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Semiclassical Electrodynamics of Bound Systems in Intense Fields

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A general approximation method is developed for the treatment of bound quantum systems in the presence of an intense external electromagnetic field of long wavelength. The method is based upon a unitary transformation which approximately removes the electromagnetic field from the problem. The accuracy of the technique increases with the number of photons involved in any given process; and in the case of large photon number, the results have a simple, concise form resembling first-order perturbation theory. It is possible to handle intense-field problems with small numbers of photons also, but then the formalism takes on a complexity analogous to the corresponding perturbation expressions for the same number of photons. The theory is extended to the case where two electromagnetic field modes are present, one or both of which may be intense. Bound-bound transitions in a hydrogenlike atom are calculated as an illustration of the application of the method. One of the explicit nonlinear effects to appear is an intensity-dependent deviation of the transition probability behavior from I^n for n-photon transitions, where I is the field intensity. It is demonstrated that intensity effects are much more probable with bound electrons than with free electrons.

I. INTRODUCTION

The only method of general applicability presently available for the treatment of bound-state problems in semiclassical electrodynamics is perturbation theory. Although perturbation theory has been remarkably successful, a new technological situation has been developing in recent years which promises to provide electromagnetic field environments so intense that the usefulness (and possibly even the correctness) of perturbation theory is in doubt. For example, atomic processes in intense fields have already been observed[1] where on the order of ten photons are involved in a single transition. Perturbation calculations of such high-order processes are extremely difficult to perform.[2] Furthermore (as will become evident later in this paper), when a transition requires a large number of photons in order to proceed, transitions involving more than the minimum number of photons can also become significant. The problem of calculating higher-order corrections to the (already high-order) lowest-order result becomes prohibitively difficult for a perturbation method.

It is possible to give an order-of-magnitude argument which indicates the likelihood of trouble with a perturbation approach. Consider an atom subjected to a plane-wave electromagnetic field given by the vector potential A. This vector potential enters the dynamical equation through the substitution $p \to p - eA$. (Throughout the paper, Gaussian units will be used, and $\hbar = c = 1$.) Let us compare the magnitude of eA with a characteristic parameter of the atom itself, such as the binding energy. If A is trigonometric with amplitude a, and if the energy E is given by a Rydberg unit $\frac{1}{2}me^4$, then

$$\frac{eA}{E} = O\left(\frac{ea}{me^4}\right) = O\left[\frac{1}{\alpha^2}\left(\frac{\Delta m^2}{m^2}\right)^{1/2}\right] ,$$

where α is the fine-structure constant, and $\Delta m^2/m^2$ is the field-intensity parameter which arises in free-electron problems.[3] Values of $(\Delta m^2/m^2)^{1/2}$ as large as 10^{-3} can be achieved at present with focused laser beams, and α^{-2} is of the order of 10^4, so that the magnitude of eA can actually exceed the atomic binding energy. Clearly, caution must be exercised in the application of perturbation methods to such a problem.

In Sec. II of this paper, a nonperturbation approximation is introduced which is particularly suited to the many-photon bound-state problem in the long-wavelength case. The basic motivation for the technique is to reduce the influence of the eA term by approximately transforming it away. The method is demonstrated first for the relativistic case, and is then developed in detail for the nonrelativistic case. The nature of the approximation is shown by an order-of-magnitude analysis. Corrections to the basic approximation are then introduced.

In Sec. III, the expectation of the energy of a bound system in the presence of an electromag-

netic field is examined.

Section IV presents the development of the formalism necessary to calculate transition probabilities without recourse to the standard results of perturbation theory.

Section V is concerned with the conversion of the general formalism into a practical calculational tool. A generalized dipole-approximation commutator theorem is demonstrated, and used to give a simple general matrix element which must be calculated. This matrix element is subjected to an angular momentum reduction valid for central potentials. The example of $1s$-$2s$ transitions in a hydrogenlike atom is calculated, and analyzed in terms of departures from perturbative intensity behavior. An important general feature is the appearance of a field-intensity parameter which is $(137)^2$ times larger than the parameter which occurs in free-electron problems.

Combined effects of intense and weak fields are treated in Sec. VI. Results analogous to those developed for a single intense field are established for the two-fields situation, and again applied to the $1s$-$2s$ transition in hydrogenlike atoms as an example. The particular cases considered are Raman-like processes involving absorption of n photons from the intense field, and emission of a single photon of appropriate energy to make up the $1s$-$2s$ energy difference; or emission of n' photons stimulated by the intense field, plus emission of a single photon sufficient to conserve energy in the decay of the $2s$ state to the ground state.

Section VII is devoted to a general comparison of the new method developed here with perturbation theory. It is shown that the present method, although designed for many-photon problems, reduces to first-order perturbation results for low field intensity. Systematic corrections to the general method developed in Sec. II can be applied to make the results reduce to any higher order of perturbation theory. A general analysis involving an intense field and a weak field is employed to show the manner in which the intense field causes real and virtual transitions into all possible states.

II. MOMENTUM-TRANSLATION METHOD

A. Rationale

We wish to develop a calculational method for bound-state problems when an intense, external electromagnetic field of long wavelength is present. By long wavelength is meant that the inequality $\omega a_0 \ll 1$ is satisfied, where ω is the frequency of the electromagnetic field, and a_0 is a characteristic elementary size of the bound system (e.g., a Bohr radius for an atom, a unit cell for a solid, a pion Compton wavelength for a nucleus). It is the long-wavelength restriction which suggests the method.[4] If the size of the bound elementary quantum system is small as compared to the wavelength, then the electromagnetic field has an approximate uniformity throughout the atom (hereafter, for convenience, we shall use "atom" in place of "bound elementary quantum system"). If the field were truly constant, it would be a simple matter to make a unitary transformation which would eliminate the electromagnetic field from the problem. We shall find it a useful technique to make this unitary transformation even when the field is not constant. The relativistic case will be presented first, since it is simpler in formal appearance. Then the nonrelativistic case, which is simpler in practical application, will be developed in detail, and applied.

B. Relativistic Case

"Natural" units ($\hbar = c = 1$) will be used. The metric is such that the scalar product of two four-vectors x^μ and y^μ is $x_\mu y^\mu = g_{\mu\nu} x^\mu y^\nu = x \cdot y = x^0 y^0 - \vec{x} \cdot \vec{y}$, where $\vec{x}$ and $\vec{y}$ are three-vectors and $g^{\mu\nu}$ is the metric tensor. The electromagnetic field is introduced through the minimal-coupling substitution $p^\mu \to p^\mu - eA^\mu$, where p^μ is the operator $i\partial^\mu$.

Set

$$\Psi = \exp(-ieA \cdot x)\overline{\Psi} \ , \tag{1}$$

where we consider Ψ to be either the spinor solution to the Dirac equation

$$[\gamma \cdot (p - eA) - \gamma^0 V - m]\Psi = 0 \ , \tag{2}$$

or the scalar solution to the Klein-Gordon equation

$$[(p^\mu - eA^\mu - V\delta_0{}^\mu)(p_\mu - eA_\mu - V\delta_\mu{}^0) - m^2]\Psi = 0 \ . \tag{3}$$

In Eqs. (2) and (3), V is the potential which gives rise to the bound states of the atom, and in Eq. (2) the γ^μ are the Dirac matrices which satisfy $\gamma^\mu\gamma^\nu + \gamma^\nu\gamma^\mu = 2g^{\mu\nu}$. If A^μ were a constant four-vector, the transformation given by Eq. (1) would amount to a momentum translation of amount eA^μ. Since A^μ is not constant

$$(p^\mu - eA^\mu)\Psi = \exp(-ieA \cdot x)[p^\mu + ex_\nu(\partial^\mu A^\nu)]\overline{\Psi} .$$

i.e., Eq. (1) amounts to a replacement of eA^μ by $ex_\nu(\partial^\mu A^\nu)$ in the wave equation. For an atom of radius a_0, x_ν can be approximated in magnitude by a_0; and for a plane wave of frequency ω, $\partial^\mu A^\nu$ has a magnitude of ω times the magnitude of A^μ. Hence,

$$\frac{|ex_\nu(\partial^\mu A^\nu)|}{|eA^\mu|} = O(\omega a_0) \ll 1 ,$$

so that the transformation of Eq. (1) leads to a suppression of electromagnetic field quantities in the wave equation by the factor ωa_0, considered to be very small by the long-wavelength hypothesis. That is, Eq. (2) becomes

$$\{\gamma_\mu[p^\mu + ex_\nu(\partial^\mu A^\nu)] - \gamma^0 V - m\}\bar{\Psi} = 0 , \tag{4}$$

and Eq. (3) becomes

$$\{[p^\mu + ex_\nu(\partial^\mu A^\nu) - V\delta_0{}^\mu]$$

$$\times [p_\mu + ex^\lambda(\partial_\mu A_\lambda) - V\delta_\mu{}^0] - m^2\}\bar{\Psi} = 0 . \tag{5}$$

Equations (4) and (5) should be much more manageable in terms of perturbation theory than Eqs. (2) and (3), even for very intense fields, as long as $\omega a_0 \ll 1$. As will be shown in detail for the nonrelativistic case, it is both nontrivial and justifiable in certain cases to employ Eq. (1) as the approximate solution of the complete problem when $\bar{\Psi}$ is replaced by the solution of Eq. (4) or (5) for $A^\mu = 0$; i.e., zeroth-order perturbation theory in the terms arising from $ex_\nu(\partial\mu A^\nu)$ leads to meaningful results.

C. Nonrelativistic Case

The Schrödinger equation for an atom in the presence of an electromagnetic field can be written

$$i\partial_t \Psi = H\Psi , \quad H = (2m)^{-1}(\vec{p} - e\vec{A})^2 + V(\vec{r}) , \tag{6}$$

where a gauge with $A_0 = 0$ has been chosen. We shall impose a restriction to source-free fields ($\partial_\mu \partial^\mu A^\nu = 0$) subject to the Lorentz condition ($\partial_\mu A^\mu = 0$), so that we have also $\vec{\nabla}\cdot\vec{A} = 0$. There will be a requirement also for the solution Φ of the field-free equation

$$i\partial_t \Phi = H_0\Phi , \quad H_0 = (2m)^{-1}\vec{p}^2 + V(\vec{r}) . \tag{7}$$

Consider the unitary transformation induced by $\exp(-ie\vec{A}\cdot\vec{x})$,

$$\bar{\Psi} = \exp(-ie\vec{A}\cdot\vec{x})\Psi , \tag{8}$$

which is the nonrelativistic analog of Eq. (1), since in the gauge with $A_0 = 0$, $A\cdot x = -\vec{A}\cdot\vec{x}$. The transformed Schrödinger equation is

$$i\bar{\partial}_t\bar{\Psi} = \bar{H}\bar{\Psi} , \tag{9}$$

where $i\bar{\partial}_t = \exp(-ie\vec{A}\cdot\vec{x})\, i\partial_t \exp(ie\vec{A}\cdot\vec{x})$,

$$\bar{H} = \exp(-ie\vec{A}\circ\vec{x})H\exp(ie\vec{A}\circ\vec{x}).$$

The transformed operators are found readily to be

$$i\bar{\partial}_t = i\partial_t - e(\partial_t\vec{A})\cdot\vec{x},$$

$$\bar{H} = H_0 + em^{-1}(\partial_i A_j)x_j p_i - \tfrac{1}{2}iem^{-1} \tag{10}$$

$$\times(\nabla^2 A_i)x_i + \tfrac{1}{2}e^2m^{-1}(\partial_i A_j)(\partial_i A_k)x_j x_k,$$

where H_0 is as given by Eq. (7), and repeated indices are to be summed from 1 to 3. Equation (9) can thus be written as the Schrödinger equation with $i\partial_t$ operator

$$i\partial_t\bar{\Psi} = (H_0 + H_I)\bar{\Psi}, \tag{11}$$

where $H_I = e(\partial_t A_i)x_i + em^{-1}(\partial_i A_j)x_j p_i$

$$-\tfrac{1}{2}iem^{-1}(\nabla^2 A_i)x_i + \tfrac{1}{2}e^2m^{-1}$$

$$\times(\partial_i A_j)(\partial_i A_k)x_j x_k. \tag{12}$$

If H_I could be neglected, then the solution of Eq. (11) would be simply Φ of Eq. (7), the field-free solution of the atomic problem. Then, by inverting Eq. (8), the general solution of the original problem would be

$$\Psi \approx \exp(ie\vec{A}\cdot\vec{x})\,\Phi . \tag{13}$$

Let us explore the circumstances under which it is reasonable to neglect H_I. Two conditions will be imposed: (a) The magnitude of H_I should be small as compared to a characteristic energy of the field-free problem; and (b) the magnitude of H_I should be small as compared to the usual perturbation operator H' of semiclassical electrodynamics, where

$$H' = H - H_0 = -em^{-1}\vec{A}\circ\vec{p} + \tfrac{1}{2}e^2m^{-1}\vec{A}^2 . \tag{14}$$

The necessary order-of-magnitude analysis is greatly expedited if we can confine our attention to only a single term of H_I. With the order-of-magnitude replacements $x_i \to a_0$, $\partial_t A_i$ or $\partial_j A_i \to \omega a$

(where a is the amplitude of A), $p_i/m \to (2E/m)^{1/2}$, then the second term of H_I as given in Eq. (12) has the relative magnitude $(2E/m)^{1/2}$ as compared to the first term. For explicitness, we can take E to be the binding energy of a hydrogenlike atom, so $(2E/m)^{1/2} = Ze^2$. For light atoms or outer electrons of heavy atoms, the effective Z is much smaller than the inverse fine-structure constant e^{-2} (or α^{-1}), so the second term of Eq. (12) is dominated by the first term. The third term of Eq. (12) bears the ratio ω/m with respect to the first term, which is certainly small for any long-wavelength atomic problem. The last term of H_I is smaller than the first term by the factor $(\omega/m)(eaa_0)$. It has been remarked already that ω/m is small. The product eaa_0 is of particular interest because it occurs throughout the analysis of this paper, and it (or, more properly, its square) can be identified as the fundamental intensity parameter of the bound-system problem. For hydrogenlike atoms, $a_0 = (Zme^2)^{-1}$, so

$$eaa_0 = (Z\alpha)^{-1}(2\Delta m^2/m^2)^{1/2} , \tag{15}$$

where, as remarked above, $\Delta m^2/m^2$ is the intensity parameter characteristic of the free-electron problem. For effective values of Z which are not much larger than unity, $(eaa_0)^2$ is then some 10^4 times larger than the intensity parameter for free electrons. A value of $\Delta m^2/m^2$ of 10^{-6} leads to $(eaa_0)^2$ about 10^{-2}. Hence, despite the much greater size of the bound-state intensity parameter as compared to the free-electron intensity parameter, the magnitude of the last term of H_I is, like the others, small as compared to the $e(\partial_t \vec{A})\cdot\vec{x}$ term of H_I.

Now it is a simple matter to examine the two criteria which are to be imposed to justify the approximate solution Eq. (13). Condition (a) was that H_I be of small magnitude as compared to a characteristic energy of the atom. Comparing $e(\partial_t \vec{A})\cdot\vec{x}$ with an atomic energy E gives the requirement

$$eaa_0(\omega/E) \ll 1. \tag{16a}$$

Condition (b) requires that $e(\partial_t \vec{A})\cdot\vec{x}$ be small as compared to $em^{-1}\vec{A}\cdot\vec{p}$. With the relation $a_0 = (2mE)^{-1/2} = p^{-1}$, condition (b) gives

$$\tfrac{1}{2}(\omega/E) \ll 1 . \tag{16b}$$

It has already been pointed out that the parameter eaa_0 is less than unity. Even with great future increases in available intensity, eaa_0 will never be a large number, so Eqs. (16a) and (16b) can both be satisfied if ω/E is taken to be sufficiently small.

Consider a transition induced in an atom by the electromagnetic field of frequency ω. The energy change of the atom in the transition is the smallest characteristic atomic energy that would enter into the problem. Hence, the requirement $\omega \ll E$, with E as the transition energy and ω as the energy of a photon, is equivalent to a many-photon condition. Thus, we conclude that if a transition can occur only through a high-order process involving the electromagnetic field, then it is a reasonable approximation to neglect H_I and employ Eq. (13) as the solution to Eq. (6). This will be referred to as the momentum-translation approximation.

Corrections to the momentum-translation approximation are derived readily. The complete solution to Eq. (11) is given by the integral equation

$$\bar{\Psi} = \Phi + \sum_n \Phi_n \times \left[-i \int_{-\infty}^{t} (\Phi_n(t'), H_I(t')\bar{\Psi}(t'))\, dt' \right] , \tag{17}$$

where the summation is over the complete set of field-free solutions Φ_n. A Neumann series solution of this integral equation gives what would commonly be termed a perturbation expansion for $\bar{\Psi}$, with H_I as the perturbing potential. For instance, the zeroth-order result $\bar{\Psi}^{(0)} = \Phi$ is just the momentum-translation approximation, Eq. (13). The first-order result is

$$\bar{\Psi}^{(1)} = \Phi + \sum_n \Phi_n \times \left[-i \int_{-\infty}^{t} (\Phi_n(t'), H_I(t')\Phi(t'))\, dt' \right] . \tag{18}$$

As shown above [and as can also be shown directly in Eq. (18)], the second term in Eq. (18) gives contributions whose order of magnitude is $eaa_0(\omega/E)$ times the contribution of the Φ term. This means that for arbitrarily intense fields, as long as ω/E is small enough, the zeroth-order result is accurate. Conversely, if we wish to explore the intense-field behavior of processes involving small numbers of photons, it may be necessary to employ higher-order approximations to Eq. (17).

III. ENERGY BEHAVIOR

Because $\vec{A}$ is time-dependent, the Hamiltonian of Eq. (6) has explicit time dependence, and thus it does not possess energy eigenvalues. Nevertheless, we can examine the energy behavior of an atom in an electromagnetic field in terms of the expectation value of the $i\partial_t$ operator.

For the momentum-translation approximation,

$$\langle \Psi | i\partial_t | \Psi \rangle = \langle \bar{\Psi} | i\partial_t | \bar{\Psi} \rangle$$

$$= E + \langle \Phi | e\vec{\mathcal{E}} \cdot \vec{x} | \Phi \rangle \quad , \tag{19}$$

where we have used Eq. (10), and introduced the electric field vector $\vec{\mathcal{E}}$, given by $\vec{\mathcal{E}} = -\partial_t \vec{A}$ in the gauge being used here. Equation (19) has a very familiar appearance. It has exactly the same form as the energy for the first-order Stark effect caused by a static electric field. In Eq. (19), $\vec{\mathcal{E}}$ is not static. If, as is generally done in the dipole approximation, $\vec{\mathcal{E}}$ is taken to be a function of time only, then Eq. (19) says that the momentum-translation approximation predicts that the average energy of an atom oscillates in phase with the applied electric field, with the same amplitude as if that field were static. It will be seen later that the momentum-translation result predicts that the $\vec{A}$ field leads to both real and virtual transitions in the atom. One can take the point of view that it is the average over all these transitions[5] that leads to the "adiabatic" Stark effect given by Eq. (19).

The Stark-effect result of Eq. (19) can be refined if $\bar{\Psi}^{(1)}$ as given in Eq. (18) is used instead of $\bar{\Psi}^{(0)}$. To first order in H_I, the result can be shown to be

$$\langle \Psi | i\partial_t | \Psi \rangle = E + \langle \Phi | e\vec{\mathcal{E}} \cdot \vec{x} | \Phi \rangle + 2\mathrm{Re}\{\langle \Phi | e\vec{\mathcal{E}} \cdot \vec{x} |$$

$$\times \sum_n \Phi_n \left[-i \int_{-\infty}^{t} (\Phi_n, H_I \Phi)\, dt' \right] \rangle\} \quad . \tag{20}$$

The last term in Eq. (20) explicitly exhibits the effects of coupling between levels in the atom caused by the electric field.

IV. FORMALISM FOR TRANSITIONS

The conventional means for calculating transition probabilities in semiclassical electrodynamics makes use of the machinery of perturbation theory. Such methods are not appropriate here, and so a more general technique will be outlined below. What is required is only a slight extension of well-known results of the formal theory of scattering, so the presentation will be quite brief.

The starting point is to split the total Hamiltonian operator H into a field-independent part H_0 and a field-dependent part H', as given in Eq. (14). The operator $\exp(iH_0 t)$ then defines a transformation to the interaction picture, in which the time-development operator $\hat{U}(t^0, t_0)$ is given by

$$\hat{U}(t^0, t_0) = 1 - i \int_{t_0}^{t_0} dt_1 \hat{H}'(t_1) \hat{U}(t_1, t_0) \ ,$$

where the time-development operator is defined by

$$\hat{\Psi}(t^0) = \hat{U}(t^0, t_0) \hat{\Psi}(t_0) \ , \tag{21}$$

and the caret denotes operators and wave functions given in the interaction picture, e.g.,

$$\hat{\Psi} = e^{iH_0 t} \Psi$$

$$\hat{H}' = e^{iH_0 t} H' e^{-iH_0 t} \ . \tag{22}$$

The time-development operator becomes the S operator when $t^0 \to \infty$, $t_0 \to -\infty$ (with appropriate behavior of $\hat{H}'$ at $t = \pm\infty$), so that

$$S = 1 - i \int_{-\infty}^{\infty} dt_1 \hat{H}'(t_1) \hat{U}(t_1, -\infty) \ .$$

The S-matrix element defined between a pair of in-states $\hat{\Psi}_f(-\infty)$ and $\hat{\Psi}_i(-\infty)$ is then

$$(\hat{\Psi}_f(-\infty), S\hat{\Psi}_i(-\infty)) \equiv S_{fi}$$

$$= \delta_{fi} - i \int_{-\infty}^{\infty} dt_1 (\hat{\Psi}_f(-\infty), \hat{H}'(t_1) \hat{\Psi}_i(t_1)) \ ,$$

where Eq. (21) has been used. To transform back to the original Schrödinger picture with the inverse of Eq. (22), it is convenient to reintroduce a limiting process in terms of t_0,

$$(S-1)_{fi} = -i \lim_{t_0 \to -\infty} \int_{t_0}^{\infty} dt_1$$

$$\times (e^{iH_0 t_0} \Psi_f(t_0), e^{iH_0 t_1} H'(t_1) \Psi_i(t_1)) \quad . \tag{23}$$

It should be noted that the S matrix is the same in both the pictures employed, so there is no need to employ the caret on S. Implicit in the introduction of S is the assumption that H' is "turned off" at $t \to \pm\infty$ by means of appropriate convergence factors, as explained in any standard text which discusses the formal theory of scattering. The absence of H' in the limit $t \to -\infty$ means that

$$e^{-iH_0 t_1} \lim_{t_0 \to -\infty} e^{iH_0 t_0} \Psi_f(t_0) = \Phi_f(t_1) \quad ,$$

i.e., the operator involving t_0 reduces Ψ_f to the time-independent field-free wave function at $t = -\infty$; and the operator involving t_1 restores the time dependence at time t_1, but retains the field-free character of the time-independent part. Thus, if we introduce a new notation τ to replace $S - 1$, Eq. (23) becomes

$$(S-1)_{fi} \equiv \tau_{fi}$$

$$= -i \int_{-\infty}^{\infty} dt (\Phi_f(t), H'(t) \Psi_i(t)) \quad . \tag{24}$$

Equation (24) is quite general. Suppose now that the momentum-translation approximation Eq. (13) is introduced. Then, with Φ split into time-depen-

dent and time-independent factors denoted by

$$\Phi(\vec{r},t)=\phi(\vec{r}) \tag{25}$$

[we assume $V(\vec{r})$ and hence H_0 to be time-independent], τ_{fi} takes on the form

$$\tau_{fi}=-i\int_{-\infty}^{\infty}dt\,e^{i(E_f-E_i)t}\,(\phi_f,H'e^{ie\vec{A}\cdot\vec{x}}\phi_i)\ . \tag{26}$$

The matrix element in the integrand of Eq. (26) depends on t through the presence of $H'\exp(ie\vec{A}\cdot\vec{x})$. This suggests the definition

$$T_{fi}^{(n)}\equiv\text{term in }(\phi_f,H'e^{ie\vec{A}\cdot\vec{x}}\phi_i)\quad\text{proportional to } e^{in\omega t}. \tag{27}$$

This definition is consistent with the usual concept of a T matrix, and leads to

$$\tau_{fi}=-2\pi i\sum_{n=-\infty}^{\infty}\delta(E_f-E_i+n\omega)T_{fi}^{(n)}\ . \tag{28}$$

Finally, the transition probability per unit time w_{fi} between an initial state Φ_i and a final state Φ_f is given by

$$w_{fi}=2\pi\sum_n|T_{fi}^{(n)}|^2\delta(E_f-E_i+n\omega)\ . \tag{29}$$

The transition probability of Eq. (29), although written in terms of a sum over n from $-\infty$ to $+\infty$, will give nonvanishing contributions only for those n for which $n\omega$ can match the energy difference E_i-E_f. Whether this is an infinite, a finite, or a null set of terms, depends on the physical problem.

V. INTENSE-FIELD TRANSITIONS

A. Commutator Theorem

The practical calculation of τ_{fi} is greatly facilitated by a simple device which has a close analogy in perturbation theory. The idea is to replace the $H'\exp(ie\vec{A}\cdot\vec{x})$ operator by a commutator involving H_0 as one of its factors. When H_0 operates on Φ_f and Φ_i in τ_{fi}, an energy difference factor appears. We shall evaluate the commutator

$$[H_0,\exp(ie\vec{A}\cdot\vec{x})]=(2m)^{-1}\times\{\vec{p}[\vec{p},\exp(ie\vec{A}\cdot\vec{x})]+[\vec{p},\exp(ie\vec{A}\cdot\vec{x})]\vec{p}\}\ ,$$

which holds for $H_0=(2m)^{-1}p^2+V(\vec{r})$. Then, since

$$[p_i,\exp(ie\vec{A}\cdot\vec{x})]=[eA_i+ex_j(\partial_iA_j)]\exp(ie\vec{A}\cdot\vec{x})\ ,$$

we get, after some rearrangement,

$$[H_0,\exp(ie\vec{A}\cdot\vec{x})]=-H'\exp(ie\vec{A}\cdot\vec{x})+\exp(ie\vec{A}\cdot\vec{x})[H_I-e(\partial_tA_i)x_i]\ . \tag{30}$$

H_I is given in Eq. (12), and the discussion following that equation led to the conclusion that $H_I-e(\partial_tA_i)x_i$ is small.

First we wish to show what happens to Eq. (30) in the perturbation limit. In a perturbation calculation, H' consists of the single term $m^{-1}e\vec{A}\cdot\vec{p}$, so that (in the long-wavelength case) the $H'\exp(ie\vec{A}\cdot\vec{x})$ term clearly dominates the second term in Eq. (30). To lowest order in $e\vec{A}\cdot\vec{x}$, Eq. (30) then becomes

$$[H_0,ie\vec{A}\cdot\vec{x}]\approx-H'\ ,$$

or, in more familiar form,

$$[H_0,\vec{x}\cdot\vec{\epsilon}]=-im^{-1}\vec{p}\cdot\vec{\epsilon}\ , \tag{31}$$

where $\vec{\epsilon}$ is the polarization vector of the $\vec{A}$ field.

Equation (30) is really useful in the nonperturbative problem only if the last term can be neglected. In the general case, H' contains $(2m)^{-1}e^2A^2$ as well as $m^{-1}e\vec{A}\cdot\vec{p}$. From the same type of order-of-magnitude analysis employed earlier, the last term in Eq. (30) will be small as compared to $(2m)^{-1}e^2A^2$ if

$$(\omega/m)\ll(ea/2m)\ . \tag{32}$$

This is true only for intense fields. It will be employed subsequently for the practical examples which are given because it simplifies the calculations so much, and because we shall be seeking to identify explicit intensity effects, i.e., we shall be exploring cases where Eq. (32) will surely be valid. It must be clearly understood, however, that Eq. (32) is not essential to the general method, and represents the only case encountered in this paper where an intensity parameter must be directly compared with a wavelength parameter. The result to be employed below is then

$$[H_0,\exp(ie\vec{A}\cdot\vec{x})]\approx-H'\exp(ie\mathrm{A}\cdot\vec{x})\ . \tag{33}$$

With Eq. (33), the matrix element which defines τ_{fi} as given in Eq. (26) is

$$(\phi_f,H'e^{ie\vec{A}\cdot\vec{x}}\phi_i)\approx(E_i-E_f)(\phi_f,e^{ie\vec{A}\cdot\vec{x}}\phi_i)\ . \tag{34}$$

B. Angular Momentum Reduction

For any central potential problem, the matrix element on the right-hand side of Eq. (34) can be reduced to a form involving only radial wave functions. For convenience, take $\vec{A}$ to be plane polarized in the z direction, which leads to the partial-wave expansion

$$e^{ie\vec{A}\cdot\vec{x}} = e^{ieAz} = \sum_{l=0}^{\infty} (2l+1) i^l j_l(eAr) P_l(\cos\theta) \ .$$

Then, since the solutions for a central potential can always be written as the product of a radial wave function and a spherical harmonic,

$$(\phi_f, e^{ie\vec{A}\cdot\vec{x}}\phi_i) = \sum_{l=0}^{\infty} (2l+1) i^l \int_0^\infty r^2 dr R_f^*(r) R_i(r) j_l(eAr) \int d\Omega\, Y_{l_f}^{m_f *}(\theta,\varphi) Y_{l_i}^{m_i}(\theta,\varphi) P_l(\cos\theta) \ . \tag{35}$$

The solid-angle integral of the product of three spherical harmonics is well-known and can be expressed as[6]

$$\int d\Omega Y_{l_f}^{m_f *} Y_{l_i}^{m_i} Y_l^0 = (-)^{m_f} \left(\frac{(2l+1)(2l_i+1)(2l_f+1)}{4\pi}\right)^{1/2} \begin{pmatrix} l & l_i & l_f \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l & l_i & l_f \\ 0 & m_i & -m_f \end{pmatrix} \tag{36}$$

The properties of the Wigner 3-j coefficients give the selection rules

$$m_i = m_f \ , \qquad l + l_i + l_f = \text{even integer} \ , \qquad |l_i - l_f| \le l \le l_i + l_f \ . \tag{37}$$

Equations (35) and (36) yield the general result

$$(\phi_f, e^{ie\vec{A}\cdot\vec{x}}\phi_i) = \sum_{l=0}^{\infty} (2l+1) i^l [(2l_i+1)(2l_f+1)]^{1/2} (-)^{m_i} \begin{pmatrix} l & l_i & l_f \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l & l_i & l_f \\ 0 & m_i & -m_i \end{pmatrix} \int_0^\infty r^2 dr R_f^*(r) R_i(r) j_l(eAr) \ . \tag{38}$$

For transitions between any given pair of states, ϕ_i and ϕ_f, Eq. (38) gives an explicit expression for the matrix element, provided only that the radial wave functions are known for the problem in the absence of the external electromagnetic field. The electromagnetic field parameters enter the problem only in the argument of the spherical Bessel function.

We can simplify the result still further if we consider the class of problems in which the initial state is an s state $l_i = 0$. Not only does this restriction require $m_i = 0$, but the conditions of Eq. (37) stipulate that the l summation reduces to the single term $l = l_f$, i.e., the angular momentum supplied by the electromagnetic field must provide the entire angular momentum of the final state. The $l_i = 0$ case of Eq. (38) is then

$$(\phi_f, e^{ie\vec{A}\cdot\vec{x}}\phi_i)_{l_i=0} = i^{l_f} (2l_f+1)^{1/2} \times \int_0^\infty r^2 dr R_f^*(r) R_i(r) j_{l_f}(eAr) \ . \tag{39}$$

C. Hydrogen Atom Example

To illustrate the procedures involved in practical application of the momentum-translation method, we shall consider the example of 1s to 2s transition in hydrogenlike atoms. The relevant matrix element is, from Eq. (39),

$$(\phi_{2s}, e^{ie\vec{A}\cdot\vec{x}}\phi_{1s}) = \int_0^\infty r^2 dr R_{2s}^*(r) R_{1s}(r) j_0(eAr) \ . \tag{40}$$

The integral is readily done in closed form with hydrogen atom radial wave functions, to yield the result

$$(\phi_{2s}, e^{ie\vec{A}\cdot\vec{x}}\phi_{1s}) = \tfrac{8}{3}\sqrt{2}(\tfrac{2}{3})^3 (\tfrac{2}{3}a_0)^2 \times e^2A^2[1 + (\tfrac{2}{3}a_0)^2 e^2A^2]^{-3} \ ,$$

where a_0 is the Bohr radius of a hydrogenlike atom of nuclear charge Ze. With $E_{1s} - E_{2s} = -\tfrac{3}{8}m^{-1}a_0^{-2}$

and Eq. (34), the matrix element required for the evaluation of the T matrix defined in Eq. (27) is

$$(\phi_{2s}, H' e^{ie\vec{A}\cdot\vec{x}}\phi_{1s}) = -\sqrt{2}\, m^{-1}(\tfrac{2}{3})^5 \times e^2A^2[1+(\tfrac{2}{3}a_0)^2 e^2A^2]^{-3} \quad . \quad (41)$$

We now substitute the dipole-approximation plane-wave form for eA

$$eA \approx ea\cos\omega t = \tfrac{1}{2}ea\beta \quad ,$$

where $\beta = e^{i\omega t} + e^{-i\omega t}$ and a is the amplitude of the vector potential. To accomplish the identification of the coefficient of $e^{in\omega t}$ required by Eq. (27), we first expand $b^2\beta^2[1+b^2\beta^2]^{-3}$ in powers of β, where

$$b = \tfrac{1}{3}eaa_0 \quad , \quad (42)$$

and β is the only time-dependent parameter appearing on the right-hand side of Eq. (41). The binomial theorem for negative integer powers

$$(1+x)^{-n} = \sum_{k=0}^{\infty}\binom{n+k-1}{k}(-x)^k$$

gives the result

$$\frac{b^2\beta^2}{(1+b^2\beta^2)^3} = \sum_{k=0}^{\infty}\binom{k+2}{k}(-)^k(b^2\beta^2)^{k+1} \quad . \quad (43)$$

The next step is to employ the usual binomial theorem to expand $\beta^{2k+2} = (e^{i\omega t}+e^{-i\omega t})^{2k+2}$, i.e.,

$$\beta^{2k+2} = \sum_{j=0}^{2k+2}\binom{2k+2}{j}e^{2i\omega t(k-j+1)} \quad . \quad (44)$$

In principle we must now single out the $e^{in\omega t}$ term from this series. For this $1s-2s$ transition, however, only even powers of $e^{i\omega t}$ can occur, corresponding to the requirements of angular momentum conservation in a transition between s states. Furthermore, we are calculating an absorption process where $E_f > E_i$. We shall thus extract the coefficient of the negative even power term $e^{-2in\omega t}$ from Eq. (43). From Eq. (44), this imposes the constraint $k-j+1=-n$. The required $e^{-2in\omega t}$ coefficient in Eq. (43) is

$$\left(\frac{b^2\beta^2}{(1+b^2\beta^2)^3}\right)^{(-2n)} = e^{-2in\omega t}\sum_{k=n-1}^{\infty}\binom{k+2}{k} \times \binom{2k+2}{n+k+1}(-)^k b^{2k+2} \quad ,$$

where the lower limit on the k sum follows from the upper limit $j \leq 2k+2$ in Eq. (44), combined with $j = n+k+1$. If the index of summation is shifted so the sum starts with zero index, and if appropriate factors are placed outside the summand to make the initial term have unit value, then

$$\left(\frac{b^2\beta^2}{(1+b^2\beta^2)^3}\right)^{(-2n)} = e^{-2in\omega t}\tfrac{1}{2}[n(n+1)](-)^{n+1} \times b^{2n}\sum_{k=0}^{\infty}\binom{n+1+k}{n+1}\binom{n+k}{n}\binom{2n+2k}{k}(-)^k b^{2k} \quad . \quad (45)$$

The required result has now been achieved for the transition from the $1s$ to $2s$ state in a hydrogenlike atom by a plane-wave field of frequency such that $E_{2S} - E_{1S} = 2n\omega$. The transition probability per unit time is given by a special case of Eq. (29),

$$w_{2s,1s} = 2\pi\left|T_{2s,1s}^{(-2n)}\right|^2\delta(E_{2S}-E_{1S}-2n\omega) \quad ,$$

where $T_{2s,1s}^{(-2n)} = \frac{1}{\sqrt{2}\,ma_0^2}(\tfrac{2}{3})^3 n(n+1)(-)^n(\tfrac{1}{3}eaa_0)^{2n} \times \sum_{k=0}^{\infty}\binom{n+1+k}{n+1}\binom{n+k}{n}\binom{2n+2k}{k}(-)^k(\tfrac{1}{3}eaa_0)^{2k}$, (46)

which follows from Eqs. (41), (42), and (45).

Equation (46) demonstrates several important points. The intensity of the electromagnetic field enters in the form $(\frac{1}{3}eaa_0)^2$. The $\frac{1}{3}$ in this parameter is specific to the $1s-2s$ transition just calculated, but the occurrence of the field intensity $(ea)^2$ multiplied by the square of the size of the bound system is an essential characteristic. As remarked earlier, this intensity parameter is on the order of $(137)^2$ times larger than the $e^2a^2/2m^2$ parameter that occurs in free-electron problems. Another point to be made concerns the radius of convergence of the sum in Eq. (46). This radius of convergence is (e.g., from the ratio test) $eaa_0 < \frac{3}{2}$. This domain is amply large enough to encompass intensity effects which can dramatically change the qualitative aspects of semiclassical radiation theory which are familiar to us from perturbation theory. Another aspect of the sum in Eq. (46) is that the numerical coefficient which multiplies the successive powers of $(\frac{1}{3}eaa_0)^2$ has a generally rather flat behavior with k, and exhibits an increase in the early part of the series. This causes an early onset of intensity effects.

To illustrate the influence of such intensity effects on hypothetical experimental observations,

consider the power-law dependence of $|T_{2s,1s}^{(-2n)}|^2$ (or the transition probability) on the field intensity. For convenience, set $x = (\frac{1}{3}eaa_0)^2$ From perturbation theory, we would get the simple result

$$\frac{d(\ln|T|^2)}{d(\ln x)} = 2n$$

for a transition of order $2n$. From Eq. (46), we get

$$\frac{d(\ln|T_{2s,1s}^{(-2n)}|^2)}{d(\ln x)} = 2n - \frac{4(n+1)(n+2)}{n}x + \cdots , \tag{47}$$

where we have displayed the effect of only the first two terms in the series of Eq. (46). Thus, the sum over all higher-order corrections (which is what the series represents) has the effect (for achievable intensities) of reducing the power-law dependence of transition probability on intensity to something less than the lowest order of the transition. Apparently this type of effect has already been observed[1] in ionization processes.

We wish to emphasize the significance of the appearance of the series in Eq. (46). This power series in the intensity parameter is associated specifically with the $e^{-2in\omega t}$ factor, and the factor $e^{-2i\omega t}$ raised to any other power also has an infinite power series in the intensity associated with it. In perturbationlike language, we can say that each energy-conserving transition comes about not only through a direct transition between initial and final states through the absorption of $2n$ photons; but that the contribution of all other processes of higher order which go to the final state by way of real or virtual states above or below the final state, are all summed in Eq. (46). The validity of this statement will be shown in a rather transparent way later in this paper, when general comparisons with perturbation theory are made.

Some of the limitations of Eq. (46) should also be noted. It is specific to the $1s-2s$ transition in hydrogen caused by an electromagnetic field whose photons are of such frequency that $2n$ of them provide precisely the difference $E_{2S}-E_{1S}$. Since it is not likely that an extremely intense source satisfying that condition will be available, a more realistic case to treat is that in which the intense-field photons do not exactly match the energy level difference, and an additional photon of different frequency must be considered in order to satisfy energy conservation. This problem will be treated in Sec. VI.

VI. COMBINED INTENSE- AND WEAK-FIELD EFFECTS

A. Extended Formalism

The problem to be considered now contains two plane-wave electromagnetic fields, with vector potentials denoted by $\vec{A}$ and $\vec{A}'$. Although $\vec{A}'$ is eventually to be regarded as a weak field, with higher orders neglected, it is actually most convenient at the outset to treat it in exactly the same way as the intense field $\vec{A}$. The results of Eqs. (33) and (34) can then be applied directly, with the simple substitution $\vec{A} \to \vec{A} + \vec{A}'$. Angular momentum reduction can be carried out in substantially the same way as for a single field. This is done in the Appendix.

B. Generalized Raman Process in Hydrogen

We shall again treat the $1s-2s$ transition in hydrogen, but we shall not require that the intense field be of such frequency that $E_{2S}-E_{1S}=2n\omega$. Rather, the conservation condition will emerge as $E_{2S}-E_{1S}=(2n+1)\omega-\omega'$ for absorption from the $1s$ level, or $E_{2S}-E_{1S}=(2n-1)\omega+\omega'$ for emission from the $2s$ level, where ω' is simply the frequency of a Raman photon required to satisfy energy conservation. The field $\vec{A}'$ of frequency ω' can certainly be treated as weak.

From the Appendix, the matrix element to be evaluated is

$$(\phi_f, \exp[ie(\vec{A}+\vec{A}')\cdot\vec{x}]\phi_i) = \int_0^\infty r^2 dr R_f^*(r)R_i(r)\sum_{l=0}^{\infty}(-)^l(2l+1) \times P_l(\vec{\epsilon}\cdot\vec{\epsilon}')j_l(eAr)j_l(eA'r) . \tag{48}$$

In accordance with the weak-field assumption for $\vec{A}'$, we retain only the j_0 and j_1 functions of $eA'r$, with $j_0(eA'r)\approx 1$ and $j_1(eA'r)\approx\frac{1}{3}eA'r$. Hence, Eq. (48) reduces to the two terms

$$(\phi_f, \exp[ie(\vec{A}+\vec{A}')\cdot\vec{x}]\phi_i) \approx \int_0^\infty r^2 dr\, R_f^*(r)R_i(r)j_0(eAr) - eA'\vec{\epsilon}\cdot\vec{\epsilon}'\int_0^\infty r^3 dr R_f^*(r)R_i(r)j_1(eAr) , \tag{49}$$

where the first term is precisely that which arose [Eq. (40)] in the problem with the intense field alone present. By hypothesis, this first term cannot satisfy energy conservation requirements, and it will therefore not contribute.

The integration indicated in the second term in

Eq. (49) is elementary for hydrogenic radial wave functions. It has the result

$$\int_0^\infty r^3 dr R_f^*(r) R_i(r) j_1(eAr) = 16\sqrt{2}\left(\tfrac{2}{3}\right)^4 a_0 b\beta\left[\tfrac{2}{3}(1+b^2\beta^2)^{-3} - (1+b^2\beta^2)^{-4}\right] \quad , \tag{50}$$

where b and β are as defined earlier [see the paragraph containing Eq. (42)], and the same result obtains for transitions in either direction between the $1s$ and $2s$ states. As in Eq. (43), we get

$$\frac{2}{3}\frac{b\beta}{(1+b^2\beta^2)^3} = \frac{2}{3}\sum_{k=0}^{\infty}\binom{k+2}{k}(-)^k(b\beta)^{2k+1} \quad ,$$

and by a similar procedure,

$$\frac{b\beta}{(1+b^2\beta^2)^4} = \sum_{k=0}^{\infty}\binom{k+3}{k}(-)^k(b\beta)^{2k+1} \quad .$$

The complete expansion in powers of β is thus

$$b\beta\left(\frac{2}{3}\frac{1}{(1+b^2\beta^2)^3} - \frac{1}{(1+b^2\beta^2)^4}\right) = -\sum_{k=0}^{\infty}\frac{(k+1)(k+2)!}{6k!}(-)^k(b\beta)^{2k+1} \quad , \tag{51}$$

which contains only odd powers of β, as expected. The required odd-power analog of Eq. (44) is

$$\beta^{2k+1} = \sum_{j=0}^{2k+1}\binom{2k+1}{j}e^{i\omega t(2k-2j+1)} \quad . \tag{52}$$

We now make a distinction between $1s \rightarrow 2s$ and $2s \rightarrow 1s$ transitions. For absorption of $2n+1$ photons from the $\vec{A}$ field by an atom initially in the $1s$ state (with concurrent emission of a single photon into the $\vec{A}'$ field), we must identify the coefficient of $e^{-(2n+1)\omega t}$, i.e., we must set $2k-2j+1=-(2n+1)$ in Eq. (52). This leads to $j=k+n+1$, so that Eq. (51) becomes

$$b\beta\left(\frac{2}{3}\frac{1}{(1+b^2\beta^2)^3} - \frac{1}{(1+b^2\beta^2)^4}\right)(-2n-1) = e^{-(2n+1)i\omega t}\tfrac{1}{6}(n+1)^2(n+2)$$

$$\times(-)^{n+1}b^{2n+1}\sum_{k=0}^{\infty}\left(\frac{n+1+k}{n+1}\right)2\left(\frac{n+2+k}{n+2}\right)\binom{2n+2k+1}{k}(-)^k b^{2k} \quad , \tag{53}$$

where again the series has been arranged to have unit value for the $k=0$ term. When combined with Eqs. (49), (50), and (34), Eq. (53) leads to the T-matrix element

$$T_{2s,1s}^{(-2n-1,\,+1)} = \frac{\sqrt{2}}{ma_0^2}\left(\tfrac{2}{3}\right)^3 e^{i\alpha}(\vec{\epsilon}\cdot\vec{\epsilon}')(n+1)^2(n+2)(-)^{n+1}(\tfrac{1}{3}ea'a_0)(\tfrac{1}{3}eaa_0)^{2n+1}$$

$$\times\sum_{k=0}^{\infty}\left(\frac{n+1+k}{n+1}\right)^2\left(\frac{n+2+k}{n+2}\right)\binom{2n+2k+1}{k}(-)^k(\tfrac{1}{3}eaa_0)^{2k} \quad , \tag{54}$$

where the parenthetical superscript on the T matrix indicates absorption of $2n+1$ ω photons and emission of a single ω' photon, and $e^{i\alpha}$ arises from the relative phase between the $\vec{A}$ and $\vec{A}'$ fields.

All of the remarks made about Eq. (46) apply to Eq. (54) as well, including the statement that the radius of convergence of the power series in b^2 is given by $eaa_0 < \frac{3}{2}$. The power-law dependence of the transition probability on the electromagnetic field intensity deviates more markedly from perturbation theory results

in Eq. (54) than was the case in Eq. (46). The power-law dependence which follows from Eq. (54) is, for the first two terms of the expansion,

$$\frac{d(\ln|T_{2s,1s}^{(-2n-1,+1)}|^2)}{d(\ln x)} = (2n+1) - \frac{2(n+2)(n+3)(2n+3)}{(n+1)^2}\, x + \cdots , \tag{55}$$

where $x = b^2$ as before. To be explicit, suppose that the $\vec{A}$ field arises from a Q-switched Nd-glass laser, with an output of 10^9 W, which is focused down to a beam of 10^{-4} cm^2 in cross section.[7] Then, for hydrogen ($Z = 1$), the intensity parameter x (or b^2) is $x \approx 1.5 \times 10^{-2}$. The $2s - 1s$ energy difference in hydrogen is 10.2 eV, and a Nd-glass laser photon has about 1.17 eV, so $2n+1 = 9$ and $\omega' = 0.3$ eV. The power-law coefficient is, from Eq. (55), $9 - (36.96)(0.015) = 8.4$.

Apart from the basic fact of deviation from $2n+1$ behavior, two important points must be made. One is that an increase of about an order of magnitude in laser power would make the second term in Eq. (55) comparable to the first term, and moderate further increases in power would bring terms of higher order in b^2 to prominence. Under such circumstances, our intuition about transitions involving photons would tend to fail, since such intuition is based upon perturbation-theoretic concepts of the dominance of lowest-order processes. If the first term in Eq. (55) is not the most important, this is evidence that the sum of all higher-order processes overshadows the lowest-order process. The second point to be made is that Eq. (55) is specific to the $1s - 2s$ generalized Raman transition in a hydrogenlike atom. The explicit form of the correction term will be different, in general, for each transition considered. That is, if we write

$$\frac{d(\ln|T|^2)}{d(\ln x)} = (2n+1) - \Delta ,$$

then Δ depends on n, on b^2, and on the process. Thus, it is likely that other processes than the one considered here might show stronger deviations from perturbative behavior.

Results for emission from the $2s$ level will be quoted here, since they are obtained very easily. The T matrix for emission of $2n-1$ photons of energy ω and one photon of energy ω' is

$$T_{2s,1s}^{(2n-1,+1)} = \frac{\sqrt{2}}{ma_0^2}\left(\tfrac{2}{3}\right)^3 e^{i\alpha}(\vec{\epsilon}\cdot\vec{\epsilon}')n^2(n+1)(-)^{n+1}(\tfrac{1}{3}ea'a_0)(\tfrac{1}{3}eaa_0)^{2n-1} \times \sum_{k=0}^{\infty}\left(\frac{n+k}{n}\right)^2\left(\frac{n+1+k}{n+1}\right)\binom{2n+2k-1}{k}(-)^k(\tfrac{1}{3}eaa_0)^{2k} . \tag{56}$$

Equation (56) can be obtained directly from Eq. (54) if the substitution $n+1 \to n$ is made in Eq. (54), and the over-all sign is changed. This happens because of the symmetry of the radial integral between final and initial states, and the symmetry of $\vec{A}$ between $e^{i\omega t}$ and $e^{-i\omega t}$. The over-all sign change is a consequence of the antisymmetry of the $E_i - E_f$ factor.

VII. RELATION TO PERTURBATION THEORY

A. Low-Intensity Limit

The method developed in this paper is designed explicitly for many-photon problems, and it retains its validity up to very high field intensity. Nevertheless, there are meaningful and instructive comparisons which can be made with a theory designed for low field intensity, i.e., perturbation theory.

Consider the low-intensity limit of the basic matrix element of Eq. (24). For $|\vec{A}| \to 0$, the perturbing Hamiltonian $H' \to -m^{-1}e\vec{A}\cdot\vec{p}$. In the momentum-translation approximation, $\Psi_i(t) \to \Phi_i(t)$ to lowest order in $\vec{A}$, and so

$$(\Phi_f, H'\Psi_i) \to (\Phi_f, -m^{-1}e\vec{A}\cdot\vec{p}\Phi_i) , \tag{57}$$

which is the usual first-order matrix element of semiclassical radiation theory. Thus, the momentum-translation approximation yields first-order perturbation theory when the low-intensity limit is taken.

For a second-order result from the momentum-translation approximation, we have

$$(\Phi_f, H'\Psi_i) \approx (\Phi_f, H'\Phi_i) + (\Phi_f, (-m^{-1}e\vec{A}\cdot\vec{p})(ie\vec{A}\cdot\vec{x})\Phi_i) \quad , \quad (58)$$

where in the second inner product an expansion of the exponential in Eq. (13) has been used. H' as it appears in the first term of Eq. (58) is the complete H' of Eq. (14). The second term of Eq. (58) can be resolved into a more familiar form by inserting a complete set of Φ states,

$$(\Phi_f, (-m^{-1}e\vec{A}\cdot\vec{p})(ie\vec{A}\cdot\vec{x})\Phi_i) = \sum_n (\Phi_f, -m^{-1}e\vec{A}\cdot\vec{p}\,\Phi_n)(\Phi_n, ieA\vec{x}\cdot\vec{\epsilon}\,\Phi_i) \quad .$$

We can then invoke the dipole approximation for $e\vec{A}$ (i.e., view $e\vec{A}$ as a function of time only), and employ Eq. (31) in an inverse sense, by using it to introduce $\vec{p}\cdot\vec{\epsilon}$ in place of $\vec{x}\cdot\vec{\epsilon}$. This gives the result

$$(\Phi_f, (-m^{-1}e\vec{A}\cdot\vec{p})(ie\vec{A}\cdot\vec{x})\Phi_i) = \sum_n \frac{(\Phi_f, -m^{-1}e\vec{A}\cdot\vec{p}\Phi_n)(\Phi_n, -m^{-1}e\vec{A}\cdot\vec{p}\Phi_i)}{E_n - E_i} \quad . \quad (59)$$

We can see from Eq. (59) that the momentum-translation approximation does not reproduce second-order time-dependent perturbation theory. It has, instead, very much the appearance of a second-order time-independent perturbation, except that $\vec{A}$ has a time-oscillatory behavior which will manifest itself when the matrix element of Eq. (59) is employed in Eq. (24).

Further development of the exponential function in the matrix element $(\Phi_f, H' e^{ie\vec{A}\cdot\vec{x}}\Phi_i)$ leads to expressions resembling, but not equivalent to, perturbation expressions to all orders. This is not surprising. Because the momentum-translation approximation is designed around a low-frequency approximation, it should not be expected to reproduce the intensity-parameter expansion which perturbation theory is. Because there are no *a priori* intensity restrictions in the momentum-translation approach [other than the condition of Eq. (16a), which is not a stringent condition on intensity and which can always be satisfied if ω/E is small enough, regardless of the magnitude of eaa_0], this approach should contain $\vec{A}$ to all orders because of the obvious fact that electrodynamics is nonlinear to arbitrarily high order when no intensity constraints are imposed. Thus, the momentum-translation method contains "diagrams," or elementary processes of all orders involving the electromagnetic field, albeit in a fashion which is accurate only when Eqs. (16a) and (16b) are satisfied.

Now consider the correction to the momentum-translation approximation which follows from Eqs. (8) and (18). The matrix element of Eq. (24) with this correction is

$$(\Phi_f, H'\Psi_i) = (\Phi_f, H' e^{ie\vec{A}\cdot\vec{x}}\Phi_i) - i\sum_n (\Phi_f, H' e^{ie\vec{A}\cdot\vec{x}}\Phi_n) \times \int_{-\infty}^{t} (\Phi_n(t'), H_I(t')\Phi_i(t'))dt' \quad . \quad (60)$$

The first term in Eq. (59) gives the only contribution as $|\vec{A}| \to 0$, and leads to the same outcome as Eq. (57). Thus, the corrected momentum-translation approximation reduces to first-order perturbation theory for weak fields. Since H' and H_I both involve $\vec{A}$, a result to second order in intensity in Eq. (60) is achieved by keeping only the leading terms in H' and H_I, and replacing $e^{ie\vec{A}\cdot\vec{x}}$ by unity in $(\Phi_f, H' e^{ie\vec{A}\cdot\vec{x}}\Phi_n)$. That is, we set $H' \approx -m^{-1} e\vec{A}\cdot\vec{p}$ and $H_I \approx e(\partial_t\vec{A})\cdot\vec{x}$ [see the discussion in the paragraph following Eq. (12)]. The integral over t in Eq. (60) can be rearranged using integration by parts and the presumption (consistent with the S-matrix formalism) that the $\vec{A}$ field is "turned off" at $t = -\infty$, to yield

$$\int_{-\infty}^{t} (\Phi_n, e(\partial_{t'}\vec{A})\cdot\vec{x}\,\Phi_i)dt' = (\Phi_n, e\vec{A}\cdot\vec{x}\,\Phi_i) - i(E_n - E_i)\int_{-\infty}^{t} (\Phi_n, e\vec{A}\cdot\vec{x}\,\Phi_i)dt' \quad .$$

Equation (31) can be employed in this last term to give

$$i(E_n - E_i)\int_{-\infty}^{t} (\Phi_n, e\vec{A}\cdot\vec{x}\,\Phi_i)dt' = \int_{-\infty}^{t} (\Phi_n, -m^{-1}e\vec{A}\cdot\vec{p}\,\Phi_i)dt' \quad .$$

If we insert these results in Eq. (60), we get

$$(\Phi_f, H'\Psi_i) \approx (\Phi_f, H' e^{ie\vec{A}\circ\vec{x}}\Phi_i) - i(\Phi_f, (-m^{-1}e\vec{A}\circ\vec{p})(e\vec{A}\cdot\vec{x})\Phi_i)$$

$$- i \sum_n (\Phi_f, - m^{-1} e\vec{A}\cdot\vec{p}\,\Phi_n) \int_{-\infty}^{t} (\Phi_n, - m^{-1} e\vec{A}\cdot\vec{p}\,\Phi_i) dt' \quad . \tag{61}$$

The first term in Eq. (61) can be treated as in Eq. (58) to give the final result

$$(\Phi_f, H'\Psi_i) \approx (\Phi_f, (- m^{-1} e\vec{A}\cdot\vec{p} + \tfrac{1}{2} m^{-1} e^2 A^2)\Phi_i) - i \sum_n (\Phi_f, - m^{-1} e\vec{A}\cdot\vec{p}\,\Phi_n) \int_{-\infty}^{t} (\Phi_n, - m^{-1} e\vec{A}\cdot\vec{p}\,\Phi_i) dt'$$

which is identically the result of second-order time-dependent perturbation theory.

Successively higher-order approximations to Eq. (17) will give successively higher-order correspondences with perturbation theory. We have already seen that the zeroth-order approximation to Eq. (17) reduces to first-order perturbation theory for small field intensity, and the first-order approximation to Eq. (17) reduces to second-order perturbation theory. This pattern continues to higher orders. It is important to keep in mind that when no low-intensity limit is taken, *all* orders of approximation to Eq. (17) lead to accurate high-intensity results.

B. Physical Interpretation of Two-Field Problem

We have seen above in what manner the present method reduces to perturbation theory when one considers the intensity of the electromagnetic field to be weak. However, much more physical insight into the nature of the momentum-translation approximation can be achieved by avoiding the low-intensity limit, and, instead, casting the complete results in a form which makes possible a very instructive physical interpretation.

Let us consider the case of two plane-wave fields $\vec{A}$ and $\vec{A}'$, where we shall consider $\vec{A}$ to be arbitrarily intense and $\vec{A}'$ to be weak. The interaction Hamiltonian is

$$H' = - m^{-1} e(\vec{A}\cdot\vec{p} + \vec{A}'\circ\vec{p}) + (2m)^{-1} e^2 (\vec{A} + \vec{A}')^2 \quad . \tag{62}$$

In Eq. (62), the term $(2m)^{-1}e^2A'^2$ can be neglected, but not $m^{-1}e^2\vec{A}\cdot\vec{A}'$, which can be of comparable importance to $m^{-1}e\vec{A}'\cdot\vec{p}$. The $\vec{A}\cdot\vec{A}'$ term is troublesome, since it intimately couples the two fields. However, H' does not appear in the theory by itself, but in the combination $H' \exp[ie(\vec{A}\circ\vec{x} + \vec{A}'\cdot\vec{x})]$. The awkward $\vec{A}\cdot\vec{A}'$ term in H' can be removed by commuting $e^{ie\vec{A}'\cdot\vec{x}}$ to the left of H'. This commutation operation leads to

$$H' \exp[ie(\vec{A} + \vec{A}')\circ\vec{x}] = \exp(ie\vec{A}'\circ\vec{x})[- m^{-1} e(\vec{A} + \vec{A}')\circ\vec{p} + (2m)^{-1} e^2 (A^2 - A'^2)] \exp(ie\vec{A}\circ\vec{x})$$

where the dipole approximation has been invoked to neglect a term $ex_j(\partial A'_j)$. Now we can drop the A'^2 term, so we achieve the convenient form

$$H' \exp[ie(\vec{A} + \vec{A}')\cdot\vec{x}] \approx \exp(ie\vec{A}'\cdot\vec{x}) \times [H'(A) + V'] \exp(ie\vec{A}\cdot\vec{x}) \quad ,$$

where $H'(A) = - m^{-1} e\vec{A}\cdot\vec{p} + (2m)^{-1} e^2 A^2$

is the interaction Hamiltonian with the $\vec{A}$ field alone present, and

$$V' = - m^{-1} e\vec{A}'\cdot\vec{p}$$

is the usual perturbation Hamiltonian that would normally be used for the weak field $\vec{A}'$ alone. If we now employ $\exp(ie\vec{A}'\cdot\vec{x}) \approx 1 + ie\vec{A}'\cdot\vec{x}$, the basic matrix element of Eq. (24) becomes

$$(\Phi_f, H'\Psi_i) \approx (\Phi_f, H'(A)\Psi_i(A)) + (\Phi_f, ie\vec{A}'\circ\vec{x} H'(A)\Psi_i(A)) + (\Phi_f, V'\Psi_i(A)) \quad . \tag{63}$$

where $\Psi(A)$ denotes the momentum-translation approximation wave function which pertains to $\vec{A}$ alone, i.e., $\Psi(A) = e^{ie\vec{A}\circ\vec{x}}\Phi$. The second term in Eq. (63) can be transformed by introducing a complete set of Φ states between $ie\vec{A}'\circ\vec{x}$ and $H'(A)$, and then using Eq. (31) to get

$$(\Phi_f, ie\vec{A}\circ\vec{x}\,\Phi_n) = (E_n - E_f)^{-1} (\Phi_f, V'\Phi_n) \quad .$$

The third term in Eq. (63) can also have a complete set of Φ states introduced between V' and $\Psi_i(A)$. Equation (63) then takes the form

$$(\Phi_f, H'\Psi_i) \approx (\Phi_f, H'(A)\Psi_i(A)) + \sum_n \frac{(\Phi_f, V'\Phi_n)(\Phi_n, H'(A)\Psi_i(A))}{E_n - E_f} + \sum_n (\Phi_n, \Psi_i(A))(\Phi_f, V'\Phi_n) \quad . \tag{64}$$

The first term in Eq. (64) describes the transitions caused by the $\vec{A}$ field alone, e.g., as calculated in Sec. V. As emphasized in the foregoing discussion, this term describes $\vec{A}$ interactions to all orders.

The second term in Eq. (64) strongly resembles second-order perturbation theory. It can be interpreted as representing a two-step process. The

initial state $\Psi_i(A)$ in the presence of the intense $\vec{A}$ field gives rise to virtual transitions to Φ_n intermediate states. [Note the similarity of the matrix element $(\Phi_n, H'(A)\Psi_i(A))$ to the first term of Eq. (64).] The A' field, through its interaction Hamiltonian V', then makes possible the second step in the transition to Φ_f. The energy denominator, $E_n - E_f$, involves the difference in energy between the two states connected by the weak field $\vec{A}'$, retained here only to first order.

The third term in Eq. (64) has a very direct interpretation. Suppose we first focus attention on the $\vec{A}'$ field only, and write the state of the atom in the presence of $\vec{A}'$ [which we denote $\Phi(A')$] as a superposition of Φ states,

$$\Phi(A') = \sum c_n(t)\Phi_n \ .$$

The expansion coefficient follows from direct application of the Schrödinger equation as

$$c_f(t) = -i \int dt \sum_n c_n(t)(\Phi_f, V'\Phi_n) \ . \tag{65}$$

Standard time-dependent perturbation theory can be developed from Eq. (65) by substituting an assumed initial distribution for $c_n(t)$ on the right-hand side. We do not use a perturbative approach here, but remark instead that the coefficients $c_f(t)$ correspond to the τ matrix of Eq. (24), and the contribution of the third term of Eq. (64) to the τ matrix is

$$\tau_{fi} = -i \int dt \sum_n (\Phi_n, \Psi_i(A))(\Phi_f, V'\Phi_n) \ . \tag{66}$$

Comparison of Eqs. (65) and (66) leads to the identification of $c_n(t)$ with $(\Phi_n, \Psi_i(A))$. That is, the overlap of $\Psi_i(A)$ onto the states Φ_n gives an initial population of Φ_n states, from which the V' term causes transitions to the final state.

In summary, the first term in Eq. (64) gives the effect of the intense field $\vec{A}$ alone. The second term in Eq. (64) is the sum of all processes where the $\vec{A}$ field causes a virtual population of states, from which the $\vec{A}'$ field causes transitions to the final state. The third term in Eq. (64) gives the sum of all first-order transitions caused by $\vec{A}'$ starting from a "conditioned medium" (a distribution of states) generated by $\vec{A}$. One could say that the second and third terms both represent transitions where a distribution of virtual (second term) or real (third term) states generated by the intense field treated in momentum-translation approximation provides an intermediate medium from which the weak $\vec{A}'$ field causes first-order transitions to the final state.

VIII. DISCUSSION

The method developed in this paper has its principal usefulness for problems involving many-photon transitions. For such problems, the method has an ease of application (and a formal appearance) similar to that of first-order perturbation theory. If the method is to be applied in cases where the transition can be effected with a small number (other than one) of photons, then corrections must be introduced which increase the complexity of the calculations. Thus it is anticipated that the method will find its principal application to problems where a large number of photons from an intense electromagnetic field act alone, or in concert with a weak field which can be treated perturbatively.

An important conclusion which emerges from this paper is that the intensity parameter appropriate to bound systems is proportional to $(eaa_0)^2$, where a is the amplitude of the vector potential of the electromagnetic field, and a_0 is the size of the bound system. This parameter can be given an obvious meaning. Since ρ, the number of photons per unit volume, is given by

$$\rho = a^2(4\lambda)^{-1} \ ,$$

where λ is the photon wavelength, then

$$(\tfrac{1}{2}eaa_0)^2 = \rho a_0 \lambda_e \lambda \ .$$

That is, the intensity parameter $(\frac{1}{2}eaa_0)^2$ is just the number of photons contained in a rectangular box whose sides are given by the size of the atom a_0, the Compton wavelength of the electron λ_e, and the wavelength of the radiation λ. The intensity parameter for free electrons is

$$(\tfrac{1}{2}ea/m)^2 = \rho r_0 \lambda_e \lambda \ ,$$

which is the number of photons in a box with sides given by λ_e, λ, and the classical electron radius r_0. Thus, the fact that the electron is bound to an atom increases its effective "radius" from r_0 to a_0, a gain of a factor of $(137)^2$ if a_0 is the Bohr radius of hydrogen.

Largely as a consequence of the size of the intensity parameter, it was shown that intensity effects can be significant with available laser powers. Furthermore, these intensity effects have the nature that when they do make themselves manifest, modest further increases in intensity can make major qualitative changes in the behavior of the system. The nature of these qualitative changes, which are such as to contradict obvious features of a perturbation treatment, lead one to suspect that perturbation theory may fail to converge for the intensities being discussed.

Much additional work remains to be done on the methods introduced here, both in terms of specific applications and further formal developments. The most obvious application, perhaps, is to the

calculation of intense-field multiphoton ionization processes, for which experimental results are already available. Other applications would be in atoms more complicated than hydrogen, in molecules, in nuclei, and in solids. There should be more investigation of the relativistic problem, specifically, to see if high intensity magnifies relativistic effects. There should also be more work on line broadening and shifting. In this context, it should be noted that the energy expectation value calculated in Sec. III is actually a line-broadening calculation, since the oscillation of the Stark-like result is generally too rapid to be time resolved, and would appear as a broadening of the line.

Formal developments which are required are numerous, and include such things as investigation of electromagnetic fields of other than plane-wave type; development of convenient methods for treating spectral distributions of plane waves (i.e., extending the monochromatic results developed here); exploration of nonperturbative means of getting corrections to the momentum-translation approximation; further investigation of path-integral methods[4] in intense-field problems; and so on.

In brief summary, the method developed is, in principle, applicable to any problem where many photons participate in the process, where the dipole approximation is valid, and where there is reasonable knowledge of the wave function for the system when no electromagnetic field is present.

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APPENDIX: ANGULAR MOMENTUM REDUCTION FOR COMBINED FIELDS

The matrix element to be considered is $\{\phi_f, \exp[ie(\vec{A}+\vec{A}')\circ\vec{x}]\,\phi_i\}$. There is no advantage in choosing any particular orientation of axes. The exponential factors can be represented by the general partial-wave expansions

$$\exp(ie\vec{A}\cdot\vec{x}) = 4\pi \sum_{l=0}^{\infty} \sum_{m=-l}^{l} i^l j_l(eAr) Y_l^{m*}(\theta_A, \varphi_A) Y_l^{m}(\theta, \varphi),$$

$$\exp(ie\vec{A}'\cdot\vec{x}) = 4\pi \sum_{\bar{l}=0}^{\infty} \sum_{\bar{m}=-\bar{l}}^{\bar{l}} i^{\bar{l}} j_{\bar{l}}(eA'r) Y_{\bar{l}}^{\bar{m}*}(\theta_{A'}, \varphi_{A'}) Y_{\bar{l}}^{\bar{m}}(\theta, \varphi) \ ,$$

where θ, φ are the angular coordinates of the vector $\vec{x}$, and θ_A, φ_A and $\theta_{A'}$, $\varphi_{A'}$ are, respectively, the angular coordinates of the $\vec{A}$ and $\vec{A}'$ vectors. Since we can set

$$\phi_i = R_i(r) Y_{l_i}^{m_i}(\theta, \varphi) \ , \qquad \phi_f = R_f(r) Y_{l_f}^{m_f}(\theta, \varphi) \ ,$$

we have the preliminary partial-wave representation of the matrix element

$$(\phi_f, \exp[ie(\vec{A}+\vec{A}')\cdot\vec{x}]\,\phi_i) = \int_0^\infty r^2\, dr\, R_f^*(r) R_i(r) (4\pi)^2 \sum_{l,m,\bar{l},\bar{m}} i^{l+\bar{l}} j_l(eAr) j_{\bar{l}}(eA'r) Y_l^{m*}(\theta_A, \varphi_A)$$
$$\times Y_{\bar{l}}^{\bar{m}*}(\theta_{A'}, \varphi_{A'}) \int d\Omega Y_{l_f}^{m_f*}(\theta, \varphi) Y_{l_i}^{m_i}(\theta, \varphi) Y_l^{m}(\theta, \varphi) Y_{\bar{l}}^{\bar{m}}(\theta, \varphi). \qquad \text{(A1)}$$

With the phase convention $Y_l^{m*} = (-)^m Y_l^{-m}$,

we can reduce the number of spherical harmonic factors by using the expressions

$$Y_{l_f}^{m_f*} Y_{l_i}^{m_i} = (-)^{m_f} \sum_{L,M} \left(\frac{(2l_i+1)(2l_f+1)(2L+1)}{4\pi}\right)^{1/2} \begin{pmatrix} l_i & l_f & L \\ m_i & -m_f & M \end{pmatrix} \begin{pmatrix} l_i & l_f & L \\ 0 & 0 & 0 \end{pmatrix} Y_L^{M*} \ ,$$

$$Y_l^m Y_{\bar{l}}^{\bar{m}} = \sum_{\lambda,\mu} \left(\frac{(2l+1)(2\bar{l}+1)(2\lambda+1)}{4\pi}\right)^{1/2} \begin{pmatrix} l & \bar{l} & \lambda \\ m & \bar{m} & \mu \end{pmatrix} \begin{pmatrix} l & \bar{l} & \lambda \\ 0 & 0 & 0 \end{pmatrix} Y_\lambda^{\mu *} \quad . \tag{A2}$$

The solid-angle integral over θ, φ coordinates then contains only two spherical harmonics, for which we get

$$\int d\Omega\, Y_\lambda^{\mu *}(\theta,\varphi) Y_L^{M*}(\theta,\varphi) = (-)^M \delta_{\lambda,L} \delta_{\mu,-M} \ , \tag{A3}$$

from the phase convention and orthogonality. The complete integral over solid angle in Eq. (A1) is, from Eqs. (A2) and (A3),

$$\int d\Omega\, Y_{l_f}^{m_f *} Y_{l_i}^{m_i} Y_l^m Y_{\bar{l}}^{\bar{m}} = \frac{(-)^{m_f}}{4\pi} \sum_{L,M} (-)^M (2L+1)[(2l+1)(2\bar{l}+1)(2l_i+1)(2l_f+1)]^{1/2}$$

$$\times \begin{pmatrix} l_i & l_f & L \\ m_i & -m_f & M \end{pmatrix} \begin{pmatrix} l & \bar{l} & L \\ m & \bar{m} & -M \end{pmatrix} \begin{pmatrix} l_i & l_f & L \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} l & \bar{l} & L \\ 0 & 0 & 0 \end{pmatrix} \quad . \tag{A4}$$

The most general result follows from Eq. (A4) employed in Eq. (A1).

The above result is rather complicated, since it involves summations over the indices l, m, $\bar{l}$, $\bar{m}$, L, and M. We get a much simpler special case if we consider initial s states, i.e., $l_i = 0$. Then the result is

$$(\phi_f, \exp[ie(\vec{A}+\vec{A}')\cdot\vec{x}]\phi_i) = \int_0^\infty r^2 dr R_f^*(r) R_i(r) 4\pi (2l_f+1)^{1/2}$$

$$\times \sum_{l,m,\bar{l},\bar{m}} [(2l+1)(2\bar{l}+1)]^{1/2} i^{l+\bar{l}} j_l(eAr) j_{\bar{l}}(eA'r) Y_l^{m*}(\theta_A, \varphi_A)\, Y_{\bar{l}}^{\bar{m}*}(\theta_{A'}, \varphi_{A'}) \begin{pmatrix} l & \bar{l} & l_f \\ m & \bar{m} & -m_f \end{pmatrix} \begin{pmatrix} l & \bar{l} & l_f \\ 0 & 0 & 0 \end{pmatrix} \quad .$$

If we set $l_f = 0$ as well as $l_i = 0$, we get

$$(\phi_f, \exp[ie(\vec{A}+\vec{A}')\cdot\vec{x}]\phi_i) = \int_0^\infty r^2 dr R_f^*(r) R_i(r) \sum_{l=0}^\infty (-)^l \ (2l+1) P_l(\vec{\epsilon}\cdot\vec{\epsilon}') j_l(eAr) j_l(eA'r) \ . \tag{48}$$

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[1]G. S. Voronov, G. A. Delone, N. B. Delone, and O. V. Kudrevatova, Zh. Eksperim. i Teor. Fiz. Pis'ma v Redaktsiyu 2, 377 (1965) [English transl.: Soviet Phys. – JETP Letters 2, 237 (1965)]; G. S. Voronov and N. B. Delone, Zh. Eksperim. i Teor. Fiz. 50, 78 (1966) [English transl.: Soviet Phys. – JETP 23, 54 (1966)]; G. S. Voronov, G. A. Delone, and N. B. Delone, *ibid.* 51, 1660 (1966) [*ibid.* 24, 1122 (1967)]; T. B. Bystrova, G. S. Voronov, G. A. Delone, and N. B. Delone, Zh. Eksperim. i Teor. Fiz. Pis'ma v Redaktsiyu 5, 223 (1967) [English transl.: Soviet Phys. – JETP Letters 5, 178 (1967)]; G. A. Delone and N. B. Delone, Zh. Eksperim. i Teor. Fiz. 54, 1067 (1968) [Soviet Phys. – JETP 27, 570 (1968)].

[2]H. B. Bebb and A. Gold, Phys. Rev. 143, 1 (1966); Y. Gontier and M. Trahin, *ibid.* 172, 83 (1968).

[3]See, for example, H. R. Reiss and J. H. Eberly, Phys. Rev. 151, 1058 (1966), and references contained therein.

[4]Actually the original development of the method, given in H. R. Reiss, Bull. Am. Phys. Soc. 12, 109 (1967), was based upon a path-integral technique. The method given here, which is less general but more direct, was first reported in H. R. Reiss, *ibid.* 13, 80 (1968).

[5]The oscillatory nature of Eq. (19) can come about only if both emissions and absorptions from a given level take place. This cannot be true for the ground state of an atom, but it is shown easily that $\langle\Phi|e\vec{\delta}\cdot\vec{x}|\Phi\rangle$ vanishes if Φ is the ground state.

[6]A. R. Edmonds, Angular Momentum in Quantum Mechanics (Princeton University Press, Princeton, 1957), p. 63.

[7]To be quite correct, one should not use plane-wave results for $\vec{A}$ (as done here) when a focused beam is considered. See A. Boivin and E. Wolf, Phys. Rev. 138, B1561 (1965); A. Boivin, J. Dow, and E. Wolf, J. Opt. Soc. Am. 57, 1171 (1967).