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A

Double Multiphotoionization of Helium

by

Jerry B. Altzman

A dissertation submitted to the Graduate Faculty in Physics
in partial fulfullment of the requirements for the degree of
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1997

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Date

Chair of Examining Commitee

July 30, 1997

Date

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תושלב״ע.

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Chapter 0

Introduction

Section 0.1: Research Program

In this thesis we consider the problem of forumlating a general theory of multiphoton double-ionization and use this theory to make predictions about the photoionization rates of helium.

In Chapter 1, we begin by creating a macroscopic picture of the double-ionization processes in terms of transition rates between the ground, ground-singly-ionized, and doubly-ionized states. We divide these processes into *sequential* ($X \to X^{+} \to X^{++}$) and *direct* ($X \to X^{++}$) ionization paths. With this in mind, we attempt to form a microscopic description of the problem using standard scattering theory. We build an S-matrix formalism to describe the ionization process, and in order to write down an answer we make the approximation of expanding in powers of the correlation interaction. We must also approximate our uncorrelated initial and final states, which we do by requiring them to satisfy the Hartree-Fock equations. By doing so our expansion in powers of the interelectronic correlation is a good one since the HF state already "accounts for" some part of the electron-electron interaction. We shall also assume that our initial state is a pure singlet ($L = 0$) state and we will ignore relativistic (spin) effects as being much smaller than the other effects we measure. The electron-laser field interaction is represented by the minimal-coupling gauge in the dipole approximation.

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We find that there is an additional, induced, state representing a third non-direct path ($X \rightarrow \tilde{X} \rightarrow X^{++}$). These three paths interfere. Finally, we connect our quantum-mechanical (microscopic) description to our measurable (macroscopic) events. The connection comes via the structure of energy singularities of the S-matrices describing the ionization dynamics, which we connect to the rates by expanding the latter in a Taylor series in time.

In Chapter 2, we apply the technique we developed in Chatper 1 to the double-ionization of helium. We restrict ourselves to a Strong Field Approxmation (SFA) where we have found we can derive analytic results. In doing this, we find that we are faced with two calculational difficulties. First, we encounter the Generalized Bessel Functions (GBFs, see Appendix 2) over which we must integrate. In the SFA, we find that we can approximate these functions by using the saddle-point method of integration on their integral representation. Also, we restrict ourselves to considering the case of a linearly-polarized laser so that we may evaluate the integrals into which the GBFs arise in a closed form. Second, we find that for the direct process, we are faced with a difficult sum-over-states. For this we make an approximation† by using the method of Stationary Closure—see Appendix 1 for details. Using these three restrictions: the SFA, the saddle-point method, and the method of Stationary Closure, we find analytic expressions for the ionization rates in terms of the laser intensity. We make a connection between experimental observations of ionization yields and our theory, and we propose that experimentalists' hypothesis, that a sudden change in the shape of the yield curve is due to the dominance of the sequential process over the direct one is not supported by our theory in the region of applicability of our theory.

Section 0.2: Notation and Units Used

Throughout, we shall use Hartree's "atomic units"[1] such that:

$$\varepsilon = \hbar = m_e = 1 \qquad 0.2.1$$

with ε being the electron charge and m_e being the electron mass. In this system, we also have:

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† a crude approximation

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$$\begin{aligned} \mathrm{H_t} = \mathrm{a_0} &= 1 \\ c &= 137 \end{aligned} \tag{0.2.2}$$

where $\mathrm{H_t}$ is a Hartree unit of energy ($-\varepsilon^2/\mathrm{a_0} = -27.6\,\mathrm{eV}$), and $\mathrm{a_0}$ is the Bohr radius $\mathrm{a_0} = \hbar^2/m_e\varepsilon^2$. Wherever there could be confusion, constants will be explicitly given. In this system, we have atomic units of time and intensity:

$$t_{\mathrm{au}} = \frac{\mathrm{a_0}}{\alpha c} = 2.42 \times 10^{-17}\,\mathrm{s} \tag{0.2.3a}$$

$$I_{\mathrm{au}} = \frac{c\varepsilon^2}{8\pi \mathrm{a_0}^4} = 3.52 \times 10^{16}\,\mathrm{W/cm^2} \tag{0.2.3b}$$

We shall attempt to adhere to the convention that upper-case Greek letters (Φ, Ψ, X, *etc.*) and upper-case Roman letters (W, H, Q, *etc.*) will be used to denote two-particle eigenfunctions, hamiltonians, energies or the like, and lower-case Greek letters (ϕ, ψ, χ, *etc.*) and lower-case Roman letters (w, h, q, *etc.*) for one-particle hamiltonians, eigenvectors, wavefunctions, momenta, and the like. Where we do not do so, we hope that context will make clear the divergence from convention.

Where we write $\dot{g}$ for some time-dependent function g we mean to take its time derivative:

$$\dot{g} \overset{\mathrm{def}}{=} \frac{\mathrm{d}}{\mathrm{d}t} g \qquad .$$

Whenever we write:

$$\langle \phi | \mathcal{A} | \psi \rangle \tag{0.2.4}$$

unless otherwise specified, we mean:

$$\underbrace{\int_{C_1} \int_{C_2} \cdots \int_{C_n}}_{n\ \textbf{integrations}} \mathrm{d}^3x_1 \mathrm{d}^3x_2 \ldots \mathrm{d}^3x_n\, \phi^\star(\vec{x}_1, \vec{x}_2, \ldots, \vec{x}_n)\; \mathcal{A}\, \psi(\vec{x}_1, \vec{x}_2, \ldots, \vec{x}_n) \tag{0.2.5}$$

where n is the number of particles which the functions represent, $\vec{x}_i$ is the position of the i^{th} particle, and $\mathcal{A}$ is an operator. The C_i's are the manifolds over which we are integrating, usually "all space"

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Chapter 1

A General Theory of Double Multi-Photon Ionization

In this chapter we present a general theory of double multiphoton ionization of heliumlike atoms. With this we determine the experimentally observable signatures of the *sequential* and *direct* ionization paths. First, we must connect macroscopic events (observables, such as ionization rates) to the microscopic terms which determine the dynamics of the system. Then, we must calculate the microscopic terms in a manner which is analytically soluble, or at least numerically tractable.† In this chapter we follow closely the method developed by Mittleman[3] in connecting the two terms via the singularity structure of the S-matrix, which determines the entire ionization process[4].

Section 1.1: The Double MPI rate equations

Characterization of double MPI is much more complicated than single MPI because there are competing processes present. For single MPI, there is only one process, for which the rate equations are:

$$\begin{aligned} \dot{n}_0 &= -\gamma_1 n_0 \\ \dot{n}_1 &= +\gamma_1 n_0 \end{aligned} \qquad 1.1.1$$

where n_0 and n_1 are the populations (as functions of time) of the ground and ionized states, respectively. γ_1 is the ionization rate. We make the assumption that all the atoms we ionize start in their ground state and leave the ionized atom in the

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† This is to be compared with the work of Parker, *et al.*[2], who take the approach of the direct numerical solution to the Schrödinger equation.

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lowest ionized state.† We also assume that the rate(s) γ_1 are only adiabatically time-dependent through the switching on-and-off of the laser. For the double MPI event, the system is more complicated‡:

$$\begin{aligned} \dot{n}_0 &= (-\gamma_{01} - \gamma_{02})\, n_0 \\ \dot{n}_1 &= +\gamma_{01} n_0 - \gamma_{12} n_1 \\ \dot{n}_2 &= +\gamma_{02} n_0 + \gamma_{12} n_1 \end{aligned} \qquad 1.1.2$$

where now n_2 is the population of the doubly-ionized state, and we write γ_{ij} for the rate of ionization from state i to state j. Once again, we assume that we only populate one singly-ionized excited state and one doubly-ionized excited state.§ The solution to this set of coupled linear ODEs is simple to get. However, we defer it until after Section 1.3 for reasons to be stated there.

Now we have two different, competing, paths for our double MPI process. Either we can ionize our atom in two steps, by going through the singly-ionized state as an intermediate step, as seen in Figure 1.1,

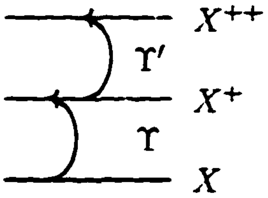

Figure 1.1: Sequential double ionization taking Υ photons to get to a singly-ionized state and Υ' photons to get to a doubly ionized state

or in one step, going directly from neutral to doubly-ionized, as in Figure 1.2.

We term the former process *sequential* and the latter *direct*.

In our analysis we will assume that the ground state is a spin singlet state, and we will ignore relativistic (spin) effects. Therefore, all the wavefunctions with which we deal shall be spin singlets and therefore symmetric under interchange of the two ejected electrons.

---

† This analysis could be carried on to include multiple final states, *e.g.* leaving the ionized atom in any of a number of final states. We shall not pursue that here.

‡ We have not found a rigorous quantum-mechanical derivation of this description of the cascading ionization processes. Mittleman[5], however, has a forthcoming letter which does describe it.

§ We could include multiple intermediate singly-ionized states (we shall do this in the next section) and multiple doubly-ionized states. We only consider for this problem the ground doubly-excited ion as the only possible final state.

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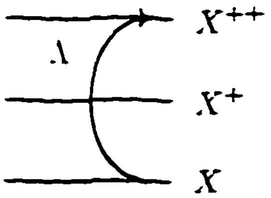

Figure 1.2: Direct MPI going from the ground state to the doubly-ionized state without going through the singly-ionized state, using Λ photons

It is these ionization rates appearing in Equation 1.1.2 which must be connected to the dynamics of the ionization process. The ionization process, as mentioned above, is completely determined by an S-matrix. Therefore, we must begin to examine this S-matrix.

Section 1.2: The S-matrix for Single-MPI of Hydrogen

Before we consider the double-ionization case, let us first examine the simpler case of the single-ionization problem of a hydrogenlike atom. In this way we establish the general principle of connecting the singularities of the S-matrix to macroscopic observables. Our starting point is Equation 1.1.1, and the time-reversed S-matrix‡ describing single ionization:

$$s = \lim_{t\to-\infty} \langle \psi_q^{(-)} | \phi_0 \rangle \qquad 1.2.1$$

where ϕ_0 is the ground-state of the field-free hamiltonian $h_0 = t + v$, where v is the nuclear potential $v = -Z/r^2$. $\psi_q^{(-)}$ is a scattering state which is an eigenstate of the hamiltonian including the laser-electron interaction:

$$\left(t + v + h'(t)\right) \psi_q^{(-)} = i\dot{\psi}_q^{(-)} \qquad 1.2.2$$

but which evolves as $t \to \infty$ into a free electron (since as $t \to \pm\infty$, the laser, h', switches off:

$$\lim_{t\to\infty} \psi_q^{(-)} \to \phi_q e^{-itw_q}. \qquad 1.2.3$$

Here we have ϕ_q a free solution to

$$(t + v)\phi_q = w_q \phi_q \qquad 1.2.4$$

---

‡ There are no problems in using this form due to the time-reversal symmetry of the scattering process. See Newton[4], §§6.4 and 7.2.

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whose energy is w_q.

Equation 1.2.1 as it stands is difficult to evaluate, so we introduce the following technique: let us write the limit as part of an improper integral:

$$s = -\int_{-\infty}^{\infty} \mathrm{d}t \, \frac{\mathrm{d}}{\mathrm{d}t} \langle \psi_q^{(-)} | \phi_0 \rangle + \lim_{t\to\infty} \langle \psi_q^{(-)} | \phi_0 \rangle. \qquad 1.2.5$$

The last term will vanish due to the limiting behavior of $\psi_q^{(-)}$ as given in Equation 1.2.3. We can use the Schrödinger equations for $\psi_q^{(-)}$ and ϕ_0 to rewrite the time derivative:

$$s = -\mathrm{i} \int_{-\infty}^{\infty} \mathrm{d}t \, \langle \psi_q^{(-)} | h' | \phi_0 \rangle. \qquad 1.2.6$$

We shall assume that our laser-atom interaction $h'(t)$ is the dipole limit of $h'(t) = \vec{p} \cdot \vec{A}(\vec{r}, t) + A^2/2$, where $\vec{A}(\vec{r}, t)$ is the laser field vector potential at the point $\vec{r}$.

We Fourier analyze the field-interaction:

$$h' = \sum_n h'_n \, \mathrm{e}^{-\mathrm{i}n\omega t} \qquad 1.2.7$$

Now, the matrix element is time-independent and the time integral may be performed immediately:

$$\begin{aligned} s &= \sum_n s_n \\ s_n &= -\mathrm{i} \, \langle \phi_q | h'_n | \phi_0 \rangle f(\Delta(n)) \\ &\text{with} \\ \Delta(n) &= w_q - w_0 - n\omega \\ f(\Delta) &= \lim_{T\to\infty} \frac{2 \sin \Delta T}{\Delta} \end{aligned} \qquad 1.2.8$$

$\Delta(n) = 0$ is the equation requiring the process to be on-shell, *i.e.*, energy-conserving. The relation[6]

$$\lim_{t\to\infty} \frac{\sin tx}{x} = \pi \delta(x). \qquad 1.2.9$$

is a standard representation of the Dirac δ-function. If we use this, we may write:

$$s_n = -2\pi \mathrm{i} \, T_n(q) \, \delta\left(\Delta(n)\right) \qquad 1.2.10$$

$$T_n(q) \stackrel{\mathrm{def}}{=} \langle \phi_q | h'_n | \phi_0 \rangle \qquad 1.2.11$$

as the usual definition of the T-matrix[4]. We explicitly show the dependence of T on q, the momentum of the outgoing electron.

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The S-matrix represents the total probability amplitude of getting "to" state f from state i. Its absolute square is then the probability of being in final state f coming from final state i as a function of q and n. If we then integrate over all possible outgoing final momenta, and sum over n, we shall have the *total probability of being in state f coming from state i.*

There is one problem with progressing in this fashion. The square of the δ-function appearing in Equation 1.2.8 is not well-defined! We work around this problem by looking directly at $|s_n|^2$, before turning $f(x)$ into $\delta(x)$:

$$|s_n|^2 = \lim_{T\to\infty} |T_n(q)|^2 f^2(\Delta T). \tag{1.2.12}$$

The function $f^2(T)$ also has an infinite-T limit which is a δ-function[7]:

$$\lim_{T\to\infty} f^2(\Delta T) = 4\pi T\delta(\Delta). \tag{1.2.13}$$

Now, the square of the S-matrix is well-defined as well:

$$|s_n|^2 = 2\pi T\delta(\Delta(n))|T_n(q)|^2 \tag{1.2.14}$$

and we can write the entire S-matrix as the sum:

$$|s|^2 = \sum_{n,n'} s_n s^*_{n'}.$$

The δ-functions, replaced by the sharply-peaked functions f, have negligible overlap when $n \neq n'$. In the infinite-time limit, we can write:

$$\begin{aligned} |s|^2 &= \sum_n |s_n|^2 \\ &= 2\pi T \sum_n \delta(\Delta(n))|T_n(q)|^2. \end{aligned} \tag{1.2.15}$$

Since $|s|^2$ is interpreted as a transition (ionization) probability, $|s|^2/T$ may be interpreted as an ionization probability per time given a specific momentum of the ionized electron. The integral of this over all possible electron momenta gives the total ionization rate. To connect this to the macroscopic observable (the rate γ_1), return to the rate equations (Equation 1.1.1). The instantaneous ionization rate is given by $\dot{n}_1(t)/n_0(t)$. To lowest order in t, this is

$$\frac{\dot{n}_1(t)}{n_0(t)} = \gamma_1 t + O(t^2) \tag{1.2.16}$$

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In the last part of the above, we have expanded the rate for small times t. If we now associate the coefficient of t in Equation 1.2.14 with the coefficient of T in $\dot{n}_1(t)/n_0(t)$, then:

$$\gamma_1 = 2\pi \sum_n \delta(\Delta(n))|T_n(q)|^2. \qquad 1.2.17$$

In arriving at Equation 1.2.17 we have used two different limits for time. In deriving the S-matrix, we have let time get very large (to turn a sharply-peaked function into a δ-function). In approximating the rate equations, we require time to be very small (to approximate the solution by its Taylor series). This need not be inconsistent. We require that the time be large on the smallest time scale of laser energy ($\omega t \gg 1$) but small when compared to the ionization rate ($\gamma_1 t \ll 1$). This condition is met if

$$\omega \gg \gamma_1, \qquad 1.2.18$$

that is, if the laser frequency is much higher than the ionization rate. Another way of saying this is that ground state must not ionize faster than the laser can probe the atom. For most realistic experiments, this condition is satisfied. This point is important and we will revisit it in Section 1.5 in regard to being able to distinguish the direct ionization process from the sequential ones.

We have now established a connection between the singularity structure (appearance of δ-functions) in the S-matrix and macroscopic observables in the single MPI case. We must now do the same for the double MPI case.

Section 1.3: The Double MPI S-Matrix for Helium

Our starting point is the most general form for the S-matrix for double ionization[4][8]:

$$S(q_1, q_2) = \lim_{t\to-\infty} \langle \Psi^{(-)}_{q_1,q_2} | \Phi_0 \rangle \qquad 1.3.1$$

where Φ_0 is the exact solution for a the helium atom in the ground state:

$$\left(H_{He} - i\frac{\partial}{\partial t} \right) \Phi_0 = 0 \qquad 1.3.2$$

$$H_{He} = t_1 + t_2 + v_1 + v_2 + V_{12} \qquad 1.3.3$$

where t_i is the kinetic energy of electron i, v_i is the nuclear attaction on electron i, and V_{12} is the interelectronic repulsion.

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$\Psi^{(-)}_{q_1,q_2}$ is the solution to the Schrödinger equation which includes the helium hamiltonian and the laser field:

$$\left(H_{He} + H' - i\frac{\partial}{\partial t}\right)\Psi^{(-)}_{q_1,q_2} = 0 \qquad 1.3.4$$

The superscript $(-)$ indicates that we are using the time-reversed solution which evolves from two free particles in the distant future, and the subscript q_1, q_2 indicates that we want the solution with the two ejected electrons having momenta $\vec{q}_1$ and $\vec{q}_2$, respectively. The electron-field interaction acts on each electron independently, and can therefore be written as the sum of two one-particle hamiltonians:

$$H'(t) = h'_1(t) + h'_2(t). \qquad 1.3.5$$

We can write the S-matrix in a somewhat more useful form, indicating explicitly the electron-laser field interaction. First, formally differentiate and antidifferentiate with respect to t:

$$S(q_1,q_2) = -\int_{-\infty}^{\infty} dt\,\frac{d}{dt}\langle\Psi^{(-)}_{q_1,q_2}|\Phi_0\rangle + \lim_{t\to\infty}\langle\Psi^{(-)}_{q_1,q_2}|\Phi_0\rangle. \qquad 1.3.6$$

(The final term arises to cancel the one introduced with the integral.) The last term will cancel because in the distant future, when the laser turns off, $\Psi^{(-)}_{q_1,q_2}$ is an eigenfunction of the field-free hamiltonian H (since $H' \to 0$) and therefore $\Psi^{(-)}_{q_1,q_2}$ and Φ_0 are orthogonal. By using the Schrödinger equations for the two states to eliminate the time derivative, we can rewrite the integral as:

$$S(q_1,q_2) = -i\int_{-\infty}^{\infty} dt\langle\Psi^{(-)}_{q_1,q_2}|H'|\Phi_0\rangle. \qquad 1.3.7$$

If we could write down exact expressions for Φ_0 and $\Psi^{(-)}_{q_1,q_2}$, we would be finished. Unfortunately, we cannot do so because the interelectronic correlation makes this impossible. Therefore, we must content ourselves with approximations. We choose, to approximate our exact solutions by "uncorrelated" Hartree-Fock solutions, perturbed slightly by the interelectronic electrostatic interaction. This is a reasonable assumption because the uncorrelated HF ground state of helium gives a ground-state energy which differs from the experimentally-determined ground-state energy of helium by 1.4%† and strong fields make interelectronic correlation less important. Our

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† See Bransden & Joachain[1], Table 7.5

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starting point is now an expansion of the initial and final states in the interelectronic correlation. We write the formal perturbation series:

$$\Phi_0 = U_0 + G_0 \delta V U_0 + \cdots \qquad 1.3.8$$

where we define the uncorrelated HF ground state U_0 and Green's function G_0 as:

$$\left(H_0 - i\frac{\partial}{\partial t}\right)\begin{Bmatrix} U_0 \\ G_0 \end{Bmatrix} = \begin{Bmatrix} 0 \\ 1 \end{Bmatrix}. \qquad 1.3.9$$

where now we have the Hartree-Fock hamiltonian:‡

$$H_0 = t_1 + t_2 + v_1 + v_2 + \Omega_1 + \Omega_2 - \nu P, \qquad 1.3.10$$

Ω_i is the Hartree-Fock potential on electron i:

$$\Omega(\vec{r}) = \int d^3x \, \frac{1}{|\vec{r} - \vec{x}|} \, u_0^2(\vec{x}), \qquad 1.3.11$$

and u_0 is the one-particle ground "Hartree-Fock" orbital from the set of orbitals defined by

$$(t + v + \Omega)u_n = e_n u_n, \qquad n \geq 0. \qquad 1.3.12$$

P is a projection operator of both particles onto the ground HF state: $P = |u_0 u_0\rangle\langle u_0 u_0|$ and ν is the shift in the ground-state HF energy due to interelectronic correlation:

$$\nu = \iint d^3r \, d^3x \, u_0^2(\vec{r}) \, \frac{1}{|\vec{r} - \vec{x}|} \, u_0^2(\vec{x}). \qquad 1.3.13$$

If we apply the Schrödinger equation 1.3.9 to the functions u_0 from Equation 1.3.12 we get the eigenenergy of U_0 to be $W_0 = 2e_0 - \nu$, the variational energy, instead of $2e_0$. This is the reason for the presence of the ν term in Equation 1.3.10. We expect the rates to be rapid functions of this variational energy[22]. Thus, by using this HF approximation we have a good approximation to W_0 where it is critical. We then write the general time-dependent wavefunction $U_0(t)$ as

$$U_0 = u_0 u_0 \, e^{-itW_0}.$$

Given H_0 from Equation 1.3.10 and H from Equation 1.3.3, we get that δV is defined by:

$$\delta V = V_{12} - \Omega_1 - \Omega_2 + \nu P. \qquad 1.3.14$$

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‡ See, *e.g.*, Landau & Lifshitz[7], §69, Equations 69.5 and 69.8. What we call Ω_i Landau calls G_{ii}.

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δV, written in this fashion, has the additional property that it will couple U_0 only to doubly-excited states. To see this, look at the expectation value of a transition between U_0 and a singly-excited state $u_k u_0$ ($k \neq 0$) driven by the potential δV:

$$\langle u_k u_0 | \delta V | u_0 u_0 \rangle = \langle u_k u_0 | V_{12} - \Omega_1 - \Omega_2 + \nu P | u_0 u_0 \rangle \qquad 1.3.15$$

Since each of the Ω's are one-particle operators, those terms are easily written down:

$$\langle u_k u_0 | \Omega_1 | u_0 u_0 \rangle = \langle u_k | \Omega_1 | u_0 \rangle \langle u_0 | u_0 \rangle = \langle u_k | \Omega | u_0 \rangle.$$
$$\langle u_k u_0 | \Omega_2 | u_0 u_0 \rangle = \langle u_k | u_0 \rangle \langle u_0 | \Omega | u_0 \rangle = 0.$$

where we have used the fact that the wavefunctions are restricted by $\langle u_m | u_n \rangle = \delta_{m,n}$. The last term in Equation 1.3.15 is zero because P acting to the left on $\langle u_k u_0 |$ is zero. The remaining term is:

$$\begin{aligned}\langle u_k u_0 | V_{12} | u_0 u_0 \rangle &= \iint d^3r\, d^3x\, u_k(\vec{r}) u_0(\vec{r}) u_0^2(\vec{x}) \frac{1}{|\vec{r} - \vec{x}|} \\ &= \int d^3r\, u_k(\vec{r}) \left(\int d^3x \frac{u_0^2(\vec{x})}{|\vec{r} - \vec{x}|} \right) u_0(\vec{r}) \\ &= \langle u_k | \Omega_1 | u_0 \rangle\end{aligned}$$

and this term cancels the one remaining from the equation above. If $k = 0$, then the first term in Equation 1.3.15 is exactly the last term, and some simple algebra like above shows that the sum of the middle two terms exactly cancel the first and last. Therefore, δV connects the ground state to only doubly-excited states. The reason why this is important becomes apparent when one attemps to include this term in the rate calculations, as we shall see in the next chapter. We call this term the *initial state correlation interaction.*

We justify treating δV as a small perturbation by noting that it measures, in effect, the difference between the full interelectronic repulsion and that approximated by the HF potentials. We have seen above that this difference is 1.4% in the ground state, which is small.

The causal Green's function defined by Equation 1.3.9 can be written as the usual eigenfunction expansion:

$$G_0^{(+)}(t - t') = -i\Theta(t - t') \sum_{n,n'} |u_n\, u_{n'}\rangle \langle u_n\, u_{n'}|\, e^{-i(W_{n,n'} - i\eta)(t - t')} \qquad 1.3.16$$

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where $W_{n,n'} = e_n + e_{n'} - \nu\,\delta_{n,0}\delta_{n',0}$. The Θ-function is the Heaviside function:

$$\Theta(x) = \begin{cases} 1, & \text{if } x \geq 0: \\ 0, & \text{otherwise.} \end{cases}$$

We proceed to write an expansion for $\Psi^{(-)}_{q_1,q_2}$ in the same fashion. Analogous to Equation 1.3.10, we write:

$$\left(H - i\frac{\partial}{\partial t}\right)\begin{Bmatrix} \Psi^{(-)} \\ G \end{Bmatrix} = \begin{Bmatrix} 0 \\ 1 \end{Bmatrix} \qquad 1.3.17$$

and an analogue to Equation 1.3.12, which includes the field and the HF potential:

$$\left(t + v + (\Omega - \frac{\nu}{2}\pi) + h'(t) - i\frac{\partial}{\partial t}\right)\psi^{(-)}_n = 0 \qquad 1.3.18$$

where $\pi = |u_0\rangle\langle u_0|$ is the one-particle projection operator onto u_0. The boundary conditions on $\psi^{(-)}_n$, that in the distant $(t \to \infty)$ future, it go to u_n, are satisfied since in that limit $h' \to 0$. Therefore,

$$\lim_{t\to\infty} \psi^{(-)}_n \to u_n e^{-ie_n t}. \qquad 1.3.19$$

This why we choose the ground state correlation $(\Omega - \frac{\nu}{2}\pi)$ in Equation 1.3.18.

Now, the full two-particle wavefunction $\Psi^{(-)}_{q_1,q_2}$ can be expanded in a perturbation series in the one-particle wavefunctions $\psi^{(-)}$:

$$\Psi^{(-)}_{q_1,q_2} = \psi^{(-)}_{q_1}\psi^{(-)}_{q_2}\left(1 + G\,\delta V' + \cdots\right)\sigma \qquad 1.3.20$$

and σ is an operator which enforces the symmetry under coördinate interchange $(x_1 \leftrightarrow x_2)$. Therefore, our perturbing potential $\delta V'$ is written as:

$$\delta V' = V_{12} - \Omega_1 - \Omega_2 + \frac{\nu}{2}(\pi_1 + \pi_2) \qquad 1.3.21$$

We consider the $\delta V'$ term small (and therefore acceptable as a perturbation) for the same reason that we accept δV in Equation 1.3.14 as small. That is, essentially, that the HF-Volkov wavefunctions well-approximate the actual wavefunctions. As well, we expect correlation to be even less important in scattering states.

With foreknowledge of the results, we write the exact Green's function as:

$$G = \Gamma_1 + G\Delta_1\Gamma_1. \qquad 1.3.22$$

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Here, Γ_1 is a propagator which has one electron in an HF-Volkov (v) state and one electron in an eigenstate w_n of the singly-ionized atom:

$$(t + v)w_n = i\frac{\partial}{\partial t}w_n \qquad 1.3.23$$

with energy† W_n^+. This decomposition is not unique but has the advantage that the first-order sequential term (Equation 1.3.51) describes the process of double-ionization by first singly-ionizing the atom, leaving the atom in the ground singly-charged state, and then ionizing the singly-charged ion in a second step. Γ_1 satisfies the Schrödinger equation:

$$\left(t_1 + t_2 + v_1 + v_2 + \Omega_2 - \frac{\nu}{2}\pi_2 + h_2' - i\frac{\partial}{\partial t}\right)\Gamma_1 = 1 \qquad 1.3.24$$

We write Γ_1 as a sum over the discrete states w_n and an integral over the continuous ones ψ_q:

$$\Gamma_1(t,t') = -i\Theta(t-t')\sum_n \int \frac{d^3q}{(2\pi)^3} e^{-i(W_n^+ - i\eta)(t-t')}|w_n(1)\psi_q(2,t)\rangle\langle w_n(1)\psi_q(2,t')| \qquad 1.3.25$$

What is left then as a "perturbing" agent in G is

$$\Delta_1 = V_{12} - \Omega_2 - h_1' + \frac{\nu}{2}\pi_2. \qquad 1.3.26$$

We shall keep only the first order term in Equation 1.3.25 for the following reason: the second term ($G\Delta_1\Gamma_1$) is essentially a rescattering term, scattering electron 1 off of an HF-Volkov state and electron 2 off of an ionic state. As well, we are only keeping first order small terms in the calculation of Φ_0.

The Lowest-Order Term

The lowest-order, and totally correlation-free, contribution to the S-matrix comes from keeping only the U_0 term in Φ_0 and the $\psi_{q1}^{(-)}\psi_{q2}^{(-)}$ term in Ψ. This gives us:

$$S^I = \lim_{t\to-\infty}\langle\psi_{q1}^{(-)}\psi_{q2}^{(-)}|\phi_0\phi_0\rangle. \qquad 1.3.27$$

Where we have set $\phi_0 = u_0\exp(-iW_0t/2)$. We can use the same technique as in the single-ionization case (Equation 1.2.5) to change this into an integral:

$$S^I = \int_{-\infty}^{\infty} dt \frac{d}{dt}\langle\psi_{q1}^{(-)}\psi_{q2}^{(-)}|\phi_0\phi_0\rangle + \lim_{t\to\infty}\langle\psi_{q1}^{(-)}\psi_{q2}^{(-)}|\phi_0\phi_0\rangle. \qquad 1.3.28$$

---

† We deviate here from our normal usage for capital and small roman letters.

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Now, we use the Schrödinger equations for ψ_q and ϕ_0 and write Equation 1.3.27 analogously to Equation 1.2.6:

$$S^f = -\mathrm{i}\int_{-\infty}^{\infty} \mathrm{d}t \langle \psi_{q_1}^{(-)} \psi_{q_2}^{(-)} |\, (h'_1 + h'_2)\, | \phi_0 \phi_0 \rangle. \qquad 1.3.29$$

Each of the $h'_{1,2}$ are *one*-particle operators, so this reduces to:

$$S^f = -\mathrm{i}\int_{-\infty}^{\infty} \mathrm{d}t \left(\langle \psi_{q_1}^{(-)} | h' | \phi_0 \rangle \langle \psi_{q_2}^{(-)} | \phi_0 \rangle + \langle \psi_{q_2}^{(-)} | h' | \phi_0 \rangle \langle \psi_{q_1}^{(-)} | \phi_0 \rangle \right). \qquad 1.3.30$$

We can introduce an integration trick to write this in a more useful form. Note that for the first integral in Equation 1.3.30:

$$\begin{aligned}\int_{-\infty}^{\infty} \mathrm{d}t \, \langle \psi_{q_1}^{(-)} | h' | \phi_0 \rangle \langle \psi_{q_2}^{(-)} | \phi_0 \rangle &= -\int_{-\infty}^{\infty} \mathrm{d}t \, \langle \psi_{q_1}^{(-)}(t) | h'(t) | \phi_0(t) \rangle \times \\ &\left(\int_t^{\infty} \mathrm{d}t' \frac{\mathrm{d}}{\mathrm{d}t'} \langle \psi_{q_2}^{(-)}(t') | \phi_0(t') \rangle - \lim_{t' \to \infty} \langle \psi_{q_2}^{(-)}(t') | \phi_0(t') \rangle \right)\end{aligned} \qquad 1.3.31$$

Once again, we can drop the last inner product because of the limiting condition on ψ_q in Equation 1.3.19 and the fact that the u's are an complete set.

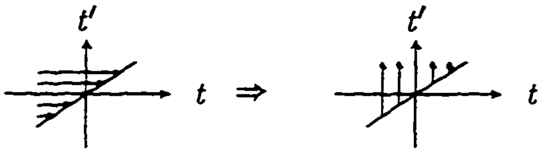

Figure 1.3: Change of integration range trick

If we once again use the Schrödinger equation on the integral in Equation 1.3.31 we get:

$$-\mathrm{i}\int_{-\infty}^{\infty} \mathrm{d}t \, \langle \psi_{q_1}^{(-)}(t) | h' | \phi_0(t) \rangle \int_t^{\infty} \mathrm{d}t' \, \langle \psi_{q_2}^{(-)}(t') | h' | \phi_0(t') \rangle \qquad 1.3.32$$

Now, we can apply the change-of-integration-range trick (given pictorially in Figure 1.3‡):

$$\int_{-\infty}^{\infty} \mathrm{d}t \, f(t) \int_t^{\infty} \mathrm{d}t' \, g(t') = \int_{-\infty}^{\infty} \mathrm{d}t' \, g(t') \int_{-\infty}^{t'} \mathrm{d}t \, f(t), \qquad 1.3.33$$

to rewrite Equation 1.3.32 as

$$\int_{-\infty}^{\infty} \mathrm{d}t \, \langle \psi_{q_2}^{(-)} | h' | \phi_0 \rangle \int_{-\infty}^{t} \mathrm{d}t' \, \langle \psi_{q_1}^{(-)} | h' | \phi_0 \rangle. \qquad 1.3.34$$

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‡ See also Sakurai[9], page 186.

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If we now use the same trick on the second integral in Equation 1.3.30, we get

$$-\mathrm{i}\int_{-\infty}^{\infty}\mathrm{d}t\,\langle\psi_{q_2}^{(-)}(t)|h'|\psi_{q_1}^{(-)}(t)\rangle\int_{t}^{\infty}\mathrm{d}t'\,\langle\psi_{q_1}^{(-)}(t')|h'|\phi_0(t')\rangle. \qquad 1.3.35$$

and the sum of these two gives:

$$\begin{aligned} S^I &= \left(\int_{-\infty}^{\infty}\mathrm{d}t\,\langle\psi_{q_1}^{(-)}|h'|\phi_0\rangle\right)\left(\int_{-\infty}^{\infty}\mathrm{d}t\,\langle\psi_{q_2}^{(-)}|h'|\phi_0\rangle\right)\\ &= S^{(1)}(q_1)\times S^{(1)}(q_2)\\ &\text{with}\\ S^{(1)} &= -\mathrm{i}\int_{-\infty}^{\infty}\mathrm{d}t\,\langle\psi_q|h'|\phi_0\rangle \end{aligned} \qquad 1.3.36$$

We have broken up the double ionization problem into a product of two single-ionization problems! This makes sense in this lowest-order context. We have "turned off" the interelectronic correlation, and so the double-ionization amplitude should be the product of two single-ionization amplitudes.

There is something peculiar to which this term gives rise. Performing the time integration for $S^{(1)}$, we get a δ-function which contains the energies of the initial and final states. The Volkov states have a time dependence which is[10]:

$$\psi_q(t) = \mathrm{e}^{\mathrm{i}\vec{q}\cdot\vec{r}}\,\mathrm{e}^{-\mathrm{i}(t\,q^2/2+\mathrm{U_p}/2\omega(t+\sin 2\omega t)+\vec{\alpha}_0\cdot\vec{q}\sin\omega t)}. \qquad 1.3.37$$

$\mathrm{U_p} = E^2/2\omega^2$ is the ponderomotive potential. We get:

$$S^{(1)}(q) = -2\pi\mathrm{i}\sum_n T_n^{(1)}(q)\,\delta\left(\frac{q^2}{2}+\mathrm{U_p}-n\omega-\frac{W_0}{2}\right) \qquad 1.3.38$$

Equation 1.3.38 defines a T-matrix $T^{(1)}$:

$$T^{(1)} = \sum_{s,n}\langle f_{q,n}|h'_s|u_0\rangle. \qquad 1.3.39$$

where we have fourier-analyzed $h' = \sum h_s \exp(\mathrm{i}\omega s t)$ and $f_{q,n}$ is the quasiperiodic expansion of the Volkov states. (See Equation 1.3.49.) We may now identify $n = j-s$ as a new summation index. This interaction "connects" the ground state to the doubly excited state through an intermediate state which is *not a state represented in the rate equations (1.1.2)!* We see this by noting that

$$S^I = (-2\pi\mathrm{i})^2\sum_{n,n'} T_n^{(1)}(q_1)\,T_{n'}^{(1)}(q_2)\,\delta\left(E_1(n)\right)\delta\left(E_2(n')\right) \qquad 1.3.40$$

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where we have defined the shorthand

$$E_\ell(n) = \frac{q_\ell^2}{2} + U_p - n\omega - \frac{W_0}{2}. \qquad 1.3.41$$

We note that when we integrate over the two δ-functions we can rewrite their product as

$$\delta\left(E_1(n)\right)\delta\left(E_2(n')\right) = \delta\left(E_1(n)\right)\delta\left(\Delta(N)\right), \qquad 1.3.42$$

where we have defined $\Delta(n)$ to represent overall energy conservation, analogously to Equation 1.2.8:

$$\Delta(N) = E_1(n) + E_2(n') \qquad 1.3.43$$

and we let $N = n + n'$. This first δ-function in Equation 1.3.42 represents overall energy conservation but the second requires that one electron have an energy which is not an eigenenergy of the singly-charged ion. Our initial set of rate equations (Equation 1.1.2) must be incomplete. To remedy this, we add in an intermediate "Hartree-Fock state" $\bar{X}$:

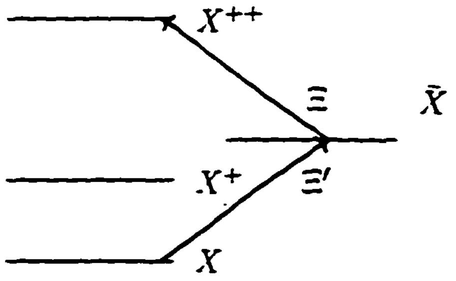

Figure 1.4: Schematic of non-sequential double ionization taking Ξ photons to get to an intermediate "Hartree-Fock" state $\bar{X}$ and Ξ' photons to get to a doubly ionized state.

Thus, we must add this HF state into our rate equations (Equation 1.1.2):

$$\begin{aligned} \dot{n}_0 &= \left(-\gamma_{01} - \gamma_{02} - \bar{\gamma}\right) n_0 \\ \dot{n}_1 &= +\gamma_{01} n_0 - \gamma_{12} n_1 \\ \dot{\bar{n}} &= \bar{\gamma} n_0 - \bar{\gamma}' \bar{n} \\ \dot{n}_2 &= +\gamma_{02} n_0 + \gamma_{12} n_1 \end{aligned} \qquad 1.3.44$$

Here, we represent the population of our new "state" by $\bar{n}$ and its rates of population and ionization by $\bar{\gamma}$ and $\bar{\gamma}'$, respectively. We note that since the two processes $X \rightarrow \bar{X}$ and $\bar{X} \rightarrow X^{++}$ have the same T-matrices, we can equate their respective rates $\bar{\gamma} = \bar{\gamma}'$.

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This result is unusual in that the lowest-order term in the S-matrix is sequential yet the intermediate step does not go through the singly-charged ion but rather through a HF intermediate state.

To make our notation more concise we write

$$\tau^{(2)}_{N,n}(q_1, q_2) = T^{(1)}_n(q_1)\, T^{(1)}_{n'}(q_2), \qquad 1.3.45$$

with $n + n' = N$.The reason for this will become more apparent. We note that $\tau^{(2)}$ is *not* a T-matrix.

The First-Order Sequential Term

The first-order sequential term arises from taking the $\delta V' G$ term in Equation 1.3.20 and approximating G by Γ_1. We also use the same technique which takes us from Equation 1.3.27 to Equation 1.3.29 to write down immediately:

$$S^{II} = -\mathrm{i}\int_{-\infty}^{\infty} \mathrm{d}t\, \langle \psi^{(-)}_{q_1} \psi^{(-)}_{q_2} | \delta V' \Gamma_1 \sigma H' | \phi_0 \phi_0 \rangle \qquad 1.3.46$$

where H' is given by Equation 1.3.5. σ, the symmetrization operator, when acting to the right is the identity since both H' and $|\phi_0\phi_0\rangle$ are symmetric under particle interchange. If we insert the expressions for Γ_1 (1.3.25) and $\delta V'$ (1.3.21) into Equation 1.3.46 the result is:

$$\begin{aligned} S^{II} &= \\ &-\mathrm{i}\int_{-\infty}^{\infty} \mathrm{d}t \sum_n \int \frac{\mathrm{d}^3 q}{(2\pi^3)} \langle \psi^{(-)}_{q_1}(t) \psi^{(-)}_{q_2}(t) | \left[\left(V_{12} - \Omega_2 + \frac{\nu}{2}\pi_2 \right) + \right. \\ &\qquad \left. \left(\frac{\nu}{2}\pi_1 - \Omega_1 \right) \right] | w_n(1,t) \psi_q(2,t) \rangle \times \qquad 1.3.47 \\ &\left(-\mathrm{i}\int_{-\infty}^{t} \mathrm{d}t'\, \mathrm{e}^{-\mathrm{i}(W_n^+ - \mathrm{i}\eta)(t-t')} \langle w_n(1,t') \psi_q(2,t') | H'(t') | \phi_0 \phi_0 \rangle \right) \end{aligned}$$

We have broken up $\delta V'$ in this fashion to emphasize that it has two parts, one which involves particle 2 (via V_{12}, Ω_2 and $(\nu/2)\pi_2$) and another which does not. If one looks at only that contribution to S^{II} from the particle-1 perturbation, the integral $\int \mathrm{d}^3 q$ can be done immediately because the inner product $\langle \psi^{(-)}_{q_2} | \psi_q \rangle$ is the δ-function: $(2\pi)^3 \delta(\vec{q} - \vec{q}_2)$. The sum over n in Equation 1.3.47 is a sum over the ionic states w_n. However, we have (in Section 1.1) already restricted ourselves to one singly-charged state† so it is consistent to take only $n = 0$. After all this, we can write:

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† The ground one.

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$$\begin{aligned} S^{IIs} = -\mathrm{i}\int_{-\infty}^{\infty} \mathrm{d}t \, \langle \psi_{q_1}^{(-)}(t)| \left(\Omega + \frac{\nu}{2}\pi\right) |w_0(t)\rangle \times \\ (-\mathrm{i}) \int_{-\infty}^{t} \mathrm{d}t' \, \mathrm{e}^{-\mathrm{i}(W_0^{+} - \mathrm{i}\eta)(t-t')} \langle w_0 \psi_{q_2}^{(-)} | H'(t') | \phi_0 \phi_0 \rangle \end{aligned} \qquad 1.3.48$$

The reason for the s will become apparent shortly.

It is convenient to write ψ_q in a quasiperiodic (Floquet)‡ series in order to "factor out" the time-dependence so that the time integrals in Equation 1.3.48 may be performed analytically. We write

$$\psi_q(t) = \sum_j \mathrm{e}^{-\mathrm{i}t(q^2/2 + \mathrm{U_p} + j\omega)} f_{q,j} \qquad 1.3.49$$

where $f_{q,j}$ is independent of time. This requires some discussion. Equation 1.3.19 requires that as $t \to \infty$, the time dependence be strictly $\exp(-\mathrm{i}t[q^2/2 + \mathrm{U_p}])$. This would force $f_{q,j}$ to be either *time-dependent* or be proportional to $\delta_{j,0}$. We therefore make the usual assumption of Floquet theory that $f_{q,j}$ is slowly varying (due to switch-on/switch-off) on an atomic and laser time scale (so it may be considered constant for all times appearing in our discussion) yet having the property that $\lim_{t\to\infty} f_{q,j} \to f_{q,0}$.

We have already Fourier-analyzed to field interaction H' so then the inner integral in Equation 1.3.48 becomes:

$$\begin{aligned} -\mathrm{i}\int_{-\infty}^{t} \mathrm{d}t' \, \mathrm{e}^{-\mathrm{i}(W_0^{+} - \mathrm{i}\eta)(t-t')} \langle w_0(t') \psi_{q_2}^{(-)}(t') | H'(t') | \phi_0 \phi_0(t') \rangle \\ = \sum_{j,s} \frac{\langle w_0 f_{q_2,j} | H'_s | u_0 u_0 \rangle}{W_0^{+} + q_2^2/2 + (j-s)\omega + \mathrm{U_p} - W_0 + \mathrm{i}\eta} \end{aligned} \qquad 1.3.50$$

where s indexes the Fourier modes of H'. The remaining time integral in Equation 1.3.48 can now be readily performed using the common definition of the δ-function

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‡ See Friedrich[10], §5.1.3 for more details on this expansion.

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in Equation 1.2.9. The result, after some short algebra, is:

$$S^{IIs} = -2\pi \mathrm{i} \sum_{j,j',s} \delta \left(\frac{q_1^2}{2} + \frac{q_2^2}{2} + 2\mathrm{U_p} - W_0 + (j + j' - s)\omega \right) \times$$

$$\frac{\langle f_{q_1,j'} | (\frac{\nu}{2}\pi - \Omega) | w_0 \rangle \langle f_{q_2,j} w_0 | H'_s | u_0 u_0 \rangle}{W_0 - W_0^+ - \mathrm{U_p} + (j - s)\omega - q_2^2/2 + \mathrm{i}\eta}$$

Let $N = s - j - j'$ and $m = s - j$. 1.3.51

$$= -2\pi \mathrm{i} \sum_N \delta \left(\Delta(N) \right) \times$$

$$\sum_{m,s} \frac{\langle f_{q_1,m-N} | \left(\frac{\nu}{2}\pi - \Omega \right) | w_0 \rangle \langle w_0 f_{q_2,s-m} | H'_s | u_0 u_0 \rangle}{W_0 - W_0^+ - \mathrm{U_p} - m\omega - q_2^2/2 + \mathrm{i}\eta}$$

The definition of $\Delta(N)$ is given in Equation 1.3.43. Now, let $\bar{E}_2(m)$ be the real part of the denominator in the last line of the above equation. We see that it is independent of s, so we can finally rewrite this as:

$$S^{IIs} = -2\pi \mathrm{i} \sum_N \delta \left(\Delta(N) \right) \sum_{m=0}^{N-1} \frac{\tau^{(1)}_{N,m}(q_1, q_2)}{\bar{E}_2(m) + \mathrm{i}\eta} \qquad 1.3.52a$$

$$\tau^{(1)}_{N,m}(q_1, q_2) = \sum_s \langle f_{q_1,m-N} | \left(\frac{\nu}{2}\pi - \Omega \right) | w_0 \rangle \langle w_0 f_{q_2,s-m} | H'_s | u_0 u_0 \rangle \qquad 1.3.52b$$

$$\bar{E}_\ell(m) = W_0 - W_0^+ - \mathrm{U_p} - m\omega - q_\ell^2/2 \qquad 1.3.52c$$

Once again we emphasize that $\tau^{(1)}$ is *not* a T-matrix.

If we assume a form for the laser potential in the dipole approximation¶ we can write:

$$H'(t) = \mathrm{U_p} \left(1 + \cos(2\omega t) \right) + \frac{1}{2} \vec{\alpha}_0 \cdot \vec{p} \cos(\omega t) \qquad 1.3.53$$

where $\alpha_0 \stackrel{\text{def}}{=} E/\omega^2$ is a laser parameter which points in the direction of the laser polarization. Therefore, the sum over s only goes from -2 to $+2$.

Note that while there is only one δ-function in Equation 1.3.51 there is a singular denominator of the form:

$$\frac{1}{\bar{E}_2(m) + \mathrm{i}\eta}.$$

We can extract this into a δ-function by using the Dirac identity[11]:

$$\frac{1}{\bar{E}_2(m) - \mathrm{i}\eta} = P \frac{1}{\bar{E}_2(m)} + \mathrm{i}\pi\delta \left(\bar{E}_2(m) \right) \qquad 1.3.54$$

¶ See Mittleman's text[8], §6.3.

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where P here means "Cauchy Princpal Part". It is because of the appearance of this second δ-function that we call this term "sequential", analogously to Equation 1.3.36. Both the δ-function and the Principal Part are equally singular and must be treated carefully when squared. This term clearly represents the process depicted in Figure 1.1. by fixing the energies to be those of the ground-state atom and that of the singly-charged ion.

We have neglected in Equation 1.3.51 the Pauli exclusion principle requirement that the wavefunction be symmetric under interchange of electrons 1 and 2. We can add this in by operating on the wavefunction $\langle\psi_{q_1}^{(-)}\psi_{q_2}^{(-)}|$ with $(1+X_{12})/2$ where X_{12} effects the $q_1 \leftrightarrow q_2$ interchange.

The First-Order Direct Term

There are two terms left in our first-order perturbation theory expansion of the S-matrix. There is the leftover term from Equation 1.3.47 containing V_{12}, Ω_2 and $(\nu/2)\pi_2$. The other term comes from taking the zero-order term in Equation 1.3.20 and the first-order term in Equation 1.3.8. These two terms have only one singular part (*i.e.* δ-function) to each of them and we identify these two terms with the "direct" ionization path depicted in Figure 1.2. We shall show this now.

We make the same assumptions on the form of ψ_q as we did for Equation 1.3.50. The first term, from Equation 1.3.47 is, after some algebra:

$$S^{IId} = -2\pi\mathrm{i} \sum_{n,s,j,k,\ell,m} \delta\left(\frac{q_1^2}{2} + \frac{q_2^2}{2} + 2\mathrm{U_p} + (j+k-\ell+m-s)\,\omega - W_0\right) \times$$
$$\int \frac{\mathrm{d}^3 q}{(2\pi^3)} \frac{M_1 M_2}{W_0 - W_0^+ - q^2/2 - \mathrm{U_p} - (m-s)\omega + \mathrm{i}\eta} \qquad 1.3.55$$

where

$$M_1 \overset{\mathrm{def}}{=} \langle f_{q_1,j}\, f_{q_2,k}| \left(V_{12} - \Omega_2 - \frac{\nu}{2}\pi_2\right) |w_n\, f_{q,\ell}\rangle$$
$$M_2 \overset{\mathrm{def}}{=} \langle w_n\, f_{q,m}|H_s'|u_0\, u_0\rangle$$

We can write this in a more familiar form by identifying as a T-matrix the integral on the second line of the above equation and writing $N = \ell + s - j - k - m$ (the total

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number of photons in the whole process):

$$S^{IId}(q_1, q_2) = -2\pi \mathrm{i} \sum_{N} \delta(\Delta(N))\, T_N^{II}(q_1, q_2)$$

where, as usual, Δ represents overall energy conservation:

$$\Delta(N) = \frac{q_1^2}{2} + \frac{q_2^2}{2} + 2\mathrm{U_p} - N\omega - W_0$$

and

$$T_N^{II}(q_1, q_2) = \sum_{n,s,j,k,m} \int \frac{\mathrm{d}^3 q}{(2\pi)^3} \frac{\langle f_{q_1,j} f_{q_2,k} | (V_{12} - \Omega_2 - \frac{\nu}{2}\pi_2) | w_n f_{q,N+j+k+m-s} \rangle \langle w_n f_{q,m} | H'_s | u_0 u_0 \rangle}{W_0 - W_n^+ - q^2/2 - \mathrm{U_p} - (m-s)\omega + \mathrm{i}\eta} \tag{1.3.56}$$

We recall that in the above sums the indices mean the following:

j, k, m: Sums over Floquet states in the expansion of $\psi^{(-)}$'s.

s: Sum over the modes of the laser field hamiltonian.

n: Sum over intermediate ionic states.

This denominator must be integrated over and that eliminates the singularity which would otherwise be present. So, while the denominator in this term looks similar to the one in Equation 1.3.51, it is made finite here by the q integral.

We do not restrict ourselves to the singly-charged ionic ground state for the following reason: In the rate equations (1.3.44) and in the first-order sequential term (Equation 1.3.51) the ionization process goes through a *real* ionic state. In this direct process, we are summing over all *virtual* ionic states. For the sake of simplicity in the rate equations, we wish to only include one real intermediate singly-charged ionic state. Preliminary calculations show that the higher virtual ionic states do not contribute significantly to the overall process and it would not be inconsistent to restrict the sum over n to just $n = 0$.

This process has been described by Corkum[12]; we term it the "boomerang" or "rescattering" effect. This direct process describes the following events: the ground state atom absorbs some number of photons from the field (via H'_s) and releases one electron leaving the system with one free electron ($f_{q,m}$) and the ion (w_n). The free electron absorbs some number of photons from the field ($f_{q,N+j+k+m-s}$) and recollides with the ion and releases the other electron (via $V_{12} - \Omega_2 - (\nu/2)\pi_2$), the screened e-e interaction.

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The remaining direct term comes from the first-order perturbation to Φ_0, the initial state (Equation 1.3.8) and the zero-order term in the final state:

$$S^{G_0} = -\mathrm{i}\int_{-\infty}^{\infty} \mathrm{d}t\,\langle \psi_{q_1}^{(-)}(t)\psi_{q_2}^{(-)}(t)|H'(t)G_0\delta V|\phi_0(t)\phi_0(t)\rangle \qquad 1.3.57$$

If we replace, once again, the $\psi^{(-)}$ states by Floquet states, insert in the Green's function from Equation 1.3.16, and turn the algebra crank, we can write:

$$\begin{aligned} S^{G_0} &= -2\pi\mathrm{i}\sum_{n,n',N,j,s} \delta\left(\frac{q_1^2}{2}+\frac{q_2^2}{2}+2\mathrm{U_p}-N\omega-W_0\right) \\ &\qquad \times \frac{\langle f_{q_1,j}f_{q_2,s-j-N}|H_s'|u_n u_{n'}\rangle\langle u_n u_{n'}|\delta V|u_0 u_0\rangle}{W_0-W_{n,n'}+\mathrm{i}\eta} \\ &= -2\pi\mathrm{i}\sum_N \delta(\Delta(N))T^{G_0} \end{aligned} \qquad 1.3.58$$

with $W_{n,n'}$ defined after Equation 1.3.16. The last line is the definition of T^{G_0}.

We call sum of the terms $T_N^{II} + T_N^{G_0} = T_N^D$ the *direct T-matrix*.

We must now justify the claim that S^{G_0} and S^{IId} are "direct" terms, and that the terms S^{IIs} and S^I are "sequential" terms.

Section 1.4: Connecting the S-matrix with Rate Parameters

Let us write the whole S-matrix at once.

$$\begin{aligned} S(q_1,q_2) &= \sum_N S_N(q_1,q_2) \\ S_N(q_1,q_2) &= -2\pi\mathrm{i}\delta\left(\Delta\left(N\right)\right)\left[T_N^D(q_1,q_2)+\left(\frac{1+\mathrm{X}_{12}}{2}\right)\sum_{m=1}^{N-1}\frac{\tau_{N,m}^{(1)}}{\bar{E}_2(m)+\mathrm{i}\eta}\right. \\ &\qquad \left. -2\pi\mathrm{i}\sum_{m=1}^{N-1}\delta\left(E_1(m)\right)\tau_{N,m}^{(2)}\right] \end{aligned} \qquad 1.4.1$$

As with the S-matrix for single-ionization, we identify the absolute-square of S as the probabability for double-ionization. Like the single-ionization rate, we must proceed with care when squaring the singular δ-functions and denominators appearing. Once again, we must also integrate over all possible outgoing electron momenta ($\int\int \mathrm{d}^3q_1\,\mathrm{d}^3q_2$) to get the total ionization probability. Therefore, we must be careful not to have any terms which are "too singular" (more than two δ-functions appearing) in our final expression.

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If we naïvely square the expression for S_N, we get:

$$\begin{aligned}|S_N|^2 = \delta(\Delta(N))^2 \Bigg[\left|T_N^D\right|^2 &+ \left(\frac{1+\mathrm{X}_{12}}{2}\right) \sum_{m,m'} \left\{ \frac{\tau^{(1)}_{N,m} \tau^{(1)}_{N,m'}{}^{*}}{(\bar{E}_2(m) + i\eta)(\bar{E}_2(m') - i\eta)} \right. \\ &\left. + \delta(E_1(m))\delta(E_1(m'))\tau^{(2)}_{N,m}\tau^{(2)}_{N,m'}{}^{*} \right\} \\ &+ 2\Re \left\{ T_N^{D*} \sum_m \left(\frac{\tau^{(1)}_{N,m}}{\bar{E}_2(m)} + \delta(E_1(m))\tau^{(2)}_m \right) \right. \\ &\left. + \sum_{m,m'} \frac{\tau^{(1)}_m}{\bar{E}_2(m)} \tau^{(2)}_{m'}{}^{*} \delta(E_1(m')) \right\} \Bigg] \end{aligned} \tag{1.4.2}$$

We will assume that the cross-term between $\tau^{(1)}$ and $\tau^{(2)}$ (the last line above) does not contribute if there are no "accidental resonances" between the first ionized state and the HF state. That is, we assume that the two singular functions $1/\bar{E}_2$ and $\delta(E_1)$ have no overlap. Since $\bar{E}_\ell(m) = q_\ell^2/2 + \mathrm{U_p} + W_0^+ - m\omega$ and $E_\ell(m') = q_\ell^2/2 + \mathrm{U_p} + W_0/2 - m'\omega$, unless $W_0/2$ becomes coupled to W_0^+ by the laser, we will have no overlap and this term will vanish. We must turn to the multiple singularities present. We show now that our S-matrix is not too singular.

First, let us consider the first term in $|S|^2$: $\delta(\Delta(N))^2|T_N^D|^2$. We can use Equation 1.2.13 to reduce this term to:

$$|S_N|^2 = 2\pi T \delta(\Delta(N)) \left|T_D^N(q_1, q_2)\right|^2 + \cdots \tag{1.4.3a}$$

We can likewise deal with the $\left|\tau^{(2)}\right|^2$ term, We see that there is no interference between $m \neq m'$ terms in the sum. This is because these two sharply-peaking functions will have negligible overlap when their arguments are not identical. The result for this term is:

$$|S_N|^2 = \cdots + (2\pi T)^2 \delta(\Delta(N)) \sum_{m=1}^{N-1} \delta(E_1(m)) \left|\tau^{(2)}_{N,m}(q_1, q_2)\right|^2 + \cdots \tag{1.4.3b}$$

where one factor of $(2\pi T)$ (as in Equation 1.2.10) comes from the square of each of the independent δ-functions.

The term in $|\tau^{(1)}|^2$ is the hardest term with which we deal. We note first that the operator $(1 + \mathrm{X}_{12})/2$ is idempotent. We must first calculate:

$$\sum_{m,m'} \frac{\tau^{(1)}_{N,m} \tau^{(1)}_{N,m'}{}^{*}}{(\bar{E}_2(m) + i\eta)(\bar{E}_2(m') - i\eta)}. \tag{1.4.4a}$$

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We first identify η with a state-width. This is the usual association of the positive infinitesimal in scattering theory†. Therefore, we can write $\eta = 1/T$, were T is a time interval. This time interval is the same one as in Equation 1.2.13.

We can imagine that the individual terms E_2 are entering not into integrals d^3q but integrals over E itself. With this in mind, scale E_2 by T to get a dimensionless variable of integration $x_2 = E_2 \times T$. The two denominators become $x_2 + \mathrm{i}$ and $x_2 + \mathrm{i} + \delta m\omega T$, werhe $\delta m = m - m'$. We are then left performing an integral of the form

$$\int_{-\infty}^{\infty} \frac{\mathrm{d}x_2}{T} \sum_{m,m'} \frac{T^2}{(x_2^2+1) + \delta m\omega T(x_2+\mathrm{i})}. \qquad 1.4.4b$$

If we rationalize the denominator to make it even in T, The result is:

$$\int_{-\infty}^{\infty} \mathrm{d}x_2 \sum_{m,m'} \frac{T\left(x_2^2+1-\delta m\omega T\left(x_2+\mathrm{i}\right)\right)}{(x_2^2+1)^2-(\delta m\omega T)^2(x_2+\mathrm{i})^2}. \qquad 1.4.4c$$

In this form it is clear that in the sum over m, m' the term odd in δm will vanish, by cancellation. In the long time $\omega T \rightarrow \infty$ limit (forced by Equations 1.2.12 *et seq.*), the terms even in δm will be of the form:

$$\int_{-\infty}^{\infty} \mathrm{d}x_2 \frac{T(x_2^2+1)}{(x_2^2+1)^2-(\delta m\omega T)^2(x_2+\mathrm{i})^2},$$

which in in the $\omega T \rightarrow \infty$ limit vanishes like $O\left((\log T)/T\right)$, which is too small to keep. What is left then is the $\delta m = 0$ term. The x_2 integral is now trivial and when we integrate over the other δ-function in the large-T limit we finally derive:

$$= \cdots + 2\pi^2 T^2 \delta(\bar{E}_2(m))\delta(\Delta(N))\delta_{m,m'} + \cdots \qquad 1.4.4d$$

This is not the only term of this form which arises. X_{12} operating in Equation 1.4.2 gives rise to a term like:

$$\sum_{m,m'} \frac{\delta^2(\Delta(N))}{(\bar{E}_1(m)+\mathrm{i}\eta)(\bar{E}_2(m')-\mathrm{i}\eta)}, \qquad 1.4.5a$$

which can be treated in a manner similar to Equations 1.4.4. Once again, we associate η with $1/T$ and note that $\Delta(N) = \bar{E}_1(m) + \bar{E}_2(N-m)$. We write $\bar{E}_2(m') = \bar{E}_2(N-m) - \delta n\omega$, and scale our two integrals (over Δ and over $\bar{E}$) by T to get dimensionless

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† See Landau & Lifshitz[7], §43.

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parameters x_1, x_2. Then our overall integral, after rationalizing the denominator as in Equation 1.4.4c turns into:

$$= \cdots \sum_{m,\delta n} \iint dx_1 dx_2 \left(\frac{\sin\left(\frac{x_1+x_2}{2}\right)}{\frac{x_1+x_2}{2}} \right)^2 \frac{T^2[x_1x_2 + 1 + i(x_2 - x_1 + \delta n\omega T)]}{[x_1x_2 + 1 + i(x_2 - x_1)]^2 + (\delta n\omega T)^2} \qquad 1.4.5b$$

The same argument which eliminated the term odd in δm in Equations 1.4.4 can be used to justify eliminating the term odd in δn here. Likewise, in the $\omega T \rightarrow \infty$ limit, the non-zero terms even in δn will vanish. What remains then in the $\delta n = 0$ term, which is

$$= \cdots \sum_{m} \delta_{\delta n,0} \iint dx_1 dx_2 \left(\frac{\sin\left(\frac{x_1+x_2}{2}\right)}{\frac{x_1+x_2}{2}} \right)^2 \frac{T^2[x_1x_2 + 1 - i(x_2 - x_1)]}{[x_1x_2 + 1]^2 + (x_2 - x_1)^2} \qquad 1.4.5c$$

This term looks very nasty but can be simplified with the transformation $y = x_1 + x_2, 2z = x_1 - x_2$. In this form, the resulting integral over z vanishes.

The result of all this is the square of the S-matrix:

$$\begin{aligned} |S_N(q_1q_2)|^2 = 2\pi T\delta(\Delta(N)) \Bigg\{ & \left|T_N^D(q_1, q_2)\right|^2 \\ & + 2\pi T \sum_{m=1}^{N-1} \Im \left[\left(\left(\frac{1 + X_{12}}{2} \right) \delta(\bar{E}_2(m))\tau_{N,m}^{(1)} + \delta(E_1(m))\tau_{N,m}^{(2)} \right) T_N^{D^*} \right] \\ & + 2\pi T \sum_{m=1}^{N-1} \left[\left(\frac{1 + X_{12}}{2} \right) \delta(\bar{E}_2(m)) \left|\tau_{N,m}^{(1)}\right|^2 + \delta(E_1(m)) \left|\tau_{N,m}^{(2)}\right|^2 \right] \Bigg\} \end{aligned} \qquad 1.4.6$$

We notice that there are two types of time dependence here: terms dependent on T and terms dependent on T^2, and each of these terms is identified with either a *single singularity (δ-function)* for T or a *pair of singularities (δ-functions)* for T^2.

Let us now turn to the rate equations, Equations 1.3.44. These coupled equations are easily solvable, given the initial conditions $n_0(t = 0) = n$, and $n_1(t = 0) = \bar{n}(t = 0) = n_2(t = 0) = 0$, and the assumption that all the γ's are constant in time:

$$\begin{aligned} n_2(t) &= n \left(1 - e^{-\Gamma t} - \frac{\bar{\gamma}}{\gamma_{01} + \gamma_{02}} \left(e^{-\bar{\gamma} t} - e^{-\Gamma t} \right) - \frac{\gamma_{01}}{\Gamma - \gamma_{12}} \left(e^{-\gamma_{12} t} - e^{-\Gamma t} \right) \right) \\ \bar{n}(t) &= n \frac{\bar{\gamma}}{\gamma_{02} + \gamma_1} \left(e^{-\bar{\gamma} t} - e^{-\Gamma t} \right) \\ n_1(t) &= n \frac{\gamma_{01}}{\Gamma - \gamma_{12}} \left(e^{-\gamma_{12} t} - e^{-\Gamma t} \right) \\ n_0(t) &= n\, e^{-\Gamma t} \end{aligned} \qquad 1.4.7$$

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Here $\Gamma \stackrel{\mathrm{def}}{=} \gamma_{01} + \gamma_{02} + \bar{\gamma}$ is used as a shorthand notation. The yield of doubly-ionized atoms as a function of time for small time t is simply the Taylor-series expansion of the instantaneous rate $\dot{n}_2(t)/n_0(t)$ (cf. Equation 1.2.16):

$$\dot{n}_2(t)/n_0(t) = \gamma_{02} + t\left[\bar{\gamma}^2 + \gamma_{01}\gamma_{12}\right] + \cdots \qquad 1.4.8$$

We identify the order-T term in Equation 1.4.6 with the order-t term in Equation 1.4.8, yielding:

$$\gamma_{02} = \sum_N \int \frac{\mathrm{d}^3q_1\mathrm{d}^3q_2}{(2\pi)^5}\delta(\Delta(N)) \left\{ \left|T_N^D(q_1,q_2)\right|^2 + 2\pi\Im T_N^D(q_1,q_2)^2 \times \right.$$
$$\left. \sum_{m=1}^{N-1}\left(\left(\frac{1+\mathrm{X}_{12}}{2}\right)\delta(\bar{E}_2(m))\tau_{N,m}^{(1)}(q_1,q_2) + 2\delta(E_1(m))\tau_{N,m}^{(2)}(q_1,q_2)\right)\right\} \qquad 1.4.9$$

We can likewise interpret the part of the T^2 term involving $\tau^{(2)}$ with the $\bar{\gamma}^2$ term, because $\tau^{(2)}$ represents a pair of transitions through the state $\bar{X}$:

$$\bar{\gamma}^2 = \sum_N \sum_{m=1}^{N-1} \int \frac{\mathrm{d}^3q_1\mathrm{d}^3q_2}{(2\pi)^4}\delta(\Delta(N))\delta(E_1(m))\left|\tau_{N,m}^{(2)}\right|^2, \qquad 1.4.10$$

and the regular "sequential" term $\gamma_{01}\gamma_{12}$ with the term involving $\tau^{(1)}$, which involves the ionic ground state w_0:

$$\gamma_{01}\gamma_{12} = \sum_N \sum_{m=1}^{N-1} \int \frac{\mathrm{d}^3q_1\mathrm{d}^3q_2}{2(2\pi)^4}\delta(\Delta(N))\left(\frac{1+\mathrm{X}_{12}}{2}\right)\delta(\bar{E}_2(m))\left|\tau_{N,m}^{(1)}(q_1,q_2)\right|^2. \qquad 1.4.11$$

These three equations are not sufficient to uniquely determine the four unknown rates. We can, however, determine from the yield of singly-charged ions the value of γ_{01}:

$$\dot{n}_1(t) = n\gamma_{01}t + \cdots \qquad 1.4.12$$

However, this requires constructing the S-matrix for single-ionization of helium, which we have not yet done.

Digression: Single-ionization of Helium

Let us look at the single ionization rate into the ionic ground state inferred from Equation 1.3.7, using the lowest-order terms in the perturbation series for the final state $\langle\Psi^{(-)}| \to \langle w_q^{(-)} w_0|$ and the initial state $|\Phi_0\rangle \to |u_0u_0\rangle$. Now, calculate in the

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same manner that led to Equation 1.3.36, still using the HF-Volkov representation for the unbound electron state. We get the single-ionization S-matrix:

$$\begin{aligned} S &= -2\pi \mathrm{i} \sum_n \delta\left(\frac{q^2}{2} + W_0^+ + U_p - W_0 - n\omega\right) T_{\mathrm{sing}}(q) \\ T_{\mathrm{sing}}(q) &= \sum_{s=-2}^{2} \left[\langle f_{q,s-n}|h'_s|u_0\rangle\langle w_0|u_0\rangle + \langle f_{q,s-n}|u_0\rangle\langle w_0|h'_s|u_0\rangle\right] \end{aligned} \qquad 1.4.13$$

When we square this, treating the square of the δ-function in our canonical fashion, we may write:

$$|S|^2 = 2\pi T \sum_n \delta\left(\frac{q^2}{2} + W_0^+ + U_p - n\omega - W_0\right) |T_{\mathrm{sing}}(q)|^2 . \qquad 1.4.14$$

This representation begs to be compared with the Single Active Electron (SAE) model, where all the electrons in an atom except for one are frozen into the initial configuration and their net effect is approximated by screening the nuclear Coulomb potential[13]. In such a situation, the T-matrix appearing in Equation 1.4.13 would have only the first term in it. The second term, where one electron is excited into an ionic state, and the other electron "suddenly" finds itself in a continuum state as the atom relaxes. (This is termed "shakeoff".) Some preliminary calculations show that these two terms are roughly the same order of magnitude. (See Table 2.2.)

Section 1.5: On the Separation between Direct and Sequential

There is a problem with the description of the various processes we gave in Figures 1.1, 1.2 and 1.4. If one of the ionization rates $\gamma_{01}\gamma_{02}$ or $\bar{\gamma}^2$ is exteremely rapid compared to γ_{02} then that "sequential" process cannot be distinguished from the "direct" process. Therefore our separation of processes into "sequential" and "direct" is very tentative.

We would like to be able to experimentally distinguish between the direct and sequential processes. The most straightforward (although most difficult) would be energy or angle correlation measurements. We can extract (*e.g.*) angular distributions from the rates, like $\gamma_{02}(\theta_1, \varphi_1; \theta_2, \varphi_2)$, where θ_i, φ_i are the directions of electron i, by replacing the 6-d integral in Equation 1.4.9 with just $\int\int dq_1\, q_1^2\, dq_2\, q_2^2$. One would then set up a triple-coincidence experiment, measuring an incoming laser pulse and

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two outgoing electrons. The results would be compared to the rates as a function of angle.

We can, in principle, devise an alternative method to measure the effects of the direct process as follows: Define a *turnover time* t_t as:

$$t_t = \frac{\gamma_{02}}{\gamma_{01}\gamma_{12} + \bar{\gamma}^2}. \qquad 1.5.1$$

This term arises from an analysis of Equation 1.4.8. For times smaller than t_t, the dominant rate into X^{++} will be γ_{02}, but for times after t_t, the sequential terms will dominate. This, however, requires that the turnover time be longer than current laser pulse lengths. As we shall see in the Chapter 2, it at least one restricted regime this is not the case.

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Chapter 2

Further approximations in the Theory of Double Ionization Yield of Helium

In Chapter 1, we developed a formal theory of double-ionization for heliumlike atoms based upon an S-matrix theory and an expansion in the correlation interaction. The singularities of the S-matrix were linked (Equation 1.4.6) to the various measurable rate parameters. We do this here in the "strong field approximation"[14].

Section 2.1: Introduction

In the last chapter we derived formulæ which connect the S-matrix (microscopic) description of double-ionization to experimentally-measurable quantities, such as the yield rate of doubly-charged ions. One could also make a connection by numerically solving the Schrödinger equation for the two-electron system[2]. This method can provide double-ionization yield curves. However, there are several drawbacks to this method. It is a numerically gargantuan task which currently requires many hours of supercomputer time. As well, the solution is dependent on the laser intensity profile. Should that change, the calculation must be repeated. The method derived in the previous chapter can, also, in the regime of a strong-field approximation, give ionization rates as a function of intensity. Under stringent conditions on laser frequency, this method provides *analytic* results. In order to do this we must use several approximations. The first is that we use the first Born approximation (FBA) for the S-matrix. This theory was also used by Reiss[15] where he called it the "Strong Field

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Approximation (SFA)". This approximation requires at the outset that[16]:

$$B \ll U_p \qquad 2.1.1$$

where B is a binding energy in the problem. (In this case, $B = 2.9 = -W_0$ is the ground-state potential of He.)

A Reminder:

Throughout this chapter we shall use the same states as defined in Chapter 1, *e.g.* $\psi^{(-)}$, u_0, w_n, etc. We introduce the additional notation that χ_q is an electron in a Volkov state with momentum q, and λ_q is a plane-wave state carrying momentum q: $\lambda_q = \exp(i\vec{q}\cdot\vec{r})$.

Section 2.2: The Sequential Processes

We begin by calculating the simplest sequential ionization rate $\bar{\gamma}$, as given by Equation 1.4.10, or, more simply, by:

$$\bar{\gamma} = \sum_N \int \frac{d^3q}{(2\pi)^2}\delta(\frac{q^2}{2} + U_p + \frac{W_0}{2} - N\omega)\left|T_N^{(1)}(q)\right|^2, \qquad 2.2.1$$

where we have taken the definition of E_1 from Equation 1.3.41, and of $T^{(1)}$ from Equation 1.3.40. In going from Equation 1.3.36 to this one we have used the fact that the terms with different n_1 (as given in Equation 1.4.10) do not interfere. This is the ionization through the "HF" state $\bar{X}$ in Figure 1.4. Its eigenenergy is $W_0/2$, with W_0 as defined above. We will write $\bar{B} = -W_0/2 = 1.45$ as a shorthand notation.

At this point we must finally write down an explicit for for the final states $\psi_q^{(-)}$. We assume the final states are approximated by the χ_q. From Equation 1.3.49, and the Volkov state expression (1.3.37) we have the expansion[8]:

$$f_{q,j} = \lambda_q \int_{-\pi}^{\pi} \frac{d\phi}{2\pi} e^{-i(n\phi + \vec{\alpha}_0\cdot\vec{q}\sin\phi + \frac{U_p}{2\omega}\sin 2\phi)}. \qquad 2.2.2$$

$\vec{\alpha}_0 = \vec{E}/\omega^2$ here, as we have previously mentioned, is a length parameter characterizing the laser. We recognize the integral as a representation of the Generalized Bessel Function. This gives us:

$$f_{q,j} = \lambda_q \mathcal{J}_{-j}(\vec{\alpha}_0\cdot\vec{q}, U_p/2\omega) \qquad 2.2.3$$

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We now write down our T-matrix from Equation 1.3.39 as:

$$T_n^{(1)}(q) = \langle\lambda_q|u_0\rangle \left[\mathrm{U_p}\, \mathcal{J}_{-n}(\vec{\alpha}_0 \cdot \vec{q}, \mathrm{U_p}/2\omega) + \frac{\omega}{2}(\vec{\alpha}_0 \cdot \vec{q})\,(\mathcal{J}_{-n-1} + \mathcal{J}_{-n+1}) + U_p/2\,(\mathcal{J}_{-n-2} + \mathcal{J}_{-n+2})\right]. \qquad 2.2.4$$

(We have suppressed the arguments to the GBF in the second and third terms for brevity.) Using the recurrence relation (Equation A2.2) we can rewrite this as:

$$T_n^{(1)}(q) = \langle\lambda_q|u_0\rangle\,(\mathrm{U_p} - n\omega)\,\mathcal{J}_{-n}(\vec{\alpha}_0 \cdot \vec{q}, \mathrm{U_p}/2\omega) \qquad 2.2.5$$

The other sequential process is the standard two-step process via the 1s state of He$^+$. The rate of population of X^+, γ_{01}, is given by Equation 1.4.13. That is, one electron is ionized from an uncorrelated initial state and the other electron is left in the 1s state of the singly-charged ion. When we approximate (as we have already done) the final states by Volkov states, we get:

$$\gamma_{01} = \sum_{n,s} \int \frac{\mathrm{d}^3q}{(2\pi)^2}\,\delta(\frac{q^2}{2} + \mathrm{U_p} + B_2 - n\omega)\,\left|\langle w_0 f_{s-n}|H_s'|u_0u_0\rangle\right|^2. \qquad 2.2.6a$$

Here, B_2 is the energy required to raise the atom to its ground ionized state:

$$B_2 = W_0^+ - W_0 = 0.9. \qquad 2.2.7a$$

γ_{12} is, in the same vein, the second electron being ionized via the electron-electron interaction from the 1s state of the ion, and its term comes from the other half of Equation 1.4.11:

$$\gamma_{12} = \sum_{n,s} \int \frac{\mathrm{d}^3q}{(2\pi)^3}\,\delta(\frac{q^2}{2} + \mathrm{U_p} + B_1 - n\omega)\left|\langle f_{q,s-n}|\left(\frac{\nu}{2}\pi - \Omega\right)|w_0\rangle\right|^2. \qquad 2.2.8a$$

Once again, B_1 is the energy required to ionize the ion:

$$B_1 = -W_0^+ = 2. \qquad 2.2.7b$$

One can verify immediately that the product of these two terms gives $\tau^{(1)}$ from Equation 1.3.52. Once again, using the recurrence relation in Equation A2.2 one may rewrite these terms in the more familiar form:

$$\gamma_{01} = \sum_n \int \frac{\mathrm{d}^3q}{(2\pi)^3}\,\delta(\frac{q^2}{2} + \mathrm{U_p} + B_2 - n\omega)\,|\langle\lambda_q|u_0\rangle\langle w_0|u_0\rangle|^2 \times \left[2(\mathrm{U_p} - n\omega)\mathcal{J}_{-n} - \frac{\omega}{2}\vec{\alpha}_0 \cdot \vec{q}\,(\mathcal{J}_{-n-1} + \mathcal{J}_{-n+1})\right]^2 \qquad 2.2.6b$$

$$\gamma_{12} = \sum_n \int \frac{\mathrm{d}^3q}{(4\pi)^2}\,\delta(\frac{q^2}{2} + \mathrm{U_p} + B_1 - n\omega) \times \left|\langle\lambda_q|\left(\frac{\nu}{2}\pi - \Omega\right)|w_0\rangle\right|^2 \mathcal{J}_{-n}^2 \qquad 2.2.8b$$

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where the arguments in all the GBFs are $(\vec{\alpha}_0 \cdot \vec{q}, U_p/2\omega)$.

Section 2.3: The Direct Processes

The direct processes stem from Equation 1.4.9. One term arises from initial state correlation, and one from final state correlation (the "Corkum term"). These two terms add coherently.

The initial-state correlation term, T^{G_0} (from Equation 1.3.58), gives:

$$T^{G_0} = \sum_{\mu,\nu,j,j',s} \frac{\langle f_{q_1,j} f_{q_2,j'} | H_s' | \mu\nu \rangle \langle \mu\nu | \delta V | u_0 u_0 \rangle}{W_0 - W_{\mu\nu}} \qquad 2.3.1$$

where δV is defined by Equation 1.3.14 and the roman indices have the relation that $N = s - j - j'$ renames one of the summations. μ and ν here label states which satisfy the same hamiltonian as u_0†. (We will sum over N later.) An approximate expression for the two-electron final state is:

$$\chi_{q_1} \chi_{q_2} = \lambda_{q_1} \lambda_{q_2} \exp(\mathrm{i}t[q_1^2/2 + q_2^2/2] + \mathrm{i}(U_p/\omega)(1 + \sin 2\omega t) + \mathrm{i}\vec{\alpha}_0 \cdot \vec{Q} \sin \omega t) \qquad 2.3.2$$

with $\vec{Q} = \vec{q}_1 + \vec{q}_2$. To convert this into the f-functions, we integrate as given by Equation 1.3.49 *twice*:

$$\begin{aligned} f_{q_1,j} f_{q_2,k} &= \lambda_{q_1} \lambda_{q_2} \sum_{j,k} \\ &\int_{-\pi}^{\pi} \int_{-\pi}^{\pi} \frac{\mathrm{d}\phi_1 \mathrm{d}\phi_2}{(2\pi)^2} \mathrm{e}^{-\mathrm{i}j\phi_1 - \mathrm{i}k\phi_1} \mathrm{e}^{\mathrm{i}(q_1^2/2 + q_2^2/2)t + \mathrm{i}tU_p/\omega} \chi_{q_1} \chi_{q_2} \qquad 2.3.3 \\ &= \lambda_{q_1} \lambda_{q_2} \mathcal{J}_{-j-k}(\vec{\alpha}_0 \cdot \vec{Q}, U_p/\omega) \end{aligned}$$

The last step comes since the expression in Equation 2.3.2 gives a term which is independent of ϕ_2.

Applying the definition of $f_{q,j}$, we simplify the expression in Equation 2.3.1:

$$T^{G_0} = \sum_{\mu,\nu} \frac{\langle \lambda_{q_1} \lambda_{q_2} | \mu\nu \rangle \langle \mu\nu | \delta V | u_0 u_0 \rangle}{W_0 - W_{\mu\nu}} (2U_p - N\omega) \mathcal{J}_{-N}(\vec{\alpha}_0 \cdot \vec{Q}, U_p/\omega), \qquad 2.3.1a$$

where in passing from Equations 2.3.1 to 2.3.1a we have made liberal use of the GBF recurrence relations again.

The final-state correlation term, T_N^{II}, from Equation 1.3.56, yields under a similar analysis (Recall the definition of N here from below Equation 1.3.55):

---

† The sum over μ, ν is a sum over the discrete eigenstates and an integral of the continuous eigenstates of the hamiltonian in Equation 1.3.12

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$$T_N^{II}(q_1, q_2) = \sum_{n,s,j,k,m} \int \frac{\mathrm{d}^3q}{(2\pi)^3} \times$$
$$\frac{\langle f_{q_1,j} f_{q_2,k} | (V_{12} - \Omega_2 - \frac{\nu}{2}\pi_2) | w_n f_{q,N+j+k+m-s} \rangle \langle w_n f_{q,m} | H_s' | u_0 u_0 \rangle}{W_0 - W_n^+ - q^2/2 - \mathrm{U_p} - (m - s)\omega + \mathrm{i}\eta} \quad 1.3.56$$

If we replace the $f_{q,j}$'s by their Volkov approximations, as usual:

$$= \sum_{\nu,s,n} \int \frac{\mathrm{d}^3q}{(2\pi)^3} \frac{\mathcal{J}_{\nu-N}(\vec{\alpha}_0 \cdot (\vec{Q} - \vec{q}), \mathrm{U_p}/2\omega)\, \mathcal{J}_{s-\nu}(\vec{\alpha}_0 \cdot \vec{q}, \mathrm{U_p}/2\omega)}{W_0 - W_0^+ - q^2/2 - \mathrm{U_p} - \nu\omega + \mathrm{i}\eta} \times$$
$$\left(\frac{1 + \mathrm{X}_{12}}{2}\right) \langle \lambda_{q_1} \lambda_{q_2} | (V_{12} - \Omega_2 - \frac{\nu}{2}\pi_2) | w_n \lambda_q \rangle \langle w_n \lambda_q | H_s' | u_0 u_0 \rangle \quad 2.3.4$$

where we have defined $\nu = N - j - k$ as a new dummy index§. We have written the first matrix element's GBFs using algebra which is a generalization of what we used in Equation 2.3.2.

We can proceed no further until we have a method of working with the GBFs, either analytical or numerical. We choose to approximate the GBFs analytically. This wise places strict bounds on both the laser frequency and intensity, as we shall see in the next section.

Section 2.4: Analytical Approximation of the T-matrices

The GBFs offer no analytic simplifications. As well, Leubner[24] has shown that even evaluating them numerically is not an easy task. Given these, we choose to evaluate the GBFs using analytic approximations. These approximations, however, require stringent requirements upon frequency and intensity.

Our ability to make our approximation stems from the appearance of δ-functions along with every GBF. The δ-function appearing controls the arguments of the GBF, and we exploit this.

Write the GBF as:

$$\mathcal{J}_{-N}(a, b) = \int_{-\pi}^{\pi} \frac{\mathrm{d}\phi}{2\pi} \mathrm{e}^{\mathrm{i}\Phi(\phi)} \quad 2.4.1$$

where $a = \vec{\alpha}_0 \cdot \vec{q}$ (in the usual case) and $b = \mathrm{U_p}/(2\omega)$ (also in the usual case). For typical experimental régimes Φ is a rapidly varying function of ϕ. A term like this will usually appear integrated against a δ-function of the type $\delta(q^2/2 + \mathrm{U_p} + B - N\omega)$

---

§ The indices j, k, m, etc., are defined in Equation 1.3.56.

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where N is interpreted as the number of photons transferred in a process. B here is a binding energy appropriate to the particular equation.

The GBF does not allow for any real stationary phase points (SPPs) but does admit two complex saddle points where the saddle-point method† of approximate integration may be used.

The saddle-point method of approximating an integral requires the vanishing of the first-derivative in a Taylor series to simplify integrals of the form $\int \exp(\alpha f(x))\,dx$, where α is very large:

$$\Phi(\phi) = \Phi(\zeta) + (\zeta - \phi)\Phi'(\zeta) + \frac{1}{2}(\zeta - \phi)^2\Phi''(\zeta) + \ldots \qquad 2.4.2$$

The saddle points occur when $\Phi'(\zeta) = 0$. (The prime here has the usual meaning of differentiation with respect to the argument.) This gives a condition on ζ:

$$N + a\cos\zeta + 2b\cos 2\zeta = 0, \qquad 2.4.3a$$

which is a condition on the saddle point(s) in terms of $\cos\zeta$:

$$\cos\zeta = \alpha \pm i\beta \qquad 2.4.3b$$

$$\alpha = \frac{a}{8b} \qquad 2.4.3c$$

$$\beta = \sqrt{\frac{N}{4b} - \frac{1}{2} - \left(\frac{a}{8b}\right)^2} = \sqrt{\frac{B + \epsilon_\perp}{2U_p}} \qquad 2.4.3d$$

where $\epsilon_\perp = \vec{q}_\perp^{\,2}/2$, and $\vec{q}_\perp$ is that part of the electon momentum perpendicular to $\vec{\alpha}_0$ (and the laser field). In the same way, $\vec{q}_\parallel$ is that part of the electron's momentum parallel to the field. In Equation 2.4.3d we have used the δ-function appearing with the GBF to rewrite the radicand in a more useful form. Using these definitions: $2\epsilon_q = q_\perp^2 + q_\parallel^2$ is the total electron energy.

The integral in Equation 2.4.1 becomes:

$$\mathcal{J}_{-N}(a,b) = e^{-i\Phi(\zeta)} \int_C d\phi\, e^{-i\Phi''(\zeta)(\zeta-\phi)^2/2} \qquad 2.4.4$$

where C is the deformed contour. Note that this requires that $\Im\Phi''(\zeta) > 0$ or the integral will diverge. We let the limits of ϕ go to $\pm\infty$. We shall see that this is

---

† Compare this method with the same one used to find asymptotic approximations to the regular Bessel function. See Morse & Feshbach[17] §5.3 for details.

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permissable in view of the expansion we shall perform later. Doing this allows us to evaluate the integral above. There are two saddle points which are complex conjugates which we must add. We therefore have:

$$\mathcal{J}_{-N}(a,b) = \frac{e^{-i\Phi(\zeta_1)}}{\sqrt{2\pi\Phi''(\zeta_1)}} + \frac{e^{-i\Phi(\zeta_2)}}{\sqrt{2\pi\Phi''(\zeta_2)}} \tag{2.4.5}$$

where $\zeta_{1,2} = i\eta \pm \xi$. These imply that

$$\alpha = \cos\xi \cosh\eta \tag{2.4.6a}$$

$$\beta = \sin\xi \sinh\eta. \tag{2.4.6b}$$

Also let $\chi = \arg\Phi''(\zeta)$. Our GBF can now be expressed in terms of the real and imaginary parts of Φ, using its symmetry properties under complex conjugation[†]:

$$\mathcal{J}_{-N}(a,b) = 2\frac{e^{-\Im\Phi(\zeta_1)}}{\sqrt{4\pi|\Phi''(\zeta_1)|}}\cos(\Re\,\Phi(\zeta_1) - \frac{\chi}{2}) \tag{2.4.7}$$

If we restrict ourselves to a low-frequency theory, such that ω is small compared to B, we can expand Φ in terms of $\epsilon_\perp$ and $\epsilon_\parallel$ and keep only first-order terms in them. We show how to proceed to do this.

Let us first write down an explicit form for $\Phi(\zeta)$. We examine the real and imaginary parts separately. From Equation 2.4.3 we have:

$$\Im\,\Phi(\zeta) = -N\eta - (a + 2b\alpha)\sinh\eta\cos\xi + 2b\beta\sin\xi\cosh\eta \tag{2.4.8}$$

Using Equation 2.4.6 we write:

$$\Im\,\Phi(\zeta)/b = (-\frac{N}{b}\eta) + \frac{6\beta\alpha^2}{\alpha\tan\xi} + 2\beta\alpha\tan\xi. \tag{2.4.9}$$

We wish to find an equation for $\alpha\tan\xi$. We use the regular trigonometric identities for sinh and cosh and we also note that $\sin^2 = \tan^2/(1+\tan^2)$. From this and Equation 2.4.6 we state:

$$\begin{aligned} &\alpha^2\tan^2\xi - \sin^2\xi = \beta^2 \\ &\qquad \to \alpha^2\tan^4\xi + (\alpha^2 - \beta^2 - 1)\tan^2\xi - \beta^2 = 0. \end{aligned} \tag{2.4.10}$$

This is a quadratic equation in $\tan^2\xi = \mathcal{Z}$. The solution to this is simple:

$$\mathcal{Z} = \frac{1}{2\alpha^2}\left(1 + \beta^2 - \alpha^2 \pm \sqrt{(1 + \beta^2 + \alpha^2)^2 - 4\alpha^2}\right) \tag{2.4.11}$$

[†] We have $\Phi(\zeta^*) = \Phi(\zeta)^*$, since $\sin(\zeta^*) = (\sin(\zeta))^*$

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Let us write

$$\beta = \beta_m\sqrt{1 + \frac{\epsilon_\perp}{2\mathrm{U_p}}\frac{2\mathrm{U_p}}{B}} \approx \beta_m\left(1 + \frac{\epsilon_\perp}{2B}\right) \qquad 2.4.12$$

where $\beta_m^2 = B/(2\mathrm{U_p})$. Also note that $\alpha^2 = \epsilon_\parallel/(2\mathrm{U_p})$. We introduce the shorthand $\mathcal{Y}$ for the common term $1 + \beta_m^2$. Our expression for $\alpha^2 \tan^2\xi$ becomes:

$$\begin{aligned}
\alpha^2 \tan^2\xi &= \frac{1}{2}\left(\mathcal{Y} + \frac{\epsilon_\perp}{2\mathrm{U_p}} - \frac{\epsilon_\parallel}{2\mathrm{U_p}} \pm \sqrt{\left(\mathcal{Y} + \frac{\epsilon_\perp}{2\mathrm{U_p}} + \frac{\epsilon_\parallel}{2\mathrm{U_p}}\right)^2 - 4\frac{\epsilon_\parallel}{2\mathrm{U_p}}}\right)\\
&= \frac{1}{2}\left(\mathcal{Y} + \frac{\epsilon_\perp}{2\mathrm{U_p}} - \frac{\epsilon_\parallel}{2\mathrm{U_p}} \pm \mathcal{Y}\sqrt{\left(1 + \frac{\epsilon_\perp}{2\mathrm{U_p}\mathcal{Y}} + \frac{\epsilon_\parallel}{2\mathrm{U_p}\mathcal{Y}}\right)^2 - \frac{2\epsilon_\parallel}{\mathrm{U_p}\mathcal{Y}^2}}\right) \qquad 2.4.13\\
&\to \frac{1}{2}\left(\mathcal{Y} + \frac{\epsilon_\perp}{2\mathrm{U_p}} - \frac{\epsilon_\parallel}{2\mathrm{U_p}} \pm \mathcal{Y}\left[1 + \frac{\epsilon_\perp}{2\mathrm{U_p}\mathcal{Y}} + \frac{\epsilon_\parallel}{2\mathrm{U_p}\mathcal{Y}} - \frac{\epsilon_\parallel}{\mathrm{U_p}\mathcal{Y}^2}\right]\right)
\end{aligned}$$

This gives for $\alpha \tan\xi$:

$$\to \sqrt{\mathcal{Y}}\left(1 + \frac{1}{2\mathcal{Y}}\frac{\epsilon_\perp}{2\mathrm{U_p}} - \frac{1}{2\mathcal{Y}^2}\frac{\epsilon_\parallel}{2\mathrm{U_p}}\right)$$

If we similarly expand the terms β and α in Equation 2.4.9, and keep only terms first order in the ϵ's we get:

$$\Im\,\Phi(\zeta)/b = -\frac{N}{b}\eta + \frac{6\beta_m}{\sqrt{\mathcal{Y}}}\frac{\epsilon_\parallel}{2\mathrm{U_p}} + 2\sqrt{\mathcal{Y}}\beta_m^2 + 2\sqrt{\mathcal{Y}}\frac{\epsilon_\perp}{2\mathrm{U_p}}\left(\frac{\beta_m}{2\mathcal{Y}}\right) - 2\beta_m\sqrt{\mathcal{Y}}\frac{\epsilon_\perp}{2\mathrm{U_p}}\frac{1}{2\mathcal{Y}^2} \qquad 2.4.14$$

We must still find an expression for η. We recall that $\tanh\eta = \beta/(\alpha\tan\xi)$, the components of which we already know. We use the expression for $\tanh x$:

$$\tanh x = \frac{1}{2}\log\frac{1+x}{1-x}.$$

The algebra turning η into a function of the ϵ's is torturous and fraught with the possibilities for minus-sign errors. We must also find an expansion of $N/(2b)$. To do this, remember that these terms all enter into an integral with a δ-function like $\delta(\epsilon + \mathrm{U_p} + B - N\omega)$. We write $N = (\epsilon_\parallel + \epsilon_\perp + B + \mathrm{U_p})/\omega$ and so $N/(2b) = 1 + \beta_m^2 + 2\frac{\epsilon_\perp}{2\mathrm{U_p}\mathcal{Y}} + 2\frac{\epsilon_\parallel}{2\mathrm{U_p}\mathcal{Y}}$. The upshot of this entire ordeal is:

$$\begin{aligned}
\Im\,\Phi(\zeta)/b &= \left[\left(1 + 2\beta_m^2\right)\,\log\frac{\sqrt{\mathcal{Y}} + \beta_m}{\sqrt{\mathcal{Y}} - \beta_m} - 2\sqrt{\mathcal{Y}}\beta_m\right] \qquad 2.4.15a\\
&\quad + \frac{\epsilon_\perp}{2\mathrm{U_p}}\left[2\log\frac{\sqrt{\mathcal{Y}} + \beta_m}{\sqrt{\mathcal{Y}} - \beta_m} - \left(1 + 2\beta_m^2\right)\left(2\sqrt{\mathcal{Y}}\beta_m^2 - \frac{\beta_m}{\sqrt{\mathcal{Y}}}\right)\right.
\end{aligned}$$

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$$
\begin{aligned}
&-2\sqrt{\mathcal{Y}}\left(\beta_m^2+\frac{\beta_m}{2\mathcal{Y}}\right)\Bigg] && \text{2.4.15}b \\
&+\frac{\epsilon_\parallel}{2U_p}\left[2\log\frac{\sqrt{\mathcal{Y}}+\beta_m}{\sqrt{\mathcal{Y}}-\beta_m}-\left(1+2\beta_m^2\right)\left(\frac{1}{2\mathcal{Y}^2}\beta_m\sqrt{\mathcal{Y}}\right)-\frac{6\beta_m}{\sqrt{\mathcal{Y}}}\right. \\
&\left.+\frac{\beta_m}{\mathcal{Y}^{3/2}}\right] && \text{2.4.15}c
\end{aligned}
$$

This, unsurprisingly, can be enormously simplified with by churning the algebra crank. There are many cancellations of similar terms and the result is written:

$$
\begin{aligned}
\Im\,\Phi(\zeta)/b &= \kappa^2+\kappa_\perp^2\frac{\epsilon_\perp}{2U_p}+\kappa_\parallel^2\frac{\epsilon_\parallel}{2U_p} \\
\kappa^2 &= \left(1+2\beta_m^2\right)\log\frac{\sqrt{\mathcal{Y}}+\beta_m}{\sqrt{\mathcal{Y}}-\beta_m}+2\sqrt{\mathcal{Y}}\beta_m \\
\kappa_\parallel^2 &= 2\log\frac{\sqrt{\mathcal{Y}}+\beta_m}{\sqrt{\mathcal{Y}}-\beta_m}+\frac{4\beta_m}{\sqrt{\mathcal{Y}}} \\
\kappa_\perp^2 &= 2\log\frac{\sqrt{\mathcal{Y}}+\beta_m}{\sqrt{\mathcal{Y}}-\beta_m}
\end{aligned}
\tag{2.4.16}
$$

In the limit of small β_m, the 3 κ's above become:

$$
\begin{aligned}
\kappa^2 &= \frac{8}{3}\beta_m^3 \\
\kappa_\perp^2 &= 4\beta_m \\
\kappa_\parallel^2 &= \frac{4}{3}\beta_m^3
\end{aligned}
\tag{2.4.16a}
$$

where we have kept only the first nonvanishing term in the Taylor series expansions for the κ's.

We need to perform a similar expansion for $\Re\,\Phi(\zeta)$. Begin with its exact expression:

$$
\begin{aligned}
\Re\,\Phi(\zeta)/b &= \frac{N}{b}\xi+\left(\frac{a}{b}+2\beta\alpha\right)\sin\xi\cosh\eta+2\beta\sinh\eta\cos\xi \\
&= \frac{N}{b}\xi-6\alpha\alpha\tan\xi+\frac{2\beta^2\alpha}{\alpha\tan\xi}
\end{aligned}
$$

Expand as is our wont in powers of ϵ:

$$
\begin{aligned}
&= \frac{N}{b}\xi-6\frac{q_\parallel}{\sqrt{4U_p}}\sqrt{\mathcal{Y}}+\frac{2\beta_m^2}{\sqrt{\mathcal{Y}}}\frac{q_\parallel}{\sqrt{4U_p}} \\
&= \frac{N}{b}\xi+\frac{q_\parallel}{\sqrt{4U_p}}\left(\frac{2\beta_m^2}{\sqrt{\mathcal{Y}}}-6\sqrt{\mathcal{Y}}\right)
\end{aligned}
\tag{2.4.17}
$$

We are left with finding an expression for ξ. Like for η, we know, from Equation 2.4.13, that:

$$
\xi=\tan^{-1}\mathcal{Y}/\alpha
$$

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. We use the expression for the arctangent:

$$\tan^{-1} x = \mathrm{i} \log \left(\frac{1 + \mathrm{i}x}{1 - \mathrm{i}x} \right)$$

and using this, and expanding the logarithm, we get an expression for ξ:

$$\xi = \left[\frac{2\alpha}{\sqrt{\mathcal{Y}}} - \pi \right] \qquad 2.4.18$$

where here π has its usual meaning 3.14159 . . . In sum, we write the real part of Φ as:

$$\Re\,\Phi(\zeta)/b \to +4 \frac{q_{\|}}{\sqrt{4\mathrm{U_p}}} \sqrt{\mathcal{Y}} + \pi \left(1 + 2\frac{\epsilon_{\perp}}{2\mathrm{U_p}} + 2\frac{\epsilon_{\|}}{2\mathrm{U_p}} \right) \qquad 2.4.19$$

All that remains is finding an explicit form for $\Phi'' = -8\mathrm{i}b\beta \sin\zeta$. We are interested in the magnitude and argument of this separately, and it is a straightforward calculation. We derive:

$$\begin{gathered} |\Phi''(\zeta)| = 8b\beta|\sin\zeta| \to 8b\beta_m \sqrt{\mathcal{Y}} \\ \tan\chi = \frac{-3\alpha}{\alpha^2 \tan^2 \xi} \to 0 \end{gathered} \qquad 2.4.20$$

where for χ we have taken the $q \to 0$ limit. (This is consistent with our small ϵ limit we have been using.) We pick the second quadrant for χ because of the signs of the real and imaginary parts of Φ''.

We may now, at long last, write down the saddle-point approximation to $\mathcal{J}_{-N}$, as an expansion in powers of $q_{\|}$ and $q_{\perp}$. It is:

$$\begin{aligned} \mathcal{J}_{-N}(a,b) = & \frac{\mathrm{e}^{-b\kappa^2}}{\sqrt{4\pi b\beta_m \sqrt{1+\beta_m^2}}} \exp\left(-\frac{\epsilon_{\|}\kappa_{\|}^2 + \epsilon_{\perp}\kappa_{\perp}^2}{2\omega} \right) \\ & \times \cos\left(\frac{n\pi}{2} + 2q_{\|} \sqrt{\frac{\mathrm{U_p}}{\omega^2}\left(1+\beta_m^2\right)} \right) \end{aligned} \qquad 2.4.21$$

We note that in equations like Equation 2.2.4 we are integrating our GBF against functions like $\langle \lambda_q | u_0 \rangle$. This function can be slowly varying compared to the GBF. In fact, the scale for q set by that matrix element is typically $q \approx \sqrt{2B}$, whereas the scale set for q by the κ's in the GBF is much smaller: $q \approx \sqrt{8\omega/b\kappa_{\perp}^2}$ or $q \approx \sqrt{8\omega/b\kappa_{\|}^2}$. The more stringent requirement for the matrix element to be slowly-varying with respect to the sharply-peaking (and exponentially-quickly shrinking) GBF is given by the latter term:

$$2B > \frac{8\omega}{\kappa_{\|}^2}. \qquad 2.4.22$$

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Along with Equations 2.1.1 and 2.4.16 this implies upper and lower bounds on the laser parameters:

$$\frac{1}{2}\left(\frac{\bar{B}}{3\omega}\right)^{2/3} > \frac{\mathrm{U_p}}{\bar{B}} > 1 \qquad 2.4.23$$

In such a case, terms which are slowly-varying on that scale, like $\langle\lambda_q|u_0\rangle$, and $\Phi''(\zeta)$, can be evaluated at $q = 0$.

Equation 2.4.23 also gives us a practial bound on laser frequency‡ by using the outer inequalities (resulting in $\bar{B} \gg 9\omega$ roughly) and laser intensity (through $\mathrm{U_p}$):

$$\omega < 5\times 10^{-3} \qquad \text{(au)} \qquad 2.4.24a$$

$$3\times 10^{-3} < I < 213 \qquad \text{(au)} \qquad 2.4.24b$$

Evaluating the Sequential Terms

To proceed to evaluate a term like Equation 2.2.5 we substitute in all of our above work. We have, then, that:

$$\bar{\gamma} = |\langle\lambda_{q=0}|u_0\rangle|^2\,\bar{B}^2\,I_1 \qquad 2.4.25$$

where we have used the δ-function to replace $\mathrm{U_p} - n\omega$ by $q^2/2 - \bar{B}$ and the q^2 term is dominated by by the decreasing exponentials in the integral I_1:

$$\begin{aligned}
I_1 &= \sum_n \int \mathrm{d}^3q\,\delta(q^2/2 + B + \mathrm{U_p} - n\omega)\,\mathcal{J}_n^2 \\
&= \sum_n \int \mathrm{d}^3q\,\delta(q^2/2 + B + \mathrm{U_p} - n\omega)\,\frac{\mathrm{e}^{-2b\kappa^2}}{4\pi b\beta_m\sqrt{1+\beta_m^2}}\exp\left(-\frac{\epsilon_{\|}\kappa_{\|}^2 + \epsilon_{\perp}\kappa_{\perp}^2}{2\omega}\right) \\
&\quad \times \cos^2\left(\frac{n\pi}{2} + 2q_{\|}\sqrt{\frac{\mathrm{U_p}}{\omega^2}(1+\beta_m^2)}\right)
\end{aligned} \qquad 2.4.26$$

and we remind ourselves that $\epsilon_{\|} = q_{\|}^2/2$ and $\epsilon_{\perp} = q_{\perp}^2/2$, so that $q^2/2 = \epsilon_{\|} + \epsilon_{\perp}$.

To write a closed-form expression for this integral, we first note that the integrand has rotational symmetry. Using this, we can express the volume element d^3q as $2\pi\,\mathrm{d}q_{\|}\,\mathrm{d}(q_{\perp}^2/2) = 2\pi\,\mathrm{d}q_{\|}\,\mathrm{d}\epsilon_{\perp}$. Now, we use the δ-function to perform the $\epsilon_{\perp}$-integration. This will imply a mininum value for n, $n_{\min} = \lceil (B + \mathrm{U_p} + \epsilon_{\|})/\omega \rceil$, restricting that sum†:

---

‡ We should note that this limit places ω outside of the current experimental range.

† Recall that $\lceil x \rceil$ is the *ceiling function*, returning the smallest integer larger than or equal to x.

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$$I_l = \frac{e^{-2b\kappa^2}}{2b\beta_m\sqrt{1+\beta_m^2}}\int dq_\| \, e^{-\kappa_\|^2\epsilon_\|/(4\omega)} \, e^{\kappa_\perp^2/(4\omega)(B+U_p+\epsilon_\|)} \times \sum_{n=n_{\min}}^{\infty} e^{-\kappa_\perp^2/4n} \cos^2\left(\frac{n\pi}{2} + 2q_\|\sqrt{\frac{U_p}{\omega^2}(1+\beta_m^2)}\right) \qquad 2.4.27$$

Now, the sum over n is a simple geometric series. Using the relation

$$\sum_{n=x}^{\infty} e^{-an} = \frac{e^{-ax}}{1-e^{-a}},$$

and representing $\cos^2 = (1+\cos)/2$, the sum term simplifies to:

$$\sum_{n=n_{\min}}^{\infty} \cdots = e^{-\kappa^2/4\left(\left\lceil (B+U_p+\epsilon_\|)/\omega \right\rceil - 1/2\right)} \left[\frac{1}{\sinh \kappa^2/4} - (-1)^{n_{\min}} \cos\left(4q_\|\sqrt{\frac{U_p}{\omega^2}(1+\beta_m^2)}\right)\right]. \qquad 2.4.28$$

Define the function $f(x) = \lceil x \rceil - x - 1/2$. This function, as seen in Figure 2.1, oscillates around 0 with period 1, and is discontinuous at every integer.

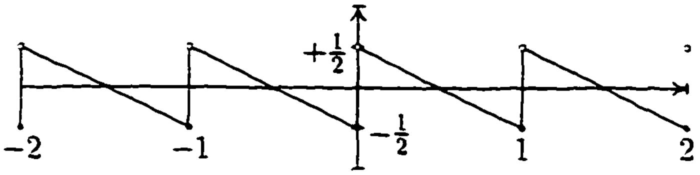

Figure 2.1: The symmetric-fractional-part function $f(x)$, as defined above.

We are left with a one-dimensional integral over $q_\|$:

$$I_l = \frac{e^{-2b\kappa^2}}{2b\beta_m\sqrt{1+\beta_m^2}}\int dq_\| \, e^{-\kappa_\|^2 q_\|^2/(2\omega)} \, e^{-\kappa_\perp^2/2f\left([U_p+B+q_\|^2/2]/\omega\right)} \times \left(\frac{1}{\sinh \kappa_\perp^2/4} - \frac{(-1)^{\lceil (U_p+B+q_\|^2/2)/\omega \rceil}}{\cosh \kappa_\perp^2/4}\cos\left(4q_\|\sqrt{\frac{U_p}{\omega^2}(1+\beta_m^2)}\right)\right) \qquad 2.4.29$$

This last integrand cannot be performed analytically but can be approximated numerically. However, as we see below in Table 2.1, we can, with small error, drop the oscillatory terms and keep only the exponentially-decreasing term. Therefore, I_l becomes:

$$I_l = \frac{e^{-2b\kappa^2}}{2b\beta_m\sqrt{1+\beta_m^2}}\frac{1}{\sinh \kappa_\perp^2/4}\int dq_\| \, e^{-q_\|^2\kappa_\|^2/(4\omega)} \rightarrow \frac{e^{-2b\kappa^2}}{2b\beta_m\sqrt{1+\beta_m^2}}\frac{1}{\sinh \kappa_\perp^2/4}\sqrt{\frac{4\pi\omega}{\kappa_\|^2}} \qquad 2.4.30$$

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	I_l approximations			
	$U_p = 100, \omega = 0.005$		$U_p = 250\,000, \omega = 0.001$	
Terms used	**Value**	*Error*	**Value**	*Error*
All terms	105.66484	—	341145.748	—
Drop cos term	105.66290	*0.002%*	341145.748	*0*
Drop all osc. terms	102.485	*3.0%*	341092.71	*0.02%*
Equation 2.4.30	102.485	*3.0%*	341092.72	*0.02%*

Table 2.1: Numerical approximations to the integral I_l in Equation 2.4.29. We do not include the exponentially small prefactor.

After replacing the values for b, β_m, etc., in the expression for $\bar{\gamma}$, and using the value for I_l in Equation 2.4.30, we may finally state:

$$\bar{\gamma} = \sqrt{\frac{3\omega^3}{16\pi^3}} \left(\frac{U_p^3 \bar{B}}{2}\right)^{1/4} |\langle\lambda_{q=0}|u_0\rangle|^2 \exp\left(-\sqrt{\frac{2^3 \bar{B}^3}{9\omega^2 U_p}}\right) \qquad 2.4.31$$

We note that Equation 2.2.8 the calculation for γ_{12} is identical to that for $\bar{\gamma}$ except for:

1. The factor of $\bar{B}$ is changed to $B_l = -W_0^+ = 2\,\text{a.u.}$
2. An extra factor of $\frac{1}{4}$
3. A factor of $U_p - n\omega$ present in Equations 2.2.1 and 2.2.5 is absent in Equation 2.2.8.
4. The matrix element appearing is different.

Because of this very simple relationship, we may without further calculation write:

$$\gamma_{12} = \sqrt{\frac{3\omega^3}{\pi^3}} \frac{1}{16 B_l^2} \left(\frac{U_p^3 B_l}{2}\right)^{1/4} \left|\langle\lambda_{q=0}| \left(\frac{\nu}{2} - \Omega\right) |w_0\rangle\right|^2 \exp\left(-\sqrt{\frac{2^3 B_l^3}{9\omega^2 U_p}}\right) \qquad 2.4.32$$

To simplify the expression for γ_{01}, we must first deal with a term which is $\bar{\mathcal{J}}_{-N} = \frac{1}{2}(\mathcal{J}_{-N-1} + \mathcal{J}_{-N+1})$. The integral representation for this is:

$$\bar{\mathcal{J}}_{-N} = \int_{-\pi}^{\pi} \frac{d\phi}{2\pi} e^{i\Phi(\phi)} \cos\phi \qquad 2.4.33$$

Since the $\cos\phi$ term will be much more slowly-varying than the other exponential term, we evaluate *it* at the saddle points as well. We know that $\cos\zeta = \alpha + i\beta$. We

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now exploit the relation:

$$\begin{aligned} \cos(\theta) \times (u + iv) + \text{c.c.} &= \sigma \cos(\theta - \tau) \\ \sigma &= |u + iv| \\ \tau &= \arg(u + iv) = \tan^{-1}\frac{v}{u} \end{aligned} \qquad 2.4.34$$

In the limit of $\epsilon \to 0$, $\alpha \to 0$ as well and therefore $\tau \to \pi/2$. Since $\cos(x - \pi/2) = \sin(x)$, we can now write $\bar{\mathcal{J}}_{-N}$, using precisely the same calculations which gave us Equation 2.4.21 as:

$$\begin{aligned} \bar{\mathcal{J}}_{-N}(a,b) = &\frac{\beta_m\, \mathrm{e}^{-2b\kappa^2}}{\sqrt{4\pi b\beta_m\sqrt{1+\beta_m^2}}}\mathrm{e}^{-\frac{\epsilon_{\|}\kappa_{\|}^2+\epsilon_{\perp}\kappa_{\perp}^2}{2\omega}} \\ &\times \sin\left(\frac{N\pi}{2} + 2q_{\|}\sqrt{\frac{\mathrm{U_p}}{\omega^2}(1+\beta_m^2)}\right) \end{aligned} \qquad 2.4.35$$

Let us take a moment to note the differences between $\mathcal{J}$ and $\bar{\mathcal{J}}$:

1. The extra factor of β_m
2. The $\cos(\cdots)$ factor has been replaced by a $\sin(\cdots)$ factor.

We now proceed to work with the expression for γ_{01} (Equation 2.2.6b) in the following way. First, we note that there will be a cross-term $\mathcal{J}_{-N}\bar{\mathcal{J}}_{-N}$, over which we integrate. These terms contain a factor which goes like $\cos(q_{\|})\sin(q_{\|})$. Over the range $-\infty$ to $+\infty$ the rapid variation in oscillations in this term will go to zero because the oscillations are out of phase and therefore interfere destructively. As well, for the same reason which led us to disregard the $\cos(q_{\|})$ term in Equation 2.4.29 we disregard the $\sin(q_{\|})$ term in $\bar{\mathcal{J}}$. Therefore, the two GBFs appearing in Equation 2.2.6b are *computationally the same* and we can write γ_{01} as:

$$\begin{aligned} \gamma_{01} = &\sum_n \int \frac{\mathrm{d}^3q}{(2\pi)^3}\delta(\frac{q^2}{2} + \mathrm{U_p} + B_2 - n\omega)\, |\langle\lambda_q|u_0\rangle\langle w_0|u_0\rangle|^2 \times \\ &\mathcal{J}_{-n}^2(a,b)\,\left[4B_2^2 - \beta_m^2\,(\omega\alpha_0 q_{\|})^2\right] \end{aligned} \qquad 2.4.36$$

We have taken the liberty of replacing $\mathrm{U_p} - n\omega$ with $B_2 + \epsilon_q$ because the presence of the δ-function guarantees that equality. We then take $\epsilon_q \to 0$ as we have previously.

We can perform ths integral in Equation 2.4.36 in the same way we calculated I_1. The only difference is the presence of the $q_{\|}$ term, which is not a problem. We

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note that

$$(\beta_{m\omega}\alpha_0)^2 = \frac{B}{2}.$$

When we perform the final $q_{\|}$ the second term in brackets in Equation 2.4.36 becomes:

$$[4B_2^2 + \frac{B}{2}\frac{\omega}{\kappa_{\|}^2}] \qquad 2.4.37$$

The term $\frac{\omega}{\kappa_{\|}^2}$ is required to be less than $B/4$ by Equation 2.4.22. Therefore, the second term in brackets in Equation 2.4.37 is bounded from above and we write the bracketed term as:

$$4B_2^2[1 + \bar{\delta}], \qquad \bar{\delta} < \frac{1}{32}. \qquad 2.4.38$$

$\bar{\delta}$ will only get smaller as we move further into the regime given by Equation 2.4.23. Therefore, we do not make a large error in ignoring the second term.

The upshot is the expression for γ_{01}:

$$\gamma_{01} = \sqrt{\frac{3\omega^3}{\pi^3}}\left(\frac{\mathrm{U_p}B_2^3}{2}\right)^{1/4} |\langle\lambda_{q=0}|u_0\rangle\langle w_0|u_0\rangle|^2 \exp\left(-\sqrt{\frac{2^3 B_2^3}{9\omega^2 \mathrm{U_p}}}\right), \qquad 2.4.39$$

where $B_2 = W_0^+ - W_0$ is the binding energy. (See Equation 2.2.7a.)

The Direct Terms

The direct terms in Equations 2.3.1 and 2.3.4 do not admit straightforward solutions like those for the sequential terms. Both involve sums over intermediate states in the matrix elements and products of GBFs. We develop a mechanism to deal with these summations.

For the ground-state correlated process (Equation 2.3.1) there is only one double sum. We can use the method described in Appendix 1 to eliminate this sum; the GBF appearing differs only in its arguments (and we shall deal with this shortly). The stationary closure method reduces the matrix elements to:

$$\sum_{\mu,\nu} \frac{\langle\lambda_{q_1}\lambda_{q_2}|\mu\nu\rangle\langle\mu\nu|\delta V|u_0u_0\rangle}{W_0 - W_{\mu\nu}} \to \frac{\langle\lambda_{q_1}\lambda_{q_2}|\delta V|u_0u_0\rangle^2}{\langle\lambda_{q_1}\lambda_{q2}|[\delta V, H_0]|u_0u_0\rangle}. \qquad 2.4.40$$

δV and H_0 are given by Equations 1.3.14 and 1.3.10, respectively. We make the same approximations about replacing $q \to 0$ in the matrix elements which we did for

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the sequential terms. The result of this is that the form for the initial-state correlated T-matrix (Equation 2.3.1 becomes:

$$T_N^{G_0} = \frac{\langle \lambda_{q_1} \lambda_{q_2} | \delta V | u_0 u_0 \rangle^2}{\langle \lambda_{q_1} \lambda q_2 | [\delta V, H_0] | u_0 u_0 \rangle} (2\mathrm{U_p} - N\omega)\, \mathcal{J}_{-N}(\vec{\alpha}_0 \cdot \vec{Q}, \mathrm{U_p}/\omega) \qquad 2.4.41$$

with $\vec{Q} = \vec{q}_1 + \vec{q}_2$ and $\vec{q} = \vec{q}_1 - \vec{q}_2$ being the sum and differences of the electron momenta.

This new GBF $\mathcal{J}_{-N}(\vec{\alpha}_0 \cdot \vec{Q}, \mathrm{U_p}/\omega)$ appears in the following double-integral:

$$\begin{aligned} I_2 = \sum_N \iint \frac{\mathrm{d}^3 q_1 \mathrm{d}^3 q_2}{(2\pi)^5} \, \delta(\epsilon_{q_1} + \epsilon_{q_2} + 2\mathrm{U_p} + B - N\omega) \\ \mathcal{J}^2_{-N}(\vec{\alpha}_0 \cdot \vec{Q}, b') \, (2\mathrm{U_p} - N\omega)^2 \end{aligned}$$

where

$$\begin{aligned} \mathcal{J}^2_{-N}(\vec{\alpha}_0 \cdot \vec{Q}, b') = {} & \frac{\mathrm{e}^{-2b'\kappa^2}}{4\pi b' \beta_m \sqrt{1+\beta_m^2}} \, \mathrm{e}^{-2b'\kappa_{\|}^2 Q_{\|}^2/2} \\ & \times \mathrm{e}^{-2b'\kappa_{\perp}^2 (Q_{\perp}^2/2 + q^2/2)} \cos^2\left(\frac{N\pi}{2} + 2Q_{\|} \sqrt{\frac{\mathrm{U_p}(1+\beta_m^2)}{\omega^2}} \right) \end{aligned} \qquad 2.4.42$$

and

$$b' \stackrel{\mathrm{def}}{=} \frac{\mathrm{U_p}}{\omega}$$

We should say a few words about the structure of $\mathcal{J}(\vec{\alpha}_0 \cdot \vec{Q}, b')$. We can consider this GBF as entering into a *6-dimensional* integral over a hyper-momentum $(\vec{q}, \vec{Q})^{\mathrm{T}} = \vec{\mathcal{Q}}$. When we solved the saddle-point method for the "regular" GBF between Equations 2.4.4 and 2.4.16, we saw that there was one particular direction of momentum $q_{\|} = \vec{\alpha}_0 \cdot \vec{q}$ which was picked out into α, and the δ-function "selected" the remaining direction $(\vec{q}_{\perp})$ into β. The situation here is analogous. The one direction we have in α is $\vec{\alpha}_0 \cdot \vec{Q}$, and the remaining 5 directions $(\vec{Q}_{\perp} + \vec{q})$ will be in β. We may, without further ado, write down an expression for $\mathcal{J}^2_{-N}(\vec{\alpha}_0 \cdot \vec{Q}, 2b)$:

$$\begin{aligned} \mathcal{J}^2_{-N}(\vec{\alpha}_0 \cdot \vec{Q}, 2b) = {} & \frac{\mathrm{e}^{-4b\kappa^2/4\omega}}{8\pi b \beta'_m \sqrt{1+{\beta'_m}^2}} \\ & \times \exp\left[-\kappa_{\|}^2 Q_{\|}^2/(2\omega) - \kappa_{\perp}^2 (Q_{\perp}^2 + q^2)/(2\omega) \right] \\ & \times \cos^2\left(\frac{N\pi}{2} + 2Q_{\|} \sqrt{\frac{\mathrm{U_p}(1+{\beta'_m}^2)}{\omega^2}} \right) \end{aligned} \qquad 2.4.43$$

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where $\beta'_m = \sqrt{B/(4U_p)}$ and the various κ's appearing here are defined in terms of β'_m.

This integral is solved in much the same way as the I_1 above. We note that the argument to the δ-function can be written $\delta(\epsilon_Q + \epsilon_q + 2U_p + B - N\omega)$. Like in I_1, the $\cos^2$ term oscillates rapidly and can be replaced by 1/2. Let us write $\epsilon_Q = \epsilon_{Q_\perp} + \epsilon_{Q_\|}$. Since the integrand is symmetric about the $\hat{\alpha}_0$ axis, we can write:

$$\mathrm{d}^3Q = 2\pi \mathrm{d}Q_\| \, \mathrm{d}Q_\perp Q_\perp = 2\pi \mathrm{d}Q_\| \, \mathrm{d}\epsilon_{Q_\perp}$$

and then use the δ-function to eliminate terms in $Q_\perp$:

$$\begin{aligned} I_2 = &\frac{1}{4}\frac{\mathrm{e}^{-2b'\kappa^2}}{(2\pi)^5 \, b' \beta'_m \sqrt{1+\beta'_m}} \sum_N \int \mathrm{d}^3q \, \mathrm{d}Q_\| \mathrm{e}^{-\kappa_\|^2 Q_\|^2/4\omega} \\ &\times \exp\left(-\kappa_\perp^2 [N\omega - 2U_p - B - \epsilon_{Q_\|} - \epsilon_q]/(4\omega)\right) \\ &\times \exp(-\kappa_\perp^2 \epsilon_q / 4\omega). \end{aligned} \qquad 2.4.44$$

Continuing, we notice that the q cancels out completely from the exponential terms. However, there is an upper limit of q set by the requirement that $Q_\perp^2$ not become negative:

$$q_{\max} = \sqrt{2\left(N\omega - 2U_p - B - \frac{{Q_\|}^2}{2}\right)} \qquad 2.4.45$$

The integral $\int \mathrm{d}^3q$ is $4\pi q_{\max}^3/3$. This requirement also bounds N from below:

$$N \geq N_{\min} = \left(\frac{2U_p + B + \epsilon_{Q_\|}}{\omega}\right) \qquad 2.4.46$$

Using this in Equation 2.4.44 results in:

$$\begin{aligned} I_2 = &\frac{1}{2}\frac{2^{3/2}\mathrm{e}^{-2b'\kappa^2}}{3(2\pi)^4 \, b' \beta'_m \sqrt{1+\beta'_m}} \\ &\sum_{N=\lceil N_{\min}\rceil}^{\infty} \int \mathrm{d}Q_\| \exp\left(-\kappa_\|^2 \epsilon_{Q_\|}/4\omega\right) \\ &\times [N\omega - N_{\min}\omega]^{3/2} \exp\left(-\kappa_\perp^2 [N - N_{\min}]/4\right). \end{aligned} \qquad 2.4.47$$

At this point, we diverge slightly from our derivation of I_1. We note that $N_{\min} \gg 1$, and so we approximate our sum over N by an integral $\int \mathrm{d}N$. In this case, our lower

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limit becomes exactly N_{min}. The integral becomes trivial with a change of variables $Y = N - N_{\mathrm{min}}$.

$$\int_0^\infty \mathrm{d}Y\, Y^{-3/2} e^{-\kappa_\perp^2/4Y} = \frac{\Gamma(5/2)}{(\kappa_\perp^2/4)^{5/2}}$$

and the remaining integral with respect to $Q_\parallel$ is likewise elementary†. The result is:

$$I_2 = \frac{3^{1/2}\omega^3}{4\pi^3}\left(\frac{\mathrm{U}_\mathrm{p}^3}{B^5}\right)^{1/2} \exp\left(-\sqrt{\frac{4B^3}{9\mathrm{U}_\mathrm{p}\omega^2}}\right). \qquad 2.4.48$$

Let us now turn to the expression for T_N^{II}, Equation 2.3.4. To perform the ν sum, we use stationary closure, employing the closure relations for the sums of GBF as given in Equations A2.3 and A2.6. We replace ν by an "average number of photons" $\bar{\nu} = (N+s)/2 - \vec{\alpha}_0 \cdot (\vec{q} - \vec{Q}/2)\bar{\mathcal{J}}_N/\mathcal{J}_N$. In the $q_1, q_2 \to 0$ limit, the last term has a leading term which is of order q_1 or q_2, and vanishes, because the ratio of the GBFs appearing there is a number $O(1)$. As well, N is large for real laser intensities‡ so we approximate $\bar{\nu} \approx N/2$. In this light, Equation 2.3.4 becomes:

$$\begin{aligned} T_N^{II}(q_1, q_2) = \sum_{n,s} \int \frac{\mathrm{d}^3 q}{(2\pi)^3} & \frac{\mathcal{J}_{s-N}(\vec{\alpha}_0 \cdot \vec{Q}, \mathrm{U}_\mathrm{p}/\omega)}{W_0 - W_n^+ - q^2/2 - \mathrm{U}_\mathrm{p} - \frac{N}{2}\omega + i\eta} \\ & \times \left(\frac{1 + \mathrm{X}_{12}}{2}\right) \langle \lambda_{q_1}\lambda_{q_2} | \delta V' | w_n \lambda_q \rangle \langle w_n \lambda_q | H_s' | u_0 u_0 \rangle \end{aligned} \qquad 2.4.49$$

where

$$\delta V' \stackrel{\mathrm{def}}{=} V_{12} - \Omega_2 - \frac{\nu'}{2}\pi_2$$

(the $\delta V'$ here is different from the one in Equation 1.3.21) and *here alone* we call $\nu' = \langle u_0 u_0 | V_{12} | u_0 u_0 \rangle$ instead of ν to avoid confusion with the summation index.

The sum over s can be done simply thanks to the recurrence relations and the special forms of H_s':

$$H_0' = 2\mathrm{U}_\mathrm{p} \qquad H_{\pm 1} = \frac{\omega}{2}(\vec{p}_1 + \vec{p}_2) \cdot \vec{\alpha}_0 \qquad H_{\pm 2} = \mathrm{U}_\mathrm{p} \qquad 2.4.50$$

With this in hand, and the GBF recurrence relations¶, we write:

---

† However, one must remember that the limits of integration $\mathrm{d}Q_\parallel$ are from $-\infty$ to $+\infty$.

‡ $N_{\mathrm{min}} \approx \mathrm{U}_\mathrm{p}/\omega$, which we assume to be large.

¶ Recall that all the arguments to the GBFs are the same

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$$\begin{aligned}\sum_s \mathcal{J}_{s-N}(a'+a'',2b)H'_s &= 2\mathrm{U_p}\mathcal{J}_{-N} + \frac{\omega(\vec{p}_1+\vec{p}_2)\cdot\alpha_0}{2}\left[\mathcal{J}_{-N-1}+\mathcal{J}_{-N+1}\right] \\ &\quad + \mathrm{U_p}\left[\mathcal{J}_{-N-2}+\mathcal{J}_{-N+2}\right] \\ &= \left(2\mathrm{U_p}-N\omega\right)\mathcal{J}_{-N} \\ &\quad - \frac{\omega(\vec{p}_1+\vec{p}_2-\vec{Q})\cdot\alpha_0}{2}\left[\mathcal{J}_{-N-1}+\mathcal{J}_{-N+1}\right]\end{aligned} \qquad 2.4.51$$

T^{II} is now given by§:

$$\begin{aligned}T^{II}_N(q_1,q_2) &= \sum_n \int \frac{\mathrm{d}^3q}{(2\pi)^3}\frac{1}{W_0-\mathrm{U_p}+N\omega/2-W_n^+-q^2/2+\mathrm{i}\eta} \\ &\quad \times \left(\frac{1+\mathrm{X}_{12}}{2}\right)\left\{\left(2\mathrm{U_p}-N\omega\right)\mathcal{J}_{-N}\langle\lambda_{q_1}\lambda_{q_2}|\delta V'|w_n\lambda_q\rangle\langle w_n\lambda_q|u_0u_0\rangle\right. \\ &\quad \left. -\frac{\omega}{2}\langle\lambda_{q_1}\lambda_{q_2}|\delta V'\vec{\alpha}_0\cdot(\vec{p}_1+\vec{p}_2-\vec{Q})|w_n\lambda_q\rangle\langle w_n\lambda_q|u_0u_0\rangle\bar{\mathcal{J}}_{-N}\right\}\end{aligned} \qquad 2.4.52$$

As before, we will evaluate this at $q_1 = q_2 = 0$ everywhere except in the GBFs. We continue in the same vein to eliminate the combination sum-over-n/integral-over-q. This sum/integral can be seen to be one single closure over the states $w_n\lambda_q$. First, we will no longer need the symmetry operator $(1+\mathrm{X}_{12})/2$ since in the limit above both the initial and final states are invariant under particle interchange. As well, we see that the second term will vanish because we will have a term which is $\vec{\alpha}_0\cdot(\vec{p}_1+\vec{p}_2)$ acting on a pair of S states, and this term will vanish. Therefore, we will have, after performing stationary closure, and employing the δ-function in Equation 1.3.56:

$$T^{II}_N(q_1,q_2) = \frac{\langle\lambda_0\lambda_0|\delta V'|u_0u_0\rangle}{\frac{W_0}{2}-\frac{\langle\lambda_0\lambda_0|\delta V'H'_0|u_0u_0\rangle}{\langle\lambda_0\lambda_0|\delta V'|u_0u_0\rangle}}\mathcal{J}_{-N} \qquad 2.4.53$$

H_0 here is the hamiltonian which $|w_n\lambda_q\rangle$ satisfies:

$$H'_0 = T_1 + V_1 + T_2. \qquad 2.4.54$$

---

§ $\bar{\mathcal{J}}$ is defined in Equation 2.4.33.

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Digression on the SC method for the ν sum.

The use of the stationary closure method to perform the ν sum requires further analysis. When we approximated the GBF by using Equation 2.4.2, we found that there were no real stationary phase points, which made the GBF itself become exponentially small. However, for each GBF of the product $\mathcal{J}_{\nu-N}(\vec{\alpha}_0\cdot(\vec{Q}-\vec{q}),b)\mathcal{J}_{s-\nu}(\vec{\alpha}_0\cdot\vec{Q},b)$ the energy δ-function is not enough to give a unique relation betwen the order and arguments of the GBF so that real stationary phase points do exist in the integral representations of the GBF. These will *not* be exponentially small. Moreover, there exist areas in the $(q_{\|},\nu)$ plane where each of the GBFs have these real SPPs and so the product is not small. In fact, in Equation 2.4.7 we would simply have:

$$\mathcal{J}_{-n} \propto C_1\cos(\Phi(\zeta_1)) + C_2\cos(\Phi(\zeta_2))$$

However, the other GBF will have a similar expression in terms of cosines, and the product of the two GBFs—two rapidly-oscillating functions—will *not* have a real SPP, and therefore the product *will* become exponentially small. We shall show this now.

For the GBF to have a real SPP, we require that β from Equation 2.4.3d be pure imaginary. Therefore:

$$\left(\frac{a}{8b}\right)^2 + \frac{\nu}{4b} + \frac{1}{2} > 0 \qquad 2.4.55$$

This is satisfied when, substituting in $b = \mathrm{U_p}/(2\omega)$ and $a = \alpha_0 q_{\|}$ (for some q):

$$\frac{q_{\|}^2}{2} + \mathrm{U_p} + \nu\omega > 0. \qquad 2.4.55'$$

(This is always true when $\nu > 0$.) Let us simplify our notation somewhat. We will call the two roots of Equation 2.4.3b $x_\pm$, where:

$$x_\pm = -\frac{a}{8b} \pm R, \qquad R = \sqrt{\left(\frac{a}{8b}\right)^2 + \frac{\nu}{4b} + \frac{1}{2}} \qquad 2.4.56$$

Note that $R > 0$ for $\nu > 0$, thus enforcing reality. The requirement that $x_\pm = \cos\phi_\pm$ have magnitude less than or equal to 1 places bounds on permissable values of ν and $q_{\|}$. The first set, coming from x_+, is:

$$2\nu\omega - 4q_{\|}\sqrt{\mathrm{U_p}} - 2\mathrm{U_p} \le 0 \le 2\nu\omega + 4q_{\|}\sqrt{\mathrm{U_p}} - 2\mathrm{U_p}. \qquad 2.4.57a$$

This provides us with part of the region in the $(q_{\|},\nu)$ plane where the GBF can be large. We have a similar set of requirements on x_-:

$$2\nu\omega + 4q_{\|}\sqrt{\mathrm{U_p}} - 2\mathrm{U_p} \le 0 \le 2\nu\omega - 4q_{\|}\sqrt{\mathrm{U_p}} - 2\mathrm{U_p}. \qquad 2.4.57b$$

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We will get a pair of $x_\pm$'s for each of the two GBFs appearing in our sum. Call one pair $x_\pm$ and the other pair $x'_\pm$. We will therefore see four contributions to the integral arising from evaluating the function at the four saddle points:

$$x_+x'_+,\quad x_+x'_-,\quad x_-x'_+,\quad x_-x'_- \tag{2.4.58}$$

These terms will contribute so long as the regions which they describe in the $(q_\parallel, \nu)$ plane overlap. Some straightforward but messy algebra shows that there *are* regions of overlap, so the terms will all contribute.

These terms will enter into a sum over ν and an integral over $\vec{q}$. Applying the saddle point technique to the GBF in the domain where there are real SPPs results in:

$$\mathcal{J}_N = \frac{\cos\left(\Phi(x_+) + \frac{\pi}{4}\,\mathrm{sgn}\Phi''(x_+)\right)}{\sqrt{2\pi|\Phi''(x_+)|}} + \frac{\cos\left(\Phi(x_-) + \frac{\pi}{4}\,\mathrm{sgn}\Phi''(x_-)\right)}{\sqrt{2\pi|\Phi''(x_-)|}} \tag{2.4.59}$$

where by $\Phi(x_+)$ we mean

$$\Phi(\phi)\Big|_{\cos\phi = x_+}. \tag{2.4.59b}$$

These are made simpler by noting that the sign of $\Phi''(x_\pm)$ is the same as that of x: that is, $\Phi''(x_+) > 0$, and $\Phi''(x_-) < 0$. To see this, write $\Phi''(\phi) = a\sin\phi + 4b\sin 2\phi$. If we expand $\sin 2\phi$ as $2\sin\phi\cos\phi$, and substitute in $x_\pm$ for $\cos\phi$, we get:

$$\Phi''(\phi) = \pm 8b\sin\phi_\pm, \qquad \sin\phi_\pm = \sqrt{1 - x_\pm^2}. \tag{2.4.60}$$

Therefore, we can write the argument to the cosine term in Equation 2.4.59 as $\cos(\Phi(x_\pm) \pm \pi/4)$. The product of the two GBFs becomes now:

$$\mathcal{J}_1\mathcal{J}_2 = \sum_{j=\pm}\sum_{m=\pm} \frac{\cos(\Phi_j + \frac{\pi}{4}\,\mathrm{sgn}\Phi_j'')\,\cos(\Phi_m + \frac{\pi}{4}\,\mathrm{sgn}\Phi_m'')}{2\pi\sqrt{|\Phi_j''\Phi_m''|}} \tag{2.4.61}$$

Here, the subscript j is summing over the $x_\pm$, which are the real SPPs of $\mathcal{J}_{s-\nu}(\vec{\alpha}_0\cdot\vec{q}, b)$ and m is summing over the $x'_\pm$, which are the real SPPs of the other GBF $\mathcal{J}_{\nu-N}(\vec{\alpha}_0\cdot(\vec{Q}-\vec{q}), b)$. Each of these four terms is integrated over its own domain in the $(q_\parallel, \nu)$ plane.

To continue on our notation-simplifying path, let us rewrite the arguments to the GBFs in Equation 2.4.49 in the following way. Let $x = \nu - N$ and $N' = -s - N$. Also, let $\vec{k} = \vec{q} - \frac{1}{2}\vec{Q}$. This lets us write the product of the GBFs as:

$$\mathcal{J}_x(\vec{\alpha}_0\cdot(\frac{\vec{Q}}{2} - \vec{k}), b)\,\mathcal{J}_{-N'-x}(\vec{\alpha}_0\cdot(\frac{\vec{Q}}{2} + \vec{k}), b)$$

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The integration formerly over $\vec{q}$ is now over $\vec{k}$. Let us symmetrize the summation indices as well by letting $x = n' - N'/2$. Equation 2.4.49 will have the (schematic) form, dropping the primes on the indices:

$$\mathcal{J}_{-N/2-n}(\vec{\alpha}_0 \cdot (\vec{Q}/2 + \vec{k}), b)\, \mathcal{J}_{n-N/2}(\vec{\alpha}_0 \cdot (\vec{Q}/2 - \vec{k}), b) \qquad 2.4.62$$

With this in hand, let us turn to the evaluation at the real SPPs. We can use the trigonometric identity $\cos a \cos b = (\cos(a+b) + \cos(a-b))/2$ to simplify the four terms appearing in Equation 2.4.61. Let us consider just the first term, with $j = +$ and $m = +$. The terms here will be rapidly oscillating and not contribute to the integral over k and sum over n unless there is a real SPP of the product of the *cosines* appearing. If there is, we will get a "large" contribution‡. Otherwise it will be exponentially small. The two Φ's appearing are:

$$\Phi_j = -(n + \frac{N}{2})\phi_j - \vec{\alpha}_0 \cdot (\frac{1}{2}\vec{Q} - \vec{k}) \sin\phi_j + b \sin 2\phi_j = \Phi_j(n, k_z)$$

$$\Phi_m = (n - \frac{N}{2})\phi_m - \vec{\alpha}_0 \cdot (\frac{1}{2}\vec{Q} + \vec{k}) \sin\phi_m + b \sin 2\phi_m = \Phi_m(n, k_z)$$

where:

$$\cos\phi_j = -\frac{\vec{\alpha}_0 \cdot (\vec{Q}/2 - \vec{k})}{8b} + \sqrt{\left(\frac{\vec{\alpha}_0 \cdot (\vec{Q}/2 - \vec{k})}{8b}\right)^2 + \frac{n + N/2}{4b} + \frac{1}{2}} \qquad 2.4.63$$

$$\cos\phi_m = -\frac{\vec{\alpha}_0 \cdot (\vec{Q}/2 + \vec{k})}{8b} + \sqrt{\left(\frac{\vec{\alpha}_0 \cdot (\vec{Q}/2 + \vec{k})}{8b}\right)^2 + \frac{n - N/2}{4b} + \frac{1}{2}}$$

For there to be a stationary phase, we require that the first partial derivatives of $\Phi_{j,m}(n, k_z)$ with respect to n and k_z vanish. The derivatives are:

$$\frac{\partial \Phi_j}{\partial n} = -\phi_j \qquad \frac{\partial \Phi_m}{\partial n} = +\phi_m$$

$$\frac{\partial \Phi_j}{\partial k_z} = +\alpha_0 \sin\phi_j \qquad \frac{\partial \Phi_j}{\partial n} = -\alpha_0 \sin\phi_m \qquad 2.4.64$$

These together require that for the $\cos(\Phi_j + \Phi_m)$ term $\phi_j = \phi_m$. For the $\cos(\Phi_j - \Phi_m)$ term, we get $\phi_j = -\phi_m$ and $\sin\phi_j = -\sin\phi_m$. For $|\phi| < \pi$, there are no solutions to this second part of the first term†. Therefore, there are no real SPPs for that term,

---

‡ See Morse & Feshbach[17], §4.6.

† The restriction to the interval $[0, \pi]$ comes about because we require $\sin\phi > 0$, otherwise there would be no real stationary phase point.

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and it will become exponentially small and vanish compared to the first part of the first term. The relation $\phi_j = \phi_m$ gives us:

$$-\frac{\alpha_0}{8b}(\frac{Q_z}{2} - k_z) + R_j = -\frac{\alpha_0}{8b}(\frac{Q_z}{2} + k_z) + R_m \qquad 2.4.65$$

where R_j and R_m are defined to be the values of R for $\phi_{j,m}$ respectively. These give us two relations between n and k_z:

$$n_\pm = \frac{-\alpha_0 k_z Q_z}{16b} \pm \alpha_0 k_z \sqrt{\frac{(\alpha_0 Q_z)^2}{(16b)^2} - \frac{1}{2}\left\{\frac{N}{4b} - 1\right\}} \qquad 2.4.66$$

The limit $Q_z \to \infty$ requires, from Equation 2.4.65 that $n \to 0$. Therefore. we need the minus sign in Equation 2.4.66:

$$n = \alpha_0 k_z \left[\sqrt{\left(\frac{\alpha_0 Q_z}{16b}\right)^2 - \frac{1}{2}\left(\frac{N}{4b} - 1\right)} - \frac{\alpha_0 Q_z}{16b}\right]. \qquad 2.4.67$$

We see that the radicand is never positive, due to the δ-function controlling the values of $\vec{Q}, \vec{k}$ and N. Exactly similar analyses show that there are *no* real SPPs for the product of these two cosines, and therefore we must proceed with the standard saddle point method with an exponentially decreasing factor — which is the *same* exponential factor one gets from the method of stationary closure! Therefore, we do not make gross errors by dropping terms exponentially larger than the ones we keep by using the SC method for the ν sum in Equation 2.3.4

We are now equipped, at long last, to write down the direct T-matrix contribution to γ_{02}. Equations 2.4.53 and 2.4.40 yield:

$$\begin{aligned} T_N^D &= \mathcal{J}_{-N}(\vec{\alpha}_0 \cdot \vec{Q}, \mathrm{U_p}/\omega)\, W_0 \\ &\times \left(\frac{\langle\lambda_0\lambda_0|\delta V|u_0u_0\rangle^2}{\langle\lambda_0\lambda_0|[\delta V, H_0]|u_0u_0\rangle} + \frac{\langle\lambda_0\lambda_0|\delta V'|u_0u_0\rangle^2}{\langle\lambda_0\lambda_0|\delta V'(W_0/2 - H_0')|u_0u_0\rangle}\right) \end{aligned} \qquad 2.4.68$$

We shall refer to the multiplicand above as $\mathcal{M}$.

We see that the second part of γ_{02}, as given in Equation 1.4.9 vanishes for the following reason: given the approximations we have used so far, all of the various T-matrices, $\tau^{(1)}$ and $\tau^{(2)}$ are all real and therefore their imaginary parts vanish uniformly. Using Equation 2.4.48 we have:

$$\gamma_{02} = \frac{3^{1/2}\omega^3}{8(2\pi)^3 B^{5/2}\mathrm{U_p^{1/2}}}\,|\mathcal{M}|^2 \exp\left(-\sqrt{\frac{4B^3}{9\omega^2\mathrm{U_p}}}\right) \qquad 2.4.69$$

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Section 2.5: Numerical Results

To continue, we must evaluate the matrix elements appearing in (*e.g.*) Equation 2.4.32. Table 2.2 lists them. We can write $u_0(r)$ as a sum of Slater orbitals[18][19]. However, in view of the coarse approximations we have made with the stationary closure method, we make no great error in approximating $u_0(r)$ by a variational form $N\exp(-\lambda r)$, where N is a normalizing factor: $N^2 = \lambda^3/\pi$, and λ is determined to be 27/16[7]. The integrals appearing below are all elementary, except that some notice must be paid to the $1/r_{12}$ terms appearing, which we treat in the manner that Landau and Lifshitz[7] do. We detail one example here. Let us calculate $\langle\lambda_0|\Omega|u_0\rangle$. In the $q \to 0$ limit, $\lambda_q \to 1$. We have then:

$$\begin{aligned}
\langle\lambda_0|\Omega|u_0\rangle &= N^3 \int \mathrm{d}^3r \int \mathrm{d}^3x\, u_0(\vec{x})^2\, u_0(\vec{r})\frac{1}{|\vec{r}-\vec{x}|} \\
\text{Our app} &= (4\pi)^2 N^3 \int r^2\mathrm{d}r\, x^2\mathrm{d}x\, u_0^2(x)\frac{1}{|\vec{r}-\vec{x}|}u_0(r) \text{ symmetric)}. \\
&= (4\pi)^2 N^3 \left(\int_0^\infty \mathrm{d}r\, u_0(r)\, r \int_0^r \mathrm{d}x\, x^2 u_0^2(x)^{r)}\right. \\
&\quad + \int_0^\infty \mathrm{d}r\, r^2 u_0(r) \int_r^\infty \mathrm{d}x\, x\, u_0^2(x)\Big) \quad x) \\
&\quad + \left.\int_0^\infty \mathrm{d}r\, r^2 u_0(r) \int_r^\infty \mathrm{d}x\, x\, u_0^2(x)\right) \\
&\text{The form of } u_0 \text{ makes these integrals simple.} \\
&= (4\pi)^2 N^3 \left(\frac{4}{27\lambda^5} + \frac{1}{18\lambda^5}\right)
\end{aligned} \qquad 2.5.1$$

All of the matrix elements in Table 2.2 are handled in a similarly straightforward fashion.

Since we are using a variational wavefunction, we shall also use a variational approximation to W_0, the ground-state He energy:

$$\langle u_0 u_0|H_0|u_0 u_0\rangle = 2\epsilon_0 - \nu = -2.848\,\text{a.u.} \qquad 2.5.2$$

We observe that $\mathcal{M}$, the matrix element appearing in the direct ionization rate γ_{02}, consists of an initial-state correlation term (with δV) and a final-state correlation term (with $\delta V'$). The data below show that the former is approximately a factor of six smaller than the latter, and so its contribution cannot be ignored. Numerically, $\mathcal{M} = -3.604\pi$

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$$\begin{aligned}
|\langle\lambda_{q=0}|u_0\rangle|^2 &= \frac{64\pi}{\lambda^3} \\
|\langle w_0|u_0\rangle|^2 &= 8\frac{(2\lambda)^{3/2}}{2+\lambda^3} = 0.989 \\
\left|\langle\lambda_{q=0}|\frac{\nu}{2}-\Omega|w_0\rangle\right|^2 &= 0.0749\pi \\
\langle\lambda_0\lambda_0|\delta V|u_0u_0\rangle &= 2.757\pi \\
\langle\lambda_0\lambda_0|\delta V'|u_0u_0\rangle &= 4.890\pi \\
\langle\lambda_0\lambda_0|[\delta V, H_0]|u_0u_0\rangle &= 10.221\pi \\
\langle\lambda_0\lambda_0|\delta V'\left(\frac{W_0}{2}-H_0'\right)|u_0u_0\rangle &= -5.50\pi
\end{aligned}$$

Table 2.2: Matrix elements appearing in the expressions for the various γ's

We note that in all the expressions for the γ's, we have a term proportional to $(U_p\omega^2)^n$ for some n. We identify this term with $(I/4)^n$, where I is the laser intensity in atomic units. This allows us to write the terms for the rates in terms of the laser intensity alone, and independent of the laser frequency:

$$\begin{aligned}
\gamma_{01} &= 3.7273\, I^{3/4}\, e^{-1.6101/\sqrt{I}} \\
\gamma_{12} &= 4.64346 \times 10^{-4}\, I^{3/4}\, e^{-5.333/\sqrt{I}} \\
\bar{\gamma} &= 3.335\, I^{3/4}\, e^{-3.2924/\sqrt{I}} \\
\gamma_{02} &= 3.169 \times 10^{-2}\, I^{3/2}\, e^{-6.585/\sqrt{I}}
\end{aligned}$$

Table 2.3: The ionization rates (γ's), as functions of laser intensity, in a.u.

We note that these rates are all monotonically rising functions of intensity. We plot them as a function of intensity in Figures 2.2, 2.3 and 2.4. All the units of time and intensity are in atomic units, given in Equations 0.2.3:

In Figure 2.5, we plot the turnover time t_t, from Equation 1.5.1 as a function of laser intensity.

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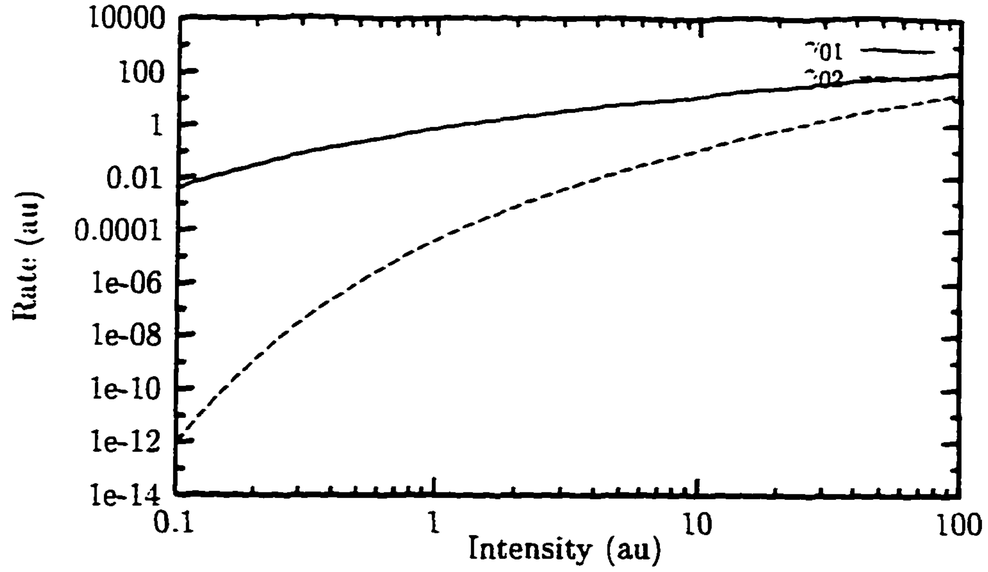

Figure 2.2: Plot of γ_{01} and γ_{02} versus laser intensity over three decades. All units are atomic units.

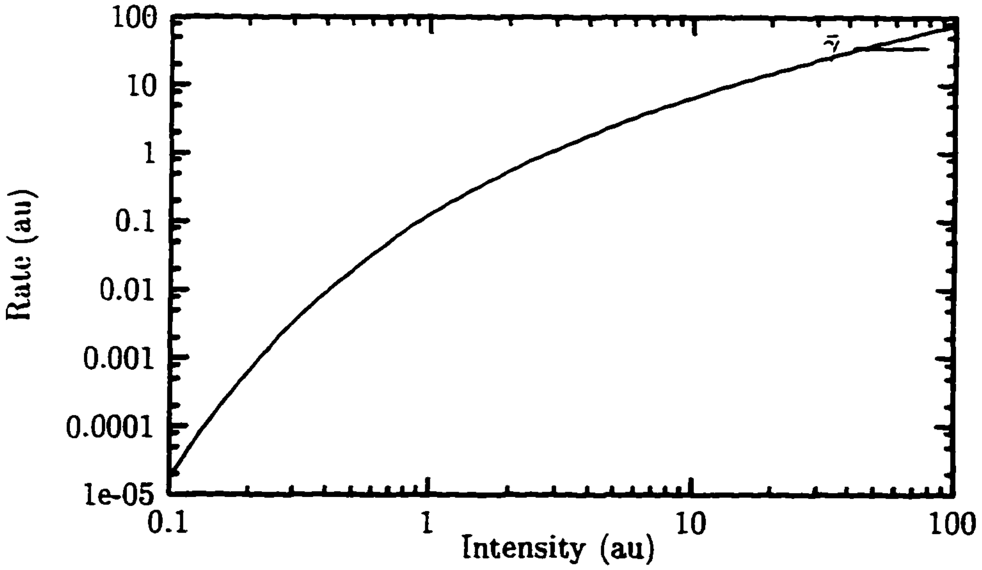

Figure 2.3: Plot of $\bar{\gamma}$ versus laser intensity over three decades.

Section 2.6: Comparison With Experiment

Up until now, direct double-ionization has not been observed directly, but rather it has been have inferred it from yield curves. For example, Walker *et al.*[20] have produced a graph of singly- and doubly-charged helium ions as a function of laser intensity—see Figure 2.6.

Walker *et al.* interpret the sudden change in shape (the "knee") of the ion-yield versus intensity curve as the changeover of the yield of He^{++}. from being dominated

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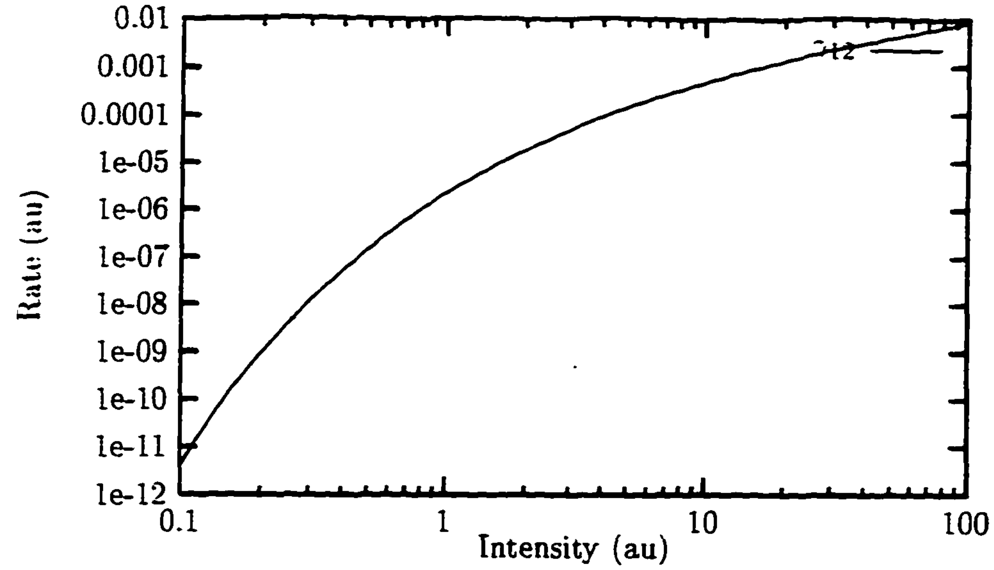

Figure 2.4: Plot of γ_{12} versus laser intensity over three decades.

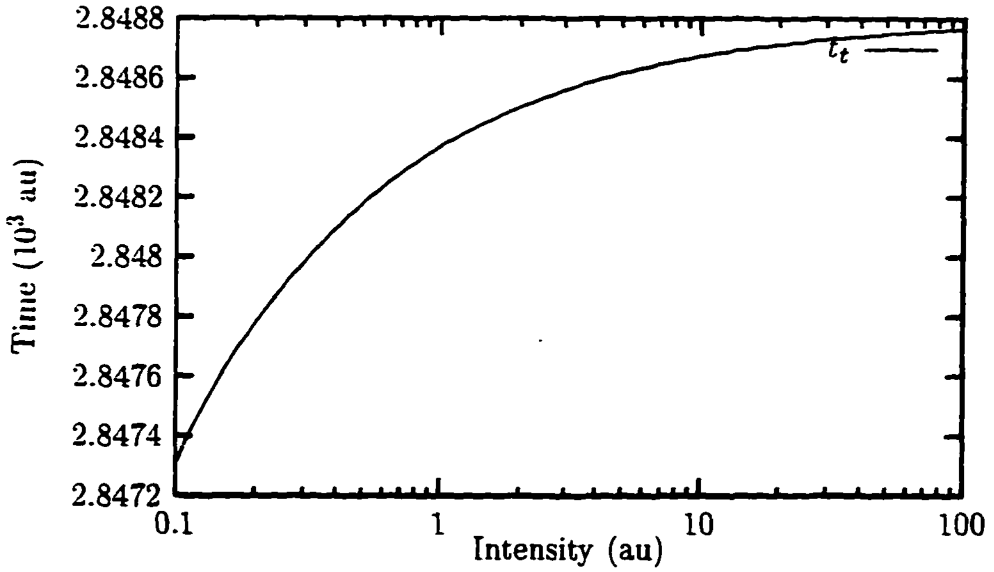

Figure 2.5: Turnover time t_t versus intensity.

by the non-sequential process to being dominated by the sequential one. However, Lambroupolous and his coworkers[21] showed that what Walker *et al.* saw might not have been the onset of a "direct" process but rather the introduction of an intensity-dependent resonance of the atom in the laser field. That is, there would be an enhancement of γ_{01} due to the atom's absorbing some number of photons and dressing the atom so that the γ_{01} becomes a resonant process. The atom would then be stabilized against ionization, and the yield would not increase as I increased as long

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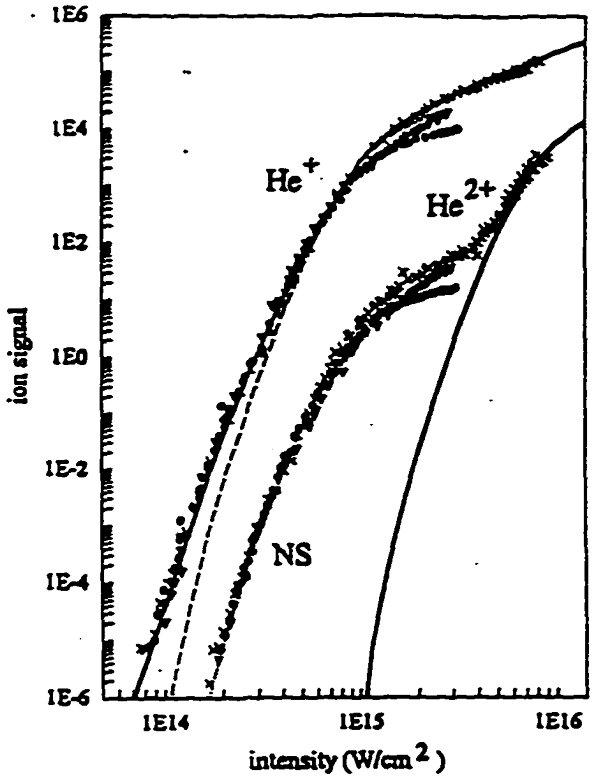
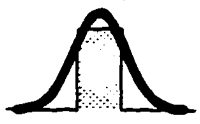


Figure 2.6: Experimental graph of ion yield vs. Intensity, taken from Walker, *et al.*[20]. Measured He ion yields for linear polarized, 100 fs, 780 nm light. Calculations are shown as solid (SAE) and dashed (ac-tunneling) lines. The measured intensities are multiplied by 1.15. The solid curve on the right is the calculated sequential He^{++} yield. (Reprinted with permission.)

as the resonance existed.

For us, with our model, to determine the onset of a changeover from the dominance of the direct process to the sequential one, we look at the fractional yield of doubly-charged ions as give by the rate equations (Equation 1.4.7). In our model, we assume that the laser intensity is "suddenly" turned on.



That is, the laser intensity is a step-function on and off in time. (In the figure above, we approximate the laser pulse, the dark curve, by the shaded area.) Therefore, we interpret the t occurring here as an effective pulse length; it is much shorter than the actual laser pulse length. We see that γ_{02} appears only as a part of Γ in Equation 1.4.7, where it is dwarfed by γ_{01} and $\bar{\gamma}$. Therefore, our model predicts that, within its range of validity, one *cannot* observe the direct process from analysis of yield curves alone.

There is another possible explanation of the "knee" which our model could pro-

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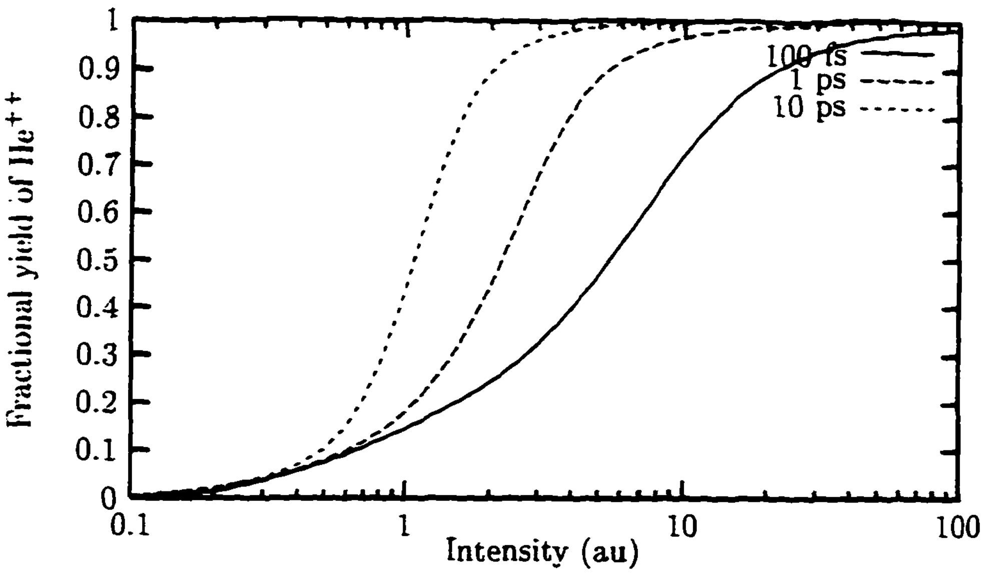

Figure 2.7: Fractional yield of He^{++} versus intensity for effective laser pulse lengths of 100 fs, 1 ns and 10 ns. Intensity is measured in atomic units of intensity $I_0 = 3.50 \times 10^{16}$ W/cm^2

vide. Suppose that there were competition between the two sequential processes. If one process were dominant for some regime of intensity and the other process dominant in another regime, then there would be a break in the slope of n_2—a "knee". For the range of I where this model is applicable, however, we have $\gamma_{01} > \bar{\gamma}$ and so this explanation is not valid.

Limitation of our model

Our model has one very serious drawback. The exponentially small factors in the GBFs make our ionization yields come out (numerically) orders of magnitude smaller[†]

Yield Rates (in au)				
	Kulander	Calculated		
Intensity (au)		100 fs	1 ns	10 ns
0.0284	**8.41 × 10^{29}**	6.51×10^{-9}	5.55×10^{-6}	2.03×10^{-3}
0.0227	**2.72 × 10^{29}**	2.93×10^{-11}	2.82×10^{-8}	1.97×10^{-5}

Table 2.4: Experimental ionization yields versus our calculated yields from Equation 2.6.1

than experimentally-measured rates. The yield is given, using the approximation of a sudden-on-sudden-off laser pulse, is:

$$n_2(I) = \frac{1}{t_{\text{pulse}}} \int_0^{t_{\text{pulse}}} dt\, n_2(I,t) \qquad 2.6.1$$

[†] Actually, orders of magnitude of orders of magnitude.

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where $n_2(I, t)$ is given in Equation 1.4.7. Kulander[22] provided us with his double ionization yield data, which we compare with our predicted yields. The results, for three different pulse lengths, are provided in Table 2.4.

We must point out that these data were for lasers outside of the regime of validity of our model (ω = 780 nm = 1.59 eV and ω = 616 nm = 2.012 eV) and so direct comparison with existing experimental data is speculative.

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Chapter 3
Conclusions

In Chapter 1, we set forth a project to look for a signature of correlation in multiphoton double ionization of atoms. We developed a definition of sequential and direct double ionization processes. We then connect the macroscopic (measurable) quantities, the fractional ionization yields, to microscopic (quantum-mechanical) properties. The two are connected through a generalization of the Fermi Golden Rule derivation for single ionization: singularities as a function of energy of the S-matrix describing the ionization process connect directly to the various ionization rates through the solutions of the coupled ODEs describing the fractional yield rates (Equations 1.4.7).

We find that the uncorrelated parts of the ionization process interfere with the correlated parts (Equation 1.4.9). In addition, our sequential process itself requires an accounting for the dynamics of both electrons, through the new "HF" state $\bar{X}$.

In writing down the S-matrix describing the ionization process we have approximated our full interaction by an expansion in powers of the screened interelectronic Coulomb interaction. This makes the connection between the S-matrix terms and the ionization parameters inexact. This is a reflection of the fact that the two processes themselves are in fact very difficult to distinguish. If both intermediate steps in the sequential path $X_1 \rightarrow X_2 \rightarrow X_3$ is slow (*i.e.* long lived) then the process will be viewed as sequential. If one rate is fast, then the process will be viewed as direct.

We define a particular signature of correlation, the *turnover time* t_t, which is a measure of how long it takes for the direct process to be dominated by the sequential processes.

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In Chapter 2, we carry out the analysis from Chapter 1 using a lowest-order First Born Approximation (FBA) to calculate the double ionization rates for helium. We approximate the ground state of helium by the simplest approximation to the Hartree-Fock orbital, and calculate the rates in an expansion in the correlation interaction. This method makes the various rates be in terms of Generalized Bessel Functions (GBFs). In order to evaluate these terms analytically, we use an asymptotic (saddle-point) approximation for the GBFs. In turn, this enforces a strict upper bound on laser frequency and upper and lower bounds on laser intensity. The return on this is a relatively simple analytic form for the various rate parameters (γ's), which are independent of laser frequency.

We find some surprising results for the rates themselves. In the regime of applicability of our theory, the sequential rate is dominated by a path which does not go through the singly-charged ion ($X \rightarrow X^{+} \rightarrow X^{++}$) but rather through a Hartree-Fock state ($X \rightarrow \tilde{X} \rightarrow X^{++}$). It is identified with a shake-off description. This is a direct result of our approximating the ground state as a product of variational approximations to the HF ground state ($u_0 u_0$).

The direct term also gives rise to new results. We found that the final state correlation—or "boomerang" (Corkum) term—dominated over the initial state correlation term, but only by a factor of approximately 6. We found our predicted ionization rates to be much smaller than the experimentally-determined rates just outside of the range of validity of our theory. We also found that our signature of correlation, the turnover time, is far too small to be used experimentally now. Likewise, we show that direct observation of yield curves alone, without other corroboration, like angular correlation, cannot stand as evidence of correlation.

A letter by Watson, *et al.*[23] was recently brought to our attention. In it, they claim to validate the work of Walker and his group[20]. They use the idea of an expansion in a correlation interaction but do not take the care that we do in defining it, and their work lies outside the regime of validity of our own.

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Appendix 1

The Method of Stationary Closure

In general, the problem of performing a sum-over-states (like the one appearing in Equation 2.3.1) is extremely difficult, if not impossible, to perform in a closed form, because of the presence of the energy of the state over which one wants to sum in the denominator appearing in the sum. If that sum-index dependence were not present, then the sum would simply sum over a complete set of eigenstates, which is a resolvent of the identity. The trick of Stationary Closure involves replacing index-dependent energy in the denominator by an "average" energy which is index-independent, at which point the sum can be done immediately. We proceed as follows:

Consider the general problem of finding a matrix element

$$\langle A|\hat{V}|B\rangle \tag{A1.1}$$

where $\hat{V}$ is some potential (or any general Hermitian operator) and where $|B\rangle$ is a sum-over-states resulting from a perturbation-series calculation.

$$B = {\sum_{\xi}}' \frac{|\xi\rangle\langle\xi|\hat{U}|C\rangle}{W_\xi - W_C} \tag{A1.2}$$

and for some appropriate Hamiltonian $\mathcal{H}$ we have $\mathcal{H}|\xi\rangle = W_\xi|\xi\rangle$ and $\mathcal{H}|C\rangle = W_C|C\rangle$. We can also write B in terms of Green's functions:

$$B = G\hat{U}C$$

$$\left(i\frac{\partial}{\partial t} - \mathcal{H}\right) G(t,t') = \delta\left(t - t'\right) \tag{A1.3}$$

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Note that were it not for the denominator in this sum, we could simply use the fact that the $|\xi\rangle$ form a complete basis in the Hilbert space and replace the sum with the identity operator†. Now, introduce an "average energy" $\mathcal{E}$ and rewrite the denominator as

$$\frac{1}{W_\xi - \mathcal{E} + \mathcal{E} - W_C}. \qquad \text{A1.4}$$

If we make the (not necessarily good) assumption that $\mathcal{E} \approx W_\xi$ in some manner, then we can expand the denominator in the following way:

$$\frac{1}{W_\xi - W_C} \approx \frac{1}{\mathcal{E} - W_C} \times \left(1 - \frac{W_\xi - \mathcal{E}}{\mathcal{E} - W_C} + \cdots\right) \qquad \text{A1.5}$$

If we substitute this back into the summation, we notice that the term W_ξ is no longer in the denominator of the sum. To determine $\mathcal{E}$, we simply require that the first-order term in this sub-expansion vanish. That is, it should be *stationary* with respect to *closure*. Hence:

$$\sum_\xi{}' \frac{\langle A|\hat{V}|\xi\rangle\langle\xi|(\mathcal{H} - \mathcal{E})\hat{U}|C\rangle}{\mathcal{E} - W_C} = 0. \qquad \text{A1.6}$$

(We have made the immediate substitution that $\langle\xi|W_\xi = \langle\xi|\mathcal{H}$.) We can now use the fact that the $|\xi\rangle$'s form a complete set (modulo the element $|C\rangle$) to immediately write

$$\mathcal{E} = \frac{\langle A|\hat{V}\mathcal{H}\hat{V}|C\rangle}{\langle A|\hat{V}\hat{U}|C\rangle}. \qquad \text{A1.7}$$

Moreover, if we write the identity $W_C = \langle A|\hat{V}\hat{U}\mathcal{H}|C\rangle/\langle A|\hat{V}\hat{U}|C\rangle$, then $1/(\mathcal{E} - W_C)$ becomes

$$\frac{1}{\mathcal{E} - W_C} = \frac{\langle A|\hat{V}\hat{U}|C\rangle}{\langle A|\hat{V}[\mathcal{H},\hat{U}]|C\rangle} \cdots \qquad \text{A1.8}$$

and therefore our total matrix element is:

$$\langle A|\hat{V}|B\rangle = \frac{|\langle A|\hat{V}\hat{U}|C\rangle|^2}{\langle A|\hat{V}[\mathcal{H},\hat{U}]|C\rangle}. \qquad \text{A1.9}$$

In the most general case, there is an added complication: instead of $\langle A|\hat{V}\mathcal{H}\hat{U}|C\rangle$ in the numerator of Equation A1.7 one gets $\langle A|\hat{V}\mathcal{H}\hat{U}|C\rangle - \langle C|\hat{V}\mathcal{H}\hat{U}|C\rangle$ and likewise in the denominator one would get $\langle A|\hat{V}\hat{U}|C\rangle - \langle C|\hat{V}\hat{U}|C\rangle$.

---

† Of course, the $\sum'$ means to exclude the state $|C\rangle$ from the summation. In the general case, we can simply add in this state later and subtract it out (adding zero) and then have the sum over a complete set. However, in our problem, as we have seen, this is unnecessary, as we will be removing the restriction on the sum.

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We apply this technique to a sample problem. the electric polarizability of hydrogen. which can be calculated explicitly by solving an inhomogenous ODE§. (the method of Dalgarno and Lewis) to estimate how well or poorly this method approximates the exact answer. This problem is not precisely the same as the one we encounter in Section 2.3. but the idea (to eliminate the sum-over-states) is the same and is instructive nontheless. The polarizability is, in second-order perturbation theory:

$$w_2 = \sum_n{}' \frac{|\langle n|H'|0\rangle|^2}{e_0 - e_n} \qquad A1.10$$

where H' is the interaction Hamiltonian:

$$H' = e\vec{E}\cdot\vec{r} \qquad A1.11$$

and the sum is over the hydrogen wavefunctions. In this case, we must keep the $\langle 0|H'|0\rangle$ term (because we cannot omit this term like we do in our two-electron case) but this term is zero due to parity, so it can be ignored.

The requirement that the first-order term in Equation A1.5 vanish is:

$$\sum_n{}' \mathcal{E}\langle 0|H'|n\rangle\langle n|H'|0\rangle - \langle 0|H'H_0|n\rangle\langle n|H'|0\rangle = 0 \qquad A1.12$$

(This follows directly from Equation A1.6.) We have made the identification that $\hat{V} = \hat{U} = H'$, and H_0 is the regular hydrogen-atom Hamiltonian, $-\frac{\hbar^2}{2m_e}\nabla^2 - \frac{e^2}{r}$. Since the sum over n is a complete set (we add in the $\langle 0|H'|0\rangle$ term, which is zero), the definition of $\mathcal{E}$ is:

$$\mathcal{E} = \frac{\langle 0|H'H'|0\rangle}{\langle 0|H'H_0|0\rangle} \qquad A1.13$$

and then $w_{2,sc}$ is:

$$w_{2,sc} = \frac{-2\,|\langle 0|H'^2|0\rangle|^2}{\langle 0|\,[H',[H_0,H']]\,|0\rangle} \qquad A1.14$$

(The -2 occurs due to the way we rewrite the denominator using the second commutator). The matrix elements appearing are:

$$\langle 0|H'^2|0\rangle = e^2E^2{a_0}^2 \qquad A1.15$$

$$\langle 0|\,[H',[H_0,H']]\,|0\rangle = \frac{\hbar^2}{m_e}e^2E^2 \qquad A1.16$$

---

§ See Schiff[6] §33

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We express $[H', [H_0, H']] = -e^2E^2[z, [T, z]] = (\hbar/m_e)e^2E^2$, where z is the z-direction position operator and T is the kinetic energy operator.

Therefore, $w_{2\_sc} = -2E^2a_0{}^3$, as compared to the exact answer obtained by Schiff of $w_2 = -\frac{9}{4}E^2a_0{}^3$. The Stationary Closure method produces an energy which is incorrect by 12.5%.

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Appendix 2

Digression: The Generalized Bessel Function (GBF)

The Generalized Bessel Function (GBF) is defined as[24]†:

$$\mathcal{J}_{-n}(a,b) \overset{\text{def}}{=} \int_{-\pi}^{\pi} \frac{\mathrm{d}\phi}{2\pi}\, \mathrm{e}^{\mathrm{i}(n\phi + a\sin\phi + b\sin 2\phi)}. \qquad A2.1$$

Note that, like the regular Bessel functions, the GBFs are also always real for real arguments. We can derive a recurrence relation for the GBFs by using the periodicity of their integral representation. Taking $\mathrm{d}/\mathrm{d}\phi$ inside the integral sign makes a perfect differential and therefore $\int \mathrm{d}\phi(\mathrm{d}/\mathrm{d}\phi)\exp(\cdots) = 0$. This gives immediately:

$$0 = \int_{-\pi}^{\pi} \mathrm{d}\phi\, \mathrm{e}^{\mathrm{i}(n\phi + a\sin\phi + b\sin 2\phi)}\; \mathrm{i}\,(n + a\cos\phi + 2b\cos 2\phi), \qquad A2.2a$$

from which we get the 5-term recurrence:

$$0 = -n\mathcal{J}_{-n}(a,b) + a\left(\mathcal{J}_{-n-1}(a,b) + \mathcal{J}_{-n+1}(a,b)\right) + 2b\left(\mathcal{J}_{-n-2}(a,b) + \mathcal{J}_{-n+2}(a,b)\right). \qquad A2.2b$$

In addition to this, the GBFs satisfy closure relations, which can be seen from their integral representations. To wit:

$$\sum_{\nu} \mathcal{J}_{s-\nu}(a',b)\, \mathcal{J}_{\nu-N}(a'',b) = \mathcal{J}_{s-N}(a'+a'',2b). \qquad A2.3$$

---

† Leubner (*loc. sit.*) also includes a third parameter θ as an argument to the last sin: $\ldots + b\sin(2\phi - \theta)$. Both we and Leubner set $\theta = 0$ uniformly here.

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To see this, consider the integral representation of the sum:

$$\sum_{\nu} \mathcal{J}_{s-\nu}(a', b)\, \mathcal{J}_{\nu-N}(a'', b) = \sum_{\nu} \int_{\pi}^{\pi} \int_{\pi}^{\pi} \frac{d\phi_1 d\phi_2}{(2\pi)^2} e^{-i(s-\nu)\phi_1 - i(\nu-N)\phi_2} \times$$
$$e^{-ia' \sin\phi_1 - ia'' \sin\phi_2}\, e^{-ib(\sin 2\phi_1 + \sin 2\phi_2)}$$

Now we recall one of the forms of the δ-function is, mod 2π,

$$2\pi\delta(x) = \sum_n \exp inx, \qquad \text{A2.4}$$

and so we can use this to eliminate the sum over ν and turn it into $\delta(\phi_1 - \phi_2)$. This allows us to immediately perform one of the ϕ integrals, and the result is:

$$= \int_{\pi}^{\pi} \frac{d\phi_1}{2\pi} e^{-i(s-N)\phi_1}\, e^{-i(a'+a'')\sin\phi_1}$$
$$\times\, e^{-i2b \sin 2\phi_1} \qquad \text{A2.5}$$

and this gives us our desired result. Using similar algebra, we can derive a second closure relation:

$$\sum_{\nu} \mathcal{J}_{s-\nu}(a', b)\, \mathcal{J}_{\nu-N}(a'', b)\nu = \frac{s+N}{2} \mathcal{J}_{s-N}(a' + a'', 2b)$$
$$+ \frac{a'}{2} \left(\mathcal{J}_{s-N+1}(a' + a'', 2b) + \mathcal{J}_{s-N-1}(a' + a'', 2b) \right). \qquad \text{A2.6}$$

This comes from the identity

$$\sum_n n\, e^{inx} = -i\frac{\partial}{\partial x} \sum_n e^{inx} = -2\pi i \frac{\partial}{\partial x} \delta(x) \qquad \text{A2.7}$$

Recall that the derivative of a δ-function is defined—like the δ-function itself—only in a "distribution" sense†:

$$\int dx\, \delta'(x)\, f(x) \overset{\text{def}}{=} -f'(0) \qquad \text{A2.8}$$

Using Equations A2.8 and A2.2 we arrive at Equation A2.6

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† See Geroch's text[25], ch. 47. Like the regular δ-function, the integral is zero if the interval of integration does not include 0.

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