Semiclassical analysis of long-wavelength multiphoton processes: The periodically driven harmonic oscillator

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The problem of multiphoton processes for intense, long-wavelength irradiation of atomic and molecular electrons is presented. The recently developed method of quasiadiabatic time evolution is used to obtain a nonperturbative analysis. When applied to the standard vector potential coupling, an exact auxiliary equation is obtained that is in the electric dipole coupling form. This is achieved through application of the Goeppert-Mayer gauge. While the analysis to this point is general and aimed at microwave irradiation of Rydberg atoms, a Floquet analysis of the auxiliary equation is presented for the special case of the periodically driven harmonic oscillator. Closed form expressions for a complete set of Floquet states are obtained. These are used to demonstrate that for the oscillator case there are no multiphoton resonances.

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I. INTRODUCTION

For a number of reasons, there has been a long standing interest in multiphoton processes involving atomic and molecular electrons. In this paper, the focus is on intense beams of long wavelength photons, such as microwave irradiation of Rydberg atoms. Experiments have been done in this context and a number of theoretical treatments have been given, some of which are directly related to questions of quantum chaos [1]. The goal of the present paper is to present a nonperturbative method for such problems when the radiation field is treated semiclassically and the wavelength of the radiation is much larger than the spatial extent of the electron states.

The approach used here is based on the quasiadiabatic method, recently developed for a different context [2]. In this approach, the original dynamics is replaced by an equivalent, auxiliary dynamics. The auxiliary dynamics may be solved using the Floquet method. This amounts to a nonperturbative solution to the original problem. In the present context, the transition from the original problem to the auxiliary problem is effectuated by the Goeppert-Mayer gauge transformation that makes use of the electric dipole approximation [3].

In order to make the method used here as clear as possible, the periodically driven harmonic oscillator is the physical system to be studied. In a sequel, microwave irradiation of Rydberg atoms will be the focus. However, a general setting and the problem for the Rydberg case will be setup in Sec. III. In Sec. IV, restriction to the oscillator case will be made. For the oscillator, explicit analytic expressions for the Floquet states are obtained. It is observed, and emphasized, that for the periodically driven harmonic oscillator, there are no multiphoton resonances. In demonstrating this result, the Keldysh approach [4], as presented by Gribakin and Kuchiev [5], is applied and found not to be valid for the oscillator case.

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The paper is organized as follows. In Sec. II, a review of the quasiadiabatic approach is given. In Sec. III, this method is applied to the general problem of the vector potential coupling of radiation to atomic and molecular electrons with the Rydberg case given explicitly. It is shown that the Goeppert-Mayer gauge provides an exact conversion to an auxiliary equation of the electric dipole coupling form when the radiation is of long wavelength, regardless of intensity. In Sec. IV, the time evolution operator for the auxiliary equation is constructed in closed form for the periodically driven harmonic oscillator. This permits determination of the eigenstates of the associated monodromy operator in closed form. These, in turn, yield the Floquet states for the periodically driven harmonic oscillator in closed form. In Sec. V, several properties of these Floquet states are elucidated. Classical correlates are discussed as well as the observation that there are no multiphoton resonances. In Sec. VI, the paper is concluded with an analysis of the Gribakin and Kuchiev treatment of the Keldysh approach applied to the periodically driven harmonic oscillator.

II. REVIEW OF THE QUASIADIABATIC TIME EVOLUTION METHOD

Quasiadiabatic time evolution is a method for Hamiltonians containing a time dependent contribution [2]. In the extreme adiabatic limit, the description is very closely related to the treatment of Berry's adiabatic geometrical phase [6]. However, the results obtained are also valid when the time dependence is not truly adiabatic, i.e., more rapid changes are allowed, hence the qualifier: *quasiadiabatic*. This was demonstrated by an analysis of the Landau-Zener dynamics at an avoided level crossing in which the passage through the avoided level crossing varied in rate by over six orders of magnitude, starting from the adiabatic regime and ending with the fast passage regime [2].

Let $\vec{R}(t)$ denote a vector of parameters that depend on time and let the Hamiltonian $H=H(\vec{R}(t))$ be given. Write $\vec{R}'=\vec{R}(t)$ where the prime signifies t dependence. At every instant of time, solve the adiabatic equation

$$H(\vec{R}')|n'\rangle = E_n(\vec{R}')|n'\rangle \tag{1}$$

for the instantaneous eigenenergies $E_n(\vec{R}')$ and the instantaneous orthonormal eigenstates, $|n'\rangle$. The prime on n in these kets signifies that the ket is really time dependent, depending on the instant of time when it was determined, and does not mean a value for n different from the n that is the subscript of the eigenenergy. This point must be kept in mind throughout the discussion. Generally, the $|n'\rangle$'s are necessarily complex because of electromagnetic coupling terms such as $\vec{A}(t)\cdot\vec{p}$ in H, but there are one parameter cases in which the $|n'\rangle$'s can be made to be real. This dichotomy has relevance for Berry's phase.

The solution to the time dependent problem, $|\psi(t)\rangle$, at time t can be expanded in terms of the adiabatic eigenstates

$$|\psi(t)\rangle = \sum_n a_n(t)|n'\rangle.$$

Noting the time dependence implicit in the $|n'\rangle$'s, it is straightforward to derive auxiliary equations for the $a_n(t)$'s given by [2]

$$i\hbar\frac{d}{dt}a_m(t) = E_m(\vec{R}')a_m(t) - i\hbar\sum_n a_n(t) \times\left\langle m'\left|\frac{\partial}{\partial\vec{R}'}\right|n'\right\rangle\cdot\frac{d\vec{R}'}{dt}. \tag{2}$$

It also follows exactly that

$$\frac{\partial}{\partial\vec{R}'}E_n(\vec{R}') = \left\langle n'\left|\frac{\partial H(\vec{R}')}{\partial\vec{R}'}\right|n'\right\rangle \tag{3}$$

and for $n\neq m$,

$$\left\langle m'\left|\frac{\partial}{\partial\vec{R}'}\right|n'\right\rangle = \frac{\left\langle m'\left|\frac{\partial H(\vec{R}')}{\partial\vec{R}'}\right|n'\right\rangle}{E_n(\vec{R}')-E_m(\vec{R}')}. \tag{4}$$

Equation (3) is of the form of the Hellmann-Feynman theorem [7]. Equation (4) converts Eq. (2) into

$$\frac{d}{dt}a_m(t) = -\frac{i}{\hbar}E_m(\vec{R}')a_m(t) - \left\langle m'\left|\frac{\partial}{\partial\vec{R}'}\right|m'\right\rangle\cdot\frac{d\vec{R}'}{dt}a_m(t) - \sum_{n\neq m}a_n(t)\frac{\left\langle m'\left|\frac{\partial H(\vec{R}')}{\partial\vec{R}'}\right|n'\right\rangle\cdot\frac{d\vec{R}'}{dt}}{E_n(\vec{R}')-E_m(\vec{R}')}. \tag{5}$$

This is the fundamental equation for quasiadiabatic time evolution [2]. The last term on the right-hand side has the possibility to become large at avoided level crossings when the denominator energy difference becomes small. The second term on the right-hand side gives rise to Berry's phase and the coefficient, $\langle m'|\partial/\partial\vec{R}'|m'\rangle$, is purely imaginary if and only if the $|m'\rangle$'s are complex. For real $|m'\rangle$'s, this term vanishes. The one-parameter case does not yield a Berry's phase as was shown in Ref. [2].

The implementation of this approach requires that the $|n'\rangle$'s and $E_n(\vec{R}')$'s be found for each time t. In a practical application, this can be done numerically at the outset for a discrete set of times, and then Eq. (5) can be used to find the quasiadiabatic coefficients, the $a_m(t)$'s.

III. VECTOR POTENTIAL COUPLING AND THE AUXILIARY EQUATION

For an atomic or molecular electron in an intense semiclassical microwave radiation field, the Hamiltonian is given by

$$H = \frac{1}{2m}\left(\vec{p}+\frac{e}{c}\vec{A}(t)\right)^2 - e\phi = \frac{p^2}{2m} - e\phi + \frac{e}{mc}\vec{A}\cdot\vec{p} + \frac{e^2}{2mc^2}\vec{A}\cdot\vec{A} \tag{6}$$

in which we take $e>0$ (the electron charge is $-e$) and use the Coulomb gauge so that $\vec{\nabla}\cdot\vec{A}=0$. The scalar potential ϕ for a Rydberg atom is given by

$$\phi(r) = \frac{Ze}{r}.$$

The microwave field is represented by a plane wave

$$\vec{A} = A_0\hat{\varepsilon}\sin(\omega t) \tag{7}$$

in which $\hat{\varepsilon}$ is the polarization unit vector and A_0 is the amplitude. The spatial dependence is replaced by 1 in the dipole approximation. If 10 GHz light is contemplated, then $\omega = 2\pi\times10^{10}\ \mathrm{s}^{-1}$ and $\lambda = 3$ cm. This is much larger than the size of a hydrogen atom even with principal quantum number $n=100$ (0.529×10^{-4} cm). Thus, even though the laser does not produce an infinite plane wave, the atomic electron sees a plane wave, constant in space and variable in time, for all practical purposes.

Denote by H_0 the noninteracting part of the Hamiltonian

$$H_0 = \frac{p^2}{2m} - e\phi. \tag{8}$$

Let the stationary solutions to this time independent problem be given by

$$H_0|\phi_n\rangle = E_n|\phi_n\rangle. \tag{9}$$

The quasiadiabatic states are solutions for the time dependent Hamiltonian at an instant of time

$$H'|\psi_n'\rangle = E_n'|\psi_n'\rangle,$$

where H' is the Hamiltonian in Eq. (6) and the prime has been included to emphasize its time dependence. The

Goeppert-Mayer gauge [3] permits an exact solution to this equation in terms of the solutions to Eq. (9). This is proved as follows:

Let

$$|\psi_n'\rangle = \exp\left[-i\frac{e}{\hbar c}\vec{r}\cdot\vec{A}'\right]|\phi_n\rangle. \tag{10}$$

Clearly

$$\vec{p}|\psi_n'\rangle = \exp\left[-i\frac{e}{\hbar c}\vec{r}\cdot\vec{A}'\right](\vec{p}|\phi_n\rangle) - \frac{e}{c}\vec{A}'|\psi_n'\rangle.$$

Therefore

$$\left(\vec{p}+\frac{e}{c}\vec{A}'\right)|\psi_n'\rangle = \exp\left[-i\frac{e}{\hbar c}\vec{r}\cdot\vec{A}'\right](\vec{p}|\phi_n\rangle) \tag{11}$$

and

$$H'|\psi_n'\rangle = \exp\left[-i\frac{e}{\hbar c}\vec{r}\cdot\vec{A}'\right](H_0|\phi_n\rangle) = E_n|\psi_n'\rangle, \tag{12}$$

which implies

$$E_n' = E_n.$$

In this special case of quasiadiabatic time evolution, the adiabatic eigenenergies are constant in time and are equal to the E_n's of Eq. (9). Moreover, the adiabatic states are precisely the $|\psi_n'\rangle$'s of Eq. (10).

The general solution for the Hamiltonian in Eq. (6) can be expanded in terms of these time dependent adiabatic states

$$|\psi(t)\rangle = \sum_n a_n(t)|\psi_n'\rangle. \tag{13}$$

Plugging this into Schrödinger's equation and using Eq. (12) yields

$$i\hbar\sum_n\left[\left(\frac{d}{dt}a_n\right)|\psi_n'\rangle + a_n\frac{d}{dt}|\psi_n'\rangle\right] = \sum_n a_n E_n|\psi_n'\rangle.$$

Therefore

$$\frac{d}{dt}a_m = -i\frac{E_m}{\hbar}a_m - \sum_n\left\langle\psi_m'\left|\frac{d}{dt}\right|\psi_n'\right\rangle a_n. \tag{14}$$

From Eq. (12) we also get

$$\left(\frac{d}{dt}H\right)|\psi_n'\rangle + H\frac{d}{dt}|\psi_n'\rangle = E_n\frac{d}{dt}|\psi_n'\rangle.$$

Therefore,

$$E_n\left\langle\psi_m'\left|\frac{d}{dt}\right|\psi_n'\right\rangle = \left\langle\psi_m'\left|\left(\frac{d}{dt}H\right)\right|\psi_n'\right\rangle + E_m\left\langle\psi_m'\left|\frac{d}{dt}\right|\psi_n'\right\rangle. \tag{15}$$

When $m=n$, it follows that

$$\left\langle\psi_n'\left|\left(\frac{d}{dt}H\right)\right|\psi_n'\right\rangle = 0$$

must hold. This is easily proved as follows:

$$\frac{d}{dt}H = \frac{e}{mc}\left(\frac{d}{dt}\vec{A}\right)\cdot\left(\vec{p}+\frac{e}{c}\vec{A}\right)$$

which implies, using Eq. (11),

$$\left(\frac{d}{dt}H\right)|\psi_n'\rangle = \left[\frac{e}{mc}\left(\frac{d}{dt}\vec{A}\right)\cdot\vec{p}\,\middle|\,\phi_n\right\rangle\exp\left[-i\frac{e}{\hbar c}\vec{r}\cdot\vec{A}\right]. \tag{16}$$

Therefore,

$$\left\langle\psi_n'\left|\left(\frac{d}{dt}H\right)\right|\psi_n'\right\rangle = \frac{e}{mc}\left(\frac{d}{dt}\vec{A}\right)\cdot\langle\phi_n|\vec{p}|\phi_n\rangle = 0. \tag{17}$$

The Goeppert-Mayer gauge factors have cancelled out and the last matrix element vanishes because the $|\phi_n\rangle$'s are parity eigenstates in the case of Rydberg atoms. When $m=n$, Eq. (15) implies

$$\left\langle\psi_m'\left|\frac{d}{dt}\right|\psi_n'\right\rangle = \frac{\left\langle\psi_m'\left|\left(\frac{d}{dt}H\right)\right|\psi_n'\right\rangle}{E_n-E_m} = \frac{e}{mc}\left(\frac{d}{dt}\vec{A}\right)\cdot\frac{\langle\phi_m|\vec{p}|\phi_n\rangle}{E_n-E_m}$$

because of Eq. (16) and the analogue to Eq. (17) for $m\neq n$. By a standard textbook argument for this case

$$\langle\phi_m|\vec{p}|\phi_n\rangle = im\frac{E_m-E_n}{\hbar}\langle\phi_m|\vec{r}|\phi_n\rangle.$$

Putting all of this into Eq. (14) yields the exact quasiadiabatic time evolution auxiliary equation

$$\frac{d}{dt}a_m = -i\omega_m a_m + i\sum_n\frac{e}{\hbar c}\left(\frac{d}{dt}\vec{A}\right)\cdot\langle\phi_m|\vec{r}|\phi_n\rangle a_n, \tag{18}$$

where $\omega_m = E_m/\hbar$.

Two observations about Eq. (18) are in order. The energy difference denominator, so important for understanding the dynamics of avoided level crossings, has cancelled out in this special case of vector potential coupling. It was not a function of time in any event. In addition, the electric field is related to the vector potential by

$$\vec{E} = -\frac{1}{c}\frac{\partial}{\partial t}\vec{A} = -\frac{A_0\omega}{c}\hat{\varepsilon}\cos(\omega t) \tag{19}$$

when Eq. (7) is used. It is convenient to write $A_0 = cE_0/\omega$ so that Eq. (18) takes the form

$$\frac{d}{dt}a_m = -i\omega_m a_m + i\sum_n\frac{eE_0}{\hbar}\cos(\omega t)\hat{\varepsilon}\cdot\langle\phi_m|\vec{r}|\phi_n\rangle a_n. \tag{20}$$

This auxiliary equation is precisely what one would obtain if one were analyzing the problem of electric dipole coupling with the Hamiltonian

$$H = \frac{p^2}{2m} - e\phi + e\vec{r}\cdot\vec{E}, \tag{21}$$

where

$$\vec{E} = -E_0\hat{\varepsilon}\cos(\omega t)$$

and the general solution $|\psi(t)\rangle$ is expanded in terms of the eigenstates of H_0, as given in Eqs. (8)–(9). That is

$$|\psi(t)\rangle = \sum_n a_n(t)|\phi_n\rangle$$

leads to

$$i\hbar \frac{d}{dt} a_m = E_m a_m - \sum_n eE_0 \cos(\omega t)\hat{\varepsilon}\cdot\langle\phi_m|\vec{r}|\phi_n\rangle a_n . \tag{22}$$

This is identical with Eq. (20). However, in Eq. (20), the a_m's are coefficients for the quasiadiabatic time evolution given in Eq. (13), which is exact and must be solved to all orders in E_0. Equation (22), by contrast, is the ordinary equation for the standard Schrödinger solution to the time dependent problem with the Hamiltonian in Eq. (21). Moreover, the $e\vec{r}\cdot\vec{E}$ coupling is an approximation and, thus Eq. (22) should be solved only to leading order in E_0. This difference is fundamental and must be emphasized to avoid confusion.

In ordinary time dependent perturbation theory, the relationship between the Hamiltonians in Eqs. (6) and (21) has been the cause of many papers. As is well known, to first order in perturbation theory, both Hamiltonians give the same results (the $\vec{A}\cdot\vec{A}$ term is ignored to this lowest order). The difficulty that arises when higher-order perturbations are considered stems from the fact that the two Hamiltonians do not in fact act on the same set of unperturbed functions. The basis states for the time independent parts of these Hamiltonians, in terms of which the perturbation expansions are rendered, are not identical but are related by the Goeppert-Mayer gauge transformation [3]. Only when this gauge is properly incorporated do the two approaches yield identical results to all orders in perturbation theory. In the present context of quasiadiabatic time evolution, the auxiliary equations in Eq. (22) are to be solved to all orders in E_0 and the resulting $a_m(t)$'s are then used in Eq. (13) along with Eq. (10) to generate a complete solution.

IV. THE PERIODICALLY DRIVEN HARMONIC OSCILLATOR

For the periodically driven harmonic oscillator, the Hamiltonians in Eqs. (6) and (21) must be changed so that they include explicitly the potential energy for a harmonic oscillator

$$\phi(q) = \frac{1}{2} m\omega_0^2 q^2,$$

where q is the generalized coordinate and the oscillator is considered one-dimensional. In this one dimensional case, the polarization of the radiation is taken to be along the coordinate q. Thus, Eq. (6) becomes

$$H = \frac{1}{2m}\left(p + \frac{e}{c}A\right)^2 + \frac{1}{2} m\omega_0^2 q^2 \tag{23}$$

and Eq. (21) becomes

$$H = \hbar\omega_0\left(a^\dagger a + \frac{1}{2}\right) - eE_0\sqrt{\frac{\hbar}{2m\omega_0}}\cos(\omega t)(a + a^\dagger), \tag{24}$$

where creation and annihilation operators have been introduced. Equation (24) is the Hamiltonian for the auxiliary equations. These take the form

$$\frac{d}{dt} a_m = -i\omega_m a_m + i\sum_n \frac{eE_0}{\hbar}\cos(\omega t)\langle\phi_m|q|\phi_n\rangle a_n ,$$

where the $|\phi_n\rangle$'s satisfy

$$H_0|\phi_n\rangle = E_n|\phi_n\rangle,$$

$$E_n = \left(n + \frac{1}{2}\right)\hbar\omega_0$$

in which H_0 is given by

$$H_0 = \hbar\omega_0\left(a^\dagger a + \frac{1}{2}\right).$$

From here on, the standard notation for the eigenstates of H_0 will be used, i.e.,

$$|\phi_n\rangle = |n\rangle.$$

Using time ordered operator algebra techniques [8], it is possible to express the evolution operator $U(t)$ for the time dependent Hamiltonian in Eq. (24) by

$$\begin{aligned} U(t) &= \underset{\leftarrow}{T}\exp\left[-\frac{i}{\hbar}\int_0^t ds H(s)\right] \\ &= \exp\left[-i\omega_0 t\left(a^\dagger a + \frac{1}{2}\right)\right]\exp[-r_0^2 J(t)] \\ &\quad\times\exp[-ir_0 I(t)a^\dagger]\exp[-ir_0 I^*(t)a] \\ &= \exp\left[-i\omega_0 t\left(a^\dagger a + \frac{1}{2}\right)\right]\exp\left[-r_0^2\left(J(t) - \frac{1}{2}|I(t)|^2\right)\right] \\ &\quad\times\exp[-ir_0(I(t)a^\dagger + I^*(t)a)], \end{aligned} \tag{25}$$

where

$$r_0 = \frac{eE_0}{\hbar}\sqrt{\frac{\hbar}{2m\omega_0}}, \tag{26}$$

which has the units of a rate, and

$$I(t)=i\{e^{i\omega_0 t}\}\frac{\omega_0[\cos(\omega t)-\cos(\omega_0 t)]-i[\omega\sin(\omega t)-\omega_0\sin(\omega_0 t)]}{\omega_0^2-\omega^2},$$

$$J(t)=\frac{1}{4}\frac{1}{\omega^2-\omega_0^2}\left\{1-\cos 2(\omega t)+i\frac{\omega_0}{\omega}\sin(2\omega t)+i2\omega_0 t+4\frac{\omega_0}{\omega^2-\omega_0^2}[\omega_0(1-\cos(\omega t))-i\omega\sin\omega t]\right\}. \tag{27}$$

Moreover, it follows that

$$|I(t)|^2=J(t)+J^*(t). \tag{28}$$

In the first equality of Eq. (25), the time ordered exponential appears. The second and third equalities represent equivalent factorizations of the time ordered exponential in terms of ordinary exponential factors.

Construction of the Floquet states is based upon the eigenstates of the monodromy operator $U(T)$ where $T=2\pi/\omega$ (see the Appendix). Using Eqs. (25)–(28) yields

$$U(T)=\exp\left[-i\omega_0 T\left(a^\dagger a+\frac{1}{2}\right)\right]\times\exp\left[-\frac{r_0^2}{2}[J(T)-J^*(T)]\right]\times\exp\{-ir_0[I^*(T)a+I(T)a^\dagger]\}.$$

Define D_α by

$$D_\alpha=e^{\alpha a^\dagger-\alpha^* a}=\exp\left[-\frac{1}{2}|\alpha|^2\right]e^{\alpha a^\dagger}e^{-\alpha^* a}. \tag{29}$$

These operators satisfy the identities

$$D_\beta D_\alpha=D_{\alpha+\beta}\exp\left[-\frac{1}{2}(\alpha\beta^*-\alpha^*\beta)\right],$$

$$D_\alpha^\dagger=D_\alpha^{-1}=D_{-\alpha}. \tag{30}$$

Let

$$\gamma(T)=ir_0 I(T)\frac{1}{e^{i\omega_0 T}-1}. \tag{31}$$

Claim: $D^\dagger_{\gamma(T)}|n\rangle$ is an eigenstate of $U(T)$ with Floquet multiplier $e^{-i\mu_n T}$, where

$$\mu_n=\left(n+\frac{1}{2}\right)\omega_0-i\frac{r_0^2}{2T}(J(T)-J^*(T))-\frac{r_0^2}{2T}|I(T)|^2\frac{\sin(\omega_0 T)}{1-\cos(\omega_0 T)}.$$

Proof. Rewrite the monodromy operator as

$$U(T)=\exp\left[-i\omega_0 T\left(a^\dagger a+\frac{1}{2}\right)\right]\times\exp\left[-\frac{r_0^2}{2}[J(T)-J^*(T)]\right]D_{-ir_0 I(T)}.$$

Therefore

$$\begin{aligned}U(T)D^\dagger_{\gamma(T)}|n\rangle&=\exp\left[-i\omega_0 T\left(a^\dagger a+\frac{1}{2}\right)\right]\times\exp\left[-\frac{r_0^2}{2}[J(T)-J^*(T)]\right]\times D_{-ir_0 I(T)}D_{-\gamma(T)}|n\rangle\\&=\exp\left[-\frac{r_0^2}{2}[J(T)-J^*(T)]\right]\times D_{-ir_0 I(T)e^{-i\omega_0 T}}D_{-\gamma(T)e^{-i\omega_0 T}}\times\exp\left[-i\omega_0 T\left(n+\frac{1}{2}\right)\right]|n\rangle\end{aligned}$$

because of the general identity

$$\exp[-i\omega_0 T a^\dagger a]D_\alpha=D_{\alpha e^{-i\omega_0 T}}\exp[-i\omega_0 T a^\dagger a].$$

Now use Eq. (30) to obtain

$$D_{-ir_0 I(T)e^{-i\omega_0 T}}D_{-\gamma(T)e^{-i\omega_0 T}}=D_{-[ir_0 I(T)+\gamma(T)]e^{-i\omega_0 T}}\exp\left[-\frac{1}{2}[-\gamma(T)ir_0 I^*(T)-\gamma^*(T)ir_0 I(T)]\right].$$

Using Eq. (31) yields

$$-[ir_0 I(T)+\gamma(T)]e^{-i\omega_0 T}=-\gamma(T)$$

and

$$-\gamma(T)ir_0 I^*(T)-\gamma^*(T)ir_0 I(T)=-ir_0^2|I(T)|^2\frac{\sin\omega_0 T}{1-\cos\omega_0 T}.$$

QED.

Further calculation yields the identities

$$I(T)=\frac{i\omega_0}{\omega_0^2-\omega^2}(e^{i\omega_0 T}-1),$$

$$J(T)-J^*(T)=\frac{1}{\omega^2-\omega_0^2}\left(i\omega_0 T+2i\frac{\omega_0^2}{\omega^2-\omega_0^2}\sin(\omega_0 T)\right),$$

$$\gamma(T)=-\frac{r_0\omega_0}{\omega_0^2-\omega^2}, \tag{32}$$

$$\mu_n=\left(n+\frac{1}{2}\left[1+\frac{r_0^2}{\omega^2-\omega_0^2}\right]\right)\omega_0.$$

The Floquet states $F_n(t)$ are given by (see the Appendix)

$$\begin{aligned}F_n(t)&=U(t)D^{\dagger}_{\gamma(T)}|n\rangle\\&=\exp\left[-\frac{r_0^2}{2}[J(t)-J^*(t)]\right]D_{-ir_0 I(t)e^{-i\omega_0 t}}D_{-\gamma(T)e^{-i\omega_0 t}}\\&\quad\times\exp\left[-i\omega_0 t\left(n+\frac{1}{2}\right)\right]|n\rangle.\end{aligned} \tag{33}$$

Using Eqs. (27), (31), and (32), it may be shown that

$$e^{i\mu_n t}F_n(t)=\exp\left[-\frac{ir_0^2\omega_0}{4\omega}\frac{\sin 2\omega t}{\omega^2-\omega_0^2}\right]D_{\eta(t)}|n\rangle, \tag{34}$$

where

$$\eta(t)=\frac{r_0}{\omega_0^2-\omega^2}[\omega_0\cos(\omega t)-i\omega\sin(\omega t)]. \tag{35}$$

This right-hand side of Eq. (34) is manifestly T periodic as is required of a genuine Floquet state when multiplied by the inverse of the Floquet multiplier. From the first equality in Eq. (33) it follows that

$$|n\rangle=D_{\gamma(T)}F_n(0)$$

and this justifies the claim that the Floquet states are a complete set of states. Moreover, they are orthonormal as follows from

$$\begin{aligned}\langle F_m(t)|F_n(t)\rangle&=\langle m|D_{\gamma(T)}U^{\dagger}(t)U(t)D^{\dagger}_{\gamma(T)}|n\rangle\\&=\langle m|n\rangle=\delta_{mn}.\end{aligned} \tag{36}$$

Armed with these Floquet states, it is now possible to solve the original problem of finding solutions for the $a_m(t)$'s. At $t=0$,

$$|\psi(0)\rangle=\sum_n a_n(0)|n\rangle$$

both for the original equations and for the auxiliary equations since they agree at $t=0$. For the auxiliary equations,

$$a_m(t)=\langle m|\psi(t)\rangle=\langle m|U(t)|\psi(0)\rangle.$$

Using the completeness of the Floquet states, this becomes

$$\begin{aligned}a_m(t)&=\sum_k\langle m|U(t)|F_k(0)\rangle\langle F_k(0)|\psi(0)\rangle\\&=\sum_k\langle m|F_k(t)\rangle\langle F_k(0)|\psi(0)\rangle.\end{aligned}$$

The solution to the original problem is

$$\begin{aligned}\sum_m a_m(t)|\psi'_m\rangle&=\sum_m a_m(t)\exp\left[-i\frac{e}{\hbar c}A(t)q\right]|m\rangle\\&=\sum_m\sum_k\exp\left[-i\frac{e}{\hbar c}A(t)q\right]|m\rangle\\&\quad\times\langle m|F_k(t)\rangle\langle F_k(0)|\psi(0)\rangle\\&=\sum_k\exp\left[-i\frac{e}{\hbar c}A(t)q\right]\\&\quad\times|F_k(t)\rangle\langle F_k(0)|\psi(0)\rangle.\end{aligned}$$

The Goeppert-Mayer gauge factor can be rewritten as

$$\begin{aligned}&\exp\left[-i\frac{e}{\hbar c}A(t)q\right]\\&\quad=\exp\left[-i\frac{e}{\hbar c}A_0\sin(\omega t)\sqrt{\frac{\hbar}{2m\omega_0}}(a+a^{\dagger})\right]\\&\quad=\exp\left[-i\frac{e}{\hbar c}\frac{cE_0}{\omega}\sin(\omega t)\sqrt{\frac{\hbar}{2m\omega_0}}(a+a^{\dagger})\right]\\&\quad=\exp\left[-ir_0\frac{\sin(\omega t)}{\omega}(a+a^{\dagger})\right]\\&\quad=D_{-ir_0\frac{\sin(\omega t)}{\omega}}\end{aligned} \tag{37}$$

that utilized the inverse of the identity following Eq. (19), $E_0=\omega A_0/c$, Eqs. (26) and (29). Therefore, the solution to the original problem is

$$|\psi(t)\rangle=D_{-ir_0\frac{\sin\omega t}{\omega}}\sum_k|F_k(t)\rangle\langle F_k(0)|\psi(0)\rangle.$$

Let N be an integer and consider

$$\begin{aligned}\langle F_m(NT)|\psi(NT)\rangle&=\sum_k\langle F_m(NT)|F_k(NT)\rangle\langle F_k(0)|\psi(0)\rangle\\&=\langle F_m(0)|\psi(0)\rangle\end{aligned} \tag{38}$$

that follows from the orthonormality of the Floquet states, Eq. (36), and from the fact that the Goeppert-Mayer gauge factor, Eq. (37), is the identity when evaluated at $t=NT$. Thus the projections of the solution onto the Floquet states

are T periodic. As an example, consider the case $|\psi(0)\rangle = |0\rangle$. Using Eqs. (34)–(35), it follows that

$$|\langle F_m(t)|0\rangle|^2 = |\langle m|D^\dagger_{-\eta(t)}|0\rangle|^2 = \exp[-|\eta(t)|^2]\frac{|\eta(t)|^{2m}}{m!}. \tag{39}$$

This satisfies Eq. (38) for $t=NT$ and it is manifestly T periodic for all t since Eq. (35) implies

$$|\eta(t)|^2 = \frac{r_0^2}{(\omega_0^2-\omega^2)^2}[\omega_0^2\cos^2\omega t + \omega^2\sin^2(\omega t)]. \tag{40}$$

Note that the right-hand side of Eq. (39) is always less than 1 and that the sum over m equals 1.

V. DRIVEN HARMONIC OSCILLATOR FLOQUET STATE PROPERTIES

The $n=0$ Floquet state, according to Eq. (34), is proportional to

$$D^\dagger_{-\eta(t)}|0\rangle = \exp\left[-\frac{1}{2}|\eta(t)|^2\right]\sum_n \frac{(-\eta(t))^n}{\sqrt{n!}}$$

that happens to be an ordinary harmonic oscillator coherent state. For $n>0$, noncoherent state generalizations are created. By using

$$q = \sqrt{\frac{\hbar}{2m\omega_0}}(a+a^\dagger),$$

$$p = -i\sqrt{\frac{m\omega_0\hbar}{2}}(a-a^\dagger),$$

and the identity

$$D^\dagger_\alpha f(a,a^\dagger)D_\alpha = f(a+\alpha,a^\dagger+\alpha^*) \tag{41}$$

the following expectation values can be computed:

$$\langle F_n(t)|q|F_n(t)\rangle = \sqrt{\frac{\hbar}{2m\omega_0}}(\eta+\eta^*) = \frac{eE_0}{m}\frac{\cos(\omega t)}{\omega_0^2-\omega^2},$$

$$\langle F_n(t)|p|F_n(t)\rangle = -i\sqrt{\frac{m\omega_0\hbar}{2}}(\eta-\eta^*) = -eE_0\omega\frac{\sin(\omega t)}{\omega_0^2-\omega^2}, \tag{42}$$

$$\langle F_n(t)|q^2|F_n(t)\rangle = \frac{\hbar}{2m\omega_0}(2n+1+\eta^2+\eta^{*2}+2|\eta|^2),$$

$$\langle F_n(t)|p^2|F_n(t)\rangle = -\frac{m\omega_0\hbar}{2}[-(2n+1)+\eta^2+\eta^{*2}-2|\eta|^2].$$

Therefore, the uncertainty product for all t is given by

$$\Delta q\Delta p = \frac{\hbar}{2}(2n+1),$$

which has the minimum possible value only for the $n=0$ case for which the Floquet state reduces to a phase factor times a coherent state.

The classical problem corresponding to the quantum Floquet problem solved here for the auxiliary equations has Hamiltonian

$$H = \frac{p^2}{2m} + \frac{1}{2}m\omega_0^2q^2 - eE_0q\cos(\omega t).$$

Defining y by $y=m\omega_0 q$, results in the classical equations of motion

$$\frac{d}{dt}\begin{pmatrix} y \\ p \end{pmatrix} = \begin{pmatrix} 0 & \omega_0 \\ -\omega_0 & 0 \end{pmatrix}\begin{pmatrix} y \\ p \end{pmatrix} + \begin{pmatrix} 0 \\ eE_0\cos(\omega t) \end{pmatrix}.$$

For arbitrary initial conditions, the solution contains a term that oscillates with frequency ω and other terms that oscillate with frequency ω_0. Only for the special initial conditions

$$q(0) = \frac{eE_0}{m}\frac{1}{\omega_0^2-\omega^2} \quad \text{and} \quad p(0)=0$$

is the solution given in terms of purely ω oscillations

$$q(t) = \frac{eE_0}{m}\frac{\cos(\omega t)}{\omega_0^2-\omega^2},$$

$$p(t) = -eE_0\omega\frac{\sin(\omega t)}{\omega_0^2-\omega^2}. \tag{43}$$

Since these solutions are T periodic, they are the classical Floquet solutions, and they agree precisely with the expectation values for the quantum Floquet states given in Eq. (42).

A similar analysis can be done for the Hamiltonian in Eq. (23). The classical result for $q(t)$ is the same as in Eq. (43) but the solution for $p(t)$ is different since the relationship between momentum and velocity now involves the vector potential. The expectation values appropriate for this perspective require the Goeppert-Mayer gauge factor given in Eq. (37). This factor must premultiply the Floquet state before the expectations are computed. Since this factor commutes with the operator q but not with the operator p the result for the positional expectation is the same as in Eq. (42), whereas the result for the momentum expectation is different, as in the classical analysis, and agrees precisely with the classical result.

VI. ARE THERE ANY MULTIPHOTON RESONANCES?

A primary motivation for this paper was to elucidate multiphoton excitation of a harmonic oscillator when the driving

frequency ω is much less than the natural oscillator frequency ω_0. The naive expectation is that when $\omega_0 = n\omega$ for integer n an n-photon resonant event occurs. Starting from the ground state, $|0\rangle$, the result in Eqs. (39)–(40) shows that there are no such events. The only singularity in Eq. (40) occurs for the simple resonance $\omega_0 = \omega$. While starting in an excited state produces a somewhat more complicated formula than appears in Eq. (39), the result is the same, there are no n-photon resonances. This is presumably a peculiarity of the harmonic oscillator and not the general situation. However, a detailed study of the general two-level quantum system gave the same result. In addition, a brute force analysis of the perturbation expansion for the oscillator case shows that this conclusion for the oscillator is justified. The reason can be seen with the three photon terms in the perturbation expansion. The perturbation expansion suggests that there are two ways to make a three photon resonance between the ground state and the first excited state. In one way the oscillator creation and annihilation operators appear in the order $aa^\dagger a^\dagger$ and in the other way they appear in the order $a^\dagger aa^\dagger$. When the expectation values of these combinations between the ground state and the first excited state are combined with the corresponding time integral factors, there is a precise cancellation. This phenomenon can be seen to be repeated in higher-order terms. It is this precise cancellation that is not to be expected in general for cases such as the microwave irradiation of Rydberg atoms. The details of what happens in the Rydberg case remain to be studied.

While engaged in this research, the work of Gribakin and Kuchiev [5] was brought to our attention. This work was inspired by an early paper of Keldysh [4]. A number of other authors developed the idea [9]. In the Gribakin and Kuchiev paper, a very intriguing and tantalizing idea is presented. It is the notion that the n-photon resonances are not to be expected between the unperturbed states of the oscillator but between an initial unperturbed state and a final Floquet state. Thus, the energy difference between the Floquet quasienergy and the initial unperturbed energy should equal the energy of n photons

$$\hbar\mu_m - \hbar\omega_0\left(k+\frac{1}{2}\right) = \hbar\omega_0(m-k) + \frac{1}{2}\hbar\omega_0\frac{r_0^2}{\omega^2-\omega_0^2} = n\hbar\omega, \tag{44}$$

wherein Eq. (32) has been used. This result is based on the Fourier components of the transition matrix element given by the expression

$$A_{mn} = \frac{1}{T}\int_0^T dt\langle e^{i\mu_m t}F_m(t)|V(t)|0\rangle e^{in\omega t}, \tag{45}$$

where $T = 2\pi/\omega$ and

$$V(t) = -eE_0\sqrt{\frac{\hbar}{2m\omega_0}}\cos(\omega t)(a+a^\dagger).$$

In Eq. (45), it is assumed that the initial state is the unperturbed ground state and that the final state is the mth

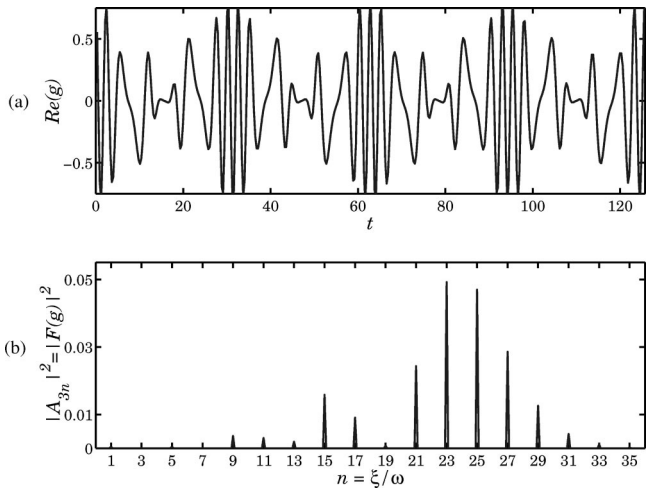

FIG. 1. (a) shows the real part of the matrix element $g(t) = \langle e^{i\mu_m t}F_m(t)|V(t)|0\rangle$ [Eq. (46)] as a function of time (dimensionless) with parameter values $m=3$, $k=0$, $\omega_0=1.0$, $\omega=0.1$, $r_0=2.312$, and $n=3$. It is clearly T periodic with period $T=62.83$. (b) shows the square of the absolute value of the Fourier transform $F(g)$ of g, as a function of the Fourier transform variable ξ divided by ω [Eq. (45)]. While there is no peak at 3, there are many other peaks at other odd numbers, but none at 5.

Floquet state, represented in the integral by its T-periodic portion. Using Eqs. (34), (35), (40), and (41), the matrix element in Eq. (45) becomes

$$\begin{aligned} g(t) &= \langle e^{i\mu_m t}F_m(t)|V(t)|0\rangle \\ &= -\hbar r_0\cos(\omega t)\frac{\exp\left[-\frac{1}{2}|\eta(t)|^2\right]}{\sqrt{m!}}\{[m-|\eta(t)|^2] \\ &\quad\times[-\eta(t)]^{m-1}\}\exp\left[i\frac{r_0^2\omega_0}{4\omega}\frac{\sin(2\omega t)}{\omega^2-\omega_0^2}\right]. \end{aligned} \tag{46}$$

In Fig. 1(a), the real part of the matrix element $g(t)$ in Eq. (46) is plotted against time for parameter values $m=3$, $k=0$, $\omega_0=1.0$, $\omega=0.1$, $r_0=2.312$, and $n=3$. These parameters refer to Eq. (44) and reflect choosing the electric field strength to have a value that implies the energy of three photons matches the difference in energy (quasienergy) between the ground state and the third Floquet state. It is clear from the figure that this quantity is T periodic ($T=62.83$). In Fig. 1(b), the square of the absolute value of the Fourier transform $F(g)$ of g, is shown in a plot of $|A_{3n}|^2$ against n, as defined in Eq. (45). There is no peak at $n=3$, as anticipated, and there are many other peaks at other odd numbers. Only the peak at 3 would have satisfied the quasienergy conservation requirement of Eq. (44). No sign of this putative resonance is seen in Eqs. (39)–(40). Indeed, at $t=0$, the largest value for $|\langle F_m(0)|0\rangle|^2$ occurs when

$$|\eta(0)|^2 = m\exp\left[\frac{1}{2m}\right]$$

as follows from Stirlings approximation. For the parameter values used in Fig. 1, this implies a value of $m=5$. Since the

results in Eq. (39) are T periodic, the $m=5$ Floquet state will be the dominant contributor periodically. By contrast, the Fourier transform in Fig. 1 shows no peak whatsoever for $n=5$. We are forced to conclude that the Gribakin and Kuchiev perspective is not valid for the periodically driven harmonic oscillator.

For the periodically driven harmonic oscillator, a completely explicit solution has been obtained in terms of closed form Floquet states. These states make it manifestly clear that there are no n-photon resonances. This quantum result is born out by the corresponding classical Floquet solution.

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APPENDIX: FLOQUET STATES

Construction of Floquet states in quantum mechanics is most easily achieved using the time ordered exponential. Let

$$H(t+T)=H(t). \tag{A1}$$

The evolution operator is expressible by

$$U(t_1,t_2)=\underset{\leftarrow}{T}\exp\left[-\frac{i}{\hbar}\int_{t_1}^{t_2}dsH(s)\right], \tag{A2}$$

wherein the right-hand side utilizes the time-order exponential [8]. The factorization property

$$\underset{\leftarrow}{T}\exp\left[-\frac{i}{\hbar}\int_{t_1}^{t_2}dsH(s)\right]=\underset{\leftarrow}{T}\exp\left[-\frac{i}{\hbar}\int_{r}^{t_2}dsH(s)\right]\times\underset{\leftarrow}{T}\exp\left[-\frac{i}{\hbar}\int_{t_1}^{r}dsH(s)\right] \tag{A3}$$

for $t_2>r>t_1$ is used below. Let $T=2\pi/\omega$ be the period of the periodic driving term. The monodromy operator, $U(0,T)$, has eigenstates with unimodular eigenvalues

$$U(0,T)|m\rangle=e^{-i\mu_m T}|m\rangle. \tag{A4}$$

Using factorization, Eqs. (A3), and (A4) yields

$$\begin{aligned}U(t,t+T)(U(0,t)|m\rangle)&=U(T,t+T)U(0,T)|m\rangle\\&=U(T,t+T)e^{-i\mu_m T}|m\rangle\\&=e^{-i\mu_m T}U(T,t+T)|m\rangle\\&=e^{-i\mu_m T}(U(0,t)|m\rangle),\end{aligned}$$

where the last equality follows from Eq. (A1) because

$$\begin{aligned}U(T,t+T)&=\underset{\leftarrow}{T}\exp\left[-\frac{i}{\hbar}\int_{T}^{t+T}dsH(s)\right]\\&=\underset{\leftarrow}{T}\exp\left[-\frac{i}{\hbar}\int_{0}^{t}ds'H(s'+T)\right]\\&=\underset{\leftarrow}{T}\exp\left[-\frac{i}{\hbar}\int_{0}^{t}ds'H(s')\right]=U(0,t).\end{aligned}$$

Thus, $U(0,t)|m\rangle$ is an eigenfunction of $U(t,t+T)$ with the same eigenvalue as the monodromy operator has for eigenfunction $|m\rangle$. Let $|m,t\rangle=U(0,t)|m\rangle$.

The Floquet states $F_m(t)$ have the form

$$F_m(t)=e^{-i\mu_m t}\phi_m(t),$$

$$\phi_m(t+T)=\phi_m(t),$$

$$i\hbar\frac{\partial}{\partial t}\phi_m(t)=H(t)\phi_m(t)-\hbar\mu_m\phi_m(t).$$

Claim:

$$F_m(t)=|m,t\rangle.$$

Proof. From Eq. (A2) it follows that

$$i\hbar\frac{\partial}{\partial t}U(0,t)=H(t)U(0,t),$$

which implies

$$i\hbar\frac{\partial}{\partial t}|m\rangle=H(t)|m,t\rangle.$$

Therefore,

$$i\hbar\frac{\partial}{\partial t}(e^{i\mu_m t}|m,t\rangle)=(H(t)-\hbar\mu_m)(e^{i\mu_m t}|m,t\rangle).$$

Thus, $\phi_m(t)=e^{i\mu_m t}|m,t\rangle$. Moreover,

$$\begin{aligned}\phi_m(t+T)&=e^{i\mu_m(t+T)}|m,t+T\rangle\\&=e^{i\mu_m(t+T)}U(t,t+T)|m,t\rangle\\&=e^{i\mu_m(t+T)}e^{-i\mu_m T}|m,t\rangle\\&=e^{i\mu_m t}|m,t\rangle=\phi_m(t).\end{aligned}$$

QED.

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