General method for determining the Floquet states of the forced rotor and other anharmonic systems

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An approximate analytical solution is developed for the Floquet states of a quantum-mechanical rigid rotor subject to an arbitrary periodically-time-dependent driving force. The rotor Hamiltonian is expanded in powers of the rotational "anharmonicity" about the angular-momentum state $|j_0\rangle$ and a perturbative Lie algebraic form of the evolution operator is derived. Except in special cases, the resulting Floquet states are localized. The localization length shows a sharp transition from narrow to broad for those Floquet states in the vicinity of a resonance with one of the Fourier components of the driving field. The special case of the periodically kicked rotor is also discussed, which, because of its unbounded frequency spectrum, can lead to delocalized Floquet states.

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I. INTRODUCTION

The past decade has seen considerable interest in exploring the quantum-mechanical analogs of classically chaotic systems. One class of these comprises periodically driven systems. Within this class the kicked rotor [1–7] and the microwave ionization of hydrogen atoms [8–15] stand out as two topics which have received a great deal of experimental and theoretical attention. As a result of these investigations there is a growing consensus that quantum mechanics suppresses chaotic behavior [16]; however, the degree to which some manifestations of chaos survive in the quantum realm is not yet entirely understood.

A wealth of information about the dynamics of strongly perturbed systems has come from studies of the kicked rotor. Classically, kick strengths below a critical value lead to trajectories bounded by Kolmogorov-Arnold-Moser [17] tori, whereas above the critical value the momentum grows diffusively. Numerical investigations have shown that this diffusive growth is mimicked for a finite time by the quantum kicked rotor, after which it is suppressed [18] via a dynamical interference, shown by Fishman and co-workers [2,6], to have a deep underlying connection with Anderson localization [19].

The localization phenomenon is intimately related to the quasienergy spectrum and Floquet eigenstates of the quantum kicked rotor, as discussed in a series of recent papers by Dittrich and Smilansky [20]. Thus, a rational ratio between the kick and natural rotor frequency leads to extended Floquet states and continuous quasienergy spectra, whereas an irrational ratio yields localized states and quasiperiodic motion [21]. The importance of the quasienergy operator is also apparent in the study of other forced anharmonic systems. For example, Blümel and Smilansky [14] based their analysis of the microwave ionization of hydrogen on the existence of broad Floquet states, which act as "ionization windows," and Tanner and Maricq [22] pursued a similar approach to explore the infrared dissociation of a Morse oscillator.

While a considerable effort has gone into the study of the kicked rotor, much less is known about how other types of periodic forces affect the quantum rigid rotor. Blümel, Fishman, and Smilansky [23] have extended the kicked rotor analysis to consider finite-duration microwave pulses, and find that the mechanism of Anderson localization continues to take place. Lin and Reichl [24] have computed quasienergies and Floquet states for a rotor in the presence of two nonlinear resonances and have examined the relationship between the localization length of the Floquet states and the nature of the corresponding classical trajectories. The purpose of the present paper is to derive a general analytical expression for the Floquet eigenstates of a forced rotor, where the applied force is periodic in time but otherwise arbitrary. Such a problem does not permit an exact solution; however, an approximate solution is possible using an algebraic framework recently introduced to calculate rotational transition probabilities [25].

The solution for the Floquet states is based on mapping the forced-rotor Hamiltonian onto the Lie algebra generated by a number operator and the associated raising and lowering operators. This mapping is exact under the two approximations that the local energy-level structure is linear in rotational quantum number and that the interaction operator between rotor and driving field is a linear combination of operators which raise and lower the total angular momentum by an integer number of quanta. With these approximations, an exact solution to the exponential form of the evolution operator becomes feasible and yields the Floquet eigenstates. The Floquet states of the real rotor are determined by introducing the "anharmonicity" in the rotational energy levels as a perturbation. The first-order corrections both significantly improve the theoretical predictions and elucidate the role of discrete and unevenly spaced energy levels on the rotational dynamics.

The solution of the forced-rotor problem shows that the Floquet states are in general localized, with a localization length which grows with the strength of the driv-

ing force and shrinks with the extent of detuning between the local free-rotor energy-level spacing and the driving frequency, as has been observed in numerical simulations [24]. Delocalized states arise in the "linearized" rotor when the level spacing is resonant with the fundamental frequency of the external force or one of its harmonic components. These are, in general, effectively suppressed by the "anharmonicity" of a real rotor; yet even in this case there is a sharp transition from narrow to much broader Floquet states in the vicinity of the resonance. A series of infinitely sharp kicks contains a sufficient number of harmonic frequencies to ensure that all or nearly all the rotor levels fall into these resonance regions and thereby produces delocalized Floquet states even though the rotor is anharmonic [26].

The body of the paper is organized as follows. Section II reviews the algebraic formalism and necessary approximations that are used in the analytical treatment of the forced rotor. The equations for the Lie coefficients governing the exponential solution to the time-dependent Schrödinger equation are developed Sec. III. A perturbative solution to these equations for the case of the forced rotor is presented in Sec. IV and the results are compared to exact numerical calculations.

II. ALGEBRAIC PRELIMINARIES

The Hamiltonian for a rigid rotor subject to a time-dependent external force,

$$\mathcal{H}=\Omega \mathbf{j}^2+f(t)\cos\theta \ , \tag{2.1}$$

depends on two noncommuting operators, $\mathbf{j}^2$ and $\cos\theta$ (with units $\hbar=1$), with θ representing position and $\mathbf{j}$ the conjugate momentum. The Floquet eigenstates are found from the evolution operator $U(\tau)$ that propagates the system forward one period in time. Because of the noncommutativity of $\mathbf{j}^2$ and $\cos\theta$, an exact solution for $U(\tau)$ is not feasible. The approximate solution proceeds by mapping the forced-rotor Hamiltonian onto the algebra of raising and lowering operators familiar from the harmonic oscillator [25]. This section describes how the Hamiltonian is rewritten in terms of this algebra and the approximations that are involved.

The first approximation consists of linearizing the rotational kinetic energy about a state $|j_0\rangle$ and treating the "anharmonicity" in the rotational energy levels as a perturbation. Linear approximations of this sort have previously been used for a semiclassical treatment of rotationally inelastic scattering [25], to analyze spectral line broadening [27], and to investigate rotational excitation by high-intensity laser fields [28]. Let $n=j-j_0$ define the number of "incremental" rotor quanta with respect to the expansion point j_0. Then we can write the rotational kinetic energy in terms of n as

$$\Omega \mathbf{j}^2|j_0+n\rangle=(C_0+\Omega_e n+\Omega_e X_e n^2)|j_0+n\rangle \ ,$$

where $\Omega_e\equiv(2j_0+1)\Omega$, $\Omega_e X_e=\Omega$, and $C_0=\Omega j_0(j_0+1)$. For $j_0\gg 0$ and $|n|\ll j_0$, the term proportional to n^2 is much smaller than the linear term and will be treated as a perturbation. Thus, upon defining the number operator $\mathbf{n}|j_0+n\rangle=n|j_0+n\rangle$, the rotational kinetic-energy operator to first order in n becomes

$$\Omega \mathbf{j}^2\simeq(\Omega_e \mathbf{n}+C_0) \ . \tag{2.2}$$

The second approximation concerns the operator $\cos\theta$. Because $\cos\theta$ is diagonal in azimuthal quantum number, we can think of it as operating in a one-dimensional space but with a parametric dependence on m. This dependence adds no complexity to the ensuing discussion and is kept for the sake of generality. By examining the action of $\cos\theta$ on the spherical harmonics, $Y_j^m(\theta,\phi)$, one easily verifies that it has matrix elements [29]

$$\langle j'm'|\cos\theta|jm\rangle = [C_m(j+1)\delta_{j',j+1}+C_m(j)\delta_{j',j-1}]\delta_{m',m} \ , \tag{2.3}$$

with the definition $C_m(j)\equiv(j^2-m^2)^{1/2}(4j^2-1)^{-1/2}$. The simpler form, $C_m(j)\simeq\frac{1}{2}(1-m^2/j^2)^{1/2}$, affords an excellent approximation except at the very lowest values of total angular momentum and is used throughout the remainder of this paper. Note that for the planar rotor $m=0$ and $C_0(j)=\frac{1}{2}$ is independent of j. Even when $m\neq 0$, $C_m(j)$ remains only weakly dependent on j, except when $j\approx|m|$.

One can take advantage of the weak-j dependence of $C_m(j)$ by introducing the phase operators $e^{i\theta}$ and $e^{-i\theta}$. These are defined by $e^{i\theta}|j\rangle=|j-1\rangle$ and $e^{-i\theta}|j\rangle=|j+1\rangle$. Their commutation relations with the number operator are

$$[\mathbf{n},e^{i\theta}]=-e^{i\theta} \ , \tag{2.4a}$$

$$[\mathbf{n},e^{-i\theta}]=e^{-i\theta} \ . \tag{2.4b}$$

The two phase operators *almost* commute with each other; that is, the matrix elements of the commutators are

$$\langle j',m|[e^{i\theta},e^{-i\theta}]|j,m\rangle=0 \tag{2.5a}$$

for all j' and j *except* when $j'=j=|m|$, in which case

$$\langle |m|,m|[e^{i\theta},e^{-i\theta}]||m|,m\rangle=1 \ . \tag{2.5b}$$

An exact representation of $\cos\theta$ in terms of the phase operators is given by

$$\cos\theta=C_m(\mathbf{j})e^{-i\theta}+e^{i\theta}C_m(\mathbf{j}) \ , \tag{2.6}$$

where $C_m(\mathbf{j})$ is an operator function. However, because of the weak-j dependence of $C_m(j)$ we can make the approximation that

$$\cos\theta\approx C_m(j_0)(e^{i\theta}+e^{-i\theta}) \ , \tag{2.7}$$

with $C_m(j_0)$ now treated as a scalar. The net result of the above two approximations is that it permits us to express the forced-rotor Hamiltonian as

$$\mathcal{H}=\mathcal{H}_0+\mathcal{H}_1+C_0 \ , \tag{2.8}$$

with

$$\mathcal{H}_0=\Omega_e \mathbf{n}+f(t)C_m(j_0)(e^{i\theta}+e^{-i\theta}) \ , \tag{2.9a}$$

$$\mathcal{H}_1=\Omega_e X_e \mathbf{n}^2 \ . \tag{2.9b}$$

The advantage of this form is that the unperturbed Hamiltonian is written entirely in terms of operators belonging to a Lie algebra,

$$\mathcal{L}_0=\{\mathbf{n},e^{i\theta},e^{-i\theta}\}\ , \tag{2.10}$$

which is approximately closed under the commutation product [i.e., ignoring the nonzero commutator matrix elements given by Eq. (2.5b), which is valid for $j_0 \gg |m|$]. The structure of $\mathcal{L}_0$ permits an exact solution for the Floquet Hamiltonian of the "linearized" rotor. By including the "anharmonicity" of the rotational energy levels, $\mathcal{H}_1$, as a perturbation, an approximate expression for the Floquet Hamiltonian of a real forced rotor can be found.

III. LIE ALGEBRAIC SOLUTION FOR THE FLOQUET HAMILTONIAN

The symmetry in time which occurs for a periodically-time-dependent Hamiltonian, with period τ, implies the existence [30] of a linear transformation, itself time independent, which maps state vectors at time t onto those at time $t+\tau$. Because it preserves vector norms and angles, the transformation is unitary; therefore,

$$|\psi(t+\tau)\rangle=\exp(-iH_F\tau)|\psi(t)\rangle\ ,$$

where H_F is defined as the Floquet Hamiltonian. In this section we will determine H_F by solving the Schrödinger equation

$$\frac{dU}{dt}=H(t)U \tag{3.1}$$

for the exponential form of the propagator, $U(t)=e^{\Omega(t)}$, and setting $H_F=i\Omega(\tau)/\tau$. The solution for $\Omega(t)$ that is presented takes advantage of the algebraic properties of the operators comprising $H(t)$ in a procedure analogous to one used by Wei and Norman [31] to develop a product representation of $U(t)$.

Let $\mathcal{L}=\{x_i\}$ denote the Lie algebra which is spanned by the elements x_i and is closed under the commutator product; that is,

$$[x_i,x_j]=\mathbf{c}_{ij}\cdot\mathbf{x}\ , \tag{3.2}$$

where the elements of $\mathbf{c}_{ij}$ are defined as the structure constants of the algebra. If $H(t)$ can be expressed in terms of the x_i, then $\Omega(t)$ is an element of $\mathcal{L}$, and we can define quantities h_i and g_i such that

$$H(t)=\sum_i h_i(t)x_i=\mathbf{h}\cdot\mathbf{x}\ , \tag{3.3}$$

$$\Omega(t)=\mathbf{g}\cdot\mathbf{x}\ . \tag{3.4}$$

According to a formula due to Wilcox [32],

$$\frac{de^{\Omega}}{dt}=\int_0^1 e^{u\Omega}\frac{d\Omega}{dt}e^{-u\Omega}du\ e^{\Omega} \tag{3.5}$$

provides a means of differentiating the exponential propagator. By combining this expression with Eqs. (3.1), (3.3), and (3.4), one obtains

$$\mathbf{h}\cdot\mathbf{x}=\frac{d\mathbf{g}}{dt}\cdot\int_0^1 e^{u\Omega}\mathbf{x}e^{-u\Omega}du\ . \tag{3.6}$$

The completeness of $\mathcal{L}$ implies that the integrand is an element of $\mathcal{L}$, whereby

$$e^{u\Omega}\mathbf{x}e^{-u\Omega}=A\cdot\mathbf{x}\ . \tag{3.7}$$

Owing to the property that $\mathcal{L}$ is closed, it is possible to define a matrix S such that

$$[\Omega,\mathbf{x}]=S\mathbf{x}\ ; \tag{3.8}$$

thus S is a function of the Lie coefficients $g_i(t)$ and the structure constants $\mathbf{c}_{ij}$. Upon differentiating Eq. (3.7) with respect to u, one finds that $dA/du=SA$. Because S is independent of u, this equation is easily integrated and yields $A=e^{uS}$. After substituting Eq. (3.7) into Eq. (3.6) and integrating with respect to u, we arrive at a differential equation for the components of $\Omega(t)$ in terms of those of $H(t)$,

$$\frac{d\mathbf{g}}{dt}=\mathbf{h}(e^S-1)^{-1}S\ , \tag{3.9}$$

subject to the initial condition $\mathbf{g}(0)=\mathbf{0}$. Note that the matrices $(e^S-1)^{-1}$ and S operate to the left on $\mathbf{h}$. This is a consequence of the convention chosen in Eqs. (3.3) and (3.4) of writing $\mathbf{h}$ and $\mathbf{g}$ to the left of $\mathbf{x}$.

The eigenstates of S would ordinarily provide a convenient basis in which to expand Eq. (3.9). Unfortunately, they are not necessarily complete, since S is not generally a normal matrix. This difficulty can be overcome by employing a basis formed by the *generalized* [33] eigenvectors of S, with the kth-order eigenvector corresponding to the eigenvalue s_i being defined by

$$\langle s_i^{(k)}|(S-s_i)^k=0\ . \tag{3.10}$$

Inserting the expansion $\mathbf{h}=\sum_i b_i\langle s_i|$ of the Hamiltonian in terms of the basis of generalized eigenvectors into Eq. (3.9) and writing $(e^S-1)^{-1}S$ in a power series about s_i yields

$$\frac{d\mathbf{g}}{dt}=\sum_i b_i\langle s_i|\left[\frac{s_i}{e^{s_i}-1}+\frac{e^{s_i}-1-s_ie^{s_i}}{(e^{s_i}-1)^2}(S-s_i)+\cdots\right]\ . \tag{3.11}$$

Owing to the definition of the generalized eigenvectors, Eq. (3.10), the series in brackets truncates at the $(k-1)$th term for an eigenvector of order k. The advantage of Eq. (3.11) over Eq. (3.9) is that the operator function present in the latter is converted into a numerical function of the eigenvalues of S.

Equation (3.9) is nonlinear due to the dependence of the matrix S on the functions g_i. Singularities appear in this equation whenever an eigenvalues of S equals $im2\pi$ for $m\neq 0$. The singularities arise from constraining the evolution operator to an exponential form, as pointed out by Magnus [34] in his original paper on the exponential solution to Eq. (3.1). A well-defined solution always exists for t sufficiently close to zero because $\det[(e^S-1)^{-1}S]\to 1$ as $t\to 0$. However, a well-behaved global solution is generally not possible [35]. In particular, series expansions of $\Omega(t)$ will at some time $t>0$ fail to converge because of the singularities. The forced rotor, however, is a special case. Under the approximations

introduced in Sec. II, $\mathcal{H}_0$ is an element of $\mathcal{L}_0$, and for this algebra an analytic solution to Eq. (3.9) is feasible.

IV. FLOQUET STATES OF THE FORCED ROTOR

A. Zeroth order

This section applies the algebraic formalism developed in the preceding section to the forced rotor and solves the differential equations for the Lie coefficients g_i. The forced rotor, to zeroth order in the "anharmonicity," evolves under the Hamiltonian $\mathcal{H}_0$ (given by Eq. (2.9a). Because $\mathcal{H}_0$ is an element of the three-dimensional algebra $\mathcal{L}_0$, the matrix S, defined by Eq. (3.8), has the form

$$S=\begin{bmatrix}0 & g_2 & -g_3\\ 0 & -g_1 & 0\\ 0 & 0 & g_1\end{bmatrix} . \tag{4.1}$$

Its eigenvalues and the corresponding eigenvectors (all of order 1) are

$$s_1=0,\quad \langle s_1|=(g_1,g_2,g_3) ,$$
$$s_2=-g_1,\quad \langle s_2|=(0,1,0) ,$$
$$s_3=g_1,\quad \langle s_3|=(0,0,1) .$$

The solution of Eq. (3.11) also requires the coefficients b_i used to expand the Hamiltonian in the basis operators of $\mathcal{L}_0$. Upon setting $\mathbf{h}\cdot\mathbf{x}=H(t)\equiv -i\mathcal{H}_0$, the coefficients are obtained from $\mathbf{h}\cdot\mathbf{x}=\sum_i b_i\langle s_i|\cdot\mathbf{x}$ (the values are listed as b_1–b_3 in Appendix A). Inserting s_i, $\langle s_i|$, and b_i into Eq. (3.11) yields the nested sequence of coupled differential equations,

$$\frac{dg_1}{dt}=-i\Omega_e , \tag{4.2a}$$

$$\frac{dg_2}{dt}=g_2\left[\frac{1}{t}-\frac{i\Omega_e}{e^{i\Omega_e t}-1}\right]+\frac{C_m f(t)\Omega_e t}{e^{i\Omega_e t}-1} , \tag{4.2b}$$

for the Lie coefficients of $\Omega(t)$, with the initial condition that $g_1(0)=g_2(0)=0$. The equation for g_3 is redundant, because the Hamiltonian is Hermitian and $g_3=-g_2^*$. Although nonlinear in general, Eq. (3.9) has uncoupled for $\mathcal{L}_0$ to yield a set of first-order ordinary differential equations. The solutions are obtained by standard procedures and are given by

$$g_1=-i\Omega_e t , \tag{4.3a}$$

$$g_2=\frac{\Omega_e t}{1-e^{-i\Omega_e t}}F(t) , \tag{4.3b}$$

where

$$F(t)\equiv C_m(j_0)\int_0^t dt' f(t')e^{-i\Omega_e t'}$$

is the component of the driving force at frequency Ω_e. Combining these solutions via $U(t)=e^{g\cdot x}$ provides the exponential form of the evolution operator for arbitrary times. By the definition $H_F=i\Omega(\tau)/\tau$, the Floquet Hamiltonian for the forced rotor to zeroth order in X_e is

$$H_{F0}=\Omega_e(\mathbf{n}+iGe^{i\theta}-iG^*e^{-i\theta}) , \tag{4.4}$$

where $G\equiv[1-\exp(-i\Omega_e\tau)]^{-1}F(\tau)$ and τ is the time for one period of the driving force.

The form of the Floquet Hamiltonian derived in Eq. (4.4) is essentially the same as that of the Hamiltonian for a charged harmonic oscillator in a static electric field [36]. Only the minor difference exists that the interaction with the field in the case of the harmonic oscillator depends on raising and lowering operators, whereas phase operators appear in Eq. (4.4). The analogy suggests that the states defined via

$$|J_F\rangle=e^{i(Ge^{i\theta}+G^*e^{-i\theta})}|j_0+n\rangle \tag{4.5}$$

are the Floquet eigenstates for the "linearized" forced rotor. It is readily verified, using the commutation relations

$$[\mathbf{n},f(e^{\pm i\theta})]=\mp e^{\pm i\theta}df/(de^{\pm i\theta}) , \tag{4.6}$$

$$[f(e^{i\theta}),g(e^{-i\theta})]=0 \tag{4.7}$$

for arbitrary functions f and g, that

$$H_{F0}|J_F\rangle=(C_0+n\Omega_e)|J_F\rangle , \tag{4.8}$$

where the constant term in the expansion of the rotational kinetic energy, Eq. (2.2), has been included.

It is important to keep in mind that the Floquet states are labeled by n, the displacement from the state $|j_0\rangle$ about which the kinetic energy is expanded and, therefore, that the labeling of states will depend on the choice of j_0. The net result is that to zeroth order in X_e the local quasienergies about any state $|j_0\rangle$ are the same as the free-rotor energy levels; however, it should be recalled that the quasienergies are only uniquely defined modulo $2\pi/\tau$.

The Floquet eigenstates, in contrast, differ markedly from the free-rotor states. The assumed commutativity between the two phase operators permits an exact calculation of the overlap between Floquet and free-rotor eigenstates; thus,

$$\langle j'|J_F\rangle=\sum_{k=0}^{\infty}\frac{i^k}{k!}\langle j_0+n'|(Ge^{i\theta}+G^*e^{-i\theta})^k|j_0+n\rangle$$
$$=\sum_{k=0}^{\infty}\frac{i^kG^{*(k-\Delta n)/2}G^{(k+\Delta n)/2}}{[(k+\Delta n)/2]![(k-\Delta n)/2]!} ,$$

where $\Delta n\equiv n'-n$. Changing the index of summation from k to $l=\frac{1}{2}(k-|\Delta n|)$ produces a series which is, except for a constant factor, identical to the series representation of the $|\Delta n|$-order cylindrical Bessel function [37]; thus,

$$\langle j_0+n'|J_F\rangle=e^{-i\Delta n(\phi-\pi/2)}J_{\Delta n}(2|G|) , \tag{4.9}$$

where the phase ϕ is defined by $e^{i\phi}\equiv G/|G|$.

Figure 1 compares the overlap of the Floquet eigenstates with the angular-momentum basis vectors that is predicted by Eq. (4.9) to exact numerical values deter-

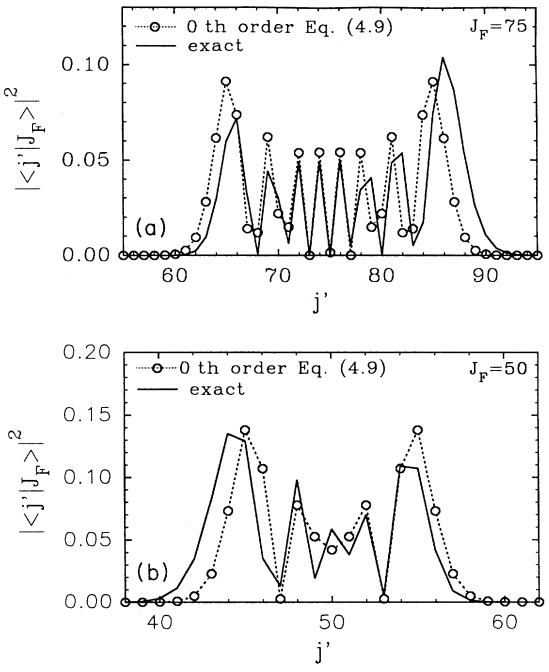

FIG. 1. Comparison of the zeroth-order approximate Floquet eigenstates to exact numerical results for a real rotor. The rotational constant is $\Omega=0.5$ cm^{-1} and $m=0$. (a) $\varepsilon=500$ cm^{-1}, $\omega=100$ cm^{-1}, and $j_0=J_F=75$. (b) $\varepsilon=400$ cm^{-1}, $\omega=27.5$ cm^{-1}, and $j_0=J_F=50$.

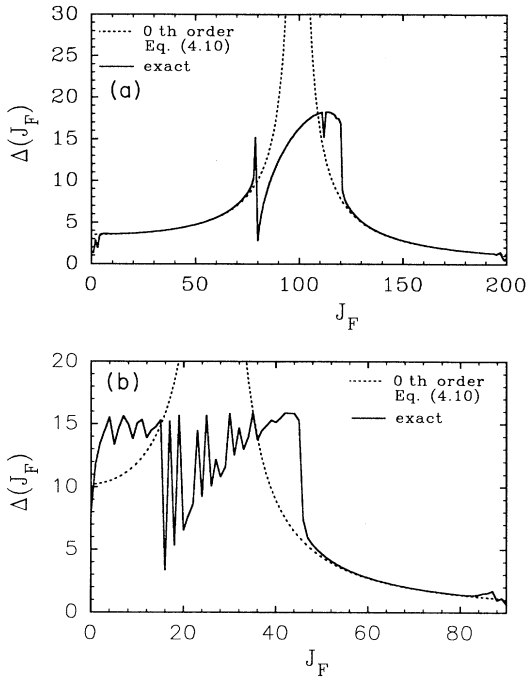

FIG. 2. Comparison between zeroth-order and exact Floquet state widths for the sinusoidally driven rotor. j_0 is set equal to J_F for each width determination. The rotational constant is $\Omega=0.5$ cm^{-1} and $m=0$. (a) $\varepsilon=500$ cm^{-1} and $\omega=100$ cm^{-1}. (b) $\varepsilon=400$ cm^{-1} and $\omega=27.5$ cm^{-1}.

mined by the procedure described in Appendix B. The example in Fig. 1 is that of a sinusoidally driven rotor; thus, $f(t)=\varepsilon\sin(\omega t)$ and $G=C_m\varepsilon\omega/(\omega^2-\Omega_e^2)$. In both cases, J_F is chosen to be removed from the resonance condition, i.e., when the local rotor energy-level spacing equals the driving frequency.

The widths of the zeroth-order Floquet states, defined by

$$\Delta_0(J_F)\equiv[\langle J_F|(\mathbf{n}-\bar{n})^2|J_F\rangle]^{1/2}=\sqrt{2}|G|\ , \tag{4.10}$$

are plotted in Fig. 2 for the example of a sinusoidally driven oscillator. The agreement with exact numerical results is very good away from the resonance condition. The singularity in the width at resonance is a consequence of the linear approximation for the local rotor energy-level spacing. While the singularity is absent, the exact widths show a pronounced increase in the resonance region. This behavior has also been observed in the numerical studies of quantum resonance overlap by Lin and Reichl [24].

While the zeroth-order approximation yields Floquet states in good qualitative agreement with those of a true rigid rotor, it falls short in a number of ways. The predicted quasienergies are identical (modulo $2\pi/\tau$) to the eigenvalues of a free rotor, a situation which is not supported by exact numerical calculations. The Bessel-function distribution of angular-momentum states obtained in zeroth order is symmetric in Δn, whereas the exact Floquet states exhibit a distinct asymmetry. The Floquet states of Eq. (4.9) exhibit singularities absent from the exact counterparts. Perhaps foremost, the special nature of the "linearized" rotor invites criticism that the good agreement between zeroth-order and exact Floquet states might be fortuitous.

B. First-order correction for the anharmonicity

In order to improve the quality of the analytical predictions and to investigate the role of the rotational "anharmonicity," the present section derives a Lie algebraic solution for the Floquet Hamiltonian that includes, to first order, the quadratic nature of the energy-level spacing of a true rigid rotor. To incorporate the anharmonicity, $\mathcal{L}_0$ must be expanded so that $\mathcal{H}_1=\Omega_e X_e\mathbf{n}^2$ can be written in terms of its elements. Because the magnitude of $\mathcal{H}_1$ relative to that of $\mathcal{H}_0$ is roughly $X_e=(2j_0+1)^{-1}$, let us temporarily attach the expansion parameter X_e to $\mathbf{n}^2$. Simply adding $X_e\mathbf{n}^2$ to $\mathcal{L}_0$ leaves it open; in fact, an infinite number of multiple commutators of the form $[\ldots[X_e\mathbf{n}^2,a],b\ldots]$ are needed to close it, where $a,b,\ldots$ are either $X_e\mathbf{n}^2$ or elements of $\mathcal{L}_0$. However, only a finite subset of such commutators is proportional to X_e^1. Thus, the forced-rotor Hamiltonian, $\mathcal{H}=\mathcal{H}_0+\mathcal{H}_1+C_0$, is an element of the algebra

$$\mathcal{L}_1=\{\mathbf{n},e^{i\theta},e^{-i\theta},\mathbf{n}^2,e^{i\theta}(2\mathbf{n}-1)\ ,$$
$$(2\mathbf{n}-1)e^{-i\theta},(e^{i\theta})^2,(e^{-i\theta})^2,1\}\ , \tag{4.11}$$

which is closed to first order in X_e (for simplicity the factor X_e is no longer attached to the operators).

The Floquet Hamiltonian for the forced rotor, to first order in X_e, is obtained by applying to $\mathcal{L}_1$ the Lie algebraic solution for the exponential form of the evolution operator developed in Sec. III. The interested reader is directed to Appendix A for an outline of the solution of Eq. (3.11) for the Lie coefficients $g_i(t)$. The result is

$$g_4=-i\Omega_e X_e t\ , \tag{4.12a}$$

$$g_5=\frac{\Omega_e X_e t}{1-e^{-i\Omega_e t}}\left[F(t)\left[1-\frac{i\Omega_e t}{e^{i\Omega_e t}-1}\right]+\Omega_e\frac{dF(t)}{d\Omega_e}\right]\ , \tag{4.12b}$$

$$g_7=\frac{2\Omega_e^2 X_e t}{1-e^{-i2\Omega_e t}}\left[F(t)\int_0^t F(t')dt'-\int_0^t F^2(t')dt'\right]-\frac{i\Omega_e^2 X_e t^2 F^2(t)}{(1-e^{-i\Omega_e t})^2}\left[\frac{1}{\sin\Omega_e t}-\frac{1}{\Omega_e t}\right]\ , \tag{4.12c}$$

$$g_9=-i2X_e\left\{\Omega_e\int_0^t|F(t')|^2dt'-\left|\frac{\Omega_e tF(t)}{1-e^{-i\Omega_e t}}\right|^2\left[\frac{\sin\Omega_e t}{1-\cos\Omega_e t}-\frac{2}{\Omega_e t}\right]\right.$$
$$\left.+\Omega_e\,\mathrm{Im}\left[\left[\frac{\Omega_e t}{1-\cos\Omega_e t}-\frac{2i}{1-e^{-i\Omega_e t}}\right]F(t)\int_0^t F^*(t')dt'\right]\right\}\ . \tag{4.12d}$$

The solutions for g_1, g_2, and g_3 are identical to those obtained for zeroth order in X_e and are given by Eqs. (4.3a) and (4.3b). Upon defining $h_{Fi}\equiv g_i(\tau)/(-i\tau)$ the Floquet Hamiltonian of a rigid rotor driven by a general time-dependent potential of the form $f(t)\cos\theta$ is

$$H_{F1}=\mathbf{h}_F\cdot\mathbf{x} \tag{4.13}$$

to first order in the "anharmonicity," X_e. Although the term $h_{F9}1$ ordinarily contributes only an additive constant to the quasienergies, it should be kept in the present circumstances, because this constant depends on the point j_0 chosen for expanding the kinetic energy.

Two approaches can be taken to calculate the eigenvalues and eigenvectors of H_{F1}: in this section an approximate unitary transformation is found which diagonalizes the Floquet Hamiltonian to first order to X_e. An exact numerical diagonalization of H_{F1}, is presented in the following section.

Let us generalize the Floquet states defined by Eq. (4.5) by introducing the unitary transformation

$$|J_F\rangle=S_1S_2|j_0+n\rangle\ ,$$
$$S_1=e^{i\{e^{i\theta}[G+b(2\mathbf{n}-1)]+[G^*+b^*(2\mathbf{n}-1)]e^{-i\theta}\}}\ , \tag{4.14}$$
$$S_2=e^{i[c(e^{i\theta})^2+c^*(e^{-i\theta})^2]}\ ,$$

where b and c are assumed to have magnitude of order X_e. The evaluation of $H_{F1}|J_F\rangle$ proceeds by factoring S_1 and S_2 into products of exponential operators and then interchanging the operator ordering of $\mathbf{n}$, and $\mathbf{n}^2$, with each of the factors; thus

$$S_1=e^{ie^{i\theta}[G+b(2\mathbf{n}-1)]}e^{i[G^*+b^*(2\mathbf{n}-1)]e^{-i\theta}}e^{i[2(Gb^*+G^*b)]}$$

using the approximation that

$$[e^{i\theta}(G+b(2\mathbf{n}-1)),(G^*+b^*(2\mathbf{n}-1))e^{-i\theta}]\approx 2Gb^*+2G^*b$$

to first order in X_e. The operator reordering is accomplished by repeated application of Eqs. (4.6) and (4.7), keeping in mind that in the spirit of the first-order treatment only terms proportional to X_e^0 and X_e are kept. The result is that

$$\mathbf{n}|J_F\rangle=\{n-ie^{i\theta}[G+b(2\mathbf{n}-1)]$$
$$+i[G^*+b^*(2\mathbf{n}-1)]e^{-i\theta}$$
$$-i2c(e^{i\theta})^2+i2c^*(e^{-i\theta})^2$$
$$-2(Gb^*+G^*b)\}S_1S_2|j_0+n\rangle\ , \tag{4.15}$$

$$\mathbf{n}^2|J_F\rangle=[n^2-iGe^{i\theta}(2\mathbf{n}-1)+iG^*(2\mathbf{n}-1)e^{-i\theta}$$
$$+G^2(e^{i\theta})^2+G^{*2}(e^{-i\theta})^2$$
$$-2GG^*]S_1S_2|j_0+n\rangle\ . \tag{4.16}$$

A comparison of Eqs. (4.15) and (4.16) to $H_{F1}|J_F\rangle$ reveals that by choosing

$$G=-ih_{F2}/\Omega_e\ , \tag{4.17}$$

$$b=(-ih_{F5}-\Omega_e X_e G)/\Omega_e\ , \tag{4.18}$$

$$c=-i(h_{F7}+\Omega_e X_e G^2)/(2\Omega_e)\ , \tag{4.19}$$

the terms proportional $e^{\pm i\theta}$ and $(e^{\pm i\theta})^2$ in Eqs. (4.15) and (4.16) cancel with the corresponding terms in H_{F1}, leaving

$$H_{F1}|J_F\rangle=[C_0+n\Omega_e+n^2\Omega_e X_e+h_{F9}$$
$$-2\Omega_e X_e GG^*-2\Omega_e(Gb^*+G^*b)]|J_F\rangle\ , \tag{4.20}$$

which is diagonal in the $|J_F\rangle$ basis. The basis vectors are labeled by n, the displacement from the state j_0 about which the rotational kinetic energy is expanded. The factor G is identical with its previous definition in Sec. IV a. Appendix C lists the quantities h_{Fi}, G, b, and c for the sinusoidally driven and kicked rotors.

The term in brackets on the right side of Eq. (4.20)

represents the quasienergy. The sum of the first three terms reproduces the free-rotor energy levels. The final two terms represent the influence of the periodic driving force on the quasienergies, which was absent in the zeroth-order treatment. Away from any resonances, these quasienergies are in very good agreement, modulo $2\pi/\tau$, with the exact values; at resonance, the first-order quasienergies given by Eq. (4.20) exhibit singularities.

The impact of the first-order correction on the approximate Floquet eigenstates is evident from the comparison made in Fig. 3 to the exact states. The projection onto the free-rotor basis states is obtained from an extension of the corresponding zeroth-order quantities, Eq. (4.9), via

$$\begin{aligned}\langle j'|J_F\rangle &= \sum_k \langle j_0+n'|S_1|j_0+k\rangle\langle j_0+k|S_2|J_F\rangle \\ &= \sum_k e^{-i(\phi_1-\pi/2)(n'-k)} J_{(n'-k)}[2|G+b(n'+k)|] \\ &\quad \times e^{-i(\phi_2-\pi/2)(k-n)/2} J_{(k-n)/2}(2|c|)\ , \qquad (4.21)\end{aligned}$$

where the operator $\mathbf{n}$ in the exponent of S_1 has been replaced by $(n'+k+1)/2$ and the phase factors are defined, respectively, by $\exp(i\phi_1)=[G+b(n'+k)]/|G+b(n'+k)|$ and $\exp(i\phi_2)=c/|c|$ (values of G, b, and c are given in Appendix C). The contrast between the first-order states in Fig. 3 and the corresponding zeroth-order states in Fig. 1 shows that the correction for the rotational "anharmonicity" introduces the required asymmetry about the state $j'=J_F$ and provides very good agreement with the exact numerical results.

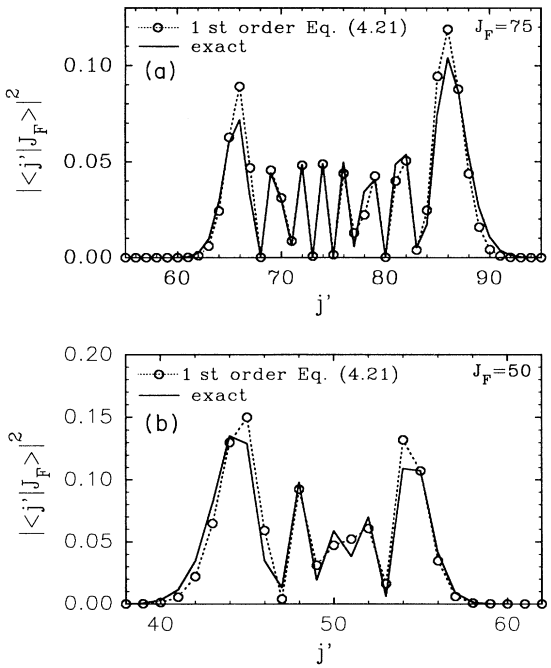

FIG. 3. Comparison of the first-order approximate Floquet eigenstates to exact numerical results for a sinusoidally driven rotor. (a) The parameters are $j_0=75$, $\Omega=0.5$ cm^{-1}, $m=0$, $\varepsilon=500$ cm^{-1}, and $\omega=100$ cm^{-1}. (b) The parameters are $j_0=50$, $\Omega=0.5$ cm^{-1}, $m=0$, $\varepsilon=400$ cm^{-1}, and $\omega=27.5$ cm^{-1}.

The effect of the "anharmonicity" can also be seen by comparing the first-order average rotational kinetic energy, of the Floquet states given by

$$\begin{aligned}\langle E_{\rm rot}\rangle_1 &= \langle J_F|\Omega j^2|J_F\rangle \\ &= C_0+\Omega_e[n+2(Gb^*+G^*b)] \\ &\quad +\Omega_e X_e(n^2+2GG^*)\ , \qquad (4.22)\end{aligned}$$

to the corresponding exact values. This is illustrated in Fig. 4 for the sinusoidally driven and kicked rotors (dotted lines). The agreement is very good except in the regions for which the "resonance" condition $\Omega_e\tau=m2\pi$ is nearly met. In the latter regions, the analytical predictions diverge, whereas the exact calculations reveal a sudden onset to a region in which the average kinetic energies of the Floquet states are nearly degenerate.

C. Removing the singularities

A persistant problem with both the zeroth- and first-order Floquet Hamiltonians is the presence of the singularities arising under the "resonance" condition. These are not simply theoretical artifacts, since they occur in situations where the exact calculations reveal a significant broadening of the Floquet eigenstates. This section explores a unitary transformation of the forced-rotor Hamiltonian into an interaction representation that, at least

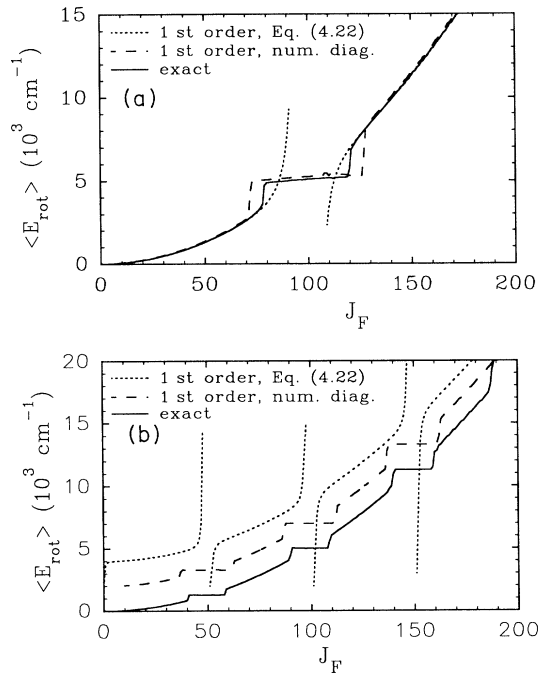

FIG. 4. Comparison of exact average kinetic energies for $|J_F\rangle$ to predictions from Eq. (4.22) and to values obtained from numerical diagonalization of H_{I1}. The rotational constant is $\Omega=0.5$ cm^{-1} and $m=0$. (a) Sinusoidally driven rotor with $\varepsilon=500$ cm^{-1} and $\omega=100$ cm^{-1}. (b) Periodically kicked rotor with $\theta=\pi$ and $\tau=\frac{2}{3}$ ps. Constant values of 2×10^3 and 4×10^3 cm^{-1} have been added to the numerical first- and analytical first-order traces, respectively, in order to separate the traces.

locally, removes the singularities and provides a Floquet Hamiltonian that remains finite.

In order to make use of Floquet's theorem, the transformation must be chosen so as to produce a periodically-time-dependent Hamiltonian in the interaction frame. The unitary operator

$$U_I = \exp(im\omega \mathbf{n}t) , \tag{4.23}$$

where $\tau = 2\pi/\omega$ represents the period of the driving force, fulfills this requirement. Because $e^{\pm i\theta}$ are "eigenoperators" of $\mathbf{n}$ under the commutation product, the phase operators transform into

$$U_I e^{\pm i\theta} U_I^{\dagger} = e^{\mp im\omega t} e^{\pm i\theta}$$

in the interaction frame, and the forced-rotor Hamiltonian $\mathcal{H}$ becomes

$$\mathcal{H}_I = (\Omega_e - m\omega)\mathbf{n} + \Omega_e X_e \mathbf{n}^2 + f(t) C_m(j_0)(e^{-im\omega t} e^{i\theta} + e^{im\omega t} e^{-i\theta}) . \tag{4.24}$$

The interesting feature about this result is that the forced-rotor Hamiltonian in the interaction frame has a form very similar to the original Schrödinger frame version; one needs only to make the replacements

$$\Omega_e \rightarrow (\Omega_e - m\omega) , \tag{4.25a}$$

$$X_e \rightarrow \Omega_e/(\Omega_e - m\omega) X_e , \tag{4.25b}$$

$$f(t) \rightarrow e^{\mp im\omega t} f(t) \tag{4.25c}$$

to convert from one frame to the other ($-$ and $+$ are used when making replacements into g_i for lowering and raising operators, respectively). The implication of this is that the interaction frame Floquet Hamiltonian, to either zeroth or first order, can be obtained simply by making the above replacements in the Lie coefficients h_{Fi} derived in Secs. IV a and IV b.

An important consequence of the above frame transformation is that it has no material effect on the quasienergies or on the *approximate* Floquet eigenstates. An examination of the quantities G, b, and c given by Eqs. (4.17)–(4.19) reveals that they are invariant to the replacements made in Eqs. (4.25a)–(4.25c). Because these quantities define the approximate Floquet eigenstates $|J_F\rangle$, these states are invariant to the frame transformation. Introducing the replacements of Eqs. (4.25a)–(4.25c) into the zeroth- and first-order quasienergies reveals that these quantities are shifted by an amount $m\omega$ in energy. Thus, modulo ω, they too are invariant to the transformation.

The frame transformation produces the zeroth-order Floquet Hamiltonian

$$H_{I0} = (\Omega_e - m\omega)(\mathbf{n} + iGe^{i\theta} - iG^* e^{-i\theta}) \tag{4.26}$$

in the interaction frame. Unlike its Schrödinger frame counterpart, H_{I0} remains finite as $\omega \rightarrow \Omega_e/m$; in fact,

$$\lim_{\omega \rightarrow \Omega_e/m} H_{I0} = \frac{1}{\tau}(Fe^{i\theta} + F^* e^{-i\theta}) , \tag{4.26a}$$

where F is the component of the driving force at Ω_e. Instead of the singularities observed in the Schrödinger frame we find that, under the "resonance" condition, the Floquet states become eigenstates of the phase operators, i.e.,

$$|J_F\rangle = \frac{1}{\sqrt{2\pi}} \sum_j e^{-ij\phi} |j\rangle , \tag{4.27}$$

in the basis of angular momentum states. Note that these Floquet states are labeled by the continuous index ϕ and that they are infinitely broad. They are not normalizable in the ordinary sense; rather

$$\langle \phi' | \phi \rangle = \frac{1}{2\pi} \sum_k e^{-ik(\phi - \phi')} = \sum_k \delta(\phi - \phi' - 2\pi k) ,$$

requiring that ϕ and ϕ' be constrained to the interval $(0, 2\pi)$. The corresponding quasienergies are given by

$$\lim_{\omega \rightarrow \Omega_e/m} E_{I0} = \frac{2}{\tau} \mathrm{Re}(Fe^{i\phi}) .$$

Such a transition from discrete to continuous quasienergies is cited as signaling the onset of quantum chaos [21]. Indeed, a linearized rotor initially in state j_0 will under resonance eventually spread to all other angular-momentum states.

The advantage of the interaction frame Floquet Hamiltonian is that it remains finite even when the resonance condition $m\omega = \Omega_e$ is met. This allows numerical diagonalization of the first-order Floquet Hamiltonian [obtained by substituting Eq. (4.25) into Eq. (4.13)]. Figure 5 compares the exact overlap probability of $|J_F = 70\rangle$ with the free-rotor basis states to the first-order predictions. Expanding the rotor kinetic energy about $j_0 = 80$ gives

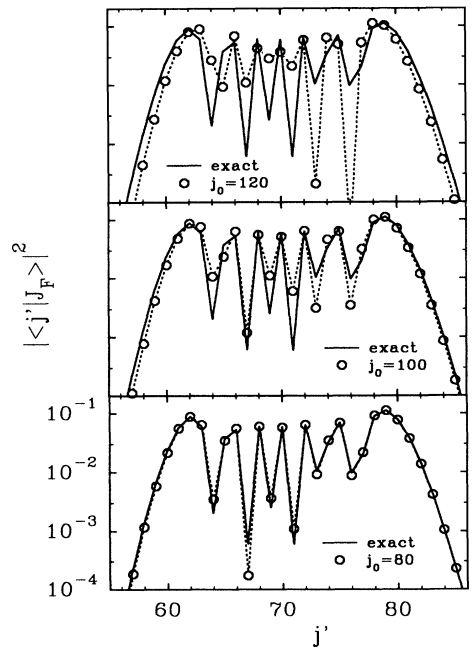

FIG. 5. Comparison of exact and first-order values for the overlap probability of $|J_F = 70\rangle$ with the free-rotor basis obtained by numerical diagonalization of H_{I1}. Three values of the expansion point j_0 are illustrated. The parameters are $\Omega = 0.5$ cm^{-1}, $m = 0$, and $\varepsilon = 500$ cm^{-1}, $\omega = 100$ cm^{-1}.

excellent agreement with the exact values. The accuracy of the predictions slowly decreases as the expansion point is chosen further from the center of the Floquet state; even at $j_0=120$ a qualitatively accurate picture of the Floquet state remains.

The question of what effect the anharmonicity has on the widths and energies of the Floquet states is addressed by Figs. 4, 6, and 7. In each case, numerical diagonalization of H_{I1} yields a qualitatively correct prediction of the exact results. When $\Omega_e \neq m\omega$, the average kinetic energy of the Floquet states is essentially the same as for the free-rotor states. These Floquet states are relatively localized, but their widths increase as a resonance is approached. As $\Omega_e \rightarrow m\omega$, assuming that the driving force has a nonzero Fourier component at frequency $m\omega$, the kinetic energy reaches a plateau [38]. Concomitantly, the Floquet states undergo a dramatic increase in width (Fig. 6). If multiple resonance regions exist, as they do for the kicked rotor, then corresponding to each region there is a plateau in the average kinetic energy (Fig. 4) and a sharp transition to delocalized Floquet states (Fig. 7). Precisely the same characteristics were observed in the numerical studies of Lin and Reichl [24]. These results can be explained as follows. In the harmonic case, a resonance region would engulf the entire free-rotor basis. This delocalization of Floquet states is suppressed by the anharmonicity, which limits the delocalized states to the resonance regions. The size of these regions is, in turn, governed by the strength of the driving field [39] relative to the magnitude of the anharmonicity. When the resonance regions are sufficiently close together and the driving field is sufficiently intense, the resonance regions can overlap to yield delocalized Floquet states.

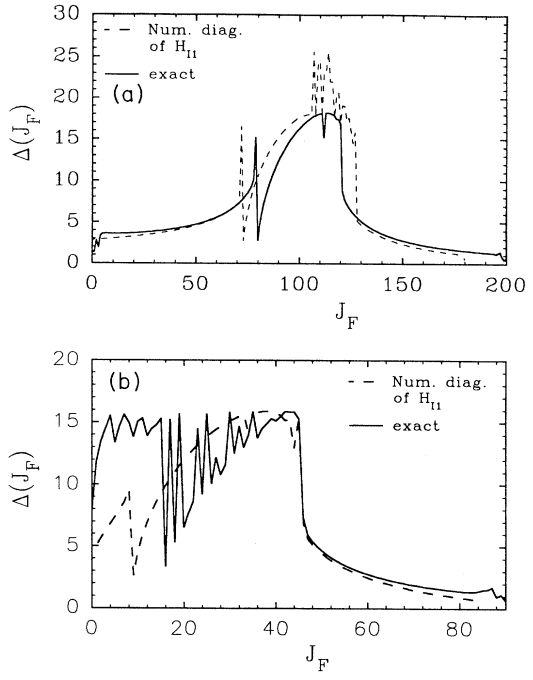

FIG. 6. Widths of the Floquet states predicted from numerical diagonalization of H_{I1} vs exact values for the sinusoidally driven rotor. The rotational constant is $\Omega=0.5$ cm^{-1} and $m=0$. (a) $\varepsilon=500$ cm^{-1}, $\omega=100$ cm^{-1}, and $j_0=100$ for all J_F. (b) $\varepsilon=400$ cm^{-1}, $\omega=27.5$ cm^{-1}, and $j_0=50$ for all J_F.

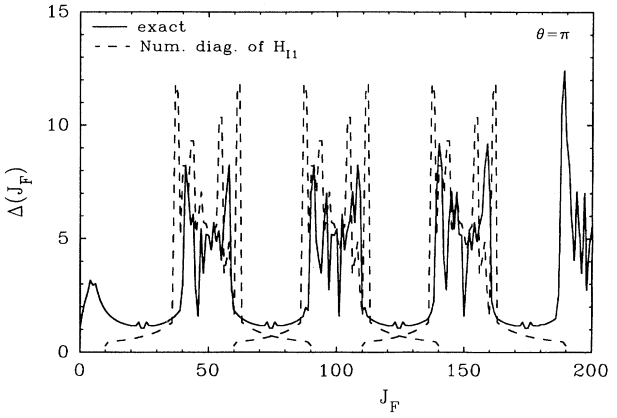

FIG. 7. Widths of the Floquet states predicted from numerical diagonalization of H_{I1} vs exact values for the periodically kicked rotor. The rotational constant is $\Omega=0.5$ cm^{-1}, $m=0$, $\theta=\pi$, and $\tau=\frac{2}{3}$ ps. The first-order predictions are made in three intervals with $j_0=50$, 100, and 150, respectively.

V. CONCLUSION

This paper has presented an analytical solution for the Floquet eigenstates and quasienergies of a rigid rotor subject to an arbitrary time-dependent force by introducing a perturbation expansion in the anharmonicity of the rotational energy-level spacing. Within this scheme both the forced rotor and its associated Floquet Hamiltonian become elements of a finite-dimensional Lie algebra. A system of differential equations for the Lie coefficients of the Floquet Hamiltonian is derived from the time-dependent Schrödinger equation by assuming an exponential form for the evolution operator. Although nonlinear in general, these equations admit analytical solutions to both zeroth- and first-order in X_e for the forced-rotor problem.

To zeroth order, the J_Fth Floquet state is composed of a Bessel-function distribution, $J_{\Delta n}(2|G|)$, of free-rotor states. This state is, in general, exponentially localized for large $|\Delta n|$, leading to a quasiperiodic motion. At resonance, however, the quasienergy spectrum becomes continuous and the Floquet states are completely delocalized.

The first-order correction for the anharmonicity in rotational energy levels introduces a number of new features. The symmetry in the zeroth-order Bessel function distribution is replaced by an asymmetry which accurately reproduces the exact Floquet states. Away from a resonance region, the Floquet states are relatively narrow. As resonance is approached, there is a sharp transition to much broader states, with nearly degenerate average kinetic energies. These states are remnants of the delocalized states that exist for the harmonic system, but are suppressed by the anharmonicity. When multiple resonance regions exist, a group of broad Floquet states is associated with each region and relatively narrow states are found in between. As the separation between resonance regions decreases, these regions will begin to

coalesce, leading to delocalized Floquet states.

The emphasis of the present paper has been on the Floquet eigenstates of the forced rotor. In two respects, a more general problem has actually been solved. For deriving the Floquet Hamiltonian, a periodic driving force was assumed and attention was directed to the evolution operator over one period. In fact, the evolution operator $e^{\Omega(t)}$, derived in Sec. IV, is valid even when the driving force is aperiodic and at arbitrary time. Secondly, the perturbation expansion in the anharmonicity that has been presented can be applied equally well to other anharmonic systems.

APPENDIX A

This appendix provides a solution for $g_i(t)$, the Lie coefficients used to expand $\Omega(t)$ in the exponential evolution operator, to first order in the anharmonicity. The commutation relations between the basis elements of $\mathcal{L}_1$ to zeroth order in X_e are identical to those for $\mathcal{L}_0$, and are given by Eqs. (2.4) and (2.5). Nonzero commutators to first order in X_e are given by

$$[\mathbf{n}^2, e^{i\theta}] = -e^{i\theta}(2\mathbf{n}-1) ,$$

$$[\mathbf{n}^2, e^{-i\theta}] = (2\mathbf{n}-1)e^{-i\theta} ,$$

$$[\mathbf{n}, e^{i\theta}(2\mathbf{n}-1)] = -e^{i\theta}(2\mathbf{n}-1) ,$$

$$[\mathbf{n}, (2\mathbf{n}-1)e^{-i\theta}] = (2\mathbf{n}-1)e^{-i\theta} ,$$

$$[e^{i\theta}, e^{i\theta}(2\mathbf{n}-1)] = 2(e^{i\theta})^2 ,$$

$$[e^{i\theta}, (2\mathbf{n}-1)e^{-i\theta}] = 2 ,$$

$$[e^{-i\theta}, e^{i\theta}(2\mathbf{n}-1)] = -2 ,$$

$$[e^{-i\theta}, (2\mathbf{n}-1)e^{-i\theta}] = -2(e^{-i\theta})^2 .$$

Using these commutation relations one constructs the matrix S from Eq. (3.8) and the expansion $\Omega(t) = \sum_i g_i(t) x_i$, where x_i are the basis elements of $\mathcal{L}_1$, with the result that

$$S = \begin{bmatrix}
0 & g_2 & -g_3 & 0 & g_5 & -g_6 & 2g_7 & -2g_8 & 0 \\
0 & -g_1 & 0 & 0 & -g_4 & 0 & -2g_5 & 0 & -2g_6 \\
0 & 0 & g_1 & 0 & 0 & g_4 & 0 & 2g_6 & 2g_5 \\
0 & 0 & 0 & 0 & g_2 & -g_3 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & -g_1 & 0 & 2g_2 & 0 & -2g_3 \\
0 & 0 & 0 & 0 & 0 & g_1 & 0 & -2g_3 & 2g_2 \\
0 & 0 & 0 & 0 & 0 & 0 & -2g_1 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 2g_1 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{bmatrix} .$$

Because it is an upper triangular matrix, the eigenvalues and generalized eigenvectors of S are easily obtained. The result is

$$s_1 = 0, \quad \langle s_1| = (g_1, g_2, g_3, g_4, g_5, g_6, g_7, g_8, 0), \quad \text{order 1} ,$$

$$s_2 = -g_1, \quad \langle s_2| = [0, 1, 0, 0, 0, 0, 2(g_2 g_4 - g_1 g_5)/g_1^2, 0, 2(g_1 g_6 - g_3 g_4)/g_1^2], \quad \text{order 2} ,$$

$$s_3 = g_1, \quad \langle s_3| = [0, 0, 1, 0, 0, 0, 0, 2(g_3 g_4 - g_1 g_6)/g_1^2, 2(g_1 g_5 - g_2 g_4)/g_1^2], \quad \text{order 2} ,$$

$$s_4 = 0, \quad \langle s_4| = (0, 0, 0, 1, g_2/g_1, g_3/g_1, g_2^2/g_1^2, g_3^2/g_1^2, 0), \quad \text{order 1} ,$$

$$s_5 = -g_1, \quad \langle s_5| = (0, 0, 0, 0, g_1, 0, 2g_2, 0, 2g_3), \quad \text{order 1} ,$$

$$s_6 = g_1, \quad \langle s_6| = (0, 0, 0, 0, 0, g_1, 0, 2g_3, 2g_2), \quad \text{order 1} ,$$

$$s_7 = -2g_1, \quad \langle s_7| = (0, 0, 0, 0, 0, 0, 1, 0, 0), \quad \text{order 1} ,$$

$$s_8 = 2g_1, \quad \langle s_8| = (0, 0, 0, 0, 0, 0, 0, 1, 0), \quad \text{order 1} ,$$

$$s_9 = 0, \quad \langle s_9| = (0, 0, 0, 0, 0, 0, 0, 0, 1), \quad \text{order 1} ,$$

where the eigenvectors have not been normalized. The solution of Eq. (3.11) for the functions $g_i(t)$ also requires the coefficients b_i used to expand the Hamiltonian in terms of the generalized eigenvectors of S, i.e., $-i(\mathcal{H}_0 + \mathcal{H}_1) = \mathbf{h}\cdot\mathbf{x} = \sum_i b_i \langle s_i| \cdot \mathbf{x}$. These coefficients are given by

$$b_1 = -i\Omega_e/g_1 ,$$

$$b_2 = -i(C_m f - \Omega_e g_2/g_1) ,$$

$$b_3 = -i(C_m f - \Omega_e g_3/g_1) ,$$

$$b_4=-i(\Omega_e X_e-\Omega_e g_4/g_1)\ ,$$

$$b_5=i(\Omega_e g_5+\Omega_e X_e g_2-\Omega_e g_4 g_2/g_1)/g_1^2\ ,$$

$$b_6=i(\Omega_e g_6+\Omega_e X_e g_3-\Omega_e g_4 g_3/g_1)/g_1^2\ ,$$

$$b_7=-i[\Omega_e X_e g_2^2-\Omega_e g_1 g_7+\Omega_e g_4 g_2^2/g_1-2(g_2 g_4-g_1 g_5)C_m f]/g_1^2\ ,$$

$$b_8=-i[\Omega_e X_e g_3^2-\Omega_e g_1 g_8+\Omega_e g_4 g_3^2/g_1-2(g_3 g_4-g_1 g_6)C_m f]/g_1^2\ ,$$

$$b_9=-i[4\Omega_e X_e g_2 g_3-2C_m f(g_1 g_5-g_2 g_4-g_3 g_4+g_1 g_6)+4\Omega_e(g_1 g_3 g_5-2g_2 g_3 g_4+g_1 g_2 g_6)/g_1]/g_1^2$$

to first order in X_e. Coefficients b_1-b_3 are the same as those used to solve for the zeroth-order Floquet states in Sec. IV a. The substitution of s_i, $\langle s_i|$, and b_i into Eq. (3.11) yields a nested sequence of nine coupled differential equations for g_i. The first three are identical to Eqs. (4.2a) and (4.2b) and negative the complex conjugate of Eq. (4.2b), respectively. Of the remaining six, two are redundant because the Hermiticity of the Hamiltonian guarantees that $g_6=-g_5^*$ and $g_8=-g_7^*$. Thus, the four equations needed to complete the first-order treatment are

$$\frac{dg_4}{dt}=-i\Omega_e X_e\ ,$$

$$\frac{dg_5}{dt}=g_5\left[\frac{1}{t}-\frac{i\Omega_e}{e^{i\Omega_e t}-1}\right]+\Omega_e X_e(C_m t f(t)-ig_2)\frac{e^{i\Omega_e t}-1-i\Omega_e t e^{i\Omega_e t}}{(e^{i\Omega_e t}-1)^2}\ ,$$

$$\frac{dg_7}{dt}=g_7\left[\frac{1}{t}-\frac{i2\Omega_e}{e^{i2\Omega_e t}-1}\right]+\left[\frac{2X_e g_2^2}{t}+2iC_m f(t)(X_e g_2-g_5)\right]\frac{1}{e^{i\Omega_e t}+1}$$
$$+2X_e g_2\left[\frac{g_2}{t}+iC_m f(t)\right]\frac{e^{i\Omega_e t}-1-i\Omega_e t e^{i\Omega_e t}}{(e^{i\Omega_e t}-1)^2}\ ,$$

$$\frac{dg_9}{dt}=2i\ \mathrm{Im}\left[2iX_e g_2^*\left[\frac{g_2}{t}+iC_m f(t)\right]\frac{\Omega_e t e^{i\Omega_e t}}{(e^{i\Omega_e t}-1)^2}-\frac{4\,\mathrm{Re}(g_2^* g_5)+2iC_m t f(t)g_5^*}{t(e^{i\Omega_e t}-1)}\right.$$
$$\left.-\frac{2C_m f(t)\Omega_e t(g_5-X_e g_2)-i2\Omega_e X_e g_2 g_2^*+i4\Omega_e g_2 g_5^*}{\Omega_e^2 t^2}\right]\ .$$

It is interesting to note that the equations for g_2 and g_5 are related by $dg_5/dt=\Omega_e X_e d^2 g_2/dt\,d\Omega_e$. As found in the zeroth-order case, the equation for dg_i/dt depends only on those g_j with $j\le i$; therefore, the above equations uncouple. While the procedure is cumbersome, these equations can be solved and yield the functions $g_i(t)$ listed in Eqs. (4.3a) and (4.3b) and Eqs. (4.12a)–(4.12d).

APPENDIX B

Exact Floquet eigenstates are determined by numerical integration of the time-dependent Schrödinger equation for the forced rotor. The evolution operator over one cycle is computed using the split propagator method [40–42],

$$U(\tau)=\prod_k e^{-i\Omega J^2\Delta t/2}$$
$$\times\exp\left[-i\int_{t_k}^{t_{k+1}}dt' f(t')\cos\theta\right]e^{-i\Omega J^2\Delta t/2}\ ,$$

and its eigenvalues and eigenvectors are determined via standard numerical procedures. By the relation $U(\tau)=\exp(-iH_F\tau)$, these eigenvectors are identical to those of H_F. Because of the ambiguity modulo $2\pi/\tau$ in the quasienergies, however, there remains the question of how to match the numerical Floquet eigenvectors to the corresponding analytic solutions developed in Sec. IV. This is accomplished by comparing the average rotational kinetic energy of the exact $|J_F\rangle$, computed from $\langle E_{\mathrm{rot}}\rangle=\sum_j\Omega j(j+1)|\langle j|J_F\rangle|^2$, to $\langle E_{\mathrm{rot}}\rangle_1$ defined by Eq. (4.22) and associating those states with nearly equal kinetic energies.

APPENDIX C

Table I lists the Lie coefficients h_{Fi} and the quantities G, b, and c for the examples of the sinusoidally driven rotor and the kicked rotor. These have been evaluated from Eqs. (4.3) and (4.12), assuming that $f(t)=\varepsilon\sin(\omega t)$ and $f(t)=\theta\sum\delta(t-(2k+1)\tau/2)$, respectively, with the definition that $h_{Fi}\equiv ig_i(\tau)/\tau$. Note that the replacements given by Eqs. (4.25a)–(4.25c) cannot be introduced directly into the expressions in Table I in order to obtain the interaction frame counterparts h_{Ii}. Instead, these substitutions must be inserted into Eqs. (4.3a) and (4.3b) and Eqs. (4.12a)–(4.12d) and these equations reevaluated in order to derive the interaction frame Floquet Hamiltonian.

TABLE I. The Lie coefficients h_{Fi} and the quantities G, b, and c for the examples of the sinusoidally driven and kicked rotors.

Coefficient	$f(t)=\varepsilon\sin(\omega t)$	$f(t)=\theta\sum_{k=0}^{\infty}\delta(t-(2k+1)\tau/2)$
h_{F1}	Ω_e	Ω_e
$h_{F2}=h_{F3}^*$	$\dfrac{i\varepsilon C_m\omega\Omega_e}{\omega^2-\Omega_e^2}$	$\dfrac{\theta C_m\Omega_e}{2\sin(\Omega_e\tau/2)}$
h_{F4}	$\Omega_e X_e$	$\Omega_e X_e$
$h_{F5}=h_{F6}^*$	$\dfrac{i\varepsilon C_m\omega\Omega_e X_e(\omega^2+\Omega_e^2)}{(\omega^2-\Omega_e^2)^2}$	$\dfrac{\theta C_m\Omega_e X_e(2-2\cos\Omega_e\tau-\Omega_e\tau\sin\Omega_e\tau}{8\sin^3(\Omega_e\tau/2)}$
$h_{F7}=h_{F8}^*$	$\dfrac{-\varepsilon^2 C_m^2\omega^2\Omega_e X_e(3\omega^2+\Omega_e^2)}{2(\omega^2-\Omega_e^2)^3}$	$\dfrac{\theta^2 C_m^2\Omega_e X_e(\sin\Omega_e\tau-\Omega_e\tau)}{4\sin^2(\Omega_e\tau/2)\sin\Omega_e\tau}$
h_{F9}	$\dfrac{\varepsilon^2 C_m^2\Omega_e X_e(3\omega^4+6\omega^2\Omega_e^2-\Omega_e^4)}{(\omega^2-\Omega_e^2)^3}$	$\dfrac{\theta^2 C_m^2\Omega_e X_e(2-2\cos\Omega_e\tau-\Omega_e\tau\sin\Omega_e 1}{4\sin^4\Omega_e\tau/2}$
G	$\dfrac{\varepsilon C_m\omega}{\omega^2-\Omega_e^2}$	$\dfrac{-i\theta C_m}{2\sin(\Omega_e\tau/2)}$
b	$\dfrac{2\varepsilon C_m\omega\Omega_e^2 X_e}{(\omega^2-\Omega_e^2)^2}$	$\dfrac{i\theta C_m\Omega_e X_e\tau\sin\Omega_e\tau}{4\sin^3(\Omega_e\tau/2)}$
c	$\dfrac{i\varepsilon^2 C_m^2\omega^2 X_e(\omega^2+3\Omega_e^2)}{4(\omega^2-\Omega_e^2)^3}$	$\dfrac{i\theta^2 C_m^2\Omega_e X_e\tau}{8\sin\Omega_e\tau\sin^2(\Omega_e\tau/2)}$

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