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Scattering theory in a time-dependent external field
I. General theory

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Abstract. A general framework is set up for the description of quantum-mechanical scattering processes for the case that both the free hamiltonian $H_0(t) = H_0 + H_1(t)$ and the full hamiltonian $H(t) = H_0(t) + V$ are time-dependent. In particular the existence of the various evolution operators is considered and the wave operators $\Omega_{\pm}(s)$, which now depend on the initial time s, are defined. The consequences of a periodic time dependence are studied and for $H_1(t)$ of the type $H_1(t) = H_1 \cos(\omega t + \delta)$ it is shown that for $|\omega| \to \infty$ the frequency-dependent wave operators converge to the wave operators which pertain to the case that $H_1(t)$ is absent.

1. Introduction

Since the advent of masers and lasers intense electric fields have become available as a tool for investigations in atomic and molecular physics. A topic that has attracted much attention during the past few years is, for instance, the system consisting of an atom or molecule placed in an intense field, which is probed by a second, weak, radiation field. In this way information is gained about the changes that are induced by the strong field in the atom, the so called dressing of the atom by the field (Cohen-Tannoudji and Haroche 1969). It is therefore somewhat remarkable that hardly any attention has been paid to the analogous case, where the weak field as a probe is replaced by a second particle (or a particle beam) which is brought into collision with the original atom or molecule and where the details of the ensuing scattering process are studied (see, however, the theoretical investigations of Hahn and Hertel 1972).

Apart from the interest in such processes from the point of view of scattering theory itself, two further reasons can be put forward for investigating atomic scattering in a radiation field.

(i) In a gas laser two-particle collisions are paramount for the actual performance of this device. It is therefore important to have some insight into the possible modifications of these processes, brought about by the radiation field.

(ii) In order to construct a kinetic theory for neutral gases subject to a strong radiation field, some knowledge is needed about the scattering of two (or more) particles in the presence of the field. For instance, within the formalism of the so called cluster expansion method (Dorfman and Cohen 1967), it is important to know that the various streaming operators possess a long time limit. This is directly related to the existence problem of the Møller wave operators in the presence of the external field.

In the present work we will investigate for suitable models some basic features of scattering theory, such as the existence of Møller wave operators and properties of the S operator in the presence of an external field. In fact our approach is semi-classical in that we consider a quantum system in the presence of an external classical field. The latter enters the formalism through a time-dependent contribution to the hamiltonian of the system.

As is customary in this area we will use a model for an atom or molecule, which consists of a point particle with internal structure (such as the rigid rotator model of a diatomic molecule). Thus our particles may possess a finite or countably infinite number of internal energy levels, so that for a particle j the one-particle hamiltonian has the form

$$H_j = K_j^{\mathrm{tr}} + K_j^{\mathrm{int}}, \tag{1.1}$$

where $K_j^{\mathrm{tr}} = \hbar k_j^2/(2m_j)$ is the translational part of the hamiltonian, whereas K_j^{int} is its internal part, which can be represented by the matrix

$$\begin{pmatrix} \omega_1 & & 0 \\ & \omega_2 & \\ 0 & & \ddots \end{pmatrix} = \{\omega_k \delta_{kl}\} \tag{1.2}$$

(we use reduced units with respect to $\hbar$).

In actual cases only a few levels ω_k play a role during radiative processes, the others being strongly 'off-resonant'.

The interaction between atoms i and j is assumed to be given by the potential 'matrix'

$$V^{(i,j)} = [V_{kl}^{(i,j)}(\boldsymbol{x}_i - \boldsymbol{x}_j)] \tag{1.3}$$

where $\boldsymbol{x}_i$ and $\boldsymbol{x}_j$ are the position vectors of particles i and j, respectively. Since we consider neutral particles an external field acts directly on the internal degrees of freedom only.

However, in case the field is spatially inhomogeneous there will be an influence on the translational motion as well. As an example we can consider the case of an external electric field $\boldsymbol{E}(\boldsymbol{x}, t)$. Then, in the dipole approximation, the one-particle hamiltonian for particle j is

$$H_j(t) = H_j + H_j^{\mathrm{ext}}(t), \tag{1.4}$$

with

$$H_j^{\mathrm{ext}}(t) = -\boldsymbol{\mu}_j \cdot \boldsymbol{E}(\boldsymbol{x}_j, t), \tag{1.5}$$

where $\boldsymbol{\mu}_j$, the dipole-moment operator, can be represented as the matrix

$$\boldsymbol{\mu}_j = \{\boldsymbol{\mu}_{j,kl}\}. \tag{1.6}$$

Although $\boldsymbol{\mu}_j$ does not contain variables pertaining to the translational motion, this may be different for (1.5) as a whole, due to the $\boldsymbol{x}_j$ dependence of $\boldsymbol{E}(\boldsymbol{x}_j, t)$.

It should be clear from the model sketched above that ionization or dissociation of the constituent particles of the system is outside the scope of the present investigation. In fact, by excluding this possibility, we are able to treat a collision between particles 1 and 2 as a potential scattering problem, the 'free' hamiltonian being

$$H_0(t) = H_1(t) + H_2(t), \tag{1.7}$$

whereas the full hamiltonian is given by

$$H(t) = H_0(t) + V^{(1,2)} \tag{1.8}$$

Note that, although break-up processes of particles 1 and 2 are not considered, these two particles may form bound states in the potential $V^{(1,2)}$, just as in ordinary potential scattering.

In order to get some feeling as to how the time dependence and spatial inhomogeneity of the external field manifest themselves during a collision process, consider the case of two particles colliding with each other at thermal velocities (say 300 m s^{-1}) in an external field in the optical region (say $\lambda = 6000$ Å wavelength). In this case the field frequency is $\nu = 0{\cdot}5 \times 10^{15}$ s^{-1}, whereas for a potential with a range of a few ångströms the average duration of a collision is in the order of 10^{-12} s so that during a collision the field oscillates rapidly. On the other hand, since the collision process effectively takes place in a spatial region of say 10 Å radius, we see that usually no large error is introduced by evaluating the external field at the centre-of-mass coordinate $\boldsymbol{X}$ rather than the coordinates $\boldsymbol{x}_1$ and $\boldsymbol{x}_2$ (long wavelength approximation).

Furthermore we can distinguish two different physical situations. The first is that of the scattering of two particles in a spatially homogeneous field, so that in the asymptotic regions the particles are still influenced by the field. This is what happens in a gas laser (although the field is not homogeneous in that case). On the other hand, one can perform scattering experiments where a beam of particles is directed into a collision chamber containing a gas of target particles as well as a radiation field, the latter being strictly confined to the chamber. Then the field is spatially localized and asymptotically the particles become free, ie they leave the field region.

The organization of the present work is as follows. In § 2 we consider the existence problem of the time-evolution operators for the case of a time-dependent hamiltonian. Here we make use of a number of results obtained previously by Kato (1953). In § 3 we define the wave operators for the case at hand and derive some general properties such as intertwining relations.

Since the system is not homogeneous in time it is not surprising that the wave operators are found to depend on the zero of the time axis (ie on the phase of the external field at $t = 0$ for sinusoidal fields). We close § 3 with a simple example for which the existence of the wave operators can be proven explicitly. Section 4 deals with the case of external fields with periodic time dependence. Under these circumstances a Hilbert space version of the usual Floquet theory leads to an expression for the wave operators as the limit of an infinite sequence which shows some resemblance to the field-free case. We also show for external fields with sinusoidal time dependence that under certain mild restrictions the wave operators converge strongly towards the wave operators pertaining to the field-free case if the field frequency tends to infinity.

In the second paper (Prugovečki and Tip 1974, to be referred to as II) we prove the existence of the wave operators for the model introduced earlier in this section. This result is obtained under some suitable conditions on the potential (1.3) and the field terms (1.5).

2. Time-evolution operators for non-conservative systems

The class of models mentioned in the introduction provides examples of systems which do not conserve energy, since, due to the presence of an external field, energy can be exchanged with the environment. Mathematically, this feature is reflected in the fact that the hamiltonian $\hat{H}(t)$ of the system is time dependent. Dealing with a family $\{\hat{H}(t)\}$ of, in general, unbounded operators depending on a parameter $t \in \mathscr{R}$ poses mathematical problems which are absent in the special case when $\hat{H}(t)$ is equal to a fixed operator $\hat{H}$ for all values of t. In order to arrive at a mathematically manageable theory we shall impose certain restrictions on $\hat{H}(t)$ by specifying in greater detail the manner in which $\hat{H}(t)$ depends on $t \in \mathscr{R}$. These restrictions are sufficiently generous to include a large class of models which are of physical interest. Let $\mathscr{H}$ be a separable complex Hilbert space with inner product $\langle \ldots | \ldots \rangle$, which is antilinear in the left variable and linear in the right one and with vector norm $\|\ldots\| = \langle \ldots | \ldots \rangle^{1/2}$. We assume that $\hat{H}(t)$ is of the form

$$\hat{H}(t) = \hat{H}_\alpha + H_\beta(t) \tag{2.1}$$

where H_α and $H_\beta(t)$ are operators with the following properties:

(i) H_α is self-adjoint with domain $\mathscr{D}$, dense in $\mathscr{H}$.

(ii) (*a*) $H_\beta(t)$ is self-adjoint and bounded for all $t \in \mathscr{R}$.

(*b*) $H_\beta(t)$ is of bounded variation on any finite interval $[a, b]$ in $\mathscr{R}$, ie there is a constant C_1 such that for any subdivision $a = t_0 < t_1 < \ldots < t_n = b$

$$\sum_{k=1}^{n} \|H_\beta(t_k) - H_\beta(t_{k-1})\| \leqslant C_1, \tag{2.2}$$

where $\|\ldots\|$ denotes the operator bound.

(*c*) $H_\beta(t)$ is uniformly bounded on $\mathscr{R}$, ie $\|H_\beta(t)\| \leqslant C_2$ for all $t \in \mathscr{R}$ and some positive constant C_2.

(iii) $H_\beta(t)$ is strongly continuous in t for all $t \in \mathscr{R}$.

Let us introduce the auxiliary operators

$$\begin{aligned} A_1(t) &= -\mathrm{i}\hat{H}(t), & A_2(t) &= \mathrm{i}\hat{H}(t), \\ A_3(t) &= -\mathrm{i}\hat{H}(-t), & A_4(t) &= \mathrm{i}\hat{H}(-t). \end{aligned} \tag{2.3}$$

Since $H_\beta(t)$ is bounded it follows that the domains of $A_j(t), j = 1, \ldots, 4$, coincide with $\mathscr{D}$.

It is a routine matter to verify that if the above conditions (i) and (ii) are satisfied, then so are the conditions C_1 and C_2 of Kato (1953). Furthermore, if condition (iii) is satisfied as well then condition C_3 of Kato is satisfied. Hence we can restate theorems 2 and 3 by Kato (1953) in the following form which is more convenient for our purposes.

Lemma 2.1. Let the above conditions (i) and (ii) hold. Then there exist evolution operators $U_j(t, s), j = 1, \ldots, 4$, for all real $t \geqslant s$ with the following properties:

(1) $U_j(t, s)$ are bounded and $\|U_j(t, s)\| \leqslant 1$.

(2) Each $U_j(t, s), j = 1, \ldots, 4$ is strongly continuous in s and t simultaneously.

(3) $U_j(t, t) = I$ and $U_j(t, s) = U_j(t, r)U_j(r, s)$ for all $t \geqslant r \geqslant s$.

(4) For any $x \in \mathscr{D}$ the following limits exist:

$$\begin{aligned} A_j^{(+)}(t) &= \operatorname*{s-lim}_{\epsilon \to +0} \epsilon^{-1}(U_j(t+\epsilon, t) - I)x, \\ A_j^{(-)}(t) &= \operatorname*{s-lim}_{\epsilon \to +0} \epsilon^{-1}(U_j(t, t-\epsilon) - I)x, \end{aligned} \tag{2.4}$$

and $A_j^{(\pm)}(t)$ coincide with $A_j(t), j = 1, \ldots, 4$, at all $t \in \mathscr{R}$ with the possible exception of a denumerable set of values.

(5) $U_j(t,s)\mathscr{D} \subset \mathscr{D}$ and for $x \in \mathscr{D}$ the vector-valued function $x(t) = U_j(t,s)x$ has a strong right derivative $D^{(+)}x(t)$ which is equal to $A_j^{(+)}(t)x(t)$.

(6) Each $U_j(t,s), j = 1, \ldots, 4$ is uniquely determined by the above properties (1)–(5).

Lemma 2.2. If the conditions (i) and (ii) are satisfied, then $U_j(t,s), j = 1, \ldots, 4$, are unitary, their adjoints $U_j^*(t,s)$ are strongly continuous in t and s simultaneously and the following relations are satisfied for all $t \geqslant s$:

$$U_1^*(t,s) = U_3(-s,-t), \qquad U_2^*(t,s) = U_4(-s,-t), \tag{2.5}$$

$$[A_{1,2}^{(+)}(t)]^* = A_{3,4}^{(-)}(-t), \qquad [A_{1,2}^{(-)}(t)]^* = A_{3,4}^{(+)}(-t). \tag{2.6}$$

Proof. According to Kato (1953)

$$U_j(t,s) = \underset{\epsilon\to+0}{s\text{-lim}} \prod_{k=1}^{n} \exp[(t_k - t_{k-1})A_j(t_k')], \tag{2.7}$$

where $s = t_0 < t_1, \ldots, < t_n = t$, $t_{k-1} \leqslant t_k' \leqslant t_k$ and $\epsilon = \max|t_k - t_{k-1}|$.

Hence $U_j(t,s)$ is the strong limit of a sequence of unitary operators and consequently it is isometric on $\mathscr{H}$, ie $U_j^*(t,s)U_j(t,s) = I$. To establish that, for example, $U_1(t,s)$ is actually unitary, note that for any $x, y \in \mathscr{H}$

$$\begin{aligned}
&\langle x|U_1(t,s)y\rangle \\
&\quad = \lim_{\epsilon\to+0}\left\langle x \,\middle|\, \prod_{k=1}^{n} \exp[-\mathrm{i}(t_k - t_{k-1})\hat{H}(t_k')]y\right\rangle \\
&\quad = \lim_{\epsilon\to+0}\left\langle \prod_{k=n}^{l} \exp[\mathrm{i}(t_k - t_{k-1})\hat{H}(t_k')]x \,\middle|\, y\right\rangle \\
&\quad = \lim_{\epsilon\to+0}\left\langle \prod_{k=n}^{l} \exp\{-\mathrm{i}[(-t_k)-(-t_{k-1})]\hat{H}[-(-t_k')]\}x \,\middle|\, y\right\rangle \\
&\quad = \langle U_3(-s,-t)x|y\rangle.
\end{aligned}$$

Thus $U_1^*(t,s) = U_3(-s,-t)$ and consequently $U_1^*(t,s)$ is isometric on $\mathscr{H}$ by the preceding result for $U_3(-s,-t)$. This shows that $U_j(t,s)$ are unitary for $j = 1, 3$; the same result for $j = 2, 4$ can be obtained in a similar manner. By letting $\epsilon \to +0$ in the equality

$$\langle \epsilon^{-1}[U_{1,2}(t+\epsilon,t) - I]x|y\rangle = \langle x|\epsilon^{-1}[U_{3,4}(-t,-t-\epsilon) - I]y\rangle$$

with $x, y \in \mathscr{D}$ we obtain

$$\langle A_{1,2}^{(+)}(t)x|y\rangle = \langle x|A_{3,4}^{(-)}(-t)y\rangle$$

and consequently (2.6) is true. The strong continuity of $U_j^*(t,s)$ in t and s follows from (2.5) and lemma 2.1.

Lemma 2.3. If the conditions (i), (ii) and (iii) are satisfied then $U_j(t,s)x$ and $U_j^*(t,s)x$, $j = 1, \ldots, 4$, have strong partial derivatives in t for any $x \in \mathscr{D}$. These derivatives are strongly continuous in the variable $t \geqslant s$ and satisfy the relations:

$$\partial_t U_j(t,s)x = A_j(t)U_j(t,s)x, \tag{2.8}$$

$$\partial_t U_j^*(t,s)x = U_j^*(t,s)A_j^*(t)x. \tag{2.9}$$

Proof. The above statements pertaining to $\partial_t U_j(t, s)$ follow from theorem 3 of Kato (1953). To derive the analogous results for $U_j^*(t, s)$ take $x, y \in \mathscr{D}$ and note that

$$\lim_{\epsilon \to +0} \langle \epsilon^{-1}[U_j^*(t+\epsilon, s) - U_j^*(t, s)]x|y\rangle$$
$$= \lim_{\epsilon \to +0} \langle x|\epsilon^{-1}[U_j(t+\epsilon, s) - U_j(t, s)]y\rangle$$
$$= \langle x|A_j(t)U_j(t, s)y\rangle = \langle U_j^*(t, s)A_j^*(t)x|y\rangle. \tag{2.10}$$

Furthermore, we get by using point 3 in lemma 2.1 and (2.5) that

$$\|\epsilon^{-1}[U_j^*(t+\epsilon, s) - U_j^*(t, s)]x\|^2$$
$$= \epsilon^{-2}\{2\|x\|^2 - 2\,\mathrm{Re}\langle x|U_j(t+\epsilon, t)x\rangle\}$$
$$= \|\epsilon^{-1}[U_j(t+\epsilon, t) - I]x\|^2.$$

The right-hand side of the above relation converges towards

$$\|A_j(t)x\|^2 = \|U_j^*(t, s)A_j^*(t)x\|^2,$$

where the equality is a consequence of the fact that $A_j^*(t) = -A_j(t)$. From this result and (2.10) we infer by means of standard algebraic manipulations (see, for instance, Prugovečki 1971, p 334) that the corresponding strong limit exists and satisfies (2.9).

The strong continuity of $\partial_t U_j^*(t, s)$ in t at any $t_0 \geqslant s$ follows from (2.9) and the estimate (the plus sign in $H_\beta(\pm t)$ refers to the case $j = 1, 2$, the minus sign to the case $j = 3, 4$)

$$\|U_j^*(t, s)A_j^*(t)x - U_j^*(t_0, s)A_j^*(t_0)x\|$$
$$\leqslant \|[H_\beta(\pm t) - H_\beta(\pm t_0)]x\| + \|[U_j^*(t, s) - U_j^*(t_0, s)]A_j^*(t_0)x\|$$

as a consequence of the strong continuity in t of $U_j^*(t, s)$ (cf lemma 2.2) and of $H_\beta(t)$ (cf condition (iii)).

We should point out at this stage that the following statement is obviously true.

Lemma 2.4. If $H_\beta(t) = H_\beta^*(t)$ has a strong derivative $\partial_t H_\beta(t)$ that is uniformly bounded on any compact set in $\mathscr{R}$, ie $\|\partial_t H_\beta(t)\| \leqslant C_3(a, b)$ for $t \in [a, b]$ and any $a \leqslant b$, then conditions (ii*b*) and (iii) are satisfied.

The above criterion provides a practically useful and easy method for establishing the validity of the presuppositions (ii*b*) and (iii) for models under consideration. It is convenient to replace in the sequel the two families $\{U_1(t, s)\}$ and $\{U_3(t, s)\}$ by a single equivalent family $\{\hat{U}(t, s)\}$.

Theorem 2.1. If the conditions (i) and (ii) are satisfied the families of operators

$$\hat{U}(t, s) = \begin{cases} U_1(t, s) & \text{for } t \geqslant s \\ U_3(-t, s) & \text{for } t < s \end{cases} \tag{2.11}$$

and $\hat{U}^*(t, s)$ are strongly continuous in $s, t \in \mathscr{R}$. The operators $\tilde{U}(t, s)$ are unitary and have the following properties:

$$\hat{U}(t, s)\mathscr{D} \subset \mathscr{D} \tag{2.12}$$

$$\hat{U}(t, t) = I \tag{2.13}$$

$$\hat{U}^*(t, s) = \hat{U}^{-1}(t, s) = \hat{U}(s, t) \tag{2.14}$$

$$\hat{U}(t, s) = \hat{U}(t, r)\hat{U}(r, s) \tag{2.15}$$

for arbitrary $r, s, t \in \mathscr{R}$.

Proof. The strong continuity properties and (2.12)–(2.13) are immediate consequences of the definition (2.11) and the corresponding properties of $U_1(t, s)$ and $U_3(t, s)$. The relation (2.14) is obtained from (2.5) and the unitarity of $U_{1,3}(t, s)$. Finally if $s \leqslant r \leqslant t$ then (2.15) follows from point 3 in lemma 2.1. If, for example, $r \leqslant t \leqslant s$ then by the same point $\hat{U}(r, s) = \hat{U}(r, t)\hat{U}(t, s)$ and (2.15) is obtained by using (2.14).

Theorem 2.2. If conditions (i), (ii) and (iii) are satisfied then $\hat{U}(t, s)x$ has strong partial derivatives in t and s for each $x \in \mathscr{D}$. These derivatives satisfy the equations

$$\partial_t \hat{U}(t, s)x = -\mathrm{i}\hat{H}(t)\hat{U}(t, s)x, \qquad \partial_t \hat{U}^*(t, s)x = \mathrm{i}\hat{U}^*(t, s)\hat{H}(t)x \tag{2.16}$$

$$\partial_s \hat{U}(t, s)x = \mathrm{i}\hat{U}(t, s)\hat{H}(s)x, \qquad \partial_s \hat{U}^*(t, s)x = -\mathrm{i}\hat{H}(s)\hat{U}^*(t, s)x \tag{2.17}$$

and are strongly continuous in the respective t and s variables. The strongly continuous operator-valued function $\hat{U}(t, s)$ is uniquely determined by the properties (2.12)–(2.16).

Proof. The statements concerning the derivatives in t are immediate consequences of lemma 2.3 applied to $U_1(t, s)$ and $U_3(t, s)$. The corresponding results for the derivatives in s follow from this observation and (2.14), while uniqueness is already contained in lemma 2.1.

As a corollary to the above theorem we can derive the integral equation:

$$\hat{U}(t, s) = \exp[-\mathrm{i}H_\alpha(t-s)] - \mathrm{i}\int_s^t \mathrm{d}u \exp[-\mathrm{i}H_\alpha(t-u)]H_\beta(u)\hat{U}(u, s), \tag{2.18}$$

which provides an iterative method (see also Phillips 1953) for obtaining a convergent perturbation series solution for $\hat{U}(t, s)$. As a matter of fact, if $x \in \mathscr{D}$ then $\hat{U}(t, s)x \in \mathscr{D}$ by (2.12) and we obtain by using (2.16)

$$\partial_u \exp[\mathrm{i}H_\alpha(u-t)]\hat{U}(u, s)x = \exp[\mathrm{i}H_\alpha(u-t)][-\mathrm{i}H_\beta(u)]\hat{U}(u, s)x. \tag{2.19}$$

The right-hand side of the above equation is strongly continuous in u since all the three u dependent factors have this property and in addition two of them have bounds not exceeding one for all $t \in \mathscr{R}$. Hence, the strong Riemann integral over $[s, t]$ of this expression exists. By integrating both sides of (2.19) with respect to u we thus arrive at (2.18).

In closing this section we remark that in practical cases where the external field is suddenly switched on or off so that condition (iii) is not fulfilled the evolution operator $\hat{U}(t, s)$ still has nice continuity properties (cf theorem 2.1). As a second remark we emphasize that $H_\beta(t)$ is assumed to be bounded.

This, however, is a limitation if one wants to consider a real system (instead of a model), such as an H atom subject to an external radiation field. Then the interaction hamiltonian (which plays the role of $H_\beta(t)$) is no longer a bounded operator and consequently problems arise in connection with the domains of definition of various unbounded operators.

3. Definition and properties of wave and scattering operators

In this section we consider two families of time-evolution operators $U_0(t, s)$ and $U(t, s)$ whose corresponding infinitesimal generators $H_0(t)$ and $H(t)$ at each t satisfy the conditions (i), (ii) and (iii) imposed on $\hat{H}(t)$ in the preceding section. Our main interest will be the existence and properties of the wave operators

$$\Omega_\pm(s) = s-\lim_{t\to\pm\infty} U^*(t, s)U_0(t, s). \tag{3.1}$$

Lemma 3.1. Suppose the two strong limits in (3.1) exist for some $s \in \mathscr{R}$. Then $\Omega_\pm(s)$ exist for all $s \in \mathscr{R}$ and have the intertwining properties

$$U(r, s)\Omega_\pm(s) = \Omega_\pm(r)U_0(r, s) \tag{3.2}$$

for all $r, s \in \mathscr{R}$. These intertwining properties will assume the form

$$U(r, s)\Omega_\pm(t) = \Omega_\pm(t)U_0(r, s) \tag{3.3}$$

for all $r, s, t \in \mathscr{R}$ if and only if $\Omega_\pm(s)$ are independent of $s \in \mathscr{R}$.

Proof. From the relation

$$U^*(t, s)U_0(t, s) = U^*(r, s)\{U^*(t, r)U_0(t, r)\}U_0(r, s)$$

obtained by means of (2.15) we infer that

$$\Omega_\pm(s) = U^*(r, s)\left(s-\lim_{t\to\pm\infty} U^*(t, r)U_0(t, r)\right)U_0(r, s)$$

exist if $\Omega_\pm(r)$ exist. Moreover, by (2.14) the above imply (3.2). By reversing the roles of r and s we conclude that the first part of the lemma is true. If $\Omega_\pm(s)$ is independent of s then (3.3) follows at once from (3.2). Conversely, if (3.3) is true then by setting $r = 0$ in (3.2) and using (2.14) we get

$$\Omega_\pm(s) = U^*(0, s)\Omega_\pm(0)U_0(0, s) = U^*(0, s)U(0, s)\Omega_\pm(0) = \Omega_\pm(0),$$

ie $\Omega_\pm(s)$ is independent of s.

The wave operators $\Omega_\pm(s)$ are strong limits of unitary operators and consequently the following result obviously holds.

Theorem 3.1. If the wave operators $\Omega_\pm(s)$ defined in (3.1) exist, then they are partial isometries with initial domain $\mathscr{H}$.

In order to derive a simple criterion for the existence of $\Omega_\pm(s)$ we need the following lemma.

Lemma 3.2. Let $\mathscr{D}_{\text{inv}}$ denote the set of all vectors x in the domain $\mathscr{D}_{H_0(t)}$ of $H_0(t)$ at all $t \in \mathscr{R}$ for which $U_0(t, s)x$ belongs to the domain $\mathscr{D}_{H(t)}$ of $H(t)$ for all $t, s \in \mathscr{R}$. For any $x \in \mathscr{D}_{\text{inv}}$ we have

$$U^*(t, s)U_0(t, s)x = x + i\int_s^t du\, U^*(u, s)[H(u) - H_0(u)]U_0(u, s)x. \tag{3.4}$$

Proof. Since $x \in \mathscr{D}_{H_0(t)}$ and $U_0(t, s)x \in \mathscr{D}_{H(t)}$ when $x \in \mathscr{D}_{\text{inv}}$ we can apply theorem 2.2

and state that the strong derivative $\partial_t U^*(t, s)U_0(t, s)x$ exists and is a strongly continuous function of $t \in \mathscr{R}$. According to (2.16) we have

$$\partial_u U^*(u, s)U_0(u, s)x = iU^*(u, s)[H(u) - H_0(u)]U_0(u, s)x.$$

After integrating both sides of the above equation in the strong Riemann sense with respect to $u \in [s, t]$ we arrive at (3.4). By a straightforward application of the above lemma we obtain the following criterion which will be used in (ii) for establishing the existence of $\Omega_\pm(s)$.

Theorem 3.2. Let $\mathscr{D}_s$ denote the set of all $x \in \mathscr{D}_{inv}$ for which

$$\pm \int_{\pm t_x}^{\pm\infty} \|[H(u) - H_0(u)]U_0(u, s)x\| \, du < \infty \tag{3.5}$$

for some values $t_x \geqslant 0$. If $\mathscr{D}_s$ is dense in $\mathscr{H}$ for some $s \in \mathscr{R}$ then $\Omega_\pm(s)$ in (3.1) exist for that value of s.

The scattering operator can be defined by

$$S(s) = \Omega_+^*(s)\Omega_-(s). \tag{3.6}$$

In general it depends explicitly on the chosen value of $s \in \mathscr{R}$. In fact, according to (3.2) we have

$$S(s) = U_0^*(r, s)S(r)U_0(r, s) \tag{3.7}$$

for any $r, s \in \mathscr{R}$. Physically such a dependence of the wave operators on the instant s where we start comparing the actual motion of the wave packet with the 'unperturbed' case depends very much on the behaviour of the external field from that moment onwards. Thus the transition probabilities will very much depend not only on the manner in which the two particles interact but also on their interaction with the field. The dependence on s can be eliminated only after averaging procedures are carried out. This matter is considered further in II, where specific models are considered.

Let us consider now a special type of $H_0(t)$ and $H(t)$ for which the existence of $\Omega_\pm(s)$ can be established directly, without resorting to theorem 3.2. Thus, take

$$H_0(t) = H^{(0)} + H^{(1)}(t), \qquad H(t) = H_0(t) + V, \tag{3.8}$$

where $H^{(0)}$ and $H^{(0)} + V$ are self-adjoint, while $H^{(1)}(t)$ satisfies in relation to both $H^{(0)}$ and $H^{(0)} + V$ the conditions imposed in § 2 on $H_\beta(t)$. Moreover, we require

$$\int_{-\infty}^{+\infty} dt \, \|H^{(1)}(t)\| < \infty. \tag{3.9}$$

We introduce the auxiliary time-evolution groups

$$\tilde{U}_0(t) = \exp(-iH^{(0)}t), \qquad \tilde{U}(t) = \exp[-i(H^{(0)} + V)t] \tag{3.10}$$

and we assume in addition that the auxiliary wave operators

$$\tilde{\Omega}_\pm = s - \lim_{t \to \pm\infty} \tilde{U}^*(t)\tilde{U}_0(t) \tag{3.11}$$

exist on $\mathscr{H}$.

Lemma 3.3. If (3.9) is satisfied then the uniform limits

$$\Lambda_{\pm}^{(0)}(s) = u-\lim_{t\to\pm\infty} U_0^*(t,s)\tilde{U}_0(t-s), \tag{3.12}$$

$$\Lambda_{\pm}(s) = u-\lim_{t\to\pm\infty} U^*(t,s)\tilde{U}(t-s) \tag{3.13}$$

are unitary operators on $\mathscr{H}$ and have the intertwining properties

$$U_0(s,t)\Lambda_{\pm}^{(0)}(t) = \Lambda_{\pm}^{(0)}(s)\tilde{U}_0(s,t), \tag{3.14}$$

$$U(s,t)\Lambda_{\pm}(t) = \Lambda_{\pm}(s)\tilde{U}(s,t). \tag{3.15}$$

Proof. By applying (2.18) to the case where $H_\alpha = H^{(0)}$ and $H_\beta(t) = H^{(1)}(t)$ we obtain

$$U_0^*(t,s)\tilde{U}_0(t-s) = I + \mathrm{i}\int_s^t \mathrm{d}u\, U_0^*(u,s)H^{(1)}(u)\tilde{U}_0(u-s).$$

Hence we have the uniform estimate

$$\|U_0^*(t_2,s)\tilde{U}_0(t_2-s) - U_0^*(t_1,s)\tilde{U}_0(t_1-s)\| \leqslant \int_{t_1}^{t_2} \mathrm{d}u\, \|H^{(1)}(u)\|,$$

from which the existence of the limits $\Lambda_{\pm}^{(0)}(s)$ in (3.14) follows as a direct consequence of (3.9). Being uniform limits of unitary operators $\Lambda_{\pm}^{(0)}(s)$ are unitary themselves. The intertwining property (3.14) can be derived as in lemma 3.1. The argument for (3.13) and (3.15) proceeds in the same manner.

Theorem 3.3. Suppose that $H_0(t)$ and $H(t)$ are as in (3.8) and that (3.9) is satisfied. Then $\Omega_{\pm}(s)$ in (3.1) exist if and only if $\tilde{\Omega}_{\pm}$ in (3.11) exist, and

$$\Omega_{\pm}(s) = \Lambda_{\pm}(s)\tilde{\Omega}_{\pm}(\Lambda_{\pm}^{(0)}(s))^*. \tag{3.16}$$

Proof. The result follows by lemma 3.3 from the obvious relation

$$U^*(t,s)U_0(t,s) = \{U^*(t,s)\tilde{U}(t-s)\}\{\tilde{U}^*(t-s)\tilde{U}_0(t-s)\}\{\tilde{U}_0^*(t-s)U_0(t,s)\}$$

and the observation that the expressions in the first and last of the above curly brackets have uniform limits and therefore their adjoints also converge uniformly to the adjoints of their respective limits.

4. Periodic time dependence

Let $H_0(t)$ and $H(t)$ be as in § 3. Thus we can write

$$H_0(t) = H^{(0)} + H^{(1)}(t), \qquad H(t) = H + H^{(1)}(t). \tag{4.1}$$

In this section we pursue the consequences of an $H^{(1)}(t)$ that is periodic in time, ie

$$H^{(1)}(t) = H^{(1)}(t+a) \tag{4.2}$$

for some fixed real a. In this case a Hilbert space version of the Floquet theory (see for instance, Ince 1956, p 381, Shirley 1965) can be based upon the following result.

Lemma 4.1. If $H_0(t)$ and $H(t)$ in (4.1) satisfy conditions (i), (ii) and (iii) of § 2 and if (4.2) is also satisfied then

$$U_0(t,s) = U_0(t+a, s+a), \qquad U(t,s) = U(t+a, s+a) \tag{4.3}$$

for all $s, t \in \mathscr{R}$ and

$$\Omega_\pm(s) = \Omega_\pm(s+a), \tag{4.4}$$

provided that the above wave operators defined according to (3.1) do exist.

Proof. The property (4.3) for $U_0(t,s)$ follows from the observation that

$$U_0(t+a, s+a) = U_0'(t,s)$$

satisfies (2.12)–(2.15) as well as the equation

$$\partial_t U_0'(t,s)x = -\mathrm{i}H_0(t+a)U'(t,s)x = -\mathrm{i}H_0(t)U'(t,s)x$$

for any $x \in \mathscr{D}_{H_0(t)}$. Therefore, due to the uniqueness of the time-evolution operator (cf theorem 2.2), $U_0'(t,s) = U_0(t,s)$. Naturally the same argument applies to $U(t,s)$. Then (4.4) follows from (4.3) and the definition of the wave operators.

When $U_0(t,s)$ and $U(t,s)$ have the properties of lemma 4.1 and since both are unitary operator families, we can define the self-adjoint operators $\tilde{H}_0$ and $\tilde{H}$ according to

$$\exp(-\mathrm{i}\tilde{H}_0 a) = U_0(a, 0), \qquad \exp[-\mathrm{i}\tilde{H}a] = U(a, 0). \tag{4.5}$$

Next we introduce the strongly continuous operator groups

$$\tilde{U}_0(t) = \exp(-\mathrm{i}\tilde{H}_0 t), \qquad \tilde{U}(t) = \exp(-\mathrm{i}\tilde{H}t), \qquad t \in \mathscr{R} \tag{4.6}$$

and the unitary operator families $W_0(t)$ and $W(t)$ through

$$W_0(t) = U_0(t, 0)\tilde{U}_0^*(t), \qquad W(t) = U(t, 0)\tilde{U}^*(t). \tag{4.7}$$

Since for $x \in \mathscr{H}$ and $t, t_0 \in \mathscr{R}$

$$\|[W(t) - W(t_0)]x\| \leqslant \|U(t,0)[\tilde{U}(-t) - \tilde{U}(-t_0)]x\| + \|[U(t,0) - U(t_0, 0)]\tilde{U}(-t_0)x\|,$$

it follows from the uniform boundedness of $U(t, 0)$ and the strong continuity of $U(t, 0)$ and $\tilde{U}(t)$ that $W(t)$ is strongly continuous in t. For similar reasons this is also the case for $W_0(t)$. In addition it is easily verified that

$$W_0(0) = W(0) = I, \qquad W_0(t) = W_0(t+a), \qquad W(t) = W(t+a). \tag{4.8}$$

Since

$$U_0(t,s) = W_0(t)\tilde{U}_0(t-s)W_0^*(s), \qquad U(t,s) = W(t)\tilde{U}(t-s)W^*(s), \tag{4.9}$$

it follows that

$$\Omega_\pm(s)x = s-\lim_{t\to\pm\infty} W(s)\tilde{U}^*(t-s)W^*(t)W_0(t)\tilde{U}_0(t-s)W_0^*(s)x \tag{4.10}$$

provided $\Omega_\pm(s)$ exist.

For $s = 0$ and $t = na$, where n runs through the positive and negative integers respectively, we then have, since $W_0(na) = W(na) = I$:

$$\Omega_\pm(0) = s-\lim_{n\to\pm\infty} \exp(\mathrm{i}a\tilde{H}n)\exp(-\mathrm{i}a\tilde{H}_0 n). \tag{4.11}$$

Consequently the following intertwining properties hold

$$\exp(ia\tilde{H}n)\Omega_{\pm}(0) = \Omega_{\pm}(0)\exp(ia\tilde{H}_0 n), \qquad n = 0, \pm 1, \pm 2, \ldots \tag{4.12}$$

Thus we have obtained an expression for the wave operators in terms of the constant self-adjoint operators $\tilde{H}_0$ and $\tilde{H}$. (These operators are Hilbert space operator versions of the so called monodromy matrices of the Floquet theory.) In this way the effect of the external field term $H^{(1)}(t)$ is entirely absorbed in $\tilde{H}_0$ and $\tilde{H}$, which operators might be called the dressed hamiltonians of the problem (see also Cohen-Tannoudji and Haroche 1969, where a dressed hamiltonian is introduced for a description of the scattering of light by an atom subjected to a strong radio-frequency field).

Next we turn to the question of what happens when the frequency $\omega = 2\pi/a$ of the external field term tends to infinity. We shall restrict our attention to the case that $H^{(1)}(t)$ in (4.2) has the specific form

$$H^{(1)}(t) = H^{(1)}\cos(\omega t + \delta). \tag{4.13}$$

We assume furthermore that the conditions of lemma 4.1 are satisfied. In particular we assume the existence of $\Omega_{\pm}(s)$. Since we are considering explicitly the dependence of various quantities on ω we shall indicate this by a superscript, ie $U(t, s) = U^{(\omega)}(t, s)$, $\Omega_{\pm}(s) = \Omega_{\pm}^{(\omega)}(s)$, etc. We define

$$U_0^{(\infty)}(t, s) = \exp[-iH^{(0)}(t-s)], \qquad U^{(\infty)}(t, s) = \exp[-iH(t-s)]. \tag{4.14}$$

Lemma 4.2. Let the conditions of lemma (4.1) be satisfied and let $H^{(1)}(t)$ have the special form (4.13). Then

$$U_0^{(\infty)}(t, s) = s-\lim_{\omega\to\pm\infty} U_0^{(\omega)}(t, s), \qquad U^{(\infty)}(t, s) = s-\lim_{\omega\to\pm\infty} U^{(\omega)}(t, s). \tag{4.15}$$

Proof. We give the proof for $U^{(\omega)}(t, s)$, the proof for $U_0^{(\omega)}(t, s)$ being analogous.

Equation (2.18) applied to the present case yields for arbitrary $x \in \mathscr{H}$:

$$U^{(\omega)}(t, s)x = U^{(\infty)}(t, s)x - i\int_s^t du\, U^{(\infty)}(t, u)H^{(1)}(u)U^{(\omega)}(u, s)x. \tag{4.16}$$

As mentioned earlier this expression can be used to generate a converging perturbation series expansion for $U^{(\omega)}(t, s)x$. In fact it has been shown by Phillips (1953), under conditions that are satisfied in the present case, that $U^{(\omega)}(t, s)$ is given by

$$U^{(\omega)}(t, s) = \sum_{n=0}^{\infty} W_n(t, s), \tag{4.17}$$

where the convergence is with respect to the uniform topology on any finite interval $[s, t]$. Here

$$W_0(t\ s) = U^{(\infty)}(t, s) \tag{4.18}$$

and

$$W_n(t, s)x = -i\int_s^t du U^{(\infty)}(t, u)H^{(1)}(u)W_{n-1}(u, s)x, \qquad n \geqslant 1.$$

Thus we obtain

$$\langle [U^{(\omega)}(t,s)-U^{(\infty)}(t,s)]x|y\rangle = \left|\left\langle \sum_{n=1}^{\infty} W_n(t,s)x\middle| y\right\rangle\right|$$

$$\leqslant \sum_{n=1}^{N} |\langle W_n(t,s)x|y\rangle| + \left\|\sum_{n=N+1}^{\infty} W_n(t,s)\right\| \ldots \|x\| \ldots \|y\|.$$

Now the sum in the last term can be made arbitrarily small, independent of ω, by choosing N sufficiently large (see Phillips 1953, equation (28), where now $M = 1$, $\omega = 0$ and $K_s = \|H^{(1)}\|$). Thus it is sufficient to show that $\langle W_n(t,s)x|y\rangle$ tends to zero for $\omega \to \pm\infty$ for arbitrary $n = 1, 2, \ldots$. As $\|U^{(\omega)}(t,s)x\| = \|U^{(\infty)}(t,s)x\| = \|x\|$ the strong convergence of $U^{(\omega)}(t,s)x$ towards $U^{(\infty)}(t,s)x$ then follows from the above result. We can write for $n \geqslant 1$

$$\langle W_n(t,s)x|y\rangle = \int_s^t \mathrm{d}t_1 \int_s^{t_1} \mathrm{d}t_2 \ldots \int_s^{t_{n-1}} \mathrm{d}t_n \cos(\omega t_1+\delta) \ldots \cos(\omega t_n+\delta) F(t, t_1, \ldots, t_n, s), \tag{4.19}$$

where

$$F(t, t_1, \ldots, t_n, s) = (-\mathrm{i})^n (U^{(\infty)}(t, t_1) H^{(1)} \ldots U^{(\infty)}(t_{n-1}, t_n) H^{(1)} U^{(\infty)}(t_{n,s})x, y) \tag{4.20}$$

is a continuous function of each of its arguments. A simple extension of the reasoning that leads to the usual Riemann–Lebesgue lemma then leads to the conclusion that $(W_n(t,s)x, y)$ tends to zero for $\omega \to \pm\infty$.

Theorem 4.1. Let the conditions of lemma 4.2 be satisfied and suppose further that the wave operators $\Omega_\pm^{(\infty)}$ exist as well as $\Omega_\pm^{(\omega)}(s)$ for every $\omega \in \mathscr{R}$. If the (strong) convergence of $(U^{(\omega)}(t,s))^* U_0^{(\omega)}(t,s)$ towards $\Omega_\pm^{(\omega)}(s)$ is uniform with respect to ω, ie for any given $\epsilon > 0$ and for given $x \in \mathscr{H}$ there exists a $t_0 = t_0(\epsilon, x)$ independent of ω, such that

$$\|\Omega_\pm^{(\omega)}(s)x - (U^{(\omega)}(t,s))^* U_0^{(\omega)}(t,s)x\| < \epsilon$$

for all $|t| > |t_0|$, then

$$\Omega_\pm^{(\infty)}x = s-\lim_{|\omega|\to\infty} \Omega_\pm^{(\omega)}x.$$

Proof. For given $\epsilon > 0$ we can find a $t_0 \in \mathscr{R}$, independent of ω, such that in the inequality

$$\begin{aligned}
&\|\Omega_+^{(\omega)}(s)x - \Omega_+^{(\infty)}x\| \\
&\quad \leqslant \|\Omega_+^{(\omega)}(s)x - (U^{(\omega)}(t,s))^* U_0^{(\omega)}(t,s)x\| \\
&\quad + \|(U^{(\omega)}(t,s))^* U_0^{(\omega)}(t,s)x - (U^{(\infty)}(t,s))^* U_0^{(\infty)}(t,s)x\| \\
&\quad + \|\Omega_+^{(\infty)}x - (U^{(\infty)}(t,s))^* U_0^{(\infty)}(t,s)x\|
\end{aligned}$$

the first and the last term at the right are smaller than $\epsilon/4$ for $t > t_0$. Keeping t fixed from now on we can choose $|\omega|$ so large that the middle term becomes smaller than

$\epsilon/2$. This follows from the inequality

$$\begin{aligned}
&\|(U^{(\omega)}(t,s))^* U_0^{(\omega)}(t,s)x - (U^{(\infty)}(t,s))^* U_0^{(\infty)}(t,s)x\| \\
&\qquad \leqslant \|(U^{(\omega)}(t,s))^*[U_0^{(\omega)}(t,s) - U_0^{(\infty)}(t,s)]x\| \\
&\qquad\quad + \|[(U^{(\omega)}(t,s))^* - (U^{(\infty)}(t,s))^*]U_0^{(\infty)}(t,s)x\| \\
&\qquad = \|[U_0^{(\omega)}(t,s) - U_0^{(\infty)}(t,s)]x\| + \|[U^{(\omega)}(s,t) - U^{(\infty)}(s,t)]U_0^{(\infty)}(t,s)x\| \qquad (4.21)
\end{aligned}$$

and lemma 4.2 applied twice.

We shall have occasion to apply theorem 4.1 in II for the special models considered there. It will be evident that lemma 4.2 and theorem 4.1 still hold for $H^{(1)}(t)$ of the type

$$H^{(1)}(t) = \sum_{k=1}^{n} H_k^{(1)} \cos(k\omega t + \delta_k). \qquad (4.22)$$

Infinite series of the above type can in general not be treated by means of the present methods. If, however,

$$\sum_{k=1}^{\infty} \|H_k^{(1)}\| < \infty, \qquad (4.23)$$

then the above results remain true. This follows from a consideration of the expression for $\langle W_n(t,s)x|y\rangle$ which replaces (4.19) in this more general case.

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References

Cohen-Tannoudji C and Haroche S 1969 *J. Phys., Paris* **30** 125–44, 153–68
Dorfman J R and Cohen E G D 1967 *J. math. Phys.* **8** 282–97
Hahn L and Hertel I V 1972 *J. Phys.* B: *Atom. molec. Phys.* **5** 1995–2015
Ince E L 1956 *Ordinary Differential Equations* (New York: Dover)
Kato T 1953 *J. Math. Soc. Japan* **5** 208–34
Phillips R S 1953 *Trans. Am. Math. Soc.* **74** 199–221
Prugovečki E 1971 *Quantum Mechanics in Hilbert Space* (New York and London: Academic Press)
Prugovečki E and Tip A 1974 *J. Phys.* A: *Math., Nucl. Gen.* **7** 586–96
Shirley J H 1965 *Phys. Rev.* **138** B979–87