PHYSICAL REVIEW A VOLUME 43, NUMBER 3 1 FEBRUARY 1991

Low-frequency expansions for laser-assisted collisions

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(Received 10 August 1990)

A variational approach to the problem of scattering in a low-frequency laser field is adopted, with trial functions chosen to be of the type originally introduced by Kroll and Watson [Phys. Rev. A **8**, 804 (1973)], who used them nonvariationally. The variational calculation leads to a low-frequency approximation that includes higher-order correction terms of a relatively simple form. This provides the basis for an analysis of the accuracy of the approximation as the strength of the external field varies over a wide range of values. The cross-section sum rule is shown, for the case of a linearly polarized monochromatic field of moderate intensity, to be more accurate than had previously been realized by virtue of the cancellation of higher-order correction terms in the transition amplitude. The approach is shown to be applicable to elastic and inelastic electron-atom scattering in a multimode laser field with arbitrary polarization properties; it represents a natural generalization of the standard Kohn-type variational procedure frequently employed for field-free scattering problems, reducing to it in the absence of the field. The dressing of the target by the field, an effect which is known to have a significant influence on the scattering cross section in certain circumstances, is not accounted for in the construction of the trial function, but is properly included as part of the variational correction.

I. INTRODUCTION

Low-frequency approximations for scattering in a laser field have been known for some time, but there is still little available in the way of systematic analyses of the domains of applicability and range of validity of these calculational techniques. The standard procedure of perturbation theory, which generates a series of terms of increasing order in some small parameter, and which therefore provides a means for judging the accuracy of an approximation, is not directly applicable for sufficiently intense external fields. The variational approach, which may be thought of as a generalization of perturbation theory with the adoption of a trial function replacing the choice of a model Hamiltonian, can be useful in this connection. A small parameter may be associated, in some sense, with the error in the trial function; with that error taken to be of first order, the variational principle guarantees that the error in the variational approximation will be of second order. In the class of problems under consideration, the frequency of the laser field provides a natural small parameter and one may then be more specific in the classification of the order of magnitude of successive terms. One of our objectives is to provide a more secure basis for this type of classification than has been available until the present time. Indeed, by examining the formal expression for the exact error in the variational approximation one may draw firm conclusions concerning its level of accuracy.

As with ordinary perturbation theory, iterative procedures may be used to generate higher-order terms in the approximation. This method was illustrated in an earlier study of low-frequency approximations.[1] In some cases it will be more efficient to employ an improved trial function, with fewer iterations required to achieve comparable accuracy. The advantages of this latter strategy in a case where a reasonably simple improved trial function is available is illustrated in the present work. An approximate wave function was introduced by Kroll and Watson[2] who used it nonvariationally to derive their now well-known low-frequency approximation. Here, following a suggestion made by Kaminski,[3] we adopt it as a trial function[4] and in so doing are able to generate a rather simpler form for the leading correction term than had been given previously[1] and to improve on its accuracy. (The accuracy of the Kroll-Watson trial function, and the variational approximation obtained from it, can be gauged in terms of two dimensionless parameters, involving the frequency and strength of the external electric field. A more careful discussion of this point is deferred until the end of Sec. II C. Here we remark that over a range of "moderate" field intensities, and for sufficiently low frequencies, both parameters will be small compared to unity. Since the error in the Kroll-Watson trial function is quadratic in these parameters it follows that the variational approximation then contains an error of *fourth* order. The approximation is useful over a wide range of field intensities, though with diminished accuracy as the intensity is increased.) The Kroll-Watson analysis dealt with potential scattering in a linearly polar-

ized laser field. It is one of the virtues of the variational approach that extensions to more complex systems can be introduced in a straightforward manner through the choice of appropriately generalized trial functions. The generalizations of the formalism treated explicitly here are to elastic and inelastic scattering by a compound target. Allowance is also made for a slowly varying field of arbitrary spectral distribution and polarization properties. The Kroll-Watson trial function contains as a factor the exact field-free scattering wave function which, of course, is rarely known in practice. The variational procedure allows for more flexibility; replacement of the exact scattering function by an approximation to it will introduce an error in the calculated transition amplitude of second order in the error in the trial function. This is a familiar feature of variational methods for field-free scattering calculations, here generalized to account for the presence of an external field.

The improved low-frequency approximation for the transition amplitude can be used to obtain a more accurate evaluation of the sum, over all final states of the field, of the partial cross sections for scattering with the emission or absorption of a definite number of photons. It is a remarkable fact that, under circumstances to be specified below, the higher-order corrections to the amplitude cancel in the calculation of the sum, which can then be evaluated in terms of the cross section for scattering in the absence of the field. The resultant sum rule is not new, but is here shown to be of higher accuracy than had been realized previously.

The basic features of the theory are outlined in Sec. II in the context of the relatively simple problem of scattering by a local, short-range potential. In Sec. III A the extension of the formalism to allow for the use of the inexact field-free scattering functions is presented and the additional complexity introduced when the target is taken to be a compound system is analyzed in Sec. III B. A summary of results appears in Sec. IV. Throughout the discussion a nonrelativistic treatment is employed and the field is taken to be spatially uniform.

II. VARIATIONAL PRINCIPLE

A. Formulation

An electron, of mass m, charge e, and initial momentum $\mathbf{p}$, scatters from a potential $V(\mathbf{r})$ in the presence of an external electric field described by the vector potential $\mathbf{A}(t)$. The field is assumed to be slowly varying, in the sense that some suitably defined average frequency of the field is small compared with any of the characteristic frequencies associated with the field-free scattering problem. (In particular, the scattering energy is assumed to be well separated from all resonance and threshold regions.) The time-dependent Schrödinger equation for this system takes the form (in units with $\hbar=1$)

$$\left[H-i\frac{\partial}{\partial t}\right]\psi_{\mathbf{p}}^{(+)}(\mathbf{r},t)=0\ , \tag{2.1}$$

with

$$H=\frac{\left[-i\nabla-\frac{e}{c}\mathbf{A}(t)\right]^2}{2m}+V(\mathbf{r})\ . \tag{2.2}$$

The motion of the electron after it has entered the field, but before it interacts with the center of force, is described by the Volkov solution

$$\varphi_{\mathbf{p}}(\mathbf{r},t)=(2\pi)^{-3/2}\exp\left[-i\int_0^t\frac{p^2(t')}{2m}dt'+i\mathbf{p}\cdot\mathbf{r}\right]\ , \tag{2.3}$$

where we have defined

$$\mathbf{p}(t)=\mathbf{p}-\frac{e}{c}\mathbf{A}(t)\ . \tag{2.4}$$

As written here, the Volkov solution does not reduce to a pure plane wave in the limit $t\to-\infty$ but rather contains a constant phase factor multiplying the plane wave; the presence of this factor is of no physical consequence. (Plane waves are used here rather than localized wave packets to simplify the presentation.) The scattering matrix may be expressed as

$$S(\mathbf{p}',\mathbf{p})=\lim_{t\to-\infty}\int d^3r\,\psi_{\mathbf{p}'}^{(-)*}(\mathbf{r},t)\varphi_{\mathbf{p}}(\mathbf{r},t)\ , \tag{2.5}$$

where the superscript $(-)$ indicates that the solution satisfies incoming-wave boundary conditions, evolving (backwards in time) from a Volkov wave function in the distant future. This exact solution may be represented formally as

$$[\psi_{\mathbf{p}'}^{(-)}(\mathbf{r},t)]^*=\varphi_{\mathbf{p}'}^*(\mathbf{r},t)+\int_{-\infty}^{\infty}dt'\int d^3r'\left[\left[H-i\frac{\partial}{\partial t'}\right]\varphi_{\mathbf{p}'}(\mathbf{r}',t')\right]^*\times G(\mathbf{r}',t';\mathbf{r},t)\ , \tag{2.6a}$$

where the retardation property of the Green's function G guarantees that

$$\psi_{\mathbf{p}'}^{(-)}-\varphi_{\mathbf{p}'}\equiv\psi_{\mathbf{p}';\mathrm{sc}}^{(-)}\to0\ ,\quad t\to\infty\ . \tag{2.6b}$$

More generally, the Volkov wave in Eq. (2.6a) may be replaced by a trial function that satisfies the proper boundary condition (2.6b). This more general version,[1] or rather its analog satisfied by the outgoing-wave solution, will be of use to us here. Thus the solution that evolves from a Volkov wave in the remote past, as expressed by the relation

$$\lim_{t\to-\infty}\int d^3r\,G(\mathbf{r}',t';\mathbf{r},t)\varphi_{\mathbf{p}}(\mathbf{r},t)=-i\psi_{\mathbf{p}}^{(+)}(\mathbf{r}',t')\ , \tag{2.7a}$$

has the representation

$$\psi_{\mathbf{p}}^{(+)}(\mathbf{r},t)=\tilde{\psi}_{\mathbf{p}}^{(+)}(\mathbf{r},t)+\int_{-\infty}^{\infty}dt'\int d^3r'G(\mathbf{r},t;\mathbf{r}',t')\times\left[H-i\frac{\partial}{\partial t'}\right]\tilde{\psi}_{\mathbf{p}}^{(+)}(\mathbf{r}',t')\ , \tag{2.7b}$$

where $\tilde{\psi}_{\mathbf{p}}^{(+)}(\mathbf{r},t)$ is a trial function which reduces to $\varphi_{\mathbf{p}}(\mathbf{r},t)$ for $t\to-\infty$. Making use of Eqs. (2.6a) and (2.7a) in Eq. (2.5), we find that

$$S(\mathbf{p}',\mathbf{p})=\delta(\mathbf{p}',\mathbf{p})-iT(\mathbf{p}',\mathbf{p}) , \tag{2.8a}$$

with

$$T(\mathbf{p}',\mathbf{p})=\int_{-\infty}^{\infty}dt\int d^3r\left[\left[H-i\frac{\partial}{\partial t}\right]\varphi_{\mathbf{p}'}(\mathbf{r},t)\right]^* \times\psi_{\mathbf{p}}^{(+)}(\mathbf{r},t) . \tag{2.8b}$$

Rather than adopting Eq. (2.8b) as the basis for approximations we replace the exact solution appearing in the integral by the representation given in Eq. (2.7b) and thereby obtain the variational identity

$$T(\mathbf{p}',\mathbf{p})=\int_{-\infty}^{\infty}dt\int d^3r\left\{\left[\left[H-i\frac{\partial}{\partial t}\right]\varphi_{\mathbf{p}'}\right]^*\tilde{\psi}_{\mathbf{p}}^{(+)} +\psi_{\mathbf{p}';\mathrm{sc}}^{(-)*}\left[H-i\frac{\partial}{\partial t}\right]\tilde{\psi}_{\mathbf{p}}^{(+)}\right\} . \tag{2.9}$$

If the exact incoming scattered wave in the second term on the right is replaced by an approximation to it, differing by a quantity of first order, the error in the transition amplitude thereby introduced will be of second order, bilinear in the errors in the two trial functions. In the following we examine in some detail the variational approximation obtained through the choice of trial functions of the form introduced by Kroll and Watson.

B. Variational approximation

Kroll and Watson considered a purely monochromatic, linearly polarized laser field. When their approximate solution is adapted to the more general class of external fields considered here we obtain the form[5]

$$\tilde{\psi}_{\mathbf{p}}^{(+)}(\mathbf{r},t)=\exp\left[-i\int_0^t dt'\frac{p^2(t')}{2m} +i\frac{e}{c}\mathbf{A}(t)\cdot\mathbf{r}\right]u_{\mathbf{p}(t)}^{(+)}(\mathbf{r}) , \tag{2.10}$$

where the function $u_{\mathbf{p}}^{(+)}$ is the field-free outgoing-wave scattering solution of the time-independent wave equation

$$\left[-\frac{\nabla^2}{2m}+V(\mathbf{r})-E_{\mathbf{p}}\right]u_{\mathbf{p}}^{(+)}(\mathbf{r})=0 , \tag{2.11}$$

with $E_{\mathbf{p}}=p^2/2m$. This function (which at this stage is assumed to be known exactly) may be decomposed into incident- and scattered-wave components as

$$u_{\mathbf{p}}^{(+)}(\mathbf{r})=(2\pi)^{-3/2}\exp(i\mathbf{p}\cdot\mathbf{r})+u_{\mathbf{p};\mathrm{sc}}^{(+)}(\mathbf{r}) . \tag{2.12}$$

A measure of the error in the trial function is obtained by determining the extent to which the Schrödinger equation (2.1) is violated. Using the (gauge transformation) property

$$\exp\left[-i\frac{e}{c}\mathbf{A}\cdot\mathbf{r}\right]H\exp\left[i\frac{e}{c}\mathbf{A}\cdot\mathbf{r}\right]=-\frac{\nabla^2}{2m}+V(\mathbf{r}) , \tag{2.13}$$

and the version of Eq. (2.11) in which $\mathbf{p}$ is replaced by $\mathbf{p}(t)$, one finds that

$$\left[H-i\frac{\partial}{\partial t}\right]\tilde{\psi}_{\mathbf{p}}^{(+)}(\mathbf{r},t)=\exp\left[-i\int_0^t dt'\frac{p^2(t')}{2m} +i\frac{e}{c}\mathbf{A}(t)\cdot\mathbf{r}\right] \times(-i\nabla_{\mathbf{p}(t)}-\mathbf{r})\cdot e\mathbf{E}(t)u_{\mathbf{p}(t)}^{(+)}(\mathbf{r}) . \tag{2.14}$$

Since the electric field $\mathbf{E}(t)$ is defined in terms of the derivative of the vector potential the error will be reduced, roughly speaking, as the characteristic frequency of the field is diminished. The linear dependence of the error on the strength of the field can also be seen from Eq. (2.14).

As mentioned, a variational approximation is obtained from the identity (2.9) by replacing the exact incoming scattered wave, appearing in the second term on the right, by a trial function. In analogy with the choice (2.10) this is taken to be

$$\psi_{\mathbf{p}';\mathrm{sc}}^{(-)}(\mathbf{r},t)=\exp\left[-i\int_0^t dt'\frac{p'^2(t')}{2m}+i\frac{e}{c}\mathbf{A}(t)\cdot\mathbf{r}\right] \times u_{\mathbf{p}'(t);\mathrm{sc}}^{(-)}(\mathbf{r}) . \tag{2.15}$$

The variational expression for the transition amplitude is the sum of two terms

$$T_v(\mathbf{p}',\mathbf{p})=T^{(1)}(\mathbf{p}',\mathbf{p})+T^{(2)}(\mathbf{p}',\mathbf{p}) , \tag{2.16}$$

corresponding to the two terms in Eq. (2.9). With the form (2.10) taken as the trial function the first term is evaluated as

$$T^{(1)}(\mathbf{p}',\mathbf{p})=\int_{-\infty}^{\infty}dt\exp[-i\Phi(t)]f(\mathbf{p}'(t),\mathbf{p}(t)) , \tag{2.17a}$$

where

$$\Phi(t)=\int_0^t dt'(E_{\mathbf{p}(t')}-E_{\mathbf{p}'(t')}) , \tag{2.17b}$$

and where

$$f(\mathbf{p}',\mathbf{p})=(2\pi)^{-3/2}\int d^3r\exp(-i\mathbf{p}'\cdot\mathbf{r})V(\mathbf{r})u_{\mathbf{p}}^{(+)}(\mathbf{r}) \tag{2.18}$$

is the (off-shell) field-free scattering amplitude. In evaluating the second term in Eq. (2.16) we make use of Eq. (2.14) to obtain

$$T^{(2)}(\mathbf{p}',\mathbf{p})=\int_{-\infty}^{\infty}dt\exp[-i\Phi(t)] \times\mathbf{g}(\mathbf{p}'(t),\mathbf{p}(t))\cdot e\mathbf{E}(t) . \tag{2.19}$$

Here we have defined the vector function

$$\mathbf{g}(\mathbf{p}',\mathbf{p})=\int d^3r\,(u_{\mathbf{p}';\mathrm{sc}}^{(-)})^*(\mathbf{r})(-i\nabla_{\mathbf{p}}-\mathbf{r})u_{\mathbf{p}}^{(+)}(\mathbf{r}) . \tag{2.20}$$

This function may be identified with the correction to the low-frequency approximation to the single-photon bremsstrahlung amplitude, as will be demonstrated below.

Suppose that the field is sufficiently slowly varying so that the correction term $T^{(2)}$, which is proportional to the electric field, is negligible compared with the leading term. This leading term may then be approximated, in many cases of interest, by an expression of the same form but with the scattering amplitude evaluated on the energy shell. Thus let us represent the scattering amplitude appearing in the expression (2.17a) as a function of the three scalar variables $E_{\mathbf{p}(t)}$, $\tau=(\mathbf{p}'-\mathbf{p})^2$, and

$$\Delta=E_{\mathbf{p}'(t)}-E_{\mathbf{p}(t)} . \tag{2.21}$$

The scattering amplitude is on the energy shell when Δ vanishes. With the energy of the incident particle taken as the reference, we note that Δ will be small if the energy transferred to or from the field is small (the basic low-frequency assumption) and if the field intensity is low enough so that the energy of interaction of the electron with the field is small. With the amplitude expanded in a Taylor series in the variable Δ about its on-shell value at $\Delta=0$, the contribution to the integral in Eq. (2.17a) arising from the term linear in Δ takes the form

$$\int_{-\infty}^{\infty} dt \exp[-i\Phi(t)]\Delta\frac{\partial f}{\partial\Delta}=-i\int_{-\infty}^{\infty} d\{\exp[-i\Phi(t)]\}\frac{\partial f}{\partial\Delta} , \tag{2.22}$$

with the derivative understood to be evaluated at $\Delta=0$. This may be integrated by parts, with the surface terms discarded in view of the infinitely rapid oscillation of the integrand for $|t|\to\infty$. [Formally, one may introduce a convergence factor $\exp(-\eta|t|)$, with η taken to be a positive infinitesimal which is allowed to vanish at the end of the calculation.] The contribution arising from the differentiation of the scattering amplitude is of higher order since it involves a time derivative of the slowly varying vector potential. The term shown in Eq. (2.22) is then seen to be of the same order as the variational correction $T^{(2)}$; they are each proportional to the electric field. When both of these terms are discarded we are left with an approximation for the transition amplitude of the form shown in Eq. (2.17a), with the field-free scattering amplitude evaluated on the energy shell. A similar result is obtained in the strong-field limit, in which the phase Φ in Eq. (2.17a) is a rapidly varying function of time. A stationary-phase evaluation of the integral places the scattering amplitude on the energy shell since the condition of stationary phase is just $\Delta=0$.

It appears likely that the arguments given above, which justify the use of the on-shell amplitude in Eq. (2.17a) in the intermediate- and strong-coupling regimes, can be generalized to apply in a uniform manner for field intensities lying in a continuous range between these limits. Indeed, when the field is taken to be purely periodic the proof is quite straightforward, as shown below in the discussion leading to Eq. (2.32)

C. Monochromatic field

To make the simplest choice for the time dependence of the vector potential, we consider a linearly polarized monochromatic field, with $\mathbf{A}(t)=\mathbf{a}\cos\omega t$. With the aid of a Fourier expansion the integration over the time variable may be performed in the variational expression for the transition amplitude, which then becomes

$$T_v(\mathbf{p}',\mathbf{p})=2\pi\sum_{n=-\infty}^{\infty}\delta(E_{\mathbf{p}'}-E_{\mathbf{p}}-n\omega)(T_n^{(1)}+T_n^{(2)}) . \tag{2.23}$$

Here we have

$$T_n^{(1)}(\mathbf{p}',\mathbf{p})=\int_0^{2\pi}\frac{d\theta}{2\pi}\exp i(n\theta-\rho\sin\theta)f(\mathbf{p}'(\theta),\mathbf{p}(\theta)) , \tag{2.24}$$

with

$$\rho=\frac{e}{mc\omega}(\mathbf{p}'-\mathbf{p})\cdot\mathbf{a} . \tag{2.25}$$

The shifted momenta are redefined as

$$\mathbf{p}(\theta)=\mathbf{p}-\frac{e}{c}\mathbf{a}\cos\theta;\quad \mathbf{p}'(\theta)=\mathbf{p}'-\frac{e}{c}\mathbf{a}\cos\theta . \tag{2.26}$$

We have effectively taken the independent variable to be $\theta=\omega t$ rather than t for notational convenience. The expression shown in Eq. (2.24) was derived nonvariationally by Kroll and Watson who then made an additional approximation by applying a stationary-phase argument, valid for $\rho\gg1$. As will be seen below, the stationary-phase approximation applied to Eq. (2.24) introduces an error of first order in the frequency. Since we are here interested in retaining corrections of this order, and since we wish to allow for ranges of field strengths and frequencies for which ρ is not large, we do not introduce the stationary-phase approximation at this stage. The variational correction term is given by expression

$$T_n^{(2)}(\mathbf{p}',\mathbf{p})=\int_0^{2\pi}\frac{d\theta}{2\pi}\exp i(n\theta-\rho\sin\theta)\times\mathbf{g}(\mathbf{p}'(\theta),\mathbf{p}(\theta))\cdot e\mathbf{E}(\theta) , \tag{2.27}$$

with $\mathbf{E}(\theta)=(\omega/c)\mathbf{a}\sin\theta$.

It is of interest to examine the weak-field limit of Eq. (2.23) since that allows us to establish a relationship between stimulated and spontaneous bremsstrahlung amplitudes; in particular, we will arrive at a physical interpretation of the matrix element $\mathbf{g}$, defined in Eq. (2.20), in terms of the amplitude for single-photon emission in the absence of an external field. This latter amplitude may be obtained by expanding the function $T_{-1}^{(1)}+T_{-1}^{(2)}$ in powers of the field strength and retaining only the linear terms. (This is exact since the error in the variational approximation is of higher order in the field strength.) For the purpose of this discussion the field-free scattering amplitude may be taken to be a function of the parameter $(e/c)a\cos\theta$ with its dependence on the remaining variables temporarily suppressed to simplify notation. Then, to first order, with a prime denoting the derivative, we

have

$$f((e/c)a\cos\theta)\cong f(0)+(e/c)a\cos\theta f' .$$

and

$$T_{-1}^{(1)}=\tfrac{1}{2}\rho\left[-f(0)+\frac{ea}{c\rho}f'\right] . \tag{2.28}$$

We see that $\cos\theta$ has effectively been replaced by $-\rho^{-1}$ in the argument of the scattering amplitude. This places the amplitude on the energy shell since, with

$$\mathbf{p}_n\equiv\mathbf{p}-\frac{nm\omega}{(\mathbf{p}'-\mathbf{p})\cdot\mathbf{a}}\mathbf{a};\quad \mathbf{p}'_n\equiv\mathbf{p}'-\frac{nm\omega}{(\mathbf{p}'-\mathbf{p})\cdot\mathbf{a}}\mathbf{a} , \tag{2.29}$$

we have $p'_n=p_n$ for a process in which n photons are absorbed ($n>0$) or emitted ($n<0$); $n=-1$ for the single-photon emission process. With the scattering amplitude once again expressed as a function of the momentum variables one finds that

$$T_{-1}^{(1)}=-\frac{e(\mathbf{p}'-\mathbf{p})\cdot\mathbf{a}}{2mc\omega}f(\mathbf{p}'_{-1},\mathbf{p}_{-1}) , \tag{2.30}$$

which is equivalent to the well-known low-frequency approximation for single-photon bremsstrahlung.[6] When the correction term in Eq. (2.27) is evaluated to first order in the field strength it reduces to

$$T_{-1}^{(2)}=-i\frac{e\omega}{2c}\mathbf{g}(\mathbf{p}',\mathbf{p})\cdot\mathbf{a} . \tag{2.31}$$

We confirm in this way that the error in the low-frequency approximation (2.30) is of first order in the frequency since the coefficient of ω in the correction term involves the integral (2.20) which converges for ω arbitrarily small. [The dependence of the integral on frequency enters through the energy-conservation condition. The fact that the integral diverges in the zero-frequency limit when the wave functions satisfy Coulomb boundary conditions at infinity is an indication that the standard approximation shown in Eq. (2.30) requires modification in that case, as does the approximation (2.24) for scattering in an external field, when terms of higher order in the frequency are included.[1]] Equation (2.31) provides us with a physical interpretation (and possibly an independent method of determination) of the matrix element $\mathbf{g}$ which appears in the correction term (2.27).

It follows from Eq. (2.24), after a process of repeated partial integrations, that

$$T_n^{(1)}(\mathbf{p}',\mathbf{p})=J_n(\rho)f(\mathbf{p}'_n,\mathbf{p}_n)+O(\varepsilon_1\varepsilon_2) , \tag{2.32}$$

where J_n is the cylindrical Bessel function; we have introduced the dimensionless parameters

$$\varepsilon_1=\frac{\omega}{p^2/2m}\ ;\quad \varepsilon_2=\frac{ea}{cp} . \tag{2.33}$$

We assume throughout that ε_1 is small compared to unity and that ε_2 is at most of order unity. [The parameter ρ in Eq. (2.25) is roughly of order $\varepsilon_2/\varepsilon_1$.] In terms of these parameters we see that the Kroll-Watson approximation,[2] reproduced as the leading term in Eq. (2.32), contains an error of second order for ε_2 comparable in magnitude to ε_1. For ε_2 of order unity—the strong-field domain—the error is of first order in the frequency and the first-order momentum shifts appearing in the arguments of the scattering amplitude should be dropped for consistency.

D. Sum rule

It was suggested some time ago,[7] in the context of a relativistic formulation of the problem of scattering in a radiation field, that the summation of the cross section over all final states of the field may, through a cancellation of higher-order corrections, give a result (a sum rule) more accurate than expected on the basis of the accuracy of the input amplitudes. A demonstration of such a cancellation was provided in an earlier treatment of the problem of nonrelativistic scattering in a laser field.[8] Here we are able to simplify the derivation of the sum rule considerably and to further extend its domain of applicability. In particular, with the parameters ε_1 and ε_2 (introduced above) both taken to be small a sum rule is derived which is correct to third order and which involves only the field-free scattering cross section. The contributions from the correction terms shown in Eq. (2.27) cancel in the evaluation of the sum, as do the off-shell components of the field-free scattering amplitude which appears in Eq. (2.24). Interestingly, the sum rule may be interpreted in terms of the classical motion of the electron in the field, with the collision taking place instantaneously and without influence from the field.

With the variational approximation (2.23) adopted, the sum to be evaluated is

$$\sigma=\frac{m(2\pi)^4}{p}\sum_{n=-\infty}^{\infty}\int d^3p'\delta(E_{\mathbf{p}'}-E_{\mathbf{p}}-n\omega)\times|T_n^{(1)}(\mathbf{p}',\mathbf{p})+T_n^{(2)}(\mathbf{p}',\mathbf{p})|^2 . \tag{2.34}$$

The unitarity relation satisfied by the transition amplitude provides us with a convenient method for the evaluation of the sum over final states. With the use of standard methods of time-independent scattering theory,[9] the exact amplitude $T_n(\mathbf{p}',\mathbf{p})$ for scattering with the exchange of n photons with the field may be shown to satisfy the generalized optical theorem

$$\mathrm{Im}T_0(\mathbf{p},\mathbf{p})=-\pi\sum_{n=-\infty}^{\infty}\int d^3q|T_n(\mathbf{q},\mathbf{p})|^2\times\delta(E_{\mathbf{q}}-E_{\mathbf{p}}-n\omega) . \tag{2.35}$$

The variational approximation $T_n\cong T_n^{(1)}+T_n^{(2)}$ is used in the evaluation of the left- and right-hand sides of this equation. It is seen from Eq. (2.24) that

$$\mathrm{Im}T_0^{(1)}(\mathbf{p},\mathbf{p})=\int_0^{2\pi}\frac{d\theta}{2\pi}\mathrm{Im}f(\mathbf{p}(\theta),\mathbf{p}(\theta)) . \tag{2.36}$$

The optical theorem satisfied by the field-free scattering amplitude may now be used to arrive at the relation

$$\mathrm{Im}T_0^{(1)}(\mathbf{p},\mathbf{p})=\int_0^{2\pi}\frac{d\theta}{2\pi}\left[-\pi\int d^3q\,|f(\mathbf{q}(\theta),\mathbf{p}(\theta))|^2 \times\delta(E_{\mathbf{q}(\theta)}-E_{\mathbf{p}(\theta)})\right] . \tag{2.37}$$

From Eq. (2.27) we have

$$\mathrm{Im}T_0^{(2)}(\mathbf{p},\mathbf{p})=\int_0^{2\pi}\frac{d\theta}{2\pi}\frac{e\omega}{c}\sin\theta\,\mathrm{Img}(\mathbf{p}(\theta),\mathbf{p}(\theta))\cdot\mathbf{a} . \tag{2.38}$$

This integral vanishes since, with $x=\cos\theta$, it can be expressed as an integral of a function of x with upper and lower limits both equal to unity. Gathering together our results we obtain the sum rule

$$\sigma=\frac{m(2\pi)^4}{p}\int_0^{2\pi}\frac{d\theta}{2\pi}\int d^3q\,|f(\mathbf{q}(\theta),\mathbf{p}(\theta))|^2 \times\delta(E_{\mathbf{q}(\theta)}-E_{p(\theta)}) . \tag{2.39}$$

The error in this sum rule is of order $(\varepsilon_1\varepsilon_2)^2$. In the domain where $\varepsilon_2\sim\varepsilon_1$ the error is of fourth order, as stated above. The level of accuracy which can be claimed decreases as ε_2 increases; for ε_2 of order unity the error is estimated to be of order ε_1^2.

The phase-space integration in Eq. (2.34) was allowed to run over all directions of the scattered electron. More generally, the integration may be restricted to an arbitrary infinitesimal domain. The sum rule holds for this case too, but the derivation is rather more complicated since the optical theorem cannot be utilized; the details of the derivation will not be presented here.

III. SOME EXTENSIONS OF THE FORMALISM

A. Inexact field-free wave functions

If, as is almost always the case, the scattering wave functions $u_{\mathbf{p}}^{(\pm)}$ are imprecisely known one must introduce trial functions

$$\tilde{u}_{\mathbf{p}}^{(\pm)}=u_{\mathbf{p}}^{(\pm)}+\delta u_{\mathbf{p}}^{(\pm)} \tag{3.1}$$

in their place. The error in the variational approximation thereby introduced will be of second order, that is, bilinear in the errors (from all sources) in the time-dependent trial functions that appear in the approximation (2.16) for the transition amplitude. Let us now verify this explicitly by demonstrating that contributions which are linear in the error functions $\delta u_{\mathbf{p}}^{(\pm)}$ cancel when the integrals are evaluated. This is equivalent to showing that the new version of the variational approximation is, to first order, identical to the one originally obtained using the exact field-free wave functions. To simplify the discussion the field will be taken to be monochromatic. The small parameters in the problem may be identified as ε_1 and $\delta u_p^{(\pm)}$, with ε_2 at most of order unity. We follow standard conventions in treating the small parameters as being, formally, of comparable order so that, for example, terms bilinear in these parameters are counted as second-order quantities.

It will be convenient at this point to make use of Dirac notation, in which case Eq. (2.18), for example, is rewritten as

$$f(\mathbf{p}',\mathbf{p})=\langle\mathbf{p}'|V|u_{\mathbf{p}}^{(+)}\rangle . \tag{3.2}$$

When the exact scattering functions are replaced by trial functions, and terms of second order are discarded, the variational approximation becomes

$$T_v(\mathbf{p}',\mathbf{p})=\int_{-\infty}^{\infty}dt\,\exp[-i\Phi(t)] \times[\tilde{f}(\mathbf{p}'(t),\mathbf{p}(t))+\tilde{\mathbf{g}}(\mathbf{p}'(t),\mathbf{p}((t))\cdot e\mathbf{E}(t)] . \tag{3.3}$$

The function $\tilde{\mathbf{g}}$ differs from $\mathbf{g}$ defined in Eq. (2.20) in the appearance of trial rather than exact field-free wave functions. Since $\mathbf{g}\cdot e\mathbf{E}$ is of first order it is clear that no first-order error is introduced by the replacement of $\mathbf{g}$ with $\tilde{\mathbf{g}}$ in this expression. The replacement for the exact scattering amplitude is

$$\tilde{f}(\mathbf{p}'(t),\mathbf{p}(t))=\langle\mathbf{p}'(t)|V|\tilde{u}_{\mathbf{p}(t)}^{(+)}\rangle +\langle\tilde{u}_{\mathbf{p}'(t);\mathrm{sc}}^{(-)}|H_0-E_{\mathbf{p}(t)}|\tilde{u}_{\mathbf{p}(t)}^{(+)}\rangle , \tag{3.4}$$

where H_0 is the field-free Hamiltonian. It remains to verify that the approximation (3.3) is unchanged to first order when $\tilde{f}$ is replaced by f, and we now turn to this matter.

We write the difference between the first term in Eq. (3.3) and the expression shown in Eq. (2.17a) as

$$\delta T^{(1)}=\int_{-\infty}^{\infty}dt\,\exp[-i\Phi(t)]\delta f(\mathbf{p}'(t),\mathbf{p}(t)) \tag{3.5}$$

with $\delta f\equiv\tilde{f}-f$. To show that the integral is of second order we prove that this is the case for each of the Fourier components

$$\delta T_n^{(1)}=\int_0^{2\pi}\frac{d\theta}{2\pi}\exp i(n\theta-\rho\sin\theta)\delta f(\mathbf{p}'(\theta),\mathbf{p}(\theta)) . \tag{3.6}$$

Following our earlier procedure, we treat δf as a function of $(e/c)a\cos\theta$ and expand the function in powers of this variable. By a process of repeated partial integrations we find that

$$\delta T_n^{(1)}=J_n(\rho)\delta f(\mathbf{p}_n',\mathbf{p}_n)[1+O(\varepsilon_1\varepsilon_2)] , \tag{3.7}$$

with the momentum shift defined in Eq. (2.29), so that $p_n'=p_n$. The error term in Eq. (3.7) may be ignored since the leading term is itself at least of first order. We now demonstrate that it is in fact of second order. Bearing in mind that the initial and final momenta are equal in magnitude we drop the subscript n, to simplify notation, and write out the expression for the error in the on-shell scattering amplitude as

$$\delta f[\mathbf{p}',\mathbf{p}]=\langle\mathbf{p}'|V|\delta u_{\mathbf{p}}^{(+)}\rangle+\langle\tilde{u}_{\mathbf{p}';\mathrm{sc}}^{(-)}|H_0-E_{\mathbf{p}}|\delta u_{\mathbf{p}}^{(+)}\rangle . \tag{3.8}$$

In the second term on the right we integrate by parts (Green's theorem), and note that the surface terms vanish

since the two functions in the integrand behave asymptotically as outgoing waves of the same energy. From the relation

$$(H-E_{\mathbf{p}})|\tilde{u}_{\mathbf{p}';\mathrm{sc}}^{(-)}\rangle=-V|\mathbf{p}'\rangle+O(\delta u_{\mathbf{p}'}^{(-)}) , \tag{3.9}$$

it is seen that the terms of first order cancel in Eq. (3.8), and hence in Eq. (3.6) as well. This completes the proof.

It may be worth remarking, in passing, that the above considerations suggest a variational principle for the field-free (on-shell) scattering amplitude based on the variational identity

$$f(\mathbf{p}',\mathbf{p})=\langle\mathbf{p}'|V|\tilde{u}_{\mathbf{p}}^{(+)}\rangle+\langle u_{\mathbf{p}';\mathrm{sc}}^{(-)}|H_0-E_{\mathbf{p}}|\tilde{u}_{\mathbf{p}}^{(+)}\rangle , \tag{3.10}$$

valid for $p'=p$. This identity represents, clearly, the time-independent analog of Eq. (2.9). The variational principle is similar to the well-known Kohn version[10] and may prove to be more convenient in some cases; the leading term is given explicitly, rather being defined in terms of the asymptotic form of the trial function, and it appears (if the use of the Born approximation for the trial function is a good indication) to provide a better first approximation than the trial amplitude in the Kohn principle. Both of these potential advantages were realized in the application of the analogous time-dependent version discussed above. We saw, in fact, that the leading term reproduced the Kroll-Watson approximation which, for fields of moderate intensity, includes the correction term of first order in the frequency. On the other hand, the amplitude defined by the asymptotic form of the trial function is difficult to represent in explicit form; for a similar, but somewhat simpler version of the trial function[1] the trial amplitude can be readily determined, and is seen to be missing the term of first order in the frequency.

B. Scattering by a compound target

The variational approach to the development of improved low-frequency approximations will now be generalized to allow for scattering by a bound system of charged particles. It is known from earlier work[11] that the Kroll-Watson approximation preserves its form under this generalization, to first order in the frequency, and that modifications appear in second order. These modifications will be determined here. [To be somewhat more specific concerning the orders of the correction terms let us consider the situation in which the parameters ε_1 and ε_2 of Eq. (2.33) are comparable in magnitude, each of the order of the small parameter ε. The variational approximation developed here is, in that case, correct to third order in ε.] With the target-field interaction described in the length gauge, one might anticipate the modifications to include that in which the dipole interaction in Eq. (2.20) is altered by the replacement of $\mathbf{r}$, the projectile coordinate, by $\mathbf{r}+\mathbf{R}$, where $\mathbf{R}$ represents the sum of all the position vectors of the bound particles. In addition, the target wave function, as it appears in the construction of the scattering amplitude present in the leading term of the low-frequency approximation [see Eq. (2.17a)], should be replaced by one which reflects the first-order perturbation caused by the electric-dipole interaction. A direct generalization of the procedure described above for potential scattering (with undistorted target wave functions used in the construction of the trial functions) shows that these expectations are correct. To focus on the essential features the target will be assumed to be in its ground state before and after the collision and the particles will be treated as distinguishable—the effect of the Pauli principle can eventually be included by forming the appropriate linear combination of direct and exchange amplitudes.

Volkov waves are defined in the case under consideration as

$$\varphi_{\mathbf{p}}=(2\pi)^{-3/2}b_1(\mathbf{R})\exp\left[-iE_1t-i\int_0^t dt'\frac{p^2(t')}{2m}+i\frac{e}{c}\mathbf{A}(t)\cdot\mathbf{R}+i\mathbf{p}\cdot\mathbf{r}\right] , \tag{3.11}$$

where the ground state of the isolated target satisfies

$$(H_T-E_1)b_1(\mathbf{R})=0 . \tag{3.12}$$

The wave equation for the scattering system in the absence of the field is

$$(H_0-E_1-E_{\mathbf{p}})u_{\mathbf{p}}^{(+)}(\mathbf{r},\mathbf{R})=0 , \tag{3.13}$$

where $H_0=H_T+K+V$ is the sum of the target Hamiltonian, the projectile kinetic-energy operator and the projectile-target interaction energy. In terms of these solutions (which we take to be known functions in the following) a trial function is introduced of the form[3]

$$\tilde{\psi}_{\mathbf{p}}^{(+)}(\mathbf{r},\mathbf{R},t)=\exp\left[-iE_1t-i\int_0^t dt'\frac{p^2(t')}{2m}+i\frac{e}{c}\mathbf{A}(t)\cdot(\mathbf{r}+\mathbf{R})\right]u_{\mathbf{p}(t)}^{(+)}(\mathbf{r},\mathbf{R}) , \tag{3.14}$$

with $\mathbf{p}(t)=\mathbf{p}-(e/c)\mathbf{A}(t)$, as defined previously. To simplify notation the variational identity (2.9), which applies directly to the problem at hand, is rewritten as

$$T(\mathbf{p}',\mathbf{p})=\left\langle\!\!\left\langle\left[H-i\frac{\partial}{\partial t}\right]\varphi_{\mathbf{p}'}\middle|\tilde{\psi}_{\mathbf{p}}^{(+)}\right\rangle\!\!\right\rangle+\left\langle\!\!\left\langle\psi_{\mathbf{p}';\mathrm{sc}}^{(-)}\middle|\left[H-i\frac{\partial}{\partial t}\right]\tilde{\psi}_{\mathbf{p}}^{(+)}\right\rangle\!\!\right\rangle , \tag{3.15}$$

where the double bracket indicates that both space and time integrations are to be performed in the definition of the scalar product. A variational approximation is obtained by replacing the exact incoming scattered wave by a trial function, constructed here along the lines indicated in Eq. (3.14). We note that the generalized Volkov function satisfies

$$\left[H-i\frac{\partial}{\partial t}\right]\varphi_{\mathbf{p}'}=(V-e\mathbf{E}\cdot\mathbf{R})\varphi_{\mathbf{p}'} . \tag{3.16}$$

and that $\langle b_1|\mathbf{R}|b_1\rangle=0$, assuming the ground state to have well-defined parity. These relations are used to write the variational approximation in the form

$$T_v=T^{(1)}+T^{(2)}+T^{(3)} . \tag{3.17}$$

Here we have

$$T^{(1)}=\langle\langle\varphi_{\mathbf{p}'}|V|\tilde{\psi}^{(+)}_{\mathbf{p}}\rangle\rangle , \tag{3.18}$$

$$T^{(2)}=\langle\langle\tilde{\psi}^{(-)}_{\mathbf{p}';\mathrm{sc}}|[-i\nabla_{\mathbf{p}(t)}-(\mathbf{r}+\mathbf{R})]\cdot e\mathbf{E}|\tilde{\psi}^{(+)}_{\mathbf{p};\mathrm{sc}}\rangle\rangle . \tag{3.19}$$

$$T^{(3)}=\langle\langle\tilde{\psi}^{(-)}_{\mathbf{p}';\mathrm{sc}}|-e\mathbf{E}\cdot\mathbf{R}|\varphi_{\mathbf{p}}\rangle\rangle+\langle\langle\varphi_{\mathbf{p}'}|-e\mathbf{E}\cdot\mathbf{R}|\tilde{\psi}^{(+)}_{\mathbf{p};\mathrm{sc}}\rangle\rangle . \tag{3.20}$$

To clarify the structure of the last term let us decompose the field-free scattering function as the sum of a plane-wave component $|\chi_{\mathbf{p}}\rangle=|b_1\rangle|\mathbf{p}\rangle$ and a scattered wave

$$|u^{(+)}_{\mathbf{p};\mathrm{sc}}\rangle=g_T(E-K)F(E)|\chi_{\mathbf{p}}\rangle , \tag{3.21}$$

where $E=E_1+E_{\mathbf{p}}$ is the scattering energy,

$$g_T(E)=(E-H_T)^{-1} \tag{3.22}$$

is the target Green's function, and

$$F(E)=V+V(E-H_0)^{-1}V \tag{3.23}$$

is the scattering operator. In evaluating the second term on the right in Eq. (3.20) we encounter the matrix element

$$\langle\chi_{\mathbf{p}'}|\mathbf{R}|u^{(+)}_{\mathbf{p};\mathrm{sc}}\rangle=\langle b_1|\mathbf{R}g_T(E-E_{\mathbf{p}'})\langle\mathbf{p}'|F(E)|\mathbf{p}\rangle|b_1\rangle . \tag{3.24}$$

This structure suggests that the sum of the amplitudes $T^{(1)}$ and $T^{(3)}$ may be expressed in terms of the matrix element of the field-free scattering operator, taken with respect to the perturbed bound-state wave function. The result will be written out here for the case of the monochromatic field considered earlier. The variational approximation is of the form

$$T_v=2\pi\sum_n\delta(E_{\mathbf{p}'}-E_{\mathbf{p}}-n\omega)(T^{(1)}_n+T^{(2)}_n+T^{(3)}_n) . \tag{3.25}$$

For the second term on the right we have

$$T^{(2)}_n=\int_0^{2\pi}\frac{d\theta}{2\pi}\exp i(n\theta-\rho\sin\theta)\langle u^{(-)}_{\mathbf{p}'(\theta);\mathrm{sc}}|[-i\nabla_{\mathbf{p}(\theta)}-(\mathbf{r}+\mathbf{R})\cdot e\mathbf{E}(\theta)]|u^{(+)}_{\mathbf{p}(\theta);\mathrm{sc}}\rangle . \tag{3.26}$$

The first and third terms are combined to give

$$T^{(1)}_n+T^{(3)}_n=\int_0^{2\pi}\frac{d\theta}{2\pi}\exp i(n\theta-\rho\sin\theta)\tilde{f}(\mathbf{p}'(\theta),\mathbf{p}(\theta)) . \tag{3.27}$$

Here we define the modified scattering amplitude

$$\tilde{f}(\mathbf{p}'(\theta),\mathbf{p}(\theta))=\langle\bar{b}(-\Delta)|\langle\mathbf{p}'(\theta)|F(E_{\mathbf{p}(\theta)}+E_1)|\mathbf{p}(\theta)\rangle|\bar{b}(\Delta)\rangle , \tag{3.28}$$

with

$$|\bar{b}(\Delta)\rangle=|b_1\rangle+g_T(E_1+\Delta)[-e\mathbf{E}(\theta)\cdot\mathbf{R}]|b_1\rangle , \tag{3.29}$$

and $\Delta=E_{\mathbf{p}'(\theta)}-E_{\mathbf{p}(\theta)}$. The modified amplitude in Eq. (3.28) differs from the field-free (off-shell) scattering amplitude in the replacement of the target wave functions by field-distorted states. With the neglect of the energy shift Δ in Eq. (3.29) these states correspond to the result of applying first-order time-independent perturbation theory in the presence of the static external field $\mathbf{E}(\theta)$. [In arriving at Eq. (3.27) a term quadratic in the electric-dipole interaction has been added, which is consistent since the error in the variational approximation is of this order.] The picture suggested by this equation is one in which the collision takes place instantaneously, with both the projectile momentum and the distortion of the target determined by the phase of the field at the moment of collision; the resultant amplitude is then averaged over the phase.

The dressing of the target by the field, in the form shown in Eq. (3.29), can lead to a dramatic enhancement of the cross section for the forward scattering of an electron by a neutral atom. Virtual excitations of the target by the field induce virtual inelastic collisions which, if optically allowed, behave in the forward direction as the inverse of the momentum transfer. This effect has been studied in detail in the context of a Born approximation approach to the scattering problem, with no restriction on the frequency.[12] The approach adopted here is confined to the low-frequency domain but the field-free scattering is treated without approximation. We emphasize that the dynamical effect leading to the forward enhancement is not accounted for in the (generalized) Kroll-Watson trial function but is properly included in the variational correction term.

IV. DISCUSSION

It is a characteristic and useful feature of low-frequency approximations that they allow one to evaluate the rate for a radiative process in terms of one of lower order, which can be either calculated or measured more easily. The Kroll-Watson approximation represents the amplitude for scattering in a laser field in terms of the field-free amplitude, with an error which is of second order in the frequency for fields of moderate intensity. Here we have seen that through the use of a variational procedure still higher accuracy can be achieved (a second-order error in the trial function introduces an er-

ror of fourth order in the amplitude) by including a correction term, shown in Eq. (2.19), which can be determined from a knowledge of the amplitude for the spontaneous emission of a single photon during the collision. These error estimates are based on the use of the exact field-free wave function in the calculation. If this function contains a first-order error one expects that an additional error of *second* order will be introduced in the final result, and this has been verified here explicitly. We have illustrated a remarkable cancellation of correction terms in the calculation of the cross section summed over final states of the field. As a consequence we obtain a sum rule, expressed in terms of the measurable field-free cross section, whose accuracy exceeds that of the input amplitude. The approach is applicable to electron-atom scattering, with the variational procedure accounting in a natural way for the dressing of the target by the field. The calculational method must be modified before it can be applied to scattering in the neighborhood of a resonance, or near an excitation threshold, where the scattering amplitude is a rapidly varying function of the energy. The appearance of the momentum gradient in the variational correction term [see Eqs. (2.19) and (2.20)] provides an indication that the numerical value of the correction can be large as a result of a strong energy dependence in the field-free scattering problem. This difficulty may be avoided through the choice of a more physically appropriate trial function, a matter which we plan to discuss in a separate paper.

ACKNOWLEDGMENTS

This work was supported in part by the National Science Foundation under Grant No. PHY87-06114. The work was completed while one of us (L.R.) was a visitor at the Joint Institute for Laboratory Astrophysics (JILA). He wishes to thank the staff and personnel of JILA for their hospitality during his visit.

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[5]With the introduction of wave packets to localize the projectile, and with the use of a wave train of finite extent to represent the field, trial functions may be constructed that properly reduce to the Volkov function at asymptotic times. The form written in Eq. (2.10) may be considered to be the limit of such a proper trial function as the wave packets are taken to be of arbitrarily large extent. The error introduced by the adoption of this limiting form may be shown to be negligible in situations of physical interest.

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