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Atoms in multi-mode radiation fields

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Abstract. The Floquet formalism for atoms in semiclassical monochromatic radiation fields has recently been extended to the multi-mode case by Chu and co-workers. In the present work we generalise a version of the Floquet formalism for the single-mode case originated by Howland to the multi-mode situation and give expressions for multi-photon ionisation amplitudes in the spirit of time-independent scattering theory. A comparison is made with the corresponding second quantised formalism.

1. Introduction

It sometimes happens that the time development of a closed physical system can be described in an approximate but still fairly accurate way in terms of the time development of a subsystem which is generated by a time-dependent Hamiltonian. The advantage is that one is now dealing with a system associated with a smaller Hilbert space (in the quantum case; in the classical case a smaller phase space), the trade-off being the time dependence of the Hamiltonian, which prevents the use of the well developed formalism of time-independent perturbation theory. As examples we mention the impact parameter method used for the description of electron-detachment processes in atomic collisions and the semiclassical method applied to atomic processes in (strong) radiation fields. In the first case the relative motion of the heavy particles is given by a prescribed classical orbit whereas in the second the radiation field is a prescribed external field. In both examples we encounter the typical situation where the influence of one subsystem on the other is strong but the converse influence is weak. Thus, in the first case, the heavy particle motion strongly influences the active electron but the behaviour of the electron does not affect the heavy particle motion appreciably. In the second case the state of an atom is significantly changed by the absorption of a number of photons but the state of the field (for instance a coherent one) remains nearly the same.

Starting from the classical concept of treating energy and time as canonical variables for non-conservative systems, Howland (1974, 1979) developed a corresponding quantum mechanical theory leading to a time-independent Hamiltonian but on a larger Hilbert space. In the first paper he considered the general situation, whereas in the second the Hamiltonian is assumed to be periodic in time which leads to a Floquet reduction. The special case of an atom in a periodic time-dependent field was studied by a number of authors (Yajima 1982, Howland 1983, Tip 1983, Graffi *et al* 1984, Graffi 1984).

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In actual experiments concerning atoms in external radiation fields it sometimes happens that two independent photon sources (lasers) are used and the above formalism ceases to be applicable. Starting from Shirley's version of the Floquet formalism (Shirley 1965), Chu and co-workers (Chu *et al* 1983, Ho and Chu 1984, 1985) developed an extension to the multi-mode situation which they applied to a number of physical processes.

In the present paper we discuss how Howland's method can be adapted to incorporate such more general situations. Basically it amounts to the multiple time scale method (see Langhoff *et al* 1972) converted to the energy domain. As a starting point we suppose that the time development of the system under consideration is governed by a time-dependent Hamiltonian

$$H(t) = H(f_1(\omega_1 t), \ldots, f_N(\omega_N t)), \tag{1.1}$$

with associated unitary time-evolution operator $U(t, s)$ ($U(t, t) = 1$, $\forall t \in \mathbb{R}$) both acting in the Hilbert space $\mathcal{H}$. Here the functions $f_j(\cdot)$ are supposed to be smooth, periodic or non-periodic. We now consider the auxiliary system (again in $\mathcal{H}$) with Hamiltonian ($\boldsymbol{\omega} = \{\omega_1, \ldots, \omega_N\}$ and $\boldsymbol{y} = \{y_1, \ldots, y_N\}$ are in $\mathcal{R}^N$)

$$H(t, \boldsymbol{y}) = H(f_1(y_1 + \omega_1 t), \ldots, f_N(y_N + \omega_N t)) = H(\boldsymbol{y} + \boldsymbol{\omega} t), \tag{1.2}$$

with associated time-evolution operator $U(t, s, \boldsymbol{y})$.

Assuming continuity in $\boldsymbol{y}$ we retrieve the true time-evolution operator by setting $\boldsymbol{y} = \boldsymbol{0}$. We now consider $\boldsymbol{y}$ as a set of new dynamical variables, i.e. as a set of coordinates and introduce a set of conjugate momenta $\boldsymbol{q} = \{q_1, \ldots, q_N\}$, $q_j = -\mathrm{i}\partial_{y_j}$. Here we have to be specific about the ranges of the y_j. In case $f_j(y_j)$ is periodic with period 2π we take $q_j = -\mathrm{i}\partial_{y_j}$ with periodic boundary conditions in 0 and 2π which makes q_j self-adjoint with spectrum $\mathbb{Z}$ on $\mathcal{H}_j = L^2([0, 2\pi], \mathrm{d}y_j)$. If $f_j(y_j)$ is defined for every real y_j and not periodic we take $\mathcal{H}_j = L^2(\mathbb{R}, \mathrm{d}y_j)$ so that q_j is essentially self-adjoint on $\mathcal{H}_j$. We denote its closure again by q_j. In the periodic case the pair $\{y_j, q_j\}$ is not canonical in the usual sense but this is of no consequence in the following. We now denote $\mathcal{H}_0 = \bigotimes_{j=1}^{N} \mathcal{H}_j$ and $\mathcal{K} = \mathcal{H}_0 \otimes \mathcal{H}$ and consider $H(t, \boldsymbol{y})$ as an operator acting in $\mathcal{K}$. Now, formally,

$$H(\boldsymbol{y} + \boldsymbol{\omega} t) = \exp(\mathrm{i}\boldsymbol{\omega} \cdot \boldsymbol{q} t) H(\boldsymbol{y}) \exp(-\mathrm{i}\boldsymbol{\omega} \cdot \boldsymbol{q} t), \tag{1.3}$$

and we can interpret (1.3) as the interaction representation associated with the Hamiltonian

$$K = \boldsymbol{\omega} \cdot \boldsymbol{q} + H(\boldsymbol{y}), \tag{1.4}$$

acting in $\mathcal{K}$. We note that in the multi-periodic case ($N > 1$) the spectrum of $\boldsymbol{\omega} \cdot \boldsymbol{q}$ is either a dense point spectrum (if at least two ω_j are incommensurate) or an infinite set of equidistant eigenvalues of infinite degeneracy ($\omega_j = n_j \omega_0$, $n_j \in \mathbb{Z}$, ω_0 real). Now

$$U(t, s, \boldsymbol{y}) = \exp(\mathrm{i}\boldsymbol{\omega} \cdot \boldsymbol{q} t) \exp[-\mathrm{i}K(t - s)] \exp(-\mathrm{i}\boldsymbol{\omega} \cdot \boldsymbol{q} s), \tag{1.5}$$

and

$$\exp(-\mathrm{i}Kt) = \exp(-\mathrm{i}\boldsymbol{\omega} \cdot \boldsymbol{q} t) U(t, 0, \boldsymbol{y}) = U(0, -t, \boldsymbol{y}) \exp(-\mathrm{i}\boldsymbol{\omega} \cdot \boldsymbol{q} t), \tag{1.6}$$

where the last line is obtained from

$$\exp(-\mathrm{i}\boldsymbol{\omega} \cdot \boldsymbol{q} u) U(t, 0, \boldsymbol{y}) \exp(\mathrm{i}\boldsymbol{\omega} \cdot \boldsymbol{q} u) = U(t, 0, \boldsymbol{y} - \boldsymbol{\omega} u) = U(t - u, -u, \boldsymbol{y}),$$

due to the definition of $H(t, \boldsymbol{y})$.

Equation (1.6) differs from Howland's definition of K in that he defines ($N=1$, $y=t$, $q=-\mathrm{i}\partial_t$, u real)

$$\exp(-\mathrm{i}Ku)=U(t,0)\exp(-\mathrm{i}qu)U(0,t)=\exp(-\mathrm{i}qu)U(t+u,t). \tag{1.7}$$

It is precisely this slight change in definition that leads to the greater flexibility of the present approach. According to the multiple time scale method the time variable is replaced by a set $\{t_j=\varepsilon_j t\}$ (Langhoff *et al* 1972). In our case $\omega_j\partial_{y_j}$ is one of the new time derivatives, i.e. $\varepsilon_j=\omega_j^{-1}$, and in fact we are dealing with a Hilbert space version of this approach. In the following we shall give a precise meaning to the above formal manipulations for the case of a one-electron atom in a spatially homogeneous multi-mode field. The starting point is the propagator properties and continuity in y of $U(t,s,y)$. It then follows that the right-hand side of (1.6) defines a strongly continuous unitary group of operators acting in $\mathscr{H}$ so that K is defined as a self-adjoint operator acting in $\mathscr{H}$. This is done in § 2 starting from (in atomic units)

$$H(t)=\tfrac{1}{2}(\boldsymbol{p}-\boldsymbol{A}(t))^2+V(\boldsymbol{x}), \tag{1.8}$$

where $\boldsymbol{A}(t)$ is the vector potential, $\boldsymbol{p}$ the momentum operator and $V(\boldsymbol{x})$ the potential, the underlying Hilbert space being $\mathscr{H}=L^2(\mathbb{R}^3,\mathrm{d}\boldsymbol{x})$. This approach does not give much information about the domain $\mathscr{D}$ of K but in § 3 we obtain more detailed information about $\mathscr{D}$ under suitable conditions on $V(\boldsymbol{x})$, treated as a perturbation. We consider vector potentials of the type

$$\boldsymbol{A}(t,\boldsymbol{y})=\sum_{j=1}^{N}\boldsymbol{a}_j\cos(\omega_j t+y_j)=\boldsymbol{A}(\boldsymbol{y}+\boldsymbol{\omega}t), \tag{1.9}$$

with $\boldsymbol{a}_j\in\mathbb{R}^3$, $y_j\in[0,2\pi]$ and $\omega_j>0$ ($\omega_j<0$ is allowed but no generality is lost by requiring $\omega_j>0$). Thus $\boldsymbol{a}_j$ is the amplitude of the jth field mode, ω_j its frequency and y_j its phase. In an actual experiment the phases of the different modes are usually uncorrelated and the initial value of $\boldsymbol{y}$ is different from one measurement to another, i.e. the y_j occur as random variables. Thus in the present case the y_j are not artificial parameters but have a physical meaning. In the case when one field mode is obtained from another by frequency doubling (or higher multiplication) then the corresponding phases remain correlated and only one phase parameter y_j should be attached to both modes, whereas the corresponding cosine function in (1.9) has to be replaced by a more complicated periodic function with the same fundamental frequency.

In § 4 we discuss, without going into mathematical details, the connection between the present formalism and actual measurable quantities such as they occur, for instance, in multi-photon ionisation.

The paper ends with a discussion section where we raise the question as to what has been gained by first going to an approximate formulation on a smaller Hilbert space and then going back to a larger one. In the case at hand the present procedure can be compared directly with that of a finite number of radiation modes in second quantisation (Grossmann and Tip (1980) studied this model for hydrogen in a single mode; for further results see also Perry (1983)).

2. One-electron atoms in a multi-mode field

We consider a system where a particle (electron) is moving in a potential $V(\boldsymbol{x})$ which is either the Coulomb potential (hydrogen atom H, He^+, in the Born-Oppenheimer

approximation with spin neglected) or an effective potential representing the combined action of the nucleus and the core electrons on the active electron (Li, Na, etc). Thus on $\mathscr{H} = L^2(\mathbb{R}^3, \mathrm{d}\boldsymbol{x}) = L^2$

$$H^{\mathrm{at}} = \tfrac{1}{2}\boldsymbol{p}^2 + V(\boldsymbol{x}), \tag{2.1}$$

where $\frac{1}{2}\boldsymbol{p}^2$ with domain $\mathscr{D}$ is the closure of $-\frac{1}{2}\partial_x^2$ and $V(\boldsymbol{x}) \in L^p + L^\infty$, $p \geqslant 2$, real, so that $V(\boldsymbol{x})$ is $\frac{1}{2}\boldsymbol{p}^2$-bounded with zero relative bound and H^{at} with domain $\mathscr{D}$ is self-adjoint. The Hamiltonian for the atom in the field $\boldsymbol{A}(t, y)$ is now ($\boldsymbol{p}$ is the closure of $-\mathrm{i}\partial_x$)

$$H(t, y) = \tfrac{1}{2}[\boldsymbol{p} - \boldsymbol{A}(t, y)]^2 + V(\boldsymbol{x}) = H(y + \boldsymbol{\omega}t), \tag{2.2}$$

which is again self-adjoint with domain $\mathscr{D}$. The associated time-evolution operator $U(t, s, y)$ is the solution of

$$\partial_t U(t, s, y) = -\mathrm{i}H(t, y)U(t, s, y), \qquad U(t, t, y) = 1. \tag{2.3}$$

$U(t, s, y)$ is uniquely defined by (2.3) for each y, is unitary and strongly continuous in t and s (see Kato (1970); for his auxiliary space $\mathscr{Y}$ we take $\mathscr{D}$ equipped with the graph norm). It can be seen by considering Kato's construction of the propagator $U(t, s, y)$, i.e. it is in addition strongly continuous in y. We note further that, since $H(t, y + \alpha\boldsymbol{\omega}) = H(t + \alpha, y)$,

$$U(t, s, y + \alpha\boldsymbol{\omega}) = U(t + \alpha, s + \alpha, y). \tag{2.4}$$

Splitting y according to

$$y = y_{\parallel} + y_{\perp}, \qquad y_{\parallel} = \omega^{-2}(y \cdot \boldsymbol{\omega})\boldsymbol{\omega}, \tag{2.5}$$

we thus have

$$U(t, s, y) = U(t + \omega^{-2}y \cdot \boldsymbol{\omega}, s + \omega^{-2}y \cdot \boldsymbol{\omega}, y_{\perp}). \tag{2.6}$$

Since for $f \in \mathscr{D}$ (see Kato 1970)

$$\begin{aligned} \partial_t U(t, s, y)f &= -\mathrm{i}H(t, y)U(t, s, y)f, \\ \partial_s U(t, s, y)f &= \mathrm{i}U(t, s, y)H(s, y)f, \end{aligned} \tag{2.7}$$

it follows from (2.6) that for $f \in \mathscr{D}$

$$-\mathrm{i}\partial_{y\cdot\omega} U(t, s, y)f = -\omega^{-2}[H(t, y)U(t, s, y)f - U(t, s, y)H(s, y)f]. \tag{2.8}$$

We now introduce $\mathscr{K}$ as discussed in the introduction. Thus $\mathscr{H}_j = L^2([0, 2\pi], \mathrm{d}y_j)$, $q_j = -\mathrm{i}\partial_{y_j}$ with periodic boundary conditions so that q_j has spectrum $\mathbb{Z}$ and the eigenfunction associated with $n \in \mathbb{Z}$ is $\phi_n(y_j) = (2\pi)^{-1/2}\exp(\mathrm{i}ny_j)$. It follows that $T = \boldsymbol{\omega} \cdot \boldsymbol{q}$, $\boldsymbol{q} = \{q_1, \ldots, q_N\}$ is self-adjoint on $\mathscr{H}_0 = \bigotimes_{j=1}^N \mathscr{H}_j$ and has pure point spectrum, its eigenvalues being $\lambda_n = \boldsymbol{\omega} \cdot \boldsymbol{n}$, $\boldsymbol{n} \in \mathbb{Z}^N$. $\{\exp(\mathrm{i}Tt) | t \in \mathbb{R}\}$ defines a unitary group on $\mathscr{H}_0$ and for $f(y) \in \mathscr{H}_0$, $[\exp(\mathrm{i}Tt)f](y) = f(y + \boldsymbol{\omega}t + \boldsymbol{v})$ where $\boldsymbol{v} \in (2\pi\mathbb{Z})^N$ is such that $y_j + \omega_j t + v_j \in [0, 2\pi)$. $\Phi(y) \in L^\infty(\mathbb{R}^N, \mathrm{d}y)$ defines a bounded multiplication operator on $\mathscr{H}_0$ and $\exp(\mathrm{i}Tt)\Phi(y)\exp(-\mathrm{i}Tt) = \Phi(y + \boldsymbol{\omega}t + \boldsymbol{v})$ with $\boldsymbol{v}$ as before. In the following we omit $\boldsymbol{v}$ if no confusion can arise. $\exp(\mathrm{i}Tt) \otimes I_{\mathscr{H}}$ defines a unitary, strongly continuous group on $\mathscr{K} = \mathscr{H}_0 \otimes \mathscr{H}$ which we again denote by $\exp(\mathrm{i}Tt)$. $U(t, s, y)$ defines a family of unitary operators on $\mathscr{K}$ which is multiplicative as far as its action in $\mathscr{H}_0$ is concerned. We now define the unitary operator $W(t)$ on $\mathscr{K}$ for each $t \in \mathbb{R}$ by

$$\begin{aligned} W(t) &\equiv \exp(-\mathrm{i}Tt)U(t, 0, y) = U(t, 0, y - \boldsymbol{\omega}t)\exp(-\mathrm{i}Tt) \\ &= U(0, -t, y)\exp(-\mathrm{i}Tt), \end{aligned} \tag{2.9}$$

where (2.4) has been used. Again using (2.5) and the propagator property $U(t, s, \boldsymbol{y}) = U(t, u, \boldsymbol{y})U(u, s, \boldsymbol{y})$; $t, s, u \in \mathbb{R}$ (see Kato 1980) we find that

$$\begin{aligned}\exp[-\mathrm{i}T(t_1+t_2)]U(t_1+t_2, 0, \boldsymbol{y}) &= \exp(-\mathrm{i}Tt_1)U(t_1+t_2, 0, \boldsymbol{y}-\boldsymbol{\omega}t_2)\exp(-\mathrm{i}Tt_2)\\ &= \exp(-\mathrm{i}Tt_1)U(t_1, -t_2, \boldsymbol{y})\exp(-\mathrm{i}Tt_2)\\ &= \exp(-\mathrm{i}Tt_1)U(t_1, 0, \boldsymbol{y})U(0, -t_2, \boldsymbol{y})\exp(-\mathrm{i}Tt_2)\\ &= \exp(-\mathrm{i}Tt_1)U(t_1, 0, \boldsymbol{y})\exp(-\mathrm{i}Tt_2)U(t_2, 0, \boldsymbol{y}),\end{aligned}$$

i.e. $W(t)$ has the group property. Since it is also strongly continuous in t Stone's theorem can be applied and we have $W(t) = \exp(-\mathrm{i}Kt)$ with K self-adjoint on $\mathscr{H}$.

Proposition 2.1. The dense set $\mathscr{D}_0 = (\mathscr{D}(T) \otimes \mathscr{H}) \cap (\mathscr{H}_0 \otimes \mathscr{D}) \subset \mathscr{H}$ is a core of K.

Proof. Let $f \in \mathscr{D}_0$ and $g(t) = \exp(-\mathrm{i}Kt)f$. Then $\|\boldsymbol{p}^2 g(t)\| = \|\boldsymbol{p}^2 U(t, s, \boldsymbol{y})f\| < \infty$ since $U(t, s, \boldsymbol{y})$ is a bounded operator on $\mathscr{D}$ equipped with the graph norm (Kato 1970) and similarly, using (2.8), $\|Tg(t)\| = \|\boldsymbol{\omega} \cdot \boldsymbol{q} U(t, s, \boldsymbol{y})f\| \leqslant \|\partial_{\boldsymbol{\omega}\cdot\boldsymbol{y}} U(t, s, \boldsymbol{y})f\| + \|Tf\| < \infty$ so that $g(t) \subset \mathscr{D}_0$. Consequently $\mathscr{D}_0$ is a core of K (Reed and Simon 1972, theorem VIII 10).

Proposition 2.1 is a simple extension of a result by Graffi *et al* (1984). Since, for $f \in \mathscr{D}_0$,

$$\begin{aligned}\partial_t \exp(-\mathrm{i}Kt)f &= -\mathrm{i}T\exp(-\mathrm{i}Kt)f - \mathrm{i}\exp(-\mathrm{i}Tt)H(t, \boldsymbol{y})U(t, s, \boldsymbol{y})f\\ &= -\mathrm{i}(T + H(\boldsymbol{y}))\exp(-\mathrm{i}Kt)f, \qquad (2.10)\end{aligned}$$

where $H(\boldsymbol{y}) = H(0, \boldsymbol{y})$, it follows that $T + H(\boldsymbol{y})$ is essentially self-adjoint on $\mathscr{D}_0$ with closure K.

At this point it is convenient to switch to the Kramers representation (Kramers 1950). Thus we introduce the Hertz vector ($\boldsymbol{A}(t) = \partial_t \boldsymbol{Z}(t, 0)$)

$$\boldsymbol{Z}(t, \boldsymbol{y}) = \sum_{i=1}^{N} \boldsymbol{a}_i \sin(\omega_i t + y_i)/\omega_i, \qquad \boldsymbol{Z}(\boldsymbol{y}) = \boldsymbol{Z}(0, \boldsymbol{y}), \qquad (2.11)$$

and the function

$$\begin{aligned}\Psi(t, \boldsymbol{y}) = \frac{1}{4}\sum_{i,j=1}^{N} \boldsymbol{a}_i \cdot \boldsymbol{a}_j(\omega_i + \omega_j)^{-1}\sin[(\omega_i + \omega_j)t + y_i + y_j]\\ + \frac{1}{4}\sum_{\substack{i=1\\ \omega_i \neq \omega_j}}^{N} \boldsymbol{a}_i \cdot \boldsymbol{a}_j(\omega_i - \omega_j)^{-1}\sin[(\omega_i - \omega_j)t + y_i - y_j], \qquad (2.12)\end{aligned}$$

$\Psi(\boldsymbol{y}) = \Psi(0, \boldsymbol{y})$.

Let now

$$M(t) = M(t, \boldsymbol{p}, \boldsymbol{y}) = \boldsymbol{p} \cdot \boldsymbol{Z}(t, \boldsymbol{y}) - \Psi(t, \boldsymbol{y}), \qquad M = M(\boldsymbol{p}, \boldsymbol{y}) = M(0, \boldsymbol{p}, \boldsymbol{y}). \qquad (2.13)$$

$M(t)$ defines a self-adjoint operator on $\mathscr{H}$ with time-dependent domain. Note, however, that

$$M(t) = \exp(\mathrm{i}Tt)M\exp(-\mathrm{i}Tt). \qquad (2.14)$$

We now consider the free (i.e. $V \equiv 0$) evolution operator

$$\exp(-\mathrm{i}K_0 t) = \exp(-\mathrm{i}Tt)U_0(t, 0, \boldsymbol{y}), \qquad (2.15)$$

where $U_0(t, s, y)$ is the propagator associated with $H_0(t) = \frac{1}{2}[p - A(t, y)]^2$. Explicitly

$$U_0(t, 0, y) = \exp\left(-i \int_0^t ds H_0(s)\right)$$
$$= \exp[-i(\tfrac{1}{2}p^2 + C)t] \exp[-i(M - M(t))], \tag{2.16}$$

where

$$C = \frac{1}{4} \sum_{i,j=1}^{N} a_i \cdot a_j \cos(y_i - y_j) \delta_{\omega_i \omega_j} \tag{2.17}$$

defines a Hermitian (=bounded, self-adjoint) operator on $\mathcal{H}$, which commutes with T. Now, using (2.14),

$$\exp(-iK_0 t) = \exp(-iTt) \exp[-i(\tfrac{1}{2}p^2 + C)t] \exp(iM(t)) \exp(-iM)$$
$$= \exp(-iTt) \exp[-i(\tfrac{1}{2}p^2 + C)t] \exp(iTt) \exp(iM) \exp(-iTt) \exp(-iM)$$
$$= \exp(iM) \exp[-i(T + \tfrac{1}{2}p^2 + C)t] \exp(-iM), \tag{2.18}$$

so that K_0 and

$$\hat{K}_0 = T + \tfrac{1}{2}p^2 + C = \check{K}_0 + C \tag{2.19}$$

are unitarily equivalent. (As T and $\frac{1}{2}p^2$ are self-adjoint, acting in $\mathcal{H}_0$, respectively in $\mathcal{H}$, their direct sum defines a unique self-adjoint operator $\check{K}_0$ in $\mathcal{H}$. Since C is Hermitian $\hat{K}_0$ is well defined.)

A special situation occurs if $\omega_i = \omega_j$ for some $i \neq j$ (two uncorrelated laser beams with the same frequency). Disregarding the constant terms in C for the moment, we can decompose $T_0 + C$ into groups

$$L_{\omega_\alpha} = \omega_\alpha(q_1 + \ldots + q_s) + \sum_{1 \leqslant i \leqslant j \leqslant s} c_{ij} \cos(y_i - y_j). \tag{2.20}$$

But now

$$\exp[iy_1(q_2 + \ldots + q_s)] L_{\omega_\alpha} \exp[-iy_1(q_2 + \ldots + q_s)]$$
$$= \omega_\alpha q_1 + \sum_{j=2}^{s} c_{1j} \cos y_j + \sum_{2 \leqslant i < j \leqslant s} c_{ij} \cos(y_i - y_j). \tag{2.21}$$

By relabelling the y_j we thus find that K_0 is unitarily equivalent to ($N_0 \leqslant N$)

$$\tilde{K}_0 = \frac{1}{2}p^2 + \sum_{j=1}^{N_0} \omega_j q_j + \frac{1}{4} \sum_{i=1}^{N} a_i^2 + D(y_{N_0+1}, \ldots, y_N) \tag{2.22}$$

where D is a real bounded function of $y_{N_0+1}, \ldots, y_N$. In the special case that $N = 2$ and $\omega_1 = \omega_2 = \omega$, we have

$$\tilde{K}_0 = \tfrac{1}{2}p^2 + \omega q_1 + \tfrac{1}{4}(a_1^2 + a_2^2 + 2a_1 \cdot a_2 \cos y_2). \tag{2.23}$$

Thus the effect of coinciding ω_j is that a representation can be found where less than N q_j occur in the free generator (but there are additional $\exp(iy_\alpha q_\beta)$, $\alpha \neq \beta$, in the original representation). We now define

$$\hat{U}(t, 0, y) = \exp(-iM(t)) U(t, 0, y) \exp(iM), \tag{2.24}$$

which is generated by

$$\hat{H}(t, y) = \tfrac{1}{2}p^2 + C + V(x - Z(t, y)) = \check{H}(t, y) + C. \tag{2.25}$$

$\hat{H}(t, y)$, which is again self-adjoint with domain $\mathscr{D}$, is the Kramers representation of the full Hamiltonian. Apart from C (which has important consequences in terms of a shift of the ionisation potential (see Muller *et al* 1983, Muller and Tip 1984, Tip 1984)) the action of the field has been moved to the potential. $\boldsymbol{Z}(t, y)$ being bounded, this has the advantage that, apart from C, the influence of the field is absent for large distances, which is a convenient feature in numerical investigations. We now introduce $\hat{K}$ through

$$\exp(-\mathrm{i}\hat{K}t) = \exp(-\mathrm{i}Tt)\hat{U}(t, 0, y). \tag{2.26}$$

In the same way as before (for K) we find that $\hat{K}$ is the self-adjoint closure of $T+\frac{1}{2}\boldsymbol{p}^2+C+V(\boldsymbol{x}-\boldsymbol{Z}(y))$, defined on $\mathscr{D}_0$. Thus, on $\mathscr{D}_0$,

$$\hat{K} = \hat{K}_0 + V(\boldsymbol{x}-\boldsymbol{Z}(y)) = \check{K}_0 + C + V(\boldsymbol{x}-\boldsymbol{Z}(y)) = \check{K} + C. \tag{2.27}$$

For bounded $V(\boldsymbol{x})$ $\hat{K}$ is self-adjoint with domain $\mathscr{D}(\hat{K}_0)$ but for more general potentials the situation can be more complicated as will be discussed in § 3. We can also consider the original operator K. On $\mathscr{D}_0$ we can split it according to

$$K = T + H^{\mathrm{at}} - \boldsymbol{p}\cdot\boldsymbol{A}(y) + \tfrac{1}{2}\boldsymbol{A}(y)^2 = K_1 - \boldsymbol{p}\cdot\boldsymbol{A}(y) + \tfrac{1}{2}\boldsymbol{A}(y)^2. \tag{2.28}$$

Here $\frac{1}{2}\boldsymbol{A}(y)^2$ is bounded and causes no problems but even for $V \equiv 0$ $-\boldsymbol{p}\cdot\boldsymbol{A}(y)$ is not a perturbation of K, with relative bound smaller than one. This is related to the circumstance that T is not bounded from below.

In conclusion we note that in the case of coinciding frequencies the development (2.20)-(2.22) also works for $\hat{K}$. On $\mathscr{D}_0$ we simply have $\hat{K} = \hat{K}_0 + V(\boldsymbol{x}-\boldsymbol{Z}(y))$.

3. Relative boundedness properties of V

In this section we turn to the question of whether $V = V(\boldsymbol{x}-\boldsymbol{Z}(y))$ in $\hat{K} = \hat{K}_0 + V$ can be interpreted as a perturbation of $\hat{K}_0$ or, alternatively, since C is bounded, of $\check{K}_0 = T + \frac{1}{2}\boldsymbol{p}^2$. This will turn out to be the case although, depending on the local singularities of V, there are situations where V has no simple $\hat{K}_0$-boundedness properties but the construction through the pseudo-Friedrichs extension (Kato 1966, Faris 1975) has to be used instead. We start with the derivation of an operator estimate. Let $\lambda \in \mathbb{R}$, $\mu > 0$ and $\alpha > 0$. Then

$$(\lambda^2+\mu^2)^{-\alpha} = (2\pi)^{-1/2}\int_{-\infty}^{+\infty} \mathrm{d}t\, F_\alpha(\mu, t)\exp(\mathrm{i}\lambda t), \tag{3.1}$$

where

$$\begin{aligned} F_\alpha(\mu, t) &= (2\pi)^{-1/2}\int_{-\infty}^{+\infty} \mathrm{d}\lambda\,(\lambda^2+\mu^2)^{-\alpha}\exp(-\mathrm{i}\lambda t) \\ &= (2/\pi)^{1/2}\mu^{1-2\alpha} g_\alpha(\mu t), \end{aligned} \tag{3.2}$$

with (Magnus *et al* 1966, p 85)

$$g_\alpha(v) = \int_0^\infty \mathrm{d}\lambda\,(\lambda^2+1)^{-\alpha}\cos(\lambda v) = \pi^{1/2}(v/2)^{\alpha-1/2}\Gamma(\alpha)^{-1}K_{\alpha-1/2}(v), \tag{3.3}$$

$K_{\alpha-1/2}(v)$ being a modified Bessel function. By partial integration

$$g_\alpha(v) = (2\alpha/v)\int_0^\infty \mathrm{d}\lambda\,\lambda(\lambda^2+1)^{-\alpha-1}\sin(\lambda v), \qquad v \neq 0 \tag{3.4}$$

so that

$$|g_\alpha(v)| \leqslant (a\alpha/|v|)\int_0^\infty \mathrm{d}\lambda\, \lambda(\lambda^2+1)^{-\alpha-1} = |v|^{-1}, \qquad v \neq 0. \tag{3.5}$$

We note further that for $\alpha > \frac{1}{2}$, $g_\alpha(v)$ is continuous in v; for $\alpha \leqslant \frac{1}{2}$, $g_\alpha(v)$ is continuous outside $v=0$ and $|g_\alpha(v)|$ behaves as $|v|^{2\alpha-1}$ in $v=0$ for $\alpha < \frac{1}{2}$ and as $\ln|v|$ in $v=0$ for $\alpha = \frac{1}{2}$.

Let now $f \in \mathscr{H}$ and $\Phi(\boldsymbol{x}) \in L^p(\mathbb{R}^3, \mathrm{d}\boldsymbol{x}) \cap L^\infty(\mathbb{R}^3, \mathrm{d}\boldsymbol{x})$, $p > 3$. Then, using (3.1) and the spectral theorem

$$\begin{aligned} I_\alpha &\equiv ((\mu^2 + \check{K}_0^2)^{-\alpha}\Phi(\boldsymbol{x}-\boldsymbol{Z}(y))f, \Phi(\boldsymbol{x}-\boldsymbol{Z}(y))f) \\ &= (2\pi)^{-1/2}\int_{-\infty}^{+\infty} \mathrm{d}t\, F_\alpha(\mu, t)(\exp(-\mathrm{i}\check{K}_0 t)\Phi f, \Phi f) \\ &= (2\pi)^{-1/2}\int_{-\infty}^{+\infty} \mathrm{d}t\, F_\alpha(\mu, t)G(t). \end{aligned} \tag{3.6}$$

Now (positive constants, which may differ from place to place, are denoted by c)

$$\begin{aligned} G(t) &\equiv (\exp(-\mathrm{i}\check{K}_0 t)\Phi f, \Phi f) \\ &= c|t|^{-3/2}\int \mathrm{d}y\, \mathrm{d}\boldsymbol{x}\, \mathrm{d}\boldsymbol{x}' \exp[-\mathrm{i}(\boldsymbol{x}-\boldsymbol{x}')^2/2t] \cdot \Phi[\boldsymbol{x}' - \boldsymbol{Z}(y+\omega t)] \\ &\quad \times f(y+\omega t, \boldsymbol{x}') \cdot \bar{\Phi}(\boldsymbol{x}-\boldsymbol{Z}(y)) \cdot \bar{f}(y, \boldsymbol{x}), \qquad t \neq 0, \end{aligned} \tag{3.7}$$

and, by familiar argument, based upon the properties of the Fourier transform (denoted by a tilde)

$$\begin{aligned} |G(t)| &\leqslant c|t|^{-3/p}\int \mathrm{d}y \|\tilde{\Phi} f(y+\omega t)\|_s \|\Phi f(y)\|_r \\ &\leqslant c|t|^{-3/p}\int \mathrm{d}y \|\Phi f(y+\omega t)\|_r \|\Phi f(y)\|_r \\ &\leqslant c|t|^{-3/p}\|\Phi\|_p^2 \int \mathrm{d}y \|f(y+\omega t)\|_{\mathscr{H}} \|f(y)\|_{\mathscr{H}} \\ &\leqslant c|t|^{-3/p}\|\Phi\|_p^2 \|f\|^2. \end{aligned} \tag{3.8}$$

Here $\|h(y)\|_q = (\int \mathrm{d}\boldsymbol{x}|h(y, \boldsymbol{x})|^q)^{1/q}$, $r^{-1} = \frac{1}{2} + p^{-1}$, $r^{-1} + s^{-1} = 1$, $r, s \geqslant 1$. It follows that

$$\begin{aligned} I_\alpha &\leqslant c\|\Phi\|_p^2\|f\|^2 \int_{-\infty}^{+\infty} \mathrm{d}t|t|^{-3/p}|F_\alpha(\mu, t)| \\ &= c\|\Phi\|_p^2\|f\|^2 \mu^{1-2\alpha}\int_{-\infty}^{+\infty} \mathrm{d}t|t|^{-3/p}|g_\alpha(\mu t)| \\ &= c\mu^{3/p-2\alpha}\|\Phi\|_p^2\|f\|^2 \int_{-\infty}^{+\infty} \mathrm{d}v|v|^{-3/p}|g_\alpha(v)|. \end{aligned} \tag{3.9}$$

It is seen from (3.5) that (3.9) is integrable in $\pm\infty$ so that the only remaining critical point is $v=0$ if $0 < \alpha \leqslant \frac{1}{2}$. $\alpha = \frac{1}{2}$ causes no problems for $p > 3$, whereas for $0 < \alpha < \frac{1}{2}$ we have to require $3/p < 2\alpha$. Thus

$$I_\alpha \leqslant c\mu^{3/p-2\alpha}\|\Phi\|_p^2\|f\|^2, \qquad p > 3, \qquad 3/p < 2\alpha, \tag{3.10}$$

which result extends to arbitrary $\Phi \in L^p$ by a density argument. In particular I_α vanishes for large μ.

Proposition 3.1. Suppose that $V(\boldsymbol{x}) \in L^p(\mathbb{R}^3, \mathrm{d}\boldsymbol{x})$, real, with $p > 3$. Then $V(\boldsymbol{x} - \boldsymbol{Z}(\boldsymbol{y}))$ is K_0-bounded with zero relative bound. Consequently $\hat{K}$ is self-adjoint with domain $\mathscr{D}(\hat{K}_0)$.

Proof. For $f \in \mathscr{H}$ and $\mu > 0$ we have by (3.10) with $\alpha = 1$

$$\|[\mathrm{i}\mu - \check{K}_0]^{-1} V(\boldsymbol{x} - \boldsymbol{Z}(\boldsymbol{y}))f\|^2 = ((\mu^2 + \check{K}_0^2)^{-1} V(\boldsymbol{x} - \boldsymbol{Z}(\boldsymbol{y}))f,\ V(\boldsymbol{x} - \boldsymbol{Z}(\boldsymbol{y}))f)$$
$$\leqslant c\mu^{3/p-2} \| V\|_p^2 \|f\|^2$$

which implies the statement.

This result is not optimal since we required $p > 3$, and among others this leaves out the Coulomb potential. (In fact its singularity in the origin is the root of this trouble since we can split the Coulomb potential in a cut-off part and a bounded tail.)

On the other hand the decay for large μ is faster than needed and indeed we can obtain a result, valid for $p > \frac{3}{2}$, by turning to the pseudo-Friedrichs extension (Kato 1966, Faris 1975). In our case Faris' formulation is the most convenient one. Thus we have to show that $V(\boldsymbol{x} - \boldsymbol{Z}(\boldsymbol{y}))$ defines a bounded sesquilinear form on $Q(\check{K}_0)$, the form domain of $\check{K}_0$, ($\mathscr{D}(|\check{K}_0|^{1/2})$, equipped with the graph norm) and that there exist constants $a, b > 0$, $a < 1$ such that

$$|(V(\boldsymbol{x} - \boldsymbol{Z}(\boldsymbol{y}))f, f)| \leqslant a((|\check{K}_0| + b)f, f), \qquad \forall f \in Q(\check{K}_0). \tag{3.11}$$

Then the form sum $\hat{K} = \check{K}_0 + V(\boldsymbol{x} - \boldsymbol{Z}(\boldsymbol{y})) + C$ is self-adjoint and $\mathscr{D}(\hat{K}) = \{f \in Q(\check{K}_0) | \check{K}_0 + V(\boldsymbol{x} - \boldsymbol{Z}(\boldsymbol{y}))f \in \mathscr{H}\}$.

Proposition 3.2. Suppose that $V(\boldsymbol{x}) \in L^p(\mathbb{R}^3, \mathrm{d}\boldsymbol{x})$, real, with $p > \frac{3}{2}$. Then $\hat{K} = \hat{K}_0 + V(\boldsymbol{x} - \boldsymbol{Z}(\boldsymbol{y}))$ defined by the pseudo-Friedrichs extension exists as a self-adjoint operator on $\mathscr{H}$ and $\mathscr{D}(\hat{K}) = \{f \in Q(\hat{K}_0) | \{\hat{K}_0 + V(\boldsymbol{x} - \boldsymbol{Z}(\boldsymbol{y}))\}f \in \mathscr{H}\}$.

Proof. We have to show that for $\mu > 0$

$$(\mu^{1/2} + |\check{K}_0|^{1/2})^{-1} | V(\boldsymbol{x} - \boldsymbol{Z}(\boldsymbol{y}))| [\mu^{1/2} + |\check{K}_0|^{1/2}]^{-1} \tag{3.12}$$

is a bounded operator on $\mathscr{H}$ which tends to zero for large μ. Since $\|(\mu^{1/2} + |\check{K}_0|^{1/2})^{-1}(\mu^2 + \check{K}_0^2)^{1/4}\| \leqslant 1$ it is sufficient to show this for $(\mu^2 + \check{K}_0^2)^{-1/4} | V(\boldsymbol{x} - \boldsymbol{Z}(\boldsymbol{y}))| \times (\mu^2 + \check{K}_0^2)^{-1/4}$. Let now $V \in L^p \cap L^\infty$, $p > \frac{3}{2}$, so that $|V|^{1/2} \in L^{2p} \cap L^\infty$, $2p > 3$. For every $f \in \mathscr{H}$, according to (3.10) with $\alpha = \frac{1}{2}$,

$$\|(\mu^2 + \check{K}_0^2)^{-1/4} | V(\boldsymbol{x} - \boldsymbol{Z}(\boldsymbol{y}))|^{1/2} f\|^2$$
$$= ((\mu^2 + \check{K}_0^2)^{-1/2} | V(\boldsymbol{x} - \boldsymbol{Z}(\boldsymbol{y}))|^{1/2} f,\ | V(\boldsymbol{x} - \boldsymbol{Z}(\boldsymbol{y}))|^{1/2} f)$$
$$\leqslant c\mu^{3/2p-1} \| |V|^{1/2} \|_{2p}^2 \|f\|^2 = c\mu^{3/2p-1} \| V\|_p \|f\|^2,$$

and it follows that

$$\| |V(\boldsymbol{x} - \boldsymbol{Z}(\boldsymbol{y}))|^{1/2} (\mu^2 + \check{K}_0^2)^{-1/4}\| \leqslant c\mu^{3/4p-1/2} \| V\|_p^{1/2}$$
$$\|(\mu^2 + \check{K}_0^2)^{-1/4} | V(\boldsymbol{x} - \boldsymbol{Z}(\boldsymbol{y}))|^{1/2}\| \leqslant c\mu^{3/4p-1/2} \| V\|_p^{1/2} \tag{3.13}$$

which results hold for every $V \in L^p$, $p > \frac{3}{2}$, by a density argument. Consequently (3.12) defines a bounded operator on $\mathscr{H}$ which vanishes for large μ and the existence of the pseudo-Friedrichs extension follows.

It remains to consider whether $\hat{K} = \hat{K}_3$ defined in this section (through propositions 3.1 or 3.2) coincides with $\hat{K} = \hat{K}_2$ defined in § 2. For $V \in L^p + L^\infty$, $p > 3$, $\mathscr{D}_0$ is in the domain of $\hat{K}_3$, and a core of $\hat{K}_2$ so that $\hat{K}_2 = \hat{K}_3$. In proposition 3.2 we encounter a larger class of potentials than that considered in § 2. Since there are no compelling physical reasons to consider such a larger class (it can probably be done by means of form techniques) we restrict ourselves to $V \in L^p + L^\infty$, $2 \leq p \leq 3$. We note that for $f \in \mathscr{D}_0$ $V(\boldsymbol{x} - \boldsymbol{Z}(\boldsymbol{y}))f \in \mathscr{H}$ since $\| V(\boldsymbol{x} - \boldsymbol{Z}(\boldsymbol{y}))f\| = \|\exp(-\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{Z}(\boldsymbol{y}))\, V(\boldsymbol{x})(1+p^2)^{-1} \times \exp(\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{Z}(\boldsymbol{y}))(1+p^2)f\| \leq \| V(\boldsymbol{x})(1+p^2)^{-1}\|_{\mathscr{H}} \|(1+p^2)f\| < \infty$, $V(\boldsymbol{x})(1+p^2)^{-1}$ being a bounded operator on $\mathscr{H}$. Next we observe that $\mathscr{D}_0 \subset \mathscr{D}(|K_0|^{1/2})$ and is a core of $|K_0|^{1/2}$. This follows from the boundedness of $(1+|K_0|^{1/2})^{-1}$ so that it is sufficient to show (Kato 1966, p 166, problem 5.19) that $(1+|K_0|^{1/2})\mathscr{D}_0$ is dense in $\mathscr{H}$. This is easily seen by turning to Fourier space and noting that the notion of a core is invariant under unitary transformations. Thus $\mathscr{D}_0$ is in the domain of $\hat{K}_3$ and a core of $\hat{K}_2$ so that also in this case $\hat{K}_2 = \hat{K}_3$.

Thus we have found that the generator $\hat{K}$ (and hence K) can be equivalently defined by the construction through the propagator (in § 2) and perturbatively (in this section).

4. Expressions for photoionisation

In §§ 2 and 3 we considered the matter of time evolution from an abstract viewpoint. Here we shall complement these results by deriving expressions for measurable quantities relevant for atomic photoionisation in terms of the resolvent of $\check{K}$. We shall however be forced to make some concessions in the way of mathematical rigour. We shall assume that $V(\boldsymbol{x}) = V_0(\boldsymbol{x}) + \lambda/|\boldsymbol{x}|$, λ real, where $V_0(\boldsymbol{x})$ is locally L^2 and is $\mathscr{O}(|\boldsymbol{x}|^{-1-\varepsilon})$, $\varepsilon > 0$, as $|\boldsymbol{x}| \to \infty$. The atomic wave operators ($H^{\mathrm{at}} = \frac{1}{2}p^2 + V$)

$$\Omega_{\pm}^{\mathrm{at}} = \lim_{t \to \pm\infty} \exp(\mathrm{i}H^{\mathrm{at}}t)\exp[-\mathrm{i}(\tfrac{1}{2}p^2 t + \lambda \ln|t|/p)], \tag{4.1}$$

then exist in the strong sense (on $\mathscr{H}$), $(\Omega_{\pm}^{\mathrm{at}})^*\Omega_{\pm}^{\mathrm{at}} = 1$, $\Omega_{\pm}^{\mathrm{at}}(\Omega_{\pm}^{\mathrm{at}})^* \subset P_{\mathrm{c}}$, the projector associated with the continuum part of H^{at}, and for real α $\Omega_{\pm}^{\mathrm{at}} \exp(\mathrm{i}\alpha\tfrac{1}{2}p^2) = \exp(\mathrm{i}\alpha H^{\mathrm{at}})\Omega_{\pm}^{\mathrm{at}}$. Suppose now that H^{at} has bound states and that at $t = 0$ the state vector of the system is equal to some bound state ϕ, for instance the ground state, of H^{at}. Then, at time t, $|\psi(t)\rangle = U(t, 0, y)|\phi\rangle$ (in this section we use Dirac notation). The probability of finding the electron with momentum $\boldsymbol{p}$ in $\Delta \subset \mathbb{R}^3$ at time t is (we suppress y for brevity)

$$\begin{aligned} w_\Delta(t, y) &= \langle\psi(t)|\chi_\Delta(\boldsymbol{p})|\psi(t)\rangle \\ &= \langle\phi|U(t,0)^*\chi_\Delta(\boldsymbol{p})U(t,0)|\phi\rangle \\ &= \langle\phi|\exp(\mathrm{i}M)\hat{U}(t,0)^*\exp(-\mathrm{i}M(t))\chi_\Delta(\boldsymbol{p}) \\ &\quad \times \exp(\mathrm{i}M(t))\hat{U}(t,0)\exp(-\mathrm{i}M)|\phi\rangle \\ &= \langle\phi|\exp(\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{Z})\hat{U}(t,0)^*\chi_\Delta(\boldsymbol{p})\hat{U}(t,0)\exp(-\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{Z})|\phi\rangle \\ &= \langle\phi|\exp(\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{Z})\hat{U}(t,0)^*\hat{U}_{\mathrm{as}}(t,0)\chi_\Delta(\boldsymbol{p})\hat{U}_{\mathrm{as}}(t,0)^*\hat{U}(t,0)\exp(-\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{Z})|\phi\rangle. \end{aligned} \tag{4.2}$$

Here $\chi_\Delta(\boldsymbol{p})$ is the characteristic function on Δ, which equals one for $\boldsymbol{p} \in \Delta$ and vanishes otherwise. $\hat{U}_{\mathrm{as}}(t, 0) = U_0(t, 0)\exp(-\mathrm{i}\lambda \ln|t|/p)$ and we used the fact that $\exp(-\mathrm{i}M(t))$

and $U_{as}(t,0)$ commute with $\chi_\Delta(\boldsymbol{p})$ and that the contributions $\exp(\mp i\psi)$ in $\exp(\pm iM)$ cancel. For V as above the wave operators

$$\Omega_\pm(y) = \lim_{t\to\pm\infty} \hat{U}(t,0,y)^* \hat{U}_{as}(t,0,y) \tag{4.3}$$

exist as strong limits (on $\mathscr{H}$). The additional factor $\exp(-i\lambda \ln|t|/p)$ for $\lambda \neq 0$ is the usual Dollard correction (Dollard 1964), necessary for potentials with Coulomb tails. We shall moreover assume that $\Omega_\pm(y)$ are unitary. This implies asymptotic completeness and

$$\Omega_\pm(y)^* = \lim_{t\to\pm\infty} \hat{U}_{as}(t,0,y)\hat{U}(t,0,y), \tag{4.4}$$

again in the strong sense on $\mathscr{H}$. We are not aware of a mathematical proof of this property although one intuitively expects that an atom in a spatially homogeneous radiation field of the type considered will eventually ionise as time proceeds. Under the above assumptions (4.2) has the limit

$$w_\Delta(y) = \langle\phi|\exp(i\boldsymbol{p}\cdot\boldsymbol{Z})\Omega_+\chi_\Delta(\boldsymbol{p})\Omega_+^* \exp(-i\boldsymbol{p}\cdot\boldsymbol{Z})|\phi\rangle. \tag{4.5}$$

As mentioned before we consider the case that the initial phases of the fields (i.e. at $t=0$) are randomly distributed from one measurement to another so that it makes sense to consider

$$\begin{aligned} w_\Delta &= (2\pi)^{-N}\int \mathrm{d}y\, w_\Delta(y) \\ &= \langle\langle \mathbf{0}\phi|\exp(i\boldsymbol{p}\cdot\boldsymbol{Z}(y))\Omega_+(y)\chi_\Delta(\boldsymbol{p})\Omega_+(y)^*\exp(-i\boldsymbol{p}\cdot\boldsymbol{Z}(y))|\mathbf{0}\phi\rangle\rangle, \end{aligned} \tag{4.6}$$

the average of $w_\Delta(y)$ over the initial phases. $\langle\langle u|v\rangle\rangle$ denotes the inner product in $\mathscr{K}$, again in Dirac notation. For $|f\rangle\in\mathscr{H}_0$ and $|g\rangle\in\mathscr{H}$, $|fg\rangle\rangle = |f\rangle\otimes|g\rangle$ and on $\mathscr{H}_0$ $\langle y|\boldsymbol{n}\rangle = (2\pi)^{-N/2}\exp(i\boldsymbol{n}\cdot y)$ is the eigenfunction of $\dot{T}$ with eigenvalue $\boldsymbol{\omega}\cdot\boldsymbol{n}$, $\boldsymbol{n}\in\mathbb{Z}^N$. For $F(y)$ a bounded operator on $\mathscr{H}$, measurable and with $\|F(y)\|_{\mathscr{H}}\in L^\infty(\mathbb{R}^N, \mathrm{d}y)$

$$\begin{aligned} \langle\langle \mathbf{0}g|F(y)|\mathbf{0}h\rangle\rangle &= \int \mathrm{d}y\langle\mathbf{0}|y\rangle\langle g|F(y)|h\rangle\langle y|\mathbf{0}\rangle \\ &= (2\pi)^{-N}\int \mathrm{d}y\langle g|F(y)|h\rangle. \end{aligned} \tag{4.7}$$

In this sense $w_\Delta(y)$ is a quantity defined on a fibre in a direct integral decomposition of $\mathscr{K}$, $\mathscr{K} = \int^\oplus \mathscr{H}\,\mathrm{d}y$, with the special property of being continuous in y ($\Omega_\pm(y)$ and $\Omega_\pm^*(y)$ are continuous in y in the strong sense). In the same way as discussed by Howland (1974) it follows that on $\mathscr{K}$ in the strong sense

$$\begin{aligned} \Lambda_\pm &= \lim_{t\to\pm\infty} \exp(i\hat{K}t)\exp[-i(\hat{K}_0 t + \lambda \ln|t|/p)] \\ &= \lim_{t\to\pm\infty} \exp(i\check{K}t)\exp[-i(\check{K}_0 t + \lambda \ln|t|/p)], \end{aligned} \tag{4.8}$$

which fact is also easily proved directly. The last line in (4.8) follows from the commutativity of C and the remaining parts of $\hat{K}$ and $\hat{K}_0$. The assumed unitarity of $\Omega_\pm(y)$ on $\mathscr{H}$ implies the unitarity of $\Lambda_\pm$ on $\mathscr{K}$. We can express the relation between $\Lambda_\pm$ and $\Omega_\pm(y)$ by means of the direct integral

$$\Lambda_\pm = \int^\oplus \mathrm{d}y\,\Omega_\pm(y), \tag{4.9}$$

and instead of (4.6) we can write

$$w_\Delta = \langle\langle \mathbf{0}\phi | \exp(\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{Z})\Lambda_+\chi_\Delta(\boldsymbol{p})\Lambda_+^* \exp(-\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{Z})|\mathbf{0}\phi\rangle\rangle$$
$$= \sum_{\boldsymbol{n}\in\boldsymbol{Z}^\wedge} \int_\Delta \mathrm{d}\boldsymbol{k}\, |f(\mathbf{0}\phi \to \boldsymbol{n}\boldsymbol{k})|^2, \tag{4.10}$$

where

$$f(\mathbf{0}\phi \to \boldsymbol{n}\boldsymbol{k}) = \langle\langle \boldsymbol{n}\boldsymbol{k}|\Lambda_+^* \exp(-\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{Z})|\mathbf{0}\phi\rangle\rangle, \tag{4.11}$$

which has the intuitive interpretation as the amplitude for ionisation with final momentum $\boldsymbol{k}$ after the absorption of n_1 photons from mode 1, n_2 from mode 2, etc. f is a square integrable function of $\boldsymbol{k}$ but does not necessarily exist pointwise.

Before proceeding we note that $\check{K}_0$ entering in (4.8) has a purely absolutely continuous spectrum. This can be seen from the fact that $\frac{1}{2}\boldsymbol{p}^2$ has a purely absolutely continuous spectrum with associated spectral measure $\chi_S(\frac{1}{2}p^2)$, S being a Borel set on the real line (we work in momentum space) and that the corresponding spectral measure for $\check{K}_0 = \boldsymbol{\omega}\cdot\boldsymbol{q} + \frac{1}{2}\boldsymbol{p}^2$ *is* $F(S) = \Sigma_{\boldsymbol{n}\in\boldsymbol{Z}^N} P_{\boldsymbol{n}}\chi_S(\frac{1}{2}\boldsymbol{p}^2 + \boldsymbol{n}\cdot\boldsymbol{\omega})$ (Foguel 1957, § 6) which is easily seen to be absolutely continuous. The intertwining relations $\Lambda_\pm \exp(\mathrm{i}\alpha K_0) = \exp(\mathrm{i}\alpha K)\Lambda_\pm$ and the unitarity of $\Lambda_\pm$ then imply the absolute continuity of the spectrum of $\check{K}$.

For $|f\rangle\rangle \in \mathscr{D}_0$ and in particular for $|f\rangle\rangle = \exp(-\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{Z})|\mathbf{0}\phi\rangle\rangle$ we have for short range V (i.e. $\lambda = 0$)

$$\Lambda_+^*|f\rangle\rangle = \left(1 - \mathrm{i}\int_0^\infty \mathrm{d}t \exp(\mathrm{i}\check{K}_0 t)\, V(\boldsymbol{x} - \boldsymbol{Z}(y)) \exp(-\mathrm{i}\check{K}t)\right)|f\rangle\rangle$$
$$= \left(1 - \mathrm{i}\lim_{\varepsilon\downarrow 0}\int_0^\infty \mathrm{d}t \exp[(-\varepsilon + \mathrm{i}\check{K}_0)t]\, V(\boldsymbol{x} - \boldsymbol{Z}(y)) \exp(-\mathrm{i}\check{K}t)\right)|f\rangle\rangle. \tag{4.12}$$

Then, formally,

$$f(\mathbf{0}\phi \to \boldsymbol{n}\cdot\boldsymbol{k}) = \delta_{\boldsymbol{n}\mathbf{0}}\langle \boldsymbol{k}|\phi\rangle_{\mathscr{H}} - \mathrm{i}\lim_{\varepsilon\downarrow 0}\int_0^\infty \mathrm{d}t \langle\langle \boldsymbol{n}\boldsymbol{k}| V(\boldsymbol{x} - \boldsymbol{Z}(y))$$
$$\times \exp[\mathrm{i}(\boldsymbol{n}\cdot\boldsymbol{\omega} + \tfrac{1}{2}k^2 + \mathrm{i}\varepsilon - \check{K})t] \exp(-\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{Z})|\mathbf{0}\phi\rangle\rangle$$
$$= \delta_{\boldsymbol{n}\mathbf{0}}\langle \boldsymbol{k}|\phi\rangle_{\mathscr{H}}$$
$$+ \lim_{\varepsilon\downarrow 0}\langle\langle \boldsymbol{n}\boldsymbol{k}|\, V(\boldsymbol{x} - \boldsymbol{Z}(y))(\boldsymbol{n}\cdot\boldsymbol{\omega} + \tfrac{1}{2}k^2 + \mathrm{i}\varepsilon - \check{K})^{-1} \exp(-\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{Z})|\mathbf{0}\phi\rangle\rangle. \tag{4.13}$$

This expression is similar to a result obtained earlier (Muller and Tip 1984) for the special case of a single circularly polarised field mode. With some more handwaving in the direction of mathematical rigour it is possible to obtain a similar result for the case $\lambda \neq 0$. Then $(|\boldsymbol{k}\rangle^{\mathrm{at}} = \Omega_+^{\mathrm{at}}|\boldsymbol{k}\rangle$ is an atomic continuum eigenstate)

$$f(\mathbf{0}\phi \to \boldsymbol{n}\boldsymbol{k}) = \delta_{\boldsymbol{n}\mathbf{0}}\langle \boldsymbol{k}^{\mathrm{at}}|\phi\rangle + \lim_{\varepsilon\downarrow 0}\langle\langle \boldsymbol{n}\boldsymbol{k}^{\mathrm{at}}|[\, V(\boldsymbol{x} - \boldsymbol{Z}(y)) - V(\boldsymbol{x})]$$
$$\times (\boldsymbol{n}\cdot\boldsymbol{\omega} + \tfrac{1}{2}k^2 + \mathrm{i}\varepsilon - \check{K})^{-1} \exp(-\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{Z})|\mathbf{0}\phi\rangle\rangle, \tag{4.14}$$

which is a form of the 'two-potential' formula. It also holds for short range V ($\lambda = 0$).

As indicated a number of assumptions have not been rigorously justified. The first, and physically most interesting, is the unitarity of the wave operators. The others are the transition from the expressions involving wave operators to those

containing the resolvent of $\check{K}$ and the existence of the limits $\varepsilon \downarrow 0$ in (4.13) and (4.14). We expect that the latter can be handled by an extension of the limiting absorption principle to quantities on $\mathscr{H}$ since $V(\boldsymbol{x}-\boldsymbol{Z})$ (or $V(\boldsymbol{x}-\boldsymbol{Z})-V(\boldsymbol{x})$) and ϕ have decay properties for large $|\boldsymbol{x}|$.

Instead of (4.13) we can write

$$f(\mathbf{0}\phi \to \boldsymbol{nk}) = \delta_{\boldsymbol{n}\mathbf{0}}\langle \boldsymbol{k} | \phi\rangle + \lim_{\varepsilon \downarrow 0} \langle\langle \boldsymbol{nk}| \exp(-\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{Z})\, V(\boldsymbol{x}) \cdot (\boldsymbol{n}\cdot\boldsymbol{\omega} + \tfrac{1}{2}k^2 + \tfrac{1}{2}\boldsymbol{A}(y)^2 + \mathrm{i}\varepsilon - K)^{-1} |\mathbf{0}\phi\rangle\rangle \tag{4.15}$$

by performing a unitary transformation. The advantage is that, for dilatation analytic $V(\boldsymbol{x})$, K has dilatation analytic properties in the single mode case (Graffi *et al* 1984, Graffi 1984, Howland 1983).

In the multi-mode case with incommensurate ω_j there is a dense set of thresholds (for $\check{K}_0$ the points $\boldsymbol{n}\cdot\boldsymbol{\omega}$, $\boldsymbol{n}\in\mathbb{Z}^N$) so that dilatation theory becomes problematic. The reason is the fact that T is not bounded from below. If we truncate T so that its truncated form is bounded from below the situation improves. This is physically quite reasonable since it means that high-order processes involving the absorption of large numbers of photons are neglected which is often justified. In the case where all ω_j are commensurate the thresholds become evenly spaced but the fact that they are infinitely degenerate still prevents the development of a dilatation theory (no compactness properties) so that here again a truncation is necessary. Equation (4.14) can be transformed in a similar way and the same remarks apply. After truncation a dilatation theory can be developed in both cases and by making a Feshbach decomposition as in Muller and Tip (1984) an approximate description of the energy spectrum of the photoionised electrons can be given along the same lines.

We end this section by noting that in two special cases truncation is not needed. The first is that of a fundamental frequency with correlated harmonics where only a single pair $\{y, q\}$ appears and the second is that where all mode frequencies coincide. Then a transformation can be made so that only a single q_j appears in K. In both cases we are essentially back to the single-mode situation.

5. Discussion

In the present paper we have given a general framework for one-electron atoms in a spatially homogeneous multi-mode radiation field. We showed that by introducing a larger Hilbert space the time evolution can be given in terms of a constant generator K and we found that K can be defined perturbatively, the atomic potential $V(\boldsymbol{x})$ being the perturbation. We obtained an expression for photoionisation amplitudes under the plausible assumption of unitarity of the relevant wave operator. An actual proof of this property seems to be an open problem which is quite interesting from a physical point of view. Disregarding some mathematical subtleties we expressed these amplitudes in terms of the resolvent of K. In an earlier investigation, concerning a single circularly polarised radiation mode, we started from a similar expression for a description of the energy spectrum of the photoionised electron. The same can be done for a single field mode in the present case but in the multi-mode situation the procedure breaks down due to problems with dilatation analyticity and analytic continuation. The reason is the lack of boundedness from below of the spectrum of K (which is typical for semiclassical theories of the type discussed here). In fact this problem is

absent in a second quantised version. Such a theory was developed by Grossmann and Tip (1980) for hydrogen in a single quantised field mode, where dilatation analyticity of the corresponding Hamiltonian was established. This formalism is easily extended to a finite number of field modes and a more general dilatation analytic potential $V(\boldsymbol{x})$. The main difference with the present formalism is that instead of T we then encounter a sum of harmonic oscillator Hamiltonians and we have boundedness from below. Since the semiclassical formalism is usually thought to be a limiting form of the second quantised one, valid in the case of initial field states containing many photons and for processes where only relatively few photons are absorbed this raises the question whether a careful consideration of the limiting process can lead to a description with a cut-off in the spectrum of T, restoring the semi-boundedness. The advantage of the semiclassical approach, on the other hand, is that a direct numerical integration of the original Schrödinger equation (with time-dependent Hamiltonian) can be performed.

Another model that is often used is that of fields that are switched off for large $|t|$, typically

$$\boldsymbol{A}(t) = \boldsymbol{A} \exp(-\gamma t^2) \cos \omega t. \tag{5.1}$$

Such fields, and in fact even more general ones, including fields depending both on $\boldsymbol{x}$ and t, were considered by Gesztesy *et al* (1985). These authors proved unitarity for the wave operators associated with the full Hamiltonian and the atomic Hamiltonian. Their situation is, however, different from the one considered in the present paper, since in their case the field is asymptotically absent. For fields of the type (5.1) the unitarity proof is rather simple due to $\exp(-\gamma t^2)$ which can be used to advantage in Cook-type estimates. One might ask the question why sinusoidal fields are considered at all instead of (5.1). The reason is that the additional symmetry due to the periodicity gives rise to a simpler spectral structure. Also experimental circumstances are usually such that the field can be assumed to be sinusoidal during the period of time that the actual ionisation process takes place. There are, however, subtle effects, related to a rise in the ionisation potential in combination with a ponderomotive energy gain by the electron leaving the field region (for a general discussion see, for instance, Tip (1984)).

By replacing (5.1) by $\exp[-\gamma(t+y_1)^2]\cos(\omega t + y_2)$ we can again introduce a generator K on a larger space $\mathcal{K}$ where now y_1 in principle ranges from $-\infty$ to $+\infty$. It would then be interesting to consider the limit where γ becomes small in connection with adiabatic behaviour.

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