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Fachbereich Mathematik

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January 8, 2010

Abstract

In this paper we present a mathematical analysis of the photoelectric effect for one-electron atoms in the framework of non-relativistic QED. We treat photo-ionization as a scattering process where in the remote past an atom in its ground state is targeted by one or several photons, while in the distant future the atom is ionized and the electron escapes to spacial infinity. Our main result shows that the ionization probability, to leading order in the fine-structure constant, α, is correctly given by formal time-dependent perturbation theory, and, moreover, that the dipole approximation produces an error of only sub-leading order in α. In this sense, the dipole approximation is rigorously justified.

1 Introduction

Even today, more than 100 years after its discovery by Hertz, Hallwachs and Lenard, the phenomenon of photoionization is still investigated, both experimentally and theoretically [4, 1]. This research is driven by novel experimental techniques that allow for the production of very strong and ultrashort laser pulses. In contrast, the photo electric effect in the early experiments is produced by weak, non-coherent radiation of high frequency. There is a third physical regime, where the radiation is weak, of high frequency, and *coherent*. This regime is the subject of the present paper. We consider one-electron atoms within the standard model of non-relativistic QED, and we present a mathematically rigorous analysis of the ionization process caused by the impact of finitely many photons. Improving on earlier results concerning more simplified models, we show that the probability of ionization, to leading order in the fine-structure constant, is proportional to the number of photons, and, in the case of a single photon, it is given correctly by the rules of formal (time-dependent) perturbation theory. It turns out that the dipole approximation produces an error of subleading order, which provides a rigorous justification of this popular approximation.

Let's briefly recall the standard model of one-electron atoms within non-relativistic QED. More elaborate descriptions may be found elsewhere [20, 31]. States of arbitrarily

many transversal photons are described by vectors in the symmetric Fock space

$$\mathcal{F} := \bigoplus_{n \geq 0} S_n \big[\otimes^n L^2(\mathbb{R}^3 \times \{1, 2\}) \big]$$

over $L^2(\mathbb{R}^3 \times \{1, 2\})$. Here S_n denotes the projection of $L^2(\mathbb{R}^3 \times \{1, 2\})^n$ onto the subspace of all symmetric functions of $(\mathbf{k}_1, \lambda_1), \ldots, (\mathbf{k}_n, \lambda_n) \in \mathbb{R}^3 \times \{1, 2\}$, and $S_0 L^2(\mathbb{R}^3 \times \{1, 2\}) := \mathbb{C}$. We shall use Ω to denote the vacuum vector $(1, 0, \ldots) \in \mathcal{F}$. N_f is the number operator in $\mathcal{F}$, and $H_f = \mathrm{d}\Gamma(\omega)$ denotes the second quantization of multiplication with $\omega(k) = |k|$ in $L^2(\mathbb{R}^3 \times \{1, 2\})$. See [27], X.7, for the notation $\mathrm{d}\Gamma(\cdot)$ and for an introduction to second quantization. The creation and annihilation operators $a^*(h)$ and $a(h)$, for $h \in L^2(\mathbb{R}^3 \times \{1, 2\})$, are densely defined, closed operators with $a^*(h) = a(h)^*$ and with

$$[a^*(h)\Psi]^{(n)} = \sqrt{n} S_n(h \otimes \Psi^{(n-1)})$$

for vectors $\Psi = (\Psi^{(0)}, \Psi^{(1)}, \ldots)$ from the subspace $D(N_f^{1/2})$. Here, $\Psi^{(n)}$ denotes the n-photon component of Ψ.

The system studied in this paper is composed of a non-relativistic, (spinless) quantum mechanical, charged particle (the electron), and the quantized radiation field which is coupled to the electron by minimal substitution. In addition, there is an external potential V, which may be due to a static nucleus. The Hilbert space is thus the tensor product

$$\mathcal{H} := L^2(\mathbb{R}^3) \otimes \mathcal{F},$$

and the Hamiltonian is of the form

$$\begin{aligned} H_\alpha &= (\mathbf{p} + \alpha^{\frac{3}{2}} \mathbf{A}(\alpha\mathbf{x}))^2 + V + H_f \\ &= H_0 + W, \end{aligned} \tag{1.1}$$

where $H_0 = H_{\mathrm{el}} + H_f$, $H_{\mathrm{el}} = -\Delta + V$, and $W = H_\alpha - H_0$. The quantized vector potential $\mathbf{A}(\alpha\mathbf{x})$, for each $\mathbf{x} \in \mathbb{R}^3$, is a triple of self-adjoint operators, each of which is a sum of a creation and an annihilation operator. Explicitly,

$$\mathbf{A}(\alpha\mathbf{x}) = a(\mathbf{G}_\mathbf{x}) + a^*(\mathbf{G}_\mathbf{x}), \qquad \mathbf{G}_\mathbf{x}(\mathbf{k}, \lambda) := \frac{\kappa(\mathbf{k})}{\sqrt{2|\mathbf{k}|}} \varepsilon(\mathbf{k}, \lambda) e^{-i\alpha \mathbf{k} \cdot \mathbf{x}}, \tag{1.2}$$

where $\varepsilon(\mathbf{k}, \lambda) \in \mathbb{R}^3$, $\lambda = 1, 2$, are orthonormal polarization vectors perpendicular to $\mathbf{k}$, and κ is an ultraviolet cutoff chosen from the space $\mathcal{S}(\mathbb{R}^3)$ of rapidly decreasing functions. No infrared cutoff is needed. Here and henceforth, the position of the electron, $\mathbf{x} \in \mathbb{R}^3$, and the wave vector of a photon, $\mathbf{k} \in \mathbb{R}^3$, are dimensionless and related to the corresponding dimensionfull quantities $\mathbf{X}, \mathbf{K}$ by $\mathbf{X} = (a_0/2)\mathbf{x}$ and $\mathbf{K} = (2\alpha/a_0)\mathbf{k}$, where $a_0 := \hbar^2/me^2$ is the Bohr-radius, $m > 0$ is the mass of the particle, e its charge, and $\alpha = e^2/\hbar c$ is the fine structure constant. It follows that $\mathbf{X} \cdot \mathbf{K} = \alpha \mathbf{x} \cdot \mathbf{k}$, and in units where $\hbar$, c, and four times the Rydberg energy $2m\alpha^2$ are equal to unity, the Hamiltonian of a one-electron atom with static nucleus at the origin takes the form (1.1) with $V(\mathbf{x}) = -Z/|\mathbf{x}|$, Z being the atomic number of the nucleus. For simplicity, we confine ourselves, in this introduction, to this particular potential V. In nature, $\alpha \approx 1/137$, but in this paper α is treated as a free parameter that can assume any non-negative value.

For all $\alpha \geq 0$, the Hamiltonian H_α is self-adjoint on $D(H_0)$ and its spectrum $\sigma(H_\alpha)$ is a half-axis $[E_\alpha, \infty)$ [24, 23]. Moreover,

$$E_\alpha := \inf \sigma(H_\alpha)$$

is an eigenvalue of H_α, and, at least for α sufficiently small, this eigenvalue is simple [2, 21]. We use Φ_α to denote a normalized eigenvector associated with E_α. Another important point in the spectrum of H_α is the ionization threshold Σ_α, which, for our system, is given by $\Sigma_\alpha = \inf \sigma(H_\alpha - V)$. In a state vector from the spectral subspace $\mathrm{Ran}\mathbf{1}_{(-\infty, \Sigma_\alpha)}(H_\alpha)$, the electron is exponentially localized in the sense that

$$\|e^{\beta|x|}\mathbf{1}_{(-\infty, \Sigma_\alpha - \varepsilon]}(H_\alpha)\| < \infty \tag{1.3}$$

for all β with $\beta^2 < \varepsilon$ [19].

The phenomenon of photo-ionization can be considered as a scattering process, where in the limit $t \to -\infty$, the atom in its ground state is targeted by a (finite) number of asymptotically free photons, while in the limit $t \to \infty$ the atom is ionized in a sense to be made precise. We begin by discussing *incoming scattering states* and their properties. To this end it is convenient to introduce the space $L^2_\omega(\mathbb{R} \times \{1, 2\})$ of all those $f \in L^2(\mathbb{R} \times \{1, 2\})$ for which

$$\|f\|^2_\omega := \sum_{\lambda=1,2} \int |f(\mathbf{k}, \lambda)|^2 \big(1 + \omega(\mathbf{k})^{-1}\big) d^3 k < \infty. \tag{1.4}$$

Given $f \in L^2_\omega(\mathbb{R} \times \{1, 2\})$, the asymptotic creation operator $a^*_-(f)$ is defined by

$$a^*_-(f)\Psi := \lim_{t \to -\infty} e^{iH_\alpha t} a^*(f_t) e^{-iH_\alpha t} \Psi, \qquad f_t := e^{-i\omega t} f, \tag{1.5}$$

and its domain is the space of all vectors $\Psi \in D(|H_\alpha|^{1/2})$ for which the limit (1.5) exists. This is know to be the case, e.g., for the ground state $\Psi = \Phi_\alpha$. Moreover, it is known that $a^*_-(f_1) \cdots a^*_-(f_n)\Phi_\alpha$ is well defined and that

$$\begin{aligned} & e^{-iH_\alpha t} a^*_-(f_1) \cdots a^*_-(f_n)\Phi_\alpha \\ = \ & a^*(f_{1,t}) \cdots a^*(f_{n,t}) e^{-iH_\alpha t} \Phi_\alpha + o(1), \qquad (t \to -\infty), \end{aligned} \tag{1.6}$$

whenever $f_i, \omega f_i \in L^2_\omega(\mathbb{R} \times \{1, 2\})$ for all $i = 1, \ldots, n$ [22]. By (1.6), $a^*_-(f_1) \cdots a^*_-(f_n)\Phi_\alpha$ describes a scattering state, which, in the limit $t \to -\infty$ is composed of the atom in its ground state and n asymptotically free photons with wave functions $f_1, \ldots, f_n$. Results analogous to those on $a^*_-(f)$ hold true for the asymptotic annihilation operators $a_-(f)$ [22].

The asymptotic annihilation and creation operators satisfy the usual canonical commutation relations: e.g.

$$[a_-(f), a^*_-(g)] = \langle f, g \rangle \tag{1.7}$$

for all $f, g \in L^2_\omega(\mathbb{R}^3 \times \{1, 2\})$. Moreover, the ground state Φ_α is a vacuum vector for asymptotic annihilation operators in the sense that

$$a_-(f)\Phi_\alpha = 0 \qquad \text{for all } f \in L^2_\omega(\mathbb{R}^3). \tag{1.8}$$

Hence, if $\underline{f} = (f_1, m_1, \ldots, f_n, m_n) \in \left[L^2(\mathbb{R}^3 \times \{1, 2\}) \times \mathbb{N} \right]^n$ with $\langle f_i, f_j \rangle = \delta_{ij}$, then it follows from (1.7) and (1.8) that

$$a_-^*(\underline{f})\Phi_\alpha := \prod_{k=1}^n \frac{1}{\sqrt{m_k!}} a_-^*(f_k)^{m_k} \Phi_\alpha \tag{1.9}$$

is a normalized vector in $\mathcal{H}$. All these properties of $a_-(f)$, $a_-^*(f)$ hold mutatis mutandis for the asymptotic operators $a_+(g)$, $a_+^*(g)$ defined in terms of the limit $t \to +\infty$.

We are interested in the probability that $e^{-iH_\alpha t} a_-^*(\underline{f})\Phi_\alpha$ describes an ionized atom in the distant future, but we are not interested in the asymptotic state of the electron or the radiation field in the limit $t \to +\infty$. We therefore shall not attempt to construct outgoing scattering states describing an ionized atom, which is a difficult open problem. Instead we base our computation of the probability of ionization on the following reasonable assumption: the atom described by $e^{-iH_\alpha t} a_-^*(\underline{f})\Phi_\alpha$ is either ionized in the limit $t \to \infty$, or else, in that limit, it *relaxes to the ground state* in the sense that $e^{-iH_\alpha t} a_-^*(\underline{f})\Phi_\alpha$, for t large enough, is well approximated by a linear combination of vectors of the form

$$a^*(g_{1,t}) \ldots a^*(g_{n,t}) e^{-iE_\alpha t} \Phi_\alpha. \tag{1.10}$$

More precisely, relaxation to the ground state occurs if $a_-^*(\underline{f})\Phi_\alpha$ belongs to the closure of the span of all vectors of the form

$$a_+^*(g_1) \ldots a_+^*(g_n)\Phi_\alpha = \lim_{t \to +\infty} e^{iH_\alpha t} a^*(g_{1,t}) \ldots a^*(g_{n,t}) e^{-iE_\alpha t}\Phi_\alpha,$$

with $g_i, \omega g_i \in L_\omega^2(\mathbb{R}^3 \times \{1, 2\})$. Let $\mathcal{H}_+^\alpha$ denote this space and let P_+^α be the orthogonal projection onto $\mathcal{H}_+^\alpha$. Then $\|P_+^\alpha a_-^*(\underline{f})\Phi_\alpha\|^2$ is the probability for relaxation to the ground state and

$$1 - \|P_+^\alpha a_-^*(\underline{f})\Phi_\alpha\|^2 = \|(1 - P_+^\alpha) a_-^*(\underline{f})\Phi_\alpha\|^2 \tag{1.11}$$

is the probability of ionization.

The assumption that relaxation to the ground state is the only alternative to ionization, is motivated by the conjecture of *asymptotic completeness for Rayleigh scattering*, which is the property, that every vector $\Psi \in \mathcal{H}$ describing a bound state in the sense that $\sup_t \|e^{\varepsilon|x|} e^{-iH_\alpha t}\Psi\| < \infty$ for some $\varepsilon > 0$, will relax the ground state in the limit $t \to \infty$. In view of (1.3), asymptotic completeness for Rayleigh scattering implies that $\mathcal{H}_+^\alpha \supseteq \mathbf{1}_{(-\infty, \Sigma_\alpha)}(H_\alpha)$, which can be proven for simplified models of atoms [30, 8, 18, 14].

The following two theorems will allow us to compute (1.11).

Theorem 1.1. *Suppose that* $f_1, \ldots, f_n \in L^2(\mathbb{R}^3 \times \{1, 2\})$ *where* $\sum_{\lambda=1}^2 \varepsilon(\cdot, \lambda) f_i(\cdot, \lambda)$ *belongs to* $C_0^2(\mathbb{R}^3 \backslash \{\mathbf{0}\}, \mathbb{C}^3)$ *for each* i, *and let* $\underline{f} = (f_1, \ldots, f_n)$. *Then:*

$$a_-^*(\underline{f})\Phi_\alpha = a_+^*(\underline{f})\Phi_\alpha - i\alpha^{3/2} \int_{-\infty}^\infty 2\mathbf{p}(s)\varphi_{el} \otimes [\mathbf{A}(\mathbf{0}, s), a^*(\underline{f})]\Omega \, ds + \mathcal{O}(\alpha^{5/2}) \tag{1.12}$$

where $\mathbf{p}(s) = e^{iH_{el}s} \mathbf{p} e^{-iH_{el}s}$ *and* $\mathbf{A}(\mathbf{0}, s) = e^{iH_f s} \mathbf{A}(\mathbf{0}) e^{-iH_f s}$.

The first term of (1.12) gives no contribution to the ionization probability (1.11) because $a_+^*(\underline{f})\Phi_\alpha \in \mathcal{H}_+^\alpha$. The second term is proportional to $\alpha^{3/2}$ and it is due to scattering processes where one of the n photons $f_1, \ldots, f_n$ is absorbed. The remainder terms are of order $\mathcal{O}(\alpha^{5/2})$ and stem from the dipole approximation $\mathbf{A}(\alpha\mathbf{x}) \to \mathbf{A}(\mathbf{0})$, from dropping $\alpha^3 \mathbf{A}(\mathbf{x})^2$ and from ignoring processes of higher order in $\alpha^{3/2}$. To isolate the contribution of order α^3 from (1.11) using (1.12), we need:

Theorem 1.2. *Suppose that $\mathcal{H}_+^\alpha \supseteq \mathbf{1}_{(-\infty, \Sigma_\alpha)}(H_\alpha)$ for α in a neighborhood of 0, and suppose that H_{el} has only negative eigenvalues. Then*

$$\lim_{\alpha \to 0} P_+^\alpha = \mathbf{1}_{pp}(H_{\mathrm{el}}) \tag{1.13}$$

in the strong operator topology.

Combining Theorem 1.1 and Theorem 1.2 we see that

$$\|(1 - P_+^\alpha)a_-^*(\underline{f})\Phi_\alpha\|^2 = \|(1 - P_+^\alpha)\Big(a_-^*(\underline{f}) - a_+^*(\underline{f})\Big)\Phi_\alpha\|^2$$

$$= \|\mathbf{1}_c(H_{\mathrm{el}})\Big(a_-^*(\underline{f}) - a_+^*(\underline{f})\Big)\Phi_\alpha\|^2 + o(\alpha^3)$$

$$= \alpha^3 \|\mathbf{1}_c(H_{\mathrm{el}}) \int_{-\infty}^\infty \mathbf{p}(s)\varphi_{el} \otimes [\mathbf{A}(\mathbf{0}, s), a^*(\underline{f})]\Omega\, ds\|^2 + o(\alpha^3)$$

where $\mathbf{1}_c(H_{\mathrm{el}}) = 1 - \mathbf{1}_{pp}(H_{\mathrm{el}})$, and where the second equation is justified by the α dependence of $a_-^*(\underline{f})\Phi_\alpha - a_+^*(\underline{f})\Phi_\alpha$ as given by (1.12). We are now going to express the coefficient of α^3 in terms of generalized eigenfunctions of H_{el}, which makes it explicitly computable in simple cases. A general and sufficient condition for the existence of a complete set of generalized eigenfunctions is the existence and completeness of a (modified) wave operator Ω_+ associated with H_{el}. This condition is satisfied for our choice of V. It means that there exists an isometric operator $\Omega_+ \in \mathcal{L}(\mathcal{H}_{el})$ with $\mathrm{Ran}\Omega_+ = \mathbf{1}_c(H_{\mathrm{el}})\mathcal{H}_{el}$ and $H_{\mathrm{el}}\Omega_+ = \Omega_+(-\Delta)$. In particular, the singular continuous spectrum of H_{el} is empty. Given the wave operator Ω_+ and the fact that $(H_{\mathrm{el}} - i)^{-1}\langle\mathbf{x}\rangle^{-2}$ is a Hilbert-Schmidt operator, it is easy to establish existence of generalized eigenfunctions $\varphi_\mathbf{q}$, $\mathbf{q} \in \mathbb{R}^3$, of H_{el} with the following properties [26]:

(i) The function $(\mathbf{x}, \mathbf{q}) \mapsto \langle\mathbf{q}\rangle^{-2}\langle\mathbf{x}\rangle^{-2}\varphi_\mathbf{q}(x)$ is square integrable on $\mathbb{R}^3 \times \mathbb{R}^3$, in particular $\langle\mathbf{x}\rangle^{-2}\varphi_\mathbf{q} \in L^2(\mathbb{R}^3)$ for almost every $\mathbf{q} \in \mathbb{R}^3$. $\langle\mathbf{x}\rangle := (1 + |\mathbf{x}|^2)^{1/2}$.

(ii) If $\psi \in D(|\mathbf{x}|^2)$ then

$$\|\mathbf{1}_{ac}(H_{\mathrm{el}})\psi\|^2 = \int_{\mathbb{R}^3} |\langle\varphi_\mathbf{q}, \psi\rangle|^2 d^3q \tag{1.14}$$

(iii) If $F : \mathbb{R} \to \mathbb{C}$ is a Borel function, $\psi \in D(|\mathbf{x}|^2) \cap D(F(H_{\mathrm{el}}))$, and $F(H_{\mathrm{el}})\psi \in D(|\mathbf{x}|^2)$, then

$$\langle\varphi_\mathbf{q}, F(H_{\mathrm{el}})\psi\rangle = F(\mathbf{q}^2)\langle\varphi_\mathbf{q}, \psi\rangle \tag{1.15}$$

for almost every $\mathbf{q} \in \mathbb{R}^3$.

In (ii) and (iii) we use $\langle\varphi_\mathbf{q}, \psi\rangle$ to denote the integral $\int \overline{\varphi_\mathbf{q}(\mathbf{x})}\psi(\mathbf{x})\, d^3x$, which is well defined by (i) and by the assumption $\psi \in D(|\mathbf{x}|^2)$.

The Theorem 1.1 in conjunction with (i)-(iii) implies the following theorem, which is our main result specialized to the case of only one asymptotic photon in the incident scattering state.

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Theorem 1.3. *For all $f \in L^2(\mathbb{R}^3 \times \{1,2\})$ with $\sum_{\lambda=1}^2 \varepsilon(\cdot,\lambda)f(\cdot,\lambda) \in C_0^2(\mathbb{R}^3 \backslash \{\mathbf{0}\}, \mathbb{C}^3)$,*

$$\left\| \mathbf{1}_{ac}(H_{\mathrm{el}}) \big(a_-^*(f)\Phi_\alpha - a_+^*(f)\Phi_\alpha \big) \right\|^2$$
$$= \alpha^3 \int_{\mathbb{R}^3} d^3q \left| \langle \varphi_{\mathbf{q}}, \mathbf{x}\varphi_{el} \rangle \cdot \int_{-\infty}^{\infty} e^{i(\mathbf{q}^2 - E_0)t} \langle \Omega, \mathbf{E}(\mathbf{0},t)a^*(f)\Omega \rangle \, dt \right|^2 + \mathcal{O}(\alpha^4) \quad (1.16)$$

as $\alpha \to 0$. Here, $\mathbf{E}(\mathbf{0},t) = -i[H_f, \mathbf{A}(\mathbf{0},t)]$, φ_{el} is a normalized ground state of H_{el} and $\varphi_{\mathbf{q}}$, $\mathbf{q} \in \mathbb{R}^3$, is any family of generalized eigenfunction of H_{el} with properties (i)-(iii) above.

The expression (1.16) for the ionization probability can be understood, *on a formal level*, by first order, time-dependent perturbation theory. To this end one considers the transitions $\varphi_{el} \otimes f \mapsto \varphi_{\mathbf{q}} \otimes \Omega$, for fixed $\mathbf{q} \in \mathbb{R}^3$, in the *interaction picture* defined by H_0. Then the time-evolution of state vectors is generated by the time-dependent interaction operator $W(t) = e^{iH_0 t} W e^{-iH_0 t} = 2\alpha^{3/2}\mathbf{p}(t) \cdot \mathbf{A}(\alpha\mathbf{x},t) + \alpha^3 \mathbf{A}(\alpha\mathbf{x},t)^2$ with $\mathbf{p}(t) = e^{iH_{\mathrm{el}}t}\mathbf{p}e^{-iH_{\mathrm{el}}t}$ and $\mathbf{A}(\alpha\mathbf{x},t) = e^{iH_0 t}\mathbf{A}(\alpha\mathbf{x})e^{-iH_0 t}$. In the computation of the transition amplitude to the order $\alpha^{3/2}$ one drops $\alpha^3 \mathbf{A}(\alpha\mathbf{x},t)^2$ and one replaces $\mathbf{A}(\alpha\mathbf{x},t)$ by $\mathbf{A}(\mathbf{0},t)$, which is known as the dipole approximation. Then, an integration by parts using that

$$2\mathbf{p}(t) = \frac{d}{dt}\mathbf{x}(t), \qquad -\frac{\partial}{\partial t}\mathbf{A}(\mathbf{0},t) = \mathbf{E}(\mathbf{0},t),$$

leads to a result for the transition amplitude which agrees with the expression in (1.16) whose modulus squared is integrated over $\mathbf{q} \in \mathbb{R}^3$. The Theorem 1.3 and its proof justify this formal derivation and the use of the dipole approximation. Note that $\alpha\mathbf{x} = \mathbf{X}$, hence the ionization probability is of order α^3 rather than of order α, as a formal computation, similar to the one above, in dimension-full quantities would suggest.

We prove a more general result than Theorem 1.3, where the incoming scattering state may contain several asymptotic photons, and where the external potential V is taken from a large class of long range potentials. In the case where the asymptotic state at $t = -\infty$ is of the form (1.9) and each of the photons $f_1, \ldots, f_n \in L^2(\mathbb{R}^3 \times \{1,2\})$ satisfies the hypotheses of Theorem 1.3, in addition to $\langle f_i, f_j \rangle = \delta_{ij}$, our result says that

$$\left\| \mathbf{1}_{ac}(H_{\mathrm{el}}) \big(a_-^*(\underline{f})\Phi_\alpha - a_+^*(\underline{f})\Phi_\alpha \big) \right\|^2 = \alpha^3 \sum_{l=1}^n m_l P^{(3)}(f_l) + \mathcal{O}(\alpha^4) \quad (1.17)$$

with

$$P^{(3)}(f_l) := \int_{\mathbb{R}^3} d^3q \left| \langle \varphi_{\mathbf{q}}, \mathbf{x}\varphi_{el} \rangle \cdot \int_{-\infty}^{\infty} \langle \Omega, \mathbf{E}(\mathbf{0},t)a^*(f_l)\Omega \rangle \, e^{i(\mathbf{q}^2 - E_0)t} dt \right|^2. \quad (1.18)$$

The integral with respect to t in (1.18) can be computed explicitly in terms of f_l and $\mathbf{G}_0$, and it gives

$$\int_{-\infty}^{\infty} e^{i(\mathbf{q}^2 - E_0)t} \langle \Omega, \mathbf{E}(\mathbf{0},t)a^*(f_l)\Omega \rangle \, dt$$
$$= i\pi \int_{|\mathbf{k}|=\mathbf{q}^2 - E_0} \kappa(\mathbf{k})\sqrt{2|\mathbf{k}|} \sum_{\lambda=1,2} \varepsilon_\lambda(\mathbf{k},\lambda)f_l(\mathbf{k},\lambda)d\sigma(\mathbf{k}),$$

where $d\sigma(\mathbf{k})$ is the surface measure of the sphere $\{\mathbf{k} \in \mathbb{R}^3 : |\mathbf{k}| = \mathbf{q}^2 - E_0\}$ in $\mathbb{R}^3$. The integration over the spheres with $|\mathbf{k}| = \mathbf{q}^2 - E_0$ expresses the conservation of energy in the scattering process, and the additivity (1.17) of the ionization probability with respect to the incoming photons corresponds to the experimental fact, that the number of photo-electrons is proportional to the intensity of the incoming radiation.

In Section 5 we give a second derivation of $\alpha^3 P_3(\underline{f})$ based on a space-time analysis of the ionization process. This approach, in a slightly different form, was introduced in the papers [3, 33], and does not assume asymptotic completeness of Rayleigh scattering.

The existence of outgoing scattering state describing an ionized atom and an electron escaping to spacial infinity is a difficult open problem in the model described above. Only for $V = 0$ such states have been constructed so far [25, 5]. Hence it is not possible yet to study the ionization probability based on transition probabilities between asymptotic states.

Previously ionization by *quantized* fields was investigated in [3, 16, 17, 33]. [3] and [33] are precursors of the present paper on simpler models of atoms and the ionization probability defined in a different, but equivalent way. In [16, 17] it is shown that a thermal quantized field leads to ionization in the sense of absence of an equilibrium state of atom and field.

There is a large host of mathematical results on ionization by *classical* electric fields: Schrader and various coauthors study the phenomenon of stabilization by providing upper and lower bounds on the ionization probability, see [10, 12, 11] and the references therein. They use the Stark-Hamiltonian with a time dependent electric field $\mathcal{E}(t)$ that vanishes unless $0 \leq t \leq \tau < \infty$. Lebowitz and various coauthors compute the probability of ionization by an electric field that is periodic in time; see [7, 29] and references therein. Most of these papers study one-dimensional Schrödinger operators with a single bound state that is produced by a δ-potential. Ionization in a three-dimensional model with a δ-potential is studied in [6].

2 Notations and Hypotheses

For easy reference, we collect in this section the definitions, our notations and all hypotheses. As usual, $L^2(\mathbb{R}^3 \times \{1, 2\})$ denotes the space of square integrable functions $f : \mathbb{R}^3 \times \{1, 2\} \to \mathbb{C}$ with inner product

$$\langle f, g \rangle := \sum_{\lambda=1,2} \int_{\mathbb{R}^3} \overline{f(\mathbf{k}, \lambda)} g(\mathbf{k}, \lambda) d^3 k.$$

We recall from the introduction that $L^2_\omega(\mathbb{R}^3 \times \{1, 2\})$ consists of those functions $f \in L^2(\mathbb{R}^3 \times \{1, 2\})$ for which the norm $\|f\|_\omega$ defined in (1.4) is finite. Regularity assumptions will be imposed on the vector-valued function

$$(\varepsilon f)(\mathbf{k}) := \sum_{\lambda=1}^{2} \varepsilon(\mathbf{k}, \lambda) f(\mathbf{k}, \lambda), \tag{2.1}$$

rather than on on $f(\cdot, 1)$ and $f(\cdot, 2)$. It is useless to impose smoothness conditions on $f(\cdot, \lambda)$ because it is (2.1) that matters and because the polarization vectors $\boldsymbol{\varepsilon}(\mathbf{k}, 1)$ and $\boldsymbol{\varepsilon}(\mathbf{k}, 2)$ are necessarily discontinuous. On the other hand, every square integrable function $f : \mathbb{R}^3 \to \mathbb{C}^3$ with $\mathbf{k} \cdot f(\mathbf{k})$, for a.e. $\mathbf{k} \in \mathbb{R}^3$, can be approximated, in the L^2-sense, by smooth functions of the form (2.1).

It is convenient to collect a family $f_1, ..., f_N \in L^2(\mathbb{R}^3 \times \{1, 2\})$ of photon wave functions in an N-tupel $\underline{f} = (f_1, ..., f_N)$. We define

$$\begin{aligned} a(\underline{f}) &:= a(f_1) \cdots a(f_N) \\ a^*(\underline{f}) &:= a^*(f_1) \cdots a^*(f_N). \end{aligned}$$

This should not lead to confusion with (1.9), where $\underline{f}$ also includes occupation numbers. For the various parts of the interaction operator $W = H_\alpha - H_0$, we use the notations

$$\begin{aligned} W^{\mathrm{dip}} &:= 2\mathbf{p} \cdot \mathbf{A}(\mathbf{0}), \\ W^{(1)} &:= 2\mathbf{p} \cdot \mathbf{A}(\alpha \mathbf{x}), \\ W^{(2)} &:= \mathbf{A}(\alpha \mathbf{x})^2. \end{aligned}$$

It follows that

$$W = \alpha^{\frac{3}{2}} W^{(1)} + \alpha^3 W^{(2)} = \alpha^{\frac{3}{2}} W^{\mathrm{dip}} + \mathcal{O}(\alpha^{\frac{5}{2}})$$

where the last equation is purely formal, but we shall give it a rigorous meaning in this paper. The Hamiltonian

$$H_\alpha = H_0 + W$$

is self-adjoint on the domain of $-\Delta + H_f$ provided that V is infinitesimally operator bounded with respect to $-\Delta$, [23, 24]. This is the case, e.g., if V is the sum of Coulomb potentials due to static nuclei; all our results are valid for such V. Nonetheless, it is useful to identify the properties of V that are essential for our analysis. From now on, we shall only assume the following hypotheses on V:

Hypotheses: *Both V and $\mathbf{x} \cdot \nabla V$ belong to $\in L^2_{loc}(\mathbb{R}^3)$, $\lim_{|\mathbf{x}| \to \infty} V(\mathbf{x}) = 0$, and there exist constants $\mu > 0$ and $R > 0$ such that for $|\beta| = 1, 2$ we have*

$$|\partial_{\mathbf{x}}^\beta V(\mathbf{x})| \le |\mathbf{x}|^{-|\beta| - \mu}, \qquad \textit{if } |\mathbf{x}| > R.$$

Moreover, $E_0 := \inf \sigma(H_{\mathrm{el}}) < 0$. We define $e_1 := \inf(\sigma(H_{\mathrm{el}}) \backslash \{E_0\})$.

From these Hypotheses it follows that $\sigma_{ess}(H_{\mathrm{el}}) = [0, \infty)$, that $\sigma_{sc}(H_{\mathrm{el}}) = \emptyset$ and that E_0 is a simple eigenvalue. In fact, the decay assumptions on V imply long-range asymptotic completeness [9], which is what we use to infer the existence of a complete set of generalized eigenfunctions. All this remains true if a singular short-range potential is added to H_{el}.

The time evolution of an operator B in the *interaction picture* will be denoted by $B(t)$, that is,

$$B(t) := e^{iH_0 t} B e^{-iH_0 t},$$

and $B_t := B(-t)$. Note that $\mathbf{p}(t) = e^{iH_{\mathrm{el}} t} \mathbf{p} e^{-iH_{\mathrm{el}} t}$, $\mathbf{A}(\mathbf{0}, t) = e^{iH_f t} \mathbf{A}(\mathbf{0}) e^{-iH_f t}$ and that $a^\#(\underline{f}_t) = e^{-iH_0 t} a^\#(\underline{f}) e^{iH_0 t} = a^\#(\underline{f})_t$.

3 Commutator estimates and scattering states

The main purpose of this section is to establish bounds on the commutators $[W^{(j)}, a^*(\underline{f}_t)]$ applied to Φ_α for $W^{(j)} \in \{W^{(1)}, W^{(2)}, W^{\mathrm{dip}}\}$. We are interested in the decay as $|t| \to \infty$ and in the dependence on α. Typically, our estimates are valid for $\alpha \leq \tilde{\alpha}$, where $\tilde{\alpha}$ is defined in Proposition A.3. As a simple application of our decay estimates in t, we will obtain existence of the scattering states

$$a_\pm^*(\underline{f})\Phi_\alpha = \lim_{t \to \pm} e^{itH_\alpha} a^*(\underline{f}_t) e^{-itH_\alpha} \Phi_\alpha,$$

which was already established in [22] in larger generality. Here $\underline{f}_t = (f_{1,t}, ..., f_{N,t})$ and $f_{j,t} := e^{-it\omega} f_j$. Given $l \in \{1, ..., N\}$, we write

$$
\begin{aligned}
a^*(\underline{f}_{[l],t}) &:= a^*(f_{1,t}) \cdots a^*(f_{l-1,t}) \mathbf{A}(\alpha \mathbf{x}) a^*(f_{l+1,t}) \cdots a^*(f_{N,t}), \\
a^*(\underline{f}_{(l),t}) &:= a^*(f_{1,t}) \cdots a^*(f_{l-1,t}) a^*(f_{l+1,t}) \cdots a^*(f_{N,t}).
\end{aligned}
$$

For $\mathbf{x} \in \mathbb{R}^3$, $\langle \mathbf{x} \rangle := (1 + |\mathbf{x}|^2)^{1/2}$.

Lemma 3.1. *Suppose that $f \in L^2(\mathbb{R}^3 \times \{1, 2\})$ with $\varepsilon f \in C_0^n(\mathbb{R}^3 \backslash \{\mathbf{0}\}, \mathbb{C}^3)$ for a given $n \in \mathbb{N}$. Then there exists is a constant $c_{1,n} = c_{1,n}(f)$ such that*

$$|\langle \mathbf{G_0}, f_t \rangle| \leq c_{1,n} \frac{1}{1 + |t|^n}, \tag{3.1}$$

$$|\langle \mathbf{G_x}, f_t \rangle| \leq c_{1,n} \frac{1 + (\alpha|x|)^n}{1 + |t|^n} \qquad \text{for all } \mathbf{x} \in \mathbb{R}^3, \tag{3.2}$$

$$|\langle \mathbf{G_x} - \mathbf{G_0}, f_t \rangle| \leq c_{1,n} \frac{\alpha|\mathbf{x}|\langle \alpha \mathbf{x} \rangle^n}{1 + |t|^n} \qquad \text{for all } \mathbf{x} \in \mathbb{R}^3. \tag{3.3}$$

Proof. Estimate (3.1) follows from (3.2). We next prove (3.2). By a stationary phase analysis of

$$\langle \mathbf{G_x}, f_t \rangle = \int_{\mathbb{R}^3} d^3k \frac{\kappa(\mathbf{k})}{\sqrt{2\omega(\mathbf{k})}} e^{i\alpha \mathbf{k} \cdot \mathbf{x} - it\omega(\mathbf{k})} (\varepsilon f)(\mathbf{k}) \tag{3.4}$$

we obtain $|\langle \mathbf{G_x}, f_t \rangle| \leq C_n |t|^{-n}$ for $\alpha|\mathbf{x}| \leq |t|/2$, [28] Theorem XI.14. It follows that

$$
\begin{aligned}
|\langle \mathbf{G_x}, f_t \rangle| \mathbf{1}_{\{2\alpha|\mathbf{x}| \leq |t|\}} &\leq \frac{C_n}{|t|^n} \\
|\langle \mathbf{G_x}, f_t \rangle| \mathbf{1}_{\{2\alpha|\mathbf{x}| > |t|\}} &\leq C \left(\frac{2\alpha|\mathbf{x}|}{|t|} \right)^n
\end{aligned}
$$

where $C := \sup_{t \in \mathbb{R}, \mathbf{x} \in \mathbb{R}^3} |\langle \mathbf{G_x}, f_t \rangle| < \infty$. This proves (3.2). To prove (3.3) we write

$$\langle \mathbf{G_x} - \mathbf{G_0}, f_t \rangle = \int_{\mathbb{R}^3} e^{-it\omega(\mathbf{k})} \mathbf{F_x}(\mathbf{k}) d^3k$$

where

$$\mathbf{F_x}(\mathbf{k}) = i\alpha \mathbf{k} \cdot \mathbf{x} \frac{\kappa(\mathbf{k})}{\sqrt{2\omega(\mathbf{k})}} (\varepsilon f)(\mathbf{k}) g(\alpha \mathbf{k} \cdot \mathbf{x})$$

and $g : \mathbb{R} \to \mathbb{C}$ denotes the real-analytic function given by $g(s) = (e^{is} - 1)/(is)$ for $s \neq 0$. g and all its derivatives are bounded, and by assumption on f, $\mathbf{F_x} \in C_0^\infty(\mathbb{R}^3 \backslash \{\mathbf{0}\}, \mathbb{C}^3)$ for each $\mathbf{x}$. It follows that

$$\sup_{\mathbf{x}, \mathbf{k} \in \mathbb{R}^3, \mathbf{x} \neq \mathbf{0}} \left| \partial_{\mathbf{k}}^\beta \mathbf{F_x}(\mathbf{k}) \right| |\mathbf{x}|^{-1} \langle \alpha \mathbf{x} \rangle^{-|\beta|} < \infty,$$

which implies (3.3), again by stationary phase arguments. $\qquad \square$

Lemma 3.2. *Suppose that $\varepsilon f_1, ..., \varepsilon f_N \in C_0^n(\mathbb{R}^3 \backslash \{\mathbf{0}\}, \mathbb{C}^3)$ for a given $n \in \mathbb{N}$, and let $\tilde{\alpha}$ be defined by Proposition A.3. Then there exist constants $\tilde{\alpha} > 0$ and $c_{2,n} = c_{2,n}(\underline{f})$, such that for all $\alpha \leq \tilde{\alpha}$, $t \in \mathbb{R}$, and $W^{(j)} \in \{W^{(1)}, W^{(2)}, W^{\mathrm{dip}}\}$,*

$$\left\| \left[W^{(j)}, a^*(\underline{f}_t) \right] \Phi_\alpha \right\| \leq \frac{c_{2,n}}{1 + |t|^n}. \tag{3.5}$$

Proof. By definition of $a^*(\underline{f}_t)$,

$$[W^{(j)}, a^*(\underline{f}_t)]\Phi_\alpha = \sum_{l=1}^N a^*(f_{1,t}) \cdots a^*(f_{l-1,t}) [W^{(j)}, a^*(f_{l,t})] a^*(f_{l+1,t}) \cdots a^*(f_{N,t})\Phi_\alpha \tag{3.6}$$

and by definition of $W^{(1)}$ and $W^{(2)}$

$$\begin{aligned}
\left[W^{(1)}, a^*(f_{l,t}) \right] &= 2 \langle \mathbf{G_x}, f_{l,t} \rangle \cdot \mathbf{p} & (3.7) \\
\left[W^{(2)}, a^*(f_{l,t}) \right] &= 2 \langle \mathbf{G_x}, f_{l,t} \rangle \cdot \mathbf{A}(\alpha \mathbf{x}) & (3.8)
\end{aligned}$$

From (3.2), (3.6), (3.7), (3.8) and Lemma A.1 it follows that

$$\begin{aligned}
\|[W^{(1)}, a^*(\underline{f}_t)]\Phi_\alpha\| &\leq \frac{Nc_n}{1 + |t|^n} \|(H_f + 1)^{\frac{N-1}{2}} \langle \alpha \mathbf{x} \rangle^n \mathbf{p}\Phi_\alpha\| & (3.9) \\
\|[W^{(2)}, a^*(\underline{f}_t)]\Phi_\alpha\| &\leq \frac{Nc_n}{1 + |t|^n} \|(H_f + 1)^{\frac{N}{2}} \langle \alpha \mathbf{x} \rangle^n \Phi_\alpha\| & (3.10)
\end{aligned}$$

with some constant c_n. Thanks to Lemma A.4, these upper bounds are bounded uniformly in $\alpha \leq \tilde{\alpha}$, $\tilde{\alpha}$ being defined by Proposition A.3. This proves (3.5) for $j = 1, 2$. The assertion for W^{dip} now follows from $W^{\mathrm{dip}} = W^{(1)}|_{\mathbf{x}=\mathbf{0}}$, which leads to a bound for $\|[W^{\mathrm{dip}}, a^*(\underline{f}_t)]\Phi_\alpha\|$ of the form (3.9) with $\mathbf{x} = \mathbf{0}$. $\qquad \square$

Proposition 3.3. *For all $\varepsilon f_1, ..., \varepsilon f_N \in C_0^2(\mathbb{R}^3 \backslash \{\mathbf{0}\}, \mathbb{C}^3)$ there exists a constant $c_3 = c_3(\underline{f})$, such that for all $\alpha \leq \tilde{\alpha}$ and for all $s \in \mathbb{R}$,*

$$\left\| \left[W^{(1)} - W^{\mathrm{dip}}, a^*(\underline{f}_s) \right] \Phi_\alpha \right\| \leq \frac{c_3 \alpha}{1 + s^2}. \tag{3.11}$$

Proof. By (3.6) for $j = 1$, (3.7), and the corresponding equations for W^{dip}

$$\left[W^{(1)} - W^{\mathrm{dip}}, a^*(\underline{f}_s) \right] \Phi_\alpha = 2 \sum_{l=1}^N \langle \mathbf{G_x} - \mathbf{G_0}, f_{l,s} \rangle \cdot \mathbf{p} a^*(\underline{f}_{(l),s}) \Phi_\alpha$$

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where

$$\begin{aligned}
\|\langle \mathbf{G_x} - \mathbf{G_0}, f_{l,s}\rangle \cdot \mathbf{p}a^*(\underline{f}_{(l),s})\Phi_\alpha\| &\leq \alpha \frac{c}{1+s^2}\||\mathbf{x}|\langle \alpha\mathbf{x}\rangle^2 a^*(\underline{f}_{(l),s})\mathbf{p}\Phi_\alpha\| \\
&\leq \alpha \frac{c}{1+s^2}\||\mathbf{x}|^2\langle \alpha\mathbf{x}\rangle^4\mathbf{p}\Phi_\alpha\|^{1/2}\|a(\underline{f}_{(l),s})a^*(\underline{f}_{(l),s})\mathbf{p}\Phi_\alpha\|^{1/2}
\end{aligned}$$

by (3.3) and the Cauchy-Schwarz inequality. The norms in the last expression are bounded uniformly in $\alpha \leq \tilde{\alpha}$ by Lemma A.1 and Lemma A.4. $\qquad\square$

Lemma 3.4. *For all $\varepsilon f_1, ..., \varepsilon f_N \in C_0^2(\mathbb{R}^3\backslash\{\mathbf{0}\}, \mathbb{C}^3)$, there exists a constant $c_4 = c_4(\underline{f}) < \infty$, such that for all $\alpha \leq \tilde{\alpha}$ and $s, t \in \mathbb{R}$*

$$\left\|\left[W_s^{\mathrm{dip}}, a^*(\underline{f}_t)\right](\Phi_\alpha - \Phi_0)\right\| \leq \frac{c_4\alpha^{\frac{3}{2}}}{1+|t-s|^2} \tag{3.12}$$

$$\left\|\left[W_s^{\mathrm{dip}}, a^*(\underline{f}_t)\right]\Phi_0\right\| \leq \frac{c_4}{1+|t-s|^2}. \tag{3.13}$$

$$\left\|\left[W, \left[W_s^{\mathrm{dip}}, a^*(\underline{f}_t)\right]\right]\Phi_\alpha\right\| \leq \frac{c_4\alpha^{\frac{3}{2}}}{1+|t-s|^2} \tag{3.14}$$

Proof. Since $\left[W_s^{\mathrm{dip}}, a^*(f_{l,t})\right] = 2\langle \mathbf{G_0}, f_{l,t-s}\rangle \cdot \mathbf{p}_s$, which commutes with the creation operators $a^*(f_{i,t})$,

$$\begin{aligned}
\left[W_s^{\mathrm{dip}}, a^*(\underline{f}_t)\right] &= \sum_{l=1}^N a^*(f_{1,t})\cdots a^*(f_{l-1,t})\left[W_s^{\mathrm{dip}}, a^*(f_{l,t})\right]a^*(f_{l+1,t})\cdots a^*(f_{N,t}) \\
&= 2\sum_{l=1}^N a^*(\underline{f}_{(l),t})\langle \mathbf{G_0}, f_{l,t-s}\rangle \cdot \mathbf{p}_s \tag{3.15}
\end{aligned}$$

where $|\langle \mathbf{G_0}, f_{l,t-s}\rangle| \leq c_l(1 + (t-s)^2)^{-1}$ by (3.1). In view of Lemma A.1 and Lemma A.5, this proves (3.12). The proof of (3.13) is similar.

From (3.15) we obtain, that

$$\left[W, \left[W_s^{\mathrm{dip}}, a^*(\underline{f}_t)\right]\right]\Phi_\alpha = 2\alpha^{\frac{3}{2}}\sum_{l=1}^N \langle \mathbf{G_0}, f_{l,t-s}\rangle \cdot \left[W^{(1)} + \alpha^{\frac{3}{2}}W^{(2)}, a^*(\underline{f}_{(l),t})\mathbf{p}_s\right]\Phi_\alpha.$$

Hence, by (3.1), it suffices to show that $\left\|W^{(j)}a^*(\underline{f}_{(l),t})\mathbf{p}_s\Phi_\alpha\right\|$ and $\left\|a^*(\underline{f}_{(l),t})\mathbf{p}_s W^{(j)}\Phi_\alpha\right\|$ are bounded uniformly in t, s and $\alpha \leq \tilde{\alpha}$. We shall do this for $a^*(\underline{f}_{(l),t})\mathbf{p}_s W^{(1)}\Phi_\alpha$ only, the proofs in the other cases being similar. Let $m \geq (N-1)/2$. Then

$$\begin{aligned}
&\left\|a^*(\underline{f}_{(l),t})\mathbf{p}_s W^{(1)}\Phi_\alpha\right\| \\
&\leq \sum_{j=1}^3 \left\|a^*(\underline{f}_{(l),t})(H_f + 1)^{-m}\mathbf{p}_s p_j(H_{\mathrm{el}} + i)^{-1}(H_{\mathrm{el}} + i)(H_f + 1)^m A_j(\alpha\mathbf{x})\Phi_\alpha\right\| \\
&\leq C\sum_{j=1}^3 \|(H_{\mathrm{el}} + i)(H_f + 1)^m A_j(\alpha\mathbf{x})\Phi_\alpha\|
\end{aligned}$$

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with a constant C, that is finite by Lemma A.1. We now want to compare $\|(H_{\text{el}} + i)(H_f + 1)^m A_j(\alpha \mathbf{x})\Phi_\alpha\|$ with $\|A_j(\alpha \mathbf{x})(H_{\text{el}} + i)(H_f + 1)^m \Phi_\alpha\|$, because the latter norm is bounded uniformly in $\alpha \leq \tilde{\alpha}$, by Lemma A.1 and by (A.17). Thus we compute the commutator of $(H_{\text{el}} + i)(H_f + 1)^m$ and $A_j(\alpha \mathbf{x}) = a^*(G_{\mathbf{x},j}) + a(G_{\mathbf{x},j})$ applied to Φ_α. Using

$$\left[H_{\text{el}}, a^*(G_{\mathbf{x},j})\right] = \alpha^2 a^*(\omega^2 G_{\mathbf{x},j}) - \alpha \sum_{m=1}^{3} 2 a^*(k_m G_{\mathbf{x},j}) p_m$$

$$\left[(H_f + 1)^m, a^*(G_{\mathbf{x},j})\right] = \sum_{l=1}^{m} \binom{m}{l} a^*(\omega^l G_{\mathbf{x},j})(H_f + 1)^{m-l}$$

and similar commutator equations for $a(G_{\mathbf{x},j})$, we see that all resulting terms have norms that are bounded, uniformly in $\alpha \leq \tilde{\alpha}$, thanks to (A.17) and Lemma A.1. $\qquad\square$

For completeness of this paper we now use Lemma 3.2 to prove existence of the asymptotic creation and annihilation operators on Φ_α. More general results can be found in [13, 22].

Proposition 3.5. *Suppose* $\underline{f} = (f_1, ..., f_N) \in [L_\omega^2(\mathbb{R}^3 \times \{1,2\})]^N$. *Then, for all* $\alpha \leq \tilde{\alpha}$,

$$a_\pm^*(\underline{f})\Phi_\alpha := \lim_{t \to \pm\infty} e^{iH_\alpha t} a^*(\underline{f}_t) e^{-iH_\alpha t}\Phi_\alpha \tag{3.16}$$

exists, and

$$\|a_\pm^*(\underline{f})\Phi_\alpha\| \leq c_5 \|f_1\|_\omega \cdots \|f_N\|_\omega, \tag{3.17}$$

with a constant c_5 *that is independent of* α *and* $\underline{f}$. *If* $\varepsilon f_l \in C_0^{n+1}(\mathbb{R}^3 \backslash \{\mathbf{0}\}, \mathbb{C}^3)$ *for* $l = 1, ..., N$, *then there exists a constant* $c_n(\underline{f})$, *such that*

$$\left\|a_\pm^*(\underline{f})\Phi_\alpha - e^{iH_\alpha t} a^*(\underline{f}_t) e^{-iH_\alpha t}\Phi_\alpha\right\| \leq \alpha^{3/2} \frac{c_n(\underline{f})}{1 + |t|^n} \tag{3.18}$$

Proof. Suppose first, that $\varepsilon f_1, ..., \varepsilon f_N \in C_0^{n+1}(\mathbb{R}^3 \backslash \{\mathbf{0}\}, \mathbb{C}^3)$. Then

$$\frac{d}{dt}\left(e^{itH_\alpha} a^*(\underline{f}_t) e^{-itH_\alpha}\Phi_\alpha\right) = i e^{i(H_\alpha - E_\alpha)t}[W, a^*(\underline{f}_t)]\Phi_\alpha,$$

and, by Lemma 3.2,

$$\pm \int_t^{\pm\infty} \|[W, a^*(\underline{f}_s)]\Phi_\alpha\| ds \leq \alpha^{3/2} \frac{c_n(\underline{f})}{1 + |t|^n}.$$

This estimate first proves existence of $a_\pm^*(\underline{f})$, by Cook's argument, and then it implies (3.18). The existence of $a_\pm^*(\underline{f})\Phi_\alpha$ in the case where $f_j \in L_\omega^2(\mathbb{R}^3 \times \{1,2\})$ now follows from the approximation argument given in [22], Proposition 2.1. By the Lemmas A.1 and A.4

$$
\begin{aligned}
\|e^{itH_\alpha} a^*(\underline{f}_t) e^{-itH_\alpha}\Phi_\alpha\| &\leq \|a^*(\underline{f}_t)(H_f + 1)^{-\frac{N}{2}}\| \|(H_f + 1)^{\frac{N}{2}}\Phi_\alpha\| \\
&\leq c_5 \|f_1\|_\omega \cdots \|f_N\|_\omega,
\end{aligned}
$$

uniformly in $t \in \mathbb{R}$ and $\alpha \in [0, \tilde{\alpha}]$. Letting $t \to \pm\infty$ in this estimate, we obtain (3.17). $\qquad\square$

4 Proofs of the main theorems

4.1 A reduction formula

In this section we first prove Theorem 4.1 below, which is a generalization of Theorem 1.1, the latter corresponding to the choice $\tau = 0$. The generalization to arbitrary $\tau \in \mathbb{R}$ will be needed in Section 5.

Theorem 4.1. *Let $\varepsilon f_1, ..., \varepsilon f_N \in C_0^2(\mathbb{R}^3 \backslash \{\mathbf{0}\}, \mathbb{C}^3)$. Then*

$$
a_+^*(\underline{f}_{\tau})\Phi_\alpha - a_-^*(\underline{f}_{\tau})\Phi_\alpha
$$
$$
= i\alpha^{\frac{3}{2}} \int\limits_{-\infty}^{\infty} e^{-i(H_0 - E_0)\tau} 2\mathbf{p}(s)\varphi_{el} \otimes [\mathbf{A}(\mathbf{0}, s), a^*(\underline{f})]\Omega \, ds + \mathcal{R}(\tau, \alpha)
$$

where $\|\mathcal{R}(\tau, \alpha)\| = \mathcal{O}(\alpha^{5/2}) + \mathcal{O}(\alpha^3|\tau|)$ as $\alpha \to 0$.

Remark. Part of the error $\mathcal{O}(\alpha^{5/2})$ stems from passing to the dipole-approximation $W^{(1)} \to W^{\mathrm{dip}}$. Hence its order $5/2 = 3/2 + 1$ cannot be improved.

Proof. Recall that $B_t = B(-t) = e^{-itH_0}Be^{itH_0}$. To compare the time-evolutions generated by H_α and H_0 we will use that

$$
e^{i(H_\alpha - E_\alpha)t}B_t\Phi_\alpha = B\Phi_\alpha + \int_0^t e^{i(H_\alpha - E_\alpha)s}[iW, B_s]\Phi_\alpha \, ds. \tag{4.1}
$$

This equation may be iterated because $[iW, B_s] = [iW_{-s}, B]_s$. From

$$
a_\pm^*(\underline{f})\Phi_\alpha = \lim_{t \to \pm\infty} e^{i(H_\alpha - E_\alpha)t}a^*(\underline{f}_t)\Phi_\alpha
$$

and (4.1) it follows that

$$
a_+^*(\underline{f})\Phi_\alpha - a_-^*(\underline{f})\Phi_\alpha = \int_{-\infty}^{\infty} e^{i(H_\alpha - E_\alpha)s}[iW, a^*(\underline{f}_{-s})]\Phi_\alpha \, ds.
$$

Only terms contributing to this integral of order $\alpha^{3/2}$ need to be kept. Since $W = \alpha^{\frac{3}{2}}W^{(1)} + \alpha^3 W^{(2)}$, we may drop $W^{(2)}$, $W^{(1)} - W^{\mathrm{dip}}$ and restrict the interval of integration to $|s| \leq \alpha^{-1}$ by Lemma 3.2 and Proposition 3.3. We obtain

$$
a_+^*(\underline{f})\Phi_\alpha - a_-^*(\underline{f})\Phi_\alpha
$$
$$
= i\alpha^{3/2} \int_{-\infty}^{\infty} e^{i(H_\alpha - E_\alpha)s}[W^{\mathrm{dip}}, a^*(\underline{f}_{-s})]\Phi_\alpha \, ds + \mathcal{O}(\alpha^{5/2})
$$
$$
= i\alpha^{3/2} \int_{|s| \leq \alpha^{-1}} e^{i(H_\alpha - E_\alpha)s}[W_{-s}^{\mathrm{dip}}, a^*(\underline{f})]_s\Phi_\alpha \, ds + \mathcal{O}(\alpha^{5/2}). \tag{4.2}
$$

Applying now (4.1) to the integrand in (4.2) and the time interval $[\tau, s]$, rather than $[0, s]$, we find

$$
\int_{|s| \leq \alpha^{-1}} e^{i(H_\alpha - E_\alpha)s}[W_{-s}^{\mathrm{dip}}, a^*(\underline{f})]_s\Phi_\alpha \, ds
$$
$$
= e^{i(H_\alpha - E_\alpha)\tau} \int_{|s| \leq \alpha^{-1}} [W_{-s}^{\mathrm{dip}}, a^*(\underline{f})]_\tau \Phi_\alpha ds \tag{4.3}
$$
$$
+ \int_{|s| \leq \alpha^{-1}} ds \int_\tau^s e^{i(H_\alpha - E_\alpha)r}[iW, [W_{r-s}^{\mathrm{dip}}, a^*(\underline{f}_{-r})]]\Phi_\alpha \, dr.
$$

By (3.14) in Lemma 3.4, the norm of the double integral is bounded by

$$\text{const} \int_{|s| \leq \alpha^{-1}} \frac{|\tau| + |s|}{1 + |s|^2} \alpha^{3/2} \, ds = \mathcal{O}(\alpha^{3/2}|\tau|) + \mathcal{O}(\alpha^{3/2} \ln(\alpha)). \tag{4.4}$$

In the integral (4.3) we use Lemma 3.4 to replace Φ_α by Φ_0 and to extend the integration over all $s \in \mathbb{R}$. We find that

$$\int_{|s| \leq \alpha^{-1}} [W_{-s}^{\text{dip}}, a^*(\underline{f})]_\tau \Phi_\alpha \, ds$$

$$= \int_{-\infty}^{\infty} [W_{\tau-s}^{\text{dip}}, a^*(\underline{f}_\tau)] \Phi_0 \, ds + \mathcal{O}(\alpha)$$

$$= \int_{-\infty}^{\infty} e^{-i(H_0 - E_0)\tau} [W_{-s}^{\text{dip}}, a^*(\underline{f})] \Phi_0 \, ds + \mathcal{O}(\alpha). \tag{4.5}$$

Equations (4.2), (4.3), (4.4) and (4.5) prove the theorem because $e^{-i(H_\alpha - E_\alpha)\tau} a_\pm^*(\underline{f})\Phi_\alpha = a_\pm^*(\underline{f}_\tau)\Phi_\alpha$ and because $\Phi_0 = \varphi_{el} \otimes \Omega$. $\qquad \square$

Theorem 4.1 in the case $\tau = 0$ becomes Theorem 1.1, which implies that

$$\left\| \mathbf{1}_{ac}(H_{\text{el}})\left(a_-^*(\underline{f})\Phi_\alpha - a_+^*(\underline{f})\Phi_\alpha\right) \right\|^2 = \alpha^3 P^{(3)}(\underline{f}) + O(\alpha^4)$$

where

$$P^{(3)}(\underline{f}) := \left\| \mathbf{1}_{ac}(H_{\text{el}}) \int_{-\infty}^{\infty} 2\mathbf{p}(s)\varphi_{el} \otimes \left[\mathbf{A}(\mathbf{0}, s), a^*(\underline{f})\right]\Omega \, ds \right\|^2. \tag{4.6}$$

We next show that $P^{(3)}(\underline{f})$ is additive in its one-photon contributions.

Proposition 4.2. *Suppose that* $\underline{f} = (f_1, m_1, \ldots, f_n, m_n) \in \left[L^2(\mathbb{R}^3) \times \mathbb{N}\right]^n$ *with* $\langle f_i, f_j \rangle = \delta_{ij}$ *and* $\varepsilon f_l \in C_0^2(\mathbb{R}^3 \backslash \{\mathbf{0}\}, \mathbb{C}^3)$. *Then* $P^{(3)}(\underline{f}) = \sum_{l=1}^n m_l P^{(3)}(f_l)$ *with*

$$P^{(3)}(f_l) = \left\| \mathbf{1}_{ac}(H_{\text{el}}) \int_{-\infty}^{\infty} 2\mathbf{p}(s)\varphi_{el} \cdot \langle \Omega, \mathbf{A}(\mathbf{0}, s)a^*(f_l)\Omega \rangle \, ds \right\|^2$$

$$= \left\| \mathbf{1}_{ac}(H_{\text{el}}) \int_{-\infty}^{\infty} \mathbf{x}(s)\varphi_{el} \cdot \langle \Omega, \mathbf{E}(\mathbf{0}, s)a^*(f_l)\Omega \rangle \, ds \right\|^2. \tag{4.7}$$

Proof. Since $a^*(\underline{f})$ is a product of creation operators $a^*(f_l)$ and since $[\mathbf{A}(\mathbf{0}, s), a^*(f_l)] = \langle \Omega, \mathbf{A}(\mathbf{0}, s)a^*(f_l)\Omega \rangle$, a scalar multiple of the identity operator, we have

$$\left[\mathbf{A}(\mathbf{0}, s), a^*(\underline{f})\right]\Omega = \sum_{l=1}^n \sqrt{m_l} \, \langle \Omega, \mathbf{A}(\mathbf{0}, s)a^*(f_l)\Omega \rangle \, a^*(\underline{f}_{(l)})\Omega$$

where $\underline{f}_{(l)} = (f_1, m_1, \ldots, f_l, (m_l - 1), \ldots, f_n, m_n)$. The vectors $a^*(\underline{f}_{(l)})\Omega$ are orthonormal by construction. Hence by definition of $P^{(3)}(\underline{f})$ and by the Pythagoras identity,

$$P^{(3)}(\underline{f}) = \sum_{l=1}^n P^{(3)}(f_l)$$

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with $P^{(3)}(f_l)$ given by the first equation in the statement of the proposition. The second equation in the proposition follows from

$$2\mathbf{p}(s)\varphi_{el} = \frac{d}{ds}\mathbf{x}(s)\varphi_{el}, \qquad \frac{d}{ds}\langle\Omega, \mathbf{A}(\mathbf{0}, s)a^*(f_l)\Omega\rangle = -\langle\Omega, \mathbf{E}(\mathbf{0}, s)a^*(f_l)\Omega\rangle$$

by an integration by parts. The differentiability of $s \mapsto \mathbf{x}(s)\varphi_{el}$ and the expression for its derivative are established in Lemma A.6. $\square$

4.2 Expansion in generalized eigenfunctions

In this section we prove Theorem 1.3 and the stronger statement expressed by the Equations (1.17) and (1.18). The ingredients are Theorem 1.1, Proposition 4.2, and a set of generalized eigenfunctions $\varphi_{\mathbf{q}}$ with the properties (i)-(iii) in the introduction. Concerning the existence of $\varphi_{\mathbf{q}}$, we recall from [9], Theorem 4.7.1, that our hypotheses on V imply existence and completeness of a (modified) wave operator Ω_+ associated with H_{el}. Moreover, $(H_{el} - i)^{-1}\langle\mathbf{x}\rangle^{-2}$ is a Hilbert-Schmidt operator.

Lemma 4.3. *Suppose that $\varphi : \mathbb{R} \to \mathcal{H}_{el} \cap D(|\mathbf{x}|^2)$ is such that $s \mapsto \varphi(s)$ and $s \mapsto |\mathbf{x}|^2\varphi(s)$ are continuous and absolutely integrable with respect to the norm of $\mathcal{H}_{el}$. Then $\int_{-\infty}^{\infty} \varphi(s)ds \in D(|\mathbf{x}|^2)$ and*

$$\left\|\mathbf{1}_{ac}(H_{el})\int_{-\infty}^{\infty}\varphi(s)ds\right\|^2 = \int_{\mathbb{R}^3}\left|\int_{-\infty}^{\infty}\langle\varphi_{\mathbf{q}}, \varphi(s)\rangle\, ds\right|^2 d^3q.$$

Proof. From the existence of the improper Riemann integrals $\int_{-\infty}^{\infty}\varphi(s)ds$ and $\int_{-\infty}^{\infty}|\mathbf{x}|^2\varphi(s)ds$ and the fact that multiplication with $|\mathbf{x}|^2$ is a closed operator, it follows that $\int_{-s}^{s}\varphi(s)ds \in D(|\mathbf{x}|^2)$ and that

$$|\mathbf{x}|^2\int_{-\infty}^{\infty}\varphi(s)ds = \int_{-\infty}^{\infty}|\mathbf{x}|^2\varphi(s)ds.$$

This equation and property (i) of $\varphi_{\mathbf{q}}$ imply that

$$\left\langle\varphi_{\mathbf{q}}, \int_{-\infty}^{\infty}\varphi(s)ds\right\rangle = \left\langle|\mathbf{x}|^{-2}\varphi_{\mathbf{q}}, \int_{-\infty}^{\infty}|\mathbf{x}|^2\varphi(s)ds\right\rangle$$

$$= \int_{-\infty}^{\infty}\left\langle|\mathbf{x}|^{-2}\varphi_{\mathbf{q}}, |\mathbf{x}|^2\varphi(s)\right\rangle ds = \int_{-\infty}^{\infty}\left\langle\varphi_{\mathbf{q}}, \varphi(s)\right\rangle ds.$$

In view of (1.14), this proves the assertion. $\square$

Proposition 4.4. *Suppose that $\varepsilon f \in C_0^2(\mathbb{R}^3\backslash\{\mathbf{0}\})$. Then*

$$P^{(3)}(f) = \int_{\mathbb{R}^3}d^3q\left|\langle\varphi_{\mathbf{q}}, \mathbf{x}\varphi_{el}\rangle \cdot \int_{-\infty}^{\infty}e^{i(\mathbf{q}^2-E_0)s}\langle\Omega, \mathbf{E}(\mathbf{0}, s)a^*(f)\Omega\rangle\, ds\right|^2$$

$$= 4\pi^2\int_{\mathbb{R}^3}d^3q\left|\langle\varphi_{\mathbf{q}}, \mathbf{x}\varphi_{el}\rangle \cdot \int_{|\mathbf{k}|=q^2-E_0}|\mathbf{k}|\sum_{\lambda=1,2}\overline{G_{\mathbf{0}}(\mathbf{k}, \lambda)}f(\mathbf{k}, \lambda)d\sigma(\mathbf{k})\right|^2,$$

where $d\sigma(\mathbf{k})$ denotes the surface measure of the sphere $|\mathbf{k}| = q^2 - E_0$ in $\mathbb{R}^3$.

Proof. We start with the expression (4.7) for $P^{(3)}(f)$ and we shall apply Lemma 4.3 to

$$\varphi(s) = \mathbf{x}(s)\varphi_{el} \cdot \langle \Omega, \mathbf{E}(\mathbf{0}, s) a^*(f)\Omega \rangle . \tag{4.8}$$

By Lemma A.6, $\mathbf{x}(s)\varphi_{el} = e^{i(H_{el}-E_0)s}\mathbf{x}\varphi_{el}$ belongs to $D(|\mathbf{x}|^2)$ and $\||\mathbf{x}|^2\mathbf{x}(s)\varphi_{el}\| \leq C(1+s^2)$. On the other hand

$$
\begin{aligned}
\langle \Omega, \mathbf{E}(\mathbf{0}, s) a^*(f)\Omega \rangle &= \sum_{\lambda=1,2} \int i\omega(\mathbf{k}) e^{-i\omega(\mathbf{k})s} \overline{\mathbf{G_0}(\mathbf{k}, \lambda)} f(\mathbf{k}, \lambda) d^3k \\
&= \int_0^\infty d\omega e^{-i\omega s} \int_{|\mathbf{k}|=\omega} i\omega \sum_{\lambda=1,2} \overline{\mathbf{G_0}(\mathbf{k}, \lambda)} f(\mathbf{k}, \lambda) d\sigma(\mathbf{k}) \quad (4.9)
\end{aligned}
$$

is the Fourier transform of a function from $C_0^\infty(\mathbb{R}_+)$, and hence rapidly decreasing as $s \to \infty$. It follows that (4.8) satisfies the hypotheses of Lemma 4.3. Hence Lemma 4.3 proves the first asserted equation because

$$\langle \varphi_{\mathbf{q}}, \mathbf{x}(s)\varphi_{el} \rangle = e^{i(\mathbf{q}^2-E_0)s} \langle \varphi_{\mathbf{q}}, \mathbf{x}\varphi_{el} \rangle .$$

The second equation follows from the first one and from (4.9) by an application of the Fourier inversion theorem. $\square$

4.3 Proof of Theorem 1.2

Lemma 4.5. *If $\underline{f} = (f_1, ..., f_n)$ with $\varepsilon f_1, ..., \varepsilon f_n \in C_0^\infty(\mathbb{R}^3\backslash\{\mathbf{0}\})$ and $F \in C_0^\infty((-\infty, 0))$, then*

$$a_+^*(\underline{f})F(H_\alpha) - a^*(\underline{f})F(H_0) = \mathcal{O}(\alpha^{\frac{3}{2}})$$

Proof. Choose $R \in \mathbb{R}$, such that $\text{supp}(f_1), ..., \text{supp}(f_n) \in \{|\mathbf{k}| < R\}$ and then choose $G \in C_0^\infty(\mathbb{R})$ with $G = 1$ on $\text{supp}(F) + [0, nR]$. Then, by the pull through formula for $a^*(\underline{f})$ and by [13], Theorem 4 (iv),

$$a^*(\underline{f})F(H_0) = G(H_0)a^*(\underline{f})F(H_0), \qquad a_+^*(\underline{f})F(H_\alpha) = G(H_\alpha)a_+^*(\underline{f})F(H_\alpha).$$

Using that $F(H_0) - F(H_\alpha) = \mathcal{O}(\alpha^{\frac{3}{2}})$, by the Helffer-Sjöstrand functional calculus, that $\left(a^*(\underline{f}) - a_+^*(\underline{f})\right)F(H_\alpha) = \mathcal{O}(\alpha^{3/2})$, by the proof of Proposition 3.5, and that $G(H_0)a^*(\underline{f})$, $a_+^*(\underline{f})F(H_\alpha)$ are bounded by Lemma A.1 and [22] Proposition 2.1, we find that

$$
\begin{aligned}
a^*(\underline{f})F(H_0) - a_+^*(\underline{f})F(H_\alpha) &= G(H_0)a^*(\underline{f})F(H_0) - G(H_\alpha)a_+^*(\underline{f})F(H_\alpha) \\
&= G(H_0)a^*(\underline{f})\left(F(H_0) - F(H_\alpha)\right) + G(H_0)\left(a^*(\underline{f}) - a_+^*(\underline{f})\right)F(H_\alpha) \\
&\quad + \left(G(H_0) - G(H_\alpha)\right)a_+^*(\underline{f})F(H_\alpha) = \mathcal{O}(\alpha^{\frac{3}{2}})
\end{aligned}
$$

as $\alpha \to 0$. $\square$

Recall from the introduction that $\mathcal{H}_+^\alpha$ is the closure of the span of all vectors of the form

$$a_+^*(\underline{h})\Phi_\alpha, \qquad \underline{h} = (h_1, ..., h_n), \quad \text{where} \quad h_i, \omega h_i \in L_\omega^2(\mathbb{R}^3 \times \{1, 2\}), \tag{4.10}$$

and that P_+^α is the orthogonal projection onto $\mathcal{H}_+^\alpha$.

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Proof of Theorem 1.2. In the first two steps of this proof we shall establish (1.13) in the weak operator topology. Then we establish norm convergence to conclude the proof.

<u>Step 1</u>: Suppose $H_{\mathrm{el}}\varphi = \lambda\varphi$, $n \in \mathbb{N}$ and $\underline{f} = (f_1, ..., f_n)$ with $\varepsilon f_1, ..., \varepsilon f_n \in C_0^\infty(\mathbb{R}^3\backslash\{\mathbf{0}\})$. Then

$$\lim_{\alpha \to 0} P_+^\alpha\Big(\varphi \otimes a^*(\underline{f})\Omega\Big) = \varphi \otimes a^*(\underline{f})\Omega \tag{4.11}$$

and the analog statement holds for $\varphi \otimes \Omega$.

Since $\lambda < 0$ there exists $F \in C_0^\infty(\mathbb{R})$ with $F(\lambda) = 1$ and $\mathrm{supp}(F) \subseteq (-\infty, 0)$. Moreover $P_+^\alpha F(H_\alpha) = F(H_\alpha)$ by the hypothesis of Theorem 1.2 and because $\Sigma_\alpha \geq 0$ for all $\alpha \in \mathbb{R}$. Using, in addition, that

$$a_+^*(\underline{f})F(H_\alpha) - a^*(\underline{f})F(H_0) = \mathcal{O}(\alpha^{\frac{3}{2}}),$$

which we know from Lemma 4.5, we conclude that

$$P_+^\alpha\Big(\varphi \otimes a^*(\underline{f})\Omega\Big) = P_+^\alpha a^*(\underline{f})F(H_0)\varphi \otimes \Omega = P_+^\alpha a_+^*(\underline{f})F(H_\alpha)\varphi \otimes \Omega + \mathcal{O}(\alpha^{\frac{3}{2}})$$

$$= a_+^*(\underline{f})F(H_\alpha)\varphi \otimes \Omega + \mathcal{O}(\alpha^{\frac{3}{2}}) = \varphi \otimes a^*(\underline{f})\Omega + \mathcal{O}(\alpha^{\frac{3}{2}}).$$

Step 1 implies that

$$\lim_{\alpha \to 0} P_+^\alpha\Phi = \Phi \quad \text{for all } \Phi \in \mathrm{Ran}\mathbf{1}_{pp}(H_{\mathrm{el}}) \otimes \mathbf{1}_{\mathcal{F}}.$$

<u>Step 2</u>: $w - \lim_{\alpha \to 0} P_+^\alpha(\mathbf{1}_c(H_{\mathrm{el}}) \otimes \mathbf{1}_{\mathcal{F}}) = 0$.

Since $\|P_+^\alpha(\mathbf{1}_c(H_{\mathrm{el}}) \otimes \mathbf{1}_{\mathcal{F}})\| \leq 1$ for all $\alpha \in \mathbb{R}$ it suffices to show that

$$\lim_{\alpha \to 0}\langle a_+^*(\underline{f})\Phi_\alpha, P_+^\alpha(\mathbf{1}_c(H_{\mathrm{el}}) \otimes \mathbf{1}_{\mathcal{F}})\varphi\rangle = 0$$

for all $\varphi \in \mathcal{H}$ and all $\underline{f} = (f_1, ..., f_n)$ with $\varepsilon f_1, ..., \varepsilon f_n \in C_0^\infty(\mathbb{R}^3\backslash\{\mathbf{0}\})$. Since $a^*(\underline{f})\Phi_\alpha \in \mathrm{Ran}P_+^\alpha$, this follows from

$$a_+^*(\underline{f})\Phi_\alpha = a^*(\underline{f})\Phi_0 + \mathcal{O}(\alpha^{\frac{3}{2}}),$$

which follows from Lemma 4.5 and Lemma A.5.

From Step 1 and Step 2 it follows that

$$w - \lim_{\alpha \to 0} P_+^\alpha = \mathbf{1}_{pp}(H_{\mathrm{el}}) \otimes \mathbf{1}_{\mathcal{F}}. \tag{4.12}$$

Since P_+^α and $\mathbf{1}_{pp}(H_{\mathrm{el}}) \otimes \mathbf{1}_{\mathcal{F}}$ are orthogonal projectors, we have

$$\|P_+^\alpha\varphi\|^2 = \langle\varphi, P_+^\alpha\varphi\rangle \xrightarrow{\alpha \to 0} \langle\varphi, \mathbf{1}_{pp}(H_{\mathrm{el}}) \otimes \mathbf{1}_{\mathcal{F}}\varphi\rangle = \|\mathbf{1}_{pp}(H_{\mathrm{el}}) \otimes \mathbf{1}_{\mathcal{F}}\varphi\|^2.$$

Combined with (4.12) this proves the desired strong convergence. $\square$

5 Space-Time Analysis of the Ionization Process

The purpose of this section is to connect our result with those of the previous papers [3, 33], where expressions for the zeroth and first non-trivial order of the ionization probability were defined. We transcribe the definitions from [33] to our model and prove their equivalence to the definitions in this paper. Let $F_R := \mathbf{1}_{\{|\mathbf{x}| \geq R\}} \otimes \mathbf{1}_{\mathcal{F}}$.

Proposition 5.1. *Let $\varepsilon f_1, ..., \varepsilon f_N \in C_0^2(\mathbb{R}^3 \backslash \{\mathbf{0}\}, \mathbb{C}^3)$. Then*

$$\lim_{R \to \infty} \limsup_{\alpha \searrow 0} \sup_{\tau \in \mathbb{R}} \| F_R a_\pm^*(\underline{f}_\tau) \Phi_\alpha \|^2 = 0. \tag{5.1}$$

Remarks. The left hand side of Equation (5.1) may be interpreted as the ionization probability to zeroth order in α [33]. Proposition 5.1 should be compared to Theorem 4.1 in [33].

Proof. As in the proof of Theorem 4.1

$$a_\pm^*(\underline{f}_\tau) \Phi_\alpha - a^*(\underline{f}_\tau) \Phi_\alpha = i \int\limits_0^{\pm\infty} e^{is(H_\alpha - E_\alpha)} [W, a^*(\underline{f}_{\tau+s})] \Phi_\alpha ds,$$

where the integral is $\mathcal{O}(\alpha^{\frac{3}{2}})$ in norm, uniformly in τ, by Lemma 3.2. Hence it remains to show that

$$\lim_{R \to \infty} \limsup_{\alpha \searrow 0} \sup_{\tau \in \mathbb{R}} \| F_R a^*(\underline{f}_\tau) \Phi_\alpha \|^2 = 0. \tag{5.2}$$

To this end, we observe that, according to Lemma A.1,

$$
\begin{aligned}
\| F_R a^*(\underline{f}_\tau) \Phi_\alpha \|^2 &\leq \| a^*(\underline{f}_\tau)^2 \Phi_\alpha \| \| F_R \Phi_\alpha \| \\
&\leq C_{2N} \prod_{l=1}^N \| f_l \|_\omega^2 \, \| (H_f + 1)^N \Phi_\alpha \| \, \| \, |\mathbf{x}| \Phi_\alpha \| \frac{1}{R}.
\end{aligned}
$$

This proves (5.2), because $\limsup_{\alpha \to 0+} \| (H_f + 1)^N \Phi_\alpha \|$ and $\limsup_{\alpha \to 0+} \| \, |\mathbf{x}| \Phi_\alpha \|$ are finite by Lemma A.4 and by (A.5). $\square$

Theorem 5.2. *Let $\varepsilon f_1, ..., \varepsilon f_N \in C_0^2(\mathbb{R}^3 \backslash \{\mathbf{0}\}, \mathbb{C}^3)$, suppose $\sigma_{sc}(H_{el}) = \emptyset$, and let $\tau(\alpha) = \alpha^{-\beta}$ for some $\beta \in (0, \frac{3}{2})$. Then*

$$P^{(3)}(\underline{f}) = \lim_{R \to \infty} \limsup_{\alpha \searrow 0} \alpha^{-3} \left\| F_R \left[a_-^*(\underline{f}_{\tau(\alpha)}) \Phi_\alpha - a^*(\underline{f}_{\tau(\alpha)}) \Phi_\alpha \right] \right\|^2. \tag{5.3}$$

Remarks. Equation (5.3) is to be compared with the expression defining $Q^{(2)}(A)$ in Equation (1.9) from [33]: if we set $g = \alpha^{3/2}$ and $\tau(g) = \alpha^{-\beta}$ in that equation, then $Q^{(2)}(A)$ coincides with the right hand side of (5.3).

Proof. From Proposition 3.5 we know that

$$\left\| a_+^*(\underline{f}_{\tau(\alpha)}) \Phi_\alpha - a^*(\underline{f}_{\tau(\alpha)}) \Phi_\alpha \right\| \leq C \frac{\alpha^{3/2}}{\tau(\alpha)} = C \alpha^{3/2+\beta},$$

hence we may replace $a^*(\underline{f}_{\tau(\alpha)})\Phi_\alpha$ by $a_+^*(\underline{f}_{\tau(\alpha)})\Phi_\alpha$ for the proof of (5.3). From Theorem 4.1 we know that

$$\lim_{R\to\infty}\limsup_{\alpha\searrow 0}\alpha^{-3}\|F_R\left[a_-^*(\underline{f}_{\tau(\alpha)})\Phi_\alpha - a_+^*(\underline{f}_{\tau(\alpha)})\Phi_\alpha\right]\|^2$$

$$= \lim_{R\to\infty}\limsup_{\alpha\searrow 0}\|F_R e^{-i\tau(\alpha)(H_0-E_0)}\Psi(\underline{f})\|^2$$

$$= \lim_{R\to\infty}\limsup_{\tau\to\infty}\|F_R e^{-i\tau H_{\mathrm{el}}}\otimes\mathbf{1}_{\mathcal{F}}\Psi(\underline{f})\|^2 \qquad (5.4)$$

where

$$\Psi(\underline{f}) := \int_{-\infty}^{\infty} 2\mathbf{p}(s)\varphi_{el}\otimes[\mathbf{A}(\mathbf{0},s),a^*(\underline{f})]\Omega\,ds = \sum_{l=1}^{N}\phi_l\otimes\eta_l. \qquad (5.5)$$

Explicit expressions for ϕ_l and η_l may be taken from the proof of Proposition 4.2, e.g., $\eta_l = a^*(\underline{f}_{(l)})\Omega$, but they are not needed here. From (5.5) it follows that

$$F_R e^{-i\tau H_{\mathrm{el}}}\Psi(\underline{f}) = \sum_{l=1}^{N}\left[\mathbf{1}_{\{|\mathbf{x}|\geq R\}}e^{-i\tau H_{\mathrm{el}}}\phi_l\right]\otimes\eta_l$$

where

$$\mathbf{1}_{\|\mathbf{x}\|\geq R\}}e^{-i\tau H_{\mathrm{el}}}\phi_l = (\mathbf{1}-\mathbf{1}_{\{\|\mathbf{x}\|<R\}})e^{-i\tau H_{\mathrm{el}}}\mathbf{1}_{ac}(H_{\mathrm{el}})\phi_l$$
$$+\mathbf{1}_{\{\|\mathbf{x}\|\geq R\}}e^{-i\tau H_{\mathrm{el}}}\mathbf{1}_{pp}(H_{\mathrm{el}})\phi_l. \qquad (5.6)$$

By the RAGE Theorem, see [32], Satz 12.8,

$$\lim_{R\to\infty}\sup_{\tau\to\infty}\|\mathbf{1}_{\{|\mathbf{x}|\geq R\}}e^{i\tau H_{\mathrm{el}}}\mathbf{1}_{pp}(H_{\mathrm{el}})\phi_l\| = 0, \qquad (5.7)$$

$$\lim_{\tau\to\infty}\|\mathbf{1}_{\{|\mathbf{x}|<R\}}e^{i\tau H_{\mathrm{el}}}\mathbf{1}_{ac}(H_{\mathrm{el}})\phi_l\| = 0. \qquad (5.8)$$

From (5.4)-(5.8) it follows, that

$$\lim_{R\to\infty}\limsup_{\tau\to\infty}\|F_R e^{-i\tau H_0}\Psi(\underline{f})\|^2 = \|(\mathbf{1}_{ac}(H_{\mathrm{el}})\otimes\mathbf{1}_{\mathcal{F}})\Psi(\underline{f})\|^2 = P^{(3)}(\underline{f}),$$

by Equation (4.6) $\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\Box$

A Uniform estimates

In this appendix we collect estimates used in the previous sections. Most of them are well-known for fixed α, but this is not sufficient for us: we need estimates holding uniformly for α in a neighborhood of $\alpha = 0$. This forces us to review some of the derivations with special attention to the dependences on α.

Lemma A.1. *For every $N\in\mathbb{N}$ there is a finite constant C_N such that for all $h_1,...,h_N\in L_\omega^2(\mathbb{R}^3\times\{1,2\})$*

$$\|a^*(\underline{h})(H_f+1)^{-\frac{N}{2}}\| \leq C_N\prod_{l=1}^{N}\|h_l\|_\omega \qquad (A.1)$$

$$\|a^*(h_1)\cdots a^*(h_{l-1})\mathbf{A}(\alpha\mathbf{x})a^*(h_{l+1})\cdots a^*(h_N)(H_f+1)^{-\frac{N}{2}}\| \leq C_N\prod_{\substack{j=1\\j\neq l}}^{N}\|h_j\|_\omega \qquad (A.2)$$

Proof. Both, (A.1) and (A.2) follow from Lemma 17 in [13]. We recall that $\mathbf{A}(\alpha\mathbf{x}) = a^*(\mathbf{G_x}) + a(\mathbf{G_x})$ and we note that $\sup_{\mathbf{x}\in\mathbb{R}^3}\|\mathbf{G_x}\|_\omega < \infty$. $\square$

Proposition A.2 ([15]). *For every $\lambda < e_1$ there exists a constant $\alpha_\lambda > 0$, such that for all $n \in \mathbb{N}$*

$$\sup_{\alpha\le\alpha_\lambda} \big\||\mathbf{x}|^n \mathbf{1}_{(-\infty,\lambda]}(H_\alpha)\big\| < \infty. \tag{A.3}$$

Proposition A.3. *There exists an $\tilde\alpha > 0$, such that:*

a) *For all $\alpha \le \tilde\alpha$*

$$E_\alpha := \inf \sigma(H_\alpha) \tag{A.4}$$

is a simple eigenvalue of H_α. In the following Φ_α denotes the unique normalized ground state of H_α whose phase is determined by $\langle\Phi_\alpha, \Phi_0\rangle \ge 0$.

b) *For every $n \in \mathbb{N}$,*

$$\sup_{\alpha\le\tilde\alpha} \big\| |\mathbf{x}|^n \Phi_\alpha \big\| < \infty. \tag{A.5}$$

c) *There exists a finite constant C, such that for all $\alpha \le \tilde\alpha$ and all $\mathbf{k} \in \mathbb{R}^3\backslash\{\mathbf{0}\}$*

$$\|a(k)\Phi_\alpha\| \quad \le \quad \alpha^{\frac{3}{2}} C \frac{|\kappa(\mathbf{k})|}{\sqrt{|\mathbf{k}|}}(1 + \alpha|\mathbf{k}|), \tag{A.6}$$

$$|E_0 - E_\alpha| \quad \le \quad \alpha^{\frac{3}{2}} C, \tag{A.7}$$

$$\|\Phi_\alpha - \Phi_0\| \quad \le \quad \alpha^{\frac{3}{2}} C. \tag{A.8}$$

d) *For every $n \in \mathbb{N}$,*

$$\sup_{\alpha\le\tilde\alpha} \|[H_f^{n-1}, H_\alpha](H_\alpha + i)^{-n+1}\| \quad < \quad \infty, \tag{A.9}$$

$$\sup_{\alpha\le\tilde\alpha} \|H_f^n (H_\alpha + i)^{-n}\| \quad < \quad \infty, \tag{A.10}$$

$$\sup_{\alpha\le\tilde\alpha} \|H_{\mathrm{el}}(H_\alpha + i)^{-1}\| \quad < \quad \infty. \tag{A.11}$$

Remark. Boundedness of $[H_f^{n-1}, H](H + i)^{-n}$ and $H_f^n(H + i)^{-n}$ has previously been established in [13], Lemma 5, for a class of Hamiltonians H that includes H_α. Yet, that results does not imply (A.9) and (A.10), and second, its proof is much more complicated than the proof of (A.9) and (A.10), because H in [13] is defined in terms of a Friedrichs' extension.

Proof. That $E_\alpha = \inf \sigma(H_\alpha)$ is an eigenvalue of H_α, for small α, was first shown in [2]. Its simplicity follows from (A.8), which hold for *every* normalized ground state vector Φ_α that satisfies the phase condition $\langle\Phi_\alpha, \Phi_0\rangle \ge 0$. A proof of (A.8) may be found, e.g., in [15], Proposition 19, Steps 4 and 5. A weaker form of (A.6) is given in Lemma 20 of [15], but the proof there actually shows (A.6). Estimate (A.7) follows from Lemma 22 in [15] by choosing the infrared cutoff in this lemma larger than the UV-cutoff. Finally, (A.5) is a consequence of Proposition A.2 and (A.7).

To prove (d) we set $R_0 := (H_0 + i)^{-1}$ and $R_\alpha := (H_\alpha + i)^{-1}$. It is a simple exercise, using (A.1) and the boundedness of $(H_f + 1)^{1/2}\mathbf{p}R_0$, to show that $\|WR_0\| = \mathcal{O}(\alpha^{3/2})$ as $\alpha \to 0$. Hence we may assume that $\sup_{\alpha \leq \tilde{\alpha}} \|WR_0\| \leq 1/2$ after making $\tilde{\alpha}$ smaller, if necessary. It follows that

$$\|(H_0 + i)R_\alpha\| = \|(1 + WR_0)^{-1}\| \leq (1 - \|WR_0\|)^{-1} \leq 2 \tag{A.12}$$

for all $\alpha \leq \tilde{\alpha}$. Since $H_{\mathrm{el}}R_0$ and $H_f R_0$ are bounded operators, we have thus proven statement (d) for $n = 1$, (A.9) being trivial in this case. We now proceed by induction, assuming that (A.9) and (A.10) hold true for all positive integers smaller or equal to a given $n \geq 1$. To prove (A.10) for n replaced by $(n + 1)$ we use that

$$
\begin{aligned}
[H_f^n, H_\alpha]R_\alpha^n &= \sum_{l=1}^n \binom{n}{l} \mathrm{ad}_{H_f}^l(W) H_f^{n-l} R_\alpha^n \\
&= \sum_{l=1}^n \binom{n}{l} \mathrm{ad}_{H_f}^l(W) R_\alpha \left(H_f^{n-l} R_\alpha^{n-1} - [H_f^{n-l}, H_\alpha]R_\alpha^n \right) \tag{A.13}
\end{aligned}
$$

where $\sup_{\alpha \leq \tilde{\alpha}} \|\mathrm{ad}_{H_f}^l(W)R_\alpha\| < \infty$ by (A.12), by explicit formulas for $\mathrm{ad}_{H_f}^l(W)$ and by the arguments above proving that $\|WR_0\| = \mathcal{O}(\alpha^{3/2})$. Hence $\sup_{\alpha \leq \tilde{\alpha}} \|[H_f^n, H_\alpha]R_\alpha^n\| < \infty$ follows from (A.13) and from the induction hypothesis. Statement (A.10) with n replaced by $n + 1$ now follows from $H_f^{n+1} R_\alpha^{n+1} = (H_f R_\alpha)(H_f^n R_\alpha^n) - H_f R_\alpha [H_f^n, H_\alpha] R_\alpha^{n+1}$, from the induction hypothesis, and from (A.9) with n replaced by $n + 1$, which we have just established. $\square$

Lemma A.4. *For all $l, m \in \mathbb{N}$:*

$$
\sup_{\alpha \leq \tilde{\alpha}} \|(H_f + 1)^m \Phi_\alpha\| < \infty, \tag{A.14}
$$

$$
\sup_{\alpha \leq \tilde{\alpha}} \|(H_f + 1)^m \langle \mathbf{x} \rangle^l \Phi_\alpha\| < \infty, \tag{A.15}
$$

$$
\sup_{\alpha \leq \tilde{\alpha}} \|\mathbf{p}^2 \Phi_\alpha\| < \infty, \tag{A.16}
$$

$$
\sup_{\alpha \leq \tilde{\alpha}} \|(H_f + 1)^m H_{\mathrm{el}} \Phi_\alpha\| < \infty, \tag{A.17}
$$

$$
\sup_{\alpha \leq \tilde{\alpha}} \|(H_f + 1)^m \langle \mathbf{x} \rangle^l \mathbf{p} \Phi_\alpha\| < \infty. \tag{A.18}
$$

Proof. The statements (A.14) and (A.16) easily follow from (A.10), (A.11), and (A.7), because $\Phi_\alpha = (H_\alpha + i)^{-n} \Phi_\alpha (E_\alpha + i)^n$; note that $\mathbf{p}^2 (H_{\mathrm{el}} + i)^{-1}$ is bounded by assumption on V. To prove (A.15) we use that

$$
\sup_{\alpha \leq \tilde{\alpha}} \|\langle \mathbf{x} \rangle^l (H_f + 1)^m \Phi_\alpha\|^2 \leq \sup_{\alpha \leq \tilde{\alpha}} \|\langle \mathbf{x} \rangle^{2l} \Phi_\alpha\| \cdot \|(H_f + 1)^{2m} \Phi_\alpha\|
$$

where the right hand side is finite thanks to (A.5) and (A.14). To prove (A.17) we write

$$
\begin{aligned}
H_{\mathrm{el}} (H_f + 1)^m \Phi_\alpha &= H_{\mathrm{el}} (H_\alpha + i)^{-1} (H_\alpha + i)(H_f + 1)^m \Phi_\alpha \\
&= H_{\mathrm{el}} (H_\alpha + i)^{-1} [H_\alpha, (H_f + 1)^m] \Phi_\alpha \\
&\quad + H_{\mathrm{el}} (H_\alpha + i)^{-1} (H_f + 1)^m \Phi_\alpha (E_\alpha + i).
\end{aligned}
$$

The vectors $[H_\alpha, (H_f + 1)^m]\Phi_\alpha$ and $(H_f + 1)^m\Phi_\alpha$, and the operator $H_{\mathrm{el}}(H_\alpha + i)^{-1}$ are bounded, uniformly in $\alpha \leq \tilde\alpha$, by (A.9), (A.10) and (A.11). This proves (A.17).

The statement (A.18) follows from (A.15) and (A.16) after moving both $\mathbf{p}$'s to one side, and both factors $\langle \mathbf{x} \rangle^l$ to the other side of the inner product $\|(H_f + 1)^m\langle \mathbf{x}\rangle^l \mathbf{p}\Phi_\alpha\|^2 = \langle (H_f + 1)^m\langle \mathbf{x}\rangle^l \mathbf{p}\Phi_\alpha, (H_f + 1)^m\langle \mathbf{x}\rangle^l \mathbf{p}\Phi_\alpha \rangle$. □

The following lemma improves upon (A.8).

Lemma A.5. *For each $m \in \mathbb{N}$ there is a finite constant K_m, such that for all $\alpha \leq \tilde\alpha$*

$$\|(H_{\mathrm{el}} + i)(H_f + 1)^m(\Phi_\alpha - \Phi_0)\| \leq K_m \alpha^{\frac{3}{2}}. \tag{A.19}$$

Proof. Let $\lambda := (E_0 + e_1)/2$. Thanks to (A.7) in Proposition A.3, we may assume that $\sup_{\alpha \leq \tilde\alpha} E_\alpha < \lambda$ by making $\tilde\alpha$ smaller, if necessary. Pick $g \in C_0^\infty(\mathbb{R})$ with $\mathrm{supp}\, g \subset (-\infty, \lambda)$ and with $g(E_\alpha) = 1$ for all $\alpha \leq \tilde\alpha$. On the one hand,

$$\|(H_{\mathrm{el}} + i)(H_f + 1)^m g(H_0)(\Phi_\alpha - \Phi_0)\|$$
$$\leq \|(H_{\mathrm{el}} + i)(H_f + 1)^m g(H_0)\|\|\Phi_\alpha - \Phi_0\| = \mathcal{O}(\alpha^{3/2})$$

by (A.8). On the other hand, $(1 - g(H_0))(\Phi_\alpha - \Phi_0) = (g(H_\alpha) - g(H_0))\Phi_\alpha$ by construction of g. Hence it remains to prove that

$$\|(H_{\mathrm{el}} + i)(H_f + 1)^m(g(H_\alpha) - g(H_0))\Phi_\alpha\| = \mathcal{O}(\alpha^{3/2}). \tag{A.20}$$

To do so, we use the Helffer-Sjöstrand functional calculus with a compactly supported almost analytic extension $\tilde{g}$ of g that satisfies an estimate $|\partial_{\bar z}\tilde{g}(z)| \leq C|y|^2$. Here and henceforth $z = x + iy$ with $x, y \in \mathbb{R}$. It follows that

$$(H_{\mathrm{el}} + i)(H_f + 1)^m(g(H_\alpha) - g(H_0))\Phi_\alpha$$
$$= -\frac{1}{\pi}\int_{\mathbb{R}^2}(H_{\mathrm{el}} + i)(H_0 - z)^{-1}(H_f + 1)^m W(H_\alpha - z)^{-1}\Phi_\alpha \frac{\partial\tilde{g}}{\partial\bar z}\, dxdy \tag{A.21}$$

where

$$(H_f + 1)^m W = \sum_{l=0}^{m}\binom{m}{l}\mathrm{ad}_{H_f}^l(W)(H_f + 1)^{m-l} =: \alpha^{3/2}\tilde{W}(m)(H_f + 1)^m. \tag{A.22}$$

From the equations $[H_f, a^*(G_x)] = a^*(\omega G_x)$ and $[H_f, a(G_x)] = -a(\omega G_x)$ it is clear that the operator $\tilde{W}(m)$, defined by (A.22), is H_0-bounded. Hence we can estimate the norm of (A.21) from above by

$$\frac{\alpha^{3/2}}{\pi}\|(H_{\mathrm{el}} + i)(H_0 + i)^{-1}\|\int\left|\frac{\partial\tilde{g}}{\partial\bar z}\right|\left\|\frac{H_0 + i}{H_0 - z}\right\|\frac{1}{|z - E_\alpha|}\, dxdy$$
$$\times \|\tilde{W}(m)(H_0 + i)^{-1}\|\|(H_0 + i)(H_f + 1)^m\Phi_\alpha\|. \tag{A.23}$$

The integral is finite by construction of $\tilde{g}$, because $|z - E_\alpha|^{-1} \leq |y|^{-1}$, and because $\|(H_0 + i)(H_0 - z)^{-1}\| \leq 1 + (1 + |x|)/|y|$ by the spectral theorem. The last factor in (A.23) is bounded uniformly in $\alpha \leq \tilde\alpha$ by (A.14) and (A.17) from Lemma A.4. This establishes (A.20) and thus concludes the proof of the lemma. □

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Lemma A.6. *Suppose that V satisfies the hypotheses in Section 2. Then*

(i) $\mathbf{x}\varphi_{el} \in D(H_{el})$ *and* $(H_{el} - E_0)\mathbf{x}\varphi_{el} = -2\nabla\varphi_{el}$.

(ii) $e^{-iH_{el}t}\mathbf{x}\varphi_{el} \in D(|\mathbf{x}|^2)$ *and there exists a constant C such that for all $t \in \mathbb{R}$,*

$$\||\mathbf{x}|^2 e^{-iH_{el}t}\mathbf{x}\varphi_{el}\| \le C(1 + t^2).$$

Proof. (i) For all $\gamma \in C_0^\infty(\mathbb{R}^3)$ we have $\mathbf{x}H_{el}\gamma = H_{el}\mathbf{x}\gamma + 2\nabla\gamma$ and hence

$$\langle H_{el}\gamma, \mathbf{x}\varphi_{el}\rangle = \langle H_{el}\mathbf{x}\gamma + 2\nabla\gamma, \varphi_{el}\rangle$$
$$= \langle \gamma, E_0\mathbf{x}\varphi_{el} - 2\nabla\varphi_{el}\rangle.$$

Since $C_0^\infty(\mathbb{R}^3)$ is a core of H_{el}, we conclude that $\mathbf{x}\varphi_{el} \in D(H_{el})$ and that

$$H_{el}\mathbf{x}\varphi_{el} = E_0\mathbf{x}\varphi_{el} - 2\nabla\varphi.$$

(ii) Let $\psi := x_i\varphi_{el}$ for some $i \in \{1, 2, 3\}$. We shall only need that $\psi \in D(|\mathbf{x}|^2) \cap D(-\Delta)$ which follows from (i). By the fundamental theorem of calculus, in a weak sense

$$e^{itH_{el}}|\mathbf{x}|^2 e^{-itH_{el}}\psi = \mathbf{x}^2\psi + \int_0^t e^{isH_{el}}[iH_{el}, |\mathbf{x}|^2]e^{-isH_{el}}\psi\, ds$$

$$= |\mathbf{x}|^2\psi + 2\int_0^t e^{isH_{el}}(\mathbf{x}\cdot\mathbf{p} + \mathbf{p}\cdot\mathbf{x})e^{-isH_{el}}\psi\, ds$$

$$= |\mathbf{x}|^2\psi + 2t(\mathbf{x}\cdot\mathbf{p} + \mathbf{p}\cdot\mathbf{x})\psi + 2\int_0^t ds \int_0^s dr\, e^{irH_{el}}(4\mathbf{p}^2 - \mathbf{x}\cdot\nabla V)e^{-irH_{el}}\psi. \quad \text{(A.24)}$$

Here $\psi \in D(|\mathbf{x}|^2) \cap D(-\Delta) \subset D(\mathbf{x}\cdot\mathbf{p} + \mathbf{p}\cdot\mathbf{x})$ and $e^{-irH_{el}}\psi \in D(H_{el}) = D(-\Delta)$ because $\psi \in D(H_{el})$ by part (i). Therefore assertion (ii) follows from (A.24) and from the hypotheses on V. $\qquad\square$

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