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Action-angle variables in quantum mechanics

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Conventional quantum mechanical treatments of many systems have worked with coordinates and momenta that are not canonically conjugate. In this work it is shown how the quantum expressions may be reformulated in terms of the canonical set of action-angle variables, and specific examples of the harmonic oscillator, linear rotor, and triaxial rotor are presented. When expressed in these terms, the quantum mechanics take on a form which can be directly related to analogous results from classical mechanics. In addition, it becomes possible to express the Hamiltonian in the minimum number of coordinates. It is also shown that the common assumption of an exponential form for the overlap of canonical coordinate and momentum eigenstates is false for an asymmetric rotor. This has important implications for the quantization rules applicable to nonseparable systems.

I. INTRODUCTION

A mechanical system can be described in terms of physical variables such as Cartesian coordinates and rotation angles. When the system is integrable, an alternative formulation in classical mechanics represents the dynamics in terms of action-angle variables—canonical coordinates and momenta for which the motion is explicitly periodic.[1,2] In the case of a linear rotor, for example, the classical momenta in action-angle variables would be the magnitude of the total angular momentum and its projection on a space-fixed axis. Since both of these are constants of the motion, the classical dynamics is particularly simple in this representation. The standard quantum mechanical treatment of the same problem keeps the same momenta but uses coordinates (the polar and azimuthal angles) which are not canonically conjugate to them. Earlier work[3-11] has considered the problem of defining quantum mechanical operators which are analogous to the angle variables of classical mechanics. This paper describes in general how the quantum mechanics may be reformulated in the angle variables which are conjugate to the usual quantized momenta (action variables).

The action-angle variable representation is particularly attractive because the correspondence between the classical and quantum expressions is very direct. Classical S-matrix theory[12] has effectively used this relationship to obtain a semiclassical approximation to molecular scattering problems. In addition, these variables play a central role in the semiclassical quantization of bound state systems.[13] The quantum mechanical description in terms of canonically conjugate coordinates and momenta will also be shown to be considerably simpler than the conventional approach.

Nonintegrable systems represent perhaps the largest area of potential applications for the quantum action-angle variables. For example, since the coordinates conjugate to constants of the motion (such as the total angular momentum) are cyclic,[1] the Hamiltonian operator in this representation will contain the minimum number of coordinates. This can be of practical benefit for direct numerical integration of the Schrödinger equation. For instance, the computational labor of the finite element method[14] is made considerably smaller when the number of variables is reduced. An application to the three-atom collision system will be the subject of a future publication[15]; in that case, the Hamiltonian can be reduced to the minimum number of four coordinates.

As mentioned above, semiclassical quantization schemes have leaned upon the assumed behavior of the quantum mechanical system when expressed in action-angle variables.[13] Some of the examples developed in this paper show that the assumptions about the quantum operators are not always correct. The asymmetric rotor represents an important and intriguing illustration of this point. For this system, it will be shown that the classical problem can be written entirely in conserved momenta and their conjugate coordinates, and that this description smoothly bridges the two symmetric top limits. Nevertheless, the overlap between conjugate coordinates and momentum quantum eigenstates cannot have the usual exponential form. The semiclassical quantization[16] of this system has required the introduction of "tunneling" corrections which have not been needed in other applications. This could have important implications for other systems which cannot be parameterized in terms of integer quantum numbers; the quantum numbers in these cases are only artificial indices for labeling the discrete states. Since most real physical and chemical problems are of this type, this initial study indicates that further work along these lines will be useful.

II. BASIC DEVELOPMENT

Consider a one-dimensional system with classical coordinate and momentum represented by q_c and p_c.[17] The spectrum of the quantum mechanical p operator will be taken to be continuous as will be the case if q_c is an unrestricted position variable. Since q and p form a canonical set, their commutator is

$$[q,p]=i\hbar \; . \tag{2.1}$$

As pointed out by Dirac,[18] this implies that the coordinate representation of p is

$$\langle q|p|q'\rangle=\left[-i\hbar\frac{\partial}{\partial q}+f(q)\right]\delta(q-q')\;, \tag{2.2}$$

[a)]Alfred P. Sloan Fellow and Camille and Henry Dreyfus Teacher–Scholar.

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where $f(q)$ is any real function of q. It is argued that the arbitrary phase factor in $|q\rangle$ can be chosen so that $f(q)$ is zero, and this leads to the usual result for $\langle q|p\rangle$:

$$\langle q|p\rangle = c\exp(ipq/\hbar)\ , \tag{2.3}$$

where c is usually chosen to give a δ function normalization.

Classical canonical transformations can be performed which take p_c, q_c over to another set P_c, Q_c, and we are particularly concerned with cases for which the quantum operator P has a discrete spectrum (with eigenvalues denoted by P_n). In all cases considered here, P is a local operator so that

$$\langle q|P|q'\rangle = P(q)\,\delta(q-q')\ .$$

The usual quantum mechanical eigenvalue problem in the coordinate representation then reduces to finding the $\langle q|P_n\rangle$ that satisfy

$$P(q)\langle q|P_n\rangle = P_n\langle q|P_n\rangle\ . \tag{2.4}$$

The overlaps $\langle q|P_n\rangle$ are the usual coordinate representation wave functions, and they are not in general "simple" periodic functions of q as in Eq. (2.3). Thus, the conventional treatment introduces the transformed momentum P but retains the old coordinate q. It is important to note that q and P are not canonical; the goal of this section is to represent the dynamics completely in terms of the canonical set Q, P. This is what is meant here as the quantum action-angle variables.

Some mathematical difficulties arise in this transformation of the coordinate and momentum. Since one can easily show that the spectrum of an operator is invariant under unitary transformations, the operators p and P are not related by such a transformation; the spectrum of p is continuous while that of P is discrete. In fact, it has been shown[5(a)] that p and P are related by a one-sided unitary transformation

$$P = U^\dagger p U$$

where

$$U^\dagger U = 1\ , \qquad UU^\dagger \neq 1\ .$$

This particular difficulty can be sidestepped by only defining the transformation implicitly without ever specifying U. It has also been questioned whether a Hermitian Q operator exists[4] which is canonical to P. Because P has a discrete spectrum, Q and P only form a Heisenberg pair[6] of canonical operators

$$[Q, P]|\psi\rangle = i\hbar|\psi\rangle\ , \tag{2.5}$$

where $|\psi\rangle$ is any element of a dense subset (the commutator space) on which the commutator is defined. In the more familiar case of a Weyl pair of canonical operators, the commutation relation is true for all $|\psi\rangle$ such that $|\psi\rangle$, $P|\psi\rangle$, and $Q|\psi\rangle$ belong to the domain of P and Q. However, in the Heisenberg sense, additional restrictions are placed on $|\psi\rangle$ to insure that the commutator is well behaved. These points will be dealt with more fully later in this section and in Appendix A, and the relevant commutator spaces for Eq. (2.5) will be defined for the specific examples to follow.

It is worthwhile to point out that standard quantum mechanical treatments in physical coordinates already include these problems. As an example, consider a free particle in three dimensions. If the Hamiltonian is written in Cartesian coordinates, all of the operators have continuous spectra. The angular part of the problem in the spherical polar coordinates θ, φ has, on the other hand, a discrete spectrum. Consequently, the transformation between these two systems is not unitary.[19] The z component of angular momentum l_z is conjugate to the azimuthal angle φ in the restricted Heisenberg sense. To show they are not a Weyl pair, consider matrix elements[4] of the commutator of φ and l_z:

$$\langle lm|[\varphi, l_z]|lm'\rangle = \langle lm|i\hbar|lm'\rangle = i\hbar\delta_{mm'}\ ,$$

but using the Hermiticity of l_z,

$$\langle lm|(\varphi l_z - l_z\varphi)|lm'\rangle = \hbar(m'-m)\langle lm|\varphi|lm'\rangle\ .$$

Setting $m=m'$, this leads to the incorrect result

$$i\hbar = \hbar(m-m)\langle lm|\varphi|lm\rangle = 0\ .$$

This paradox can be resolved[4] by noting that the usual coordinate representation of l_z, i.e., $-i\hbar(\partial/\partial\varphi)$, is only Hermitian when all functions of φ that enter are periodic with period 2π. Since φ itself does not have this periodic property, combinations of φ and $i\hbar(\partial/\partial\varphi)$ (as in the commutator) are not Hermitian. In the usual applications, only trigonometric functions of φ enter, and the difficulty is removed when considering $\exp(\pm i\varphi)$ rather than φ as the basic operator. The commutator space for φ and $-i\hbar(\partial/\partial\varphi)$, in the sense of Eq. (2.5), must be such that $\varphi\psi(\varphi)$ is periodic[6] or $\psi(\varphi)$ is the set of all functions of φ analytic on the unit circle with the additional property

$$\psi(\varphi = 2\pi) = 0\ .$$

Despite these mathematical questions associated with the use of spherical polar coordinates, if proper care is exercised, the dynamics can be described in either set without changing any quantities of physical interest.

When $[P, Q]$ are canonical in the sense of Eq. (2.5), one might think that this leads to the analog of Eq. (2.3):

$$\langle Q|P_n\rangle = c\exp(iP_nQ/\hbar)\ , \tag{2.6}$$

but this may not always be true. If the system is periodic in Q with period ξ, it is necessary that P_n be an integer multiple of $2\pi\hbar/\xi$ in order for Eq. (2.6) to be single valued. As will be seen later, the asymmetric rotor is one example for which the known quantum values of P_n do not satisfy this property. As a result, in the analog of Eq. (2.2), i.e.,

$$\langle Q|P|Q'\rangle = \left[-i\hbar\frac{\partial}{\partial Q} + f(Q)\right]\langle Q|Q'\rangle\ , \tag{2.7}$$

there are cases in which $f(Q)$ is not zero and Eq. (2.6) does not follow. This point will be discussed in more detail with regard to the asymmetric rotor in Sec. III C.

Let us assume for the present that the form given in Eq. (2.6) is correct and that the solutions of Eq. (2.4) are known. As discussed above, this implies that P_n is an integer multiple of $2\pi\hbar/\xi$, which also yields that these overlaps are orthogonal on any closed interval of length ξ:

$$\int_Q^{Q+\xi} dQ' \langle P_n | Q' \rangle \langle Q' | P_m \rangle = \delta_{nm} |c|^2 \xi \ . \tag{2.8}$$

Because of the periodic property, we can restrict Q to the interval $[0, \xi]$, and the choice $|c|^2 = 1/\xi$ then makes $\langle Q | P_n \rangle$ orthonormal on this interval. The transformation between q and Q is then easily obtained from the completeness of the eigenstates

$$\langle q | Q \rangle = \sum_n \langle q | P_n \rangle \langle P_n | Q \rangle = c^* \sum_n \langle q | P_n \rangle \exp(-iP_n Q/\hbar) \ . \tag{2.9}$$

The discrete Fourier transform of the conventional wave functions $\langle q | P_n \rangle$ is thus seen to play the role of the transformation from the physical coordinate q to the angle variable Q.

By analogy with the φ, l_z case discussed above, the commutator space in the coordinate representation consists of the set of analytic functions $\psi(Q) = \langle Q | \psi \rangle$ such that $\psi(\xi) = 0$. Note that this restriction is only necessary to establish the validity of Eq. (2.5), and other more general $|\psi\rangle$ are allowed when the Q operator occurs in functions with the proper period [i.e., $\exp(\pm 2\pi i Q/\xi)$].

It might occur that the *classical* Hamiltonian in terms of Q_c, P_c may have somewhat different properties than for the set q_c, p_c. For example, in the case of a harmonic oscillator, the action-angle variable classical Hamiltonian allows solutions over a wider range of energies than are admissable in Cartesian coordinates. In order to construct a proper quantum mechanical description (including a Hermitian Q operator) in action-angle variables, it is necessary to allow the quantum operators to have a spectrum consistent with the entire range admissable for the classical Hamiltonian in action-angle variables. Physical quantities in terms of q and p are then obtained from this larger quantum Hilbert space by projection. Appendix A treats these matters in detail using the harmonic oscillator as an illustration. Let us introduce an operator $\mathcal{P}$ which projects out the part of space relevant for the set q, p (which will be referred to as the physical subspace); thus, the allowed eigenvalues P_n in the physical subspace will not include all possible values on the complete space. The exact nature of $\mathcal{P}$ depends on the system in question, and specific cases are outlined in the examples in the following section. In this context, Eq. (2.9) must actually represent the quantity $\langle q | \mathcal{P} | Q \rangle$. An arbitrary operator which is known in terms of q can then be expressed in the Q representation by

$$\begin{aligned} \langle Q | \mathcal{P} A \mathcal{P} | Q' \rangle &= \int dq \int dq' \langle Q | \mathcal{P} | q \rangle \langle q | A | q' \rangle \langle q' | \mathcal{P} | Q' \rangle \\ &= |c|^2 \sum_n \sum_m \exp i(P_n Q - P_m Q')/\hbar \\ &\quad \times \int dq \, \langle q | P_n \rangle^* A(q) \langle q | P_m \rangle \ , \end{aligned} \tag{2.10}$$

where it has been assumed that A is local in the q representation

$$\langle q | A | q' \rangle = A(q) \delta(q - q') \ .$$

The completeness and orthogonality of the states $\mathcal{P} | Q \rangle$ in the physical subspace may now be investigated. For completeness consider

$$\begin{aligned} \int_0^\xi & dQ \langle q | \mathcal{P} | Q \rangle \langle Q | \mathcal{P} | q' \rangle \\ &= \sum_n \sum_m \langle q | P_n \rangle \langle P_m | q' \rangle |c|^2 \int_0^\xi dQ \exp i(P_m Q' - P_n Q)/\hbar \\ &= \sum_n \langle q | P_n \rangle \langle P_m | q' \rangle \delta_{nm} = \delta(q - q') \ , \end{aligned} \tag{2.11}$$

where the last step follows from the completeness of the functions $\langle q | P_n \rangle$ of q. Orthogonality comes from Eq. (2.10) after setting A to the unit operator

$$\begin{aligned} \langle Q | \mathcal{P}\mathcal{P} | Q' \rangle &= \langle Q | \mathcal{P} | Q' \rangle \\ &= |c|^2 \sum_n \sum_m \delta_{nm} \exp i(P_n Q - P_m Q')/\hbar \\ &= |c|^2 \sum_n \exp i P_n (Q - Q')/\hbar \ . \end{aligned} \tag{2.12}$$

Since $\langle Q | Q' \rangle$ is a delta function, Eq. (2.12) is not a delta function when $\mathcal{P}$ is not the unit operator on the larger space. Thus, the states $\mathcal{P} | Q \rangle$ are complete but not orthogonal in general.

The above difficulty can be dealt with by restricting the class of functions of Q which are admissable. If $|\psi\rangle$ is an arbitrary state and $g(Q)$ is defined by

$$g(Q) = \langle Q | \mathcal{P} | \psi \rangle \ ,$$

then $g(Q)$ may be expressed as a sum of $\langle Q | P_n \rangle$'s:

$$g(Q) = \sum_n \alpha_n \exp(i P_n Q/\hbar) \ , \tag{2.13}$$

where

$$\alpha_n = \langle P_n | \mathcal{P} | \psi \rangle \ .$$

The presence of the projection operator in the definition of α_n restricts the sum in Eq. (2.13) to only those values of n which are in the physical subspace. There are, therefore, three function spaces applicable to this discussion. The first of these is the one for the usual physical coordinates and requires no further comment (although it should be remembered that the structure will be different depending on whether or not these coordinates are angles). The second is that for the full space of $|Q\rangle$, and in the cases considered in this paper, it is the set of functions square integrable on the unit circle. Finally, the last is the projected subspace which consists of all functions which may be expanded as above. Now, from Eq. (2.12), the integral of $g(Q)$ with $\langle Q | \mathcal{P} | Q' \rangle$ is

$$\begin{aligned} \int_0^\xi dQ' g(Q') \langle Q | \mathcal{P} | Q' \rangle &= |c|^2 \sum_n \alpha_n \sum_m \exp i P_m Q/\hbar \\ &\quad \times \int_0^\xi dQ' \exp i(P_n - P_m) Q'/\hbar \\ &= \sum_n \alpha_n \exp i P_n Q/\hbar = g(Q) \ . \end{aligned}$$

Therefore, $\langle Q | \mathcal{P} | Q' \rangle$ is a "reproducing kernel" for an arbitrary function $g(Q)$ having the form of Eq. (2.13) and is effectively a delta function when the admissable functions of Q are restricted in this manner. Note that if the sum in Eq. (2.13) included values of n outside the physical subspace, $\langle Q|\mathcal{P}|Q'\rangle$ would not have the indicated behavior.

Since the above construction has rested on the exponential functional form in Eq. (2.6), it is worthwhile to investigate the circumstances when this is expected to be valid. Some information can be gained from classical mechanics which is extremely useful because the classical canonical transformations are well understood. In many systems, Q_c has the properties of a true angle; the motion is periodic in this variable with a constant period ξ. It is certainly to be expected that the classical periodicity carries over into quantum mechanics, or

$$\langle Q+k\xi|\psi\rangle=\langle Q|\psi\rangle ,$$

where $|\psi\rangle$ is an arbitrary state and k is an integer. Under these circumstances, the exponential form given in Eq. (2.6) is likely correct; the only possible objection is that $P_n=2\pi\hbar n/\xi$ does not give the correct quantum energy levels.

It may occur, however, that the classical "angle" variable Q_c exhibits a different behavior. For example, the classical period for Q_c may not be a constant; it may depend on the magnitudes of the conserved momenta. Such a case will actually be shown to arise later for the rigid asymmetric rotor. A basic objection to the exponential form can be raised when the period does depend on the constants of the motion. Either the overlaps $\langle Q|P_n\rangle$ will not have the correct periodic properties or else they will not be orthogonal. It should be stressed that this is a criterion that can be tested solely from the classical mechanics, and it is not necessary to know whether the true quantum energy levels are predicted.

When the classical period is dependent on P_c, it may be true that it is impossible to construct a canonical quantum Q operator as in Eq. (2.5). Alternatively, this operator might exist, but the function $f(Q)$ in Eq. (2.7) must then be unequal to zero. Perhaps the simplest example of a momentum dependent classical period is a one-dimensional system for which the classical motion is described by elliptic rather than circular functions. The general triaxial rotor problem will be shown in Sec. III C to essentially be of this type, and these matters will be discussed more fully with regard to this important case. In order to complete the quantum construction, it would be necessary to find an $f(Q)$ for Eq. (2.7) such that the solutions of the differential equation for $\langle Q|P_n\rangle$ have the correct periodic structure. This differential equation would also have to yield the proper energy levels, and it has been shown that this reduces to finding the roots of a continued fraction.[20] These points are worthy of further study.

III. SELECTED EXAMPLES

A. Harmonic oscillator

Several other workers have considered quantum action-angle variables for this very important system.[3-11] The present construction is somewhat different from the previous formulations, and it is set down in detail in Appendix A. This Appendix also deals with the mathematical objections that have been raised to the existence of a quantum angle operator.[3,4]

The classical Hamiltonian in Cartesian coordinates is

$$H(p,q)=p^2/2m+m\omega^2q^2/2 . \tag{3.1}$$

A canonical transformation to the set n, q_n (the analogs of P_c, Q_c in the previous section) is obtained from the F_2 type generating function[1]

$$F_2(q,n)=(n+\hbar/2)\sin^{-1}[q\sqrt{m\omega/(2n+\hbar)}] + q/2[(2n+\hbar)m\omega-m^2\omega^2q^2]^{1/2} , \tag{3.2}$$

which gives for the classical coordinate q_n the result

$$q_n=\sin^{-1}[q\sqrt{m\omega/(2n+\hbar)}] . \tag{3.3}$$

Note that the additional constant term of $\hbar/2$ appears in the above expressions for the purpose of a more direct comparison of quantum and classical mechanics. The Hamiltonian in the new set becomes

$$H(n,q_n)=\omega(n+\hbar/2) \tag{3.4}$$

and q_n is thus a cyclic coordinate.

If one were to consider the Hamiltonian in Eq. (3.4) as separate and independent from its origin in Eq. (3.1), an interesting feature arises. The classical dynamics are perfectly well behaved for all real values (including negative ones) of the classical momentum n. Thus, Eq. (3.4) allows for proper negative energy solutions of the classical equations of motion while Eq. (3.1) is restricted to positive energies (in order to assure real coordinate and momentum). As discussed in Appendix A, this leads to the construction of quantum operators on a larger Hilbert space. The relevant projection operator $\mathcal{P}$ (as in the previous section) then projects out the positive energy solutions.

In quantum mechanics, the coordinate representation of the Hamiltonian operator is

$$H=-\hbar^2/2m\ \frac{\partial^2}{\partial q^2}+m\omega^2/2q^2 , \tag{3.5}$$

which has the well known normalized eigenstates $\varphi_n(q)=\langle q|n\rangle$ and the eigenvalues $\hbar\omega(n+\frac{1}{2})$, where n is a positive integer.[21] Thus, the standard treatment does introduce the new momentum n, as in the above classical expressions, but combines it with the old coordinate q which is not canonically conjugate to n. Applying the methods of the previous section, the transformation to the quantum coordinate q_n is given by

$$\langle q|\mathcal{P}|q_n\rangle=(2\pi)^{-1/2}\sum_{n'=0}^{\infty}\varphi_{n'}(q)\exp(-in'q_n) , \tag{3.6}$$

where the projection operator $\mathcal{P}$ is

$$\mathcal{P}=\sum_{n=0}^{\infty}|n\rangle\langle n|$$

(note that the sum only runs over *positive* values of n). The coordinate space operator corresponding to n is

$$\langle q_n|\mathcal{P}n\mathcal{P}|q_n'\rangle=(2\pi)^{-1}\sum_{n_1=0}^{\infty}\sum_{n_2=0}^{\infty} \times\exp i(n_1q_n-n_2q_n')\hbar n_1\delta_{n_1n_2}=\hbar\hat{n}\langle q_n|\mathcal{P}|q_n'\rangle , \tag{3.7}$$

in which

$$\hat{n} = -i \frac{\partial}{\partial q_n} \quad .$$

Therefore, the Hamiltonian in terms of q_n is

$$\langle q_n | \mathcal{P} H \mathcal{P} | q_n' \rangle = \hbar\omega(\hat{n} + \tfrac{1}{2}) \langle q_n | \mathcal{P} | q_n' \rangle \ . \tag{3.8}$$

It immediately follows from the above that

$$\hbar\hat{n} \langle q_n | n \rangle = \hbar n \langle q_n | n \rangle$$

and therefore the eigenfunctions may be expressed as

$$\langle q_n | n \rangle = (2\pi)^{-1/2} \exp(inq_n) \ . \tag{3.9}$$

The Hamiltonian operator and its eigenfunctions are thus seen to take on a particularly simple form when written in terms of the quantum action-angle variables. Since the classical period of q_n is 2π, the requirement that Eq. (3.9) be single valued gives directly the result that n is an integer.

A number of mathematical fine points are worthy of consideration in regard to the above development. It was seen in Sec. II that the class of functions $g(q_n)$ which are admissable must be restricted in order for the states $\mathcal{P}|q_n\rangle$ to be orthogonal. In this case, only functions which may be expressed as a sum containing only *positive* integers

$$g(q_n) = \sum_{n'=0}^{\infty} \alpha_{n'} \exp(in'q_n) \tag{3.10}$$

are allowed. In the conventional q, p picture, this is reminiscent of requiring that all proper functions of q be expandable in a complete set of the eigenfunctions $\varphi_n(q)$. It can then be seen that Eqs. (3.7)–(3.9) lead to the same results as in the usual q, p treatment. In particular, suppose that H' is given by

$$H' = H + V(q) \ , \tag{3.11}$$

where $V(q)$ is any function such that

$$V(q) = \sum_{n=0}^{\infty} \beta_n \varphi_n(q) \ .$$

From Eq. (2.10),

$$\begin{aligned}\langle q_n | \mathcal{P} V \mathcal{P} | q_n' \rangle &= (2\pi)^{-1} \sum_{n_1=0}^{\infty} \sum_{n_2=0}^{\infty} \exp i(n_1 q_n - n_2 q_n') \\ &\quad \times \int_{-\infty}^{\infty} dq \, \varphi_{n_1}^*(q) V(q) \varphi_{n_2}(q) \\ &= (2\pi)^{-1} \sum_{n_1=0}^{\infty} \sum_{n_2=0}^{\infty} V_{n_1 n_2} \exp i(n_1 q_n - n_2 q_n') \ . \end{aligned} \tag{3.12}$$

Now form the matrix of $\mathcal{P} H' \mathcal{P}$ in the basis of eigenstates of H given by Eq. (3.9):

$$\begin{aligned}\langle n | \mathcal{P} H' \mathcal{P} | n' \rangle &= \int_0^{2\pi} dq_n \int_0^{2\pi} dq_n' \langle n | q_n \rangle \langle q_n | \mathcal{P} H' \mathcal{P} | q_n' \rangle \langle q_n' | n' \rangle \\ &= \hbar\omega(n + \tfrac{1}{2}) \delta_{nn'} + \sum_{n_1 n_2} \delta_{nn_1} \delta_{n' n_2} V_{n_1 n_2} \\ &= \hbar\omega(n + \tfrac{1}{2}) \delta_{nn'} + V_{nn'} \ , \end{aligned} \tag{3.13}$$

where the following identity was used:

$$(2\pi)^{-1} \int_0^{2\pi} dq_n \exp i q_n (n_1 - n) = \delta_{nn_1} \ .$$

Thus, the matrix elements of $\mathcal{P} H' \mathcal{P}$ are the same in either the q, p or q_n, n treatments and therefore so are the eigenvalues and eigenvectors of $\mathcal{P} H' \mathcal{P}$.

For purposes of comparing with the classical canonical transformation, let us form the matrix of $\mathcal{P} q \mathcal{P}$ in the new representation. This is

$$\begin{aligned}\langle q_n | \mathcal{P} q \mathcal{P} | q_n' \rangle &= (2\pi)^{-1} \sum_{n_1=0}^{\infty} \sum_{n_2=0}^{\infty} \exp i(n_1 q_n - n_2 q_n') \\ &\quad \times i(\hbar/2m\omega)^{1/2} (\sqrt{n_1+1}\, \delta_{n_1 n_2 - 1} - \sqrt{n_1}\, \delta_{n_1 n_2 + 1}) \\ &= i(\hbar/2m\omega)^{1/2} [\exp(iq_n)\sqrt{\hat{n}+1} - \exp(-iq_n)\sqrt{\hat{n}}\,] \langle q_n | \mathcal{P} | q_n' \rangle \ . \end{aligned} \tag{3.14}$$

Before proceeding with the classical comparison, two points about Eq. (3.14) are worthy of consideration. Since the functions of q_n are restricted to the form of Eq. (3.10), the action of the square root operators is well defined:

$$\sqrt{\hat{n}+1} \sum_{n'=0}^{\infty} \alpha_{n'} \exp(in'q_n) = \sum_{n'=0}^{\infty} \alpha_{n'} \sqrt{n'+1} \exp(in'q_n) \ ,$$

and similar expressions will also be found to appear in the examples to follow. The two terms in brackets are seen to correspond to the raising and lowering operators[21] $a^\dagger$ and a, respectively:

$$\langle q_n | \mathcal{P} a^\dagger \mathcal{P} | q_n' \rangle = \exp(iq_n) \sqrt{\hat{n}+1} \langle q_n | \mathcal{P} | q_n' \rangle \ , \tag{3.15a}$$

$$\langle q_n | \mathcal{P} a \mathcal{P} | q_n' \rangle = \exp(-iq_n) \sqrt{\hat{n}} \langle q_n | \mathcal{P} | q_n' \rangle \ . \tag{3.15b}$$

It may easily be seen (by taking n, n' matrix elements) that

$$\exp(-iq_n)\sqrt{\hat{n}} = \sqrt{\hat{n}+1} \exp(-iq_n),$$

$$\exp(iq_n)\sqrt{\hat{n}+1} = \sqrt{\hat{n}} \exp(iq_n) \ ,$$

so that the two expressions are indeed compatible. These forms have been objected to on the grounds that $\exp(-iq_n)$ cannot be unitary,[3,4] and this difficulty is dealt with in Appendix A. Now if both of the operators in the square roots of Eq. (3.14) were $(\hat{n} + \frac{1}{2})^{1/2}$, the matrix element would become

$$\langle q_n | \mathcal{P} q \mathcal{P} | q_n' \rangle \simeq \sin q_n [(2\hbar/m\omega)(\hat{n} + \tfrac{1}{2})]^{1/2} \langle q_n | \mathcal{P} | q_n' \rangle \ , \tag{3.16}$$

while the classical result [from Eq. (3.3)] is

$$q = \sin q_n [(2\hbar/m\omega)(n/\hbar + \tfrac{1}{2})]^{1/2} \ . \tag{3.17}$$

The classical and quantum expressions are therefore easily seen to become equivalent in the limit of large quantum numbers.

B. Linear rotor

The quantum Hamiltonian in spherical polar coordinates is

$$H = (p_\theta^2 - i\hbar \cot\theta p_\theta + \csc^2\theta p_\varphi^2)/2I \ , \tag{3.18}$$

where I is the moment of inertia. In the standard treatment, the conserved momenta—the magnitude of the angular momentum l and its projection m on the space fixed z axis—are introduced. Thus, the representation of eigenstates $|lm\rangle$ of these momenta in the rotation angles θ and φ are the usual spherical harmonics $\langle \theta\varphi | lm \rangle = Y_{lm}(\theta, \varphi)$. As before, the angles θ and φ can be replaced by the coordinates q_l, q_m which are canonically conjugate to l and m.

One must first examine the classical mechanics in order to determine the nature of the projection operator $\mathcal{P}$ for this system. The classical Hamiltonian in the polar angles is

$$H=(\mathbf{l}\cdot\mathbf{l})/2I=(p_\theta^2+\csc^2\theta p_\varphi^2)/2I\ , \tag{3.19}$$

where p_φ $(=m)$ is just the z component of $\mathbf{l}$. When a canonical transformation is performed[22] to replace p_θ by l, the Hamiltonian in classical action-angle variables becomes

$$H=l^2/2I\ . \tag{3.20}$$

The equations of motion from Eq. (3.20) allow l to be negative and also permit m to be greater than l. Therefore, the projection operator is

$$\mathcal{P}=\sum_{l=0}^{\infty}\sum_{m=-l}^{l}|lm\rangle\langle lm|\ ,$$

which restores the restrictions on l and m inherent in Eq. (3.19) but not in Eq. (3.20).

Applying the general methods of Sec. II, the transformation matrix elements between the spherical angles and the angle variables are

$$\langle\theta\varphi|\mathcal{P}|q_lq_m\rangle=(2\pi)^{-1}\sum_{l'=0}^{\infty}\sum_{m'=-l'}^{l'}\exp[-i(l'q_l+m'q_m)]\,Y_{l'm'}(\theta,\varphi)\ , \tag{3.21}$$

so that the expression for an arbitrary local operator A known in terms of θ, φ is

$$\langle q_lq_m|\mathcal{P}A\mathcal{P}|q_l'q_m'\rangle=(2\pi)^{-2}\sum_{l_1m_1}\sum_{l_2m_2}\exp i(l_1q_l+m_1q_m-l_2q_l'-m_2q_m')\times\left[\int_{-1}^{1}d(\cos\theta)\int_0^{2\pi}d\varphi Y^*_{l_1m_1}(\theta,\varphi)A(\theta,\varphi)Y_{l_2m_2}(\theta,\varphi)\right]\ . \tag{3.22}$$

Thus, the major role of the spherical harmonics in this approach is to transform a function of θ and φ to one of q_l and q_m.

If $\hat{\mathbf{l}}$ is the vector angular momentum operator, it is readily seen from Eq. (3.22) that

$$\langle q_lq_m|\mathcal{P}(\hat{\mathbf{l}})^2\mathcal{P}|q_l'q_m'\rangle=(2\pi)^{-2}\sum_{l'm'}\hbar^2l'(l'+1)\exp i[l'(q_l-q_l')+m'(q_m-q_m')]=-\hbar^2\left(\frac{\partial^2}{\partial q_l^2}+i\frac{\partial}{\partial q_l}\right)\langle q_lq_m|\mathcal{P}|q_l'q_m'\rangle \tag{3.23a}$$

and

$$\langle q_lq_m|\mathcal{P}\hat{l}_z\mathcal{P}|q_l'q_m'\rangle=-i\hbar\frac{\partial}{\partial q_m}\langle q_lq_m|\mathcal{P}|q_l'q_m'\rangle\ . \tag{3.23b}$$

The extra $i(\partial/\partial q_l)$ term in Eq. (3.23a) gives rise to the eigenvalue being l^2+l rather than just l^2, and this is a consequence of the presence of the nonclassical quantity $-i\hbar\cot\theta p_\theta$ in the Hamiltonian (3.18). Also of interest are the raising and lowering operators $\hat{l}_\pm=\hat{l}_x\pm i\hat{l}_y$:

$$\langle q_lq_m|\mathcal{P}\hat{l}_\pm\mathcal{P}|q_l'q_m'\rangle=\hbar\exp(\pm iq_m)\times[\hat{l}(\hat{l}+1)-\hat{m}(\hat{m}\pm1)]^{1/2}\langle q_lq_m|\mathcal{P}|q_l'q_m'\rangle\ , \tag{3.24}$$

where $\hat{l}=-i(\partial/\partial q_l)$ and $\hat{m}=-i(\partial/\partial q_m)$. The classical expression analogous to Eq. (3.24) is

$$l_x\pm il_y=\exp(\pm iq_m)\sqrt{l^2-m^2} \tag{3.25}$$

and this is again the quantum result in the limit of large quantum numbers.

As seen above and also for the harmonic oscillator, one can directly compare quantum mechanical and classical mechanical expressions in action-angle variables. The difficulty of doing this in the standard treatment stems from the nature of the wave functions $Y_{lm}(\theta,\varphi)=\langle\theta\varphi|lm\rangle$. Since it involves old coordinates and new momenta, its best classical equivalent is a canonical transformation. Indeed, the semiclassical limit of $\langle\theta\varphi|lm\rangle$ is the exponential of the classical F_2 type generating function.[12] It is not possible to describe the classical dynamics in such a mixed set of coordinates and momenta and so the comparison must be indirect.

C. The triaxial rotor

This is the case of the rotation of a general rigid body, and for simplicity we shall work in the principal axis (PA) system of the rotor (the axes are denoted by x, y, z). The three rotational constants are

$$A=\hbar^2/2I_x\ ,\qquad B=\hbar^2/2I_y\ ,\qquad C=\hbar^2/2I_z\ ,$$

where I_x is the moment of inertia about the x principal axis and, by convention, $A\geq B\geq C$. In terms of the three Euler angles[1] θ, φ, ψ, the components of $\mathbf{j}$ (the total angular momentum vector) are given by

$$\mathbf{j}(\mathrm{PA})=\begin{pmatrix}\cos\psi p_\theta+\sin\psi\csc\theta p_\varphi-\cot\theta\sin\psi p_\psi\\-\sin\psi p_\theta+\cos\psi\csc\theta p_\varphi-\cot\theta\cos\psi p_\psi\\p_\psi\end{pmatrix} \tag{3.26}$$

and the Hamiltonian is

$$H=A/\hbar^2j_x^2+B/\hbar^2j_y^2+C/\hbar^2j_z^2\ . \tag{3.27}$$

In the case of an oblate symmetric top, $A=B$ and the three conserved action variables are j, m, k, where j is the magnitude of $\mathbf{j}$, m is its projection on the space-fixed (SF) z axis, and k is the projection of $\mathbf{j}$ on the z principal axis. For a prolate symmetric top, $B=C$ and k is replaced by the x component of $\mathbf{j}$(PA) as the third conserved momentum. The eigenstates of the oblate symmetric top Hamiltonian in the θ, φ, ψ representation are the well known representations of the rotation group[23]

$$\langle\theta\varphi\psi|jmk\rangle=\mathcal{D}^j_{mk}(\psi,\theta,\varphi)\ ,$$

with eigenvalues

$$Bj(j+1)-(B-C)k^2\ .$$

In the symmetric top limit, it remains true that

$$\langle q_jq_mq_k|jmk\rangle=(2\pi)^{-3/2}\exp i(jq_j+mq_m+kq_k)\ ,$$

but it will shortly be shown that this exponential form is no longer correct when the rotor is asymmetric. Given this expression, the $q_jq_mq_k$ representation of a general local operator $A(\theta, \varphi, \psi)$ can be obtained in a similar fashion to previous examples as

$$\langle q_j q_m q_k | \mathcal{P} A \mathcal{P} | q_j' q_m' q_k' \rangle = (2\pi)^{-3} \sum_{j_1 m_1 k_1} \sum_{j_2 m_2 k_2} \exp i(j_1 q_j + m_1 q_m + k_1 q_k - j_2 q_j' - m_2 q_m' - k_2 q_k') \times \left[\int_{-1}^{1} d(\cos\theta) \int_0^{2\pi} d\varphi \int_0^{2\pi} d\psi\, \mathfrak{D}_{m_1 k_1}^{j_1 *}(\psi, \theta, \varphi) A(\theta, \varphi, \psi) \mathfrak{D}_{m_2 k_2}^{j_2}(\psi, \theta, \varphi) \right], \tag{3.28}$$

which comes from the transformation elements

$$\langle \theta\varphi\psi | \mathcal{P} | q_j q_m q_k \rangle = (2\pi)^{-3/2} \sum_{j_1=0}^{\infty} \sum_{m_1=-j_1}^{j_1} \sum_{k_1=-j_1}^{j_1} \times \exp -i(j_1 q_j + m_1 q_m + k_1 q_k) \mathfrak{D}_{m_1 k_1}^{j_1}(\psi, \theta, \varphi) . \tag{3.29}$$

The projection operator $\mathcal{P}$ is the same form as for the linear rotor with the additional restriction that $k \le j$. A major difference between this case and the linear rotor is the existence of two sets of ladder operators

$$j_\pm(\mathrm{SF}) = j_x(\mathrm{SF}) \pm i j_y(\mathrm{SF}) ,$$

$$j_\pm(\mathrm{PA}) = j_x(\mathrm{PA}) \pm i j_y(\mathrm{PA}) .$$

From Eq. (3.28), the new coordinate representations of these operators are

$$\hat{j}_\pm(\mathrm{SF}) = \langle q_j q_m q_k | \mathcal{P} j_\pm(\mathrm{SF}) \mathcal{P} | q_j' q_m' q_k' \rangle / \hbar = \exp(\pm i q_m) [\hat{j}(\hat{j}+1) - \hat{m}(\hat{m} \pm 1)]^{1/2} \times \langle q_j q_m q_k | \mathcal{P} | q_j' q_m' q_k' \rangle , \tag{3.30a}$$

$$\hat{j}_\pm(\mathrm{PA}) = \langle q_j q_m q_k | \mathcal{P} j_\pm(\mathrm{PA}) \mathcal{P} | q_j' q_m' q_k' \rangle / \hbar = \exp(\pm i q_k) [\hat{j}(\hat{j}+1) - \hat{k}(\hat{k} \pm 1)]^{1/2} \times \langle q_j q_m q_k | \mathcal{P} | q_j' q_m' q_k' \rangle , \tag{3.30b}$$

where $\hat{j} = -i(\partial/\partial q_j)$, $\hat{m} = -i(\partial/\partial q_m)$, and $\hat{k} = -i(\partial/\partial q_k)$.

For a general asymmetric rotor in this representation, the Hamiltonian is

$$H = A[\hat{j}_+^2 + \hat{j}_-^2 + 2\hat{j}(\hat{j}+1) - 2\hat{k}^2]/4 - B[\hat{j}_+^2 + \hat{j}_-^2 - 2\hat{j}(\hat{j}+1) + 2\hat{k}^2]/4 + C\hat{k}^2 , \tag{3.31}$$

in which all the operators are understood to be in reference to the PA frame. In the oblate symmetric top limit, $A = B$ and it is easy to see that Eq. (3.31) yields the correct eigenvalues. Since the total angular momentum is conserved, $\hat{j}$ can be replaced by its constant eigenvalue (m does not appear in the PA frame), and then Eq. (3.31) reduces to an effectively one-dimensional operator in q_k. The operator $\hat{\eta}^2$, defined in Appendix B, plays an important role for the asymmetric rotor. In the coordinate representation, it is given by

$$2\hat{\eta}^2 = (1+\kappa)\hat{j}_z^2 - (1-\kappa)\hat{j}_x^2 = (1+\kappa)\hat{k}^2 - (1-\kappa)[\hat{j}_+^2 + \hat{j}_-^2 + 2\hat{j}(\hat{j}+1) - 2\hat{k}^2]/4 , \tag{3.32}$$

where κ is the asymmetry parameter $(2B - A - C)/(A - C)$. If one were to form the matrix of $\hat{\eta}^2$ in the $\langle q_k | k \rangle$ basis (j being held fixed), it is readily verified that the eigenvalues of this matrix yield the correct quantum energy levels. In particular, for $j = 1$ (although not generally true), the eigenvalues and eigenfunctions[24] can be obtained in closed form as

$$\eta^2(1_{10}) = (\kappa - 1)/2 , \quad \langle q_k | \eta \rangle = (2\pi)^{-1/2} ,$$

$$\eta^2(1_{11}) = \kappa , \quad \langle q_k | \eta \rangle = (\pi)^{-1/2} \cos q_k ,$$

$$\eta^2(1_{01}) = (\kappa + 1)/2 , \quad \langle q_k | \eta \rangle = (\pi)^{-1/2} \sin q_k .$$

(The eigenfunctions presented above result from combining the eigenvectors of the matrix with the exponential form for $\langle q_k | k \rangle$ given in this work.) These eigenvalues are labeled by the usual notation $(j_{k_{-1} k_{+1}})$, and it is somewhat surprising that these values are predicted correctly by setting $\hat{j}_x$ to k_{-1} and $\hat{j}_z$ to k_{+1} in Eq. (3.32).

The above development illustrates one of the areas of potential utility for the quantum action-angle variables. In the usual treatment, the Hamiltonian and the ladder operators for the asymmetric rotor are two-dimensional partial differential operators in the angles θ, ψ. However, without introducing any basis set expansion, full advantage has been taken of the conservation of total angular momentum to reduce Eq. (3.31) to a one-dimensional ordinary differential operator. Besides being conceptually simpler, the elimination of one coordinate greatly reduces the computational labor of directly integrating the Schrödinger equation. A future paper[15] will consider the practical aspects of dealing with operators of the type given in Eqs. (3.30a) and (3.30b).

Although Eq. (3.32) represents an effectively one-dimensional eigenvalue problem, the usual Bohr–Sommerfeld semiclassical quantization rule is not valid.[16] Two difficulties are the failure to give the correct number of distinct energy levels, and the inability to make a continuous transition between the symmetric top limits. In Appendix B, it is shown how the set q_η, η does make the transition smoothly in classical mechanics. The quantum limit of these classical expressions is therefore worthy of investigation.

Since q_η and η are canonical, the commutator of the operator is (presuming a q_η operator exists)

$$[q_\eta, \eta] = i\hbar \tag{3.33}$$

and this implies—as in Sec. II—that

$$\langle q_\eta | \eta | q_\eta' \rangle = \left[-i\hbar \frac{\partial}{\partial q_\eta} + f(q_\eta) \right] \langle q_\eta | q_\eta' \rangle . \tag{3.34}$$

The eigenstates $\langle q_\eta | \eta \rangle$ are then the solution of the one-dimensional differential equation

$$\left[-i\hbar \frac{\partial}{\partial q_\eta} + f(q_\eta) \right] \langle q_\eta | \eta \rangle = \eta \langle q_\eta | \eta \rangle \tag{3.35}$$

subject to the appropriate boundary conditions. However, the periodic properties of the classical motion introduce significant complications. The classical dynamics is described in terms of Jacobian elliptic functions[25] (see Appendix B) which are doubly periodic with periods that depend on the magnitudes of the classical momenta j, η. Although the classical motion only samples one of these periods, the successful semiclassical quantization[16] of this system has also required an action integral over the other period (tunneling or reflection contributions). This is in sharp contrast to the previous examples where the angle variables all had periods of 2π, independent of the action variables. If these periodicities are carried over into quantum mechanics and a q_η operator does exist, then it would seem to be true that $f(q_\eta)$ in Eq. (3.34) cannot be made to vanish. Were $f(q_\eta)$ to be zero, the unique (unnormalized) solution of Eq. (3.35) would be

$$\langle q_\eta | \eta \rangle = \exp(i\eta q_\eta/\hbar) \; . \tag{3.36}$$

For this to be single valued, it would have to be true that $\eta/(2\pi\hbar)$ be an integer multiple of the reciprocal of the period. Because this period depends upon η, this would not yield orthogonal eigenfunctions. The classical doubly periodic q_η dependence can only be obtained if $\langle q_\eta | \eta\rangle$ is an elliptic function rather than the exponential form above.

Unfortunately, there does not seem to be any straightforward procedure for determining the unknown function $f(q_\eta)$. Although the classical mechanics is solvable in closed form, there is no known method for constructing a parallel quantum formulation (if it indeed exists). The successful modified semiclassical quantization of this system[16] suggests, however, that the classical behavior carries over into quantum mechanics at least to a good approximation. A resolution of these questions will shed some light on the more general class of problems for which the classical motion is given by elliptic functions, with resulting momentum dependent periods. The development presented here also indicates that the standard semiclassical quantization rules[13] will need to be modified for systems of this type. Since it has been shown that the quantum energy levels for one-dimensional classical elliptic systems are the roots of a continued fraction,[20] this may well provide some insight into those large numbers of quantum problems for which the eigenvalues cannot be simply parameterized in terms of integer quantum numbers. These points are worthy of further investigation.

IV. CONCLUSION

Conventional quantum mechanics has worked in a representation where the momenta are analogous to the action variables of classical mechanics but the coordinates are not their conjugate angle variables. It was shown in this work how the quantum dynamics may be reformulated in a canonical set. The resulting coordinate space operators are often of a simpler form than the standard expression in terms of physical coordinates. In many important cases, the eigenstates have a simple exponential form. The more complex functions, such as the spherical harmonics, are no longer needed as eigenfunctions; they only play a role in transforming physical coordinate functions into the action-angle variables.

The examples developed in this paper are just a few of the integrable systems which can be expressed in this manner. Other cases whose quantum solution is known, such as the Morse oscillator, may be treated in a similar fashion. However, nonintegrable systems should represent the largest class of problems for which the action-angle variables are useful. There the variables would be chosen in accord with some reference Hamiltonian H_0 which describes an important part of the problem. Consider, for example, a system of coupled oscillators; the introduction of new momenta which are the energies of the individual oscillators is reminiscent of the usual procedure of expanding in a basis of eigenstates of H_0. In scattering problems, the coupling vanishes in the asymptotic regime, and so it is natural to use variables appropriate to the free Hamiltonian. The angular parts of a scattering system can also be simplified by the introduction of action variables corresponding to the various angular momenta.

In addition, when the quantum action-angle variables are used, full advantage can be taken of the known constants of the motion. For example, the rigid asymmetric rotor Hamiltonian can be written as a two-dimensional partial differential operator in the conventional Euler angles θ and ψ. As was demonstrated in Sec. III C, the eigenvalue problem can be reduced to a one-dimensional ordinary differential equation in the variable q_k. This simplification can be of enormous practical significance for techniques based on the direct numerical integration of the Schrödinger equation. It will be shown in a future publication[15] that the appropriate action-angle variables reduce the atom–diatom scattering problem from five coordinates to the minimum number (four). This lowering of the dimensionality should greatly decrease the computational labor of numerical partial differential equation techniques (e.g., the finite element method).

Much of the work on semiclassical quantization has relied upon the overlap between canonical coordinate and momentum eigenstates having an exponential form. For the important case of an asymmetric rotor, where the classical dynamics may be expressed in closed form, it was demonstrated that this assumption is false. Whenever the classical "angle" variables have nonconstant periods, it was shown that the quantum mechanics does not permit an exponential overlap. This behavior may indeed be typical of the quantum mechanical problems where the "quantum numbers" are nothing more than indices to label the discrete states. Further work in this area may greatly improve our understanding of nonseparable quantum systems.

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APPENDIX A: QUANTUM TREATMENT OF THE HARMONIC OSCILLATOR IN TERMS OF ACTION-ANGLE VARIABLES

The existence of Hermitian operators corresponding to the classical harmonic oscillator action-angle variables has been questioned in the literature.[3,4] Similar mathematical objections could be raised with regard to the other examples developed in this paper. Therefore, it will be shown in detail how the apparent problems may be dealt with for the harmonic oscillator and also how the formulation is related to previous work. Many of the quantum constructions were guided by some suggestions of Newton.[26]

Consider first the *classical* harmonic oscillator Hamiltonian as given in Eq. (3.1):

$$H(p,q)=p^2/2m+m\omega^2q^2/2\ . \tag{A1}$$

The classical generating function of the F_2 type[1]

$$F_2(q,n)=(n+\hbar/2)\sin^{-1}[q\sqrt{m\omega/(2n+\hbar)}]+q/2[(2n+\hbar)m\omega-m^2\omega^2q^2]^{1/2} \tag{A2}$$

converts this to a Hamiltonian in terms of action-angle variables

$$\tilde{H}(n,q_n)=\omega(n+\hbar/_2)\ . \tag{A3}$$

Note that the classical action variable has been written as $n+\hbar/2$ in anticipation of the quantum developments to follow. An important point here is that H and $\tilde{H}$ do not represent the same dynamical system. Examining Eq. (A1), the restriction that p and q be real yields that the energy must be greater than or equal to zero. The equations of motion obtained from Eq. (A3) are

$$\begin{aligned}\dot{n}&=\partial\tilde{H}/\partial q_n=0\ ,\\ \dot{q}_n&=\partial\tilde{H}/\partial n=\omega\ ,\end{aligned} \tag{A4}$$

and these are perfectly well behaved for *all* real values of n and q_n. If one considers the Hamiltonian in Eq. (A3) independently from its origins, both positive and negative energies are classically allowed. Therefore, $\tilde{H}$ is only equivalent to H if the additional (external) restriction of positive energies is imposed on $\tilde{H}$. These observations about the classical mechanics are crucial to the construction of a proper quantum mechanical theory in terms of action-angle variables.

By analogy with the classical situation, let us consider the quantum Hamiltonian operator

$$\tilde{H}=\omega(\hat{n}+\hbar/2)\ , \tag{A5}$$

where the operator $\hat{n}$ has a canonically conjugate operator $\hat{q}_n$. The eigenstates of these operators are denoted by $|n\rangle$ and $|q_n\rangle$:

$$\hat{n}|n\rangle=\hbar n|n\rangle\ , \tag{A6a}$$

$$\hat{q}_n|q_n\rangle=q_n|q_n\rangle\ . \tag{A6b}$$

From the classical description, it will be further presumed that $\hat{q}_n$ is an angle operator; for any state $|\psi\rangle$ in the Hilbert space, it is true that

$$\langle q_n+2\pi k|\psi\rangle=\langle q_n|\psi\rangle \tag{A7}$$

when k is any integer. The eigenvalues of $\hat{q}_n$ are continuous and the states $|q_n\rangle$ are complete

$$\int dq_n|q_n\rangle\langle q_n|=1 \tag{A8}$$

and orthonormal

$$\langle q_n|q_n'\rangle=\sum_{k=-\infty}^{\infty}\delta(q_n-q_n'+2\pi k)\ . \tag{A9}$$

Because of the periodic property (A7), without any loss of generality we may restrict q_n to all real numbers in $[0,2\pi]$ and Eq. (A9) is then a single δ function.

Since $\hat{q}_n$ is an angle operator, the same problems arise as were described for the φ, l_z pair in Sec. II. The operators $\hat{n}$ and $\hat{q}_n$ therefore constitute a Heisenberg rather than a Weyl canonical pair

$$[\hat{q}_n,\hat{n}]\,|\psi\rangle=i\hbar|\psi\rangle \tag{A10a}$$

only for states $|\psi\rangle$ such that $\psi(q_n)=\langle q_n|\psi\rangle$ vanishes when q_n is equal to 2π (the commutator space). If the unitary operator $\mathcal{E}=\exp(i\hat{q}_n)$ were regarded as the basic operator with the commutation relation

$$[n,\mathcal{E}]=\mathcal{E}\ , \tag{A10b}$$

this restriction of the commutator space would not need to be considered. The entire development that follows could then be expressed in terms of $\mathcal{E}$ and $\mathcal{E}^\dagger$ with $|q_n\rangle$ being a quasieigenvector of $\mathcal{E}$:

$$\mathcal{E}|q_n\rangle=e^{iq_n}|q_n\rangle\ .$$

However, it may be noted that Eq. (A10a) is compatible with Eq. (A10b), and the canonical commutation relation for $\hat{q}_n$ alone is conceptually useful. The difficulties associated with the commutator space never really arise since only periodic functions of $\hat{q}_n$ actually occur in practice. However, it should be pointed out that the Heisenberg form of the commutator cannot be used to derive uncertainty relations for $\hat{n}$, $\hat{q}_n$ in the usual way, and Eq. (A10b) must be used to obtain the correct uncertainty products.[26]

The commutation relation given in Eq. (A10a) [or equivalently Eq. (A10b)] implies that the q_n representation of $\hat{n}$ is

$$\langle q_n|\hat{n}|q_n'\rangle=-i\hbar\frac{\partial}{\partial q_n}\delta(q_n-q_n')\ . \tag{A11}$$

From this one obtains

$$\langle q_n|n\rangle=c\exp(inq_n)\ ,$$

and the periodic boundary conditions give the requirement that *n be a positive or negative integer*. Thus, the operator $\hat{n}$ has a discrete spectrum and

$$\langle n|n'\rangle=\delta_{nn'}\ . \tag{A12}$$

The q_n representation of the Hamiltonian is now

$$\langle q_n|\tilde{H}|q_n'\rangle=\hbar\omega\left(-i\frac{\partial}{\partial q_n}+\frac{1}{2}\right)\delta(q_n-q_n') \tag{A13}$$

and the normalized wave functions are

$$\langle q_n|n\rangle=(2\pi)^{-1/2}\exp(inq_n)\ . \tag{A14}$$

Therefore, the function space appropriate to the q_n representation is the space spanned by functions having the form of Eq. (A14), which is the same as the space of square-integrable functions on the unit circle.

As in the classical problem, $\tilde{H}$ is not the Hamiltonian for the harmonic oscillator; it allows for both positive and negative energies. The analog of the classical restriction to positive energies is provided by the projection operator $\mathcal{P}$ defined by

$$\mathcal{P}=\sum_{n=0}^{\infty} |n\rangle\langle n| \,, \tag{A15}$$

and this has a complement $\mathcal{Q}$

$$\mathcal{Q}=1-\mathcal{P}=\sum_{n=-\infty}^{-1} |n\rangle\langle n| =\sum_{n=0}^{\infty} |-n-1\rangle\langle -n-1| \,. \tag{A16}$$

The space will now be divided into two subspaces represented by the column matrices

$$\begin{pmatrix}\mathcal{P}|\psi\rangle \\ \mathcal{Q}|\psi\rangle\end{pmatrix},$$

where $|\psi\rangle$ is any state in the complete space. An arbitrary operator $\mathcal{O}$ becomes the matrix

$$\mathcal{O}=\begin{pmatrix}\mathcal{P}\mathcal{O}\mathcal{P} & \mathcal{P}\mathcal{O}\mathcal{Q} \\ \mathcal{Q}\mathcal{O}\mathcal{P} & \mathcal{Q}\mathcal{O}\mathcal{Q}\end{pmatrix} \;: \qquad \text{A17)}$$

Thus, since

$$\exp(-i\hat{q}_n)|n\rangle=|n-1\rangle \,,$$

this operator may be expressed as

$$\exp(-i\hat{q}_n)=\begin{pmatrix}\sum_{n=0}^{\infty}|n\rangle\langle n+1| & 0 \\ |-1\rangle\langle 0| & \sum_{n=0}^{\infty}|-n-2\rangle\langle -n-1|\end{pmatrix} \tag{A18}$$

and similarly

$$\hat{n}=\hbar\begin{pmatrix}\sum_{n=0}^{\infty} n|n\rangle\langle n| & 0 \\ 0 & -\sum_{n=0}^{\infty}(n+1)|-n-1\rangle\langle -n-1|\end{pmatrix} \tag{A19}$$

It may easily be verified from the completeness of the states $|n\rangle$ that $\exp(-i\hat{q}_n)$ is unitary; this requires the consideration of all values (positive and negative) of n.

The introduction of some additional operators permits the construction of the harmonic oscillator Hamiltonian. Define the operators $\mathfrak{M}$ and $\mathcal{A}$ by

$$\mathfrak{M}=\begin{pmatrix}\sum_{n=0}^{\infty} n^{1/2}|n\rangle\langle n| & 0 \\ 0 & \sum_{n=0}^{\infty}(n+1)^{1/2}|-n-1\rangle\langle -n-1|\end{pmatrix} \tag{A20}$$

$$\mathcal{A}=\exp(-i\hat{q}_n)\mathfrak{M}=\begin{pmatrix}\sum_{n=0}^{\infty}(n+1)^{1/2}|n\rangle\langle n+1| & 0 \\ 0 & \sum_{n=0}^{\infty}(n+1)^{1/2}|-n-2\rangle\langle -n-1|\end{pmatrix} \tag{A21}$$

The operators $\mathcal{A}$ and $\mathcal{A}^\dagger$ do not commute; from Eq. (A21), their commutator is

$$[\mathcal{A},\mathcal{A}^\dagger]=\begin{pmatrix}\mathcal{P} & 0 \\ 0 & -\mathcal{Q}\end{pmatrix}=\mathcal{R} \,. \tag{A22}$$

A new canonical set of Hermitian coordinate and momentum operators can be constructed from $\mathcal{A}$; these are given by

$$\hat{q}=i(\hbar/2m\omega)^{1/2}\mathcal{R}(\mathcal{A}-\mathcal{A}^\dagger) \,, \tag{A23a}$$

$$\hat{p}=(\hbar m\omega/2)^{1/2}(\mathcal{A}+\mathcal{A}^\dagger) \,. \tag{A23b}$$

(Note that $\mathcal{R}$ commutes with $\mathcal{A}$ and $\mathcal{A}^\dagger$.) Define the operator H by

$$H=\hat{p}^2/2m+m\omega^2\hat{q}^2/2=\hbar\omega[\mathcal{A}^\dagger\mathcal{A}+\tfrac{1}{2}\mathcal{R}] \,, \tag{A24}$$

and this may be compared with $\tilde{H}$ (using $\hat{n}=\hbar\mathcal{R}\mathfrak{M}^2$):

$$\tilde{H}=\omega[\hat{n}+\hbar/2]=\hbar\omega[\mathcal{R}\mathfrak{M}^2+\tfrac{1}{2}]=\hbar\omega\mathcal{R}[\mathcal{A}^\dagger\mathcal{A}+\tfrac{1}{2}\mathcal{R}]=\mathcal{R}H \,. \tag{A25}$$

The above equation can also be expressed in terms of the operators $\hat{p}$ and $\hat{q}$ by

$$\tilde{H}=\hat{p}\mathcal{R}\hat{p}/2m+m\omega^2\hat{q}\mathcal{R}\hat{q}/2 \,. \tag{A26}$$

It can be seen from the above that the system is not periodic in $\hat{p}$ or $\hat{q}$ and that there are no restrictions on their eigenvalues. Thus, these operators have a spectrum consisting of all real numbers, and the function space appropriate to the operators consists of all functions square integrable on the real line.

One can now construct the eigenstates $|q\rangle$ of the operator $\hat{q}$:

$$\hat{q}|q\rangle=q|q\rangle \,.$$

Expanding $|q\rangle$ in terms of the states $|n\rangle$ gives

$$q|q\rangle=\hat{q}\sum_{n=-\infty}^{\infty} c_n(q)|n\rangle$$

$$=i(\hbar/2m\omega)^{1/2}\sum_{n=-\infty}^{\infty} c_n(q)\mathcal{R}(\mathcal{A}-\mathcal{A}^\dagger)|n\rangle \,,$$

which yields the recurrence relations

$$qc_0=ic_1(\hbar/2m\omega)^{1/2} \,, \tag{A27a}$$

$$-qc_{-1}=ic_{-2}(\hbar/2m\omega)^{1/2} \,, \tag{A27b}$$

$$qc_n=i(\hbar/2m\omega)^{1/2}[n^{1/2}c_{n-1}-(n+1)^{1/2}c_{n+1}] \quad (n>0) \,, \tag{A27c}$$

$$-qc_{-n}=i(\hbar/2m\omega)^{1/2}[n^{1/2}c_{-n-1}-(n-1)^{1/2}c_{-n+1}]$$
$$(n<-1)\,. \tag{A27d}$$

It may be noted that there are two independent sets of coefficients because the operator $\hat{q}$ does not connect the two subspaces. One can readily see that for positive n the c's have the form

$$c_n(z)=(-i)^n P_n(z)\,c_0(z)\,,$$

where $z=q(\hbar/m\omega)^{1/2}$ and P_n is a polynomial. The P_n's satisfy the recurrence ($P_0=1,\ P_1=z$)

$$(n+1)^{1/2}P_{n+1}=2^{1/2}zP_n-n^{1/2}P_{n-1}\,, \tag{A28}$$

which yields that

$$P_n(z)=\frac{2^{-n/2}}{\sqrt{n!}}\,H_n(z)\,,$$

where H_n is the usual Hermite polynomial.[27] In a similar fashion, one obtains

$$c_{-n-1}=(-i)^n\frac{2^{-n/2}}{\sqrt{n!}}\,H_n(-z)c_{-1}(z)\,.$$

The two functions c_0 and c_{-1} may be obtained from the completeness requirement for $|q\rangle$:

$$\int_{-\infty}^{\infty}dq\,|q\rangle\langle q|$$
$$=\mathcal{P}+\mathcal{Q}=\sum_{n=0}^{\infty}\sum_{n'=0}^{\infty}|n\rangle\langle n'|\left[\int_{-\infty}^{\infty}dqP_n(q)P_{n'}(q)|c_0(q)|^2\right]$$
$$+\sum_{n=0}^{\infty}\sum_{n'=0}^{\infty}|-n-1\rangle\langle -n'-1|$$
$$\times\left[\int_{-\infty}^{\infty}dqP_n(-q)P_{n'}(-q)|c_{-1}(q)|^2\right]\,, \tag{A29}$$

which is equivalent to the two conditions

$$\int_{-\infty}^{\infty}dqP_n(q)P_{n'}(q)|c_0(q)|^2=\delta_{nn'}\,,$$
$$\int_{-\infty}^{\infty}dq\,P_n(-q)P_{n'}(-q)|c_{-1}(q)|^2=\delta_{nn'}\,.$$

From the known properties of the Hermite polynomials, this leads to the unique choices for c_0 and c_{-1}:

$$c_0(z)=c_{-1}(z)=(m\omega/\pi\hbar)^{1/4}\exp(-z^2/2)\,.$$

If the wave functions of the usual harmonic oscillator[21] are denoted by $\varphi_n(q)$, the expression for $|q\rangle$ is finally given by

$$|q\rangle=\sum_{n=0}^{\infty}[\varphi_n(q)|n\rangle+\varphi_n(-q)|-n-1\rangle]\,. \tag{A30}$$

It may also be verified from the above that the states $|q\rangle$ are orthogonal

$$\langle q|q'\rangle=\delta(q-q')(\mathcal{P}+\mathcal{Q})\,. \tag{A31}$$

The q representation of the operators are readily obtained from Eq. (A30). First of all, the canonical momentum is

$$\langle q|\hat{p}|q'\rangle=-i\hbar\begin{pmatrix}\frac{\partial}{\partial q} & 0\\ 0 & \frac{\partial}{\partial q}\end{pmatrix}\delta(q-q')\,, \tag{A32}$$

which is clearly compatible with the canonical commutation relation

$$[\hat{q},\hat{p}]=i\hbar\,.$$

The two Hamiltonian operators in the q representation are

$$\langle q|H|q'\rangle=\begin{pmatrix}-\hbar^2/2m\,\frac{\partial^2}{\partial q^2}+m\omega^2q^2/2 & 0\\ 0 & -\hbar^2/2m\,\frac{\partial^2}{\partial q^2}+m\omega^2q^2/2\end{pmatrix}\delta(q-q')\,, \tag{A33a}$$

$$\langle q|\tilde{H}|q'\rangle=\begin{pmatrix}-\hbar^2/2m\,\frac{\partial^2}{\partial q^2}+m\omega^2q^2/2 & 0\\ 0 & +\hbar^2/2m\,\frac{\partial^2}{\partial q^2}-m\omega^2q^2/2\end{pmatrix}\delta(q-q')\,, \tag{A33b}$$

so that H can be immediately identified with the harmonic oscillator Hamiltonian. Although $\tilde{H}$ is not the same as H, these two operators do have the same projection on the positive energy subspace. This projection is reminiscent of the classical mechanical restriction of positive energies on Eq. (A3).

All of the results of Ref. 4 can be obtained from the above formulation by projection onto the $\mathcal{P}$ subspace. In particular, it is readily seen that the $E_\pm$ of Carruthers and Nieto are given by

$$E_\pm=\mathcal{P}\exp(\pm i\hat{q}_n)\,\mathcal{P}\,.$$

Other identifications (after noting that $[\mathcal{P},\mathcal{A}]=0$) include the harmonic oscillator raising and lowering operators $a^\dagger$ and a, respectively, and the number operator $a^\dagger a$:

$$a^\dagger=\mathcal{P}\mathcal{A}^\dagger\mathcal{P}\,,$$
$$a=\mathcal{P}\mathcal{A}\mathcal{P}\,,$$
$$a^\dagger a=\mathcal{P}\hat{n}\mathcal{P}/\hbar=\mathcal{P}\mathcal{A}^\dagger\mathcal{A}\mathcal{P}\,.$$

Because of the presence of the projection operators, $E_\pm$ are not unitary even though $\exp(\pm i\hat{q}_n)$ are. The recognition that these are obtained by projection from a larger space removes the objection[3,4] to the existence of the quantum mechanical angle operator $\hat{q}_n$.

APPENDIX B. CLASSICAL TREATMENT OF THE ASYMMETRIC ROTOR

The three principal axes of the rotor have moments of inertia I_x, I_y, and I_z. Associated with these are the rotational constants A, B, and C defined by

$$A=\hbar^2/2I_x\ , \quad B=\hbar^2/2I_y\ , \quad C=\hbar^2/2I_z\ ,$$

where by convention $A\geq B\geq C$. The classical Hamiltonian is[28]

$$H=A/\hbar^2 j_x^2+B/\hbar^2 j_y^2+C/\hbar^2 j_z^2\ , \tag{B1}$$

and j_x, j_y, j_z are the principal axis components of the angular momentum vector. In the limit of a prolate symmetric top, $B=C$, while $A=B$ for an oblate symmetric top.

The classical problem may be expressed in two momenta which are constants of the motion[29]; these are

$$j^2=j_x^2+j_y^2+j_z^2\ , \tag{B2}$$

$$\eta^2=(1+\kappa)/2j_z^2-(1-\kappa)/2j_x^2\ , \tag{B3}$$

where j is the magnitude of the total angular momentum and κ is the asymmetry parameter of Ray

$$\kappa=(2B-A-C)/(A-C)\ .$$

It may thus be seen that in the oblate limit $\kappa=+1$ and j_z $(=k_+)$ is conserved while in the prolate case $\kappa=-1$ and j_x $(=k_-)$ becomes a constant of the motion. The quantity η^2 is consequently a weighted average of forms appropriate in the two symmetric top limits since

$$\eta^2(\kappa=\pm 1)=\pm k_\pm^2\ .$$

In terms of j^2 and η^2, the Hamiltonian is

$$H=Bj^2-(A-C)\eta^2\ . \tag{B4}$$

If the oblate top is used as a starting point, the principal axis components of $\mathbf{j}$ can be written in terms of the momenta j, k_+ and their conjugate coordinates. Here, j is the magnitude of $\mathbf{j}$ and k_+ is the component of $\mathbf{j}$ on the oblate symmetry axis (the z axis). The projection of $\mathbf{j}$ on the space fixed z axis does not enter into any of the dynamics in the principal axis frame and so it will not be considered here. The result for $\mathbf{j}$ is

$$\mathbf{j}=\begin{pmatrix} -\sqrt{j^2-k_+^2}\cos q_{k_+} \\ +\sqrt{j^2-k_+^2}\sin q_{k_+} \\ k_+ \end{pmatrix} \tag{B5}$$

and also

$$\eta^2=(1+\kappa)k_+^2/2-(1-\kappa)/2(j^2-k_+^2)\cos^2 q_{k_+}\ . \tag{B6}$$

A canonical transformation can be performed[29] to replace k_+ by η and this results in the substitutions

$$k_+=\sqrt{\frac{2}{1+\kappa}}\ \frac{\eta}{\sqrt{1-m}}\, dn(u|m)\ , \tag{B7a}$$

$$\tan q_{k_+}=\sqrt{\frac{2}{1+\kappa}}\ \frac{sn(u|m)}{cn(u|m)}\ , \tag{B7b}$$

where

$$m=\frac{(1-\kappa)[j^2(1+\kappa)-2\eta^2]}{(1+\kappa)[j^2(1-\kappa)+2\eta^2]}\ , \tag{B8a}$$

$$u=q_\eta/\sqrt{1-m}\ , \tag{B8b}$$

and $sn(u|m)$, $cn(u|m)$, and $dn(u|m)$ are Jacobian elliptic functions.[25] The expression for $\mathbf{j}$ is now

$$\mathbf{j}=\eta/\sqrt{1-m}\begin{pmatrix} -\sqrt{\frac{2}{1-\kappa}}\sqrt{m}\ cn\,u \\ \sqrt{\frac{2}{1-\kappa}}\sqrt{\frac{2}{1+\kappa}}\sqrt{m}\ sn\,u \\ \sqrt{\frac{2}{1+\kappa}}\,dn\,u \end{pmatrix}\ . \tag{B9}$$

In the oblate limit $\kappa\to 1$, $m\to 0$ and Eq. (B9) therefore reduces to

$$\mathbf{j}(\kappa=+1)=\begin{pmatrix} -\sqrt{j^2-\eta^2}\cos q_\eta \\ \sqrt{j^2-\eta^2}\sin q_\eta \\ \eta \end{pmatrix}\ , \tag{B10}$$

which is identical to Eq. (B5) with $\eta=k_+$ and $q_\eta=q_{k_+}$. The investigation of the prolate limit is facilitated by the known[25] transformation relations for the elliptic functions

$$sn(u|m)=\mu^{1/2}sn(v|\mu)\ , \tag{B11a}$$

$$cn(u|m)=dn(v|\mu)\ , \tag{B11b}$$

$$dn(u|m)=cn(v|\mu)\ , \tag{B11c}$$

where

$$\mu=1/m\ , \quad v=u\sqrt{m}\ .$$

These are appropriate because m becomes infinite as κ approaches -1. Inserting these expressions into Eq. (B9), one obtains

$$\mathbf{j}=-i\eta/\sqrt{1-\mu}\begin{pmatrix} -\sqrt{\frac{2}{1-\kappa}}\,dn\,v \\ \sqrt{\frac{2}{1-\kappa}}\sqrt{\frac{2}{1+\kappa}}\ \sqrt{\mu}\ sn\,v \\ \sqrt{\frac{2}{1+\kappa}}\ \sqrt{\mu}\ cn\,v \end{pmatrix}\ , \tag{B12}$$

$$\mu=1/m\ , \quad v=-iq_\eta/\sqrt{1-\mu}\ .$$

Taking the limit $\kappa\to -1$, this becomes

$$\mathbf{j}(\kappa=-1)=\begin{pmatrix} i\eta \\ +\sqrt{j^2+\eta^2}\sin(+iq_\eta) \\ -\sqrt{j^2+\eta^2}\cos(+iq_\eta) \end{pmatrix}\ , \tag{B13}$$

which is exactly the correct prolate limit with $i\eta=k_-$ and $iq_\eta=q_{k_-}$. It may also be verified that no pathological results are obtained when $\eta^2=0$, the transition point between the two symmetric top forms. Thus, the expression for $\mathbf{j}$ given in Eq. (B9) includes correctly both limiting symmetric top forms and smoothly connects them.

Further insight into the motion of the system can be obtained from Eqs. (B9) and (B12). The time dependence of q_η is given by [from Eq. (B4)]

$$q_\eta(t) = q_\eta(t_0) - \eta(A - C)(t - t_0) , \tag{B14}$$

in which η is either purely real ($\eta^2 > 0$) or purely imaginary ($\eta^2 < 0$). When η^2 is positive, $m \le 1$ and Eq. (B9) is appropriate, while negative η^2 results in $\mu \le 1$ and indicates that Eq. (B12) should be used for j. It can thus be seen that, regardless of the sign of η^2, the arguments of the elliptic functions and the components of j are always purely real. Examining Eq. (B9) in light of the known properties of the elliptic functions, it may be noted that the system is *doubly* periodic in q_η with periods $P_\pm$ given by

$$P_+ = 4\sqrt{1 - m}\, K(m) , \tag{B15a}$$

$$P_- = i4\sqrt{1 - m}\, K(1 - m) , \tag{B15b}$$

where $K(m)$ is the complete elliptic integral of the first kind.[30] However, the classical motion of the system only "samples" the real period P_+. Since m is not a function of time, the value of this parameter is crucial to characterizing the dynamics of the rotor. It also depends on the magnitude of the two conserved momenta j and η so that the periods of the angle variable q_η are not independent of the conserved action variables.

[1]See, for example, H. Goldstein, *Classical Mechanics* (Addison-Wesley, Reading, Mass., 1950).
[2]The term will be used here in a somewhat broader sense as applying to nonintegrable systems as well. For example, a linear rotor in a collision system might be advantageously treated by the angular momentum variables for the free rotor.
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[17]To avoid confusion, classical coordinates and momenta will, in this section only, be given the subscript c. When the subscript is absent in this section, it is to be understood that the symbols represent quantum mechanical quantities. Later in the paper, this convention is no longer adhered to in order to avoid making the notation overly cumbersome. However, the meaning should then be clear from the context.
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[21]See, for example, pp. 256–260 of Ref. 19.
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[25]*Applied Mathematics Series*, Vol. 55, edited by M. Abramowitz and I. Stegun (National Bureau of Standards, Washington, D.C., 1964), Chap. 16.
[26]Roger Newton (to be published).
[27]See Chap. 22 of Ref. 25.
[28]See, for example, C. H. Townes and A. Schalow, *Microwave Spectroscopy* (McGraw-Hill, New York, 1955).
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[30]See Chap. 17 of Ref. 25.