Strong-field S-matrix theory with final-state Coulomb interaction in all orders

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Phys. Rev. A **94**, 031401 — Published 6 September 2016
DOI: 10.1103/PhysRevA.94.031401

Strong-Field S-Matrix Theory with Final State Coulomb Interaction in All Orders

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During the last several decades the so-called Keldysh-Faisal-Reiss (KFR) or strong-filed approximation (SFA) has been highly usefulness for the analysis of atomic and molecular processes in intense laser fields. However, it is well known that SFA does *not* account for the final-state Coulomb interaction which is, however, unavoidable for the ubiquitous ionization process. In this Letter we solve this long standing problem and give a complete strong-field S-matrix expansion that accounts for the final-state Coulomb interaction in all orders, explicitly.

PACS numbers: 32.80.Rm,42.50.Hz, 34.80.Qb

Over the past several decades the well-known strong-field approximation in the form of the so-called Keldysh-Faisal-Reiss or strong field approximation (KFR/SFA) [1–3] has provided much fruitful insights into the highly non-perturbative processes which occur during the interaction of intense laser fields with atoms and molecules. However, it is also well-known that SFA, based as it is on the plane-wave Volkov wavefunction, which is the wavefunction of a free electron interacting with a laser field, does not account for the Coulomb interaction in the final state. The latter interaction due to its long range, however, is unavoidable for the ubiquitous ionization and related processes in strong laser fields. Consequently, besides full numerical simulations of the time-dependent Schroedinger equation (if and when feasible) many attempts were made in the past to account for the Coulomb effect within the SFA using various *heuristic* corrections. Early examples of them are Coulomb corrections to the strong-field electron-detachment models [4–6], and Coulomb correction via WKB-like approximations [7, 8], or using semi-classical and/or "quantum trajectory" approach [9–11]. Another early work [12] employed the so-called Coulomb-Volkov state (that was defined a long-time ago [13]) in an *ad hoc* one-step model of strong-field ionization. Later on a similar model using an "adiabatic" version of the same was used [14]. More recently, a semi-analytic R-matrix approach [15] and an approach employing mixed ansätze with phase corrections and an inhomogeneous differential equation [16] have been introduced to account for the Coulomb effect. However, till now, no strong-field S-matrix expansion has been found that, unlike the usual SFA, can account for the final-state Coulomb interaction in the laser field, systematically to all orders. The purpose of this paper is to solve this long standing problem and give a complete Coulomb-Volkov S-matrix series that accounts for the same in all orders, explicitly.

The rest of the work is organised as follows: first, we determine (a) the exact Coulomb-Volkov Hamiltonian, (b) find the complete set of linearly independent time-dependent solutions of the corresponding Schroedinger equation, (c) construct the exact Coulomb-Volkov propagator (or Green's function) and, (d) identify the rest-interaction associated with the final state. Then, the above analytical information are used to derive the complete Coulomb-Volkov S-matrix series of interest. We also consider the limiting case in the absence of the Coulomb interaction. The final S-matrix expansions are given in the commonly used "velocity" and "length" gauges. We conclude with a few additional observations on the results obtained.

The Schroedinger equation of the interacting atom+ laser field is

$$(i\hbar\frac{\partial}{\partial t} - H(t))|\Psi(t)\rangle = 0 \tag{1}$$

where $H(t)$ is the total Hamiltonian of the system,

$$H(t) = H_a + V_i(t) \tag{2}$$

For the sake of concreteness, we assume an effective one electron atomic system interacting with a laser field and take

$$H_a = (\frac{\mathbf{p}_{op}{}^2}{2m} - \frac{Ze^2}{r} + V_{s.r.}(\mathbf{r})) \tag{3}$$

where Z is the core charge and $V_{s.r.}(\mathbf{r})$ is a short-range potential that goes to zero for asymptotically large r faster than the Coulomb potential $-\frac{Ze^2}{r}$. The laser-atom interaction is taken in the "velocity gauge" (i.e. the minimal coupling radiation gauge in "dipole" approximation)

$$V_i(t) = (-\frac{e}{mc}\mathbf{A}(t)\cdot\mathbf{p}_{op} + \frac{e^2A^2(t)}{2mc^2}) \tag{4}$$

where $\mathbf{A}(t)$ is the vector potential of the laser field, and $\mathbf{p}_{op} \equiv -i\hbar\nabla$.

For the final state, we intend to take account of the laser and the long-range Coulomb interaction explicitly. This can be done employing the so-called Coulomb-Volkov state. This was originally defined heuristically in

[13] by taking the stationary Coulomb-wave, $\phi_{\mathbf{p}}(\mathbf{r})$, and augmenting it with the time-dependent Volkov-phase:

$$\Phi_{\mathbf{p}}(\mathbf{r},t) = \phi_{\mathbf{p}}(\mathbf{r}) e^{-\frac{i}{\hbar}\int^t (\frac{p^2}{2m} - \frac{e}{c}\mathbf{A}(t)\cdot\mathbf{p} + \frac{e^2 A^2(t)}{2mc^2})dt'} \quad (5)$$

where [18],

$$\phi_{\mathbf{p}}(\mathbf{r}) = \frac{1}{L^{\frac{3}{2}}} e^{\frac{\pi}{2}\eta_p}\Gamma(1+i\eta_p) e^{\frac{i}{\hbar}\vec{p}\cdot\vec{r}} \times {}_1F_1(-i\eta_p, 1, -i(pr + \mathbf{p}\cdot\mathbf{r})) \quad (6)$$

($\eta_p \equiv \frac{Z\hbar}{a_0 p}$, $a_0 = \frac{\hbar^2}{me^2}$) are the "in-going" continuum eigenstates of energy $E_p = \frac{p^2}{2m}$, of the Coulomb Hamiltonian

$$\begin{aligned} H_{Cou} &= H_a - V_{s.r.}(\mathbf{r}) \\ &= (\frac{\mathbf{p}_{op}^2}{2m} - \frac{Ze^2}{r}). \end{aligned} \quad (7)$$

Note that the ansatz (5) does *not* fully satisfy the Schroedinger equation (1) of the interacting system. We may, therefore, ask: what is the Hamiltonian or, the Schroedinger equation of which the Coulomb-Volkov state, Eq. (5), is an *exact* solution? Essentially it is the lack of this information that had so far hindered the development of a systematic S-matrix theory based on the Coulomb-Volkov state. To overcome this hindrance here we shall first determine the Coulomb-Volkov Hamiltonian (to be denoted H_{CV} below) and the complete set of linearly independent solutions of the associated Schroedinger equation. This will allow us to construct the exact Coulomb-Volkov propagator, G_{CV}, and identify the corresponding rest-interaction in the final state, $V_{CV}(t)$. Once they are known, we can use them directly in a convenient form of the total wavefunction $\Psi_i(t)$ evolving from a given initial state $\phi_i(t)$. We use a formal expression of $\Psi_i(t)$ that was first obtained in connection with the so-called intense-field many-body S-matrix theory or IMST (e.g. review [17], sec. 3) and write it here as:

$$\begin{aligned} |\Psi_i(t)\rangle &= |\phi_i(t)\rangle + \int dt_1 G_{CV}(t,t_1)V_i(t_1)|\phi_i(t_1)\rangle \\ &+ \int dt_2 dt_1 G_{CV}(t,t_2)V_{CV}(t_2) \\ &\times G(t_2,t_1)V_i(t_1)|\phi_i(t_1)\rangle \end{aligned} \quad (8)$$

where the propagator $G(t,t')$ satisfies the equation

$$G(t,t') = G_{CV}(t,t') + \int G_{CV}(t,t_1)V_{CV}(t_1)G(t_1,t')dt_1 \quad (9)$$

The final state Coulomb-Volkov propagator $G_{CV}(t,t')$ is to be found, once $H_{CV}(t)$ is known, from the solution of the inhomogeneous equation

$$(i\hbar\frac{\partial}{\partial t} - H_{CV}(t))G_{CV}(t,t') = \delta(t-t') \quad (10)$$

To determine the Hamiltonian $H_{CV}(t)$ we first introduce a vector operator defined by

$$\mathbf{h}_{op} \equiv \sum_{\mathbf{s}} |\phi_{\mathbf{s}}\rangle\mathbf{s}\langle\phi_{\mathbf{s}}| \quad (11)$$

where, $\Sigma_{\mathbf{s}}(\cdots) \equiv (\frac{L}{2\pi})^3\int d^3s(\cdots)$, and $|\phi_{\mathbf{s}}\rangle$ stands for the Coulomb *continuum* waves with momentum $\mathbf{s}$ (cf. Eq. (6)). Next, we consider the exponential operator

$$T(\mathbf{h}_{op}) = e^{i\mathbf{a}(t)\cdot\mathbf{h}_{op}} \quad (12)$$

where $\mathbf{a}(t) = \frac{e}{mc}\int^t \mathbf{A}(t')dt'$. By expanding the exponential as a power series and using the projection operator nature of the individual terms, it can be reduced to the simple form

$$T(\mathbf{h}_{op}) = 1 - \sum_{\mathbf{s}} |\phi_{\mathbf{s}}\rangle(1 - e^{i\mathbf{a}(t)\cdot\mathbf{s}})\langle\phi_{\mathbf{s}}| \quad (13)$$

With the help of the operator $\mathbf{h}_{op}$, we can write down the Coulomb-Volkov Hamiltonian $H_{CV}(t)$:

$$H_{CV}(t) = \frac{\mathbf{p}_{op}^2}{2m} - \frac{Ze^2}{r} + \frac{e^2A^2(t)}{2mc^2} - \frac{e}{mc}\mathbf{A}(t)\cdot\mathbf{h}_{op} \quad (14)$$

The corresponding Schroedinger equation is

$$i\hbar\frac{\partial}{\partial t}\Phi_j(t) = (\frac{\mathbf{p}_{op}^2}{2m} - \frac{Ze^2}{r} + \frac{e^2A^2(t)}{2mc^2} - \frac{e}{mc}\mathbf{A}(t)\cdot\mathbf{h}_{op}) \times \Phi_j(t) \quad (15)$$

The complete set of linearly independent solutions of Eq. (15) is

$$|\Phi_j(t)\rangle = e^{-\frac{i}{\hbar}\int^t (E_j + \frac{e^2A^2(t')}{2mc^2})dt' + \frac{i}{\hbar}\mathbf{a}(t)\cdot\mathbf{h}_{op}}|\phi_j\rangle \quad (16)$$

where $j \equiv \mathbf{p}$, stands for the momentum $\mathbf{p}$ of the Coulomb wave state $|\phi_{\mathbf{p}}\rangle$ and $j \equiv D$ stands for the discrete indices of the bound states $|\phi_D\rangle$ of the Coulomb potential.

To establish that Eq.(16) indeed satisfies Eq. (15), let us first consider the case $\{j \equiv \mathbf{p}\}$ and use Eq. (13) to calculate,

$$\begin{aligned} e^{\frac{i}{\hbar}\mathbf{a}(t)\cdot\mathbf{h}_{op}}|\phi_{\mathbf{p}}\rangle &= T(\mathbf{h}_{op})|\phi_{\mathbf{p}}\rangle \\ &= |\phi_{\mathbf{p}}\rangle - \sum_{\mathbf{s}}|\phi_{\mathbf{s}}\rangle(1 - e^{\frac{i}{\hbar}\mathbf{a}(t)\cdot\mathbf{s}})\langle\phi_{\mathbf{s}}|\phi_{\mathbf{p}}\rangle \\ &= |\phi_{\mathbf{p}}\rangle - |\phi_{\mathbf{p}}\rangle(1 - e^{\frac{i}{\hbar}\mathbf{a}(t)\cdot\mathbf{p}}) \\ &= e^{\frac{i}{\hbar}\mathbf{a}(t)\cdot\mathbf{p}}|\phi_{\mathbf{p}}\rangle \end{aligned} \quad (17)$$

Also we have

$$-\frac{e}{mc}\mathbf{A}(t)\cdot\mathbf{h}_{op}|\phi_{\mathbf{p}}\rangle = -\frac{e}{mc}\mathbf{A}(t)\cdot\mathbf{p}|\phi_{\mathbf{p}}\rangle \quad (18)$$

Thus, substituting Eq. (16) in Eq. (15) for the continuum case we get on the left hand side

$$\begin{aligned} l.h.s. &= e^{-\frac{i}{\hbar}(\int^t (E_p + \frac{e^2A^2(t')}{2mc^2})dt' - \mathbf{a}(t)\cdot\mathbf{p})} \\ &\times (E_p + \frac{e^2A^2(t)}{2mc^2} - \dot{\mathbf{a}}(t)\cdot\mathbf{p})|\phi_{\mathbf{p}}\rangle \end{aligned} \quad (19)$$

and on the right hand side

$$r.h.s. = e^{-\frac{i}{\hbar}(\int^t (E_p + \frac{e^2A^2(t')}{2mc^2})dt' - \mathbf{a}(t)\cdot\mathbf{p})} \times ((\frac{\mathbf{p}_{op}{}^2}{2m} - \frac{Ze^2}{r}) + \frac{e^2A^2(t')}{2mc^2} - \frac{e}{mc}\mathbf{A}(t)\cdot\mathbf{p})|\phi_\mathbf{p}\rangle \tag{20}$$

Noting that $\dot{\mathbf{a}}(t) = \frac{e}{mc}\mathbf{A}(t)$ and $(\frac{\mathbf{p}_{op}{}^2}{2m} - \frac{Ze^2}{r})|\phi_\mathbf{p}\rangle = E_p|\phi_\mathbf{p}\rangle$, where, $E_p = \frac{p^2}{2m}$, one easily sees that the $l.h.s = r.h.s$ and hence the given solution is exactly fulfilled. In a similar way it is seen that,

$$\begin{aligned} T(\mathbf{h}_{op})|\phi_D\rangle &= |\phi_D\rangle - \sum_\mathbf{s} |\phi_\mathbf{s}\rangle(1 - e^{\frac{i}{\hbar}\mathbf{a}(t)\cdot\mathbf{s}})\langle\phi_\mathbf{s}|\phi_D\rangle \\ &= |\phi_D\rangle + 0 \end{aligned} \tag{21}$$

and,

$$\begin{aligned} -\frac{e}{mc}\mathbf{A}(t)\cdot\mathbf{h}_{op}|\phi_D\rangle &= -\frac{e}{mc}\mathbf{A}(t)\cdot\sum_\mathbf{s}|\phi_\mathbf{p}\rangle\mathbf{s}\langle\phi_\mathbf{s}|\phi_D\rangle \\ &= 0 \end{aligned} \tag{22}$$

since, the overlap integrals between the continuum and the discrete eigenstates of the Coulomb Hamiltonian vanish by orthogonality, $\langle\phi_\mathbf{s}|\phi_D\rangle = 0$. Hence, substituting Eq. (16) in Eq.(15) in the discrete case we get

$$l.h.s. = e^{-\frac{i}{\hbar}\int^t (E_D + \frac{e^2A^2(t')}{2mc^2}dt' + 0)} \times (E_D + \frac{e^2A^2(t)}{2mc^2} + 0)|\phi_D\rangle \tag{23}$$

and

$$r.h.s. = e^{-\frac{i}{\hbar}(\int^t (E_D + \frac{e^2A^2(t')}{2mc^2})dt' + 0)} \times ((\frac{\mathbf{p}_{op}{}^2}{2m} - \frac{Ze^2}{r}) + \frac{e^2A^2(t')}{2mc^2} + 0)|\phi_D\rangle \tag{24}$$

Also, $(\frac{\mathbf{p}_{op}^2}{2m} - \frac{Ze^2}{r})|\phi_D\rangle = E_D|\phi_D\rangle$, where $E_D \equiv E_{nlm} = -\frac{Z^2e^2}{2n^2a0}$ are the $((l,m)$-degenerate) eigen-energies of the Coulomb system. Hence, clearly, the $l.h.s = r.h.s$ in the discrete case as well, and the verification is complete. Thus, the complete set of solutions of the CV-Schroedinger equation defined by (15) is given by Eq.(16) or, more simply, by

$$\Phi_{j(\mathbf{p},D)}(\mathbf{r},t) = \phi_{j(\mathbf{p},D)}(\mathbf{r}) \times e^{-\frac{i}{\hbar}\int^t (\frac{p^2}{2m} + \frac{A(t')^2}{2mc^2} - (\frac{e}{c}\mathbf{A}(t')\cdot\mathbf{p})\delta_{\mathbf{p},j})dt'} \tag{25}$$

where for the continuum states $|\phi_\mathbf{p}\rangle$ are given by the Coulomb waves (6) and, for the discrete states $|\phi_D\rangle$, one has the well known bound states of the hydrogenic atom [18],

$$\begin{aligned} \phi_{D\equiv(nlm)}(\mathbf{r}) &= N_{nl}R_{nl}(r)Y_{lm}(\theta,\phi) \\ R_{nl}(r) &= (2\kappa_n r)^l e^{-\kappa_n r}{}_1F_1(-n+l+1, 2l+2, 2\kappa_n r) \\ N_{nl} &= \frac{(2\kappa_n)^{3/2}}{\Gamma(2l+2)}\sqrt{\frac{\Gamma(n+l+1)}{2n\Gamma(n-l)}} \end{aligned} \tag{26}$$

where $\kappa_n = \frac{Z}{na_0} = \sqrt{\frac{-2mE_D}{\hbar^2}}$. Having thus found the explicit form of both $H_{CV}(t)$, Eq. (14), and the complete set of solutions (25) of the Coulomb-Volkov Schroedinger equation (15), we can explicitly construct the solution of the Coulomb-Volkov propagator equation (10):

$$\begin{aligned} G_{CV}(t,t') = -\frac{i}{\hbar}\theta(t-t') &\times \{\sum_\mathbf{p}|\phi_\mathbf{p}\rangle e^{-\frac{i}{\hbar}\int_{t'}^t \frac{(\mathbf{p}-\frac{e}{c}\mathbf{A}(t''))^2}{2m}dt''}\langle\phi_\mathbf{p}| \\ &+ \sum_{nlm}|\phi_{nlm}\rangle e^{-\frac{i}{\hbar}\int_{t'}^t (E_{nl} + \frac{e^2A^2(t'')}{2mc^2})dt''}\langle\phi_{nlm}|\} \end{aligned} \tag{27}$$

Also, the final-state rest-interaction can be obtained self-consistently:

$$\begin{aligned} V_{CV}(t) &\equiv H(t) - H_{CV}(t) \\ &= (-\frac{e}{mc}\mathbf{A}(t)\cdot(\mathbf{p}_{op} - \mathbf{h}_{op}) + V_{s.r.}(\mathbf{r})) \end{aligned} \tag{28}$$

To determine the S-matrix amplitude of interest, we now substitute the initial and the final rest-interactions, V_i, and V_{CV} (Eqs. (4) and (28), respectively) as well as the final-state Coulomb-Volkov propagator (27), in the expression for the full wavefunction (8) and, project on to the Coulomb-Volkov state $\langle\Phi_\mathbf{p}(t)|$ of momentum $\mathbf{p}$. Thus, we get

$$\begin{aligned} S_{fi} &= \langle\Phi_\mathbf{p}(t)|\Psi(t)\rangle \\ &= \langle\Phi_\mathbf{p}(t)|\phi_i(t)\rangle + \int dt_1\langle\Phi_\mathbf{p}(t_1)|V_i(t_1)|\phi_i(t_1)\rangle \\ &+ \int dt_2dt_1\langle\Phi_\mathbf{p}(t_2)|V_{CV}(t_2)G(t_2,t_1)V_i(t_1)|\phi_i(t_1)\rangle \end{aligned} \tag{29}$$

To complete the derivation we expand, by iteration, the full propagator $G(t,t')$ appearing in between the initial and the final interactions in Eq. (29), in terms of the Volkov propagator G_{Vol} and the intermediate interaction $V_0(t)$, as follows:

$$\begin{aligned} G(t,t') &= G_{Vol}(t,t') + \int dt_1 G_{Vol}(t,t_1)V_0(t_1)G(t_1,t') \\ &= G_{Vol}(t,t') + \int dt_1 G_{Vol}(t,t_1)V_0(t_1)G_{Vol}(t_1,t') \\ &+ \int dt_2dt_1 G_{Vol}(t,t_2)V_0(t_2)G_{Vol}(t_2,t_1)V_0(t_1) \\ &\times G_{Vol}(t_1,t') + \cdots \end{aligned} \tag{30}$$

where (e.g. [17])

$$G_{Vol}(t,t') = -\frac{i}{\hbar}\theta(t-t')\sum_\mathbf{p}\frac{1}{L^3}|\mathbf{p}\rangle e^{-\frac{i}{\hbar}\int_{t'}^t \frac{p_\tau^2}{2m}d\tau}\langle\mathbf{p}| \tag{31}$$

and

$$\begin{aligned} V_0(t) &\equiv H(t) - H_{Vol}(t) \\ &= (-\frac{Ze^2}{r} + V_{s.r}(\mathbf{r})) \end{aligned} \tag{32}$$

Finally, collecting the resulting terms from Eq. (29) explicitly, we arrive at the desired all-order Coulomb-Volkov S-matrix series:

$$S_{fi} = \sum_{n=0}^{\infty} S_{fi}^{(n)} \tag{33}$$

$$S_{fi}^{(0)} = \langle \Phi_{\mathbf{p}}(\mathbf{r}, t_f)|\phi_i(\mathbf{r}, t_i)\rangle \tag{34}$$

$$\begin{aligned} S_{fi}^{(1)} = &-\frac{i}{\hbar}\int dt_1 \langle \Phi_{\mathbf{p}}(\mathbf{r}_1, t_1)| \\ &\times (-\frac{e}{mc}\mathbf{A}(t_1)\cdot \mathbf{p}_{op} + \frac{e^2A^2(t_1)}{2mc^2})|\phi_i(\mathbf{r}_1, t_1)\rangle \end{aligned} \tag{35}$$

$$\begin{aligned} S_{fi}^{(2)} = &-\frac{i}{\hbar}\int dt_2 dt_1 \langle \Phi_{\mathbf{p}}(\mathbf{r}_2, t_2)| \\ &\times (-\frac{e}{mc}\mathbf{A}(t_2)\cdot (\mathbf{p}_{op} - \mathbf{h}_{op}) + V_{s.r.}(\mathbf{r}_2)) \\ &\times G_{Vol}(\mathbf{r}_2, t_2; \mathbf{r}_1, t_1) \\ &\times (-\frac{e}{c}\mathbf{A}(t_1)\cdot \mathbf{p}_{op} + \frac{e^2A^2(t_1)}{2mc^2})|\phi_i(\mathbf{r}_1, t_1)\rangle \end{aligned} \tag{36}$$

and, for the general n-th order amplitude we get:

$$\begin{aligned} S_{fi}^{(n)} = &-\frac{i}{\hbar}\int dt_n dt_{n-1}\cdots dt_1 \langle \Phi_{\mathbf{p}}(\mathbf{r}_n, t_n)| \\ &\times (-\frac{e}{mc}\mathbf{A}(t_n)\cdot (\mathbf{p}_{op} - \mathbf{h}_{op}) + V_{s.r.}(\mathbf{r}_n)) \\ &\times G_{Vol}(\mathbf{r}_n, t_n; \mathbf{r}_{n-1}, t_{n-1})(-\frac{Ze^2}{r_{n-1}} + V_{s.r.}(\mathbf{r}_{n-1})) \\ &\times \cdots \times \\ &\times G_{Vol}(\mathbf{r}_2, t_2; \mathbf{r}_1, t_1)(-\frac{e}{mc}\mathbf{A}(t_1)\cdot \mathbf{p}_{op} + \frac{e^2A^2(t_1)}{2mc^2}) \\ &\times |\phi_i(\mathbf{r}_1, t_1)\rangle \end{aligned} \tag{37}$$

$n = 3, 4, \cdots \infty$. The angle brackets above stand for the integration with respect to the space coordinates, and the symbol $\int$ stands for *all* time integrations in the same range, from the initial time t_i to the final time t_f. We note that the Heaviside-functions of the propagators automatically control the appropriate intermediate time intervals.

It should be noted that the S-matrix series derived above, like most other well-known (perturbative/iterative) S-matrix series, is not proven by us to be convergent or otherwise. This, therefore, is an open mathematical question for the future. At present it should be understood as a *systematic* tool of theoretical investigation in strong-field physics in the same way as most other S-matrix series e.g. the various "Born series" of collision physics (in nuclear, atomic or molecular physics) or the usual SFA (in strong-field physics) have been used for decades usefully (in the absence of a proof of their convergence) i.e. by testing case by case against experimental data and/or alternative theoretical results (if and when available). One may surmise that the most well-known perturbative/iterative S-matrix series perhaps constitute (as they tend to behave like) a class of "asymptotic series" that can provide good estimates of the series sum from a finite number of terms, even when they may not be convergent in the standard sense (cf. Pade', Borel, or Shank's generalised summation of asymptotic series, e.g. [19]).

We next point out briefly how the method presented above can be extended, if desired, to take into account the *combined* effect of the Coulomb *and* the short-range potentials in the final state directly, rather than treating the $V_{s.r.}(r)$ in successive terms of the series, as above. This also automatically ensures any desired orthogonality between the initial and final states. This extension requires a predetermined effectively complete orthogonal set of (bound and continuum) eigenstates of the *unperturbed* target atom (e.g. from an atomic "structure" calculation). To be specific we consider again an active one-electron atom modelled by a spherically symmetric atomic potential $V_{at}(r)$. The bound states, $\phi_{nlm}(\mathbf{r})$, are then be of the general form

$$\phi_{nlm}(\mathbf{r}) = R_{nl}(r)Y_{lm}(\theta, \phi) \tag{38}$$

The continuum state $\phi_{\mathbf{p}}(\mathbf{r})$ of momentum $\mathbf{p}$ (orthogonal to the bound states) can be constructed as follows:

$$\phi_{\mathbf{p}}(\mathbf{r}) = 4\pi \sum_{lm} (i)^l e^{-i\delta_l(p)} R_{pl}(r) Y_{lm}(\theta, \phi) Y_{lm}^*(\theta_p, \phi_p) \tag{39}$$

where $\delta_l(p)$ is the lth partial wave total phase-shift (sum of Coulomb and short-range) defined by the asymptotic form of the radial waves,

$$r \to \infty, R_{pl}(r) \to \frac{\sin\left(kr - l\pi/2 + \eta_p ln(2pr) + \delta_l(p)\right)}{kr} \tag{40}$$

The operator $\mathbf{h}_{op}$ is then defined by the same Eq. (11) as before except that, $\phi_{\mathbf{p}}(\mathbf{r})$ is now given by Eq. (39). The reference Hamiltonian also takes the same form as Eq. (14) except that, the Coulomb potential $-\frac{Z}{r}$ is replaced by the full potential $V_{at}(r)$. The associated reference propagator also has the same form as Eq. (27) before, except that the radial bound states $R_{nl}(r)$ and the radial continuum momentum states of momentum $\mathbf{p}$, are now defined by Eq. (38) and Eq. (39), respectively. With the help of the above quantities, the desired S-matrix series can be constructed exactly in the same way as before and the final result takes the same form as the CV S-matrix series, Eqs. (33)-(37), with the interaction terms appearing in them replaced simply as follow:

$$\begin{aligned} &(-\frac{e}{mc}\mathbf{A}(t_n)\cdot (\mathbf{p}_{op} - \mathbf{h}_{op}) + V_{s.r.}(\mathbf{r}_n)) \\ &\to -\frac{e}{mc}\mathbf{A}(t_n)\cdot (\mathbf{p}_{op} - \mathbf{h}_{op}) \end{aligned} \tag{41}$$

and

$$(-\frac{Ze^2}{r_{n-1}} + V_{s.r.}(\mathbf{r}_{n-1})) \to V_{at}(r_{n-1}) \quad (42)$$

We next consider the interesting limiting case of the S-matrix series for a system without the long-range Coulomb interaction (e.g. interaction of the outer electron with the neutral core of a negative ion). This is easily found by simply putting $Z = 0$ in the CV series, Eqs. (33)-(37), and noting that now the Coulomb wave $\langle\phi_{\mathbf{p}}|$ simplifies to the plane wave $\langle\mathbf{p}|$, and, hence, the Coulomb-Volkov state $\Phi_{\mathbf{p}}(t)$ (Eq. (5)) reduces to the plane wave Volkov state $\langle\psi_{\mathbf{p}}|$, and V_0, defined in Eq. (32), reduces to $V_{s.r.}$. Also, the final-state rest-interaction simplifies to the short range potential $V_{s.r.}$ only, since in this limit

$$\begin{aligned}&\langle\phi_{\mathbf{p}}|(-\frac{e}{mc}\mathbf{A}(t)\cdot(\mathbf{p}_{op} - \mathbf{h}_{op}) + V_{s.r.})\\ \to &(-\frac{e}{mc}\mathbf{A}(t)\cdot(\mathbf{p} - \mathbf{p})\langle\mathbf{p}| + \langle\mathbf{p}|V_{s.r.}\\ = &\langle\mathbf{p}|V_{s.r.}\end{aligned} \quad (43)$$

Hence, the Coulomb-Volkov S-matrix series, Eq. (33), goes over to the reduced series:

$$\begin{aligned}S_{fi}^{[Z\equiv 0]} &= \langle\psi_{\mathbf{p}}(t_f)|\phi_i(t_i)\rangle\\ &+ (-\frac{i}{\hbar})\int dt_1\langle\psi_{\mathbf{p}}(\mathbf{r}_1,t_1)|V_i(\mathbf{r}_1,t_1)|\phi_i(\mathbf{r}_1,t_1)\rangle\\ &+ (-\frac{i}{\hbar})\int dt_2 dt_1\langle\psi_{\mathbf{p}}(\mathbf{r}_2,t_2)|V_{s.r.}(\mathbf{r}_2)\\ &\times G_{Vol}(\mathbf{r}_2,t_2;\mathbf{r}_1,t_1)V_i(\mathbf{r}_1,t_1)|\phi_i(\mathbf{r}_1,t_1)\rangle\\ &\ldots\ldots\\ &+ (-\frac{i}{\hbar})\int dt_n dt_{n-1}\cdots dt_1\langle\psi_{\mathbf{p}}(\mathbf{r}_n,t_n)|\\ &\times V_{s.r.}(\mathbf{r}_n)G_{Vol}(\mathbf{r}_n,t_n;\mathbf{r}_{n-1},t_{n-1})\\ &\times V_{s.r.}(\mathbf{r}_{n-1})\\ &\times \cdots\times G_{Vol}(\mathbf{r}_2,t_2;\mathbf{r}_1,t_1)\\ &\times V_i(\mathbf{r}_1,t_1)|\phi_i(\mathbf{r}_1,t_1)\rangle\end{aligned} \quad (44)$$

$n = 3, 4, \cdots\infty$. We point out that (44) has the same form as the usual SFA but its potential dependence is through $V_{s.r.}$ only. This is self-consistent with the absence of the Coulomb interaction also in the final state.

In this work, for the sake of concreteness, we have explicitly derived our main result – Eq. (33)-(37) – in the so-called "velocity" gauge. In an exactly analogous manner, or by a gauge transformation, one can obtain the corresponding result in the "length" gauge. We may, therefore, simply quote the final result of the Coulomb-Volkov S-matrix series in the "length" gauge (indicated by the superscript L) below:

$$S_{fi}^{(L)} = \sum_{n=0}^{\infty} S_{fi}^{(L;n)} \quad (45)$$

$$S_{fi}^{(L;0)} = \langle\Phi_{\mathbf{p}}^{(L)}(\mathbf{r},t_f)|\phi_i(\mathbf{r},t_i)\rangle \quad (46)$$

$$S_{fi}^{(L;1)} = -\frac{i}{\hbar}\int dt_1\langle\Phi_{\mathbf{p}}^{(L)}(\mathbf{r}_1,t_1)(-e\mathbf{F}(t_1)\cdot\mathbf{r}_1)|\phi_i(\mathbf{r}_1,t_1)\rangle \quad (47)$$

$$\begin{aligned}S_{fi}^{(L;2)} &= -\frac{i}{\hbar}\int dt_2 dt_1\langle\Phi_{\mathbf{p}}^{(L)}(\mathbf{r}_1,t_1)|\\ &\times(-\frac{e}{mc}\mathbf{A}(t_2)\cdot(\mathbf{p}_{op} + \frac{e\mathbf{A}(t_2)}{c} - \mathbf{h}_{op}^{(L)}) + V_{s.r.}(\mathbf{r}_2))\\ &\times G_{Vol}^{(L)}(\mathbf{r}_2,t_2;\mathbf{r}_1,t_1)(-e\mathbf{F}(t_1)\cdot\mathbf{r}_1)|\phi_i(\mathbf{r}_1,t_1)\rangle\end{aligned} \quad (48)$$

and, for the general nth order, $n = 3, 4, \cdots\infty$,

$$\begin{aligned}S_{fi}^{(L;n)} &= -\frac{i}{\hbar}\int dt_n dt_{n-1}\cdots dt_1\langle\Phi_{\mathbf{p}}^{(L)}(\mathbf{r}_n,t_n)|\\ &\times(-\frac{e}{mc}\mathbf{A}(t_n)\cdot(\mathbf{p}_{op} + \frac{e\mathbf{A}(t_2)}{c} - \mathbf{h}_{op}^{(L)}) + V_{s.r.}(\mathbf{r}_n))\\ &\times G_{Vol}^{(L)}(\mathbf{r}_n,t_n;\mathbf{r}_{n-1},t_{n-1})(-\frac{Ze^2}{r_{n-1}} + V_{s.r.}(\mathbf{r}_{n-1}))\\ &\times\cdots\times\\ &\times G_{Vol}^{(L)}(\mathbf{r}_2,t_2;\mathbf{r}_1,t_1)(-e\mathbf{F}(t_1)\cdot\mathbf{r}_1)|\phi_i(\mathbf{r}_1,t_1)\rangle\end{aligned} \quad (49)$$

Before concluding, a few additional observations are in order:
(a) The first order term in the present S-matrix series (Eq. (35) or Eq. (47)) reproduces the heuristic expression introduced a long time ago (e.g. [12]) and justifies it as a lowest order contribution. It is worth noting here that for very short interaction times the contribution of the first order term alone gives good agreement for ionization rates and angular distributions of the ejected electrons as shown for H atom in [20] by comparison with the exact numerical simulation (in length gauge), for laser pulses lasting *less* than two cycles.

(b) Beginning with the second order term the present theory opens up the possibility of systematic investigations of the role of final-state Coulomb interaction in "re-scattering" processes in a wide range of strong-field phenomena including the observation of low and very low energy structures [21–23], or the so-called zero-energy structure (ZES) [24]. Despite recent progress in their understanding (cf. e.g. [25–28]) they remain to be fully understood. Thus, for example, the specific role played by the asymptotically long-range (Coulomb) and the short-range atomic potentials in their formation, their actual numbers, the "threshold law" of strong-field ionization process etc. are apparently yet to be fully understood.

(c) We point out that the terms of the S-matrix series (33), for example, the amplitudes $S_{if}^{(1)}$ and $S_{if}^{(2)}$, can be evaluated efficiently by a combination of stationary phase method and/or numerical integration, provided the coordinates dependent integrals with the Coulomb-Volkov state can be evaluated analytically. They are of the form

$$M^{(1)}_{\mathbf{p},i} = \int \phi^{(-)*}_{\mathbf{p}}(\mathbf{r})(-\frac{e}{mc}\mathbf{A}(t)\cdot\mathbf{p}_{op})e^{-\kappa r}d^3r \quad (50)$$

where $\eta(p) \equiv \frac{Z\hbar}{pa0}$, $a0 = \hbar^2/me^2$. and

$$M^{(2)}_{\mathbf{p},\mathbf{k}} = \int \phi^{(-)*}_{\mathbf{p}}(\mathbf{r})(-\frac{e}{mc}\mathbf{A}(t)\cdot(\mathbf{p}_{op}-\mathbf{h}_{op})e^{i\mathbf{k}\cdot\mathbf{r}}d^3r$$
$$= (-\frac{e}{mc}\mathbf{A}(t)\cdot(\mathbf{k}-\mathbf{p}))\int \phi^{(-)*}_{\mathbf{p}}(\mathbf{r})e^{i\mathbf{k}\cdot\mathbf{r}}d^3r \quad (51)$$

The coordinate integrals in the 3rd and higher order terms can also be obtained by parametric differentiation from them. We give the necessary analytical results for the two prototypical integrals, evaluated analytically by Nordsieck's method [29], explicitly below (in a.u., $|e| = m = \hbar = \alpha c = 1$):

$$I_1 = \int e^{-i\mathbf{s}\cdot\mathbf{r}}{}_1F_1(i\eta_s, 1, i(sr+\mathbf{s}\cdot\mathbf{r}))(\epsilon\cdot\mathbf{p}_{op})e^{-\kappa r}d^3r$$
$$= 8\pi\kappa(1+i\eta_s)(\epsilon\cdot\mathbf{s})\frac{(\kappa-is)^{(2+i\eta_s)}}{(\kappa+is)^{(2-i\eta_s)}} \quad (52)$$

$$I_2 = lim_{\lambda\to 0}\int e^{-i\mathbf{s}\cdot\mathbf{r}}{}_1F_1(i\eta_s, 1, i(sr+\mathbf{s}\cdot\mathbf{r})$$
$$\times\, \epsilon\cdot(\mathbf{p}_{op}-\mathbf{h}_{op})e^{i\mathbf{k}\cdot\mathbf{r}}e^{-\lambda r}d^3r$$
$$= (\epsilon\cdot\mathbf{q})\times\frac{8\pi s\eta_s}{q^2(q^2+2\mathbf{q}\cdot\mathbf{s})}(\frac{q^2}{q^2+2\mathbf{q}\cdot\mathbf{s}})^{i\eta_s} \quad (53)$$

where, $\mathbf{q} \equiv \mathbf{k} - \mathbf{s}$, $\eta_s = \frac{Z}{s}$, and ϵ stands for the unit polarisation vector. The additional integration over the intermediate momentum $\mathbf{k}$ can be performed e.g. by the stationary phase method (or otherwise), and the first time-integration can be done either analytically or by the stationary phase method, while the additional time-integration can be done e.g. numerically. (Calculations are in currently in progress for a problem of much current interest namely, threshold behavior of ionization in intense long-wavelength laser fields. They are envisaged to be reported in a subsequent paper.)

(d) The explicit expression of the Coulomb-Volkov propagator derived here (Eq. (27)) suggests that the theory can be used also to investigate the role of strong-field excitation processes involving the *discrete* states, either as a final state or as intermediate/doorway states or both. For example, the theory could be used to analyse the mechanisms of "frustrated ionization" observed some time ago [30] for near-infrared wavelengths and of "ionization reduction" (e.g [23]) observed more recently for mid-infrared wavelengths, at very low electron energy.

(e) Finally, we point out that the (differential) probability of ionization, $d^3P_{fi}(\mathbf{p})$, by an ultra-short pulse of duration $\tau_d = t_f - t_i$, can be obtained directly from the absolute square of the S-matrix amplitude:

$$d^3P_{fi}(\mathbf{p}) = |S_{fi}|^2 d^3p \quad (54)$$

For a long pulse (with an effectively constant amplitude), on the other hand, it is useful to Fourier transform the periodic part of the S-matrix amplitude and rewrite it as:

$$S_{fi} = -\frac{i}{\hbar}\sum_{n=-\infty}^{\infty}\int_{t_i}^{t_f} dte^{\frac{i}{\hbar}(\frac{p^2}{2m}+Up+|E_i|-n\hbar\omega)t}T^{(n)}_{fi}(\mathbf{p}) \quad (55)$$

In this case the quantity of interest is the ionization *rate* (or, the probability of ionization per unit interaction time) which can be then determined from the generalised Fermi golden rule, in terms of the Fourier components $T^{(n)}_{fi}(\mathbf{p})$:

$$d^3R_{fi}(\mathbf{p}) = \sum_{n=n_0}^{\infty}\sum_{\mathbf{p}}\frac{2\pi}{\hbar}|T^{(n)}_{fi}(\mathbf{p})|^2$$
$$\times\, \delta(\frac{p^2}{2m}+U_p+|E_i|-n\hbar\omega)d^3p \quad (56)$$

where $n_0 = [\frac{\frac{p^2}{2m}+U_p+|E_i|)}{\hbar\omega}]_{int.}+1$, $|E_i|$ is the initial binding energy, $U_p = \frac{e^2F^2}{4m\omega^2}$ is the ponderomotive energy, F is the peak field strength and ω is the laser frequency.

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