Modified perturbation theory for scattering in a laser field

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A variation-iteration procedure is described for determining the amplitude for scattering in the presence of a laser field. The essential limitation of the method lies in the requirement that the interaction of the charged projectile with the external field be sufficiently weak, relative to its interaction with the target, to justify use of perturbation theory to account for the effect of the field in intermediate states of the collision process. This still allows for fields which are strong enough to significantly affect the motion of the projectile in initial and final states (leading, for example, to multiphoton transitions) and this strong interaction is treated nonperturbatively. The first two terms in the modified perturbation expansion are analyzed in detail. They are expressed in terms of those matrix elements which describe one- and two-photon free-free transitions in the *absence* of an external field. The method is not restricted to a consideration of fields of low frequency but the first-order amplitude obtained here does reduce, in that limit, to the known form of low-frequency approximation and the second-order amplitude provides a correction. The theory is described in the context of nonrelativistic potential scattering. A discussion is included of some of the special features of the second-order amplitude associated with potentials having a long-range Coulomb tail.

I. INTRODUCTION

When an electron scatters from a target in the presence of a laser field, radiative interactions can play a significant role, leading, for example, to the stimulated emission or absorption of a large number of photons.[1] It might be thought, then, that perturbation theory, which has been the traditional tool in theoretical studies of spontaneous radiation effects, would be ineffectual in the strong-field regime. Indeed, the ratio of some suitably defined average of the electron-field interaction energy to the photon energy, a parameter which provides a measure of the effective strength of the radiative interaction in initial and final states of the scattering process, can be of order unity or greater even for fields of only moderate intensity provided the frequency ω is small enough. An enhancement in the effective strength of the radiative interaction at low frequencies can be understood as a consequence of the near degeneracy of adjacent states of the electron-field system—asymptotically, when the electron energy is well defined, the level spacing is $\hbar\omega$. In many cases of interest the asymptotic electron-field states can be constructed analytically so that the inapplicability of perturbation expansion techniques causes no difficulty. Since the electron energy is not well defined during the scattering process itself the near-degeneracy argument no longer applies. A more appropriate coupling parameter in this case would be the ratio of the external field strength to some characteristic atomic strength, which we take to be 10^9 V/cm (the field produced by a proton at a distance of a Bohr radius). This ratio is of order unity when the external field intensity reaches the "critical" value $I_c = 10^{16}$ W/cm^2, *extremely* strong by laboratory standards. It then seems reasonable to expect that for external fields with intensity $I \ll I_c$ a type of modified perturbation theory, involving an exact description of the asymptotic motion and a perturbative treatment of the radiative interaction in intermediate states, should be useful. Such an approach will be described here, in the context of the relatively simple model of nonrelativistic potential scattering.

The essential feature of the approximation procedure outlined below lies in its decoupling of the electron-field and electron-target interactions. While this is a considerable simplification one is still left with the problem of evaluating matrix elements involving exact solutions (wave functions and Green's functions) of the scattering system in the absence of the field. It should be emphasized that the approach adopted here includes the low-frequency domain but it is not restricted to it. Since much of the earlier work on external-field scattering theory has dealt with low-frequency fields[2] we shall point out explicitly how our results reduce, in the appropriate limit, to those already obtained. A characteristic property of the low-frequency approximation (in any of its standard versions)[2] is that it requires only on-shell scattering parameters as input. In the more general results obtained here off-shell information (in the form of field-free wave functions and Green's functions) is required, as we have mentioned.[3]

While there are several possible starting points for the development of a modified perturbation theory we have found it convenient to adopt the previously derived variational formulation[4] as the basis for the discussion. By suitable choice of trial function the combined effect of radiative and scattering interactions can be built in approximately at the outset; the error in the trial function can then be accounted for to first order. Extending the earlier work[4] we here derive a formal expression for the second-

order error term—the difference between the exact transition amplitude and the variational estimate. Intermediate-state radiative interactions are included in a systematic way through a perturbative expansion of the propagator which appears in the second-order error term. Only the lowest-order contribution to such an expansion is examined explicitly here. There are no obstacles, in principle, in including still higher-order corrections, though of course the computational difficulties will escalate rapidly. In breaking off the expansion as we have done here, we have included the electron-field interaction in asymptotic states to all orders and in intermediate states to second order.

As seen previously,[4] the first-order (variational) term factorizes into a field-dependent part and a matrix element which represents *single*-photon bremsstrahlung. This latter matrix element was evaluated analytically by Sommerfeld for the case of a purely Coulombic scattering potential.[5] No such analytic form is available for an arbitrary potential. A soft-photon approximation for this matrix element is known,[6] expressed in terms of the on-shell field-free scattering amplitude. However, use of this approximation is restricted to the low-frequency limit, and even there it will introduce errors of the same order as the correction terms we now wish to include. Therefore, in the context of the present procedure, we must rely on numerical evaluation of the single-photon bremsstrahlung matrix element, the procedure for which is now well established.[7] The second-order correction to the transition amplitude can be represented as a sum of terms, each taking the form of a product of a known field-dependent factor and a matrix element very closely related to the amplitude for *two*-photon spontaneous bremsstrahlung. Interestingly, in the special case where the potential is purely Coulombic an extension of the Sommerfeld procedure has been developed which allows for accurate evaluations of matrix elements of this type.[8] With some effort approximate numerical determinations of the two-photon matrix element could be obtained for a wider class of potentials. This would open the way to careful quantitative studies of the range of validity of the low-frequency approximation, and, more generally, of the modified perturbation theory approach to the scattering problem.

II. FORMAL DEVELOPMENT

The radiation field is described, in the Coulomb gauge, by the classical vector potential $\mathbf{A}(t)$, a function only of the time in the dipole approximation to be adopted here. With $\mathbf{A}(t)$ assumed to vanish for $t\to\pm\infty$ well-defined momenta can be assigned to the electron in initial and final states. While greater generality is possible we shall specialize at the outset to a potential of the form

$$\mathbf{A}(t)=\boldsymbol{\lambda}\mathscr{A}\cos(\omega t)e^{-\eta|t|}\ , \tag{2.1}$$

with $\boldsymbol{\lambda}$ a real, unit polarization vector, and with $\eta>0$. The Hamiltonian of the electron-field system is taken to be (in units with $\hbar=1$)

$$H=(-i\nabla-e\mathbf{A}/c)^2/2m+V\ , \tag{2.2}$$

where V is the scattering potential, falling off as g/r at great distances from the center of force. Following the discussion in Ref. 4 we introduce asymptotic solutions of the time-dependent Schrödinger equation of the form

$$\chi_{\mathbf{p}}^{(\pm)}(\mathbf{r},t)=(2\pi)^{-3/2}\exp[-iE_{\mathbf{p}}t+i\mathbf{p}\cdot\mathbf{r} \pm i(gm/p)\ln(pr\mp\mathbf{p}\cdot\mathbf{r}) +i\Phi_{\mathbf{p}}^{(\pm)}(t)]\ , \tag{2.3}$$

with $E_{\mathbf{p}}=p^2/2m$ and

$$\Phi_{\mathbf{p}}^{(+)}(t)=-\int_{-\infty}^{t}\left[\frac{-e\mathbf{p}\cdot\mathbf{A}(t')}{mc}+\frac{e^2A^2(t')}{2mc^2}\right]dt'\ , \tag{2.4a}$$

$$\Phi_{\mathbf{p}}^{(-)}(t)=\int_{t}^{\infty}\left[\frac{-e\mathbf{p}\cdot\mathbf{A}(t')}{mc}+\frac{e^2A^2(t')}{2mc^2}\right]dt'\ . \tag{2.4b}$$

We evaluate these integrals using the form (2.1) for $\mathbf{A}(t)$ and, to simplify subsequent formulas, then allow η to vanish. Thus, with the aid of relations of the type

$$\int_{-\infty}^{t}\cos(\omega t')e^{-\eta|t'|}dt'\to\omega^{-1}\sin(\omega t),\quad \eta\to 0+$$

we find $\Phi_{\mathbf{p}}^{(+)}(t)=\Phi_{\mathbf{p}}^{(-)}(t)\equiv\Phi_{\mathbf{p}}(t)$, with

$$\Phi_{\mathbf{p}}(t)=\rho_{\mathbf{p}}\sin(\omega t)-(\Delta/2\omega)\sin(2\omega t)-\Delta t\ ; \tag{2.5}$$

here we introduced

$$\rho_{\mathbf{p}}=(e\mathscr{A}/mc\omega)\mathbf{p}\cdot\boldsymbol{\lambda}\ , \tag{2.6}$$

$$\Delta=\tfrac{1}{2}e^2\mathscr{A}^2/2mc^2\ . \tag{2.7}$$

A constant term, singular in the limit $\eta\to 0$, has been omitted in arriving at Eq. (2.5); this is permissible since it affects only the overall phase of the solution. The wave functions $\psi_{\mathbf{p}}^{(\pm)}(\mathbf{r},t)$ may now be defined as solutions of

$$\left[H-i\frac{\partial}{\partial t}\right]\psi_{\mathbf{p}}^{(\pm)}(\mathbf{r},t)=0 \tag{2.8}$$

subject to the conditions $\psi_{\mathbf{p}}^{(\pm)}(\mathbf{r},t)\to\chi_{\mathbf{p}}^{(\pm)}(\mathbf{r},t)$ for $t\to\mp\infty$.[9]

In the variational approach we introduce trial functions $\psi_{\mathbf{p}\tau}^{(\pm)}$ which have the correct asymptotic behavior but which need not satisfy Eq. (2.8). A convenient starting point is the identity[4,10]

$$S=S_\tau-iT\ , \tag{2.9a}$$

where S is the S matrix element for a scattering process which changes the electron momentum from $\mathbf{p}$ to $\mathbf{p}'$ (momentum labels on matrix elements are omitted to simplify notation), S_τ is a trial S matrix element defined as

$$S_\tau=\lim_{t\to\infty}\int d^3r[\chi_{\mathbf{p}'}^{(-)}(\mathbf{r},t)]^*\psi_{\mathbf{p}\tau}^{(+)}(\mathbf{r},t)\ , \tag{2.9b}$$

and

$$T\equiv\int_{-\infty}^{\infty}dt\int d^3r[\psi_{\mathbf{p}'}^{(-)}(\mathbf{r},t)]^*\left[H-i\frac{\partial}{\partial t}\right]\psi_{\mathbf{p}\tau}^{(+)}(r,t)\ . \tag{2.9c}$$

With $(H-i\partial/\partial t)\psi_{\mathbf{p}\tau}^{(+)}$ treated as a quantity of first order the replacement of the exact solution $\psi_{\mathbf{p}'}^{(-)}$ in Eq. (2.9c) by the trial function $\psi_{\mathbf{p}'\tau}^{(-)}$ introduces an error of second order and this provides the basis for the variational principle.

Rather than simply discarding the second-order error term we proceed now to set up a successive approximation procedure for estimating it, thereby allowing one to improve on the variational approximation in a systematic way. Toward this end we first derive a formal expression for the error. Let $G(\mathbf{r}',t';\mathbf{r},t)$ represent the retarded Green's function, satisfying the condition

$$G(\mathbf{r}',t';\mathbf{r},t)=0, \quad t'<t \tag{2.10}$$

and the differential equation

$$\left[H'-i\frac{\partial}{\partial t'}\right]G(\mathbf{r}',t';\mathbf{r},t)=-\delta^3(\mathbf{r}'-\mathbf{r})\delta(t'-t)\ . \tag{2.11}$$

Here H' is the Hamiltonian expressed in terms of the primed coordinates. The fundamental time-evolution property of G allows us to write

$$i\int d^3r''[\chi_{\mathbf{p}'}^{(-)}(\mathbf{r}'',t'')]^*G(\mathbf{r}'',t'';\mathbf{r},t) =\Theta(t''-t)[\psi_{\mathbf{p}'}^{(-)}(\mathbf{r},t)]^*\ , \tag{2.12}$$

where Θ is the step function. The identity

$$[\psi_{\mathbf{p}'}^{(-)}(\mathbf{r},t)]^*=[\psi_{\mathbf{p}'\tau}^{(-)}(\mathbf{r},t)]^*+\int_{-\infty}^{\infty}dt'\int d^3r'\left[\left[H'-i\frac{\partial}{\partial t'}\right]\psi_{\mathbf{p}'\tau}^{(-)}(\mathbf{r}',t)\right]^*G(\mathbf{r}',t';\mathbf{r},t) \tag{2.13}$$

is now readily verified. The procedure to do so, briefly outlined, is to move the operators H' and $-i\partial/\partial t'$ to the right, so they act on G rather than on $\psi_{\mathbf{p}'\tau}^{(-)}$, and then to make use of the differential equation (2.11) for G. The Hamiltonian is Hermitian, so no surface terms are introduced when H' is switched over.[11] When the time-derivative operator is switched over, as part of an integration-by-parts procedure, one finds that, by virtue of the retardation property (2.10), the end-point contribution evaluated at $t'=-\infty$ vanishes. The contribution from $t'=+\infty$ is nonvanishing, and is determined with the aid of the asymptotic condition $\psi_{\mathbf{p}'\tau}^{(-)}(\mathbf{r}',t')\to\chi_{\mathbf{p}'}^{(-)}(\mathbf{r}',t')$ for $t'\to\infty$, along with Eq. (2.12). The result of these manipulations is to transform the right-hand side (rhs) of Eq. (2.13) into $[\psi_{\mathbf{p}'}^{(-)}(\mathbf{r},t)]^*$, as claimed. The identity just derived may now be combined with Eq. (2.9c), leading to the decomposition $T=T^{(1)}+\Delta T$, with

$$T^{(1)}=\int_{-\infty}^{\infty}dt\int d^3r[\psi_{\mathbf{p}'\tau}^{(-)}(\mathbf{r},t)]^*\left[H-i\frac{\partial}{\partial t}\right]\psi_{\mathbf{p}\tau}^{(+)}(\mathbf{r},t) \tag{2.14}$$

and

$$\Delta T=\int_{-\infty}^{\infty}dt\int d^3r\int_{-\infty}^{\infty}dt'\int d^3r'\left[\left[H'-i\frac{\partial}{\partial t'}\right]\psi_{\mathbf{p}'\tau}^{(-)}(\mathbf{r}',t')\right]^*G(\mathbf{r}',t';\mathbf{r},t)\left[H-i\frac{\partial}{\partial t}\right]\psi_{\mathbf{p}\tau}^{(+)}(\mathbf{r},t)\ . \tag{2.15}$$

The variational approximation for the S matrix is

$$S_v\equiv S_\tau-iT^{(1)} \tag{2.16}$$

with ΔT providing an exact formal representation of the error.

An estimate of the second-order error is obtained by replacing G in Eq. (2.15) by a trial Green's function G_τ. Corrections of still higher order may be generated by an iterative procedure which we describe, somewhat schematically, as follows. With the solution to Eq. (2.11) represented symbolically as

$$G=\left[i\frac{\partial}{\partial t}-H\right]^{-1}, \tag{2.17}$$

we introduce the identity

$$G=G_\tau-G(G^{-1}G_\tau-1)\ . \tag{2.18}$$

If G_τ is sufficiently accurate $G^{-1}G_\tau-1$ may be treated as a first-order quantity, in which case the replacement of G with G_τ on the rhs of Eq. (2.18) will lead to an approximation for G correct to second order. This variational approximation for G, inserted into Eq. (2.15), provides us with the estimate $\Delta T\cong T^{(2)}+T^{(3)}$, where $T^{(2)}$ is obtained from (2.15) by replacing G with G_τ, and $T^{(3)}$ is obtained by replacing G with $-G_\tau(G^{-1}G_\tau-1)$. It should be clear how this iterative procedure could be continued to generate corrections of still higher order.

III. FIRST AND SECOND ORDERS OF A MODIFIED PERTURBATION EXPANSION

The trial functions adopted previously,[4] and assumed here as well, are of the form

$$\psi_{\mathbf{p}\tau}^{(\pm)}(\mathbf{r},t)=\exp[-iE_{\mathbf{p}}t+i\Phi_{\mathbf{p}}(t)]u_{\mathbf{p}}^{(\pm)}(\mathbf{r})\ , \tag{3.1}$$

where the $u_{\mathbf{p}}^{(\pm)}$ are solutions of the field-free wave equation

$$\left[-\frac{\nabla^2}{2m}+V(r)-E_{\mathbf{p}}\right]u_{\mathbf{p}}^{(\pm)}(\mathbf{r})=0\ , \tag{3.2}$$

satisfying outgoing-wave $(+)$ and incoming-wave $(-)$ boundary conditions. The functions (3.1) account for the dominant asymptotic interaction of the electron with the field but fail to provide a proper treatment of the radiative interaction during the collision. The variational approximation (2.16) introduces a partial compensation for this deficiency; to this must be added the error term (2.15) to include the full effect of the electron-field interaction in intermediate states. A perturbative construction of this error term is based on the choice of trial Green's function G_τ as the field-free Green's function G_0. The latter func-

tion satisfies Eq. (2.11) with $\mathbf{A}=\mathbf{0}$ and can be represented by the eigenfunction expansion

$$G_0(\mathbf{r}'t';\mathbf{r},t)=-i\Theta(t'-t)\int d^3q\exp[-iE_q(t'-t)] \times u_q^{(+)}(\mathbf{r}')[u_q^{(+)}(\mathbf{r})]^* , \tag{3.3}$$

where the symbol $\int d^3q$ is meant to include a sum over discrete states as well as an integration over the continuum. In the following we examine the form of the first- and second-order contributions to the S matrix corresponding to the above choice of trial wave functions and trial Green's function.

A. First-order term

The trial S matrix element defined in Eq. (2.9b) can be evaluated by recognizing that as a consequence of the rapid oscillations of the integrand in the limit $t\to\infty$ only the singular part of the spatial integration contributes, and to determine that contribution we may confine our attention to the asymptotic region of configuration space. Following the methods outlined in Ref. 4 for carrying out such an asymptotic evaluation, and adopting the form (3.1) for $\psi_{\mathbf{p}\tau}^{(+)}$, we find that

$$S_\tau=t(\mathbf{p}',\mathbf{p})\lim_{t\to\infty}\frac{\exp[i(E_{\mathbf{p}'}-E_\mathbf{p})t+i\Phi_\mathbf{p}(t)-i\Phi_{\mathbf{p}'}(t)]}{E_\mathbf{p}-E_{\mathbf{p}'}+i\epsilon} , \tag{3.4}$$

where $t(\mathbf{p}',\mathbf{p})$ is the conventionally defined t matrix for scattering in the absence of the field, and ϵ is an infinitesimal positive parameter which, according to the standard procedure of scattering theory, is allowed to vanish at the end of the calculation. [Its appearance in Eq. (3.4) can be traced to the inclusion of a small imaginary contribution to the energy of the outgoing scattered wave, an inclusion which is in fact necessary to keep the spatial integration well defined at infinity.] To complete the calculation we note that, according to Eq. (2.5), $\Phi_{\mathbf{p}'}(t)-\Phi_\mathbf{p}(t)=\rho\sin(\omega t)$ with

$$\rho=(e\mathscr{A}/mc\omega)(\mathbf{p}'-\mathbf{p})\cdot\boldsymbol{\lambda} . \tag{3.5}$$

The expansion

$$e^{-i\rho\sin(\omega t)}=\sum_{n=-\infty}^{\infty}e^{-in\omega t}J_n(\rho) , \tag{3.6}$$

where $J_n(\rho)$ is the cylindrical Bessel function, may be used in Eq. (3.4) and the infinite limit evaluated. Only the term with $n=0$ survives and we obtain the result

$$S_\tau=-2\pi i\delta(E_{\mathbf{p}'}-E_\mathbf{p})t(\mathbf{p}',\mathbf{p})J_0(\rho) . \tag{3.7}$$

In the evaluation of $T^{(1)}$, defined in Eq. (2.14), we make use of the relation

$$\left[H-i\frac{\partial}{\partial t}\right]\psi_{\mathbf{p}\tau}^{(+)}(\mathbf{r},t) =-(e/mc)\mathbf{A}(t)\cdot(-i\nabla-\mathbf{p})\psi_{\mathbf{p}\tau}^{(+)}(\mathbf{r},t) , \tag{3.8}$$

appropriate to the form (3.1) for $\psi_{\mathbf{p}\tau}$. With the aid of the recursion relation

$$J_{n-1}(\rho)+J_{n+1}(\rho)=(2n/\rho)J_n(\rho) \tag{3.9}$$

we then find

$$T^{(1)}=2\pi\sum_{n=-\infty}^{\infty}\delta(E_{\mathbf{p}'}-E_\mathbf{p}-n\omega)\left[\frac{-n\omega J_n(\rho)}{(\mathbf{p}'-\mathbf{p})\cdot\boldsymbol{\lambda}}\right]M^{(1)} , \tag{3.10}$$

with

$$M^{(1)}=\boldsymbol{\lambda}\cdot\int d^3r[u_{\mathbf{p}'}^{(-)}(\mathbf{r})]^*(-i\nabla-\mathbf{p})u_\mathbf{p}^{(+)}(\mathbf{r}) . \tag{3.11}$$

For $E_{\mathbf{p}'}\neq E_\mathbf{p}$ the functions $u_{\mathbf{p}'}^{(-)}$ and $u_\mathbf{p}^{(+)}$ are orthogonal so that we may replace $-i\nabla-\mathbf{p}$ by $-i\nabla$ in the matrix element (3.11), which is then identified (to within a normalization factor) with the amplitude for single-photon spontaneous bremsstrahlung. Since the bremsstrahlung amplitude is singular for $E_{\mathbf{p}'}-E_\mathbf{p}\to 0$ this limiting case requires special consideration. We note first that the integral which vanishes due to orthogonality for $E_{\mathbf{p}'}\neq E_\mathbf{p}$ should more properly be written as

$$\int d^3r[u_{\mathbf{p}'}^{(-)}(\mathbf{r})]^*u_\mathbf{p}^{(+)}(\mathbf{r})=-2\pi i\delta(E_{\mathbf{p}'}-E_\mathbf{p})t(\mathbf{p}',\mathbf{p}) \tag{3.12}$$

[as may be verified using an asymptotic evaluation of the radial integral analogous to that employed in the derivation of Eq. (3.4)]. This contribution may be ignored, however, since the vanishing factor $n\omega=E_{\mathbf{p}'}-E_\mathbf{p}$ multiplies $M^{(1)}$ in Eq. (3.10). We now observe that the same asymptotic-evaluation technique applied to the bremsstrahlung amplitude leads to the result

$$\boldsymbol{\lambda}\cdot\int d^3r[u_{\mathbf{p}'}^{(-)}(\mathbf{r})]^*(-i\nabla)u_\mathbf{p}^{(+)}(\mathbf{r}) \cong-(E_{\mathbf{p}'}-E_\mathbf{p}-i\epsilon)^{-1}(\mathbf{p}'-\mathbf{p})\cdot\boldsymbol{\lambda}t(\mathbf{p}',\mathbf{p}) \tag{3.13}$$

for $E_{\mathbf{p}'}-E_\mathbf{p}\to 0$. The origin of the infinitesimal positive parameter ϵ is similar to that discussed in connection with Eq. (3.4). (Here one must add $-i\epsilon$ to the energy of the incoming scattering wave in $u_{\mathbf{p}'}^{(-)}$ as well as adding $+i\epsilon$ to the energy of the outgoing scattered wave in $u_\mathbf{p}^{(+)}$.) Since the limit $\epsilon\to 0$ is taken last the factor $(E_{\mathbf{p}'}-E_\mathbf{p})(E_{\mathbf{p}'}-E_\mathbf{p}-i\epsilon)^{-1}$ which appears in Eq. (3.10) is interpreted as zero in the limit $E_{\mathbf{p}'}-E_\mathbf{p}\to 0$.

The result of these considerations can be summarized by writing the variational estimate (2.16) as

$$S_v=-2\pi i\sum_{n=-\infty}^{\infty}\delta(E_{\mathbf{p}'}-E_\mathbf{p}-n\omega)T_n^{(1)} , \tag{3.14}$$

with

$$T_n^{(1)}=J_0(\rho)t(\mathbf{p}',\mathbf{p}),\quad n=0 =-\frac{n\omega J_n(\rho)}{(\mathbf{p}'-\mathbf{p})\cdot\boldsymbol{\lambda}}M^{(1)},\quad n\neq 0 , \tag{3.15}$$

the index n having the obvious interpretation as the net number of photons absorbed ($n>0$) or emitted ($n<0$) in the scattering process.

It should be emphasized that a low-frequency approximation has not been invoked in arriving at Eq. (3.15); as discussed in Sec. I we do not wish to confine our attention

to low-frequency fields. For the sake of orientation, however, let us briefly consider that special case. If we were to make use of the approximation (3.13) for $M^{(1)}$ we would find, in the limit $\omega \to 0$ with ρ fixed, and for all n,

$$T_n^{(1)} \cong J_n(\rho) t(\mathbf{p}',\mathbf{p}) . \tag{3.16}$$

For potentials of short range the leading correction to the approximation (3.16) [which can be obtained by improving the estimate (3.13) for $M^{(1)}$] is of order ω and it too can be expressed in terms of $t(\mathbf{p}',\mathbf{p})$.[2] (When potentials with a Coulomb tail are admitted one finds[4] that the correction introduces additional terms depending logarithmically on the frequency.) Such low-frequency approximations will play no role in the present approach, however, since the errors thereby introduced are comparable in magnitude to the correction term $T^{(2)}$ to be included here.

B. Second-order term

The approximation adopted here for the second-order correction term $T^{(2)}$ is obtained from Eq. (2.15) by replacing G with G_0, the field-free Green's function. Higher-order corrections can be generated in a straightforward way but will not be considered here. This perturbative approach should be valid for field intensities well below the critical level. (Modifications would be required if there were resonances present—for simplicity we exclude that possibility here.) Adopting the representation (3.3) for G_0 we find, with the aid of the identity

$$-i\Theta(t'-t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dW \frac{e^{-iW(t'-t)}}{W+i\epsilon} , \tag{3.17}$$

the expression

$$T^{(2)} = \frac{1}{2\pi} \int_{-\infty}^{\infty} dW \left[\int_{-\infty}^{\infty} dt \exp[iWt - iE_{\mathbf{p}}t + i\Phi_{\mathbf{p}}(t)] \frac{eA(t)}{mc} \right] \times \left[\int_{-\infty}^{\infty} dt' \exp[-iWt' + iE_{\mathbf{p}'}t' - i\Phi_{\mathbf{p}'}(t')] \frac{eA(t')}{mc} \right] M^{(2)}(W) . \tag{3.18}$$

Here we have written $\mathbf{A}(t) = \boldsymbol{\lambda} A(t)$ and have defined

$$M^{(2)}(W) = \int d^3r \int d^3r' [\boldsymbol{\lambda}\cdot(-i\nabla' - \mathbf{p}') u_{\mathbf{p}'}^{(-)}(\mathbf{r}')]^* \times G_0(\mathbf{r}',\mathbf{r};W) [\boldsymbol{\lambda}\cdot(-i\nabla - \mathbf{p}) u_{\mathbf{p}}^{(+)}(\mathbf{r})] , \tag{3.19}$$

where

$$G_0(\mathbf{r}',\mathbf{r};W) = \int d^3q \frac{u_{\mathbf{q}}^{(+)}(\mathbf{r}')[u_{\mathbf{q}}^{(+)}(\mathbf{r})]^*}{W - E_{\mathbf{q}} + i\epsilon} \tag{3.20}$$

is the time-independent field-free Green's function. The integrations over time in Eq. (3.18) can be carried out with the aid of Fourier expansions of the type shown in Eq. (3.6). Use of the recursion relation (3.9) to combine terms then leads us to the form

$$T^{(2)} = 2\pi \sum_{n=-\infty}^{\infty} \delta(E_{\mathbf{p}'} - E_{\mathbf{p}} - n\omega) T_n^{(2)} \tag{3.21}$$

with

$$T_n^{(2)} = -(e\mathscr{A}/mc)^2 (\rho_{\mathbf{p}'}\rho_{\mathbf{p}})^{-1} \times \sum_{n'=-\infty}^{\infty} (n-n')(n') J_{n-n'}(\rho_{\mathbf{p}'}) J_{n'}(-\rho_{\mathbf{p}}) \times M^{(2)}(E_{\mathbf{p}} + n'\omega) . \tag{3.22}$$

The approximate S matrix, correct to second order in the modified perturbation expansion defined by the choice (3.1) of trial functions, is then given by

$$S \cong -2\pi i \sum_{n=-\infty}^{\infty} \delta(E_{\mathbf{p}'} - E_{\mathbf{p}} - n\omega)(T_n^{(1)} + T_n^{(2)}) , \tag{3.23}$$

with $T_n^{(1)}$ and $T_n^{(2)}$ represented by Eqs. (3.15) and (3.22), respectively.

We conclude this subsection with several general remarks which bear on the problem of evaluating the expression (3.22) for $T_n^{(2)}$. We first observe that if the potential has a Coulomb tail the matrix elements $M^{(2)}(E_{\mathbf{p}})$ and $M^{(2)}(E_{\mathbf{p}'})$ are each singular. [Such singularities do not appear if $V(r)$ is short-ranged. These points are discussed in more detail in the Appendix.] Nevertheless, the amplitude $T_n^{(2)}$ is well defined since $M^{(2)}$ appears in Eq. (3.22) multiplied by $(n-n')n'$ and as a result terms corresponding to $n'=0$ and $n'=n$ do not contribute to the sum. The rapid energy dependence of the matrix element for energies near $E_{\mathbf{p}}$ or $E_{\mathbf{p}'}$ must be kept in mind, however, in approximate evaluations of the sum in Eq. (3.22).

To put this last remark in clearer focus let us consider the situation where the field is of low frequency, and suppose first that the potential is of short range. In that case the replacement of $M^{(2)}(E_{\mathbf{p}} + n'\omega)$ by $M^{(2)}(E_{\mathbf{p}} + \bar{n}\omega)$, with this factor then removed from underneath the summation sign, represents a reasonable approximation, since for ω small enough, the result will be fairly insensitive to the choice of average photon number $\bar{n}$. The sum (3.22) can then be performed explicitly.[12] In this way we obtain

$$T_n^{(2)} = \left[\frac{\omega}{(\mathbf{p}'-\mathbf{p})\cdot\boldsymbol{\lambda}} \right]^2 \times [n^2 J_n(\rho) - \rho J_n'(\rho)] M^{(2)}(E_{\mathbf{p}} + \bar{n}\omega) , \tag{3.24}$$

where

$$J_n'(\rho) = \frac{dJ_n(\rho)}{d\rho} = \tfrac{1}{2}[J_{n-1}(\rho) - J_{n+1}(\rho)] \tag{3.25}$$

and we have used the definition (3.5) of ρ.[13] On the other

hand, if the potential is Coulombic at great distances a more careful treatment of the energy dependence of $M^{(2)}$ is required; one cannot simply set $\bar{n}=0$, for example. (We return briefly to this point later on.) The case where the potential is *purely* Coulombic, $V(r)=g/r$, may provide an opportunity for quantitative studies of this matter since very accurate evaluations of the one- and two-photon amplitudes $M^{(1)}$ and $M^{(2)}$ can be obtained for such a potential.[5,8]

The utility of the expression (3.22) for $T_n^{(2)}$ depends on the rate of convergence of the sum over n'. The convergence is of course most rapid at low field intensities. In the extreme weak-field limit where the small-argument representation $J_n(\rho)\cong(\rho/2)^n/n!$ is valid one finds that to order e^2 only the terms with $n'=1$ and $n'=-1$ contribute to $T_0^{(2)}$ and only the terms with $n'=\pm1$ contribute to $T_{\pm2}^{(2)}$. As a check, one can verify that the expression for $T_n^{(1)}+T_n^{(2)}$ for $n=\pm2$, with terms of order e^3 and higher ignored, gives the correct two-photon absorption ($n=2$) and emission ($n=-2$) amplitudes in lowest-order perturbation theory. Similarly, one can verify that the correct lowest-order perturbation theory amplitudes for $n=0$ and $n=\pm1$ are contained in the weak-field limit of the results obtained here.

Turning now to the strong-field case of present interest we introduce, for the purpose of discussion, the parameter

$$x=\left[\frac{\omega_1}{\omega}\right]^2\left[\frac{I}{I_1}\right]^{1/2}\left[\frac{E_p}{E_1}\right]^{1/2}, \tag{3.26}$$

where $I=\omega^2\mathscr{A}^2/8\pi c$ is the field intensity and we choose, as reference values, $\omega_1=0.1$ eV, $I_1=10^8$ W/cm^2, and $E_1=10$ eV. (These parameters correspond approximately to one of the experimental situations reported in Ref. 1.) As defined above x represents, roughly speaking, the ratio of the electron-field interaction energy $ep\mathscr{A}/mc$ to the photon energy ω and, with angular factors ignored, provides a measure of the order of magnitude of the parameters ρ_p, $\rho_{p'}$, and ρ. Under conditions such that x is of order unity or smaller, and with the possibility of rapid fluctuations of $M^{(2)}$ with energy ignored, the convergence of the sum (3.22) should be rapid enough to make feasible explicit evaluation of the amplitude $T_n^{(2)}$ based on this expression. (It is important to note that the requirement $x\lesssim1$ does not restrict one to the low-frequency domain.) On the other hand, for $x\approx10^2$, say, a large number of terms (of order 10^2) may be expected to contribute significantly to the sum, and an evaluation of $M^{(2)}(E_p+n'\omega)$ for each relevant value of n' would be tedious if not prohibitively difficult. Considerable simplification can be expected, however, if $M^{(2)}$ varies slowly enough with energy over the range of energies $E_p+n'\omega$ that are significant. More specifically, suppose that $M^{(2)}$ can be adequately represented by the first three terms in a Taylor series expansion about E_p. (Here we exclude consideration of Coulombic potentials for reasons discussed above.) Defining the quantities

$$\sigma_i=\sum_{n'=-\infty}^{\infty}(n'\omega)^i(n-n')n'J_{n-n'}(\rho_{p'})J_{n'}(-\rho_p) \tag{3.27}$$

(which can be explicitly evaluated using known sum rules), and approximating the first and second derivatives by finite differences, we may estimate the sum (3.22) as

$$T_n^{(2)}\cong(e\mathscr{A}/mc)^2(\rho_p\rho_p)^{-1}\left[M^{(2)}(E_p)\left[\sigma_0-\frac{\sigma_2}{\Delta E^2}\right]+M^{(2)}(E_p+\Delta E)\left[\frac{\sigma_1}{2\Delta E}+\frac{\sigma_2}{2\Delta E^2}\right]+M^{(2)}(E_p-\Delta E)\left[-\frac{\sigma_1}{2\Delta E}+\frac{\sigma_2}{2\Delta E^2}\right]\right], \tag{3.28}$$

where ΔE is a suitably chosen energy increment. If $M^{(2)}$ can be represented with sufficient accuracy by only the first two terms in the Taylor series expansion, the terms involving σ_2 in Eq. (3.28) may be dropped and the approximation for $T_n^{(2)}$ reduces to that shown in Eq. (3.24) with $\bar{n}=\sigma_1/\omega\sigma_0$; this may be evaluated as

$$\bar{n}=\frac{\sum_{s=\pm1}[\rho_{p'}J'_{n-s}(\rho)-(n-s)^2(\rho_p/\rho)J_{n-s}(\rho)]}{(2n^2/\rho)J_n(\rho)-2J'_n(\rho)}. \tag{3.29}$$

The approximation expressed in Eqs. (3.24) and (3.29) should be valid if $\bar{n}\omega$ is small compared with the energy range over which $M^{(2)}$ changes by an appreciable fraction of itself; the criterion $y\equiv|\bar{n}\omega/E_p|\ll1$ might serve as a rule of thumb in this regard. For the purpose of obtaining simple, order-of-magnitude estimates of the parameter y, let us introduce, in Eq. (3.29), the approximations $(J'_{n+1}+J'_{n-1})/2\approx J'_n$ and $[(n+1)^2J_{n+1}+(n-1)^2J_{n-1}]/2\approx n^2J_n$ and replace ρ_p, $\rho_{p'}$, and ρ by x. This leads to $|\bar{n}|\approx x$ and $y\approx x\omega/E_p$. (In this form y can be interpreted as the ratio of the electron-field interaction energy to the initial electron energy.) With x represented by Eq. (3.26) the above estimate for y becomes

$$y\cong10^{-2}\left[\frac{\omega_1}{\omega}\right]\left[\frac{I}{I_1}\right]^{1/2}\left[\frac{E_1}{E_p}\right]^{1/2}. \tag{3.30}$$

As an example, the parameters $I=2.5\times10^{13}$ W/cm^2, $\omega=2$ eV, and $E_p=0.25$ keV lead to the values $x\approx10$ and $y\approx\frac{1}{10}$. Under these circumstances use of the approximation (3.24) would appear to be preferable to an attempt to evaluate the sum (3.22) directly.

If the matrix element $M^{(2)}$ does vary rapidly in some restricted energy region (as is the case, for example, for energies near E_p and $E_{p'}$ when the potential has a Coulomb tail) the averaging procedure leading to Eq. (3.24) may be generalized slightly to account for such variations. For example, let us write

$$M^{(2)}(E_p+n'\omega)=M^{(2)}(E_p+\bar{n}\omega)+[M^{(2)}(E_p+n'\omega)-M^{(2)}(E_p+\bar{n}\omega)] .$$

Substitution of the first term on the rhs into Eq. (3.22) gives back the result (3.24). When the remaining term is introduced one may limit the summation to that range of values of n' over which $M^{(2)}(E_p+n'\omega)$ is rapidly varying, with $\bar{n}$ chosen to maximize the rate of convergence of this sum.

The discussion given here concerning approximate methods for evaluating the sum (3.22) is meant to be suggestive rather than definitive. The point to be emphasized, in summary, is that traditional perturbative methods, suitably modified to account for the effect of the field on the projectile in initial and final states, can play a useful role in analysis of the external-field scattering problem. Considerable attention has been given to the low-frequency approximations in theoretical studies of this problem over the past several years. Methods of the type discussed here can (at some cost in increased calculational complexity) be used to relax the requirement of low field frequencies and widen significantly the range of applicability of available techniques.

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APPENDIX

Here we examine the convergence properties of the integral (3.19) which defines the matrix element $M^{(2)}(W)$. Since integration over a finite domain will converge it is sufficient, in looking for possible divergences, to consider only the asymptotic region of integration. Our method of analysis (applied previously in Ref. 4) makes use of partial-wave representations of the wave functions along with the known asymptotic behavior of the radial functions which appear there. We need concern ourselves here only with radial integrations; these appear in the form

$$\lim_{\epsilon\to 0+}\int_0^\infty e^{-(\epsilon+ia)r}r^b dr=(ia)^{-1-b}\Gamma(1+b) . \tag{A1}$$

(The integration has been extended down to the origin for convenience—this does not affect the convergence property of interest.) A number of terms of the form (A1) appear in the calculation. We confine our attention to those with the smallest value of a and the largest power b. This is sufficient to prove convergence or to determine the nature of the leading singularity.

With the energy parameter W set equal to $q^2/2m$ for convenience we may represent the asymptotic behavior of the Green's function (to within a multiplicative constant) as

$$G_0(\mathbf{r}',\mathbf{r};q^2/2m)\sim u^{(+)}_{-q\hat{\mathbf{r}}}(\mathbf{r}')\frac{1}{r}e^{iqr}(2qr)^{-igm/q} \tag{A2}$$

for $r\to\infty$ with $r'<r$. (The contribution from the domain $r'\to\infty$ with $r<r'$ can be studied in a similar way and the result will be included below.) It is useful to decompose the wave function into incident and scattered components, $u^{(+)}_p=u^{(+)}_{p;\mathrm{inc}}+u^{(+)}_{p;\mathrm{sc}}$, with

$$u^{(+)}_{p;\mathrm{inc}}(\mathbf{r})=(2\pi)^{-3/2}\exp[i\mathbf{p}\cdot\mathbf{r}+i(gm/p)\ln(pr-\mathbf{p}\cdot\mathbf{r})] . \tag{A3}$$

The scattered wave has the form

$$u^{(+)}_{p;\mathrm{sc}}(\mathbf{r})\sim(2\pi)^{-3/2}\frac{1}{r}f^{(+)}_p(\hat{\mathbf{r}})e^{ipr}(2pr)^{-igm/p} \tag{A4}$$

for $r\to\infty$. We note that $[u^{(-)}_{p'}(\mathbf{r})]^*=u^{(+)}_{-p'}(r)$.

A distinction must be drawn between the case in which the potential is of short range and that in which it is Coulombic asymptotically. In the former case the relation $(-i\nabla-\mathbf{p})u^{(+)}_{p;\mathrm{inc}}(\mathbf{r})=0$ is satisfied and as a result the integrand in Eq. (3.19) is proportional to

$$[\boldsymbol{\lambda}\cdot(-i\nabla-\mathbf{p}')u^{(-)}_{p;\mathrm{sc}}(\mathbf{r}')]^*[u^{(+)}_{-q\hat{\mathbf{r}}}(\mathbf{r}')e^{iqr}/r]\times[\boldsymbol{\lambda}\cdot(i\nabla-\mathbf{p})u^{(+)}_{p;\mathrm{sc}}(\mathbf{r})] ,$$

with $u^{(+)}_{p;\mathrm{sc}}(\mathbf{r})$ taking the form of an outgoing spherical wave for $r\to\infty$. One sees that the factor $e^{i(q+p)r}$ appears in the integration over the radial variable r which, owing to the rapid oscillations of the integrand at infinity, is convergent. Additional r-dependent factors do appear as a result of the r' integration but their presence does not affect the convergence. Considering, for example, the contribution which is potentially most "dangerous" we may write

$$\int^r e^{i(p'-q)r'}dr'=r+\tfrac{1}{2}i(p'-q)r^2+\cdots$$

which, when multiplied by $e^{i(q+p)r}$ leads to a convergent integral. It may be concluded that $M^{(2)}(W)$ is nonsingular for a short-range potential. In particular, for $W=E_p+n'\omega$, $M^{(2)}(W)$ is finite in the limit $\omega\to 0$.

We turn now to the Coulomb problem and observe that the effect of the operator $(-i\nabla-\mathbf{p})$ on the modified plane wave $u^{(+)}_{p;\mathrm{inc}}(\mathbf{r})$ is not to annihilate it but merely (with regard to the radial dependence of interest here) to introduce an additional factor of $1/r$. Noting that $u^{(+)}_{p;\mathrm{inc}}$ has an incoming-wave as well as an outgoing-wave component at infinity, we identify the most singular part of the radial integrations to be of the form

$$\int^r e^{i(p'-q)r'}(r')^{i\alpha}dr'\int^\infty\frac{1}{r}e^{i(q-p)r}r^{i\beta}dr ,$$

where the powers of $i\alpha$ and $i\beta$ arise from the logarithmic phase factors in the Coulomb wave functions. We expand the exponential $e^{i(p'-q)r'}$ and perform the integration over r' term by term. Then, with the aid of Eq. (A1) to evaluate the r integral, we find that in leading order the singularity is of the form $(q-p)^{-1}$; this is proportional to $(W-E_p)^{-1}=(n'\omega)^{-1}$. In a similar way, by including the contribution from the asymptotic region $r'\to\infty$ with $r<r'$, we identify a singular term proportional to $(W-E_{p'})^{-1}=[(n-n')\omega]^{-1}$. Recalling the implicit presence of a small imaginary addition to the energy in these denominators we see that singularities in the sum (3.22) for $n'=0$ and $n'=n$ are avoided owing to the ap-

pearance of the factor $(n-n')n'$ multiplying the matrix element. The significant conclusion which emerges from these considerations is that $M^{(2)}(E_p+n'\omega)$ behaves as ω^{-1} for $\omega\to 0$. Accordingly, one may not assume $M^{(2)}$ to be a slowly varying function of the energy in the low-frequency limit and this must be borne in mind in devising approximate methods to evaluate the sum (3.22), as discussed in the text.

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[7]See, for example, H. K. Tseng and R. H. Pratt, Phys. Rev. A **3**, 100 (1971).

[8]M. Gavrila, A. Maquet, and V. Véniard, Phys. Rev. A **32**, 2537 (1985).

[9]These asymptotic conditions are to be understood, in the usual sense, as relations satisfied by wave-packet solutions. To simplify the presentation we do not explicitly carry out the procedure of constructing wave packets.

[10]For a general discussion of variational identities for time-evolution operators, leading to variational approximations to, and variational bounds on, transition amplitudes see L. Spruch, in *Lectures in Theoretical Physics,* edited by S. Geltman, K. T. Mahanthappa, and W. E. Brittin (Gordon and Breach, New York, 1969), Vol. XI C, p. 77.

[11]Surface terms do of course appear in analogous derivations of variational principles for time-independent scattering problems. The time-dependent approach, based on the picture of localized wave packets, is simpler in this regard since at any finite time the particle flux at infinity is zero. Further discussion of this point can be found in Ref. 10.

[12]The required Bessel-function sum rules can be derived very simply by making use of the integral representation of the Bessel function [obtained by inverting Eq. (3.6)].

[13]When $M^{(2)}$ in Eq. (3.24) is evaluated in the zero-frequency limit one obtains, for a short-range potential, an approximation to $T_n^{(2)}$ of order ω^2 (for a given fixed value of ρ). One might wish to express the complete n-photon S matrix element as an expansion in powers of ω, thereby obtaining an expression for the ω^2 correction to the standard version (Ref. 2) of the low-frequency approximation. To accomplish this, in the context of the present approach, one must add to $T_n^{(2)}$ the contribution $T_n^{(1)}$, Eq. (3.15), with $M^{(1)}$ replaced by the first *three* terms in its low-frequency expansion. [The coefficients of the first two terms, of order ω^{-1} and ω^0, can be expressed in terms of on-shell scattering parameters, as shown by Low (Ref. 6). The third term involves off-shell information.] The ω^2 correction to the standard low-frequency approximation obtained in this way could then be compared with that derived by Mittleman, Ref. 3, who used a different method. The two expressions should be formally identical but this has not yet been explicitly demonstrated.