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Gauge-invariant time-dependent perturbation theory: II. Degenerate case

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Abstract. This paper concludes our formulation of a manifestly gauge-invariant, time-dependent perturbation theory for a non-relativistic, spinless charged particle interacting with a classical, external electromagnetic radiation field. In this paper, we consider the treatment of the degenerate bound states and the states in the continuum. We have found that the gauge-invariant and the conventional perturbative solutions agree on the net transition rates but disagree on the transition amplitudes and transition probabilities. The treatment of a realistic physical system can be obtained by combining the treatment here with that in a previous paper for a non-degenerate system.

1. Introduction

In the investigation of the gauge-invariant formulation of a quantised particle interacting with classical, external electromagnetic fields, there are theoretical and practical aspects. The theoretical investigation concerns the *exact* quantities and the basic physical principles involved (Yang 1976, 1982a, b, Cohen-Tannoudji *et al* 1977, Kobe and Smirl 1978, Leubner and Zoller 1980, Kobe and Wen 1980, 1982, Leubner 1981, Shirokov 1981, Kobe *et al* 1982, Kobe 1983). It therefore provides the necessary foundations for an understanding of the basic mechanism of how a charged particle interacts with the fields. An analysis by Shirokov (1981) illustrates the usual confusion inherent in the theoretical investigation: that the proofs valid for processes pertaining to the S-matrix formalism are often mistakenly applied to processes for which the usual S-matrix formalism does not apply. (See also Yang (1982b) for a different analysis.)

The practical aspect of the investigation deals with how to devise some approximation procedures to solve the differential equations for the probability amplitudes formulated in the theoretical investigation. Here, one overriding concern is still the manifest gauge invariance. Because of this requirement, only those approximation procedures with the correct gauge properties can be used to solve for the probability amplitudes in our formulation. Our previous paper dealing with the time-dependent perturbation theory (TDPT) of a non-degenerate system is just one such example (Yang 1982c, to be referred to as I). An excellent review of the applications of the gauge-invariant formulation can be found in Lee and Albrecht (1983).

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The main purposes of I are to establish our notation, and to introduce the concept and carry out the proofs of manifest gauge invariance in the perturbation theory. (The gauge invariance in an exact theory and that in a perturbation theory demand some careful distinctions.) The treatment in I alone is not sufficient to deal with the electromagnetic interaction of any realistic systems since all such systems usually exhibit some kinds of degeneracy. It can, however, be considered to be a good approximation when the complete Hilbert space of the particle system is truncated into a finite number of non-degenerate states and if the effects due to the truncation are estimated to be small. For example, in a process involving the near resonant interaction of an atom with a coherent laser field (e.g. Sargent *et al* 1977, Knight and Milonni 1980), a two-state approximation is often sufficient.

The purpose of this second paper is to conclude our formulation of the gauge-invariant TDPT by extending our treatment to include the degenerate states. The system to be considered is still that of a non-relativistic, spinless charged particle interacting with classical, external electromagnetic fields. Here, we will consider two kinds of degeneracy: that of the discrete (bound) states and that of the states in the continuum (scattering states). The treatment to be developed here is intended for an *arbitrary* radiation field. If the field encountered exhibits some special characteristics, e.g. a circularly polarised radiation field, then a different treatment is more appropriate. It will not be discussed here.

The arrangement of this paper is as follows. In § 2, we shall briefly review the exact gauge-invariant formulation and establish our notation suitable for the particular situation here. In § 3, we first examine the eigenfunction and eigenvalue corrections of the energy operator (H_B) obtained by the usual Rayleigh–Schrödinger (RS) procedure (e.g. Messiah 1966) for bound states. Due to the degeneracy, the 'zeroth-order' eigenfunctions obtained from this procedure generally involve the fields (through the $\boldsymbol{A}$ dependence in V_i defined in (3.2)). Because of this property, the 'nth-order' eigenfunctions will not have the simple A^n dependence. Thus, for the purpose of developing a TDPT, we investigate an alternative procedure that does permit perturbative solutions (using the potentials and/or fields as the expansion parameters). Also in this section, we review the usual procedure for obtaining the exact and perturbative solutions of the scattering states (e.g. Goldberger and Watson 1964).

In § 4, we use the perturbative solutions discussed in § 3 to formulate our TDPT and derive the perturbative rates of transition due to a single-frequency radiation field. Here, all the formal proofs of manifest gauge invariance will be omitted since the methods for a non-degenerative system still apply. Finally in § 5, we conclude this series of two papers by examining once again the simplest gauge problem from which this author started his investigation on the gauge-invariant formulation (Yang 1976)—the problem whether $\boldsymbol{A}\cdot\boldsymbol{p}$ or $\boldsymbol{r}\cdot\boldsymbol{E}$ should be used as the transition operator when the time-varying field is treated in the dipole approximation $\boldsymbol{E}(\boldsymbol{r}, t)\approx\boldsymbol{E}(\boldsymbol{0}, t)$ and $\boldsymbol{B}(\boldsymbol{r}, t)\approx 0$ (Lamb 1952).

2. The gauge-invariant formulation

In this section, we shall briefly review the gauge-invariant formulation and the physical principles involved, and establish our notation necessary for the particular situation to be considered. A perspective of the theory and a very detailed discussion of the exact manifest gauge invariance can be found in Yang (1976, 1982a), Cohen-Tannoudji

et al (1977), Kobe and Smirl (1978), Leubner and Zoller (1980), Leubner (1981), Kobe *et al* (1982), Kobe (1983) and Lee and Albrecht (1983). A discussion of the experimental basis of the formulation can be found in Yang (1982b).

Let us consider a non-relativistic, spinless charged particle of mass m and charge e in the presence of a conservative electrostatic field $\boldsymbol{E}_0(\boldsymbol{r}) = -\boldsymbol{\nabla} V_0(\boldsymbol{r})$ and a time-varying electromagnetic radiation field $\boldsymbol{E}(\boldsymbol{r}, t)$ and $\boldsymbol{B}(\boldsymbol{r}, t)$. If we use the potential $\boldsymbol{A}(\boldsymbol{r}, t)$ and $\Phi(\boldsymbol{r}, t)$ in an arbitrary gauge to represent the time-varying fields, then

$$\boldsymbol{E}(r, t) = -\boldsymbol{\nabla}\Phi(\boldsymbol{r}, t) - c^{-1}\partial \boldsymbol{A}(\boldsymbol{r}, t)/\partial t, \qquad \boldsymbol{B}(\boldsymbol{r}, t) = \boldsymbol{\nabla} \times \boldsymbol{A}(\boldsymbol{r}, t). \tag{2.1}$$

The Schrödinger equation and the Hamiltonian in this gauge are then†

$$\mathrm{i}\hbar\dot{\Psi}(\boldsymbol{r}, t) = H(t)\Psi(\boldsymbol{r}, t), \tag{2.2}$$

$$H(t) = (\boldsymbol{p} - e\boldsymbol{A}/c)^2/2m + eV_0 + e\Phi. \tag{2.3}$$

According to the gauge-invariant formulation, we first construct the energy operator $H_{\mathrm{B}}(t)$ that is determined by

$$(\mathrm{d}H_{\mathrm{B}}/\mathrm{d}t)_H = \tfrac{1}{2}(\boldsymbol{J} \cdot \boldsymbol{E} + \boldsymbol{E} \cdot \boldsymbol{J}) \equiv P(t), \tag{2.4}$$

where $(\mathrm{d}H_{\mathrm{B}}/\mathrm{d}t)_H = \partial H_{\mathrm{B}}/\partial t + [H_{\mathrm{B}}, H]/\mathrm{i}\hbar$. The symbol $\boldsymbol{J}$ is the current operator associated with the Hamiltonian in (2.3), and is therefore, using $\boldsymbol{n} = (\boldsymbol{p} - e\boldsymbol{A}/c)/m$ to denote the velocity operator,

$$\boldsymbol{J} = e\boldsymbol{v} = e(\boldsymbol{p} - e\boldsymbol{A}/c)/m. \tag{2.5}$$

The operator $P(t)$ in (2.4) will be referred to as the power (or Poynting) operator. Equation (2.4) is obtained by applying the correspondence principle (Bohr 1928) to the classical Poynting theorem and the conservation of energy (e.g. Jackson 1975, Yang 1982a, b, Kobe *et al* 1982). From the definition of $(\mathrm{d}H_B/\mathrm{d}t)_H$ and (2.3)–(2.5), it can be shown that

$$H_{\mathrm{B}}(t) = (\boldsymbol{p} - e\boldsymbol{A}/c)^2/2m + eV_0, \tag{2.6}$$

which is just the sum of the particle's *Newtonian* kinetic energy and the potential energy.

If we use $\{E_s(t)\}$ and $\{\Psi_s(\boldsymbol{r}, t)\}$ to denote the eigenvalues and the orthonormal and complete (assumed) set of eigenfunctions of $H_{\mathrm{B}}(t)$, then‡

$$H_{\mathrm{B}}(t)\Psi_s(\boldsymbol{r}, t) = E_s(t)\Psi_s(\boldsymbol{r}, t), \tag{2.7}$$

$$\langle \Psi_s(t) | \Psi_{s'}(t) \rangle = \delta_{ss'}. \tag{2.8}$$

The gauge-invariant probability amplitudes $\{a_s(t)\}$ are then defined by

$$a_s(t) = \langle \Psi_s(t) | \Psi(t) \rangle. \tag{2.9}$$

† Throughout this paper, we use $\dot{f} = \mathrm{d}f/\mathrm{d}t$ if f is a function of time only, and $\dot{f} = \partial f/\partial t$ if f is a function of time and position.

‡ Let us note here that the word 'degenerate' in the title of this paper refers to the spectrum of the 'unperturbed Hamiltonian' $H_0 = \boldsymbol{p}^2/2m + eV_0$, not to the spectrum of the energy operator H_{B} in (2.6). Because of the presence of the vector potential $\boldsymbol{A}$ in H_{B}, some degeneracy in the spectrum of H_0 may be removed if there is a magnetic field (see equation (A7)). I am grateful to one referee for kindly pointing out that this point should be explained carefully.

These amplitudes satisfy the differential equation

$$i\hbar\dot{a}_s = E_s a_s + \sum_{s'} a_{s'}\langle\Psi_s|(e\Phi - i\hbar\partial/\partial t)|\Psi_{s'}\rangle. \tag{2.10}$$

As has been shown before (e.g. Yang 1976, Kobe and Smirl 1978), the transition matrix elements can be expressed in terms of the power operator. If $E_s \neq E_{s'}$, then

$$\langle\Psi_s|(e\Phi - i\hbar\partial/\partial t)|\Psi_{s'}\rangle = i\hbar\langle\Psi_s|P(t)|\Psi_{s'}\rangle/(E_s - E_{s'}). \tag{2.11}$$

In the rest of this paper, we shall develop a particular type of TDPT to solve for the probability amplitudes from (2.10). The procedure will parallel that for a non-degenerate system (I). However, because of the extra complexity introduced by the degeneracy of bound states and the problems associated with the states in the continuum, we will have to be cautious in solving the eigenvalue problem (2.7) perturbatively. This will be discussed in detail in the next section. Finally, we note that for the purpose of the TDPT involving the bound states, we shall require that *all* scalar potentials vanish at the origin at all times, which can be accomplished simply by the substitution $\Phi(\boldsymbol{r}, t) \to \Phi(\boldsymbol{r}, t) - \Phi(\boldsymbol{0}, t)$. As has been explained before (I, appendix), such a requirement entails no consequences in both the transition matrix elements and the probabilities.

3. Perturbative solutions of the eigenvalue equation

In this section, we shall discuss in detail the procedures by which the eigenvalues $\{E_s(t)\}$ and the eigenfunctions $\{\Psi_s(\boldsymbol{r}, t)\}$ are to be solved and the consequences on our development of a TDPT. We shall also discuss the physical interpretations associated with the ingoing and outgoing scattering states according to the usual static (time-independent) scattering theory (e.g. Goldberger and Watson 1964). Such a discussion is necessary to provide a basis for using the outgoing scattering states of equation (2.7) to define the initial condition for $\{a_s(0)\}$ and the ingoing scattering states to define the gauge-invariant probability amplitudes.

3.1. The Rayleigh–Schrödinger procedure

For clarity of notation, we will use $E_s^{(n)}$ and $\Psi_s^{(n)}$ for the nth-order corrections in the eigenvalue and eigenfunction of state s *strictly* obtained by the RS procedure for (2.7) with the decomposition (e.g. Messiah 1966)

$$H_B(t) = H_0 + V_1(t) + V_2(t), \tag{3.1}$$

where $H_0 = \boldsymbol{p}^2/2m + eV_0$ is the usual 'unperturbed Hamiltonian' and

$$V_1 = -e(\boldsymbol{p}\cdot\boldsymbol{A} + \boldsymbol{A}\cdot\boldsymbol{p})/2mc, \qquad V_2 = (e\boldsymbol{A})^2/2mc^2. \tag{3.2}$$

For bound-state solutions the RS procedure gives, using $\hbar\omega_s$ for $E_s^{(0)}$,

$$(H_0 - \hbar\omega_s)\Psi_s^{(0)}(\boldsymbol{r}, t) = 0, \tag{3.3}$$

$$(H_0 - \hbar\omega_s)\Psi_s^{(1)} + (V_1 - E_s^{(1)})\Psi_s^{(0)} = 0, \tag{3.4}$$

$$(H_0 - \hbar\omega_s)\Psi_s^{(2)} + (V_1 - E_s^{(1)})\Psi_s^{(1)} + (V_2 - E_s^{(2)})\Psi_s^{(0)} = 0, \tag{3.5}$$

and so on. Here, the relations between $\{E_s^{(n)}\}$ and $\{E_s\}$ and between $\{\Psi_s^{(n)}\}$ and $\{\Psi_s\}$ are

$$E_s = \sum_{n=0}^{\infty} E_s^{(n)}, \qquad \Psi_s = \sum_{n=0}^{\infty} \Psi_s^{(n)}. \tag{3.6}$$

The renormalisation required is

$$\langle \Psi_s | \Psi_{s'} \rangle = \langle \Psi_s^{(0)} | \Psi_{s'}^{(0)} \rangle = \delta_{ss'}. \tag{3.7}$$

As is well known from the RS *degenerate* perturbation theory (e.g. Messiah 1966, pp 698–700), the zeroth-order eigenfunctions $\{\Psi_s^{(0)}\}$ are determined by both (3.3) and (3.4). That is, the $\{\Psi_s^{(0)}\}$ not only must satisfy (3.3) but must also diagonalise V_1 such that

$$\langle \Psi_s^{(0)} | V_1(t) | \Psi_{s'}^{(0)} \rangle = E_s^{(1)} \delta_{ss'} \qquad \text{for} \quad \hbar\omega_s = \hbar\omega_{s'}. \tag{3.8}$$

Equation (3.8) characterises the differences in the perturbative solutions between the non-degenerate and degenerate cases (see appendix). Thus, in general, these zeroth-order eigenfunctions are time dependent through the $\boldsymbol{A}$ dependence in V_1. Because of this $\boldsymbol{A}$ dependence in the zeroth-order eigenfunctions, it will complicate our formulation of a time-dependent perturbation theory using the potentials, and hence the fields, as the expansion parameters. Let us now investigate this situation more closely.

To begin with, we use $\{\phi_{j,\alpha}(\boldsymbol{r})\}$ to denote the eigenfunctions of the 'unperturbed' Hamiltonian H_0 and some other operators that commute with H_0 (e.g. $\boldsymbol{L}^2$ and L_z for the hydrogen atom), with the associated eigenvalues $\{\hbar\omega_j\}$, where α is the degeneracy index:

$$H_0 \phi_{j,\alpha}(\boldsymbol{r}) = \hbar\omega_j \phi_{j,\alpha}(\boldsymbol{r}), \qquad \langle \phi_{j,\alpha} | \phi_{k,\beta} \rangle = \delta_{jk}\delta_{\alpha\beta}. \tag{3.9}$$

By comparing (3.3) and (3.9), we may choose $s = (j, \alpha)$ and $\omega_s = \omega_j$. Furthermore, it also follows that, for each j, there exists a unitary matrix $(D_{j,\alpha\beta})$ such that

$$\Psi_{j,\alpha}^{(0)}(\boldsymbol{r}, t) = \sum_{\beta} D_{j,\alpha\beta}^{*} \phi_{j,\beta} \qquad \text{or} \qquad \phi_{j,\alpha} = \sum_{\beta} \Psi_{j,\beta}^{(0)} D_{j,\beta\alpha} \tag{3.10}$$

where * denotes the complex conjugate. From now on, we shall use (j, α) for s and similar notation for s' in all of our previous perturbative equations and results. The values of $E_{j,\alpha}^{(1)}$ and $D_{j,\alpha\beta}$ are determined by diagonalising the matrix $(\varepsilon_{j,\alpha\beta}^{(1)})$ where

$$\varepsilon_{j,\alpha\beta}^{(1)}(t) = \langle \phi_{j,\alpha} | V_1(t) | \phi_{j,\beta} \rangle. \tag{3.11}$$

It can be shown (see appendix) that all $\varepsilon_{j,\alpha\beta}^{(1)}(t)$ are gauge invariant, which means that all $E_{j,\alpha}^{(1)}$ and $D_{j,\alpha\beta}$ are gauge invariant. Since $\varepsilon_{j,\alpha\beta}^{(1)}$ in general depend on V_1 unless $[V_1, H_0] = 0$ or $\nabla \times \boldsymbol{A} = 0$, it follows that all $E_{j,\alpha}^{(1)}$ and $D_{j,\alpha\beta}$ have time and field dependence. So, $\Psi_{j,\alpha}^{(0)}$ will in general have these two kinds of dependence. Because of this characteristic property, which pertains only to degenerate bound states, the 'zeroth-order' transition matrix elements $\{\langle \Psi_{j,\alpha}^{(0)} | -i\hbar\partial\Psi_{j,\beta}^{(0)}/\partial t \rangle\}$ will generally be time and field dependent (through the dependence of $D_{j,\alpha\beta}$ on $\boldsymbol{A}(\boldsymbol{r}, t)$)†. This means that if we use the eigenfunctions obtained from the usual RS procedure in (2.10), the

† For some special field situations, such as a precessing or rotating magnetic field (Güttinger 1931, Schwinger 1937, Rabi 1937, Yang 1976), the exact matrix elements $\{\langle \Psi_{j,\alpha} | -i\hbar\partial\Psi_{j,\beta}/\partial t \rangle\}$ are independent of both the time and the magnetic field. The eigenvalues $\{E_{j,\alpha}\}$ do depend upon the field (but are time independent). For such a situation, the TDPT to be developed here still applies (with less accurate results), although a different treatment is more appropriate.

resulting TDPT for the $\{a_{j,\alpha}\}$ will not be in the power series of the fields. To circumvent this problem, we will transform (2.10) into another equation in terms of other gauge-invariant coefficients such that their nth-order quantities will have exactly the nth power dependence on the fields.

3.2. An alternative procedure

Our objective is, in preparation for the TDPT, to solve *perturbatively* for the set of functions $\{u_{j,\alpha}(\boldsymbol{r}, t)\}$ that are defined by

$$u_{j,\alpha}(\boldsymbol{r}, t) = \sum_{\beta} \Psi_{j,\beta} D_{j,\beta\alpha}, \tag{3.12}$$

which is chosen according to (3.10). From (2.7) and the above, we get

$$H_{\mathrm{B}} u_{j,\alpha} = \sum_{\beta} u_{j,\beta} \varepsilon_{j,\beta\alpha} \qquad \text{where} \quad \varepsilon_{j,\beta\alpha} = \sum_{\gamma} D^{*}_{j,\gamma\beta} E_{j,\gamma} D_{j,\gamma\alpha}. \tag{3.13}$$

As is clear from the above equation, the functions $\{u_{j,\alpha}(\boldsymbol{r}, t)\}$ are in general not the eigenfunctions of $H_{\mathrm{B}}(t)$.

If we expand $u_{j,\alpha}$ and $\varepsilon_{j,\alpha\beta}$ using the potential $\boldsymbol{A}$ as the expansion parameter, then

$$u_{j,\alpha} = \sum_{n=0}^{\infty} u^{(n)}_{j,\alpha} \qquad \text{and} \qquad \varepsilon_{j,\alpha\beta} = \sum_{n=0}^{\infty} \varepsilon^{(n)}_{j,\alpha\beta}. \tag{3.14}$$

Clearly, $u^{(0)}_{j,\alpha} = \phi_{j,\alpha}(\boldsymbol{r})$ by (3.9), (3.10) and (3.12). Substituting (3.14) into (3.13) and using (3.1), we get

$$(H_0 - \hbar\omega_j) u^{(1)}_{j,\alpha} + V_1 \phi_{j,\alpha} = \sum_{\beta} \phi_{j,\beta} \varepsilon^{(1)}_{j,\beta\alpha}, \tag{3.15}$$

$$(H_0 - \hbar\omega_j) u^{(2)}_{j,\alpha} + V_1 u^{(1)}_{j,\alpha} + V_2 \phi_{j,\alpha} = \sum_{\beta} \{u^{(1)}_{j,\beta} \varepsilon^{(1)}_{j,\beta\alpha} + \phi_{j,\beta} \varepsilon^{(2)}_{j,\beta\alpha}\}, \tag{3.16}$$

and so on. The normalisation required is

$$\sum_{m=0}^{n} \langle u^{(m)}_{j,\alpha} | u^{(n-m)}_{j,\beta} \rangle = 0 \qquad \text{for all } j \text{ and all } n \geqslant 1. \tag{3.17}$$

The ε's in (3.15) and (3.16) can be derived using a method identical to that for the non-degenerate case. Thus, the $\{\varepsilon^{(1)}_{j,\alpha\beta}\}$ are given by (3.11) and

$$\varepsilon^{(2)}_{j,\alpha\beta} = \langle \phi_{j,\alpha} | V_1 | u^{(1)}_{j,\beta} \rangle + \langle \phi_{j,\alpha} | V_2 | \phi_{j,\beta} \rangle - \sum_{\gamma} \delta^{(1)}_{j,\alpha\gamma} \varepsilon^{(1)}_{j,\gamma\beta}, \tag{3.18}$$

where $\delta^{(n)}_{j,\alpha\gamma} = \langle \phi_{j,\alpha} | u^{(n)}_{j,\gamma} \rangle$ for $n \geqslant 1$. Our choices for the first- and second-order δ's are (I, appendix):

$$\delta^{(1)}_{j,\alpha\gamma} = (ie/c\hbar) \langle \phi_{j,\alpha} | F[\boldsymbol{A}] | \phi_{j,\gamma} \rangle, \tag{3.19}$$

$$\delta^{(2)}_{j,\alpha\gamma} = -\tfrac{1}{2} \langle u^{(1)}_{j,\alpha} | u^{(1)}_{j,\gamma} \rangle + (ie/2c\hbar) \{ \langle \phi_{j,\alpha} | F[\boldsymbol{A}] | u^{(1)}_{j,\gamma} \rangle + \langle u^{(1)}_{j,\alpha} | F[\boldsymbol{A}] | \phi_{j,\gamma} \rangle \} \tag{3.20}$$

where

$$F[\boldsymbol{A}] = \int_0^1 \mathrm{d}s\, \boldsymbol{r} \cdot \boldsymbol{A}(s\boldsymbol{r}, t). \tag{3.21}$$

This concludes our investigation into the bound degenerate states. In § 3.3, we will discuss the scattering states.

3.3. The scattering-state eigenfunctions

Here we shall briefly discuss the scattering-state eigenfunctions of both the 'unperturbed' Hamiltonian and the energy operator H_B. Most of these discussions can be found in any textbook (e.g. Goldberger and Watson 1964) dealing with the scattering of a particle by a static potential. In the case of the scattering-state eigenfunctions of $H_B(t)$, the time t is treated as a parameter.

Let us use $\{\phi_k^{(+)}(r)\}$ for the outgoing and $\{\phi_k^{(-)}(r)\}$ for the ingoing eigenfunctions of H_0, i.e.,

$$H_0\phi_k^{(\pm)}(r) = \varepsilon_k\phi_k^{(\pm)}(r), \qquad \varepsilon_k = (\hbar k)^2/2m. \tag{3.22}$$

Similarly, if we use $\{u_k^{(+)}(r, t)\}$ for the outgoing and $\{u_k^{(-)}(r, t)\}$ for the ingoing eigenstates of $H_B(t)$, then

$$H_B(t)u_k^{(\pm)}(r, t) = \varepsilon_k u_k^{(\pm)}(r, t). \tag{3.23}$$

On equation (3.23) we shall also impose the condition that the fields $\boldsymbol{E}$ and $\boldsymbol{B}$ vanish outside some *finite* region.

The conventional interpretation of $\phi_k^{(+)}(r)$ is that the particle has an incident (initial) momentum $\hbar k$ before it interacts with eV_0 in H_0, and is then scattered. The physical process associated with $\phi_k^{(-)}(r)$ is that the particle is measured to have a final momentum $\hbar k$ after it has interacted with eV_0. These same physical interpretations will be applied to $u_k^{(+)}$ and $u_k^{(-)}$, with the understanding that the fields $\boldsymbol{E}(r, t)$ and $\boldsymbol{B}(r, t)$ are 'suddenly frozen' at the values at time t. It then follows that the initial wavefunction $\Psi(r, 0)$ must be denoted by $u_i^{(+)}(r, 0)$ with $\hbar i$ as the incident momentum. Similarly, the gauge-invariant coefficients in (2.9) must be defined by $a_k(t) = \langle u_k^{(-)}(t)|\Psi(t)\rangle$ since we are interested in the probability amplitudes after the interaction with the radiation fields.

By the standard procedure, we have

$$u_k^{(\pm)} = \phi_k^{(\pm)} + G_B^{(\pm)}(\varepsilon_k)[V_1(t) + V_2(t)]u_k^{(\pm)}, \tag{3.24}$$

$$\langle u_k^{(\pm)}|u_{k'}^{(\pm)}\rangle = \langle\phi_k^{(\pm)}|\phi_{k'}^{(\pm)}\rangle = \delta(k - k'), \tag{3.25}$$

where V_1 and V_2 are listed in (3.2) and

$$G_B^{(\pm)}(\varepsilon_k) = [\varepsilon_k - H_0 \pm i0]^{-1}. \tag{3.26}$$

For our later convenience we shall henceforth suppress the superscripts $(-)$ in all wavefunctions and operators pertaining to the ingoing states. That is, we shall use ϕ_k for $\phi_k^{(-)}$, $G_B(\varepsilon_k)$ for $G_B^{(-)}(\varepsilon_k)$, etc. If we expand u_k as

$$u_k(r, t) = \sum_{n=0}^{\infty} u_k^{(n)}(r, t), \tag{3.27}$$

then $u_k^{(0)}(r, t) = \phi_k(r)$ and

$$u_k^{(1)} = G_B(\varepsilon_k)V_1\phi_k, \tag{3.28}$$

$$u_k^{(2)} = G_B(\varepsilon_k)[V_1G_B(\varepsilon_k)V_1 + V_2]\phi_k, \tag{3.29}$$

and so on. Note, the normalisation in (3.25) is what we want; there is no need (unlike the bound states) to choose the values of $\langle\phi_k|u_{k'}^{(n)}\rangle$ for $n \geq 1$ and $k'^2 = k^2$.

If we assume that the fields $\boldsymbol{E}(\boldsymbol{r}, t)$ and $\boldsymbol{B}(\boldsymbol{r}, t)$ vanish identically everywhere at $t \leqslant 0$, then it can be shown that

$$a_{\boldsymbol{k}}(0) = \langle u_{\boldsymbol{k}}(0) | u_{\boldsymbol{i}}^{(+)}(0) \rangle = \langle \phi_{\boldsymbol{k}} | \phi_{\boldsymbol{i}}^{(+)} \rangle, \tag{3.30}$$

regardless of the values of $\boldsymbol{A}(\boldsymbol{r}, 0)$ and $\Phi(\boldsymbol{r}, 0)$. Note, $a_{\boldsymbol{k}}(0)$ may not vanish for $\boldsymbol{k} \neq \boldsymbol{i}$. However, due to the conservation of energy in the static scattering processes, $a_{\boldsymbol{k}}(0) = 0$ unless $\boldsymbol{k}^2 = \boldsymbol{i}^2$. This completes our review of the scattering-state eigenfunctions of the energy operator.

3.4. Discussions of the bound and scattering states

One may note that the perturbative solutions of the degenerate bound states are obtained differently from those of the scattering states. This is because the bound states have very different physical properties from the scattering states. One notes that the bound states carry no flux at infinity (in space) whereas the scattering states do. Because of this the degenerate bound states have to adjust their distributions of probabilities, which causes a change in their eigenenergies. On the other hand, the scattering states can always adjust the flow of flux in such a way as to match the initial (for outgoing states) or final (for ingoing states) conditions.

4. Time-dependent perturbation theory

As we have mentioned in § 3.1 in connection with the degenerate *bound* states, physically we would like to use $\{\Psi_{j,\alpha}^{(n)}(\boldsymbol{r}, t)\}$ in (2.10). This is because each $\Psi_{j,\alpha}^{(n)}$ is an nth-order eigenfunction correction of H_{B}. However, the $\{\Psi_{j,\alpha}^{(n)}\}$ as determined from the RS procedure do not have the simple A^n dependence. This implies that their use in (2.10) will not result in a perturbation theory using potentials and/or fields as the expansion parameters.

To circumvent this problem, we shall use $\{u_{j,\alpha}^{(n)}\}$ discussed in § 3.3 in (2.10) to develop a perturbation theory for a different set of *gauge-invariant* expansion coefficients. We will then discuss how to construct physical quantities from the perturbative solutions of these coefficients. After this is done, we will investigate the net rates of transition due to the interaction with a single-frequency external radiation field.

4.1. Formulation

For simplicity of notation and arguments, we shall assume that all bound states have degeneracy.

First, for bound states we define $(d_{j,\alpha}(t)\}$ by

$$d_{j,\alpha}(t) = \langle u_{j,\alpha}(t) | \Psi(t) \rangle, \tag{4.1}$$

where $\Psi(\boldsymbol{r}, t)$ is the wavefunction, and $\{u_{j,\alpha}(\boldsymbol{r}, t)\}$ are related to $\{\Psi_{j,\alpha}(\boldsymbol{r}, t)\}$ by (3.12) and are solved perturbatively from (3.15) and (3.16). For the scattering states, we define

$$d_{\boldsymbol{k}}(t) = a_{\boldsymbol{k}}(t) = \langle u_{\boldsymbol{k}}(t) | \Psi(t) \rangle, \tag{4.2}$$

where $u_{\boldsymbol{k}}(\boldsymbol{r}, t)$ is an ingoing eigenstate of $H_{\mathrm{B}}(t)$.

If we use the completeness of the direct sum of $\{u_{j,\alpha}(\boldsymbol{r}, t)\}$ and $\{u_{\boldsymbol{k}}(\boldsymbol{r}, t)\}$, then

$$\mathrm{i}\hbar\dot{d}_{j,\alpha} = \sum_{\beta} \varepsilon_{j,\alpha\beta} d_{j,\beta} + \sum_{n,\gamma} d_{n,\gamma}\langle u_{j,\alpha}|(e\Phi - \mathrm{i}\hbar\partial/\partial t)|u_{n,\gamma}\rangle + \sum_{\boldsymbol{k}'} d_{\boldsymbol{k}'}\langle u_{j,\alpha}|(e\Phi - \mathrm{i}\hbar\partial/\partial t)|u_{\boldsymbol{k}'}\rangle, \quad (4.3a)$$

$$\mathrm{i}\hbar\dot{d}_{\boldsymbol{k}} = \varepsilon_{\boldsymbol{k}} d_{\boldsymbol{k}} + \sum_{n,\gamma} d_{n,\gamma}\langle u_{\boldsymbol{k}}|(e\Phi - \mathrm{i}\hbar\partial/\partial t)|u_{n,\gamma}\rangle + \sum_{\boldsymbol{k}'} d_{\boldsymbol{k}'}\langle u_{\boldsymbol{k}}|(e\Phi - \mathrm{i}\hbar\partial/\partial t)|u_{\boldsymbol{k}'}\rangle, \quad (4.3b)$$

where $\dot{d}_{j,\alpha}$ is the time derivative of $d_{j,\alpha}$, and similarly for $\dot{d}_{\boldsymbol{k}}$. These two equations do permit perturbative solutions using potentials and/or fields as the expansion parameters.

In order to put the perturbative form of (4.3*a*) and (4.3*b*) into one equation, let us now alter the notation for the *scattering* states. From now on, we shall use $(j, \alpha) = \boldsymbol{j}$ where $j = |\boldsymbol{j}|$ and α denotes the solid angle of $\boldsymbol{j}$, and similarly for $\boldsymbol{k} = (k, \beta)$. Furthermore, we shall also use $\hbar\omega_j = \varepsilon_j$. Since there is no perturbed eigenvalue for the scattering states, we have $\varepsilon^{(n)}_{j,\alpha\beta}$ for all $n \geq 1$ if (j, α) refers to a scattering state.

Using this notation convention and substituting the expansions in (3.14), (3.27) and

$$d_{j,\alpha} = \sum_{n=0}^{\infty} b^{(n)}_{j,\alpha} \exp(-\mathrm{i}\omega_j t) \quad (4.4)$$

into equations (4.3), we have

$$\mathrm{i}\hbar\dot{b}^{(0)}_{j,\alpha} = 0, \quad (4.5)$$

$$\mathrm{i}\hbar\dot{b}^{(n)}_{j,\alpha} = \sum_{k,\beta} \sum_{m=1}^{n} M^{(m)}_{j\alpha,k\beta} b^{(n-m)}_{k,\beta} \exp(\mathrm{i}\omega_{jk} t), \qquad n \geq 1. \quad (4.6)$$

Here, $\omega_{jk} = \omega_j - \omega_k$ and

$$M^{(m)}_{j\alpha,k\beta} = \varepsilon^{(m)}_{j,\alpha\beta}\delta_{jk} + \sum_{n=1}^{m} \langle u^{(m-n)}_{j,\alpha}|\xi^{(n)}_{k,\beta}\rangle, \qquad m \geq 1, \quad (4.7)$$

$$\xi^{(n)}_{k,\beta} = e\Phi u^{(n-1)}_{k,\beta} - \mathrm{i}\hbar\partial u^{(n)}_{k,\beta}/\partial t. \quad (4.8)$$

Let us note here that (4.5)–(4.8) are practically identical in form to those developed for the non-degenerate case (I). Hence, all the proofs of manifest gauge invariance and all the results for obtaining the perturbative rates of transitions in I still apply here for the expansion coefficients $\{b^{(n)}_{j,\alpha}(t)\}$. In the following, we shall first discuss how to identify physical quantities from the perturbative solutions of these coefficients.

4.2. Physical interpretation of perturbative solutions

From the unitarity of the matrix $(D_{j,\alpha\beta})$ in (3.12) relating $\{u_{j,\alpha}\}$ to $\{\Psi_{j,\alpha}\}$, and from the definition in (4.1), we therefore get, for bound states,

$$\mathscr{P}_j(t) = \sum_{\alpha} |a_{j,\alpha}(t)|^2 = \sum_{\alpha} |d_{j,\alpha}(t)|^2. \quad (4.9)$$

Thus, $\mathscr{P}_j(t)$ is the probability for finding the particle in the j subspace. (The designation of the j subspace is convenient for the perturbative solutions only.) The matrix $(D_{j,\alpha\beta})$ does not have any physical meaning except that, mathematically, it relates the *non*-eigenfunctions $\{u_{j,\alpha}\}$ to the eigenfunctions $\{\Psi_{j,\alpha}\}$ of H_{B}.

Based on (4.9), we may therefore interpret

$$\mathscr{P}_j^{(n)}(t) = \sum_{m=0}^{n} \sum_{\alpha} a_{j,\alpha}^{(n-m)} [a_{j,\alpha}^{(n+m)}]^* = \sum_{m=0}^{n} \sum_{\alpha} b_{j,\alpha}^{(n-m)} [b_{j,\alpha}^{(n+m)}]^* \tag{4.10}$$

as the nth-order probability in the j subspace, where * denotes the complex conjugate and

$$a_{j,\alpha}^{(n)} = \sum_{\beta} D_{j,\alpha\beta} b_{j,\beta}^{(n)} \exp(-i\omega_j t) \tag{4.11}$$

is the nth-order probability amplitude for the particle in the energy eigenstate (j, α). As discussed in the appendix, if there is not external field or if the fields are treated in the dipole approximation, then each $d_{j,\alpha}(t)$ is a physically meaningful probability amplitude, and hence each $d_{j,\alpha}^{(n)} = b_{j,\alpha}^{(n)} \exp(-i\omega_j t)$ is an nth-order meaningful amplitude.

The physical interpretation for $b_{j,\alpha}^{(n)}$ when (j, α) is a scattering state, on the other hand, is completely determined by the physical processes associated with the ingoing states. According to the discussion in § 3.3, $b_{j,\alpha}^{(n)}$ can be interpreted as a (reduced) probability amplitude only after the particle has emerged from the interaction with $\boldsymbol{E}$ and $\boldsymbol{B}$, the time-varying radiation fields.

4.3. Transition matrix elements

The transition matrix elements $\{M_{j\alpha,k\beta}^{(n)}\}$ can be evaluated very simply by using the method described for the non-degenerate case (I) when at least one of the states involved is a bound state. If both states are the scattering states, then one simply uses the perturbative forms in (3.28) and (3.29) in (4.7) and (4.8). Most of these matrix elements can be expressed in terms of the first- and second-order power operators:

$$P^{(1)} = e(\boldsymbol{p} \cdot \boldsymbol{E} + \boldsymbol{E} \cdot \boldsymbol{p})/2mc, \qquad P^{(2)} = -e^2 \boldsymbol{A} \cdot \boldsymbol{E}/mc. \tag{4.12}$$

It can be shown that

$$M_{j\alpha,k\beta}^{(1)} = i[\omega_{jk} + i0]^{-1} \langle \phi_{j,\alpha} | P^{(1)} | \phi_{k,\beta} \rangle \tag{4.13}$$

which is valid *except* when $j = k$ and (j, α) is a bound state. For *bound* states,

$$M_{j\alpha,j\beta}^{(1)} = \varepsilon_{j,\alpha\beta}^{(1)} + \eta_{j,\alpha\beta}^{(1)}, \qquad \eta_{j,\alpha\beta}^{(1)} = \langle \phi_{j,\alpha} | F[-e\boldsymbol{E}] | \phi_{j,\beta} \rangle, \tag{4.14}$$

where $F[-e\boldsymbol{E}]$ can be obtained from (3.21). Under the same conditions that (4.13) is valid, we get

$$M_{j\alpha,k\beta}^{(2)} = i[\omega_{jk} + i0]^{-1} \{ \langle \phi_{j,\alpha} | P^{(1)} | u_{k,\beta}^{(1)} \rangle + \langle u_{j,\alpha}^{(1)} | P^{(1)} | \phi_{k,\beta} \rangle + \langle \phi_{j,\alpha} | P^{(2)} | \phi_{k,\beta} \rangle \}$$

$$+ [i\hbar(\omega_{jk} + i0)^2]^{-1} \Big\{ \sum_{\gamma} \varepsilon_{j,\alpha\gamma}^{(1)} \langle \phi_{j,\gamma} | P^{(1)} | \phi_{k,\beta} \rangle - \sum_{\gamma} \langle \phi_{j,\alpha} | P^{(1)} | \phi_{k,\gamma} \rangle \varepsilon_{k,\gamma\beta}^{(1)} \Big\}. \tag{4.15}$$

According to our notation convention, if both (j, α) and (k, β) are scattering states, then those terms involving the ε's in (4.15) vanish identically. Once again, we will not evaluate $M_{j\alpha,j\beta}^{(2)}$ for bound states. It can be derived by using (3.20) and relevant definitions.

From (4.12)–(4.15) and our previous knowledge for the non-degenerate case, we can immediately write down the first- and second-order net transition rates for bound states if the external field is a single-frequency field.

4.4. Perturbative rates of transition

For simplicity of manipulations, we consider an external field of the form

$$\boldsymbol{E}(\boldsymbol{r}, t) = \boldsymbol{E}_+(\boldsymbol{r})\, e^{i\omega t} + \boldsymbol{E}_-(\boldsymbol{r})\, e^{-i\omega t}, \qquad (\boldsymbol{E}_+)^* = \boldsymbol{E}_-, \tag{4.16}$$

and similarity for $\boldsymbol{B}(\boldsymbol{r}, t)$ where * denotes the complex conjugate. Since we have already shown that the gauge-invariant procedure is capable of eliminating the arbitrariness in the potentials, we may now choose the most convenient set of potentials of the form

$$\boldsymbol{A}(\boldsymbol{r}, t) = \boldsymbol{A}_+(\boldsymbol{r})\, e^{i\omega t} + \boldsymbol{A}_-(\boldsymbol{r})\, e^{-i\omega t}, \qquad \Phi(\boldsymbol{r}, t) = 0, \tag{4.17}$$

with

$$(\boldsymbol{A}_+)^* = \boldsymbol{A}_- \qquad \text{and} \qquad \boldsymbol{A}_\pm = \mp(c/i\omega)\boldsymbol{E}_\pm. \tag{4.18}$$

By our previous convention of using the subscripts $\pm$ and ± 2 (I), we have

$$P^{(1)}_\pm = e(\boldsymbol{E}_\pm \cdot \boldsymbol{p} + \boldsymbol{p} \cdot \boldsymbol{E}_\pm)/2m, \qquad P^{(2)}_{\pm 2} = -e^2 \boldsymbol{A}_\pm \cdot \boldsymbol{E}_\pm / mc = \pm(e\boldsymbol{E}_\pm)^2/im\omega, \tag{4.19}$$

$$V_{1,\pm} = -e(\boldsymbol{A}_\pm \cdot \boldsymbol{p} + \boldsymbol{p} \cdot \boldsymbol{A}_\pm)/2mc = \pm(i\omega)^{-1} P^{(1)}_\pm, \tag{4.20}$$

$$V_{2,\pm 2} = (e\boldsymbol{A}_\pm)^2/2mc^2 = \mp(i/2\omega) P^{(2)}_{\pm 2}, \tag{4.21}$$

where V_1 and V_2 are listed in (3.2). Similarly, we will decompose the matrix elements $\{M^{(n)}_{j\alpha,k\beta}\}$ according to their frequency dependence as in I.

From (4.5), (4.6), (4.10) and the above definitions, the first-order net transition rates are

$$T^{(1)}_{fI,\pm} = (2\pi/\hbar^2) \sum_\beta |N^{(1)}_{f\beta,I;\pm}|^2 \delta(\omega_{fi} \pm \omega), \tag{4.22}$$

where $I = (i, \alpha_i)$ refers to the initial state and

$$N^{(1)}_{f\beta,I;\pm} = \sum_{k,\gamma} M^{(1)}_{f\beta,k\gamma;\pm} b^{(0)}_{k,\gamma}(0), \tag{4.23}$$

where $\{b^{(0)}_{k,\gamma}(0)\}$ are the initial expansion coefficients that are determined by the initial wavefunction $\Psi(\boldsymbol{r}, 0)$. If the initial state I is a *bound* state, then

$$N^{(1)}_{f\beta,I;\pm} = M^{(1)}_{f\beta,I;\pm} = (i/\omega_{fi}) \langle \phi_{f,\beta} | P^{(1)}_\pm | \phi_I \rangle. \tag{4.24a}$$

However, if I refers to a scattering state, then it can be shown that (4.23) leads to

$$N^{(1)}_{f\beta,I;\pm} = (i/\omega_{fi}) \langle \phi_{f,\beta} | P^{(1)}_\pm | \phi^{(+)}_I \rangle, \tag{4.24b}$$

where $\phi^{(+)}_I$ is an outgoing state with incident momentum $\hbar\boldsymbol{I} = (\hbar i, \alpha_i)$. In either case, we may write the two expressions in (4.24*a*) and (4.24*b*) in terms of the initial wavefunction $\Psi(\boldsymbol{r}, 0)$. Henceforth, this argument will be implicitly assumed in our later results.

Similarly, one can show that the second-order rates of transition for the double-frequency excitation and de-excitation are

$$T^{(2)}_{fI,\pm 2} = (2\pi/\hbar^2) \sum_\beta |N^{(2)}_{f\beta,I;\pm 2}|^2 \delta(\omega_{fi} \pm 2\omega), \tag{4.25}$$

where†

$$N^{(2)}_{f\beta,I;\pm 2} = \sum_{k,\gamma}{}'' P^{(1)}_{\pm;f\beta,k\gamma}[\hbar(\mp\omega_{fi}/2)\omega(\omega_{ki}\pm\omega)]^{-1}P^{(1)}_{\pm;k\gamma,I}$$
$$+(2/\mathrm{i}\hbar\omega_{fi})\Big\{(\omega_{fi}\pm\omega)^{-1}\sum_{\gamma}\varepsilon^{(1)}_{f,\beta\gamma;\pm}P^{(1)}_{\pm;f\gamma,I}+(\pm\omega)^{-1}\sum_{\gamma}P^{(1)}_{\pm;f\beta,i\gamma}\varepsilon^{(1)}_{i,\gamma\alpha_i;\pm}\Big\}$$
$$+(\mathrm{i}/\omega_{fi})P^{(2)}_{\pm 2;f\beta,I}+(\omega_{fi}\pm 2\omega)\Delta^{(2)}_{f\beta,I;\pm 2}, \tag{4.26}$$

$$\Delta^{(2)}_{f\beta,I;\pm 2} = \sum_{k,\gamma}{}'' P^{(1)}_{\pm;f\beta,k\gamma}[\hbar\omega\omega_{fi}\omega_{fk}(\omega_{ki}\pm\omega)]^{-1}P^{(1)}_{\pm;k\gamma,I}$$
$$+(\mathrm{i}\hbar\omega_{fi}^2)^{-1}\Big\{(\omega_{fi}\pm\omega)^{-1}\sum_{\gamma}\varepsilon^{(1)}_{f,\beta\gamma;\pm}P^{(1)}_{\pm;f\gamma,I}-(\pm\omega)^{-1}\sum_{\gamma}P^{(1)}_{\pm;f\beta,i\gamma}\varepsilon^{(1)}_{i,\gamma\alpha_i;\pm}\Big\}$$
$$+[\mathrm{i}\hbar\omega_{fi}(\pm\omega)(\omega_{fi}\pm\omega)]^{-1}\sum_{\gamma}\eta^{(1)}_{f,\beta\gamma;\pm}P^{(1)}_{\pm;f\gamma,I}. \tag{4.27}$$

Here, the double prime in the summation means excluding only the *bound-state* subspace(s) of the initial and the final states. For example, if I refers to a scattering state whereas (f,β) is a bound state, then the summation over (k,γ) excludes only those bound states (k,γ) with $k=f$. The matrix elements involving the first- and second-order power operators are defined by

$$P^{(1)}_{\pm;f\beta,k\gamma} = \langle\phi_{f,\beta}|P^{(1)}_{\pm}|\phi_{k,\gamma}\rangle, \qquad P^{(1)}_{\pm;k\gamma,I} = \langle\phi_{k,\gamma}|P^{(1)}_{\pm}|\Psi(0)\rangle,$$
$$P^{(2)}_{\pm 2;f\beta,I} = \langle\phi_{f,\beta}|P^{(2)}_{\pm 2}|\Psi(0)\rangle,$$

where we note that the matrix elements with subscript I are evaluated using the initial wavefunction $\Psi(\boldsymbol{r},0)$ as explained in connection with equations (4.24). We also note that, according to our notation convention, $\varepsilon^{(1)}_{i,\gamma\alpha_i;\pm}=0$ if $I=(i,\alpha_i)$ is a scattering state. Finally, the η's are defined in (4.14). This formally concludes our formulation of the TDPT for a degenerate system.

5. Conclusions

In this series of two papers, we have formulated a manifestly gauge-invariant TDPT for a non-relativistic, spinless charged particle interacting with classical, external electromagnetic fields. We have shown that the gauge-invariant first- and second-order perturbative rates of transition agree with their conventional counterparts. But, our transition amplitudes and probabilities differ from the conventional ones‡. As has

† There was a missing ω in the first term on the right-hand side of equation (4.24) for the non-degenerate case (Yang 1982c). This term should read

$$\sum_{k}{}'' P^{(1)}_{\pm,fk}[\hbar(\mp\omega_{fi}/2)\omega(\omega_{ki}\pm\omega)]^{-1}P^{(1)}_{\pm,ki}.$$

‡ When translated into conventional language, these two statements read as follows: 'The gauge-invariant and the conventional first- and second-order amplitudes agree on-the-energy-shell but disagree off-the-energy-shell.' We will by all means avoid using this conventional language since it is one major source of confusion on the subject discussed. For a very detailed explanation of the relationship between gauge transformations and the quantum mechanical interpretation of transition probabilities, see Leubner and Zoller (1980) and Lee and Albrecht (1983).

been shown by Leubner and Zoller (1980) and Lee and Albrecht (1983), it is precisely this difference in the transition amplitudes and probabilities that has enabled the gauge-invariant formulation to resolve important conceptual and practical difficulties in the conventional interpretation of quantum mechanics.

One important purpose of this series of two papers on the TDPT is to demonstrate one simple fact: that the approximations used in solving the differential equations for the probability amplitudes, if these equations are not initially manifestly gauge-invariant, can alter the gauge properties of some particular—but not all—solutions. Shirokov (1981) has also observed this conclusion in his investigations. Thus, the investigations of this paper and Yang (1982a, b, c), of Kobe and Yang (1982) and Lee and Albrecht (1983) explain why there has been such a confusion over whether the conventional interpretation of the quantum mechanical probabilities and amplitudes is gauge invariant. (For a list of references, see Yang (1982a).)

However, the most important purpose of our investigation of the gauge-invariant formulation is to try to understand the 'hidden issues' in the 'controversy' of $\boldsymbol{A}\cdot\boldsymbol{p}$ versus $\boldsymbol{r}\cdot\boldsymbol{E}$, first raised by Lamb (1952). He observed that his experimental data favoured $\boldsymbol{r}\cdot\boldsymbol{E}$ as the 'interaction Hamiltonian' in a *non*-perturbative calculation involving the two-state approximation, the rotating-wave approximation and the decay constants added phenomenologically to simulate the effects of the spontaneous emissions. Let us now examine the advantages and the disadvantages of these two forms of 'interaction Hamiltonian' as the transition operator.

The main advantage of $\boldsymbol{r}\cdot\boldsymbol{E}$ as a transition operator is that it involves explicitly the electric field which is directly measurable by classical electromagnetic theory. The main disadvantage lies in its explicit dependence on the position operator $\boldsymbol{r}$. Because of this explicit $\boldsymbol{r}$ dependence, the matrix element $\langle\phi_m|-e\boldsymbol{r}\cdot\boldsymbol{E}(\boldsymbol{0},t)|\phi_n\rangle$ can be physically interpreted only when at least one of ϕ_m and ϕ_n is a bound state (i.e. a localised state). If both states ϕ_m and ϕ_n are in the continuum, such a matrix element loses its physical significance since it is rather difficult to imagine the 'significance' of the electric dipole of a system extending to infinity (in space). To illustrate this difficulty in the multi-photon processes, let us consider a third-order resonant transition between two states ϕ_i and ϕ_f of a realistic system arising from the interaction with a single-frequency external radiation field of angular frequency ω. According to the conventional TDPT using $\boldsymbol{r}\cdot\boldsymbol{E}$ as the 'interaction Hamiltonian,' the third-order transition amplitude will have a term of the form

$$\sum_{m,n}\langle\phi_f|-e\boldsymbol{r}\cdot\boldsymbol{E}|\phi_m\rangle\langle\phi_m|-e\boldsymbol{r}\cdot\boldsymbol{E}|\phi_n\rangle\langle\phi_n|-e\boldsymbol{r}\cdot\boldsymbol{E}|\phi_i\rangle[(\omega_{mi}\pm 2\omega)(\omega_{ni}\pm\omega)]^{-1}.$$

Here, m and n must run through the complete set, including the continuum. It is here when both m and n are the states in the continuum that, we believe, the $\boldsymbol{r}\cdot\boldsymbol{E}$ form as a transition operator is less appropriate.

We now examine also the advantages and the disadvantages of $\boldsymbol{A}\cdot\boldsymbol{p}$ as a transition operator. The main advantage of this form is that it is explicitly independent of the position operator $\boldsymbol{r}$ and, since $\boldsymbol{p}=-i\hbar\boldsymbol{\nabla}$, any displacement in the origin of the coordinate system still results in the same gradient operator. The main disadvantage of this form is that it involves explicitly the vector potential $\boldsymbol{A}$ which, by classical electromagnetic theory, cannot be physically measured. However, if we ignore this explicit dependence upon the unphysical potentials, $\boldsymbol{A}\cdot\boldsymbol{p}$ is actually more appropriate for the matrix elements involving the states in the continuum because of the presence of $\boldsymbol{p}$.

We now summarise the above discussions. Both $\boldsymbol{r}\cdot\boldsymbol{E}$ and $\boldsymbol{A}\cdot\boldsymbol{p}$ have nice properties and conceptual defects. Hence either cannot be 'the true transition operator.' If we combine the nice properties of both operators, we would get $\boldsymbol{p}\cdot\boldsymbol{E}$. However, $\boldsymbol{p}$ is not gauge invariant (e.g. Cohen-Tannoudji *et al* 1977). A more appropriate form for the exact transition operator should take the form $[e(\boldsymbol{p}-e\boldsymbol{A}/c)\cdot\boldsymbol{E}+\boldsymbol{E}\cdot e(\boldsymbol{p}-e\boldsymbol{A}/c)]/2m$. Such an operator is both gauge invariant and explicitly dependent upon $\boldsymbol{E}$. A comparison of this operator with $\boldsymbol{p}\cdot\boldsymbol{E}$ also indicates that $\boldsymbol{p}\cdot\boldsymbol{E}$ is a valid first-order transition operator in the perturbation theory (only).

Having arrived at the operator $P=\frac{1}{2}(e\boldsymbol{v}\cdot\boldsymbol{E}+\boldsymbol{E}\cdot e\boldsymbol{v})$ where $\boldsymbol{v}=(\boldsymbol{p}-e\boldsymbol{A}/c)/m$, we then realise that the arguments by which this operator is derived are phenomenological in nature and that a theoretical foundation is lacking. A quick check through any textbook on classical electromagnetic theory at once provides us with a sound foundation: Poynting's theorem and its interpretation as a conservation law of energy (e.g. Jackson 1975). The connection between the classical Poynting theorem and the necessary quantum mechanical formulation is then made by invoking the correspondence principle (Bohr 1928) through the equations of motion of operators. The use of Poynting's theorem is especially important when dealing with spinning particles since the Foldy–Wouthuysen (1950) transformations are involved and the consistency between the non-relativistic and the relativistic (Dirac) probability amplitudes and probabilities becomes a major concern (Yang 1977a, 1982a, Kobe and Yang 1980, Yang and Hirschfelder 1980, Yang *et al* 1981, Hirschfelder *et al* 1982).

Appendix. Proofs of gauge invariance

In this appendix, we shall provide a simple proof of the gauge invariance of $\varepsilon^{(1)}_{j,\alpha\beta}$, $D_{j,\alpha\beta}$ and $E^{(1)}_{j,\alpha}$ discussed in connection with §§ 3.1 and 3.3, especially with (3.11). Let us assume that we have another set of potentials $(\boldsymbol{A}', \Phi')$ that are related to $\boldsymbol{A}$ and Φ by

$$\boldsymbol{A}'=\boldsymbol{A}+\boldsymbol{\nabla}\chi \qquad \text{and} \qquad \Phi'=\Phi-c^{-1}\partial\chi/\partial t. \tag{A1}$$

In this gauge, the energy operator H'_{B} is

$$H'_{\mathrm{B}}=(\boldsymbol{p}-e\boldsymbol{A}'/c)^2/2m+eV_0. \tag{A2}$$

Parallelling (3.1), we write $H'_{\mathrm{B}}=H_0+V'_1+V'_2$, where $H_0=\boldsymbol{p}^2/2m+eV_0$ is the 'unperturbed Hamiltonian', and

$$V'_1=-e(\boldsymbol{p}\cdot\boldsymbol{A}'+\boldsymbol{A}'\cdot\boldsymbol{p})/2mc, \qquad V'_2=(e\boldsymbol{A}')^2/2mc^2. \tag{A3}$$

We note that V'_1 has the same form as V_1 in (3.2), and V'_2 the same form as V_2. This is a basic requirement in the proof of the gauge invariance of the quantities $\{\varepsilon^{(1)}_{j,\alpha\beta}\}$ and so on.

The gauge invariance of $\{\varepsilon^{(1)}_{j,\alpha\beta}\}$ follows from the equality

$$\langle\phi_{j,\alpha}|V'_1|\phi_{j,\beta}\rangle=\langle\phi_{j,\alpha}|V_1|\phi_{j,\beta}\rangle, \tag{A4}$$

which is a mathematical consequence of the relation

$$V'_1=V_1+(ie/c\hbar)[\chi, H_0]. \tag{A5}$$

Note, (A4) implies the gauge invariance of the quantities involved because the left-hand side has the same form and same value as the right-hand side. The gauge invariance

of all higher-order quanties $\{\varepsilon^{(n)}_{j,\alpha\beta}\}$ in § 3.2 can be shown using a procedure similar to the one used for the non-degenerate case (e.g. Yang 1977b). Equation (A5) also implies that $\langle\phi_{j,\alpha}|V_1|\phi_{j,\beta}\rangle = 0$ if $\nabla\times\boldsymbol{A} = 0$.

Because the matrix $(D_{j,\alpha\beta})$ diagonalises the matrix $(\varepsilon^{(1)}_{j,\alpha\beta})$ (assuming V_1 breaks up the degeneracy in H_0), and because the eigenvalue corrections $\{E^{(1)}_{j,\alpha}\}$ are the matrix elements in the resultant diagonal matrix, it follows that each $D_{j,\alpha\beta}$ and $E^{(1)}_{j,\alpha}$ is gauge invariant. However, since the values of $\{E^{(1)}_{j,\alpha}\}$ are determined by solving the secular equation,

$$\det(\lambda\delta_{\alpha\beta} - \varepsilon^{(1)}_{j,\alpha\beta}) = 0, \tag{A6}$$

where det denotes the determinant, it follows that $E^{(1)}_{j,\alpha}$ will in general be a complicated function of the magnetic field and will not be linear in the fields when degeneracy is involved.

As an example, let us consider a particular j with two degenerate states $(j, 1)$ and $(j, 2)$. By (A6), the RS first-order eigenenergies $E^{(1)}_{j,1}$ and $E^{(1)}_{j,2}$ will assume the values

$$\lambda_\pm = \tfrac{1}{2}\{(\varepsilon^{(1)}_{j,11} + \varepsilon^{(1)}_{j,22}) \pm [(\varepsilon^{(1)}_{j,11} - \varepsilon^{(1)}_{j,22})^2 + 4|\varepsilon^{(1)}_{j,12}|^2]^{1/2}\}. \tag{A7}$$

Thus, in general $E^{(1)}_{j,1}$ and $E^{(1)}_{j,2}$ will differ from $\varepsilon^{(1)}_{j,11}$ and $\varepsilon^{(1)}_{j,22}$ unless $\nabla\times\boldsymbol{A} = 0$ or $[V_1, H_0] = 0$. This means that the matrix $(D_{j,\alpha\beta})$ cannot in general be chosen to be $D_{j,\alpha\beta} = \delta_{\alpha\beta}$. Furthermore, (A7) also indicates that $(D_{j,\alpha\beta})$ will in general be both time and magnetic field dependent.

Let us now discuss some special field (or potential) situations of particular interest. If $\nabla\times\boldsymbol{A} = 0$, which corresponds to no magnetic field regardless of whether the electric field is present, then it can be shown that $E^{(n)}_{j,\alpha} = 0$ or $\varepsilon^{(n)}_{j,\alpha\beta} = 0$ for all $n \geq 1$ (e.g. Yang 1977b). Under this situation, we set $D_{j,\alpha\beta} = \delta_{\alpha\beta}$. The consequence is that the coefficient $\langle u_{j,\alpha}|\Psi(t)\rangle$, where $u_{j,\alpha}$ is defined in (3.12) and $\Psi(r, t)$ is the wavefunction, can be interpreted as the physically meaningful probability amplitude for finding the particle in energy $\hbar\omega_j$. This simply follows from (3.12). A better discussion on this point is presented by Leubner and Zoller (1980).

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