Nonperturbative analysis of the two-level atom: Applications to multiphoton excitation

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Excitation of an atomic system subjected to a slowly varying external electromagnetic field is studied using a two-level model. Time evolution of the system is found using an approach which is nonperturbative in the field strength. There is no constraint to small values of the applied field, that is, the field (in appropriate energy units) need not be small compared to the difference in energies of the two levels. Rather, we use the fact that the situation of interest to us is where the frequency of the exciting field is small compared to the frequency associated with the level difference. Transition probabilities and resonance conditions are found which circumscribe both the large- and small-field limits. In the weak-field limit the previous results of high-order perturbation theory are readily recovered. For a monochromatic field the characteristic features of resonance excitation at high harmonic number of the applied field are (a) extremely narrow resonance widths and (b) shifts in resonance positions which are strong functions of field intensity. Because of this sensitivity, we are able to demonstrate that when slow temporal evolution of the field amplitude is taken into account (e.g., due to finite pulse duration) the appropriate mean excitation rate is that due to the uncorrelated contribution of many resonances. The results of this analysis are used to estimate excitation rates in a specific atomic system, Cd^{12+}, which are then compared to multiphoton-ionization rates. Our calculations suggest that the ionization rate exceeds the excitation rate by several orders of magnitude.

I. INTRODUCTION

There has been a great deal of interest, both experimental[1] and theoretical,[2] in atomic processes due to very strong electromagnetic fields. In fact, current laser technology enables production of oscillating electric fields comparable to internal atomic electric fields. Transitions involving the emission or absorption of many field quanta, multiphoton processes, can occur under these circumstances. Most experimental studies in this area have investigated multiphoton ionization. There is the suggestion, however, based on Rhodes's observations in krypton,[3] that excitation by multiphoton processes has been observed experimentally. In light of these results, it has been proposed that selective excitation by comparatively-low-frequency electromagnetic fields may be feasible.[4]

In this report we address this problem of multiphoton excitation of an atomic system, as might result from its interaction with such an intense laser field. We use a simple model of this interaction between two levels in such a system

$$i\frac{d\mathbf{a}}{dt}=\underline{H}(t)\mathbf{a}\ ,$$

or equivalently,

$$i\frac{d}{dt}\begin{bmatrix}a_1\\ a_2\end{bmatrix}=\begin{bmatrix}E_1 & F\sin(\omega t)\\ F\sin(\omega t) & E_2\end{bmatrix}\begin{bmatrix}a_1\\ a_2\end{bmatrix}. \tag{1}$$

Here a_1 and a_2 are the state amplitudes, with associated frequencies in the absence of the external fields E_1 and E_2, respectively, whose time development we wish to find. The coupling parameter F is proportional to the amplitude of an electromagnetic field of frequency ω, and, in general, is itself a slowly varying function of time (with $\dot{F}/F \ll \omega$), because of effects such as finite pulse duration. We take $E_2 > E_1$, $\omega > 0$, and $F > 0$. The electromagnetic field is treated semiclassically and in the dipole approximation. The correspondence between this model and the quantized treatment of the electromagnetic field is given in the fundamental paper by Shirley.[5] (The model also describes various other physical problems, for example, the state of a spin-$\frac{1}{2}$ particle immersed in an oscillating magnetic field oriented perpendicular to a static magnetic field.)

Solutions of Eq. (1) have been obtained from perturbation theory with F small (e.g., see Shirley[5]). However, we observe that the characteristic feature of high-order multiphoton processes is that $\omega \ll \Delta \equiv (E_2 - E_1)/2$. Here we develop a theory which is asymptotic in ω/Δ, spans the domain $F \sim \Delta$, and is constrained only by the condition $F \ll \Delta^2/\omega$. The identification with the quantized treatment of the electromagnetic field is that $2\Delta/\omega$ is identified with the number of photons involved in the transition.

In Sec. II the theory is developed for the case $F=$const. In order to find the time evolution of Eq. (1), we introduce the WKB representation in the complex time plane. Solving the problem then amounts to finding the small changes in this representation over each period of the applied field. The most essential feature of our approach is that the *small size of these changes,* on the real time axis, *necessitates moving our path into the complex plane.* For various complex values of time, there occur jumps in the WKB representation which are comparable in size to the solution itself. By moving our path into such regions, we find these jumps and thus determine a transfer matrix which advances the system over one period of the applied field. The Nth power of this matrix then determines the state of the system after N periods. This method is to be contrasted with Shirley's approach using Floquet theory, where it is necessary to find the eigenvalues of very large matrices.

We find general expressions for the time-dependent excitation probability P, Eq. (39), and exhibit its dependence on field frequency and intensity. In the limit $F/\Delta \ll 1$, we recover the resonance shifts and the $(F^2)^{\Delta/\omega}$ power law for the generalized Rabi frequency[6] as found by the perturbative treatment of Shirley's Floquet Hamiltonian. Simplified asymptotic expressions are also given for each of these quantities in the limit $F/\Delta \gg 1$.

In Sec. III the effects of the temporal intensity envelope ($\dot{F}/F \ll \omega$) of the applied field are considered. Because of the strong dependence of the resonance positions on intensity and because of their narrowness, any given resonance will be excited only during a small interval of the laser pulse. Any slight randomness introduced into the system will ensure that the contribution to the excited-state amplitude from successive resonances will be uncorrelated. Given these conditions, an average rate of excitation is then found.

We apply our results in Sec. IV to model a Cd^{12+} ion which has been proposed as a candidate for multiphoton excitation. Using the results of Sec. III, the rate of excitation between the $4s^24p^6$ and $4s4p^65p$ levels is considered and is compared with estimates of rates for multiphoton ionization from the upper level.[7] It is found that the rate of ionization clearly dominates the rate of excitation that was predicted by considering the two-state interaction alone. We realize that application of the two-level model to atomic systems is certainly suspect if intermediate resonant states can participate in the transition from initial to final state. However, the results of Sec. III suggest that maintenance of individual high-order intermediate resonances will be extremely difficult experimentally.

II. TIME EVOLUTION IN THE PRESENCE OF A LOW-FREQUENCY, PERIODIC ELECTRIC FIELD

Rather than deal directly with the system of two coupled first-order equations for **a**, we generate an equivalent second-order system of equations which is coupled only through the specification of initial data. To this end, we define two new dependent variables, y and z, through the relation

$$\mathbf{a} = \exp(-iAt)\underline{R}\begin{bmatrix} y \\ z \end{bmatrix} , \tag{2}$$

with the unitary matrix $\underline{R}$ defined by

$$\underline{R} \equiv \frac{1}{\sqrt{2}}\begin{bmatrix} 1 & -1 \\ 1 & 1 \end{bmatrix} , \tag{3}$$

and with $A \equiv (E_2+E_1)/2$. Differentiation of Eq. (1) yields the second-order system

$$\frac{d^2y}{d\tau^2} + Q_y y = 0 \tag{4a}$$

and

$$\frac{d^2z}{d\tau^2} + Q_z z = 0 , \tag{4b}$$

for y and z, respectively. Here we have transformed to the dimensionless time variable $\tau \equiv \omega t$ and introduced the quantities

$$Q_y \equiv \left[\frac{\Delta}{\omega}\right]^2 \left[1 + \left[\frac{F}{\Delta}\right]^2 \sin^2\tau + i\frac{\omega F}{\Delta^2}\cos\tau\right] \tag{5a}$$

and

$$Q_z \equiv \left[\frac{\Delta}{\omega}\right]^2 \left[1 + \left[\frac{F}{\Delta}\right]^2 \sin^2\tau - i\frac{\omega F}{\Delta^2}\cos\tau\right] , \tag{5b}$$

which are complex periodic functions of τ. The periodic nature of the problem allows us to construct a solution for arbitrary time from the general solution obtained over one period. [On the surface it would appear that the solution of the equations for y and z would require initial data for four independent complex amplitudes, whereas solution of Eq. (1) requires initial data for only two complex amplitudes. The extra information is provided by differentiating Eq. (2) and substituting into Eq. (1). That is, if **a** is known initially, then so is $d\mathbf{a}/dt$.] The development of the solutions for y and z proceeds in a parallel fashion. Therefore, we present the details of the method for y alone. For notational simplicity, we shall temporarily write Q for Q_y.

So far, Eqs. (4) are exact. However, by observing that the situation of interest to us (multiphoton processes) is that when

$$\frac{\Delta}{\omega} \gg 1 , \tag{6}$$

we can make use of the adiabatic (WKB) approximation since Q is large and slowly varying for τ real. Most importantly, this does not, however, preclude the situation where $F/\Delta > 1$.

The accuracy of the WKB approximation is reflected in the fact that if one were to attempt to integrate directly Eq. (4) along the real τ axis, one would find that the changes in the amplitudes of the two eikonal solutions are extremely small and difficult to compute. However, near the complex points τ_j where Q vanishes, the changes in these amplitudes are relatively finite. The essential feature of our method is to determine the behavior of the solution near these points and to carry the solution asymptotically back to the real time axis. Our method is equivalent to that of Zwaan[8,9] (see, e.g., Pokrovskii and Khalatnikov[10]). It has just recently come to our attention that similar but less complete techniques have been used on this problem in the past (see, for example, Delone and Krainov[11]).

There are infinitely many zeros of Q. For each integer k, there is a pair of zeros τ_k^+, τ_k^-, such that

$$\tau_k^{\pm} = k\pi \pm s_k , \tag{7}$$

where

$$s_k = \mathrm{Im}\, s + (-1)^k \mathrm{Re}\, s \ , \tag{8}$$

with s the solution of

$$\cos s = \frac{\Delta}{F}\left\{ i\frac{\omega}{2\Delta} + \left[1 + \left[\frac{F}{\Delta}\right]^2 - \left[\frac{\omega}{2\Delta}\right]^2 \right]^{1/2} \right\} , \tag{9}$$

for which $\mathrm{Im}\, s > 0$ and $|\mathrm{Re}\, s| < \pi/2$.

The anti-Stokes lines, defined by

$$\mathrm{Im}\left[\int_{\tau_j}^{\tau} Q^{1/2} d\tau \right] = 0 \ ,$$

emanating from the points $Q(\tau_j)=0$, with τ_j any zero of Q, have special significance in that, asymptotically, the relative size of the two independent solutions to Eq. (4) is maintained along such lines. Because of the reflection symmetry of Q about the lines $\mathrm{Re}\tau = (n+\frac{1}{2})\pi$ for integer n, one sees that a series of anti-Stokes lines connects the set of zeros τ^+ and another series connects the set τ^-. The location of the anti-Stokes lines and the Stokes lines, for which

$$\mathrm{Re}\left[\int_{\tau_j}^{\tau} Q^{1/2} d\tau \right] = 0 \ ,$$

are plotted as solid and dashed lines, respectively, in Fig. 1, where k is chosen to be even.

If, as we assume, $F\omega/\Delta^2 \ll 1$, then the phase integral between the zeros $j \neq j'$ is large, i.e.,

$$\int_{\tau_j}^{\tau_{j'}} Q^{1/2} d\tau \gg 1 \ .$$

Therefore, the nonasymptotic regions,

$$\int_{\tau_j}^{\tau} Q^{1/2} d\tau < 1 \ ,$$

surrounding each zero do not overlap. Furthermore, the phase integral between the zeros τ_k^+ and τ_k^- for a given value of k (which are roughly vertically displaced with respect to each other in the complex τ plane) is much less than that between zeros of differing k. This suggests that it is possible to develop the asymptotic representation of the solution in an interval of the real τ axis including $\tau = k\pi$ by taking explicit account of at most τ_k^+, τ_k^-. Thus motivated, we apply Langer's method[12] to Eq. (4) in order to determine systematically the asymptotic behavior of y in a region surrounding a particular zero τ_j.

Specifically, we transform both independent and dependent variables according to

$$\left[\frac{d\tau}{d\xi}\right]^2 Q = \xi \tag{10}$$

and

$$u = \left[\frac{d\tau}{d\xi}\right]^{-1/2} y \ , \tag{11}$$

and obtain the resulting equation,

$$\frac{d^2u}{d\xi^2} + \xi u = \tfrac{1}{2}\{\tau,\xi\} u \ . \tag{12}$$

Here $\{\tau,\xi\}$ is the Schwarzian derivative defined by

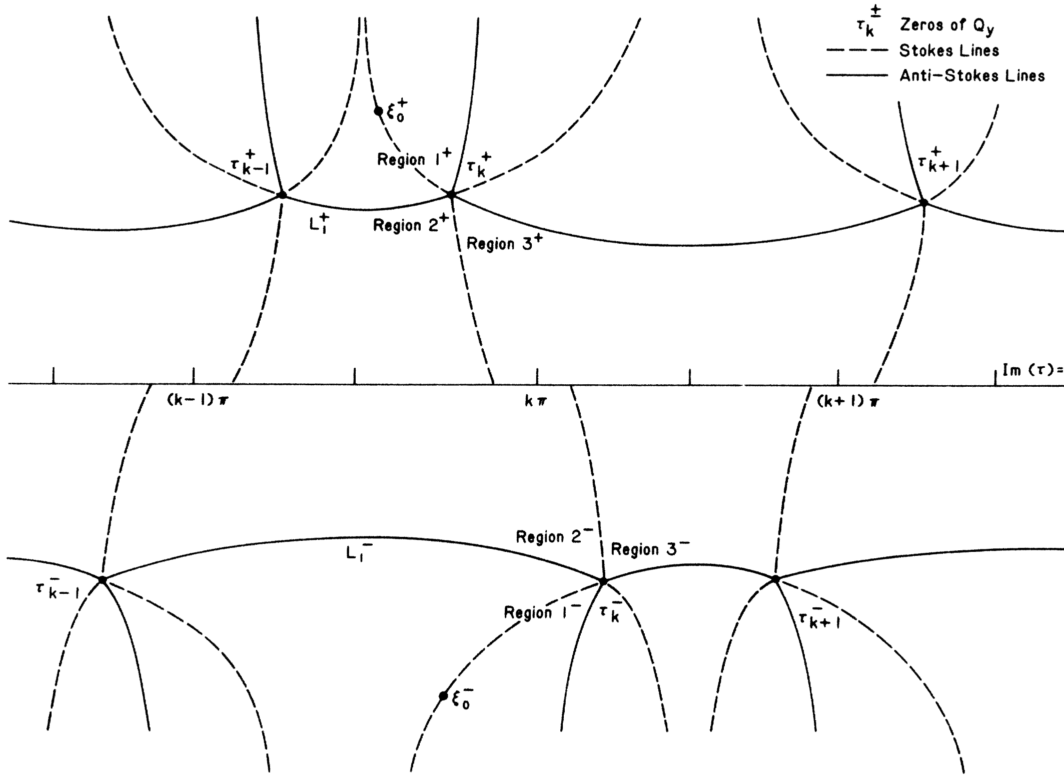

FIG. 1. Stokes and anti-Stokes line structure for Q_y in the complex τ plane for an interval surrounding $\tau = k\pi$, where k is taken to be even. The structure repeats as τ changes by 2π.

$$\{\tau,\xi\}\equiv\frac{\tau'''}{\tau'}-\frac{3\tau''^2}{2\tau'^2}\ , \tag{13}$$

with primes denoting differentiation with respect to ξ.

The solution of Eq. (10) requires the specification of a constant of integration in order to determine completely the functional form $\xi(\tau)$. The choice $\xi(\tau_j)=0$ makes τ' finite at the point $\tau=\tau_j$ which guarantees the boundedness of $\{\tau,\xi\}$. With this choice, we have

$$\tfrac{2}{3}[\xi(\tau,\tau_j)]^{3/2}=\int_{\tau_j}^{\tau}Q^{1/2}d\tau\equiv\Phi(\xi)\ . \tag{14}$$

A comparison of the right-hand side of Eq. (12) with the second term on the left shows the former to be smaller, nominally by $(\omega/\Delta)^2$. If the right-hand side were zero, then the general solution for u would be a linear combination of the two Airy functions $Z_1(\xi)$ and $Z_2(\xi)$. Without approximation, Eq. (12) can be recast as an integral equation,

$$u=AZ_1(\xi)+BZ_2(\xi)-\tfrac{1}{2}\int_{\xi_0}^{\xi}d\eta\,G(\xi,\eta)\{\tau,\eta\}u(\eta)\ , \tag{15}$$

by formal use of the method of variation of parameters. Here the kernel,

$$G(\xi,\eta)\equiv\frac{Z_1(\xi)Z_2(\eta)-Z_2(\xi)Z_1(\eta)}{W}\ ,$$

with the Wronskian,

$$W\equiv Z_1(\xi)Z_2'(\xi)-Z_2(\xi)Z_1'(\xi)\ ,$$

a constant. An asymptotic series for u in inverse powers of ξ is developed by treating the integral term in Eq. (15) as small, relative to the first two terms. We now construct such a solution whose domain of validity extends away from τ_j to include an interval of the real τ axis. Referring to Fig. 1, we consider the region surrounding the particular zero $\tau_j=\tau_k^+$. Let $Z_1(\xi)$ be that solution of

$$Z_j''+\xi Z_j=0\ , \tag{12'}$$

which is subdominant in region 1^+. Choose $B=0$. Then, inspection of the integral term shows that it will be asymptotically small throughout that region for which the integral from ξ_0 to ξ can be performed along a path for which

$$\mathrm{Im}[\Phi(\eta)-\Phi(\xi)]>0$$

everywhere. Choosing $\xi_0=\xi_0^+$, a point which lies on the Stokes line in region 1^+, guarantees that this inequality is satisfied in a domain which encompasses the real τ axis throughout an interval of size $|\mathrm{Re}(\tau-k\pi)|\simeq\pi$. A second independent solution is constructed completely analogously by making the replacements (see Fig. 1),

$$\tau_k^+\to\tau_k^-\ ,$$

region $1^+\to$ region 1^- ,

$$\xi_0^+\to\xi_0^-\ .$$

The representation of the functions Z_1 outside of regions $1^\pm$ is accomplished by applying the well-known connection formulas[13] developed for second-order equations with a single first-order turning point. To this end, we define

$$(a,\tau)\equiv Q^{-1/4}(\tau)\exp\left[+i\int_a^{\tau}Q^{1/2}d\tau\right] \tag{16a}$$

and

$$(\tau,a)\equiv Q^{-1/4}(\tau)\exp\left[-i\int_a^{\tau}Q^{1/2}d\tau\right]\ , \tag{16b}$$

and introduce the notation $Z_k^\pm$ for $Z_1(\xi(\tau,\tau_k^\pm))$. Then, in regions $1^\pm$ we have

$$Z_k^+\sim(\tau_k^+,\tau)(\tau')^{1/2}\ , \tag{17a}$$

$$Z_k^-\sim(\tau,\tau_k^-)(\tau')^{1/2}\ , \tag{17b}$$

respectively. These forms continue unchanged upon crossing the anti-Stokes lines $L_1^\pm$, respectively, but each solution becomes dominant in its region 2. As the solutions are continued across the Stokes lines emanating from τ_k^+ and τ_k^-, each acquires a subdominant part,

$$Z_k^+\sim[(\tau_k^+,\tau)+i(\tau,\tau_k^+)](\tau')^{1/2}\ , \tag{18a}$$

$$Z_k^-\sim[(\tau,\tau_k^-)-i(\tau_k^-,\tau)](\tau')^{1/2}\ , \tag{18b}$$

in region $3^\pm$. Upon returning to the line $\mathrm{Im}\tau=0$, Eqs. (17) and (18) provide sufficient information to evolve the asymptotic representation for y over a full period $\Delta\tau=2\pi$.

We represent y asymptotically in the WKB form

$$y\sim c_k^+(\tau_k^+,\tau)+c_k^-(\tau,\tau_k^-),\quad |\tau-k\pi|<\pi\ . \tag{19}$$

With this representation, the Stokes phenomenon appears as a discontinuous jump in the c_k as $\tau=k\pi$ is crossed. Denoting $\mathbf{c}_k=\binom{c_k^+}{c_k^-}$, the relation between $\mathbf{c}_k^>$ defined through Eq. (19) for $\tau-k\pi>0$ and $\mathbf{c}_k^<$ defined for $\tau-k\pi<0$ is

$$\mathbf{c}_k^>=\underline{S}\mathbf{c}_k^<\ , \tag{20}$$

where

$$\underline{S}=\begin{bmatrix}1 & -i[\tau_k^-,\tau_k^+]\\ i[\tau_k^-,\tau_k^+] & 1\end{bmatrix}\ , \tag{21}$$

with

$$[a,b]\equiv\exp\left[i\int_a^bQ^{1/2}d\tau\right]\ .$$

The functions τ_k^-,τ_k^+ satisfy $[\tau_k^-,\tau_k^+]=(-1)^k\varepsilon$, where

$$\varepsilon\equiv[\tau_0^-,\tau_0^+] \tag{22}$$

can be written in terms of elliptic functions. Its magnitude has the asymptotic forms

$$|\varepsilon|\sim\left[\frac{e}{4}\frac{F}{\Delta}\right]^{2\Delta/\omega},\quad F/\Delta\ll1$$

$$\sim\exp\left[-\frac{\pi}{2}\frac{\Delta}{\omega}\frac{\Delta}{F}\right],\quad F/\Delta\gg1 \tag{23}$$

with e the base of the natural logarithm, and is small whenever our theory applies. A direct numerical evaluation of ε versus F/Δ for $\Delta/\omega=50$ is shown together with these asymptotic forms in Fig. 2. As long as $\omega/F \ll 1$, the phase of ε is nearly constant,

$$\phi(\varepsilon) \simeq -\frac{\pi}{2} . \tag{24}$$

With the introduction of ε, we have

$$\underline{S}=\begin{bmatrix} 1 & -i(-1)^k\varepsilon \\ i(-1)^k\varepsilon & 1 \end{bmatrix} . \tag{25}$$

When τ lies in the interval $k\pi<\tau<(k+1)\pi$, y can as well be written in terms of $\mathbf{c}_{k+1}^{<}$. Using the definition, Eq. (19), we obtain the relation,

$$\mathbf{c}_{k+1}^{<}=i(-1)^k \begin{bmatrix} \exp(i\theta_0) & 0 \\ 0 & \exp(-i\theta_0) \end{bmatrix} \mathbf{c}_k^{>} , \tag{26}$$

with

$$\theta_0 \equiv [0,\pi]=\frac{\Delta}{\omega}\int_0^{\pi}\left[1+\left[\frac{F}{\Delta}\right]^2 \sin^2\tau + i\frac{\omega F}{\Delta^2}\cos\tau\right]^{1/2} d\tau , \tag{27}$$

evidently real. In the small- and large-field limits, θ_0 has the asymptotic forms

$$\theta_0 \sim \pi\frac{\Delta}{\omega}\left[1+\frac{1}{4}\left[\frac{F}{\Delta}\right]^2\right], \quad F/\Delta \ll 1$$

$$\sim 2F/\omega, \quad F/\Delta \gg 1 . \tag{28}$$

If we repeat this procedure for the interval $\pi \le \tau - k\pi \le 2\pi$, we now obtain a transfer matrix relating $\mathbf{c}_{k+2}^{<}$ to $\mathbf{c}_k^{<}$,

$$\mathbf{c}_{k+2}^{<}=\underline{M}_c \mathbf{c}_k^{<} . \tag{29}$$

With accuracy to order ε, we obtain

$$\underline{M}_c=\begin{bmatrix} \exp(i2\theta_0) & 2\varepsilon\sin\theta_0\exp(i\theta_0) \\ 2\varepsilon\sin\theta_0\exp(-i\theta_0) & \exp(-i2\theta_0) \end{bmatrix} . \tag{30}$$

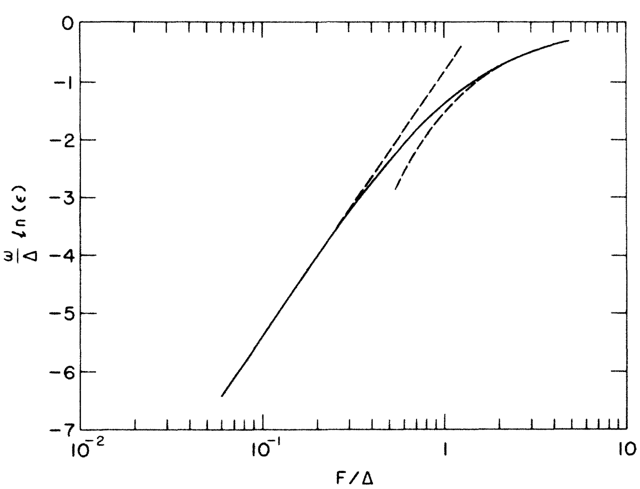

FIG. 2. $(\omega/\Delta)\ln|\varepsilon|$ vs F/Δ for $\Delta/\omega=50$. The solid curve shows the numerical estimate. The dashed curves show the asymptotic estimates from Eq. (23). The numerical results are insensitive to Δ/ω as long as $\Delta/\omega \ge 2$.

The procedure can be repeated in a completely analogous fashion for z wherein Q_y is everywhere replaced by Q_z and the corresponding coefficients $d_k^{\pm}$ are defined through the asymptotic relation

$$z \sim d_k^{+}(\tau_k^{+},\tau)+d_k^{-}(\tau,\tau_k^{-}), \quad |\tau-k\pi|<\pi . \tag{31}$$

The values $\tau_k^{\pm}$ are now those for which $Q_z(\tau_k^{\pm})=0$. The corresponding matrix $\underline{M}_d$ is the same as $\underline{M}_c$ except for the replacement of ε with $-\varepsilon$.

Having obtained evolution of y and z over one period of the applied field, we are now equipped to determine the evolution of $\mathbf{a}$. In the limit of interest here, $\Delta/\omega \gg 1$, we are led naturally to represent the state vector $\mathbf{a}$ in terms of its projections onto the instantaneous (normalized) eigenvectors $\mathbf{e}^{\pm}$ of the Hamiltonian $\underline{H}(\tau)$, defined through

$$\underline{H}\mathbf{e}^{\pm}=-\omega\lambda^{\pm}\mathbf{e}^{\pm} , \tag{32}$$

with

$$\lambda^{\pm} \equiv \pm\left[\frac{\Delta}{\omega}\right]\left[1+\left[\frac{F}{\Delta}\right]^2\sin^2\tau\right]^{1/2} , \tag{33}$$

the corresponding instantaneous eigenvalues. Writing

$$\mathbf{a}(t)=a_k^{+}\mathbf{e}^{+}\exp\left[i\int_{k\pi}^{\tau} d\tau\lambda^{+}\right]+a_k^{-}\mathbf{e}^{-}\exp\left[i\int_{k\pi}^{\tau} d\tau\lambda^{-}\right] , \quad |\tau-k\pi|<\pi \tag{34}$$

we are to determine the transfer matrix for $a_k^{\pm}$ in terms of $\underline{M}_c$ or $\underline{M}_d$.

In order to find the relations between $c_k^{\pm}$, $d_k^{\pm}$, and $a_k^{\pm}$, we begin by making the expansion

$$\int d\tau Q^{1/2} \simeq \left[\frac{\Delta}{\omega}\right]\int d\tau(1+f^2)^{1/2} \pm\frac{i}{2}\int d\tau\frac{\dot f}{(\Delta^2+f^2)^{1/2}} ,$$

where $f=F/\Delta\sin\tau$, in Eqs. (19) and (31). The resulting expressions are substituted into Eq. (2) which, when it is then compared to Eq. (34), determines $c_k^{\pm}$ and $d_k^{\pm}$ in terms of $a_k^{\pm}$. For $c_k^{\pm}$, we have

$$\mathbf{a}_k \equiv \begin{bmatrix} a_k^{+} \\ a_k^{-} \end{bmatrix} = \left[\frac{2\omega}{\varepsilon\Delta}\right]^{1/2} \mathbf{c}_k , \tag{35}$$

so that the transfer matrix which evolves $\mathbf{a}_k$ over an integral number N of periods is, finally,

$$\mathbf{a}_{k+2N}=\underline{M}^N\mathbf{a}_k , \tag{36}$$

where we have dropped the subscript c on $\underline{M}_c$ from here on. Since $\underline{H}$ is Hermitian, the norm $|\mathbf{a}|^2$ is preserved by Eq. (1). It will also be preserved by the result, Eq. (36), if $\underline{M}$ is unitary. In order to enforce unitarity, we simply divide $\underline{M}$ by the quantity $(|M_{11}|^2+|M_{12}|^2)^{1/2}$. The WKB derivation leading to Eq. (30) is, in any event, not correct to $O(\varepsilon^2)$ which is the size of the difference between $\|\underline{M}\|$ and unity. By diagonalizing $\underline{M}$ we can find $\underline{M}^N$ quite easily. Given that $a_k^{+}=1$ and $a_k^{-}=0$, the result then becomes

$$a^-_{k+2N} = \frac{-(m_2/m_1)e^{-i[\gamma_2+2\pi(A/\omega)N]}}{[\sin^2\gamma_1+(m_2/m_1)^2]^{1/2}} \sin\left[\tan^{-1}\left[\frac{[\sin^2\gamma_1+(m_2/m_1)^2]^{1/2}}{\cos\gamma_1}\right]N\right] , \quad (37)$$

where $m_i = |M_{1i}|$ and $\gamma_i = \phi(M_{1i})$. From Eq. (30) we know that $\phi(M_{11}) \simeq 2\theta_0$; therefore, the condition for a resonance is defined by $\sin(2\theta_0)=0$ or $2\theta_0 - l\pi = 0$ for some integer l. If Eq. (1) is used to model an atomic quantum-mechanical system, then l is identified with the number of quanta separating the resonant energy levels.

From Eq. (37), we see that, on a resonance, the transition rate is proportional to $\tan^{-1}(m_2/m_1) \sim m_2/m_1$. From Eq. (30), we have

$$m_1 \simeq 1 ,$$
$$m_2 \simeq 2|\varepsilon \sin\theta_0| . \quad (38)$$

On a resonance $\theta_0 = (\pi/2)l$. Therefore, for l even, the resonance value of m_2 will be zero to lowest order. This means that the transition rate will also be zero. We note that only *odd* resonances will be observed. In previous work on this problem, this condition seems to have been proven by appealing to quantum-mechanical perturbation theory (e.g., see Delone and Krainov[11] and Shirley[5]).

With the identification $P \equiv |a^-_{k+2N}|^2$ and the replacement $N \to \omega t/2\pi$, we obtain, from Eq. (37),

$$P \simeq \frac{(2|\varepsilon|)^2}{\sin^2(2\theta_0)+(2|\varepsilon|)^2} \times \sin^2\left[\frac{\omega t}{2\pi}[\sin^2(2\theta_0)+(2|\varepsilon|)^2]^{1/2}\right] . \quad (39)$$

For $\omega t \sim \varepsilon^{-1}$, P assumes a resonance structure. Near a resonance, we have $\sin(2\theta_0) \simeq 2[\theta_0-(\pi/2)l]$. Defining $u=(2\omega/\pi)|\varepsilon|$, and $\omega_{\rm res}=(2\omega/\pi)\theta_0$, the result is

$$P \simeq \frac{u^2}{[\omega_{\rm res}-(2p+1)\omega]^2+u^2} \times \sin^2(\tfrac{1}{2}\{[\omega_{\rm res}-(2p+1)\omega]^2+u^2\}^{1/2}t) , \quad (40)$$

where $l=2p+1$, and p is an integer. The resonance condition is

$$\omega_{\rm res}-(2p+1)\omega=0 . \quad (41)$$

In the small-field limit (F/Δ small), we have, from Eqs. (23) and (28),

$$u \sim \frac{2\omega}{\pi}\left[\frac{eF}{4\Delta}\right]^{2\Delta/\omega} ,$$
$$\omega_{\rm res} \sim 2\Delta\left[1+\frac{1}{4}\left[\frac{F}{\Delta}\right]^2\right] . \quad (42)$$

We show in the Appendix that these results are equivalent to those obtained perturbatively by Shirley. For the case of large fields (F/Δ large), we have

$$u \sim \frac{2\omega}{\pi}\exp\left[-\frac{\pi}{2}\frac{\Delta}{\omega}\frac{\Delta}{F}\right] ,$$
$$\omega_{\rm res} \sim \frac{4F}{\pi} . \quad (43)$$

Equations (40)–(43) constitute the major result of our theory when $\dot{F}=0$.

For all the ranges of F/Δ that are considered here, u is much less than ω. From Eq. (40), we see that this means that the resonance widths for P are very small. (This property is not surprising if we consider the close analogy of the equation of motion, Eq. (4), to the Mathieu equation, $d^2y/d\tau^2+[a-2q\cos(2\tau)]y=0$. A well-known property of this equation[14] is the presence of unstable solutions near integer values of $a^{1/2}$. The successively higher-order cusping of the stability boundaries with increasing $a^{1/2}$ is analogous to the narrowness of the resonances contained in Eq. (40).) Furthermore, to the extent that our model is applicable, we see, from both Eqs. (42) and (43), that variations of $\omega_{\rm res}$ that are large compared with u will occur in practice because of intensity variations associated with the finite duration of the applied field. These considerations place very stringent criteria on the required constancy of the intensity, if any particular resonance is to be maintained for as long as one Rabi period $\simeq u^{-1}$. In Sec. III we consider some implications of this limitation.

III. EFFECTS OF FINITE PULSE LENGTH

In this section the effects of the finite length of the laser pulse are considered. Because $\omega_{\rm res}$ varies with intensity, the condition, Eq. (41), for resonance of a given order p is satisfied precisely at only a single time t_p. The system remains effectively on resonance up to times t^*, such that

$$\int_{t_p}^{t_p+t^*} dt\,[\omega_{\rm res}-(2p+1)\omega] < 1 .$$

During such intervals, coherent additions to the amplitude of an initially empty state a^- will occur twice each period of the applied field and will accumulate to a size

$$a^- \simeq (\omega t^*)\varepsilon \exp\left[i\omega\int_{t_p}^{t} dt\,\lambda^-\right] ,$$

where $\omega t^* \gg 1$, if the amplitude varies sufficiently slowly. Outside of such resonant intervals, the contributions to a^- will be $\simeq \varepsilon$, i.e., negligible. Any uncertainty in the temporal behavior of the applied field will introduce corresponding uncertainties in t_p and λ^-. If the integral $\omega\int_{t_p}^{t} dt\,\lambda^-$ accumulates an uncertainty of $O(1)$, then the contribution to a^- from resonances of adjacent orders will be incoherent. This is the short-autocorrelation-time approximation, considered below. If the autocorrelation time of the phase integral approaches the interval between resonances, then these results will be modified. Defining $\Delta|a^-|^2$ as the change in $|a^-|^2$ which occurs upon passing through one resonance, and t_{step} as the time interval during which the system is between adjacent resonances, then we may further define an average rate of excitation R as

$$R=\frac{\Delta|a^-|^2}{t_{\text{step}}} . \tag{44}$$

We now develop quantitative expressions for these quantities.

To find $\Delta|a^-|^2$, we go back to the transfer matrix, Eq. (30). The matrix $\underline{M}$ has the form

$$\underline{M}=\begin{bmatrix} m_{11} & m_{12} \\ -m_{12}^* & m_{11}^* \end{bmatrix} .$$

Assuming $|a^+|\approx 1$, then over each period of the applied field, a^- will suffer a small "kick" of size equal to $-m_{12}^*$. From Eq. (30), $m_{12}^*=i2|\varepsilon|\sin\theta_0 e^{-i\theta_0}$. If we assume variation of the form

$$\theta_0=(2p+1)\frac{\pi}{2}+\alpha j , \tag{45a}$$

where j is an integer such that

$$t=t_p+\left[\frac{2\pi}{\omega}\right]j , \tag{45b}$$

and $\alpha\equiv(2\pi/\omega)(d\theta_0/dF)(dF/dt)$ is the change in θ_0 per period of the applied field, then

$$m_{11}(j)=-\exp(+i2\alpha j) ,$$

$$m_{12}(j)=-i2|\varepsilon|\exp(i\alpha j) .$$

The contributions to a^- which occur from $j=-M$ to $j=M$ are now considered. Taking $a^+(-M)=1$ and $a^-(-M)=0$, we have

$$a^-(M)=\sum_{j=-M}^{M}\prod_{q=j+1}^{M}\prod_{q'=-M}^{j-1} m_{11}^*(q)m_{12}(j)m_{11}(q') , \tag{46}$$

which yields, for the magnitude $|a^-(M)|$, the result

$$|a^-(M)|=|2\varepsilon|\left|\sum_{j=-M}^{M-1}\exp[i2\alpha(j-\tfrac{1}{4})^2]\right| . \tag{47}$$

In the case of interest, that of a slowly varying envelope, we have $\alpha\ll 1$ (corresponding to $\omega t^*\gg 1$). It is evident that only those terms for which $j\sim\alpha^{-1/2}$ contribute in phase to $|a^-|$. As M becomes large, the sum can be replaced by an integral, with one consequence, that the contributions of all resonances other than that centered at $j=0$ are eliminated. These operations give the result

$$|a^-(M)|\simeq|2\varepsilon|\left[\frac{\pi}{2\alpha}\right]^{1/2} , \tag{48}$$

or

$$\Delta|a^-|^2\simeq\left[\frac{2\pi}{\alpha}\right]|\varepsilon|^2 .$$

To find t_{step}, the interval between adjacent resonances, we consider the resonance condition, Eq. (45), with $j=0$. Changing p by unity yields the condition for an adjacent resonance. Therefore

$$t_{\text{step}}=\pi\left[\frac{d\theta_0}{dt}\right]^{-1} ,$$

$$=\frac{2\pi^2}{\alpha\omega} ,$$

where the second form follows upon using Eq. (45). With this result, we obtain the average rate of excitation,

$$R=\frac{\pi u^2}{4\omega} , \tag{49}$$

and note that R depends only on the instantaneous field intensity, and not on its time derivative.

IV. COMPETITION BETWEEN DIRECT N-PHOTON EXCITATION AND IONIZATION

In a real atomic system, under experimental conditions, the possibility of coupling two levels needs to be considered in the light of competing processes. Here we consider one such process, that of multiphoton ionization. Keldysh[7] has shown that ionization from the upper level of a transition pair is much more likely than from the lower energy (more deeply bound) state. We consider the case of an electromagnetic pulse with temporal intensity variation $I(t)$ incident upon an atomic system with dipole moment $d=qa$ for the selected bound-bound transition. Here q is the electron charge and a is the effective size of the dipole. The coupling frequency $F=E_0d/\hbar$, where E_0 is the amplitude of the applied electric field. In experimental units,

$$\frac{F}{\Delta}=\frac{2qaE_0}{W}=0.55\frac{I_{16}^{1/2}a_{\text{Å}}}{W_{100}} , \tag{50}$$

where W_{100} is the transition energy expressed in units of 100 eV, I_{16} is the applied electromagnetic (em) field intensity in units of 10^{16} W/cm^2, and $a_{\text{Å}}$ is the dipole size in angstroms. The asymptotic parameter,

$$\frac{\omega}{\Delta}=2.48\times10^{-2}\lambda_{\mu\text{m}}^{-1}W_{100}^{-1} , \tag{51}$$

where $\lambda_{\mu\text{m}}$ is the wavelength of the applied field in micrometers. Two additional characteristic parameters, the adiabaticity parameter,

$$\gamma=\frac{\omega(2m\chi)^{1/2}}{qE_0}=0.23\frac{\chi_{100}^{1/2}}{\lambda_{\mu\text{m}}I_{16}^{1/2}} , \tag{52}$$

and $n_i\equiv\chi/\hbar\omega=1.24\times10^{-2}\chi_{100}\lambda_{\mu\text{m}}$, the minimum number of photons needed for ionization, enter the computation of the multiphoton-ionization rate. Here m is the electron mass and χ_{100} is the ionization potential, again in units of 100 eV. If $\gamma\ll 1$, then the tunneling (adiabatic) limit of Keldysh's result applies whereas, if $\gamma\gg 1$, the "multiphoton" limit applies.

For illustration, we consider the particular transition $4s^2 4p^6 \rightarrow 4s4p^6 5p$ between the ground state and a core excited state in Kr-like Cd^{12+}, suggested by Clark *et al.*[4] as a candidate for selective multiphoton excitation and possible consequent lasing action. The pumping is to occur through the application of a short pulse (1 psec) of high-intensity radiation ($\simeq 10^{16}$ W/cm^2) from a KrF laser ($\lambda = 0.248$ μm). They compute the transition energy $W_{100} = 1.9$, and the ionization potential from the upper level $\mathcal{X}_{100} = 1.6$, and estimate $a_{\text{Å}} = 1$. Keldysh's result for the ionization rate in the limit $\gamma \ll 1$, applicable here, is

$$\omega_{\text{ion}} = \omega 2^{-5/4} (3\pi)^{1/2} n_i^{1/2} \gamma^{-1/2} \exp[-\tfrac{4}{3}\gamma n_i (1 - \tfrac{1}{10}\gamma^2)] \ . \tag{53}$$

A comparative plot of ω_{ion} and the mean excitation rate R, Eq. (49), is shown versus intensity in Fig. 3. Clearly, the ionization rate is always much larger than the excitation rate, a result which casts doubt upon the prospects for efficient excitation in this case.

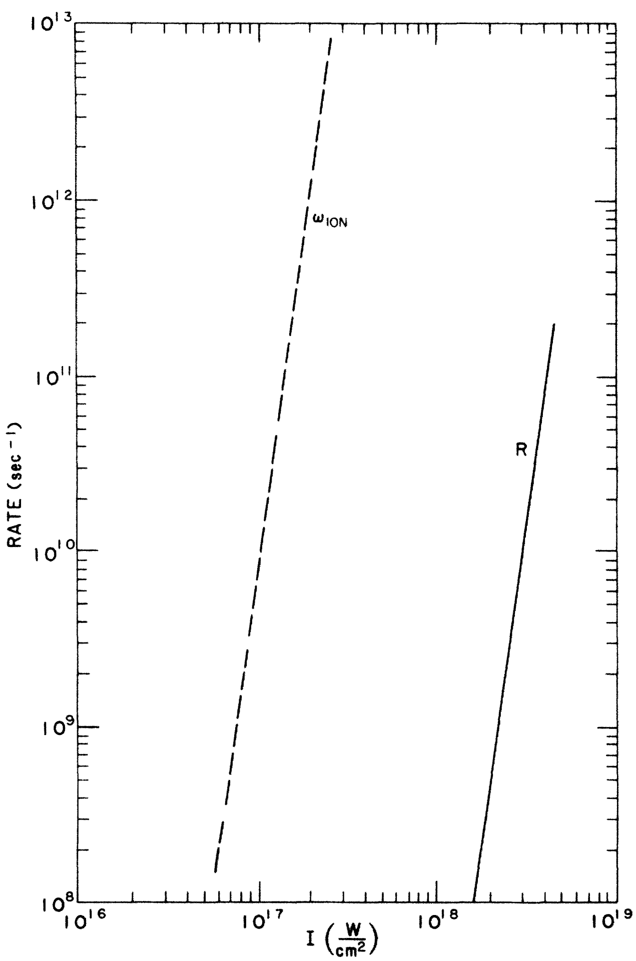

FIG. 3. Solid curve shows the excitation rate R vs intensity. The dashed curve shows the (Keldysh) ionization rate ω_{ion} vs intensity.

V. CONCLUSIONS

We have analyzed the evolution of a two-level system in the presence of an externally imposed finite, but slowly varying, potential. We have given general expressions for the transition rates and resonance conditions when the potential is strictly periodic. Simple limiting forms are given, which both reproduce the results of perturbation theory in the weak-external-field limit, Eq. (42), and which apply in the strong-field case, Eq. (43), where perturbation theory is no longer valid. Finite transition probabilities are found when conditions for high-order (multiphoton) resonances are satisfied. Such resonances are extremely narrow, however, and have relatively much larger nonlinear shifts in position as the external field amplitude is varied. These characteristics motivated the further calculation of an average transition rate which includes the decorrelating effects, on these resonances, of external uncertainties. For strong decorrelation, i.e., when adjacent resonances are decorrelated, Eq. (49) is obtained for the mean rate. This rate was evaluated for parameters relevant to the case of multiphoton excitation in Cd^{12+}, and was compared to the rate of multiphoton ionization from the excited state, which was found to dominate at all intensities of interest.

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APPENDIX: COMPARISON TO SHIRLEY'S RESULTS

By means of Floquet theory, Shirley[5] has perturbatively obtained results for Eq. (1) in the limit $F/\Delta \ll 1$. The same results have also been found by Aravind and Hirsfelder.[15] Near an odd resonance, Shirley obtained Eq. (40) with u and ω_{res} replaced by

$$u_{\text{Sh}} = \frac{(F/2)^{2p+1}}{2^{2p-1}(p!)^2 \omega^{2p}} \tag{54}$$

and

$$\omega_{\text{Sh}} = E_2 - E_1 + \left[\frac{2p+1}{p(p+1)} \right] \frac{F/2}{\omega} \ ,$$

respectively. In the limit $F/\Delta \ll 1$, we see that $\omega_{\text{res}} \simeq 2\Delta$. Therefore, when near the $2p+1$ resonance, 2Δ can be replaced by $(2p+1)\omega$ in our expressions for u and ω_{res}. For large p, which corresponds to the many-photon limit (large Δ/ω), ω_{Sh} then agrees with our ω_{res}.

To show the correspondence between u and u_{Sh}, we first use Stirling's formula for large p,

$$p!=\Gamma(p+1)=(2\pi)^{1/2}(p+1)^{p+(1/2)}e^{-(p+1)}\times\left[1+O\left[\frac{1}{p}\right]\right],$$

in Eq. (54). We now have

$$u_{\rm Sh}=\left[\frac{(F/2)^{2p+1}}{2^{2p-1}(2\pi)(p+1)^{2p+1}e^{-(2p+1)}\omega^{2p}}\right]\times\left[1+O\left[\frac{1}{p}\right]\right],$$

$$=\left[\frac{2(F/2)^{2p+1}}{\pi(2p+2)^{2p+1}e^{-(2p+2)}\omega^{2p}}\right]\left[1+O\left[\frac{1}{p}\right]\right].$$

Also, for large p,

$$(2p+2)^{2p+1}=(2p+1)^{2p+1}\left[1+\frac{1}{2p+1}\right]^{2p+1}$$

$$=e(2p+1)^{2p+1}\left[1+O\left[\frac{1}{p}\right]\right].$$

Therefore

$$u_{\rm Sh}=\left[\frac{2(F/2)^{2p+1}}{\pi(2p+1)^{2p+1}e^{-(2p+1)}\omega^{2p}}\right]\left[1+O\left[\frac{1}{p}\right]\right].$$

The replacement $2p+1\rightarrow 2\Delta/\omega$ is valid near a resonance for $F/\Delta \ll 1$. If this replacement is made in the above expression, it becomes identical to our form for u, Eq. (42), to order ω/Δ.

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