is a very good approximation to treat the field classically ("laser approximation" [1]) has rendered nonperturbative approaches a viable proposition. For studying steady-state phenomena like change in the atomic or molecular spectra due to the induced level shifts, the use of Floquet's theorem to search for quasiperiodic states of the interacting system has been known [2] to be particularly advantageous. This approach has also been extended to encompass transitions to a continuum, by letting the induced level shift be complex, and interpreting transitions as decay in a steady field [3]. However, from the point of view of time-dependent perturbation theory, if the wave functions are properly normalized, the imaginary part of the level shift would be absorbed by the normalization factor, leaving only a real part, as argued by Langhoff, Epstein, and Karplus [4]. In particular, Sambe [5] has shown this to be the case when there is no continuum. In this circumstance, it would then appear that no transitions are possible at all. Such a null result was in fact obtained recently [6], using an S-matrix approach. In this work we show how this result gets modified when two factors known to be of considerable importance in formal scattering theory [7] and quantum electrodynamics [8], viz., adiabatic switching of the interaction and wave-function renormalization, are taken into account. First of all, it is no longer true that the existence of quasiperiodic states forbids transitions; however, if the level shifts of the initial (in) and final (out) states happen to be equal, the corresponding transition does not occur. The example of the harmonic oscillator considered in Ref. [6] belongs to the special case where all level shifts are equal, and therefore the null result applies there. Second, only those transitions are permitted which correspond to the absorption of an integral number of photons present in the external field, regardless of the field strength; the level shifts, which are intensity dependent, do not lead to any violation of this requirement, because of their transient nature. Finally, the wave-function renormalization factor is essential to ensure agreement with lowest-order perturbation theory (LOPT) in the weak-field limit.

This paper is organized as follows: In Sec. II the basic formula, which expresses the transition matrix element governing the absorption of a given number of photons, in terms of the level shifts of the initial and finals states and certain Floquet coefficients, is derived, and its first-order approximation shown to be the familiar golden-rule matrix element. Gauge dependence is discussed in Sec. III, where the need to use the "preferred gauge" [9], in which an expansion of the total wave function in terms of the eigenfunctions of the unperturbed Hamiltonian has direct physical significance, is brought out. Section IV presents a model numerical calculation using both the "length" and "velocity" forms of the interaction, which demonstrates that (a) the relative level shifts are gauge independent at any intensity or frequency; (b) the transition amplitudes have this property only in the LOPT regime (in accordance with the analysis of Sec. III); (c) at intensities beyond this regime, the amplitudes increase at a rate lower than the power law characteristic of LOPT, until ultimately they start to decrease; and (d) amplitudes for the simultaneous excitation of more than one level, which involves intermediate resonance, can be calculated without any difficulty.

II. LEVEL SHIFTS AND TRANSITION AMPLITUDES

We shall develop the formalism, as far as possible, parallel to the earlier treatment [6] in which adiabatic switching of the interaction was not imposed. Consider a one-electron system specified by a Hamiltonian $H_0=p^2/2+W(\mathbf{r})$, with $H_0\phi_a=\varepsilon_a\phi_a$. Since we are interested in transitions between the energy eigenstates ϕ_a due to an external electromagnetic interaction, we shall choose at the outset a gauge in which H_0 coincides with the unperturbed energy operator [9], so that an expansion of the interacting wave function in terms of ϕ_a is directly amenable to physical interpretation. This is of particular importance here, since the transition amplitudes are ultimately expressed in terms of the Floquet expansion coefficients of the basis states embedded in the field. A more detailed discussion of this point is taken up in Sec. III; for the present, we follow Forney, Quattropani, and Bassani [9] and consider a monochromatic beam propagating in the y direction with electric and magnetic fields given by

$$\mathbf{E}(\mathbf{r},t)=E_0\hat{\mathbf{z}}\cos\left[\omega\left[t-\frac{y}{c}\right]\right]$$

and

$$\mathbf{B}(\mathbf{r},t)=E_0\hat{\mathbf{x}}\cos\left[\omega\left[t-\frac{y}{c}\right]\right] .$$

Using atomic units in which $c=137$, the vector and scalar potentials in the "preferential gauge" [9] are

$$\mathbf{A}=-\frac{cE_0}{\omega}\hat{\mathbf{z}}\left\{\sin\left[\omega\left[t-\frac{y}{c}\right]\right]-\sin(\omega t)\right\} \tag{1}$$

and

$$\phi=-E_0 z\cos(\omega t) . \tag{2}$$

The interaction operator is then given by

$$V=\frac{cE_0}{\omega}\left\{\sin\left[\omega\left[t-\frac{y}{c}\right]\right]-\sin(\omega t)\right\}p_z + \frac{c^2E_0^2}{2\omega^2}\left\{\sin\left[\omega\left[t-\frac{y}{c}\right]\right]-\sin(\omega t)\right\}^2 + E_0 z\cos(\omega t) , \tag{3}$$

which can be written in the general form [6]

$$V=\sum_m V_m e^{-im\omega t} , \tag{4}$$

with $V_{-m}=V_m^\dagger$, so that V is Hermitian. It is assumed that any terms which commute with the total Hamiltonian $H=H_0+V$, such as those arising from the second term in Eq. (3), do not appear in Eq. (4) (see Appendix A). Further, it is understood that V is accompanied by a switching factor $\exp(-\eta|t|)$, with $\eta\to 0$ finally. Except where its presence is essential, this factor will be suppressed. We now look for an adiabatic Floquet solution to the Schrödinger equation,

$$i\frac{\partial\psi_a}{\partial t}=H\psi_a, \quad H=H_0+V , \tag{5}$$

evolving from the unperturbed state $\phi_a\exp(-i\varepsilon_a t)$ as $t\to-\infty$, of the form

$$\psi_a=e^{-i(\varepsilon_a+\Delta_a e^{-\eta|t|})t}[\phi_a+\{F_a(\mathbf{r},t)-\phi_a\}e^{-\eta|t|}] , \tag{6}$$

where F_a is a periodic function of t with period $2\pi/\omega$, i.e., we may write

$$F_a(\mathbf{r},t)=\sum_n e^{-in\omega t}\chi_n^a(\mathbf{r}) . \tag{7}$$

Equation (6) clearly satisfies the stipulated initial condition, and also reduces to the usual Floquet form [6] as $\eta\to 0$. Note that ψ_a will go over to the reference state ϕ_a as $|t|\to\infty$, since the evolution is adiabatic. However, what one is interested in is the amplitude for switching the reference state from ϕ_i to ϕ_f due to scattering, which happens, according to the adiabatic hypothesis (e.g., Ref. [7]), around $t=0$. This is also clear from the representation of the S operator in terms of the evolution operator U as [7]

$$S=U(\infty,0)U(0,-\infty) .$$

Before proceeding to the calculation of the S matrix, we shall indicate how Δ_a and F_a can be determined in practice. To this end, we expand χ_n in terms of the basis set $\{\phi_a\}$ as

$$\chi_n^a(\mathbf{r})=\sum_\mu \alpha_{n\mu}^a\phi_\mu . \tag{8}$$

Since the switching factors have been separated out in Eq. (6), i.e., Δ_a and $\alpha_{n\mu}^a$ are independent of η, we may substitute Eqs. (4), (6), (7), and (8) in Eq. (5), and take the limit $\eta\to 0$ to get

$$(\varepsilon_a+\Delta_a+n\omega-\varepsilon_\mu)\alpha_{n\mu}^a=\sum_m\sum_\nu \alpha_{n-m,\nu}^a\langle\mu|V_m|\nu\rangle . \tag{9}$$

As described in Sec. IV, Δ_a and $\alpha_{n\mu}^a$ can be determined numerically, using Eq. (9).

To calculate the S matrix, we proceed as in Ref. [6], except that the adiabatic prescription is followed and wave-function renormalization is also carried out. Thus we introduce the adiabatic level-shift operator

$$\Delta=\sum_c |\phi_c\rangle\Delta_c e^{-\eta|t|}\langle\phi_c| , \tag{10}$$

so that

$$\Delta\phi_a=\Delta_a\exp(-\eta|t|)\phi_a ,$$

and split H as $H=(H_0+\Delta)+(V-\Delta)$, with $(H_0+\Delta)$ defining a new reference system and $(V-\Delta)$ an interaction which does not produce any level shifts. The new reference states evolve as

$$\phi_a\exp[-i(\varepsilon_a+\Delta_a e^{-\eta|t|})t] ,$$

while ψ_a is still given by Eq. (6) since it is independent of the splitting of H, and can be normalized (in the sense of Sambe [5]) by requiring that

$$\sum_n \sum_\mu |\alpha^a_{n\mu}|^2 = 1 \ . \tag{11}$$

This takes care of the "in" state ψ_i [7]. As for the "out" state ψ_f, we note that the final state $\phi_f \exp(-i\varepsilon_f t)$ (to which ψ_f goes over as $t \to \infty$) has an amplitude α^f_{0f} in ψ_f at $t=0$. The renormalized S-matrix element connecting ϕ_i and ϕ_f ($f \neq i$) is therefore given by

$$S_{fi} = -i \lim_{\eta \to 0} \alpha^{f*}_{0f} \int_{-\infty}^{\infty} e^{i(\varepsilon_f + \Delta_f e^{-\eta|t|})t} \langle \phi_f | (V - \Delta) | \psi_i \rangle dt \ . \tag{12}$$

It is important to remember here that the operations $\eta \to 0$ and $\int dt$ cannot be interchanged, since the presence of η is required to render the evolution operator $U(t,t_0)$ well defined as $t \to \infty$ or $t_0 \to -\infty$ [7]. In fact, letting $\eta \to 0$ in the integrand will only lead to the null result of Ref. [6], which forbids any transition, even when perturbation theory permits it, e.g., for the model considered in Sec. IV.

The time integrals in Eq. (12) can be evaluated, after substituting for V, Δ, and ψ_i and using the following results proved in Appendix B:

$$\lim_{\eta \to 0} \int_{-\infty}^{\infty} \exp[i(x + Ye^{-\eta|t|})t] \, dt = 2\pi\delta(x) \tag{13}$$

and

$$\lim_{\eta \to 0} \int_{-\infty}^{\infty} \exp[i(x + Ye^{-\eta|t|})t - \eta|t|] dt = 2\pi\delta(x) \ . \tag{14}$$

After some simple rearrangements, we get

$$S_{fi} = -2\pi i \sum_n T^{(n)}_{fi} \delta(\varepsilon_f - \varepsilon_i - n\omega) \ , \tag{15}$$

$$T^{(n)}_{fi} = \alpha^{f*}_{0f} \left[\sum_n \langle f | V_m | \chi^i_{n-m} \rangle - \Delta_f \langle \phi_f | \chi^i_n \rangle \right] \ . \tag{16}$$

From Eq. (15) we see that the $T^{(n)}_{fi}$ satisfying the energy-conservation condition $\varepsilon_f - \varepsilon_i = n\omega$ are the transition amplitudes for the absorption of n *net* photons, from which the corresponding transition rates can be deduced as usual [1]. Using Eqs. (8) and (9), we may express these on-the-energy-shell T-matrix elements compactly as

$$T^{(n)}_{fi} = (\Delta_i - \Delta_f) \alpha^{f*}_{0f} \alpha^i_{nf}, \quad \varepsilon_f = \varepsilon_i + n\omega \ . \tag{17}$$

Two notable features of Eqs. (15) and (17) are the following: First of all, despite the induced level shifts, which are not constrained to be commensurate with the photon energy, the final energy transfer strictly corresponds to the absorption of an integral number of photons. This is as it should be, since the system is in one of its eigenstates before and after the scattering event, so that the transition energy does not involve any level shift. Since the field can lose energy only in integral multiples of ω, overall energy conservation demands that $\varepsilon_f - \varepsilon_i = n\omega$. Second, the transition amplitudes are proportional to the difference between the level shifts of the initial and final states. Thus level shifts are necessary, but not sufficient, to induce a transition. The example of the harmonic oscillator considered in Ref. [6] is a case in point. Here no transitions are permitted, since all level shifts are equal.

The intimate relationship between level shifts and transition amplitudes can be made more transparent by means of a detailed analysis of first-order transitions. It is worth stressing here that truncating Eq. (9) at any n does not imply nth-order perturbation theory, since n denotes the *net* number of photons absorbed. Further, the presence of Δ_a (which, being entirely due to the interaction, is at least of order V) on the left-hand side makes a general perturbation analysis extremely cumbersome. However, the first-order case can be handled with some facility by noting that in this case only the states ϕ_i and ϕ_f are involved, i.e., all virtual transitions are ignored. One may therefore make use of the well-known results for a two-level atom in a spatially homogeneous electromagnetic field in the rotating-wave approximation [10] to calculate the relevant parameters, as follows: Two normalized solutions can be formed, one of which goes over to $\phi_i \exp(-i\varepsilon_i t)$ and the other to $\phi_f \exp(-i\varepsilon_f t)$ as $V \to 0$, depending on whether $\omega > (\varepsilon_f - \varepsilon_i)$ or $\omega < (\varepsilon_f - \varepsilon_i)$. In the former case, these can be reduced to the desired form:

$$\psi_i = e^{-i(\varepsilon_i + \Delta_i)t} [\alpha^i_{0i} \phi_i + \alpha^i_{1f} e^{-i\omega t} \phi_f]$$

and

$$\psi_f = e^{-i(\varepsilon_f + \Delta_f)t} [\alpha^f_{-1i} e^{-i\omega t} \phi_i + \alpha^f_{0f} \phi_f] \ ,$$

where

$$\Delta_i = \frac{\Omega - \epsilon}{2}, \quad \epsilon = \omega - (\varepsilon_f - \varepsilon_i)$$

$$\Omega = [\epsilon^2 + 4|V^{fi}_1|^2]^{1/2}, \quad V^{fi}_1 = \langle f | V_1 | i \rangle$$

$$\alpha^i_{0i} = (\epsilon + \Omega)/[(\epsilon + \Omega)^2 + 4|V^{fi}_1|^2]^{1/2} \ ,$$

$$\alpha^i_{1f} = -2\alpha^i_{0i} V^{fi}_1 / (\epsilon + \Omega) \ ,$$

$$\Delta_f = \frac{\epsilon - \Omega}{2}, \quad \alpha^f_{-1i} = -\alpha^i_{1f}, \quad \text{and} \quad \alpha^f_{0f} = -\alpha^i_{0i} \ .$$

When energy conservation permits one photon absorption, $\epsilon = 0$, and we get

$$|\alpha^f_{0f}| = |\alpha^i_{1f}| = 2^{-1/2}$$

and

$$\Delta_i = -\Delta_f = |V^{fi}_1| \ ,$$

so that $|T^{(1)}_{fi}| = |V^{fi}_1|$, which is the standard first-order formula. A similar calculation for the case $\omega < \varepsilon_f - \varepsilon_i$ leads to the same result. In other words, the first-order transition amplitude is simply the lowest-order level shift of the initial state at resonance. (Note that the usual second-order ac Stark-shift formula [11,12] is not valid near resonance.) Rather than attempt to pursue this type of analysis for higher-order processes, we shall demonstrate the equivalence of the present nonperturbative theory (NPT) and LOPT for low field strengths by a numerical calculation (cf. Sec. IV).

III. GAUGE DEPENDENCE

As has already been stressed, the present scheme is designed for use in a gauge in which the unperturbed

Hamiltonian coincides with the energy operator. In fact, it is easy to see that, in an arbitrary gauge, there may not be any quasiperiodic solutions at all. After a gauge transformation,

$$\mathbf{A}\rightarrow\mathbf{A}+\nabla\Lambda(\mathbf{r},t),\quad \phi\rightarrow\phi-\frac{1}{c}\frac{\partial\Lambda(\mathbf{r},t)}{\partial t}\ , \tag{18}$$

the new wave function will be

$$\tilde{\psi}_a=e^{ie\Lambda/c}\psi_a\ . \tag{19}$$

Therefore, in order for $\tilde{\psi}_a$ to be quasiperiodic, given ψ_a is, Λ should be of the general form

$$\Lambda(\mathbf{r},t)=\gamma t+\tilde{\Lambda}(\mathbf{r},t)\ , \tag{20}$$

where γ is a constant and $\tilde{\Lambda}$ is a periodic function of t with period $2\pi/\omega$. The linear term only contributes a constant to the scalar potential [cf. Eq. (18)] and therefore, in accordance with the discussion in Appendix A, need no longer be considered. Clearly, a gauge transformation induced by $\tilde{\Lambda}$ leaves the level shifts unaffected. However, unwanted terms may yet arise from $\nabla\tilde{\Lambda}$. For example, in the dipole approximation, the preferred gauge interaction assumes the so-called "length" form $\mathbf{E}(t)\cdot\mathbf{r}$. Choosing $\Lambda=\mathbf{A}(t)\cdot\mathbf{r}$, where $\mathbf{A}=\mathbf{A}_0\sin(\omega t)$, $\mathbf{A}_0=-(cE_0/\omega)\hat{\mathbf{z}}$, one gets the "velocity" form $\mathbf{A}\cdot\mathbf{p}/c+A^2/2c^2$. Here, the A^2 term, which arises from $\nabla\Lambda$, commutes with H and has both time-dependent and time-independent Fourier components. In the light of Appendix A, the former must be discarded. If the constant term $A_0^2/4c^2$ is retained, the total level shift will be the same as in the length gauge. That this is so up to second order has been shown by Milonni and Ackerhalt [12]; our analysis generalizes this conclusion to all orders.

Since all terms in the interaction potential, which commute with H, can be removed by a unitary transformation involving $\nabla\Lambda$ and $\partial\Lambda/\partial t$, the new wave function will still have the form of Eq. (19), with Λ replaced by, say, $\bar{\Lambda}$, which does not produce any spurious effects. It is readily verified that the new Floquet coefficients $\tilde{\alpha}$ are related to the old α's through

$$\tilde{\alpha}^a_{n\mu}=\sum_m\sum_\nu\alpha^a_{n-m,\nu}\langle\phi_\mu|\xi_m|\phi_\nu\rangle\ , \tag{21}$$

where

$$\xi_m(\mathbf{r})=\frac{1}{2\pi}\int_0^{2\pi}e^{i[e(\bar{\Lambda}/c)+m\omega t]}dt\ . \tag{22}$$

To lowest order in $\bar{\Lambda}$, $\xi_m=\delta_{m0}$ and $\tilde{\alpha}^a_{n\mu}=\alpha^a_{n\mu}$. If, as in the transformation between the length and velocity forms, $\bar{\Lambda}$ is of the order of the interaction potential, this means that the Floquet coefficients are gauge independent when LOPT holds. Since we have already shown that the *relative* level shifts are exactly gauge independent, the transition amplitudes [Eq. (17)] are also gauge independent in LOPT. (It may be mentioned here that the explicit verification of the gauge independence of transition amplitudes for multiphoton processes, discussed in Ref. [13], is actually at this level.) When higher orders are important, the Floquet coefficients can be gauge dependent and only the preferred gauge can be expected to yield the correct transition amplitudes.

IV. NUMERICAL APPLICATION

Implementation of the above formalism in practice may proceed as follows: Let the maximum number of photon exchanges to be considered be N_p and let the Hilbert space of the unperturbed system be represented by N_a basis states. These can be finally fixed by means of an appropriate convergence criterion. Arranging $\alpha^a_{n\mu}$ in ascending order from $n=-N_p,\ldots,N_p$ and $\mu=1,\ldots,N_a$, Eq. (9) can be recast as an eigenvalue equation for Δ_a, with $N_a(2N_p+1)$ rows and columns. At low field strengths, Δ_a will be given by the eigenvalue such that the corresponding eigenvector will have α^a_{0a} as the dominant element. Δ_a at progressively higher field strengths may be determined by requiring it to be a smooth function of E_0 (adiabatic evolution). For any transition, two calculations are needed: one with the initial state as the reference state and the other with the final state as the reference state. The first one determines Δ_i and α^i_{nf}, and the second Δ_f and α^f_{0f}. Equation (17) then gives the amplitude for the transition $\phi_i\rightarrow\phi_f$ such that $\epsilon_f-\epsilon_i=n\omega$.

The model system chosen to illustrate the above scheme consists of an electron in a one-dimensional infinite potential well, described by the Hamiltonian

$$H_0=\frac{p^2}{2}+W(z)\ , \tag{23}$$

$$W(z)=\begin{cases}0, & -L/2\le z\le L/2\\ \infty, & |z|>L/2\ .\end{cases} \tag{24}$$

The interaction in the preferred gauge [Eq. (3)] is exactly equivalent to the length form in the dipole approximation. This gauge will be referred to as G1 in this section. The Fourier components [Eq. (4)] of V are

$$V^{\mathrm{G1}}_{\pm1}=\frac{E_0 z}{2}\ . \tag{25}$$

Measuring energy with respect to the ground state, the energy levels and wave functions of H_0 are [14]

$$E_n=\frac{\pi^2}{2L^2}(n^2-1) \tag{26}$$

and

$$\phi_n=\sqrt{2/L}\ \sin\left\{\left[\frac{n\pi}{L}\right]\left[z+\frac{L}{2}\right]\right\}\ . \tag{27}$$

The required matrix elements are easily evaluated to be

$$(V^{\mathrm{G1}}_{\pm1})_{nn'}=\begin{cases}\dfrac{-4nn'LE_0}{\pi^2(n^2-n'^2)^2}, & n+n'\ \text{odd}\\ 0, & n+n'\ \text{even}\ .\end{cases} \tag{28}$$

To study the effect of gauge transformation, we may also specify the interaction in the velocity gauge, referred to as G2,

$$V^{\mathrm{G2}}=Ap_z/c+A^2/2c^2\ , \tag{29}$$

where $A=-(E_0c/\omega)\sin(\omega t)$. As discussed earlier, the

time-dependent Fourier components of the A^2 terms should not be retained; however, to facilitate comparison of absolute level shifts, it is advantageous to keep the time-independent part. With this convention, the required matrix elements in this gauge are

$$(V_{\pm 1}^{\mathrm{G2}})_{nn'}=\begin{cases}\mp\dfrac{2E_0 nn'}{\omega(n^2-n'^2)L}, & n+n' \text{ odd}\\ 0, & n=n' \text{ even}\end{cases} \tag{30}$$

and

$$(V_0^{\mathrm{G2}})_{nn'}=\frac{E_0^2}{4\omega^2}\delta_{nn'}\ . \tag{31}$$

For comparison with LOPT, we shall use the well-known expressions for the level shifts [11] and transition amplitudes [15], viz.,

$$\Delta_n=\sum_{n'}\frac{2|V_{nn'}|^2(\epsilon_n-\epsilon_{n'})}{(\epsilon_n-\epsilon_{n'})^2-\omega^2} \tag{32}$$

and

$$T_{fi}^{(N)}=\sum_{j_1,\ldots,j_{N-1}}\frac{V_{fj_{N-1}}V_{j_{N-1}j_{N-2}}\cdots V_{j_1 i}}{[(N-1)\omega-\varepsilon_{j_{N-1}}][(N-2)\omega-\varepsilon_{j_{N-2}}]\cdots[\omega-\varepsilon_{j_1}]},\quad \varepsilon_f-\varepsilon_i=N\omega\ , \tag{33}$$

where $V_{ji}=\langle j|V_1|i\rangle$, in either gauge. All the results discussed in this section refer to a binding potential [Eq. (24)] with $L=2$ a.u. As with the numerical calculation of the second-order matrix elements for hydrogen by Bassani, Forney, and Quattropani [16], it was found that the convergence is much better in G1, both for LOPT and NPT. Typically, the value of N_a or N_p required in this gauge for convergence varied from about 7 at low field strengths to about 12 at high E_0.

Figure 1 shows the variation of Δ_1 and Δ_2 with ω for $E_0=1$ a.u. It might be worth stressing once again that these are obtained from two separate calculations with different initial conditions. Thus, $\Delta_f-\Delta_i$ *cannot* be interpreted as the change in $\varepsilon_f-\varepsilon_i$ due to the interaction, for any given initial condition. As expected, the level shifts were found to be gauge independent and the values displayed correspond to G1 as well as to G2. The level shifts diverge at exact resonance in LOPT [cf. Eq. (32)], whereas the NPT results exhibit a finite discontinuity at a slightly lower frequency. As expected, far away from resonance, NPT and LOPT are in full agreement.

A comparison of the level shifts and transition amplitudes in the two gauges and LOPT is presented in Table I, for $\omega=\varepsilon_2-\varepsilon_1$. Also given are the two Floquet coefficients governing the excitation of the level $n=2$. Convergence to the figures quoted could be achieved in G1 with $N_a=N_p=7$, while $N_a=14$ and $N_p=20$ were required in G2 at $E_0=2.0$. Since Eqs. (9) and (11) fix α_{nj}^a only to within an overall phase (for a given a), the NPT calculation actually determines only $|T_{fi}^{(n)}|$, which, of course, completely determines the transition probability. In Table I the level shifts in LOPT are not shown since they are divergent for this case. At $E_0=0.05$, representative of the low-intensity regime, both level shifts and the transition amplitude are gauge independent, and the latter is also in agreement with the LOPT result. At $E_0=2$, the level shifts are still gauge independent, while the transition amplitude is not. Further, NPT predicts a lower transition amplitude as compared to LOPT.

Data presented in Table II demonstrate how unsuitable gauge G2 is for calculations of multiphonon absorption cross sections. The results pertain to $\omega=(\epsilon_2-\epsilon_1)/3$ and $E_0=0.05$. We shall henceforth denote $\epsilon_j-\epsilon_i$ by ϵ_{ji}. The NPT results in G1 are seen to agree with LOPT, while the G2 data are approaching the G1 values slowly as N_p and N_a are increased. Calculations in G2 were stopped at $N_p=15$ and $N_a=24$, by which time the size of the matrix involved had reached 744×744. In the rest of this section, NPT refers to the gauge G1.

Figure 2 illustrates the field-strength dependence of the excitation of the level $n=4$ via one-photon absorption ($\omega=\epsilon_{21}$) and three-photon absorption ($\omega=\epsilon_{41}/3$), both of which can occur in LOPT also. [It is clear from the structure of the matrix elements $(V_{\pm 1})_{nn'}$, Eq. (28), that $(i+j+n)$ has to be even for $T_{ij}^{(n)}$ to be nonvanishing.] It is seen that nonperturbative effects set in at a much lower field strength for multiphoton excitation as compared to single-photon absorption. Further, in both cases, these effects are such as to diminish the rate of increase of the transition amplitudes in comparison with LOPT.

Finally, Fig. 3 shows the variation of transition amplitudes in the presence of an intermediate resonance. It is readily seen from Eq. (26) that if a level r can be excited through the absorption of l photons, i.e., if $l\omega=\epsilon_{r1}$, any level s satisfying

$$l(s^2-l)=k(r^2-1)$$

can also be excited through the absorption of k photons.

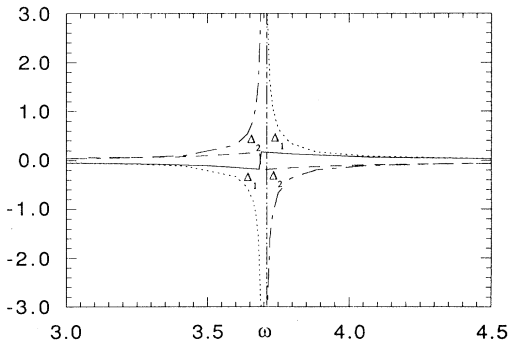

FIG. 1. Level shifts Δ_1 and Δ_2 at $E_0=2$ a.u., as a function of ω. Solid line, Δ_1 (NPT); dotted line, Δ_1 (LOPT); dashed line, Δ_2 (NPT); dot-dashed line, Δ_2 (LOPT).

TABLE I. Level shifts and transition amplitudes for single-photon excitation of the level $n=2$ of the system considered in Sec. IV, with $L=2$.

	Δ_1	Δ_2	$\lvert\alpha_{12}^1\rvert$	$\lvert\alpha_{02}^2\rvert$	$\lvert T_{21}^{(1)}\rvert$
		$E_0=0.05$			
LOPT					0.009 006
G1	0.089 82	−0.090 30	0.706 6	0.707 6	0.009 006
G2	0.089 82	−0.090 30	0.704 9	0.709 3	0.009 006
		$E_0=2.0$			
LOPT					0.360 3
G1	0.317 7	−0.394 8	0.675 1	0.718 3	0.345 5
G2	0.317 7	−0.394 8	0.584 1	0.766 1	0.318 8

Thus, when single-photon excitation of level 2 is permitted, five-photon excitation of level 4 is also allowed. $T_{21}^{(1)}$ refers to the former and $T_{41}^{(5)}$ to the latter, which is clearly beyond LOPT. The interesting feature here is the decrease of $T_{21}^{(1)}$ after $E_0 \simeq 11$. This is evidently the analog of "peak suppression," observed in above-threshold ionization [17], where the final states are in the continuum.

In conclusion, the formalism presented in this paper is numerically tractable, whenever the system of interest can be represented by a basis set of square-integrable functions. From a formal point of view, the chief merits of this theory are its adherence to the guidelines provided by scattering theory in the presence of level shifts and its ability to explain energy absorption in integral multiples of photon energy despite these level shifts. Its limitation is the uncertainty regarding its extension to systems having a continuous spectrum, but, then, as pointed out by Chu and Reinhardt [3], in this situation, the use of Floquet's theorem is itself questionable and, as such, this limitation is shared by any Floquet theory of multiphoton ionization. As is well known, in this case one would expect the level shift to be complex, making it impossible to normalize the wave function in a time-independent manner. Again, any Floquet theory of transition rates makes sense only if $\mathrm{Im}\Delta_i$ is sufficiently small, in which case the present scheme may also be expected to work, to the same degree of approximation. This is currently being explored. For the present, it can be stated without qualification that, under conditions where the interacting system has properly normalizable Floquet states, the exact transition probabilities can be calculated nonperturbatively, in a manner consistent with time-dependent scattering theory.

APPENDIX A: DIAGONAL MATRIX ELEMENTS OF THE INTERACTION

If the interaction potential V has nonvanishing matrix elements between the same basis states $|a\rangle$, we may write

$$V(t)=\tilde{V}(t)+v(t)\ ,$$

where

$$\tilde{V}=\sum_a \sum_{b\neq a} |a\rangle\langle b|V_{ab} \tag{A1}$$

and

$$v=\sum_a |a\rangle\langle a|V_{aa}\ , \tag{A2}$$

with $V_{ab}=\langle a|V|b\rangle$. Since $[H_0,v]|a\rangle=0$ for every a and the basis states form a complete set, v commutes with H_0. However, this is not sufficient to warrant omitting v. It is easy to see that

$$\langle b|[V,v]|a\rangle=V_{ba}(V_{aa}-V_{bb})\ . \tag{A3}$$

Therefore, if, and only if, the diagonal matrix elements of V are the same for all states, v also commutes with V, i.e., v commutes with the total Hamiltonian and cannot produce any genuine transitions. In general, let $V=\tilde{V}+\bar{V}$, where $[\bar{V},H]=0$. Any time-independent part of $\bar{V}$ simply contributes a uniform extra level shift; however, higher Fourier components of $\bar{V}$ can lead to unphysical transitions, and hence should be discarded. This has been amply demonstrated in Ref. [18]; the potential well considered in Sec. IV provides yet another example. Had we chosen the origin not at the center, but such that $V=0$, $0<z<L$, we would have gotten

TABLE II. Convergence of various parameters in the velocity (G2) gauge, for three-photon excitation of the level $n=2$ ($\omega=\epsilon_{21}/3$) at $E_0=0.05$. The numbers in brackets indicate powers of 10.

N_p	N_a	Δ_1	Δ_2	$\lvert\alpha_{32}^1\rvert$	$\lvert\alpha_{02}^2\rvert$	$\lvert T_2^{(3)}\rvert$
14	18	−4.93[−5]	1.76[−5]	0.1074	0.9999	7.182[−6]
15	20	−4.94[−5]	1.75[−5]	0.1078	0.9999	7.206[−6]
15	24	−4.94[−5]	1.74[−5]	0.1084	0.9999	7.233[−6]
G1		−4.94[−5]	1.73[−5]	0.1093	1.0	7.277[−6]
LOPT		−4.94[−5]	1.73[−5]			7.277[−6]

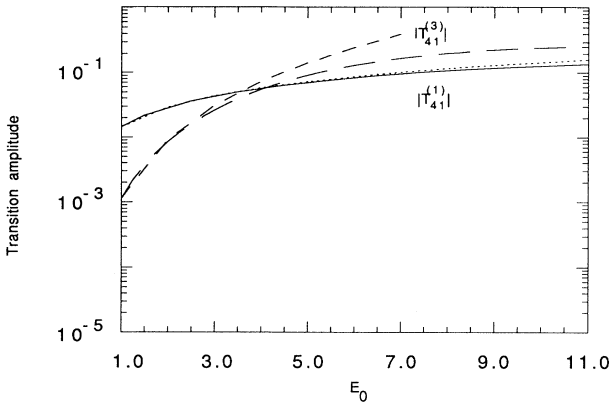

FIG. 2. Transition amplitudes for the excitation of $n=4$ as a function of field strength E_0. Solid line, $\omega=\varepsilon_{41}$ (NPT); dotted line, $\omega=\varepsilon_{41}$ (LOPT); long-dashed line, $\omega=\varepsilon_{41}/3$ (NPT); short-dashed line, $\omega=\varepsilon_{41}/3$ (LOPT).

$$\langle n|V_{\pm 1}|n\rangle=\frac{E_0 L}{4}\ , \tag{A4}$$

the nondiagonal matrix elements remaining unaltered. Inclusion of (A4) in numerical calculations affects transition amplitudes significantly for sufficiently high field strengths. This is clearly a spurious effect, since physical quantities cannot depend on the choice of the origin of the coordinate system. The matrix elements (A4) have no place in a calculation of transition amplitudes, since they are independent of $|n\rangle$.

APPENDIX B: PROOF OF EQS. (13) AND (14)

The integral of Eq. (13) may be written as

$$I=\lim_{\eta\to 0}[I(x)+I(-x)]\ , \tag{B1}$$

where

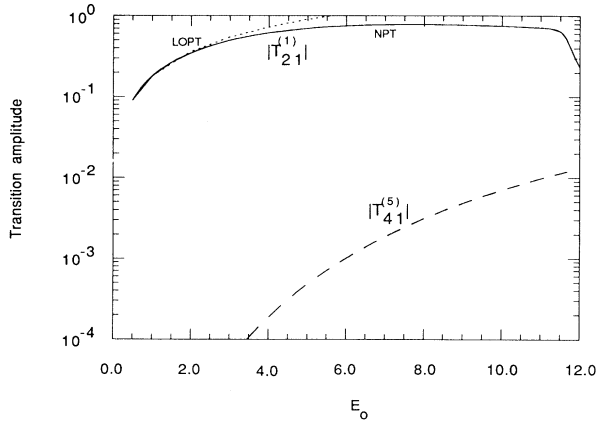

FIG. 3. Simultaneous excitation of $n=2$ with the absorption of one photon and $n=4$ with the absorption of five photons, at $\omega=\varepsilon_{21}$.

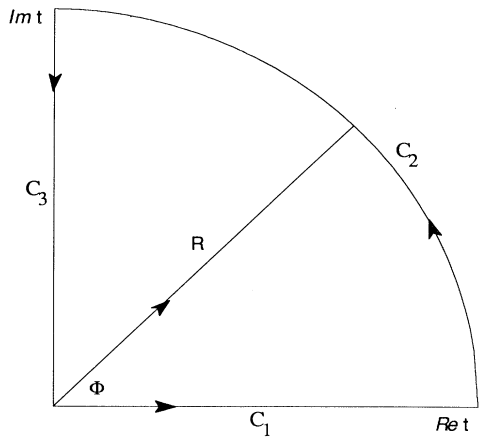

FIG. 4. Integration contour for $I(X)$ (Appendix B), for $X>0$.

$$I(x)=\int_0^\infty \exp[i(x+Ye^{-\eta t})t]dt\ . \tag{B2}$$

First we note that, if $x=0$, the integrand tends to 1 as $t\to\infty$, so that

$$I(0)=\infty\ . \tag{B3}$$

If $x>0$, we may carry out the integration in the complex t plane using the contour of Fig. 4 to write

$$I(x)=-\lim_{R\to\infty}[I_{c_2}+I_{c_3}]\ . \tag{B4}$$

It is readily verified that as $R\to\infty$, $I_{c_2}\to 0$, and we get

$$I(x)=i\int_0^\infty \exp[-(x+Ye^{-i\eta t})t]dt\ . \tag{B5}$$

To evaluate $I(-x)$, we may use the above contour reflected about the Ret axis so that the contribution from the arc again vanishes as $R\to\infty$. This gives

$$I(-x)=-i\int_0^\infty \exp[-(x+Ye^{i\eta t})t]dt\ . \tag{B6}$$

Using Eqs. (B5) and (B6) in Eq. (B1), we get

$$I=\lim_{\eta\to 0} i\int_0^\infty e^{-xt}\{\exp(-Ye^{-i\eta t}t)-\exp(-Ye^{i\eta t}t)\}\ . \tag{B7}$$

The integrand is clearly zero at $t=\infty$, whether η is zero or not. For any finite t, we may expand the exponentials in the curly braces to show that they cancel each other in the limit $\eta\to 0$. That is, the integrand tends to zero as $\eta\to 0$, for any t. Therefore,

$$I(x>0)=0\ . \tag{B8}$$

The case $x<0$ can be handled in the same way by interchanging the contours to get

$$I(x<0)=0 \ . \tag{B9}$$

From Eqs. (B3), (B8), and (B9), we see that $I(x)$ must be proportional to $\delta(x)$; the normalization constant can be determined by noting that these relations also imply that I is actually independent of Y, so that one may set $Y=0$ in (B2). This immediately yields Eq. (13). Equation (14) can be proved in exactly the same way; the extra factor, $\exp(-\eta|t|)$, makes essentially no difference to the arguments.

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