cordingly, the success or failure of such an attempt should be judged by a comparison with the hard-sphere static structure factor as obtained by machine calculations. Since such data is not available to us, we can simply express the belief that at least in this case, our attempt to modify the AL model so as to extend it to higher densities should be well worth the effort. This is obvious because a variable parameter is built in our model.

However, for any comparison with real liquids or liquid metals, this attempt seems to be as much a failure as the AL hard-sphere model itself. This is, of course, not very surprising in view of the hard-sphere nature of such models.

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Steady States and Quasienergies of a Quantum-Mechanical System in an Oscillating Field*†

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A general formalism is presented for a system whose Hamiltonian is periodic in time. The formalism is intended to deal with the interactions between *bond* electrons and an external electromagnetic field, which can be treated semiclassically, such as electric and magnetic polarizations, optical rotation, and transitions among discrete levels. A particular bound-state solution of the Schrödinger equation which belongs to an irreducible representation of the time-translation symmetry group is defined as a steady state, and the characteristic number of the irreducible representation as a quasienergy. It is shown that the defined steady states and quasienergies behave in a newly constructed Hilbert space like stationary states and energies of a conservative system in many respects. It is also shown that for a resonant case the unperturbed quasienergy becomes degenerate and the transitions among discrete levels can be accounted for by the familiar degenerate perturbation procedure. Using a suitable Hilbert space, the steady states are established as firmly as the stationary states stand in the theory of a conservative system.

I. INTRODUCTION

It is well known in solid-state physics that for a spatially periodic Hamiltonian, there exist quasimomenta and corresponding Bloch wave functions. Analogously, for a periodically time-dependent Hamiltonian, one expects the existence of quasienergies and Bloch-type states. For these states Young *et al.*[1] coined the term *quasiperiodic states*; we prefer to use the term *steady states*. Such steady states have been discussed and used in the theories of susceptibilities,[1,2] and in the theories of multiple-quantum transitions among discrete levels.[3-5]

In spite of the widespread utilization of steady states for the study of the semiclassical interation between bound electrons and an external electromagnetic field, many aspects of steady states have been discussed only partially and superficially in the literature and apparently require further investigation. The essential points missed by previous workers are the introduction of a Hilbert space suitable for steady states and the uniform treatment of steady states in this space. The introduction of such a Hilbert space not only makes the formalism transparent, but also introduces new

aspects of steady states. Above all, it makes possible the unification of two seemingly different theories, namely, the theory of susceptibilities and the theory of transitions among discrete levels. Furthermore, the approximate nature of the previous theories of transitions[3-5] is removed in the new formalism. The main purpose of this paper is to show that, using a suitable Hilbert space, the steady states of a periodically time-dependent system can be placed on a foundation equally as firm as that possessed by the stationary states of time-independent quantum mechanics.

In Sec. II of this paper, we shall study the properties of steady states from a more fundamental point of view than has been done before. We first construct a Hilbert space suitable for steady states, and then show that steady states and quasienergies behave in this Hilbert space in many respects like stationary states and energies of a conservative system: Quasienergies and steady states are eigenvalues and eigenfunctions of a Hermitian operator (which we call the "Hamiltonian" for steady states); the variational principle for steady states takes the familiar form of the Ritz variational principle; and theorems analogous to the Hellmann–Feynman theorem and to the hypervirial theorem for stationary states hold for steady states. The "Hamiltonian" for steady states, which is a sum of the periodically time-dependent Hamiltonian and the time-derivative operator $-i\hbar\partial/\partial t$, plays a central role in this formalism. Unlike energies (or like quasimomenta), quasienergies are only defined *modulo* $n\hbar\omega$, where ω is the frequency of external field and n is an integer; a zone analogous to the Brillouin zone is introduced in order to obtain only physically different steady states.

In Sec. III, a perturbation theory for steady states is formulated analogously to the Rayleigh–Schrödinger perturbation theory for stationary (bound) states. The nonresonant cases (e.g., linear and nonlinear optical susceptibilities) can be accounted for by the nondegenerate perturbation procedure. In a resonant case, the unperturbed quasienergy becomes degenerate or almost degenerate; multiple-quantum transitions and the attendant Stark shift can be accounted for by the degenerate or almost-degenerate perturbation procedure. Previously, these two cases (nonresonant and resonant cases) were treated with quite different formalisms; we treat them on an equal footing, as just described. Furthermore, we do not need to restrict ourselves to a finite-dimensional Hilbert space, the use of which was essential in the previous theories of transitions.[3-5] Another advantage of the present formalism is that it provides the validity conditions for the obtained formulas. These aspects are demonstrated in Secs. III and IV.

In order to avoid the "secular divergences," Langhoff *et al.* write a wave function as a product of a time-dependent regular part and a phase factor; certain conditions imposed on the regular part render this partition unique.[2] Although these authors used the fact that for a periodic perturbation, the regular part is a periodic function of time, they did not show that the conditions imposed on the regular part go hand in hand with the periodic properties of the regular part. We shall clarify this point in Sec. III.

In Sec. IV, we apply the formalism to two specific examples in order to demonstrate the potential of this formalism.

II. STEADY STATE AND QUASIENERGY

A. Definition of Steady State and Quasienergy

We shall study a system whose Hamiltonian $H(t)$ is periodic in time with period τ: $H(t+\tau)=H(t)$. The period τ is positive, finite, and fixed at some value. The corresponding frequency is denoted by $\omega(\equiv 2\pi/\tau)$. The Schrödinger equation for the system is given by

$$\left[H(t)-i\hbar\frac{\partial}{\partial t}\right]\psi(\vec{\mathbf{r}},t)=0 \quad . \tag{2.1}$$

The vector $\vec{\mathbf{r}}$ in the wave function $\psi(\vec{\mathbf{r}},t)$ symbolizes all the spatial and spin coordinates of the system; we use this convention throughout.

Let us assume that there exists a solution $\psi(\vec{\mathbf{r}},t)$ of the form

$$\begin{aligned}&\psi(\vec{\mathbf{r}},t)=u(\vec{\mathbf{r}},t)\,e^{-i\mathcal{E}t/\hbar} \quad ,\\ &u(\vec{\mathbf{r}},t+\tau)=u(\vec{\mathbf{r}},t) \quad ,\end{aligned} \tag{2.2}$$

$$\left[H(t)-i\hbar\frac{\partial}{\partial t}\right]u(\vec{\mathbf{r}},t)=\mathcal{E}u(\vec{\mathbf{r}},t) \quad , \tag{2.3}$$

where $u(\vec{\mathbf{r}},t)$ is *square integrable* and $\mathcal{E}$ is a *real* number. If a state of the system is represented by such a solution, we call the state a *steady bound state* (or simply *steady state*) and the characteristic real number $\mathcal{E}$ the *quasienergy* of the state.

We define a time-translation operator $T(\Delta t)$ by means of

$$T(\Delta t)\psi(\vec{\mathbf{r}},t+\Delta t)=\psi(\vec{\mathbf{r}},t) \quad . \tag{2.4}$$

The time-translation operators

$$T(q\tau), \quad q=0,\ \pm 1,\ \pm 2,\ \ldots \tag{2.5}$$

commute with operator $H(t)-i\hbar(\partial/\partial t)$ and form a symmetry group of the Schrödinger equation (2.1). Since the time-translation group (2.5) is Abelian, all its irreducible representations are one-dimensional. The steady-state solution $\psi(\vec{\mathbf{r}},t)$ given by (2.2) satisfies

$$T(q\tau)\psi(\vec{\mathbf{r}},t)=e^{iq\mathcal{E}\tau/\hbar}\psi(\vec{\mathbf{r}},t) \ ; \tag{2.6}$$

hence it belongs to an irreducible representation

given by $e^{iq\mathcal{E}\tau/\hbar}$ for $q=0, \pm 1, \pm 2, \ldots$, where the quasienergy $\mathcal{E}$ characterizes the irreducible representation. We could define a steady-state solution as a bound solution which belongs to an irreducible representation of the time-translation symmetry group (2.5).

There is a close analogy between the stationary states of a time-independent Hamiltonian and the steady states of a periodically time-dependent Hamiltonian. For a time-independent Hamiltonian, the time-translation operators

$$T(t), \quad -\infty < t < \infty \tag{2.7}$$

form a symmetry group of the Schrödinger equation. A stationary state can be defined as a state which belongs to an irreducible representation of the time-translation group (2.7); the energy eigenvalue characterizes the irreducible representation.

We shall discuss the existence of steady states in Sec. V; for the time being we assume the existence of steady states.

B. Hilbert Space for Steady States

For the definition of terminology used here, we refer to textbooks on abstract Hilbert space.[6,7]

It is well known that a linear space consisting of all square-integrable functions on configuration space $\vec{r}$ [i.e., all functions $f(\vec{r})$ with finite $\int |f(\vec{r})|^2 d\vec{r}$] with the inner product $\langle f, g\rangle$ defined as $\int f^*(\vec{r})g(\vec{r})\,d\vec{r}$ is a Hilbert space, where the range of integration is the entire configuration space.[6] This Hilbert space shall be denoted by $\mathcal{R}$, and a complete orthonormal set in $\mathcal{R}$ by $\{f_1(\vec{r}), f_2(\vec{r}), \ldots\}$, which contains countable infinite basis functions. This is the Hilbert space which plays an important role for the study of stationary bound states of conservative systems.

Let us introduce another well-established Hilbert space $\mathcal{T}$, which consists of all possible periodic functions $a(t)$ of time t with the period τ with finite $\int_{-\tau/2}^{\tau/2} |a(t)|^2 dt$ and which is furnished with the inner product

$$(a, b) \equiv (1/\tau) \int_{-\tau/2}^{\tau/2} a^*(t)b(t)\,dt \quad , \tag{2.8}$$

where τ is a fixed, finite, positive, real number.[7] The function $e^{iq\omega t}$, for $q=0, \pm 1, \pm 2, \ldots$, form a complete orthonormal set in Hilbert space $\mathcal{T}$, where $\omega = 2\pi/\tau$.

We construct the composite space $\mathcal{R}+\mathcal{T}$ consisting of all possible functions $u(\vec{r}, t)$ which are periodic in the time with period τ and for which

$$\int_{-\tau/2}^{\tau/2} \int |u(\vec{r}, t)|^2 d\vec{r}\,dt \tag{2.9}$$

is finite, where the range of integration variable $\vec{r}$ is the entire configuration space as before. This composite space $\mathcal{R}+\mathcal{T}$ is a linear space; the inner product of the functions $u(\vec{r}, t)$ and $v(\vec{r}, t)$ in $\mathcal{R}+\mathcal{T}$ is defined by

$$\langle\langle u(\vec{r}, t), v(\vec{r}, t)\rangle\rangle \equiv (1/\tau) \int_{-\tau/2}^{\tau/2} \int u^*(\vec{r}, t)v(\vec{r}, t)\,d\vec{r}\,dt \quad , \tag{2.10}$$

which satisfies the conditions required to be an inner product in Hilbert space. The composite space $\mathcal{R}+\mathcal{T}$ furnished with this inner product is again a Hilbert space, and the functions $u_{nq}(\vec{r}, t)$,

$$u_{nq}(\vec{r}, t) \equiv f_n(\vec{r})\, e^{iq\omega t}, \quad n=1, 2, \ldots, \quad q=0, \pm 1, \pm 2, \ldots \tag{2.11}$$

form a complete orthonormal set in the composite Hilbert space $\mathcal{R}+\mathcal{T}$. This is the Hilbert space which we shall use to study steady states.

Once we have defined the composite Hilbert space, we can define operators in that space according to the theory of abstract Hilbert space. The definition of a linear operator in $\mathcal{R}+\mathcal{T}$ is apparent. A Hermitian operator $\mathcal{A}$ in $\mathcal{R}+\mathcal{T}$ is defined as an operator which satisfies

$$\langle\langle u, \mathcal{A}v\rangle\rangle = \langle\langle \mathcal{A}u, v\rangle\rangle \tag{2.12}$$

for any function $u(\vec{r}, t)$ and $v(\vec{r}, t)$ in $\mathcal{R}+\mathcal{T}$. A linear Hermitian operator in $\mathcal{R}$ (or $\mathcal{T}$) is also one in the composite Hilbert space $\mathcal{R}+\mathcal{T}$. The time-derivation operator $-i\hbar(\partial/\partial t)$ is a linear Hermitian operator in $\mathcal{T}$ and $\mathcal{R}+\mathcal{T}$.

I should mention here that Okuniewicz also has been using the similar Hilbert space for the study of steady states.[8]

C. Properties of Steady State and Quasienergy

"Hamiltonian" for steady states. Let us introduce the operator defined by

$$\mathcal{K} \equiv H(t) - i\hbar \frac{\partial}{\partial t} \quad , \tag{2.13}$$

where $H(t)$ is the Hamiltonian of the system concerned, which is periodic in time with period τ as before. This operator $\mathcal{K}$ is linear and Hermitian in the composite Hilbert space $\mathcal{R}+\mathcal{T}$. Using this operator $\mathcal{K}$, the steady-state Schrödinger equation (2.3) can be written in the form

$$\mathcal{K}u(\vec{r}, t) = \mathcal{E}u(\vec{r}, t) \quad , \tag{2.14}$$

where the solution $u(\vec{r}, t)$ is located in $\mathcal{R}+\mathcal{T}$. Clearly $\mathcal{K}$ is analogous to the Hamiltonian for stationary states of the time-independent Schrödinger equation; we shall call the operator the "Hamiltonian" for steady states. Quasienergies and steady states are eigenvalues and eigenfunctions of the "Hamiltonian" $\mathcal{K}$. Since $\mathcal{K}$ is Hermitian, every eigenvalue (quasienergy) is real, and two eigenfunctions (steady states) belonging to different eigenvalues (quasienergies) are orthogonal.

Physically equivalent steady states. If $\{\mathcal{E}, u(\vec{r}, t)\}$ is a solution of the steady-state eigenvalue equation (2.14), then

$$\mathcal{E}' \equiv \mathcal{E} + q\hbar\omega \ ; \ u'(\vec{r}, t) \equiv u(\vec{r}, t)\, e^{iq\omega t} \tag{2.15}$$

is also a solution for any integer q; the complete wave functions of them are, however, the same:

$$u(\vec{r}, t)\, e^{-i\mathcal{E}t/\hbar} = u'(\vec{r}, t)\, e^{-i\mathcal{E}'t/\hbar} \ . \tag{2.16}$$

In other words, all solutions given by (2.15) are physically equivalent. It is evident that one can always reduce any quasienergy $\mathcal{E}$ to a point in a zone

$$E - \tfrac{1}{2}\hbar\omega < \mathcal{E} \leq E + \tfrac{1}{2}\hbar\omega \tag{2.17}$$

specified by a real number E; therefore physically different steady states can be characterized (partially) by their *reduced quasienergies*, which lie in the same zone. The choice of zone (i.e., the choice of E) is, however, arbitrary; we shall make use of this freedom from time to time.

If $\{\mathcal{E}_m, u_m(\vec{r}, t)\}$ and $\{\mathcal{E}_n, u_n(\vec{r}, t)\}$ are solutions of Eq. (2.14) and if the quasienergies $\mathcal{E}_m$ and $\mathcal{E}_n$ lie in the same zone, then the eigenfunctions $u_m(\vec{r}, t)$ and $u_n(\vec{r}, t)$ satisfy

$$\langle u_m(\vec{r}, t),\ u_n(\vec{r}, t)\rangle = \langle\langle u_m(\vec{r}, t),\ u_n(\vec{r}, t)\rangle\rangle \ . \tag{2.18}$$

This relation implies that one can always choose the eigenfunctions $u_n(\vec{r}, t)$ such that $\langle u_m, u_n\rangle = \delta_{mn}$, since it is always possible to choose the eigenfunctions such that $\langle\langle u_m, u_n\rangle\rangle = \delta_{mn}$.

From now on, we assume that quasienergies of a "Hamiltonian" lie in the same zone, so that Eq. (2.18) holds and corresponding complete wave functions represent different physical situations.

Variational principle. The variational form of the steady-state Schrödinger equation (2.14) is given by

$$\delta\mathcal{E}[u] = 0 \ , \quad \mathcal{E}[u] \equiv \langle\langle u, \mathcal{K}u\rangle\rangle / \langle\langle u, u\rangle\rangle \ . \tag{2.19}$$

where $u(\vec{r}, t)$ and its variation $\delta u(\vec{r}, t)$ are both in $\mathfrak{R} + \mathfrak{T}$. The eigenfunctions $u_n(\vec{r}, t)$ of Eq. (2.14) are given by the stationary solutions of the variational equation (2.19), and the corresponding eigenvalues $\mathcal{E}_n$ are given by the stationary values $\mathcal{E}[u_n]$ of the functional $\mathcal{E}[u]$. We can easily show, analogously to the time-independent case, that the variational principle (2.19) is equivalent to the steady-state Schrödinger equation (2.14). The variational principle plays a central role for the determination of approximate eigenfunctions and eigenvalues, as in the case of stationary states.

Hellmann–Feynman theorem. A theorem analogous to the Hellmann–Feynman theorem[9] for stationary states in a conservative system holds also for steady-state solutions in a periodically time-dependent system. If the Hamiltonian $H(t, \lambda)$ of a system depends on a *time-independent parameter* λ and the periodic relation $H(t+\tau, \lambda) = H(t, \lambda)$ holds for any λ, then the solution $\{\mathcal{E}(\lambda), u(\vec{r}, t, \lambda)\}$ of the steady-state Schrödinger equation (2.14) satisfies the relation

$$\frac{d\mathcal{E}(\lambda)}{d\lambda} = \langle\langle u, (\partial\mathcal{K}/\partial\lambda)u\rangle\rangle / \langle\langle u, u\rangle\rangle \ , \qquad \mathcal{E}(\lambda) \equiv \langle\langle u, \mathcal{K}u\rangle\rangle / \langle\langle u, u\rangle\rangle \ . \tag{2.20}$$

The proof is analogous to the corresponding proof for stationary states,[10] and will be omitted.

Hypervirial theorem. The steady-state solutions also satisfy a theorem analogous to the hypervirial theorem[11] for stationary states: If $u(\vec{r}, t)$ is a solution of Eq. (2.14) and if operator $\mathfrak{A}$ is periodic in time with period τ, this theorem states that

$$\langle\langle u, [\mathcal{K}, \mathfrak{A}]u\rangle\rangle = 0 \ , \tag{2.21}$$

where $[\mathcal{K}, \mathfrak{A}]$ is the commutator of $\mathcal{K}$ and $\mathfrak{A}$. This hypervirial relation (2.21) has a wide range of application depending upon the choice of the operator $\mathfrak{A}$. For a particular choice of $\mathfrak{A}$, namely, $\mathfrak{A} = \frac{1}{2}\sum_n \times (\vec{r}_n \cdot \vec{p}_n + \vec{p}_n \cdot \vec{r}_n)$, where $\vec{r}_n$ and $\vec{p}_n$ are the position and linear momentum operators of the nth particle in the system concerned, Eq. (2.21) yields the virial-theorem analog for steady states:

$$2\langle\langle u, Tu\rangle\rangle = \langle\langle u, \{\sum_n \vec{r}_n \cdot \vec{\nabla}_n V(t)\}u\rangle\rangle \ , \tag{2.22}$$

where $H(t) = T + V(t)$, T is the kinetic energy, and $V(t)$ is the potential energy, which is of course periodic.

A remark. Relations with $\langle\langle\ ,\ \rangle\rangle$ for steady states have analogs of $\langle,\rangle$ relations in the stationary case, as seen before; relations with $\langle,\rangle$ for steady states, however, have no special standing and must be expected to differ from the stationary case in general.

III. STEADY-STATE PERTURBATION THEORY

A. Preliminary Remarks

Let the Hamiltonian $H(t, \lambda)$ of a given system be given by

$$H(t, \lambda) = H^{(0)} + \lambda V(t) \ , \tag{3.1}$$

where $H^{(0)}$ is a time-independent Hermitian operator, the operator $V(t)$ is also Hermitian but periodic in time with period τ, and λ is a small, real, expansion parameter.

The steady-state Schrödinger equation for the system is given by

$$[\mathcal{K}^{(0)} + \lambda V(t) - \mathcal{E}(\lambda)]u(\vec{r}, t, \lambda) = 0 \ , \tag{3.2}$$

where

$$\mathcal{K}^{(0)} \equiv H^{(0)} - i\hbar\frac{\partial}{\partial t} \ , \tag{3.3}$$

which is a Hermitian operator in the composite Hilbert space $\mathfrak{R} + \mathfrak{T}$; the solution $u(\vec{r}, t, \lambda)$ is located in $\mathfrak{R} + \mathfrak{T}$ for any λ. Note that the complete wave function $\psi(\vec{r}, t, \lambda)$ is given by

$$\psi(\vec{r},t,\lambda)=u(\vec{r},t,\lambda)\,e^{-i\mathcal{E}(\lambda)t/\hbar}\ ,$$
$$u(\vec{r},t+\tau,\lambda)=u(\vec{r},t,\lambda)\ , \tag{3.4}$$

where $\{\mathcal{E}(\lambda),\ u(\vec{r},t,\lambda)\}$ is a solution of Eq. (3.2).

We demand, of course, that $u(\vec{r},t,\lambda)$ varies continuously with λ, and adopt the normalization

$$\langle\langle u(\vec{r},t,\lambda),\ u(\vec{r},t,\lambda)\rangle\rangle=1\ , \tag{3.5}$$

which is equivalent to $\langle u,u\rangle=1$ so long as $u(\vec{r},t,\lambda)$ is a solution of Eq. (3.2), and which assures, therefore, the normalization of the complete wave function, namely, $\langle\psi,\psi\rangle=1$. The phase factor of $u(\vec{r},t,\lambda)$ will be fixed by the standard phase convention,[12] namely,

$$\langle\langle u(\vec{r},t,0),\ u(\vec{r},t,\lambda)\rangle\rangle=\langle\langle u(\vec{r},t,\lambda),\ u(\vec{r},t,0)\rangle\rangle\ , \tag{3.6}$$

which is always possible.

The unperturbed eigenvalue equation is given by

$$\mathcal{K}^{(0)}u(\vec{r},t,0)=\mathcal{E}(0)u(\vec{r},t,0)\ , \tag{3.7}$$

where $u(\vec{r},t,0)$ is located in $\mathcal{R}+\mathcal{T}$. Let E_n and $f_n(\vec{r})$ be discrete eigenvalues and eigenfunctions of the operator $H^{(0)}$, namely,

$$H^{(0)}f_n(\vec{r})=E_nf_n(\vec{r})\ ; \tag{3.8}$$

then the solutions of Eq. (3.7) are given by

$$\mathcal{E}(0)=E_n+q\hbar\omega\ ,\quad u(\vec{r},t,0)=f_n(\vec{r})\,e^{iq\omega t}\ , \tag{3.9}$$

where q is any integer. A choice of the zone (2.17) for the unperturbed quasienergies $\mathcal{E}(0)$ determines the integers q uniquely. As mentioned before, if $\{\mathcal{E}(\lambda),\ u(\vec{r},t,\lambda)\}$ is a solution of Eq. (3.2), then $\{\mathcal{E}(\lambda)+q\hbar\omega,\ u(\vec{r},t,\lambda)\,e^{iq\omega t}\}$ is also a solution representing the same physical situation. Owing to the continuity of the solutions $\{\mathcal{E}(\lambda),\ u(\vec{r},t,\lambda)\}$ with respect to λ, a choice of the zone (2.17) for the unperturbed solutions $\{\mathcal{E}(0),u(\vec{r},t,0)\}$ fixes the time-dependent phase factors $e^{iq\omega t}$ for all λ.

Consider now an eigenvalue E_k of $H^{(0)}$ and suppose that $H^{(0)}$ has eigenvalues E_m, $E_{m'}$, $E_{m''}$, ..., which satisfy

$$E_k=E_m+p\hbar\omega\ ,\quad E_{m'}+p'\hbar\omega\ ,\quad E_{m''}+p''\hbar\omega,\ \ldots \tag{3.10}$$

for some integers p, p', p'', ...; then the functions

$$f_k(\vec{r})\ ,\quad f_m(\vec{r})\,e^{ip\omega t},\quad f_{m'}(\vec{r})\,e^{ip'\omega t},$$
$$f_{m''}(\vec{r})\,e^{ip''\omega t},\ \ldots \tag{3.11}$$

are eigenfunctions of $\mathcal{K}^{(0)}$ and belong to the eigenvalue E_k of $\mathcal{K}^{(0)}$. (Note that several E_n may be the same.) This shows that even if the eigenvalue E_k of $H^{(0)}$ is nondegenerate in $\mathcal{R}$, the eigenvalue E_k of $\mathcal{K}^{(0)}$ could be degenerate in $\mathcal{R}+\mathcal{T}$. If the eigenvalue E_k of $H^{(0)}$ is degenerate in $\mathcal{R}$, then E_k is certainly a degenerate eigenvalue of $\mathcal{K}^{(0)}$ in $\mathcal{R}+\mathcal{T}$.

Since $\mathcal{K}^{(0)}$ is linear, the $\mathcal{E}(0)$ and $u(\vec{r},t,0)$ given by

$$\mathcal{E}(0)=E_k\ ,$$
$$u(\vec{r},t,0)=c_kf_k(\vec{r})+c_mf_m(\vec{r})\,e^{ip\omega t}+c_{m'}f_{m'}(\vec{r})\,e^{ip'\omega t}+\cdots \tag{3.12a}$$

is also a solution of Eq. (3.7), where c_k, c_m, $c_{m'}$, ... are arbitrary complex numbers; the corresponding complete wave function $\psi(\vec{r},t,0)$ is given by

$$\psi(\vec{r},t,0)=u(\vec{r},t,0)\,e^{-i\mathcal{E}(0)t/\hbar}$$
$$=c_kf_k(\vec{r})\,e^{-iE_kt/\hbar}+c_mf_m(\vec{r})\,e^{-iE_mt/\hbar}+c_{m'}f_{m'}(\vec{r})\,e^{-iE_{m'}t/\hbar}+\cdots\ . \tag{3.12b}$$

Equation (3.12b) clearly shows the physical significance of the coefficients, c_k, c_m, $c_{m'}$, ..., namely, the probability amplitudes of finding in the stationary states with the energies E_k, E_m, $E_{m'}$, ... One can see here the reason why degenerate perturbation theory for steady states can explain transitions among discrete levels.

B. Perturbation Theory

The Rayleigh–Schrödinger (stationary-bound-state) perturbation theory is formulated for an eigenvalue equation in the Hilbert space $\mathcal{R}$; the analogous theory for the eigenvalue equation (3.2) in $\mathcal{R}+\mathcal{T}$ can be formulated by simply translating the formulas for $\mathcal{R}$ into the corresponding ones for $\mathcal{R}+\mathcal{T}$. We shall simply write down the formulas which will be used in Sec. IV.

Nondegenerate case. Expanding $\mathcal{E}(\lambda)$ and $u(\vec{r},t,\lambda)$ in Eq. (3.2) according to

$$\mathcal{E}(\lambda)=\mathcal{E}^{(0)}+\lambda\mathcal{E}^{(1)}+\lambda^2\mathcal{E}^{(2)}+\cdots\ ,$$
$$u(\vec{r},t,\lambda)=u^{(0)}(\vec{r},t)+\lambda u^{(1)}(\vec{r},t)+\lambda^2u^{(2)}(\vec{r},t)+\cdots\ , \tag{3.13}$$
$$u^{(n)}(\vec{r},t+\tau)=u^{(n)}(\vec{r},t)\ ,\quad n=0,\ 1,\ 2,\ \ldots$$

and equating the coefficients of the same powers of λ, one obtains the following sequence of equations:

$$[\mathcal{K}^{(0)}-\mathcal{E}^{(0)}]u^{(0)}=0\ , \tag{3.14a}$$
$$[\mathcal{K}^{(0)}-\mathcal{E}^{(0)}]u^{(1)}+[V(t)-\mathcal{E}^{(1)}]u^{(0)}=0\ , \tag{3.14b}$$
$$[\mathcal{K}^{(0)}-\mathcal{E}^{(0)}]u^{(2)}+[V(t)-\mathcal{E}^{(1)}]u^{(1)}-\mathcal{E}^{(2)}u^{(0)}=0\ . \tag{3.14c}$$

The combination of normalization and phase conditions (3.5) and (3.6) yields another sequence of equations,

$$\langle\langle u^{(0)},u^{(0)}\rangle\rangle=1\ , \tag{3.15a}$$
$$\langle\langle u^{(0)},u^{(1)}\rangle\rangle=0\ , \tag{3.15b}$$
$$\langle\langle u^{(0)},u^{(2)}\rangle\rangle=-\tfrac{1}{2}\langle\langle u^{(1)},u^{(1)}\rangle\rangle\ . \tag{3.15c}$$

Expressions for the purturbation eigenvalues are

$$\mathcal{E}^{(1)} = \langle\langle u^{(0)},\ V(t)u^{(0)}\rangle\rangle, \tag{3.16a}$$

$$\mathcal{E}^{(2)} = \langle\langle u^{(0)},\ V(t)u^{(1)}\rangle\rangle. \tag{3.16b}$$

Degenerate case. Suppose that the unperturbed eigenvalue $\mathcal{E}^{(0)}$ in question is degenerate and that one set of corresponding orthonormal eigenfunctions are $u_1^{(0)}(\vec{r}, t), \ldots, u_N^{(0)}(\vec{r}, t)$; then the first-order eigenvalues $\mathcal{E}_a^{(1)}$ and the corresponding "correct" zeroth-order eigenfunctions $u_{a\alpha}^{(0)}(\vec{r}, t)$ are given by

$$\sum_{n=1}^{N}\left(\langle\langle u_m^{(0)}, V(t)u_n^{(0)}\rangle\rangle - \mathcal{E}_a^{(1)}\delta_{mn}\right)c_{n,a\alpha} = 0\ ,$$
$$m = 1, \ldots, N \tag{3.17}$$
$$u_{a\alpha}^{(0)}(\vec{r}, t) = \sum_{n=1}^{N} u_n^{(0)}(\vec{r}, t)c_{n,a\alpha}\ ,$$

where the index a distinguishes between the values of eigenvalues $\mathcal{E}_a^{(1)}$ and the index α in the eigenvector $\{c_{1,a\alpha}, c_{2,a\alpha}, \ldots, c_{N,a\alpha}\}$ distinguishes between the eigenvectors belonging to the same eigenvalue $\mathcal{E}_a^{(1)}$. We can always choose the coefficients $c_{n,a\alpha}$ to constitute a unitary matrix; then the N eigenfunctions $u_{a\alpha}^{(0)}(\vec{r}, t)$ are again orthornomal.

Almost-degenerate case (Ref. 13). Let $\mathcal{E}_1^{(0)}$ and $\mathcal{E}_2^{(0)}$ be two nondegenerate eigenvalues of $\mathcal{H}^{(0)}$ and $u_1^{(0)}(\vec{r}, t)$ and $u_2^{(0)}(\vec{r}, t)$ be the corresponding eigenfunctions, respectively. The expansion parameter λ is now considered as a fixed finite number, which is small enough so that one can still put forward the solution of Eq. (3.2) as a power series. If the unperturbed eigenvalues $\mathcal{E}_1^{(0)}$ and $\mathcal{E}_2^{(0)}$ are so close together that they satisfy the relation

$$\left|(\mathcal{E}_1^{(0)} - \mathcal{E}_2^{(0)})/\lambda\right| \lesssim \left|\langle\langle u_1^{(0)}, V(t)u_2^{(0)}\rangle\rangle\right|\ , \tag{3.18}$$

then the first-order approximate solution

$$\mathcal{E}(\lambda) = \tfrac{1}{2}(\mathcal{E}_1^{(0)} + \mathcal{E}_2^{(0)}) + \lambda\mathcal{E}^{(1)} + O(\lambda^2)\ ,$$
$$u(\vec{r}, t, \lambda) = \{c_1u_1^{(0)}(\vec{r}, t) + c_2u_2^{(0)}(\vec{r}, t)\} + O(\lambda) \tag{3.19}$$

is given by the secular equation

$$\begin{pmatrix} V_{11} + \Delta - \mathcal{E}^{(1)} & V_{12} \\ V_{21} & V_{22} - \Delta - \mathcal{E}^{(1)} \end{pmatrix}\begin{pmatrix} c_1 \\ c_2 \end{pmatrix} = 0\ ,$$
$$\Delta \equiv (\mathcal{E}_1^{(0)} - \mathcal{E}_2^{(0)})/2\lambda\ ,\quad V_{mn} \equiv \langle\langle u_m^{(0)},\ V(t)u_n^{(0)}\rangle\rangle\ . \tag{3.20}$$

C. Transformed Perturbation Equations

We now transform the eigenvalue equation (3.2) by introducing a factor $e^{i\theta(t,\lambda)/\hbar}$, where $\theta(t, \lambda)$ is a function of t and λ:

$$v(\vec{r}, t, \lambda) = e^{i\theta(t,\lambda)/\hbar}u(\vec{r}, t, \lambda)\ , \tag{3.21}$$

$$\left(\mathcal{H}^{(0)} + \lambda V(t) - \frac{\partial\theta(t, \lambda)}{\partial t} - \mathcal{E}(\lambda)\right)v(\vec{r}, t, \lambda) = 0\ . \tag{3.22}$$

The complete wave functions $\psi(\vec{r}, t, \lambda)$ is now given by

$$\psi(\vec{r}, t, \lambda) = v(\vec{r}, t, \lambda)\, e^{-i\{\mathcal{E}(\lambda)t + \theta(t,\lambda)\}/\hbar}\ . \tag{3.23}$$

Let us first make the transformation such that $v(\vec{r}, t, \lambda)$ satisfies the conditions

$$\langle v(\vec{r}, t, \lambda),\ v(\vec{r}, t, \lambda)\rangle = 1\ ,$$
$$\left\langle v(\vec{r}, t, \lambda),\ \frac{\partial}{\partial t}v(\vec{r}, t, \lambda)\right\rangle = \left\langle\frac{\partial}{\partial t}v(\vec{r}, t, \lambda),\ v(\vec{r}, t, \lambda)\right\rangle. \tag{3.24}$$

The corresponding $\theta(t, \lambda)$ has to be real and satisfy

$$\frac{\partial\theta(t, \lambda)}{\partial t} = \langle u, [H^{(0)} + \lambda V(t)]u\rangle - \mathcal{E}(\lambda)\ , \tag{3.25}$$

where we have used the fact that the operators $V(t)$ and $e^{i\theta(t,\lambda)/\hbar}$ commute. Suppose that $\theta(t+\tau, \lambda) = \theta(t, \lambda)$; then, integrating Eq. (3.25) with respect to t over the period τ, one has

$$\mathcal{E}(\lambda) = \langle\langle u, [H^{(0)} + \lambda V(t)]u\rangle\rangle. \tag{3.26}$$

This equation, of course, does not hold in general. Hence neither $\theta(t, \lambda)$ nor $v(\vec{r}, t, \lambda)$ can be a periodic function of time with period τ. In other words, the conditions (3.24) and the periodic relation

$$v(\vec{r}, t+\tau, \lambda) = v(\vec{r}, t, \lambda) \tag{3.27}$$

do not hold simultaneously. It is important to notice the close relation between the conditions imposed on $v(\vec{r}, t, \lambda)$ and its periodic property (3.27).

As stated by Langhoff *et al.*,[2] one can avoid the "secular divergences" by imposing the conditions

$$\langle v(\vec{r}, t, \lambda),\ v(\vec{r}, t, \lambda)\rangle = 1\ , \tag{3.28a}$$

$$\langle v(\vec{r}, t, 0),\ v(\vec{r}, t, \lambda)\rangle = \langle v(\vec{r}, t, \lambda),\ v(\vec{r}, t, 0)\rangle \tag{3.28b}$$

on $v(\vec{r}, t, \lambda)$, or by imposing another set of conditions,

$$\langle\psi(\vec{r}, t, \lambda),\ \psi(\vec{r}, t, \lambda)\rangle = 1\ ,$$
$$\langle v(\vec{r}, t, 0),\ v(\vec{r}, t, \lambda)\rangle = 1\ . \tag{3.29}$$

In the following paragraphs we shall show that one can always choose $\theta(t, \lambda)$ so that $v(\vec{r}, t, \lambda)$ satisfies both the conditions (3.28) and the periodic relation (3.27).

In order to satisfy Eqs. (3.27) and (3.28a), the corresponding $\theta(t, \lambda)$ has to be *real* and periodic in time with period τ. For any given $\theta(t, \lambda)$ which is real and periodic in time with period τ, the corresponding solution $v(\vec{r}, t, \lambda)$ of Eq. (3.22) satisfies

$$\frac{\partial}{\partial t}\langle v, v\rangle = 0\ , \tag{3.30}$$

$$\frac{i\hbar}{2}\frac{\partial}{\partial t}\ln\left[\frac{\langle v^{(0)}, v\rangle}{\langle v, v^{(0)}\rangle}\right] = \frac{\partial}{\partial t}[\theta(t, 0) - \theta(t, \lambda)] + [\mathcal{E}^{(0)} - \mathcal{E}(\lambda)] + \lambda\,\mathrm{Re}\left[\frac{\langle v, V(t)v^{(0)}\rangle}{\langle v, v^{(0)}\rangle}\right], \tag{3.31}$$

where $\mathcal{E}^{(0)} \equiv \mathcal{E}(0)$ and $v^{(0)} \equiv v(\vec{r}, t, 0)$. Equation (3.31) is the key relation to prove the statement.

The eigenvalue $\mathcal{E}(\lambda)$ is given by

$$\mathcal{E}(\lambda) = (1/\tau) \int_{-\tau/2}^{\tau/2} \mathcal{E}(t,\lambda)\, dt \ , \tag{3.32}$$

$$\mathcal{E}(t,\lambda) \equiv \mathcal{E}^{(0)} + \lambda \mathrm{Re}\left[\frac{\langle v, V(t)v^{(0)}\rangle}{\langle v, v^{(0)}\rangle}\right], \tag{3.33}$$

as is seen from Eq. (3.31). We choose the function $\theta(t,\lambda)$ such that

$$\frac{\partial}{\partial t}\theta(t,\lambda) = \mathcal{E}(t,\lambda) - \mathcal{E}(\lambda) \ , \quad \int_{-\tau/2}^{\tau/2} \theta(t,\lambda)\, dt = 0 \ , \tag{3.34}$$

where the function $v(\vec{r},t,\lambda)$ in the $\mathcal{E}(t,\lambda)$ is the corresponding solution for this chosen $\theta(t,\lambda)$. In order to be self-consistent, the function $\theta(t,\lambda)$ defined by Eqs. (3.34) must be real and periodic in time with period τ; using Eq. (3.32), one can easily show the self-consistency.

For this specially chosen $\theta(t,\lambda)$, the corresponding $v(\vec{r},t,\lambda)$ satisfies

$$[\mathcal{H}^{(0)} + \lambda V(t) - \mathcal{E}(t,\lambda)]v(\vec{r},t,\lambda) = 0 \ , \tag{3.35}$$

$$\frac{\partial}{\partial t}[\langle v^{(0)}, v\rangle / \langle v, v^{(0)}\rangle] = 0 \ , \tag{3.36}$$

where the second equation follows from Eq. (3.31). If $v(\vec{r},t,\lambda)$ is a solution of Eq. (3.35), then $c(\lambda)v(\vec{r},t,\lambda)$ is also a solution, where $c(\lambda)$ is an arbitrary complex function of λ. Using this freedom and Eqs. (3.30) and (3.36), one can always make a solution $v(\vec{r},t,\lambda)$ to satisfy the conditions (3.28a) and (3.28b).

Thus we have shown that the conditions (3.28) imposed on $v(\vec{r},t,\lambda)$ go hand in hand with the periodic relation (3.27). Similarly one can show that Eqs. (3.27) and (3.29) hold simultaneously. For this case, however, the corresponding $\theta(t,\lambda)$ is not real, and the transformed "Hamiltonian," $\mathcal{H}^{(0)} + \lambda V(\lambda) - \partial\theta(t,\lambda)/\partial t$, is no longer Hermitian in $\mathfrak{R}+\mathfrak{T}$. Because of this disadvantage, we prefer the conditions (3.28) to (3.29) in order to avoid the secular divergences.

To sum up, the equations for the desired $v(\vec{r},t,\lambda)$ are Eqs. (3.35), (3.33), and (3.28), and the solution $v(\vec{r},t,\lambda)$ of them must be located in $\mathfrak{R}+\mathfrak{T}$. The eigenvalue $\mathcal{E}(\lambda)$ and the phase function $\theta(t,\lambda)$ are given by Eq. (3.32) and Eqs. (3.34), respectively, where $v(\vec{r},t,\lambda)$ in the $\mathcal{E}(t,\lambda)$ is the solution of Eqs. (3.35) and (3.33). The complete wave function $\psi(\vec{r},t,\lambda)$ is given by (3.23). Since the solution $v(\vec{r},t,\lambda)$ of Eq. (3.35) automatically satisfies Eqs. (3.30) and (3.36), the conditions (3.38) are equivalent to the conditions

$$\langle\langle v(\vec{r},t,\lambda),\ v(\vec{r},t,\lambda)\rangle\rangle = 1 \ , \tag{3.37}$$

$$\langle\langle v(\vec{r},t,0),\ v(\vec{r},t,\lambda)\rangle\rangle = \langle\langle v(\vec{r},t,\lambda),\ v(\vec{r},t,0)\rangle\rangle \ ,$$

so long as $v(\vec{r},t,\lambda)$ is a solution of Eq. (3.35). The solution $v(\vec{r},t,\lambda)$ of Eqs. (3.35), (3.33), and (3.28) satisfies

$$\langle\langle v^{(0)},\ V(t)v\rangle\rangle = \langle\langle v, V(t)v^{(0)}\rangle\rangle \ . \tag{3.38}$$

Note that in general the relation $\langle v^{(0)},\ V(t)v\rangle = \langle v, V(t)v^{(0)}\rangle$ cannot be expected to hold.

Expanding $\mathcal{E}(\lambda)$, $\mathcal{E}(t,\lambda)$, $\theta(t,\lambda)$, and $v(\vec{r},t,\lambda)$ according to

$$\mathcal{E}(\lambda) = \sum_{k=0}^{\infty} \lambda^k \mathcal{E}^{(k)}, \quad \mathcal{E}(t,\lambda) = \sum_{k=0}^{\infty} \lambda^k \mathcal{E}^{(k)}(t) \ ,$$
$$\theta(t,\lambda) = \sum_{k=0}^{\infty} \lambda^k \theta^{(k)}(t) \ , \quad v(\vec{r},t,\lambda) = \sum_{k=0}^{\infty} \lambda^k v^{(k)}(\vec{r},t), \tag{3.39}$$

and substituting into Eqs. (3.35), (3.33), and (3.28), one obtains

$$[\mathcal{H}^{(0)} - \mathcal{E}^{(0)}]v^{(0)} = 0 \ , \quad \langle v^{(0)}, v^{(0)}\rangle = 1 \ ,$$
$$\mathcal{E}^{(0)}(t) = \mathcal{E}^{(0)} \ , \tag{3.40}$$

and for $n = 1, 2, \ldots,$

$$[\mathcal{H}^{(0)} - \mathcal{E}^{(0)}]v^{(n)} + [V(t) - \mathcal{E}^{(1)}(t)]v^{(n-1)} - \sum_{k=2}^{n} \mathcal{E}^{(k)}(t)v^{(n-k)} = 0 \ , \tag{3.41a}$$

$$\mathcal{E}^{(n)}(t) = \mathrm{Re}\langle v^{(0)},\ V(t)v^{(n-1)}\rangle - \sum_{k=1}^{n-1} \mathcal{E}^{(k)}(t)\langle v^{(0)}, v^{(n-k)}\rangle \ , \tag{3.41b}$$

$$\langle v^{(0)},\ v^{(n)}\rangle = -\tfrac{1}{2}\sum_{k=1}^{n-1} \langle v^{(k)},\ v^{(n-k)}\rangle \ . \tag{3.41c}$$

One can solve the sequence of equations (3.40) and (3.41) progressively. The $\mathcal{E}^{(k)}$ and $\theta^{(k)}(t)$ are given by

$$\mathcal{E}^{(k)} = (1/\tau)\int_{-\tau/2}^{\tau/2} \mathcal{E}^{(k)}(t)\, dt \ ,$$
$$\frac{d}{dt}\theta^{(k)}(t) = \mathcal{E}^{(k)}(t) - \mathcal{E}^{(k)}, \quad \int_{-\tau/2}^{\tau/2} \theta^{(k)}(t)\, dt = 0 \ . \tag{3.42}$$

Incidentally, the variational equation for the $v^{(1)}(\vec{r},t)$ is given by

$$\delta F[u(\vec{r},t)] = 0,$$
$$F[u] \equiv \langle\langle u, [\mathcal{H}^{(0)} - \mathcal{E}^{(0)}]u\rangle\rangle + 2\,\mathrm{Re}\langle\langle v^{(0)}, [V(t) - \langle v^{(0)}, V(t)v^{(0)}\rangle]u\rangle\rangle, \tag{3.43}$$

where $u(\vec{r},t)$ and $\delta u(\vec{r},t)$ are in $\mathfrak{R}+\mathfrak{T}$.

Remarks. Let us consider an inhomogeneous equation with an auxiliary condition

$$\left[\mathrm{H}^{(0)} - i\hbar\left(\frac{\partial}{\partial t}\right) - E_0\right]v(\vec{r},t) = w(\vec{r},t),$$
$$\langle f_0(\vec{r}),\ v(\vec{r},t)\rangle = 0 \ , \tag{3.44}$$

where $H^{(0)}f_0(\vec{r}) = E_0 f_0(\vec{r})$ and the given function $w(\vec{r},t)$ is periodic in time with period τ. If $v(\vec{r},t)$ is a solution of Eqs. (3.44), then the $v'(\vec{r},t)$ given by

$$v'(\vec{r},t) = v(\vec{r},t) + \sum_{n(\neq 0)} c_n f_n(\vec{r})\, e^{i(E_0 - E_n)t/\hbar} \tag{3.45}$$

is also a solution of Eqs. (3.44), where E_n and $f_n(\vec{r})$ are discrete eigenvalues and eigenfunctions of $H^{(0)}$, and the coefficients c_n are arbitrary. This shows that the solution of Eqs. (3.44) is not unique and in general not periodic in time with period τ. The fact that $w(\vec{r},t)$ is a periodic function of time *does not* ensure that solutions $v(\vec{r},t)$ of Eqs. (3.44) are periodic in time.[14] One should establish the periodicity of the solution $v^{(n)}(\vec{r},t)$ of Eq. (3.41a) on the basis of the steady state $u(\vec{r},t)$, as we have done before. Finally we emphasize that the transformed equation and the original one are equivalent as long as the period τ is finite.

IV. APPLICATIONS

We shall now apply the steady-state perturbation theory to the case when the perturbing operator $V(t)$ is harmonic, namely,

$$V(t)=2V^{(1)}\cos\omega t \quad , \tag{4.1}$$

where $V^{(1)}$ is a time-independent Hermitian operator, and $\omega=2\pi/\tau\neq 0$; we shall study two examples for demonstration.

A. One-Level System

In this section we consider the case when the discrete eigenvalue E_0 of $H^{(0)}$ in question is nondegenerate in $\mathfrak{R}$, and when there is, besides E_0, no discrete eigenvalue of $H^{(0)}$ in the vicinity of E_0, $E_0\pm\hbar\omega$, $E_0\pm 2\hbar\omega$, and $E_0\pm 3\hbar\omega$.

Time dependence of the perturbed wave function. Let us first assume that there is no discrete eigenvalue E_n of $H^{(0)}$ which satisfies $E_n=E_0+q\hbar\omega$ for some nonzero integer q; namely, the eigenvalue E_0 of $\mathfrak{H}^{(0)}$ is nondegenerate. We shall use the transformed perturbation equations (3.40) and (3.41), because of the desirable limiting behavior of the perturbed wave function at $\omega=0$; the same notations as in Sec. III C will be used in this section.

We choose the zone (2.17) such that the zeroth-order eigenfunction $v^{(0)}(\vec{r},t)$ is time independent:

$$[H^{(0)}-E_0]f_0(\vec{r})=0 \ , \ \langle f_0(\vec{r}), f_0(\vec{r})\rangle=1 \ ,$$
$$\mathcal{E}^{(0)}=E_0 \ , \ v^{(0)}(\vec{r},t)=f_0(\vec{r}) \ . \tag{4.2}$$

Knowing $v^{(0)}(\vec{r},t)$, one can calculate $\mathcal{E}^{(1)}(t)$ from Eq. (3.14b), namely,

$$\mathcal{E}^{(1)}(t)=2E^{(1)}\cos\omega t \ , \quad E^{(1)}\equiv\langle f_0, V^{(1)}f_0\rangle \ . \tag{4.3}$$

Since the first-order eigenfunction $v^{(1)}(\vec{r},t)$ is a periodic function of t with period $2\pi/\omega$, it can be expanded in a Fourier series, namely,

$$v^{(1)}(\vec{r},t)=\sum_q f_q^{(1)}(\vec{r})\,e^{iq\omega t} \quad , \tag{4.4}$$

where function $f_q^{(1)}(\vec{r})$ are in $\mathfrak{R}$. Substituting (4.3) and (4.4) into the first-order equation of (3.41a) and (3.41c) and using the fact that $\omega\neq 0$, one obtains

$$(H^{(0)}-E_0\pm\hbar\omega)f_{\pm1}^{(1)}+(V^{(1)}-E^{(1)})f_0=0 \ , \tag{4.5a}$$

$$\langle f_0, f_{\pm1}^{(1)}=0\rangle, \tag{4.5b}$$

$$(H^{(0)}-E_0+q\hbar\omega)f_q^{(1)}=0 \ , \ \langle f_0, f_q^{(1)}\rangle=0 \ , \quad \text{for } q\neq\pm1 \ . \tag{4.6}$$

From the assumption we made, there are no nonvanishing functions in $\mathfrak{R}$ which satisfy Eqs. (4.6); therefore, the functions $f_q^{(1)}(\vec{r})$ must vanish except for $q=\pm1$. Thus the first-order eigenfunction $v^{(1)}(\vec{r},t)$ is given by

$$v^{(1)}(\vec{r},t)=f_{+1}^{(1)}(\vec{r})\,e^{i\omega t}+f_{-1}^{(1)}(\vec{r})e^{-i\omega t} \quad , \tag{4.7}$$

where the functions $f_{\pm1}^{(1)}(\vec{r})$ satisfy Eqs. (4.5), respectively. Note that Eqs. (4.5a) yield Eqs. (4.5b).

From Eq. (3.41b), one has

$$\mathcal{E}^{(2)}(t)=2E^{(2)}(1+\cos2\omega t) \ ,$$
$$E^{(2)}\equiv\tfrac{1}{2}\langle f_0, V^{(1)}(f_{+1}^{(1)}+f_{-1}^{(1)})\rangle \quad ; \tag{4.8}$$

it is easy to see from Eqs. (4.5a) that $\langle f_0, V^{(1)}f_{\pm1}^{(1)}\rangle$ are real. By similar manipulation, one obtains

$$v^{(2)}(\vec{r},t)=f_{+2}^{(2)}(\vec{r})e^{i2\omega t}+f_{-2}^{(2)}(\vec{r})e^{-i2\omega t}+2f_0^{(2)}(\vec{r}) \ , \tag{4.9}$$

where the functions $f_{\pm2}^{(2)}(\vec{r})$ and $f_0^{(2)}(\vec{r})$ satisfy

$$(H^{(0)}-E_0\pm2\hbar\omega)\,f_{\pm2}^{(2)}+(V^{(1)}-E^{(1)})\,f_{\pm1}^{(1)}-E^{(2)}f_0=0 \ ,$$
$$(H^{(0)}-E_0)\,f_0^{(2)}+\tfrac{1}{2}(V^{(1)}-E^{(1)})(f_{+1}^{(1)}+f_{-1}^{(1)})-E^{(2)}f_0=0; \tag{4.10}$$

$$\langle f_0, f_{+2}^{(2)}\rangle=\langle f_0, f_{-2}^{(2)}\rangle=-\tfrac{1}{2}\langle f_{+1}^{(1)}, f_{-1}^{(1)}\rangle$$
$$=-\tfrac{1}{2}\langle f_{-1}^{(1)}, f_{+1}^{(1)}\rangle \quad , \tag{4.11a}$$

$$\langle f_0, f_0^{(2)}\rangle=-\tfrac{1}{4}(\langle f_{+1}^{(1)}, f_{+1}^{(1)}\rangle+\langle f_{-1}^{(1)}, f_{-1}^{(1)}\rangle) \ ; \tag{4.11b}$$

note that Eqs. (4.5a) and (4.10) yield Eqs. (4.11a).

From the formulas (3.42), one obtains the eigenvalue $\mathcal{E}^{(k)}$ and the phase functions $\theta^{(k)}(t)$, namely,

$$\mathcal{E}^{(0)}=E_0 \ , \quad \mathcal{E}^{(1)}=0 \ , \quad \mathcal{E}^{(2)}=2E^{(2)}, \quad \mathcal{E}^{(3)}=0 \ , \tag{4.12}$$

$$\theta^{(0)}(t)=0 \ , \quad \theta^{(1)}(t)=2E^{(1)}\sin\omega t/\omega \ ,$$
$$\theta^{(2)}(t)=2E^{(2)}\sin2\omega t/2\omega \ . \tag{4.13}$$

Thus the complete wave function $\psi(\vec{r},t,\lambda)$ to the second order, is given by

$$\psi(\vec{r},t,\lambda)=[f_0+\lambda(f_{+1}^{(1)}e^{i\omega t}+f_{-1}^{(1)}e^{-i\omega t})+\lambda^2(f_{+2}^{(2)}e^{i2\omega t}$$
$$+f_{-2}^{(2)}e^{-i2\omega t}+2f_0^{(2)})+\cdots]e^{-i\eta(t,\lambda)/\hbar} \ , \tag{4.14a}$$

$$\eta(t,\lambda)=E_0t+2\lambda E^{(1)}\frac{\sin\omega t}{\omega}+2\lambda^2E^{(2)}\left(t+\frac{\sin2\omega t}{2\omega}+\cdots\right) , \tag{4.14b}$$

where $f_{\pm1}^{(1)}$ satisfy Eqs. (4.5), and $f_{\pm2}^{(2)}$ and $f_0^{(2)}$

satisfy Eqs. (4.10) and (4.11).

The second-order quasienergy eigenvalue $\mathcal{E}^{(2)}$ is the quantity of physical interest; for example, when $V^{(1)}$ is the x component of the dipole moment operator, $-\mathcal{E}^{(2)}$ gives the frequency-dependent polarizability $\alpha_{xx}(\omega)$.

Applicability conditions. Suppose this time that there are functions $g_{q\alpha}(\vec{r})$ in $\mathfrak{R}$ which satisfy

$$[H^{(0)} - E_0 + q\hbar\omega]g_{q\alpha}(\vec{r}) = 0 \ , \quad \langle g_{q\alpha},\ g_{q\beta}\rangle = \delta_{\alpha\beta} \tag{4.15}$$

for some nonzero integer q, where the second index α in $g_{q\alpha}(\vec{r})$ distinguishes between the eigenfunctions belonging to the same eigenvalue $E_0 - q\hbar\omega$ of $H^{(0)}$; then the functions $f_0(\vec{r})$ and $g_{q\alpha}(\vec{r})\,e^{iq\omega t}$ belong to the eigenvalue E_0 of $\mathcal{K}^{(0)}$, and E_0 is no longer a nondegenerate eigenvalue of $\mathcal{K}^{(0)}$. For this case, one has to use the degenerate perturbation method.

If the functions $v^{(0)}(\vec{r},t)$, $v^{(1)}(\vec{r},t)$, and $v^{(2)}(\vec{r},t)$ given by Eqs. (4.2), (4.7), and (4.9) satisfy

$$\langle\langle g_{q\alpha}e^{iq\omega t},\ \{[V(t) - \mathcal{E}^{(1)}(t)]v^{(m-1)} - \sum_{k=2}^{m}\mathcal{E}^{(k)}(t)v^{(m-k)}\}\rangle\rangle = 0 \quad \text{for all } q\alpha \tag{4.16}$$

for $m = 1, \ldots, (n+1)$, where $\mathcal{E}^{(1)}(t)$ and $\mathcal{E}^{(2)}(t)$ are given by Eqs. (4.3) and (4.8), then the function $v^{(0)} + \ldots + \lambda^n v^{(n)}$ satisfies the degenerate perturbation equations up the to nth-order with the eigenvalue $\mathcal{E}^{(0)} + \ldots + \lambda^n\mathcal{E}^{(n)}$, and $\mathcal{E}^{(n+1)}$ is an eigenvalue of the $(n+1)$th-order equation, where the eigenvalue $\mathcal{E}^{(k)}$ are given by Eqs. (4.12).

We still assume that E_0 is a nondegenerate eigenvalue of $H^{(0)}$. For $m = 1$, Eq. (4.16) yields

$$\langle g_{q\alpha}, V^{(1)}f_0\rangle(\delta_{q,+1} + \delta_{q,-1}) = 0 \quad \text{for all } q\alpha \ ; \tag{4.17}$$

hence if $E_0 \pm \hbar\omega$ are not eigenvalues of $H^{(0)}$, then $\{\mathcal{E}^{(1)} = 0,\ v^{(0)}(\vec{r},t) = f_0(\vec{r})\}$ is a solution of the first-order degenerate perturbation equation, and furthermore the solutions $f_{\pm1}^{(1)}(\vec{r})$ of Eqs. (4.5) are unique. If there exist the eigenvalues of $H^{(0)}$ which are close to the $E_0 + \hbar\omega$ or $E_0 - \hbar\omega$, then the solution $f_{+1}^{(1)}(\vec{r})$ or $f_{-1}^{(1)}(\vec{r})$ becomes large. Therefore the applicability condition for $\{\mathcal{E}^{(0)} + \lambda\mathcal{E}^{(1)},\ v^{(0)}\}$ is that there exists no eigenvalue of $H^{(0)}$ at the vicinity of $E_0 \pm \hbar\omega$.

For $m = 2$, Eq. (4.16) yields

$$\langle g_{q\alpha}, [V^{(1)} - E^{(1)}]f_{+1}^{(1)}\rangle\delta_{q,+2} + \langle g_{q\alpha},\ [V^{(1)} - E^{(1)}]f_{-1}^{(1)}\rangle\delta_{q,-2} = 0 \quad \text{for all } q\alpha \ ; \tag{4.18}$$

if $E_0 \pm 2\hbar\omega$ are not eigenvalues of $H^{(0)}$, then $\{\mathcal{E}^{(2)}, v^{(2)}\}$ is a solution of the second-order degenerate perturbation equation, and the solution $f_{\pm2}^{(2)}(\vec{r})$ and $f_0^{(2)}(\vec{r})$ of Eqs. (4.10) and (4.11) are unique. If there exist the eigenvalues of $H^{(0)}$ which are close to the $E_0 + 2\hbar\omega$, $E_0 - 2\hbar\omega$, or E_0, then the solution $f_{+2}^{(2)}(\vec{r})$, $f_{-2}^{(2)}(\vec{r})$, or $f_0^{(2)}(\vec{r})$ again becomes large. Hence the applicability condition for $\{\mathcal{E}^{(0)} + \lambda\mathcal{E}^{(1)} + \lambda^2\mathcal{E}^{(2)},\ v^{(0)} + \lambda v^{(1)}\}$ is that besides E_0, there exists no eigenvalue of $H^{(0)}$ at the vicinity of E_0, $E_0 \pm \hbar\omega$, and $E_0 \pm 2\hbar\omega$. Similarly the applicability condition for $\{\mathcal{E}^{(0)} + \lambda\mathcal{E}^{(1)} + \lambda^2\mathcal{E}^{(2)} + \lambda^3\mathcal{E}^{(3)},\ v^{(0)} + \lambda v^{(1)} + \lambda^2 v^{(2)}\}$ is that besides E_0 there exists no eigenvalue of $H^{(0)}$ at the vicinity of E_0, $E_0 \pm \hbar\omega$, $E_0 \pm 2\hbar\omega$, and $E_0 \pm 3\hbar\omega$.

Limiting behavior at $\omega = 0$. If $\hbar\omega$ is much smaller than the difference between E_0 and the closest eigenvalue of $H^{(0)}$, then there will be, besides E_0, no eigenvalue of $H^{(0)}$ in the vicinity of E_0, $E_0 \pm \hbar\omega$, $E_0 \pm 2\hbar\omega$, and $E_0 \pm 3\hbar\omega$; hence one may consider Eqs. (4.1)–(4.14) valid in the neighborhood of $\omega = 0$.

At the limit $\omega = 0$, the functions $f_{\pm1}^{(1)}(\vec{r})$, $f_{\pm2}^{(2)}(\vec{r})$, and $f_0^{(2)}(\vec{r})$ become

$$\begin{aligned} f^{(1)}(\vec{r}) &\equiv f_{+1}^{(1)}(\vec{r}) = f_{-1}^{(1)}(\vec{r}) \ , \\ f^{(2)}(\vec{r}) &\equiv f_{+2}^{(2)}(\vec{r}) = f_{-2}^{(2)}(\vec{r}) = f_0^{(2)}(\vec{r}) \ , \end{aligned} \tag{4.19}$$

where $f^{(1)}(\vec{r})$ and $f^{(2)}(\vec{r})$ satisfy the stationary perturbation equations, namely,

$$\begin{aligned} &[H^{(0)} - E_0]f_0 = 0 \ , \\ &[H^{(0)} - E_0]f^{(1)} + [V^{(1)} - \langle f_0,\ V^{(1)}f_0\rangle]f_0 = 0 \ , \\ &[H^{(0)} - E_0]f^{(2)} + [V^{(1)} - \langle f_0,\ V^{(1)}f_0\rangle]f^{(1)} \\ &\qquad - \langle f_0,\ V^{(1)}f^{(1)}\rangle f_0 = 0 \ , \end{aligned} \tag{4.20a}$$

$$\begin{aligned} &\langle f_0,\ f_0\rangle = 1 \ , \quad \langle f_0,\ f^{(1)}\rangle = 0 \ , \\ &\langle f_0,\ f^{(2)}\rangle = -\tfrac{1}{2}\langle f^{(1)},\ f^{(1)}\rangle \ . \end{aligned} \tag{4.20b}$$

When $\omega \to 0$, the complete wave function $\psi(\vec{r},t,\lambda)$ smoothly joins the stationary solution of the Hamiltonian $H^{(0)} + (2\lambda)V^{(1)}$,

$$\begin{aligned} \psi(\vec{r},t,\lambda) &= [f_0 + (2\lambda)f^{(1)} + (2\lambda)^2 f^{(2)} + \ldots]\,e^{-i\eta(t,\lambda)/\hbar} , \\ \eta(t,\lambda) &= t[E_0 + (2\lambda)\langle f_0,\ V^{(1)}f_0\rangle \\ &\qquad + (2\lambda)^2\langle f_0,\ V^{(1)}f^{(1)}\rangle + \cdots] \ , \end{aligned} \tag{4.21}$$

for any finite t.

This limiting behavior is due to the transformation we made; the original eigenfunction $u(\vec{r},t,\lambda)$ does not have this limiting property. If one wishes to expand the perturbed wave function in powers of ω, then the limiting property we obtain is indispensable.

Variational method. The variational equations for the solutions $f_{\pm1}^{(1)}(\vec{r})$ of Eqs. (4.5) are given by

$$\begin{aligned} &\delta F_+[h_+(\vec{r})] = 0 \ , \quad \delta F_-[h_-(\vec{r})] = 0 \ , \\ &F_\pm[h_\pm] \equiv \langle h_\pm,\ [H^{(0)} - E_0 \pm \hbar\omega]h_\pm\rangle \\ &\qquad + 2\,\mathrm{Re}\langle f_0,\ [V^{(1)} - \langle f_0,\ V^{(1)}f_0\rangle]h_\pm\rangle \ . \end{aligned} \tag{4.22}$$

These equations can be obtained from Eq. (3.43), or merely by inspection.

A remark. If one adopts another normalization and phase convention, namely, $\langle\psi,\ \psi\rangle=1$ and $\langle v^{(0)},\ v\rangle=1$ with complex $\theta(t,\lambda)$, then one obtains somewhat more complex equations than Eqs. (4.1)–(4.14).[15]

B. Two-Level System Connected with Single-Quantum Transition

We shall now study the case which obtains when two discrete nondegenerate eigenvalues E_1 and E_2 of $H^{(0)}$ satisfy $E_2\approx E_1+\hbar\omega$, and there are, besides E_1 and E_2, no eigenvalues of $H^{(0)}$ in the vicinity of $E_1\pm\hbar\omega$ and $E_2\pm\hbar\omega$; the eigenvalue E_1 of $\mathcal{H}^{(0)}$ is then almost degenerate. We can treat this problem by the almost-degenerate perturbation method as developed in Sec. III. In particular, the eigenfunctions and eigenvalues are determined by Eqs. (3.19) and (3.20); we shall use the same notation here as was used there.

Let $f_1(\vec{r})$ and $f_2(\vec{r})$ be the eigenfunctions of $H^{(0)}$ belonging to the eigenvalues E_1 and E_2, respectively; choosing the zone (2.17) suitably, one has the eigenvalues and eigenfunctions of $\mathcal{H}^{(0)}$ in the form

$$\begin{aligned}&\mathcal{E}_1^{(0)}=E_1\ ,\quad u_1^{(0)}(\vec{r},t)=f_1(\vec{r})\ ,\\&\mathcal{E}_2^{(0)}=E_2-\hbar\omega\ ,\quad u_2^{(0)}(\vec{r},t)=f_2(\vec{r})\,e^{-i\omega t}\ .\end{aligned}\tag{4.23}$$

The eigenvalues of Eq. (3.20) are given by

$$\begin{aligned}&\mathcal{E}_{\pm1}^{(1)}=\pm\left[\Delta^2+\left|\langle f_1,\ V^{(1)}f_2\rangle\right|^2\right]^{1/2}\ ,\\&\Delta=(E_1-E_2+\hbar\omega)/2\lambda\ ;\end{aligned}\tag{4.24}$$

the corresponding eigenvectors $\{c_{1+},\ c_{2+}\}$ and $\{c_{1-},\ c_{2-}\}$ are determined up to a phase factor from the equations

$$R_\pm\equiv\frac{c_{1\pm}}{c_{2\pm}}=\frac{\langle f_1,\ V^{(1)}f_2\rangle}{\mathcal{E}_\pm^{(1)}-\Delta}\ ,\quad |c_{1\pm}|^2+|c_{2\pm}|^2=1\ .\tag{4.25}$$

Thus the first-order solutions are given by

$$\begin{aligned}\psi_\pm(\vec{r},t,\lambda)=&\left[c_{1\pm}u_1^{(0)}(\vec{r},t)+c_{2\pm}u_2^{(0)}(\vec{r},t)\right]\\&\times e^{-i(\mathcal{E}^{(0)}+\lambda\mathcal{E}_\pm^{(1)})t/\hbar}\ ,\end{aligned}\tag{4.26}$$

$$\mathcal{E}^{(0)}\equiv\tfrac{1}{2}(\mathcal{E}_1^{(0)}+\mathcal{E}_2^{(0)})\ .$$

Suppose that the system is in the state $f_1(\vec{r})$ at $t=0$; then the wave function $\psi(\vec{r},t,\lambda)$ at subsequent values of t is, in first-order, given by

$$\begin{aligned}\psi(\vec{r},t,\lambda)&=(c_{2-}\psi_+-c_{2+}\psi_-)/(c_{2-}c_{1+}-c_{2+}c_{1-})\\&=\frac{e^{-i\mathcal{E}^{(0)}t/\hbar}}{R_+-R_-}\left[(R_+e^{-i\lambda\mathcal{E}_+^{(1)}t/\hbar}-R_-e^{-i\lambda\mathcal{E}_-^{(1)}t/\hbar})f_1\right.\\&\left.\quad+e^{-i\omega t}(e^{-i\lambda\mathcal{E}_+^{(1)}t/\hbar}-e^{-i\lambda\mathcal{E}_-^{(1)}t/\hbar})f_2\right],\end{aligned}\tag{4.27}$$

since $\psi(\vec{r},t,\lambda)$ is the first-order solution of the Schrödinger equation, and satisfies $\psi(\vec{r},0,\lambda)=f_1(\vec{r})$. The probability $P_2(t)$ of finding the system in the state $f_2(\vec{r})$ at the time t is given by

$$P_2(t)=\left|(e^{-i\lambda\mathcal{E}_+^{(1)}t/\hbar}-e^{-i\lambda\mathcal{E}_-^{(1)}t/\hbar})/(R_+-R_-)\right|^2\ ;\tag{4.28}$$

substituting (4.25) into (4.28), one obtains

$$P_2(t)=\frac{\lambda^2|\langle f_1,\ V^{(1)}f_2\rangle|^2}{\hbar^2}\left[\frac{\sin(\omega^{(1)}t)}{\omega^{(1)}}\right]^2\ ,\tag{4.29}$$

$$\omega^{(1)}=\left[\tfrac{1}{4}(E_1-E_2+\hbar\omega)^2+\lambda^2\left|\langle f_1,\ V^{(1)}f_2\rangle\right|^2\right]^{1/2}/\hbar\ .$$

It is easy to show that the probability $P_1(t)$ of finding the system in the state $f_1(\vec{r})$ at the time t is given by $P_1(t)+P_2(t)=1$. The formula (4.29) is nothing but the well-known Rabi formula.[16]

The applicability conditions for the formula (4.29) are given by Eq. (3.18), namely,

$$\left|(E_1-E_2+\hbar\omega)/\lambda\right|\lesssim\left|\langle f_1,\ V^{(1)}f_2\rangle\right|\ ,\tag{4.30}$$

and by the requirement that there are, besides E_1 and E_2, no eigenvalues of $H^{(0)}$ in the vicinity of $E_1\pm\hbar\omega$ and $E_2\pm\hbar\omega$. The presence of eigenvalues of $H^{(0)}$ in the vicinity of $E_1+q\hbar\omega$ and $E_2+q\hbar\omega$ for $|q|\geq2$ does not change the final result (4.29). These applicability conditions give the conditions for the two-level-system *model* to be valid in the first-order-transition-probability calculation.

V. DISCUSSIONS

Existence of steady-state solutions. Most of the Hamiltonians which one encounters in practice are of the form $H^{(0)}+\lambda V(t)$, where $H^{(0)}$ is a time-independent Hermitian operator, $V(t)$ is also Hermitian but periodic in time, and λ is a small real parameter. If the steady-state Schrödinger equation (3.2) and (3.3) for the system has a discrete eigenvalue $\mathcal{E}(\lambda)$ and its eigenfunction $u(\vec{r},t,\lambda)$ in $\mathfrak{R}+\mathfrak{T}$, then we certainly have a steady-state solution, since $u(\vec{r},t,\lambda)e^{-i\mathcal{E}(\lambda)t/\hbar}$ is a bound solution of the Schrödinger equation and has the required form. Hence the question of the existence of steady states can be reduced to the question of the existence of the perturbation solutions of Eq. (3.2) and (3.3).

The unperturbed Hamiltonian $H^{(0)}$ that we are interested in usually has bound-state solutions in $\mathfrak{R}$ and therefore the operator $\mathcal{H}^{(0)}$ $(=H^{(0)}-i\hbar\,\partial/\partial t)$ has discrete eigenvalues and corresponding eigenfunctions in $\mathfrak{R}+\mathfrak{T}$, namely, steady-state solutions [see Eqs. (3.7)–(3.9)]. The solutions that we are interested in are such that the eigenvalue $\mathcal{E}(\lambda)$ approaches one of the discrete eigenvalues of $\mathcal{H}^{(0)}$, when $\lambda\to0$. The question on the existence of such perturbation solutions can be treated analogously to the static case[17]; again the difference is the Hilbert spaces we use, $\mathfrak{R}$ or $\mathfrak{R}+\mathfrak{T}$.

By analogy, one can expect that for some $V(t)$ (including the perturbing operator for the Stark effect), there exist only asymptotic eigenvalues and eigenfunctions; in other words, the perturbation equations have solutions only up to some order.[17]

For this case, one has asymptotic steady states, which is sufficient to explain phenomena such as the Stark effect. Young *et al.* have also given an argument on the existence of asymptotic steady states.[1]

Switching function. In this paper we have intentionally avoided use of a switching function, which describes how the oscillating part $V(t)$ is turned on and reaches its asymptotic form. We simply regard steady-state solutions as asymptotic solutions of the Schrödinger equation which has a switching function, and expect that steady-state solutions are valid at times long after the oscillating part has reached its asymptotic form, namely, a periodically time-dependent form. As is well known, the static Stark effect has been treated in similar manner. In this way, we avoid tricky arguments on switching functions and hope the above statement is correct. Langhoff *et al.* have included a switching function in their formalism and somehow obtained essentially the same equations as ours for the one-level system.[2]

Prospects. Just recently the multilevel theory was proposed for the simultaneous occurrence of Stark shifts and multiple-quantum transitions by Hicks *et al.*[5]; in essence, they solve a steady-state Schrödinger equation for a perturbed system [for example, Eq. (3.2) with a finite λ] within a specially chosen subspace of the composite Hilbert space $\mathcal{R}+\mathcal{T}$, which is composed from several eigenfunctions of the unperturbed operator $H^{(0)}$ and the functions $e^{iq\omega t}$ with small integers q. As is well known for the Stark-effect calculation, the unperturbed eigenfunctions of $H^{(0)}$ are not suited to expand the perturbed portion of the wave function, since so many unperturbed eigenfunctions, including those belonging to the continuous spectrum, are required in order to obtain reasonably accurate susceptibilities. One avoids this difficulty by choosing the basis functions properly. We can reformulate the multilevel theory within our formalism by developing a higher-order almost-degenerate perturbation theory. Research along this line is in progress and the results will published in the near future.

The Hellmann–Feynman theorem and the hypervirial theorem are expected to yield useful relations which can be used to check the accuracy of calculated, induced charge and current densities of an atom (or a molecule) in an external electromagnetic field. This will be considered subsequently elsewhere.

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