Chapter 14 Theoretical Aspects of Laser-Assisted *(e,* **2***e)* **Collisions in Atoms**

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Abstract An overview of theoretical approaches to ionization of atomic systems by electron impact in the presence of laser radiation is given. Basic approximations for calculating multiphoton (*e*, 2*e*) transition amplitudes are discussed, with special emphasis on the first Born approximation in the projectile-target interaction. Various methods for the treatment of the dressing of initial and final (ionized) atomic-target states by a laser field are brought into focus, ranging respectively from the Floquet theory to the time-dependent perturbation theory and two-level approximation and from the Coulomb-Volkov models to the Sturmian-Floquet approach.

14.1 Introduction

The advance in laser technologies stimulates laser applications in various fields of atomic physics $[1-3]$ $[1-3]$. One of such promising developments is laser-assisted elastic and inelastic electron-atom scattering [\[4](#page-15-2)], which also includes an ionization channel, namely, the $(e, 2e)$ ionization of atoms in the presence of laser radiation. The latter was measured for the first time by Höhr et al. [\[5,](#page-15-3) [6\]](#page-15-4) in the case of a helium atomic target and a Nd:YAG laser beam ($\lambda = 1064$ nm) with intensity $I = 4 \times 10^{12}$ W/cm². While the pioneering experiment was performed not so long ago, theoretical investigations of laser-assisted (*e*, 2*e*) processes on atoms began much earlier, dating back to the 1970s (see, for instance, [\[7\]](#page-15-5)). A number of results concerning the dependence of the multiphoton (*e*, 2*e*) cross sections on laser-field parameters, such as polarization, frequency and intensity, have been obtained (see [\[4\]](#page-15-2) for a review of some earlier works and also more recent articles $[8-18]$ $[8-18]$). The main theoretical findings can be briefly summarized as follows: (i) the cross sections are seriously modified even by the presence of low-intensity laser radiation and (ii) they strongly depend on the dressing of the atomic target states. The aforementioned experiment of Höhr et al. [\[5,](#page-15-3) [6\]](#page-15-4) confirmed the existence of distinct differences in the (*e*, 2*e*) differential cross sections

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between laser-on and laser-off conditions. This indicates the importance of further developing the theory of laser-assisted (*e*, 2*e*) collisions for future experiments.

The present contribution aims at outlining a general formalism along with the main methods and approximations, which are typically employed in the theoretical treatment of laser-assisted $(e, 2e)$ collisions in atoms. Atomic units (a.u., $\hbar = e$) *me*) are used throughout unless otherwise specified.

14.2 General Formulation

We consider the process

$$
e^- + A + \ell \omega \to 2e^- + A^+, \tag{14.1}
$$

where, in the presence of a laser field, an electron impinges on an atomic target A and induces an ionizing collision, in which a net number ℓ of photons with frequency ω is exchanged between the colliding system and the field. As a result, an outgoing electron pair emerges which is formed by the scattered and ejected electrons. In what follows, the incident, scattered, and ejected electron energies and momenta are specified respectively by (E_0, \mathbf{p}_0) , (E_s, \mathbf{p}_s) , and (E_e, \mathbf{p}_e) .

The laser field is assumed to switch on and off adiabatically at $t \to -\infty$ and $t \to +\infty$, respectively. More specifically, the turn on and off time δT of the laser pulse as well as the laser pulse duration *T* are very long on a time scale typical for the target and much longer than the (*e*, 2*e*) collision duration. We consider the case of a monochromatic elliptically polarized laser wave with a wave vector **k** $(k = \omega/c)$. The frequency ω and intensity *I* of the wave are such that the laser electric-field amplitude F_0 is much less than the typical intra-atomic field F_A and the Keldysh parameter [\[19\]](#page-15-8) is $\gamma = \omega F_A/(2E_I F_0) \gg 1$, where E_I is the atomic ionization energy. This means that the ionization due to the laser electric field occurs in the perturbative regime via multiphoton transitions and hence it does not produce any appreciable effect compared to that due to the electron-atom collision. Without loss of generality we suppose that the z axis is directed along k . A typical situation is when the laser wavelength $\lambda = 2\pi/k$ is much greater than the spatial extent both of the target and of the region where the electron-electron collision takes place. This validates the use of the dipole approximation for the electric component of the laser field:

$$
\mathbf{F}(t) = F_x \mathbf{e}_x \cos \omega t + F_y \mathbf{e}_y \sin \omega t, \qquad (14.2)
$$

where $F_x > 0$ and $F_y > 0$ ($F_y < 0$) for right (left) polarization. The vector potential corresponding to [\(14.2\)](#page-1-0) is

$$
\mathbf{A}(t) = A_x \mathbf{e}_x \sin \omega t + A_y \mathbf{e}_y \cos \omega t, \qquad (14.3)
$$

with $A_x = -cF_x/\omega$, $A_y = cF_y/\omega$. Note that the case of linear polarization derives from [\(14.2\)](#page-1-0) upon setting $F_x = F_0$ and $F_y = 0$, while that of circular polarization amounts to $F_x = |F_y| = F_0 / \sqrt{2}$.

14.2.1 S Matrix

The rate of the discussed laser-assisted $(e, 2e)$ reaction is governed by the matrix element of the scattering operator, which is usually called the *S* operator. Using the Furry representation [\[20](#page-15-9)], in which the effect of an external field is included in the asymptotic Hamiltonians of the initial and final channels of the reaction, the *S* matrix can be presented as

$$
S_{(e,2e)} = -i \int_{-\infty}^{\infty} dt \left\langle \Psi_f^{(-)}(\mathbf{p}_s, \mathbf{p}_e; t) | V_{eA} | \chi_{\mathbf{p}_0}(t) \Phi_i(t) \right\rangle, \tag{14.4}
$$

where V_{eA} is the projectile-atom potential, $\Phi_i(t)$ is the laser-dressed initial atomic state, and $\Psi_f^{(-)}(\mathbf{p}_s, \mathbf{p}_e; t)$ is the final (time-reversed) scattering state of the colliding system in the presence of the laser field. The incident electron state $\chi_{p_0}(t)$ is given by the Gordon-Volkov function, which solves the following Schrödinger equation:

$$
i\frac{\partial}{\partial t}\chi_{\mathbf{p}_0}(\mathbf{r},t) = \frac{1}{2}\left[\hat{\mathbf{p}} + \frac{1}{c}\mathbf{A}(t)\right]^2 \chi_{\mathbf{p}_0}(\mathbf{r},t).
$$
 (14.5)

For the vector potential given by [\(14.3\)](#page-1-1) one has (see, for instance, [\[2\]](#page-15-10))

$$
\chi_{\mathbf{p}_0}(\mathbf{r}, t) = \exp\left\{i\left[\mathbf{p}_0 \mathbf{r} - \alpha_{\mathbf{p}_0} \sin(\omega t + \delta_{\mathbf{p}_0}) - E_0 t - \zeta(t)\right]\right\},\tag{14.6}
$$

where $E_0 = p_0^2/2$ and

$$
\alpha_{\mathbf{p}_0} = \frac{\sqrt{F_x^2 p_{0,x}^2 + F_y^2 p_{0,y}^2}}{\omega^2}, \qquad \delta_{\mathbf{p}_0} = \arcsin\left(\frac{F_x p_{0,x}}{\sqrt{F_x^2 p_{0,x}^2 + F_y^2 p_{0,y}^2}}\right),
$$

$$
\zeta(t) = \frac{1}{2c^2} \int_{-\infty}^t A^2(t') dt'.
$$

By definition, the *S* matrix remains invariant under unitary transformations. Therefore, [\(14.4\)](#page-2-0) is gauge-invariant, since the gauge transformation of the vector and scalar potentials of the laser field is equivalent to the unitary transformation in quantum mechanics (see [\[21\]](#page-15-11) for detail).

14.2.2 Cross Sections

It can be shown that, after integrating over time in [\(14.4\)](#page-2-0), the *S* matrix has the general form

$$
S_{(e,2e)} = -2\pi i \sum_{\ell=-\infty}^{\infty} T_{fi}^{(\ell)} \delta(E_0 + \mathscr{E}_i - \mathscr{E}_f - E_s - E_e - U_p + \ell \omega), \qquad (14.7)
$$

where \mathcal{E}_i and \mathcal{E}_f are the quasienergies (see below) of the laser-dressed initial atomic and final ionic states, respectively, $U_p = F_0^2 / 4\omega^2$ is a ponderomotive potential, and $T_{fi}^{(\ell)}$ are the ℓ -photon transition amplitudes. For the fully differential cross section (FDCS), which provides the most detailed information about the scattering process, one thus has

$$
\frac{\mathrm{d}^4\sigma}{\mathrm{d}E_s\mathrm{d}\Omega_s\mathrm{d}\Omega_e} = \sum_{\ell=-\infty}^{\infty} \frac{\mathrm{d}^3\sigma^{(\ell)}}{\mathrm{d}E_e\mathrm{d}\Omega_s\mathrm{d}\Omega_e} \,\delta(E_0 + \mathscr{E}_i - \mathscr{E}_f - E_s - E_e - U_p + \ell\omega),\tag{14.8}
$$

where the ℓ -photon triple differential cross section (TDCS) is

$$
\frac{\mathrm{d}^3 \sigma^{(\ell)}}{\mathrm{d}E_e \mathrm{d}\Omega_s \mathrm{d}\Omega_e} = \frac{p_s p_e}{(2\pi)^5 p_0} |T_{fi}^{(\ell)}|^2. \tag{14.9}
$$

14.3 Theoretical Methods and Approximations

For calculating the *S* matrix and cross sections one has to know the two states: (i) the initial laser-dressed atomic state $\Phi_i(t)$ and (ii) the final scattering state $\Psi_f^{(-)}(\mathbf{p}_s, \mathbf{p}_e; t)$ of the colliding system in the presence of the laser field. The first is the solution of the time-dependent Schrödinger equation (TDSE) for an atom in the laser field, and the second solves the TDSE for the interacting projectile-target system in the laser field and obeys the proper asymptotic behavior (when $t \to \infty$ and relative positions of the final ion and two outgoing electrons tend to infinity). Since both Hamiltonians are periodic in time, the TDSE can be solved employing the Floquet theory [\[22\]](#page-15-12).

14.3.1 Initial Laser-Dressed Atomic State

In the Floquet theory, one seeks the solution to the TDSE for the initial state,

$$
i\frac{\partial}{\partial t}|\Phi_i(t)\rangle = [H_A + H_{int}(t)]|\Phi_i(t)\rangle,
$$
\n(14.10)

where H_A is the field-free atomic Hamiltonian and $H_{int}(t)$ is the atom-field interaction Hamiltonian, in the form of the following (Floquet-Fourier) expansion:

$$
|\Phi_i(t)\rangle = e^{-i\mathcal{E}_i t} \sum_{n=-\infty}^{\infty} e^{-in\omega t} |\Phi_i^{(n)}(\mathcal{E}_i)\rangle.
$$
 (14.11)

The Floquet-Fourier components $|\Phi_i^{(n)}(\mathscr{E}_i)\rangle$ satisfy the system of coupled timeindependent equations

$$
(H_{\mathcal{A}} - n\omega - \mathscr{E}_i)|\Phi_i^{(n)}(\mathscr{E}_i)\rangle + \sum_{k=-\infty}^{\infty} (H_{\text{int}})_{n-k}|\Phi_i^{(k)}(\mathscr{E}_i)\rangle = 0, \qquad n = 0, \pm 1, \pm 2, \dots
$$
\n(14.12)

Here $(H_{int})_{n-k}$ are the components of the Fourier expansion

$$
H_{\text{int}}(t) = \sum_{n = -\infty}^{\infty} e^{-in\omega t} (H_{\text{int}})_n.
$$
 (14.13)

It can be seen that the solution of the system (14.12) does not define the quasienergy \mathcal{E}_i uniquely, since the latter can be changed to $\mathcal{E}_i + m\omega$, where *m* is an arbitrary integer. The customary way of defining the quasienergy consists in using the boundary condition at $t \to -\infty$,

$$
|\Phi_i(t \to -\infty)\rangle \to e^{-iE_A^{(i)}t}|\Phi_A^{(i)}\rangle, \tag{14.14}
$$

where $E_A^{(i)}$ is the energy of the field-free atomic state $|\Phi_A^{(i)}\rangle$, and requiring that only the $n = 0$ component of the expansion (14.11) remains nonvanishing in the limit $t \rightarrow -\infty$.

14.3.1.1 Time-Dependent Perturbation Theory

If the laser field is nonresonant with atomic transitions, the laser-atom interaction $H_{\text{int}}(t)$ appears to be weak due to the imposed condition $F_0 \ll F_A$ and, hence, can be treated as a perturbation. It is efficient to develop the time-dependent perturbation theory in the length gauge (L-gauge), where

$$
H_{int}^{L}(t) = \mathbf{F}(t) \cdot \mathbf{R}, \qquad \mathbf{R} = \sum_{k=1}^{Z} \mathbf{r}_k,
$$
 (14.15)

with *Z* being the number of atomic electrons (or the nuclear charge) and \mathbf{r}_k being their positions. The choice of the L-gauge is based on the observation that over the atomic region the electron-laser interaction [\(14.15\)](#page-4-2) is much weaker than the intra-atomic potential experienced by electrons. Developing the time-dependent perturbation theory for

$$
|\Phi_i^{\mathcal{L}}(t)\rangle = \exp\left(\frac{i}{c}\mathbf{A}(t)\cdot\mathbf{R}\right)|\Phi_i(t)\rangle
$$
 (14.16)

to first order, one obtains

$$
|\Phi_i^{\mathrm{L}}(t)\rangle = e^{-iE_{\mathrm{A}}^{(i)}}t \left[|\Phi_{\mathrm{A}}^{(i)}\rangle - \frac{1}{2}\sum_{j\neq 0} \left(e^{i\omega t} \frac{M_{ji}^+}{\omega_{ji} + \omega} + e^{-i\omega t} \frac{M_{ji}^-}{\omega_{ji} - \omega}\right)|\Phi_{\mathrm{A}}^{(j)}\rangle\right],\tag{14.17}
$$

where $\omega_{ji} = E_A^{(j)} - E_A^{(i)}$ are the field-free atomic transition energies, and

$$
M_{ji}^{\pm} = \left\langle \Phi_{\mathbf{A}}^{(j)} | F_x R_x \mp i F_y R_y | \Phi_{\mathbf{A}}^{(i)} \right\rangle
$$

are the dipole transition matrix elements.

When $\omega \ll |\omega_{ii}|$ for all $j \neq i$ (the low-frequency regime), one can readily perform the *n* summation in [\(14.17\)](#page-5-0) using the low-frequency ($\omega_{ii} \pm \omega \approx \omega_{ii}$) and closure $(\omega_{ii} \approx \omega_{c1})$ approximations. This yields

$$
\Phi_i^{\mathcal{L}}(X,t) = e^{-iE_{\mathcal{A}}^{(i)}t} \left[1 - \frac{1}{\omega_{\mathcal{C}i}} \left(F_x R_x \cos \omega t + F_y R_y \sin \omega t \right) \right] \Phi_{\mathcal{A}}^{(i)}(X), \quad (14.18)
$$

where $X = \{r_1, r_2, \ldots, r_Z\}.$

14.3.1.2 Two-Level Approximation

When the laser frequency ω is close to or coincides with a particular atomic transition energy ω_{ii} , the perturbation theory [\(14.17\)](#page-5-0) is not applicable anymore. A more appropriate theoretical approach in such a case consists in using the two-level approximation¹

$$
|\Phi_i^{\mathcal{L}}(t)\rangle = e^{-iE_{\mathcal{A}}^{(i)}t}a_i(t)|\Phi_{\mathcal{A}}^{(i)}\rangle + e^{-iE_{\mathcal{A}}^{(j)}t}a_j(t)|\Phi_{\mathcal{A}}^{(j)}\rangle, \tag{14.19}
$$

where the coefficients $a_i(t)$ and $a_j(t)$ are determined by solving the TDSE [\(14.10\)](#page-3-0) in the L-gauge with ansatz (14.19) . The latter can be done using the so-called rotating wave approximation (RWA) [\[1](#page-15-0)] which neglects the fast oscillating terms $\alpha e^{\pm i(\omega_{ji}+\omega)t}$ and $\propto e^{\pm i\omega t}$ in comparison with the slow oscillating terms $\propto e^{\pm i(\omega_{ji}-\omega)t}$. This procedure yields

¹Here the field-free atomic states are assumed to be nondegenerate. Generalization to the case of degenerate states is straightforward (see, for instance, [\[13](#page-15-13)]).

$$
a_i^{\pm}(t) = \sqrt{\frac{|\Delta| + \Omega}{2\Omega}} \exp\left[-\frac{i}{2}(\Delta \mp \Omega)t\right],
$$

\n
$$
a_j^{\pm}(t) = \mp \frac{M_{ji}^-}{\sqrt{2\Omega(|\Delta| + \Omega)}} \exp\left[\frac{i}{2}(\Delta \pm \Omega)t\right].
$$
\n(14.20)

Here

$$
\Delta = \omega_{ji} - \omega, \qquad \Omega = \sqrt{\Delta^2 + \chi_{ji}^2} \tag{14.21}
$$

are the resonance detuning and the generalized Rabi frequency, respectively, while

$$
\chi_{ji}=|M_{ji}^-|
$$

stands for the Rabi frequency. Note that according to [\(14.20\)](#page-6-0) the following relations hold true:

$$
|a_i^{\pm}(t)|^2 + |a_j^{\pm}(t)|^2 = 1, \qquad |a_i^{\pm}(t)|^2 \ge \frac{1}{2}, \qquad |a_j^{\pm}(t)|^2 \le \frac{1}{2}.
$$
 (14.22)

The laser-dressed target state is thus given by

$$
\Phi_i^{\mathcal{L} \pm}(X, t) = \exp\left\{-i \left[E_A^{(i)}t + \frac{1}{2}(\Delta \mp \Omega)t\right]\right\}
$$

$$
\times \sqrt{\frac{|\Delta| + \Omega}{2\Omega}} \left[\Phi_A^{(i)}(X) \mp e^{-i\omega t} \frac{M_{ji}}{|\Delta| + \Omega} \Phi_A^{(j)}(X)\right].
$$
(14.23)

The target wave function evolves into $\Phi_i^{\mathsf{L}+}(X, t)$ or $\Phi_i^{\mathsf{L} -}(X, t)$ according to whether $\Delta > 0$ or $\Delta < 0$.

14.3.2 Final Laser-Dressed Scattering State

In general, finding the final state $\Psi_f^{(-)}(\mathbf{p}_s, \mathbf{p}_e; t)$ is a more difficult task than in the $\Phi_i(t)$ case. Apart from dealing with a system that has an additional interacting electron (i.e., the projectile electron), one faces the three-body scattering problem involving Coulomb-tail potentials in the presence of a laser field. Since the solution is not known already in the field-free case [\[23](#page-15-14)], one has to resort to approximate treatments.

14.3.2.1 Asymptotic Behavior

Due to specifics of scattering on long-range potentials, such as Coulomb-tail potentials, a nontrivial issue which arises in the case of the presence of a laser field consists in imposing a proper asymptotic condition on the solution of the TDSE for $\Psi_f^{(-)}(\mathbf{p}_s, \mathbf{p}_e; t)$ when r_s, r_e , and $r_{se} = |\mathbf{r}_s - \mathbf{r}_e| \to \infty$. This issue is convenient to address in the accelerated, or Kramers-Henneberger (KH), frame [\[24\]](#page-15-15):

$$
\Psi_f^{\text{KH}(-)}(\mathbf{p}_s, \mathbf{p}_e; X_0, t) = \exp\left[\mathbf{a}(t) \cdot \sum_{k=0}^{Z+1} \nabla_k + i(Z+1)\zeta(t)\right] \Psi_f^{(-)}(\mathbf{p}_s, \mathbf{p}_e; X_0, t),\tag{14.24}
$$

where $X_0 = {\mathbf{r}_0, X}$, and

$$
\mathbf{a}(t) = \frac{1}{c} \int_{-\infty}^{t} dt' \mathbf{A}(t') = a_x \mathbf{e}_x \cos \omega t + a_y \mathbf{e}_y \sin \omega t
$$
 (14.25)

is the displacement vector of a classical electron in the laser field, with $a_{x(y)} =$ $F_{x(y)}/\omega^2$. When working within the KH frame, a laser field is effectively absent, and the scattered and ejected electrons move in a Coulomb-tail potential of an ion $A⁺$ that oscillates in time (the oscillations are equivalent to those of a classical free electron in a laser field in the laboratory frame). At large distances from the ion, the role of its oscillating motion vanishes: the outgoing electrons experience therefore a usual, time-independent, Coulomb-tail force and, accordingly, the leading asymptotic behavior of the wave function [\(14.24\)](#page-7-0) is

$$
\left\langle \mathbf{r}_s, \mathbf{r}_e | \Psi_f^{\mathrm{KH}(-)}(\mathbf{p}_s, \mathbf{p}_e; t) \right\rangle \xrightarrow[r_s, r_e, r_{se} \to \infty]{} e^{i(\mathbf{p}_s \cdot \mathbf{r}_s - E_s t)} e^{i(\mathbf{p}_e \cdot \mathbf{r}_e - E_e t)} C_{\mathbf{p}_s}^{(-)}(\eta_s, \mathbf{r}_s)
$$
\n
$$
\times C_{\mathbf{p}_e}^{(-)}(\eta_e, \mathbf{r}_e) C_{\mathbf{p}_{se}}^{(-)}(\eta_{se}, \mathbf{r}_{se}) | \tilde{\Phi}_f^{\mathrm{KH}}(t) \rangle. \tag{14.26}
$$

Here the Coulomb-distortion factors are given by

$$
C_{\mathbf{p}}^{(-)}(\eta, \mathbf{r}) = \exp\left[-i\eta \ln(pr + \mathbf{p} \cdot \mathbf{r})\right],
$$

with the Sommerfeld parameters $\eta_{s(e)} = -1/p_{s(e)}$ and $\eta_{se} = 1/p_{se}$ ($p_{se} = |\mathbf{p}_s - \mathbf{p}_s|$) \mathbf{p}_e), and $\tilde{\Phi}_f^{\text{KH}}(t)$ is the laser-dressed ionic state satisfying the boundary condition

$$
|\tilde{\Phi}_f^{\text{KH}}(t \to \infty)\rangle \to e^{-iE_{\text{A}^+}^{(f)}t}|\tilde{\Phi}_{\text{A}^+}^{(f)}\rangle,\tag{14.27}
$$

where $E_{A^+}^{(f)}$ and $|\tilde{\Phi}_{A^+}^{(f)}\rangle$ are the field-free energy and state of the final ion.

14.3.2.2 3C-Volkov Wave Function

The approximate final-state wave function that accounts for the proper asymptotic behavior [\(14.26\)](#page-7-1) can be formulated on the basis of the 3C model [\[25](#page-15-16)], which proved to be useful in the theoretical treatment of field-free (*e*, 2*e*) collisions in atoms, and Coulomb-Volkov approximation [\[26\]](#page-15-17). In the KH frame, it reads [\[27](#page-15-18)]

$$
\left\langle \mathbf{r}_s, \mathbf{r}_e | \Psi_f^{\mathrm{KH}(-)}(\mathbf{p}_s, \mathbf{p}_e; t) \right\rangle = e^{-i(E_s + E_e)t} e^{-i(\mathbf{p}_s + \mathbf{p}_e) \cdot \mathbf{a}(t)} e^{-i\mathbf{p}_{se} \cdot \mathbf{r}_{se}} \psi_{\mathbf{p}_s}^{c(-)}(\eta_s, \mathbf{r}_s + \mathbf{a}(t)) \times \psi_{\mathbf{p}_e}^{c(-)}(\eta_e, \mathbf{r}_e + \mathbf{a}(t)) \psi_{\mathbf{p}_{se}}^{c(-)}(\eta_{se}, \mathbf{r}_{se}) | \tilde{\Phi}_f^{\mathrm{KH}}(t) \rangle, (14.28)
$$

where $\psi_{\mathbf{p}}^{c(-)}(\eta, \mathbf{r})$ stands for a stationary Coulomb wave function with incoming spherical wave behavior (see, for instance [\[28\]](#page-15-19)):

$$
\psi_{\mathbf{p}}^{c(-)}(\eta, \mathbf{r}) = e^{-\frac{1}{2}\pi\eta} \Gamma(1 - i\eta) e^{i\mathbf{p}\cdot\mathbf{r}} {}_{1}F_{1}(i\eta, 1; -i(\mathbf{p}\mathbf{r} + \mathbf{p}\mathbf{r})), \tag{14.29}
$$

where $_1F_1$ is the confluent hypergeometric function. In the field-free case, (14.28) reduces to the field-free 3C model [\[25](#page-15-16)]. It should be also noted that the laser-dressed final ionic state in [\(14.28\)](#page-8-0) can be calculated using the methods and approximations outlined above in regard to the laser-dressed initial atomic state.

14.4 First Born Approximation

One of the most frequently used approaches in the theory of (*e*, 2*e*) collisions is the first Born approximation (FBA), which treats the projectile-atom interaction V_{eA} in the *S* matrix [\(14.4\)](#page-2-0) only to first order. It is supposed to be generally applicable if both the incident and scattered electrons are fast. In the FBA, the laser-dressed final scattering state of the colliding system is approximated as

$$
|\Psi_f^{(-)}(\mathbf{p}_s, \mathbf{p}_e; t)\rangle = |\chi_{\mathbf{p}_s}(t)\Phi_f^{(-)}(\mathbf{p}_e; t)\rangle, \tag{14.30}
$$

where $|\Phi_f^{(-)}(\mathbf{p}_e; t)\rangle$ is the laser-dressed final atomic state with one (ejected) electron in continuum having the asymptotic momentum \mathbf{p}_e . Note that the final state [\(14.30\)](#page-8-1) can be derived from the 3C-Volkov model [\(14.28\)](#page-8-0) upon setting $\eta_s = \eta_{se} = 0$.

Using [\(14.30\)](#page-8-1) and the explicit form of the Gordon-Volkov functions [\(14.6\)](#page-2-1), one obtains for the *S* matrix the following expression:

$$
S_{(e,2e)}^{\text{FBA}} = -i \frac{4\pi}{Q^2} \int_{-\infty}^{\infty} dt \, e^{-i[\Delta E t + \alpha_Q \sin(\omega t + \delta_Q)]} \Bigg\langle \Phi_f^{(-)}(\mathbf{p}_e; t) | \sum_{k=1}^Z e^{i\mathbf{Q} \cdot \mathbf{r}_k} - Z | \Phi_i(t) \Bigg\rangle, \tag{14.31}
$$

where $\Delta E = E_0 - E_s$ and $\mathbf{Q} = \mathbf{p}_0 - \mathbf{p}_s$ are the energy and momentum transfers. Provided the momentum-transfer value is small, the contributions due to exchange between the projectile and atomic electrons are typically omitted in [\(14.31\)](#page-8-2) due to large p_0 and p_s values (on the atomic scale) in the case of fast incident and scattered electrons.

If neglecting the dressing of the initial and final atomic states by the laser field, one can readily perform the time integration in (14.31) using the following formula [\[29](#page-16-0)]:

$$
e^{iz\cos\xi} = \sum_{\ell=-\infty}^{\infty} J_{\ell}(z) e^{i\ell z},
$$

where J_{ℓ} are the Bessel functions of integer order. As a result, one obtains the ℓ photon TDCS [\(14.9\)](#page-3-1) in the form [\[7](#page-15-5)]

$$
\frac{\mathrm{d}^3 \sigma_{\text{FBA}}^{(\ell)}}{\mathrm{d}E_e \mathrm{d}\Omega_s \mathrm{d}\Omega_e} = |J_\ell(\alpha_\mathbf{Q})|^2 \frac{\mathrm{d}^3 \sigma_{\text{FBA}}}{\mathrm{d}E_e \mathrm{d}\Omega_s \mathrm{d}\Omega_e},\tag{14.32}
$$

where $d^3\sigma_{\text{FBA}}$ is the field-free TDCS in the FBA approach, where, however, the energy balance is

$$
E_0 + E_{\rm A}^{(i)} + \ell \omega = E_{\rm A^+}^{(f)} + E_s + E_e,
$$

i.e., it takes into account the transfer of ℓ photons in the laser-assisted (*e*, 2*e*) collision. From the properties of the Bessel functions it follows that the cross section [\(14.32\)](#page-9-0) turns to zero for all $\ell \neq 0$ if $\alpha_0 = 0$ or, in other words, if the momentum transfer is perpendicular to the laser polarization.

14.4.1 Laser-Dressed Final Atomic State

The asymptotic behavior of the state $\Phi_f^{(-)}(\mathbf{p}_e; t)$ is also convenient to formulate in the KH frame:

$$
\langle \mathbf{r}_e | \Phi_f^{\text{KH}(-)}(\mathbf{p}_e; t) \rangle \xrightarrow[r_e \to \infty]{} e^{i(\mathbf{p}_e \cdot \mathbf{r}_e - E_e t)} C_{\mathbf{p}_e}^{(-)}(\eta_e, \mathbf{r}_e) | \tilde{\Phi}_f^{\text{KH}}(t) \rangle. \tag{14.33}
$$

Below the approaches are described, in which the laser-dressed final atomic state is constructed to obey the proper asymptotic behavior.

14.4.1.1 Coulomb-Volkov Approximation

The condition [\(14.33\)](#page-9-1) can be fulfilled by neglecting correlations between the ejected and ionic electrons and using the Coulomb-Volkov wave function [\[26](#page-15-17)] for the laserdressed ejected electron state, namely,

$$
|\Phi_f^{\text{KH}(-)}(\mathbf{p}_e; t)\rangle = |\psi_{\text{CV}}^{\text{KH}(-)}(\mathbf{p}_e, \eta_e; t)\tilde{\Phi}_f^{\text{KH}}(t)\rangle, \tag{14.34}
$$

where the Coulomb-Volkov function in the KH frame is given by

$$
\psi_{\text{CV}}^{\text{KH}(-)}(\mathbf{p}_e, \eta_e; \mathbf{r}_e, t) = e^{-i[E_e t + \mathbf{p}_e \cdot \mathbf{a}(t)]} \psi_{\mathbf{p}_e}^{c(-)}(\eta_e, \mathbf{r}_e + \mathbf{a}(t)).
$$
\n(14.35)

If now using the approximation [\(14.34\)](#page-10-0) in the FBA *S*-matrix [\(14.31\)](#page-8-2) and neglecting the dressing of the initial atomic $\Phi_i(t)$ and final ionic $\Phi_f(t)$ states by the laser field, it is also possible to relate the multiphoton TDCS to the field-free FBA cross section. In particular, in the case of a circular polarized laser beam or when

$$
|\alpha_{\mathbf{Q}}| \gg \left| \frac{F_x^2 - F_y^2}{8\omega^3} \right|,\tag{14.36}
$$

one derives [\[30\]](#page-16-1)

$$
\frac{\mathrm{d}^3 \sigma_{\text{FBA}}^{(\ell)}}{\mathrm{d}E_e \mathrm{d}\Omega_s \mathrm{d}\Omega_e} = |J_\ell(\alpha_{\mathbf{q}_{\text{ion}}})|^2 \frac{\mathrm{d}^3 \tilde{\sigma}_{\text{FBA}}}{\mathrm{d}E_e \mathrm{d}\Omega_s \mathrm{d}\Omega_e},\tag{14.37}
$$

where the field-free FBA cross section $d^3 \tilde{\sigma}_{\text{FRA}}$ is calculated with the model field-free final atomic state

$$
|\phi_{\mathbf{A}}^{(f)}\rangle = |\psi_{\mathbf{p}_e}^{c(-)}\tilde{\phi}_{\mathbf{A}^+}^{(f)}\rangle
$$

and for the energy balance

$$
E_0 + E_{\rm A}^{(i)} + \ell \omega = E_{\rm A^+}^{(f)} + E_s + E_e + U_p,
$$

and $\mathbf{q}_{\text{ion}} = \mathbf{Q} - \mathbf{p}_e$ is the recoil-ion momentum. The cross section [\(14.37\)](#page-10-1) has a similar structure as that given by [\(14.32\)](#page-9-0). However, it turns to zero for all $\ell \neq 0$ if the laser polarization is perpendicular to the recoil-ion momentum q_{ion} rather than the momentum transfer **Q**.

The approximation [\(14.35\)](#page-10-2) can be further improved by taking into account the role of all unperturbed electron states in the dressing of the ejected-electron wave function by the laser field. This can be done within the first-order time-dependent perturbation theory as was initially proposed by Joachain et al. [\[31\]](#page-16-2). For atomic hydrogen and a linearly polarized laser beam such a modification of the Coulomb-Volkov wave function in the laboratory frame reads

$$
\psi_{\text{MCV}}^{(-)}(\mathbf{p}_e, \eta_e; \mathbf{r}_e, t) = e^{-i[E_e t + \mathbf{a}(t) \cdot \mathbf{r}_e + \alpha_{\mathbf{p}_e} \sin \omega t]} \left[(1 + i\alpha_{\mathbf{p}_e} \sin \omega t) \psi_{\mathbf{p}_e}^{c(-)}(\eta_e, \mathbf{r}_e) \right]
$$

$$
-i \frac{F_0}{2} \sum_j \left(\frac{e^{i\omega t}}{\omega_{j\mathbf{k}_e} + \omega} - \frac{e^{-i\omega t}}{\omega_{j\mathbf{k}_e} - \omega} \right) M_{j\mathbf{k}_e} \psi_j(\mathbf{r}_e) \right], (14.38)
$$

where

$$
M_{j\mathbf{k}_e} = -\langle \psi_j | x | \psi_{\mathbf{p}_e}^{c(-)}(\eta_e) \rangle
$$

are the dipole matrix elements, with ψ_j being the unperturbed hydrogen wave functions.

14.4.1.2 Sturmian-Floquet Approach

An alternative, nonperturbative method for calculating the laser-dressed ejectedelectron state consists in using the Hermitian Floquet theory and employing the basis set of Sturmian functions, which proved to be efficient in the treatment of the laser-assisted scattering processes [\[3](#page-15-1), [33](#page-16-3)[–36\]](#page-16-4). The dynamics of the ejected electron in the KH frame is governed by the following TDSE:

$$
i\frac{\partial}{\partial t}\,\psi_{\mathbf{p}_e}^{\mathrm{KH}(-)}(\mathbf{r}_e,t) = \left(-\frac{1}{2}\,\Delta + V[\mathbf{r}_e + \mathbf{a}(t)]\right)\psi_{\mathbf{p}_e}^{\mathrm{KH}(-)}(\mathbf{r}_e,t),\tag{14.39}
$$

where $V[r_e + a(t)]$ is a space-translated electron-ion potential, which has the Coulomb tail

$$
V[\mathbf{r}_e + \mathbf{a}(t)] \xrightarrow[r_e \to \infty]{} -\frac{1}{r_e}.
$$

Using the Floquet-Fourier expansion

$$
\psi_{\mathbf{p}_e}^{\mathrm{KH}(-)}(\mathbf{r}_e, t) = e^{-iE_e t} \sum_{n=-\infty}^{\infty} e^{-in\omega t} \mathcal{F}_n^{\mathrm{KH}(-)}(\mathbf{p}_e, \mathbf{r}_e), \tag{14.40}
$$

one arrives at an infinite set of coupled time-independent equations:

$$
(H_n + \widetilde{V}_0(a_x, a_y; \mathbf{r}_e) - E_e) \mathcal{F}_n^{\text{KH}(-)}(\mathbf{p}_e, \mathbf{r}_e)
$$

+
$$
\sum_{v \neq n} V_{n-v}(a_x, a_y; \mathbf{r}_e) \mathcal{F}_v^{\text{KH}(-)}(\mathbf{p}_e, \mathbf{r}_e) = 0, \qquad n = 0, \pm 1, \pm 2, \dots
$$
 (14.41)

Here

$$
H_n = H_C - n\omega, \qquad H_C = -\frac{1}{2}\,\Delta - \frac{1}{r_e}, \qquad \widetilde{V}_0(a_x, a_y; \mathbf{r}_e) = V_0(a_x, a_y; \mathbf{r}_e) + \frac{1}{r_e},
$$
\n(14.42)

and V_n are the Fourier components of $V[r_e + \mathbf{a}(t)],$

$$
V_n(a_x, a_y; \mathbf{r}_e) = \frac{1}{T} \int_0^T dt \exp(in\omega t) V[\mathbf{r}_e + \mathbf{a}(t)], \qquad (14.43)
$$

with $T = 2\pi/\omega$ being the optical cycle.

The solutions $\mathscr{F}_n^{\text{KH}(-)}$ must satisfy the incoming boundary conditions in the form [\[32\]](#page-16-5)

$$
\mathscr{F}_{n}^{\text{KH}(-)}(\mathbf{p}_{e}, \mathbf{r}_{e}) \xrightarrow[r_{e} \to \infty]{} \delta_{n0} \exp \left[i \mathbf{p}_{e} \cdot \mathbf{r}_{e} - i \eta_{e} \ln (p_{e} r_{e} + \mathbf{p}_{e} \cdot \mathbf{r}_{e}) \right] + f_{n}^{(-)}(p_{e}, \hat{\mathbf{r}}_{e}) \frac{\exp \left[-i p_{e}^{(n)} r_{e} + i \eta_{e}^{(n)} \ln (2 p_{e}^{(n)} r_{e}) \right]}{r_{e}}, \quad (14.44)
$$

where $\eta_e^{(n)} = -1/p_e^{(n)}$, and

$$
p_e^{(n)} = \begin{cases} \sqrt{2(E_e + n\omega)}, & E_e + n\omega \ge 0, \\ \pm i\sqrt{-2(E_e + n\omega)}, & E_e + n\omega < 0. \end{cases} \tag{14.45}
$$

The Coulomb-specific asymptotic behavior [\(14.44\)](#page-12-0) can be taken into account by recasting the system of Floquet equations into the Lippman–Schwinger form

$$
\mathscr{F}_{n}^{\text{KH}(-)}(\mathbf{p}_{e}, \mathbf{r}_{e}) = \delta_{n0} \psi_{\mathbf{p}_{e}}^{c(-)}(\eta_{e}, \mathbf{r}_{e}) \n+ \sum_{v} \int d\mathbf{r}_{e}^{\prime} G_{c}^{(-)}(p_{e}^{(n)}; \mathbf{r}_{e}, \mathbf{r}_{e}^{\prime}) \mathscr{V}_{n-v}(a_{x}, a_{y}; \mathbf{r}_{e}^{\prime}) \mathscr{F}_{v}^{\text{KH}(-)}(\mathbf{p}_{e}, \mathbf{r}_{e}^{\prime}), \nn = 0, \pm 1, \pm 2, ...
$$
\n(14.46)

Here the notation $\mathcal{V}_n = \delta_{n0} \tilde{V}_0 + (1 - \delta_{n0}) V_n$ is introduced, and $G_c^{(-)}$ is the advanced Coulomb Green's function, which satisfies the equation

$$
\left(\frac{p_e^{(n)2}}{2} + \frac{1}{2}\Delta + \frac{1}{r_e}\right) G_c^{(-)}\left(p_e^{(n)}; \mathbf{r}_e, \mathbf{r}'_e\right) = \delta(\mathbf{r}_e - \mathbf{r}'_e). \tag{14.47}
$$

The solution of the system of the Lippmann–Schwinger–Floquet equations [\(14.46\)](#page-12-1) can be sought using the expansion of the Floquet-Fourier components in terms of Sturmian functions. Recently, a new efficient method has been proposed [\[37\]](#page-16-6) based on employing the basis set of quasi-Sturmian functions [\[38](#page-16-7)] in parabolic coordinates. A marked advantage of the quasi-Sturmians is that they possess an appropriate incoming Coulomb asymptotic behavior, thus providing the proper asymptotic form [\(14.44\)](#page-12-0) of the solution.

14.4.2 Laser-Assisted Electron Momentum Spectroscopy

The $(e, 2e)$ reactions involving large momentum transfer under kinematical conditions close to a free electron-electron collision are usually referred to as electron momentum spectroscopy (EMS) [\[39](#page-16-8), [40\]](#page-16-9). EMS is a well-known method for explor-

ing the electronic structure of various systems ranging from atoms and molecules to clusters and solids.

The theoretical formulation of EMS in the presence of laser radiation was given in [\[13\]](#page-15-13). According to this formulation, the *S* matrix of the laser-assisted (*e*, 2*e*) EMS process can be presented as

$$
S_{(e,2e)}^{\text{EMS}} = -i \frac{4\pi}{Q^2} \int_{-\infty}^{\infty} dt \, \exp\left[-i\left(\varepsilon - \frac{q^2}{2}\right)t\right] \langle \chi_{\mathbf{q}}(t) | \varphi_{fi}(t) \rangle, \tag{14.48}
$$

where $\varepsilon = E_s + E_e - E_0$, $\mathbf{q} = -\mathbf{q}_{ion}$, and

$$
|\varphi_{fi}(t)\rangle = \langle \Phi_f(t)|\Phi_i(t)\rangle \tag{14.49}
$$

is the laser-dressed Kohn-Sham orbital. The expression [\(14.48\)](#page-13-0) follows from the FBA result [\(14.31\)](#page-8-2) using the binary-encounter approximation, which accounts only for the interaction between the colliding electrons in the projectile-atom potential *Ve*A, and the laser-dressed final atomic state in the form

$$
|\Phi_f^{(-)}(\mathbf{p}_e; t)\rangle = |\chi_{\mathbf{p}_e}(t)\tilde{\Phi}_f(t)\rangle.
$$
 (14.50)

The time integration in [\(14.48\)](#page-13-0) can be readily performed using the Floquet expansion of the laser-dressed Kohn-Sham orbital

$$
|\varphi_{fi}(t)\rangle = e^{-i\mathcal{E}_{fi}t} \sum_{n=-\infty}^{\infty} e^{-in\omega t} |\varphi_{fi}^{(n)}(\mathcal{E}_{fi})\rangle, \qquad (14.51)
$$

where \mathcal{E}_{fi} is the Kohn-Sham quasienergy.

If the laser-dressing effect in [\(14.51\)](#page-13-1) is negligible, then

$$
|\varphi_{fi}(t)\rangle = e^{-iE_{fi}t}|\varphi_{fi}\rangle, \qquad (14.52)
$$

where $E_{fi} = E_A^{(i)} - E_{A^+}^{(f)}$ and φ_{fi} are the field-free (unperturbed) Kohn-Sham energy and orbital. In such a case, provided the laser beam is circularly polarized or the inequality [\(14.36\)](#page-10-3) holds, one derives the ℓ -photon TDCS as (cf. [14.37\)](#page-10-1)

$$
\frac{d^3 \sigma_{\text{EMS}}^{(\ell)}}{dE_e d\Omega_s d\Omega_e} = |J_\ell(\alpha_\mathbf{q})|^2 \frac{d^3 \sigma_{\text{EMS}}}{dE_e d\Omega_s d\Omega_e},\tag{14.53}
$$

where $d^3\sigma_{\text{EMS}}$ is the field-free EMS cross section² [\[39\]](#page-16-8)

²The exchange between the colliding electrons is taken into account, for in the EMS kinematics both outgoing electrons (scattered and ejected) are fast.

$$
\frac{d^3 \sigma_{\text{EMS}}}{dE_e d\Omega_s d\Omega_e} = \frac{p_s p_e}{2\pi^3 p_0} \left(\frac{1}{|\mathbf{p}_0 - \mathbf{p}_s|^4} + \frac{1}{|\mathbf{p}_0 - \mathbf{p}_e|^4} - \frac{1}{|\mathbf{p}_0 - \mathbf{p}_s|^2 |\mathbf{p}_0 - \mathbf{p}_e|^2} \right) |\varphi_{fi}(\mathbf{q})|^2
$$
(14.54)

which is calculated for the energy balance

$$
E_{if} + \ell \omega = \varepsilon + U_p.
$$

If the laser-dressing effect in (14.51) is substantial, one can minimize the role of the interaction of the fast ingoing and outgoing electrons with the laser field by considering such laser-field orientations that $\alpha_q = 0$ [\[15\]](#page-15-20). Assuming again a circularly polarized laser beam or the validity of (14.36) , the resultant ℓ -photon TDCS is given by

$$
\frac{d^3 \sigma_{\text{EMS}}^{(\ell)}}{dE_e d\Omega_s d\Omega_e} = \frac{p_s p_e}{2\pi^3 p_0} \left(\frac{1}{|\mathbf{p}_0 - \mathbf{p}_s|^4} + \frac{1}{|\mathbf{p}_0 - \mathbf{p}_e|^4} - \frac{1}{|\mathbf{p}_0 - \mathbf{p}_s|^2 |\mathbf{p}_0 - \mathbf{p}_e|^2} \right) |\varphi_{fi}^{(\ell)}(\mathbf{q})|^2,
$$
\n(14.55)

and the energy balance is

$$
\mathscr{E}_{if} + \ell \omega = \varepsilon + U_p.
$$

Thus, the cross section [\(14.55\)](#page-14-0) contains the direct information about the ℓ th Floquet-Fourier component of the laser-dressed Kohn-Sham orbital in momentum space.

14.5 Concluding Remarks

In this work an account of the basic theoretical methods and approximations in the field of laser-assisted (*e*, 2*e*) collisions in atoms has been given. At the same time, a number of issues related to the theory of laser-assisted (*e*, 2*e*) processes inevitably remained beyond the scope of the present contribution. In particular, the high-order Born approximations, for example, such as the second Born approximation, has not been discussed here. The reason is that no theoretical study of the Born series in the laser-assisted (*e*, 2*e*) case has been carried out so far, except for the second-Born calculations performed in [\[14,](#page-15-21) [17](#page-15-22)]. Owing to the long-range Coulomb-tail potentials involved in the $(e, 2e)$ scattering processes, the higher Born approximations are known to diverge in the field-free case. There are theoretical methods allowing to cope with these divergences (see [\[41\]](#page-16-10) and references therein), but they are not directly applicable to the laser-assisted case. It should be noted that the authors of the works [\[14,](#page-15-21) [17](#page-15-22)] left the problem of divergences unaddressed, and therefore it still awaits a rigorous theoretical analysis.

Some comments should be made about testing the presented theoretical approaches. Currently the laser-assisted (*e*, 2*e*) measurements are lacking: only one experimental study [\[5\]](#page-15-3) has been conducted so far. Notable discrepancies were found [\[6\]](#page-15-4) between the experimental data and the FBA calculations using the initial laser-dressed atomic wave function in the form [\(14.18\)](#page-5-3) and the CoulombVolkov model [\(14.34\)](#page-10-0) for the final laser-dressed atomic state. The disagreement was attributed in [\[6](#page-15-4)] mainly to the deficiencies of the Coulomb-Volkov approximation, thus suggesting that more advanced non-perturbative treatments (for example, such as the *R*-matrix-Floquet theory [\[42\]](#page-16-11)) are needed. One of novel advanced non-perturbative approaches, namely, the quasi-Sturmian-Floquet approach [\[37](#page-16-6)], has been outlined above. It might be expected that the further progress in laser-assisted (*e*, 2*e*) experimental studies, including the improvement of energy and momentum resolutions and investigation of the atomic targets other than helium, will provide more stringent tests for the current theoretical understanding.

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