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Continued fraction perturbation theory: applications to radiative processes in the dipole approximation

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Abstract. A general perturbation procedure is described for writing down the contributions to any order for the probability of a quantum system described by a time-independent Hamiltonian making a transition from an arbitrary initial state to an arbitrary final state. The results are expressed in terms of certain propagators which involve continued fractions, so that some processes are summed to all orders. The general theory is then applied to a single multilevel atom interacting with any number of modes of the electromagnetic field in the electric dipole approximation. This work may be regarded as a generalization of earlier treatments of the corresponding single-mode, two-level atom problem involving the continued fraction approach. We are mainly concerned with the fundamental theory here, but a detailed discussion is given of spontaneous and stimulated emission. Expressions for the lineshape are derived incorporating the generalized Lamb shifts and damping constants.

1. Introduction

A very common model for describing the properties of atomic systems interacting with electromagnetic fields is that in which the coupling is taken to be electric dipole (Power and Zienau 1959), and this is the situation we are concerned with here. We first develop an improved form of perturbation theory for calculating the probability of transitions between two arbitrary states of a system whose Hamiltonian may be decomposed into a perturbed and unperturbed part. We describe rules for calculating the transition probability to any order. This general theory is then specialized to deal with the interacting single atom/electromagnetic field problem. Our method is applicable to atomic systems with any number of energy levels, and to any number of electromagnetic field modes. The treatment is non-relativistic but fully quantum electrodynamical. No approximations beyond the electric dipole approximation are introduced.

In two previous publications (Swain 1973a, to be referred to as I, 1973b) we have derived continued fraction solutions to the problem of a single two-level atom interacting with a single quantized field mode in the dipole approximation. The semi-classical version of the continued fraction approach allows an elegant treatment of various problems in radio frequency spectroscopy (see for example Autler and Townes 1955, Stenholm 1972, Tsukada and Ogawa 1973, Stenholm and Aminoff 1973, Swain 1974). The connection between the semi-classical and quantum electrodynamical solutions has been discussed by Swain (1973c). In view of its usefulness, it is natural to enquire whether the continued fraction approach for a single mode and two atomic levels can be generalized to deal with any number of modes and atomic levels. The present work

had its source in this consideration. Although we are mainly concerned with establishing the fundamental method here, by way of illustration we give an account of stimulated and spontaneous emission in § 5. This is perhaps the most fundamental of radiative phenomena, and it is a first consideration to see how our method deals with it.

The system we consider may be described by the Hamiltonian

$$H = H_A + H_F + H_{AF} \quad (1a)$$

where

$$H_A = \sum_i |i\rangle E_i \langle i| \quad (1b)$$

$$H_F = \sum_\lambda a_\lambda^\dagger a_\lambda \omega_\lambda \quad (1c)$$

$$H_{AF} = \sum_\lambda \sum_i \sum_j g_{ij}^\lambda |i\rangle \langle j| (a_\lambda - a_\lambda^\dagger). \quad (1d)$$

Here H_A is the Hamiltonian for the isolated atom which has eigenstates $|i\rangle$ with energies E_i , and H_F is the Hamiltonian for the free field with the zero-point energy omitted. a_λ and a_λ^\dagger are the usual Bose annihilation and creation operators for photons of mode λ and frequency ω_λ . (We understand λ to include the mode wavevector \mathbf{k} and the polarization index σ : $\lambda \equiv \mathbf{k}, \sigma$.) We use rationalized MKS units, but for simplicity we set $\hbar = 1$ throughout. E_i and ω_λ thus have the same dimensions. H_{AF} describes the interaction between the atom and the field in the electric dipole approximation. The coupling constant, g_{ij}^λ , has the value

$$g_{ij}^\lambda = i \left(\frac{\omega_\lambda}{2\epsilon_0 V} \right)^{1/2} \mu_{ij} \cos \theta_\lambda = -(g_{ji}^\lambda)^* \quad (2a)$$

where θ_λ is the angle between the atomic dipole moment $\boldsymbol{\mu}$ and the electric vector of the λ th mode, and V is the volume of the system.

Other systems of physical interest, for example an isolated paramagnetic ion interacting with phonons in a crystal, or an atom situated in a DC magnetic field with an oscillating magnetic field at right angles to it, may be described by the same Hamiltonian (1), but with g_{ij}^λ having a different form to that given in (2). It is only in § 5 that we use the explicit form for g_{ij}^λ , so the previous analysis applies equally well to such situations.

In the examples we give we will assume

$$g_{jj}^\lambda = 0, \quad \text{all } j \quad (2b)$$

ie, that g_{ij}^λ is strictly off-diagonal, but we only do this to simplify the expressions obtained; it is not essential.

The first term of (1d) describes processes in which a photon of mode λ is absorbed, and the second those in which such a photon is emitted. In each case the atom changes its state from $|i\rangle$ to $|j\rangle$. These processes, and the associated matrix elements, are shown in figure 1 with the corresponding matrix elements. (Only photons which are emitted or absorbed are shown in the diagrams, and we assume that there are n_λ photons in the mode λ initially.)

According to Power and Zienau (1959), (1d) is the lowest approximation to the interaction between the atoms and the EM field; additional terms representing magnetic dipole, electric quadrupole, and higher multipole contributions should be added.

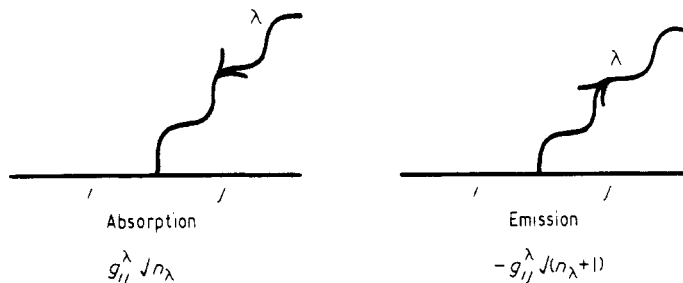


Figure 1. The primary processes of the electric dipole interaction Hamiltonian: the transitions $i_{\underline{n}} \rightarrow j_{\underline{n}-\lambda}$ (absorption) and $i_{\underline{n}} \rightarrow j_{\underline{n}\lambda}$ (emission). Here $\underline{n} \pm \lambda \equiv n_1, n_2, \dots, n_{\lambda} \pm 1, \dots, n_N$. The corresponding matrix elements are indicated on the diagrams.

Furthermore, there is the term

$$H_P = \frac{1}{2\epsilon_0} \int d^3x |\mathbf{P}(\mathbf{x})|^2 \tag{3a}$$

where $\mathbf{P}(\mathbf{x})$ is the polarization vector operator of the atom. Using the approach of Power and Zienau (§ 4.2) this may be written

$$H_P = \frac{1}{6\pi^2\epsilon_0} \int k^2 dk \sum_i \sum_j \langle i | \boldsymbol{\mu}^2 | j \rangle |i\rangle \langle j|. \tag{3b}$$

The integral over k in (3b) extends from 0 to ∞ , so that it diverges (this is just one of the divergences of quantum electrodynamics). However, for the moment we work with it as though it were a finite quantity. We may split H_P into diagonal and off-diagonal parts:

$$H_P = H_P^{(d)} + H_P^{(od)}$$

where

$$H_P^{(d)} \equiv \frac{1}{6\pi^2\epsilon_0} \int k^2 dk \sum_i \sum_j \boldsymbol{\mu}_{ij} \boldsymbol{\mu}_{ji} |i\rangle \langle i| \tag{3c}$$

$$H_P^{(od)} \equiv \frac{1}{6\pi^2\epsilon_0} \int k^2 dk \sum_i \sum_j \langle i | \boldsymbol{\mu}^2 | j \rangle |i\rangle \langle j| (1 - \delta_{ij}). \tag{3d}$$

The important term is (3c): this may be written

$$H_P^{(d)} = \sum_i |i\rangle \delta E_i \langle i| \tag{3e}$$

where

$$\delta E_i = \sum_j \frac{|\boldsymbol{\mu}_{ij}|^2}{6\pi^2\epsilon_0} \int k^2 dk. \tag{3f}$$

On comparing (1b) and (3f) it is clear that $H_P^{(d)}$ just shifts each level E_i by the (divergent) amount δE_i . We return to a discussion of $H_P^{(d)}$ in § 5, and for simplicity we neglect $H_P^{(od)}$, although we describe how to account for it at the end of § 3.

In § 2 we derive the fundamental equations governing the transition probabilities, and in § 3 we show how these equations may be solved. In the latter section we also

summarize the rules for calculating the transition probabilities to any order. In § 4 we show how the previously derived results for the two-level atom, single-mode problem may be obtained from this formalism, and finally, in § 5 we show how the fundamental phenomena of stimulated and spontaneous emission may be treated by our method.

2. Form of the general solutions

The approach we use is the generalized Fourier/Laplace transform technique described by Pike and Swain (1971) (see also I). We wish to calculate quantities of the type $P_i^f(t)$, the probability that the system will be in state $|f\rangle$ at time t if it was in state $|i\rangle$ at time 0. To do this we first calculate the generalized Laplace transform, $L_i^f(E)$ of the matrix element of the time development operator, $\langle i | \exp(iHt) | f \rangle$, and then obtain $P_i^f(t)$ from the inverse transform

$$P_i^f(t) = \left| \frac{1}{2\pi i} \oint dE L_i^f(E) e^{-iEt} \right|^2. \quad (4)$$

Here E is the Laplace variable, and the contour lies above the real axis. It is easy to see that $L_i^f(E)$ is given explicitly by the relation

$$L_i^f(E) = \sum_{\xi} \frac{\langle i | \xi \rangle \langle \xi | f \rangle}{E - E_{\xi}} \quad (5)$$

where $|\xi\rangle$ and E_{ξ} are exact eigenvectors and eigenvalues of the complete Hamiltonian. Clearly $L_i^f(E)$ is a matrix element of the resolvent operator (Goldberger and Watson 1964)

$$G(E) = (E - H)^{-1}. \quad (6)$$

It follows from the definition (5) that $L_i^f(E)$ obeys the equation of motion

$$EL_i^f(E) = \langle i | f \rangle + L_{Hf}^f(E) \quad (7)$$

where the suffix Hf is shorthand for the state $H|f\rangle$. This equation is somewhat similar to that obeyed by the Green functions of many-body theory. The connection between the present approach and the traditional Green function method is discussed by Pike and Swain (1971).

The fact that the states i and f are arbitrary complicates the argument, and we find it convenient to express $L_i^f(E)$ in terms of the functions $L_b^a(E)$, where the states $|a\rangle, |b\rangle, |c\rangle, \dots$ form a complete orthonormal set: $\sum_a |a\rangle \langle a| = 1$. Introducing two such complete sets into the definition (5) for $L_i^f(E)$ we obtain

$$L_i^f(E) = \sum_a \sum_b \langle i | a \rangle \left(\sum_{\xi} \frac{\langle a | \xi \rangle \langle \xi | b \rangle}{E - E_{\xi}} \right) \langle b | f \rangle \quad (8)$$

$$= \sum_a \sum_b \langle i | a \rangle \langle b | f \rangle L_b^a(E) \quad (9)$$

where we have interchanged the order of summation, and then noted that the expression in large parentheses in (8) is just the definition of $L_b^a(E)$. Thus instead of calculating $L_i^f(E)$ directly, we may instead first find $L_b^a(E)$, and then use (9) to obtain $L_i^f(E)$. The advantage of this approach is that $L_b^a(E)$ has a particularly simple equation of motion, because $\langle a | b \rangle = \delta_{a,b}$.

Now we further assume that the Hamiltonian may be decomposed into a perturbed and an unperturbed part,

$$H = H_0 + V \quad (10)$$

and that the states $|a\rangle, |b\rangle, \dots$ are eigenvectors of the unperturbed Hamiltonian H_0 .

$$H_0|a\rangle = W_a|a\rangle. \quad (11)$$

From the definition (5) we have

$$L_{Hb}^a(E) = \sum_c \sum_{\xi} \frac{\langle a|\xi\rangle \langle \xi|c\rangle \langle c|H|b\rangle}{E - E_{\xi}}$$

and from (10) and (11)

$$\langle c|H|b\rangle = W_b\delta_{c,b} + V_{cb},$$

where $V_{cb} \equiv \langle c|V|b\rangle$. Hence we easily obtain

$$L_{Hb}^a(E) = W_b L_b^a + \sum_c V_{cb} L_c^a. \quad (12)$$

Using (12) in the equation of motion (7) for $L_b^a(E)$ leads to the modified equation of motion

$$(E - W_b)L_b^a(E) = \delta_{a,b} + \sum_c V_{cb} L_c^a(E)$$

or

$$(E - W_b - V_{bb})L_b^a(E) = \delta_{a,b} + \sum_{c \neq b} V_{cb} L_c^a. \quad (13)$$

For a fixed value of a , equation (13) defines in general an infinite set of coupled equations for the $L_j^a(E)$, $j = a, b, c, \dots$. However, this set is particularly simple in that only when $b = a$ is there a constant term on the right-hand side, and then this is unity. Thus the equations for the $L_j^a(E)$, $j \neq a$ form a homogeneous set; one may therefore solve this set for the $L_j^a(E)$ in terms of $L_a^a(E)$, then substitute these expressions for the $L_j^a(E)$ into equation (13) for $b = a$. This determines $L_a^a(E)$. We shall illustrate this procedure shortly. Equation (13) is the most convenient for developing the perturbation series, and we shall refer to it as the fundamental equation. We note that only off-diagonal elements of V appear on the right-hand side.

Finally we note the symmetry relation

$$L_y^x(E) = (L_x^y(E))^* \quad (14a)$$

which follows trivially from the definition (5). It implies the property

$$P_y^x(t) = P_x^y(t), \quad (14b)$$

so that mathematically it does not matter which state we take to be the initial state and which the final one.

3. Solution of the fundamental equations

In order to develop the perturbation series it is convenient to change our notation somewhat. We now use A to label the initial state, B to label the set of all states,

B_1, B_2, B_3, \dots for which $V_{B_i,A} \neq 0$ but excluding the initial state A , C the set C_1, C_2, C_3, \dots for which $V_{C_j,B_i} \neq 0$, but excluding A and any member of the set B , D the set D_1, D_2, D_3, \dots for which $V_{D_k,C_j} \neq 0$ but excluding A and any members of the sets B and C , and so on. Thus the states B_i are connected to state A in the first order of perturbation theory, the states C_j in second order (via the states B_i), etc.

With this notation the first few members of the set may be written

$$(E - \tilde{W}_A)L_A^A - \sum_B L_B^A V_{B,A} = 1 \tag{15a}$$

$$(E - \tilde{W}_B)L_B^A - L_A^A V_{A,B} - \sum_{B' \neq B} L_{B'}^A V_{B',B} - \sum_C L_C^A V_{C,B} = 0 \tag{15b}$$

$$(E - \tilde{W}_C)L_C^A - \sum_B L_B^A V_{B,C} - \sum_{C' \neq C} L_{C'}^A C_{C',C} - \sum_D L_D^A V_{D,C} = 0 \tag{15c}$$

$$(E - \tilde{W}_D)L_D^A - \sum_C L_C^A V_{C,D} - \sum_{D' \neq D} L_{D'}^A V_{D',D} - \sum_E L_E^A V_{E,D} = 0 \quad \text{etc} \tag{15d}$$

where $\tilde{W}_J \equiv W_J + V_{J,J}$ and for clarity we have not displayed the E dependence of the L functions explicitly.

Let S be the determinant of the system of equations (15). Then according to standard theory (Margenau and Murphy 1956) the solution is

$$L_N^A = \frac{S^{A,N}}{S} \tag{16}$$

where S^{IJ} is a cofactor of S . The determinant S may be expanded in terms of its elements S_{IJ} and its cofactors as

$$S = \sum_I S_{KI} S^{KI} \tag{17}$$

where the sum over I extends over all the unperturbed eigenstates A, B, C, D, \dots and K may be any one of these states.

Choosing $K = A$ in (17) we obtain

$$S = (E - \tilde{W}_A)S^{A,A} - \sum_B V_{B,A} S^{A,B}. \tag{18}$$

We may expand $S^{A,B}$ using an expression similar to (17) but now we choose $K = B$. Then (18) may be written

$$S = (E - \tilde{W}_A)S^{A,A} - \sum_B \left(V_{AB} V_{B,A} S^{A,B,BA} + \sum_{B' \neq B} V_{B'B} V_{B,A} S^{A,B,BB'} + \sum_C V_{CB} V_{B,A} S^{A,B,BC} \right) \tag{19}$$

where $S^{A,B,IJ}$ is a cofactor of $S^{A,B}$. The final two terms in (19) may also be expanded using (17), the object being to choose K in each case so that only diagonal cofactors tend to appear. The process may be repeated indefinitely so that an infinite series is generated. By diagonal cofactors, we mean those of the form $S^{IJ,JK,KL,LI}$, which in fact is the determinant obtained from S by striking out the I th, J th, K th, and L th rows and columns. As the order in which the rows and columns are struck out is of no significance, we could have written this as $S^{II,JJ,KK,LL}$. To simplify the notation it is convenient to write these diagonal cofactors as

$$S^{II,JJ,KK,LL} \equiv \varphi^{IJKL}. \tag{20}$$

We may now write the expansion for S as

$$S = (E - \tilde{W}_A) \mathcal{S}^A - \sum_B V_{AB} V_{BA} \mathcal{S}^{AB} - \sum_{B' \neq B} \sum_B V_{AB'} V_{B'B} V_{BA} \mathcal{S}^{AB'B} - \sum_C \sum_{B' \neq B} \sum_B V_{AB'} V_{B'C} V_{CB} V_{BA} \mathcal{S}^{AB'CB} - \dots \quad (21)$$

and thus, from (16), we have

$$(L_A^A)^{-1} \equiv \frac{S}{\mathcal{S}^A} = E - \tilde{W}_A - \sum_B V_{AB} V_{BA} \frac{\mathcal{S}^{AB}}{\mathcal{S}^A} - \sum_{B' \neq B} \sum_B V_{AB'} V_{B'B} V_{BA} \frac{\mathcal{S}^{AB'B}}{\mathcal{S}^A} - \dots \quad (22)$$

If we introduce the notation

$$\mathcal{D}_K^{AB\dots IJ} \equiv \frac{\mathcal{S}^{AB\dots IJ}}{\mathcal{S}^{AB\dots IJK}}, \quad \mathcal{D}^A = \frac{S}{\mathcal{S}^A} \quad (23)$$

expression (22) may be written

$$(L_A^A)^{-1} \equiv \mathcal{D}_A = E - \tilde{W}_A - \sum_B \frac{V_{AB} V_{BA}}{\mathcal{D}_B^A} - \sum_{B' \neq B} \sum_B \frac{V_{AB'} V_{B'B} V_{BA}}{\mathcal{D}_B^A \mathcal{D}_B^{AB'}} - \dots \quad (24)$$

where we have made use of the relation

$$\frac{\mathcal{S}^{AB'B}}{\mathcal{S}^A} = \frac{\mathcal{S}^{AB'B}}{\mathcal{S}^{AB'}} \frac{\mathcal{S}^{AB'}}{\mathcal{S}^A} \equiv (\mathcal{D}_B^{AB'} \mathcal{D}_B^A)^{-1}. \quad (25a)$$

Note that the decomposition (25a) is not unique; we could equally well have written

$$\frac{\mathcal{S}^{AB'B}}{\mathcal{S}^A} = \frac{\mathcal{S}^{AB'B}}{\mathcal{S}^{AB}} \frac{\mathcal{S}^{AB}}{\mathcal{S}^A} \equiv (\mathcal{D}_B^{AB} \mathcal{D}_B^A)^{-1}. \quad (25b)$$

It remains to obtain expressions for the \mathcal{D} functions. Consider first \mathcal{D}_B^A : this is defined by the relation

$$\mathcal{D}_B^A = \frac{\mathcal{S}^A}{\mathcal{S}^{AB}}$$

which is similar to the definition of \mathcal{D}_1 except that S has been replaced by \mathcal{S}^A and A by B . We may therefore obtain an expansion for \mathcal{D}_B^A from (24) by replacing A by B , B by B' (where $B' \neq B$) or C , etc, and by making sure that no matrix elements involving the state A appear. Thus

$$\mathcal{D}_B^A = E - \tilde{W}_B - \sum_{B' \neq B} \frac{V_{BB'} V_{B'B}}{\mathcal{D}_B^{AB'}} - \sum_C \frac{V_{BC} V_{CB}}{\mathcal{D}_C^{AB}} - \sum_{B'' \neq B, B'} \sum_{B'' \neq B} \frac{V_{BB''} V_{B''B'} V_{B'B}}{\mathcal{D}_B^{AB} \mathcal{D}_B^{AB''}} - \dots \quad (26)$$

(The superscripts of the \mathcal{D} functions indicate the states which are *not* allowed to appear in their expansion: this is evident from the definition (23).)

If we again use I and J to label any eigenstate of H_0 , (26) may be written more concisely as

$$\mathcal{D}_B^A = E - \tilde{W}_B - \sum_{I \neq A, B} \frac{V_{BI}V_{IB}}{\mathcal{D}_I^{AB}} - \sum_{J \neq A, B, I} \sum_{I \neq A, B} \frac{V_{BJ}V_{JI}V_{IA}}{\mathcal{D}_J^{AB}\mathcal{D}_I^{ABJ}} \dots \tag{27}$$

We may repeat the procedure to obtain expansions for $\mathcal{D}_B^{AB'}$, etc. In the general case we find

$$\begin{aligned} \mathcal{D}_N^{IJ\dots LM} &= E - \tilde{W}_N - \sum_{0 \neq I, J, \dots, L, M} \frac{V_{N0}V_{0N}}{\mathcal{D}_0^{IJ\dots LMN}} \\ &- \sum_{P \neq I, J, \dots, N, 0} \sum_{0 \neq I, J, \dots, M, N} \frac{V_{NP}V_{P0}V_{0N}}{\mathcal{D}_P^{IJ\dots MN}\mathcal{D}_0^{IJ\dots NP}} - \dots \end{aligned} \tag{28}$$

Equation (28) defines a recurrence relation for the \mathcal{D} functions. By repeated use of it one can obtain explicit expressions for them. It is apparent that they consist of a sum of products of (in general) infinite continued fractions. On examining expression (28), it is clear that $(\mathcal{D}_N^{IJ\dots LM})^{-1}$ has a propagator type of structure for the state N in which transitions to the states $IJ\dots LM$ are forbidden.

The zeros of S give the eigenvalues (or quasi-particle energies) of the interacting system. From (23), these are also given by the zeros of \mathcal{D}_A , and expression (24) gives this quantity in a convenient form for finding the zeros by iteration.

Having obtained the perturbation expansion for L_A^A , let us now do the same for L_B^A . We have

$$L_B^A \equiv \frac{\mathcal{S}^{AB}}{S} = \frac{1}{\mathcal{D}_A^A} \frac{\mathcal{S}^{AB}}{\mathcal{S}^A} \tag{29}$$

By repeatedly using expressions similar to (17) we obtain the expansion

$$\begin{aligned} \mathcal{S}^{AB} &= V_{AB}\mathcal{S}^{AB} + \sum_{B' \neq B} V_{AB'}V_{B'B}\mathcal{S}^{ABB'} \\ &+ \sum_{B'' \neq B, B'} \sum_{B' \neq B} V_{AB''}V_{B''B'}V_{B'B}\mathcal{S}^{ABB'B''} \\ &+ \sum_C \sum_{B' \neq B} V_{AB'}V_{B'C}V_{CB}\mathcal{S}^{ABB'C} + \dots \end{aligned} \tag{30}$$

Consequently, employing relations of the same nature as (25), we obtain

$$\begin{aligned} L_B^A &= \frac{V_{AB}}{\mathcal{D}_A^A\mathcal{D}_B^A} + \sum_{B' \neq B} \frac{V_{AB}V_{B'B}}{\mathcal{D}_A^A\mathcal{D}_{B'}^A\mathcal{D}_B^{AB'}} + \sum_{B'' \neq B, B'} \sum_{B' \neq B} \frac{V_{AB''}V_{B''B'}V_{B'B}}{\mathcal{D}_A^A\mathcal{D}_{B'}^A\mathcal{D}_{B''}^{AB''}\mathcal{D}_B^{AB'B''}} \\ &+ \sum_C \sum_{B' \neq B} \frac{V_{AB'}V_{B'C}V_{CB}}{\mathcal{D}_A^A\mathcal{D}_{B'}^A\mathcal{D}_C^{AB'}\mathcal{D}_B^{AB'C}} + \dots \end{aligned} \tag{31a}$$

$$\begin{aligned} &= \frac{V_{AB}}{\mathcal{D}_A^A\mathcal{D}_B^A} + \sum_{I \neq A, B} \frac{V_{AI}V_{IB}}{\mathcal{D}_A^A\mathcal{D}_I^A\mathcal{D}_B^{AI}} + \sum_{J \neq A, B, I} \sum_{I \neq A, B} \frac{V_{AJ}V_{JI}V_{IB}}{\mathcal{D}_A^A\mathcal{D}_J^A\mathcal{D}_I^{AJ}\mathcal{D}_B^{AJI}} \\ &+ \dots \end{aligned} \tag{31b}$$

In a similar fashion we obtain

$$L_C^A = \sum_{I \neq A, C} \frac{V_{AI}V_{IC}}{\mathcal{D}_A^A\mathcal{D}_I^A\mathcal{D}_C^{AI}} + \sum_{J \neq A, C, I} \sum_{I \neq A, C} \frac{V_{AJ}V_{JI}V_{IC}}{\mathcal{D}_A^A\mathcal{D}_J^A\mathcal{D}_I^{AJ}\mathcal{D}_C^{AJI}} + \dots \tag{32}$$

$$\begin{aligned}
 L_D^A = & \sum_{J \neq A, D, I} \sum_{I \neq A, D} \frac{V_{AJ} V_{JI} V_{ID}}{\mathcal{D}_A \mathcal{D}_J^A \mathcal{D}_I^{AJ} \mathcal{D}_D^{AJI}} \\
 & + \sum_{K \neq A, D, J, I} \sum_{J \neq A, D, I} \sum_{I \neq A, D} \frac{V_{AK} V_{KJ} V_{JI} V_{ID}}{\mathcal{D}_A \mathcal{D}_K^A \mathcal{D}_J^{AK} \mathcal{D}_I^{AKJ} \mathcal{D}_D^{AKJJ}} + \dots \\
 & \text{etc.}
 \end{aligned} \tag{33}$$

Although the formal solution, (24), (31), (32), (33) is complicated it can be given an interpretation more in line with conventional many-body theory. In (31) for example we may associate the various terms with corresponding processes which take us from one unperturbed state to another under the action of the interaction V . Thus the following three processes correspond to the first three terms respectively of (31b):

$$\begin{aligned}
 & A \rightarrow B \\
 & A \rightarrow I \rightarrow B \quad (I \neq A, B) \\
 & A \rightarrow J \rightarrow I \rightarrow B \quad (I \neq A, B, J \neq A, B, I).
 \end{aligned} \tag{34}$$

We define an *irreducible* process to be one in which all the intermediate states are different from each other and from the initial and final states (however, the initial and final states may be identical). All the processes which occur in our expressions for the L and \mathcal{D} functions are of this type. Only irreducible processes need be considered because the contributions from reducible processes are incorporated within the continued fraction structure of the \mathcal{D} functions. If one expands the \mathcal{D} functions in Taylor series the reducible processes reappear. Consider for example the second-order approximation to L_A^A obtained by truncating expression (24):

$$L_A^A \equiv \mathcal{D}_A^{-1} \simeq \left(E - \tilde{W}_A - \sum_B \frac{V_{AB} V_{BA}}{E - W_B} \right)^{-1}. \tag{35}$$

The Taylor expansion of this quantity is (if it exists):

$$L_A^A \simeq \frac{1}{E - \tilde{W}_A} \sum_{k=0}^{\infty} \left(\frac{1}{E - \tilde{W}_A} \sum_B \frac{V_{AB} V_{BA}}{E - W_B} \right)^k. \tag{36}$$

All the contributions for $k > 1$ are represented by reducible processes. For example, the term for $k = 2$, namely

$$\frac{1}{(E - \tilde{W}_A)^3} \sum_B \sum_{B'} \frac{V_{AB'} V_{B'A} V_{AB} V_{BA}}{(E - W_{B'}) (E - W_B)}, \tag{37}$$

is represented by the process

$$A \rightarrow B' \rightarrow A \rightarrow B \rightarrow A \tag{38}$$

which is clearly reducible. In fact the solutions we have obtained may be regarded as having been derived from ordinary perturbation theory by a renormalization procedure in which all the reducible processes are summed and incorporated into the continued fractions of the \mathcal{D} functions. The present approach is more flexible than ordinary perturbation theory in that one can approximate the series or the \mathcal{D} functions at different orders if necessary.

We may represent the process (38) as

$$A \rightarrow \boxed{A \rightarrow B' \rightarrow A} \rightarrow A \rightarrow B \rightarrow A \tag{39}$$

in which the boxed-in portion may be regarded as a sub-process inserted into the propagation of the system in the state A . The situation is analogous to that of the many-body theory of interacting systems (eg Abrikosov *et al* 1963) in which the important diagrams are those which cannot be considered as having been formed by the insertion of one or more self-energy parts into one or more particle lines of an abbreviated diagram. (We take the word ‘particle’ here to include photons, holes etc.) For brevity, we shall describe these important diagrams as irreducible, others as reducible. Now a particle line represents the propagation of a particle in a certain state, so that if a self-energy part is inserted into that line, then the state of the particle just before the point of entrance of the self-energy part is the same as just after the point of exit. Thus if a diagram is reducible, it must correspond to the same state being occupied at two different points of the diagram. It follows that a reducible diagram corresponds to a reducible process.

However, the correspondence between the conventional theory and the present one is not complete. Consider for example an interacting system of particles and photons ; in the conventional theory one associates one type of Green function with the particle lines, and another with the photon lines whereas in the present theory there is just one type of propagator (the reciprocal of the \mathcal{D} functions) which is associated with both particle and photon states.

The processes which occur in expression (28) for the \mathcal{D} functions, in addition to being irreducible, are also *exclusive*, where by exclusive we mean that the states which appear as superscripts (ie the states, I, J, \dots, L, M in $\mathcal{D}_N^{IJ\dots LM}$) may not occur as intermediate states.

Making use of these concepts we may formulate simple rules for calculating the solutions as follows.

Rule 1

To calculate L_N^A to k th order in V write down all the irreducible processes by which one may proceed from state A to state N through no more than $(k - 1)$ intermediate states. With the initial state A , associate the propagator \mathcal{D}_A^{-1} , and with each intermediate state, M say, and with the final state, associate the propagator $(\mathcal{D}_M^{IJ\dots L})^{-1}$ where the states I, J, \dots, L are all the intermediate states which precede state M . With each transition, $J \rightarrow K$, associate the interaction matrix element V_{JK} . Take the product of all these factors, sum over all allowed intermediate states, and add the contributions of each irreducible process.

Thus to calculate L_B^A to third order, we have the contributions :

$$\begin{aligned} A \xrightarrow{\mathcal{D}_A} \xrightarrow{V_{AB}} \mathcal{D}_B^A B : & \quad \frac{V_{AB}}{\mathcal{D}_A \mathcal{D}_B^A} \\ A \xrightarrow{\mathcal{D}_A} \xrightarrow{V_{AI}} I \xrightarrow{\mathcal{D}_I^A} \xrightarrow{V_{IB}} \mathcal{D}_B^{AI} B : & \quad \sum_{I \neq A, B} \frac{V_{AI} V_{IB}}{\mathcal{D}_A \mathcal{D}_I^A \mathcal{D}_B^{AI}} \\ (I \neq A, B) & \end{aligned}$$

$$\begin{aligned}
 \overset{A}{\mathcal{D}} \xrightarrow[\mathcal{D}_A]{V_{AJ}} J \xrightarrow[\mathcal{D}_J^{\dagger}]{V_{JI}} I \xrightarrow[\mathcal{D}_I^{\dagger}]{V_{IB}} B : & \sum_{J \neq A, B, I} \sum_{I \neq A, B} \frac{V_{AJ} V_{JI} V_{IB}}{\mathcal{D}_A \mathcal{D}_J^{\dagger} \mathcal{D}_I^{\dagger} \mathcal{D}_B} \\
 (I \neq A, B; J \neq A, B, I) &
 \end{aligned}$$

Adding these gives the first three terms of equation (31b). An exception to this rule is the case $N = A$: then L_A^A is given simply by the relation

$$L_A^A = \mathcal{D}_A^{-1}. \tag{39a}$$

Rule 2

To calculate $\mathcal{D}_N^{IJ\dots LM}$ to k th order write down all the irreducible, exclusive processes by which one can proceed from the state N through no more than $(k - 1)$ intermediate states and back to the state N . With each intermediate state, eg U , associate the propagator $(\mathcal{D}_U^{IJ\dots LMNR\dots T})^{-1}$ where the states R, \dots, T are all the intermediate states which precede state U . Propagators are not associated with the initial and final states. The procedure subsequently is the same as in rule 1 except that all the contributions are subtracted from $E - \tilde{W}_N$.

Thus to calculate \mathcal{D}_C^{AB} to third order we have to consider the following processes:

$$\begin{aligned}
 C \xrightarrow[\mathcal{D}_C^{ABC}]{V_{CI}} I \xrightarrow[\mathcal{D}_I^{\dagger}]{V_{IC}} C : & \sum_{I \neq A, B, C} \frac{V_{CI} V_{IC}}{\mathcal{D}_I^{\dagger}} \\
 C \xrightarrow[\mathcal{D}_C^{ABC}]{V_{CJ}} J \xrightarrow[\mathcal{D}_J^{\dagger}]{V_{JI}} I \xrightarrow[\mathcal{D}_I^{\dagger}]{V_{IC}} C : & \sum_{J \neq A, B, C, I} \sum_{I \neq A, B, C} \frac{V_{CJ} V_{JI} V_{IC}}{\mathcal{D}_J^{\dagger} \mathcal{D}_I^{\dagger}}
 \end{aligned}$$

Combining these contributions as described, we obtain

$$\mathcal{D}_C^{AB} = E - \tilde{W}_C - \sum_{I \neq A, B, C} \frac{V_{CI} V_{IC}}{\mathcal{D}_I^{\dagger}} - \sum_{J \neq A, B, C, I} \sum_{I \neq A, B, C} \frac{V_{CJ} V_{JI} V_{IC}}{\mathcal{D}_J^{\dagger} \mathcal{D}_I^{\dagger}} - \dots$$

Finally, we note the alternative way of writing the products of the \mathcal{D} functions implied by expressions (25). For example, in the third-order process for L_B^A given above equation (39a), we could have written the \mathcal{D} functions as

$$\overset{A}{\mathcal{D}_A^{\dagger}} \rightarrow \overset{J}{\mathcal{D}_J^{\dagger}} \rightarrow \overset{I}{\mathcal{D}_I^{\dagger}} \rightarrow \overset{B}{\mathcal{D}_B}$$

ie we could have worked backwards in labelling the \mathcal{D} functions instead of forwards as we have done in our previous examples.

As our main interest here is with the interacting atom-radiation system described by the Hamiltonian (1) we consider in more detail the application of the foregoing rules to this case. We henceforth use a, b, c, \dots, i, \dots to label the eigenstates of the atomic system in isolation, and we use \underline{n} as shorthand for the set of non-negative integers n_1, n_2, \dots, n_N which define the photon occupation numbers. N is the total number of modes, which may be infinite. Thus a typical eigenstate of the unperturbed Hamiltonian $H_0 \equiv H_A + H_F$ may be written $|i\rangle|\underline{n}\rangle \equiv |i\underline{n}\rangle$ and the corresponding unperturbed eigenvalue is $E_i + \sum_{\lambda} n_{\lambda} \omega^{\lambda}$. The primary processes of the interaction (1d), $i\underline{n} \rightarrow j\underline{n} \pm \lambda$, are shown in figure 1 together with the corresponding matrix elements.

To illustrate the method, it is convenient to consider the especially simple case of an atomic system which has only two eigenstates, which we denote as $|\alpha\rangle$ and $|\beta\rangle$ to distinguish them from the states of the multilevel atom, $|a\rangle, |b\rangle, |c\rangle, \dots$. The processes

which need to be considered in, for example, the calculation of \mathcal{D}_{an} to fourth order are

$$\left. \begin{aligned}
 \alpha n &\rightarrow \beta n \lambda \rightarrow \alpha n \\
 \alpha n &\rightarrow \beta n - \lambda \rightarrow \alpha n
 \end{aligned} \right\} \text{(second order)}$$

$$\left. \begin{aligned}
 \alpha n &\rightarrow \beta n \lambda \rightarrow \alpha n \lambda \lambda_1 \rightarrow \beta n \lambda_1 \rightarrow \alpha n \\
 \alpha n &\rightarrow \beta n \lambda \rightarrow \alpha n \lambda - \lambda_1 \rightarrow \beta n - \lambda_1 \rightarrow \alpha n \\
 \alpha n &\rightarrow \beta n - \lambda \rightarrow \alpha n - \lambda \lambda_1 \rightarrow \beta n \lambda_1 \rightarrow \alpha n \\
 \alpha n &\rightarrow \beta n - \lambda \rightarrow \alpha n - \lambda - \lambda_1 \rightarrow \beta n - \lambda_1 \rightarrow \alpha n
 \end{aligned} \right\} \begin{array}{l} (\lambda \neq \lambda_1) \\ \text{(fourth order).} \end{array} \quad (40)$$

The condition, $\lambda_1 \neq \lambda$ is imposed in order that the processes be irreducible. If we allow $\lambda = \lambda_1$ in the first of the fourth-order processes for example, it becomes reducible because the second and fourth intermediate states are then identical. It may then be considered as a renormalized second-order process :

$$\alpha n \xrightarrow{\alpha n 2 \lambda} \beta n \lambda \rightarrow \alpha n. \quad (41)$$

In figure 2 we represent the processes in (40) and (41) by diagrams. However, as it is quite straightforward to write out the processes in the manner described, we do not adopt the diagram approach subsequently.

To demonstrate the calculation of the contributions from each process we consider some simpler examples which will be made use of in a later section. To calculate \mathcal{D}_{an} to second order the relevant processes and their contributions are

$$\alpha n \xrightarrow[\mathcal{D}_{bn-\lambda}^{an}]^{g_{ab}^{\lambda} \sqrt{n_{\lambda}}} \beta n - \lambda \xrightarrow[\mathcal{D}_{bn\lambda}^{an}]^{-g_{ba}^{\lambda} \sqrt{n_{\lambda}}} \alpha n : \quad - \sum_{\lambda} \sum_b \frac{g_{ab}^{\lambda} g_{ba}^{\lambda} n_{\lambda}}{\mathcal{D}_{bn-\lambda}^{an}}$$

$$\alpha n \xrightarrow[\mathcal{D}_{bn\lambda}^{an}]^{-g_{ab}^{\lambda} \sqrt{(n_{\lambda}+1)}} \beta n \lambda \xrightarrow[\mathcal{D}_{bn\lambda}^{an}]^{g_{ba}^{\lambda} \sqrt{(n_{\lambda}+1)}} \alpha n : \quad - \sum_{\lambda} \sum_b \frac{g_{ab}^{\lambda} g_{ba}^{\lambda} (n_{\lambda}+1)}{\mathcal{D}_{bn\lambda}^{an}}.$$

Hence, subtracting these two contributions from $E - E_a - \sum_{\lambda} n_{\lambda} \omega_{\lambda}$ we obtain

$$\mathcal{D}_{an} = E - E_a - \sum_{\lambda} n_{\lambda} \omega_{\lambda} - \sum_{\lambda} \sum_b |g_{ab}^{\lambda}|^2 \left(\frac{n_{\lambda}}{\mathcal{D}_{bn-\lambda}^{an}} + \frac{n_{\lambda}+1}{\mathcal{D}_{bn\lambda}^{an}} \right) \quad (42a)$$

where we have used $g_{ab}^{\lambda} g_{ba}^{\lambda} = -|g_{ab}^{\lambda}|^2$, which follows from (2a). In (42a) the propagators $\mathcal{D}_{bn-\lambda}^{an}$ and $\mathcal{D}_{bn\lambda}^{an}$ appear. To calculate $\mathcal{D}_{bn\lambda}^{an}$ to second order for example, consider the processes

$$\begin{aligned}
 \beta n \lambda &\xrightarrow[\mathcal{D}_{cn\lambda\lambda_1}^{an}]^{-g_{cb}^{\lambda} (n_{\lambda_1} + 1 + \delta_{\lambda\lambda_1})^{1/2}} \alpha n \lambda \lambda_1 \xrightarrow[\mathcal{D}_{bn\lambda}^{an}]^{g_{cb}^{\lambda} (n_{\lambda_1} + 1 + \delta_{\lambda\lambda_1})^{1/2}} \beta n \lambda \\
 \beta n \lambda &\xrightarrow[\mathcal{D}_{cn\lambda-\lambda_1}^{an}]^{-g_{cb}^{\lambda} (n_{\lambda_1} + \delta_{\lambda\lambda_1})^{1/2}} \alpha n \lambda - \lambda_1 \xrightarrow[\mathcal{D}_{bn\lambda}^{an}]^{-g_{cb}^{\lambda} (n_{\lambda_1} + \delta_{\lambda\lambda_1})^{1/2}} \beta n \lambda \quad (\lambda \neq \lambda_1 \text{ if } c = a)
 \end{aligned}$$

which give

$$\mathcal{D}_{bn\lambda}^{an} = E - E_b - \sum_{\lambda_1} n_{\lambda_1} \omega_{\lambda_1} - \omega_{\lambda} - \sum_{\lambda_1} \sum_c |g_{bc}^{\lambda}|^2 \left(\frac{n_{\lambda_1} + 1 + \delta_{\lambda\lambda_1}}{\mathcal{D}_{cn\lambda\lambda_1}^{an, bn\lambda}} + \frac{(n_{\lambda_1} + \delta_{\lambda\lambda_1})(1 - \delta_{\lambda\lambda_1} \delta_{ca})}{\mathcal{D}_{cn\lambda-\lambda_1}^{an, bn\lambda}} \right). \quad (42b)$$

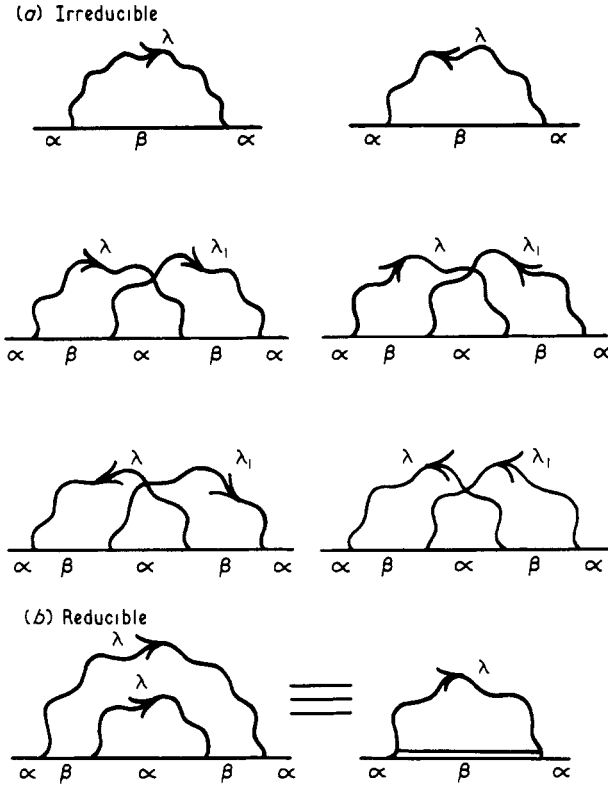


Figure 2. (a) the irreducible and (b) the reducible diagrams corresponding to processes (40) and (41) respectively. As in figure 1 only those photons which undergo emission or absorption are shown. The reducible fourth-order diagram (b) is equivalent to a second-order diagram in which the free atomic propagator --- is replaced by a renormalized propagator ==== .

The condition, $\lambda \neq \lambda_1$ if $c = a$, is imposed in the second process to ensure that it is always exclusive: if we set $\lambda = \lambda_1$ and $c = a$ the intermediate state becomes $a\underline{n}$, which is not allowed.

To give an example of the calculation of a probability amplitude, consider $L_{\beta\underline{n}\lambda}^{a\underline{n}}$. To third order, the processes which contribute are

$$\left. \begin{aligned} a\underline{n} &\rightarrow \beta\underline{n}\lambda \\ a\underline{n} &\rightarrow \beta\underline{n}\lambda_1 \rightarrow a\underline{n}\lambda\lambda_1 \rightarrow \beta\underline{n}\lambda \\ a\underline{n} &\rightarrow \beta\underline{n}-\lambda_1 \rightarrow a\underline{n}\lambda-\lambda_1 \rightarrow \beta\underline{n}\lambda \end{aligned} \right\} \lambda_1 \neq \lambda.$$

Using rule 1 we easily obtain

$$L_{\beta\underline{n}\lambda}^{a\underline{n}} = \frac{g_{\beta a}^\lambda (n_\lambda + 1)^{1/2}}{\mathcal{D}_{a\underline{n}}} \left[1 + \sum_{\lambda_1 \neq \lambda} |g_{\beta a}^{\lambda_1}|^2 \times \left(\frac{n_\lambda + 1}{\mathcal{D}_{\beta\underline{n}\lambda_1}^{a\underline{n}} \mathcal{D}_{a\underline{n}\lambda_1}^{a\underline{n}} \mathcal{D}_{\beta\underline{n}\lambda}^{a\underline{n}, \beta\underline{n}\lambda_1, a\underline{n}\lambda\lambda_1}} + \frac{n_{\lambda_1}}{\mathcal{D}_{\beta\underline{n}-\lambda_1}^{a\underline{n}} \mathcal{D}_{a\underline{n}\lambda-\lambda_1}^{a\underline{n}} \mathcal{D}_{\beta\underline{n}\lambda}^{a\underline{n}, \beta\underline{n}-\lambda_1, a\underline{n}\lambda-\lambda_1}} \right) \right] \quad (43)$$

where we have used (2a) to write $g_{a\beta}^\lambda (g_{\beta a}^{\lambda_1})^2$ as $-g_{\beta a}^\lambda |g_{\beta a}^{\lambda_1}|^2$.

So far we have ignored the contributions of $H_p^{(od)}$, which we may write as

$$H_p^{(od)} = \sum_i \sum_j h_{ij} |i\rangle \langle j| \tag{44a}$$

where

$$h_{ij} = \frac{\langle i | \mu^2 | j \rangle (1 - \delta_{ij})}{6\pi^3 \epsilon_0} \int k^2 dk = (h_{ji})^* \tag{44b}$$

As expressions (44) indicate, the primary action of $H_p^{(od)}$ is to cause the atom to change its state without affecting the photon occupation numbers.

From the rules, it is clear that $H_p^{(od)}$ may be taken into account simply by allowing transitions of the form

$$i\underline{n} \rightarrow j\underline{n} : \quad h_{ij}$$

for which the matrix element is, as indicated, h_{ij} .

Thus to calculate $L_{a\underline{n}}^{an}$ to second order taking into account $H_p^{(od)}$ we must consider the following processes:

$$\begin{aligned} a\underline{n} &\rightarrow b\underline{n} \rightarrow a\underline{n} && (b \neq a) \\ a\underline{n} &\rightarrow b\underline{n}\lambda \rightarrow a\underline{n} \\ a\underline{n} &\rightarrow b\underline{n} - \lambda \rightarrow a\underline{n} \end{aligned}$$

which give

$$L_{a\underline{n}}^{an} = \left[E - E_a - \sum_{\lambda} n_{\lambda} \omega_{\lambda} - \sum_{b \neq a} \frac{|h_{ab}|^2}{D_{b\underline{n}}^{an}} - \sum_{\lambda} \sum_b |g_{ab}^{\lambda}|^2 \left(\frac{n_{\lambda}}{\mathcal{D}_{b\underline{n}-\lambda}^{an}} + \frac{n_{\lambda} + 1}{\mathcal{D}_{b\underline{n}\lambda}^{an}} \right) \right]^{-1} \tag{45}$$

Note, however, that $h_{ab} \sim e^2$, whereas $g_{ab}^{\lambda} \sim e$ (e being the electronic charge). Hence, if one is working to order e^2 in (45), the contribution $-\sum_{b \neq a} |h_{ab}|^2 / \mathcal{D}_{b\underline{n}}^{an}$ should be neglected, or, if one is working to order e^4 , additional terms should be included involving terms of order $|g|^4$ and $|h||g|^2$.

It is a general rule that the lowest-order contribution from $H_p^{(od)}$ is of the order $e^2 \times$ the lowest-order contribution from H_{AF} , and for this reason it may often be neglected. Finally we note that for a two-level system where μ is off-diagonal, $H_p^{(od)}$ vanishes identically. This may easily be inferred from (3d).

4. The single-mode, two-level problem

The rules described in the previous section apply to any number of modes; for the case of a single mode and two atomic levels they must lead to the expressions derived earlier in I. It is instructive to examine briefly how this comes about. An analysis of this situation also assists in the interpretation of the multimode case.

Consider for example \mathcal{D}_{as}^{as} , $\mathcal{D}_{\beta s + n}^{as, \beta s + 1, as + 2, \dots, as + n - 1}$ and $\mathcal{D}_{as + n}^{as, \beta s + 1, \dots, \beta s + n - 1}$. The only processes which contribute to these are, respectively,

$$\left. \begin{aligned} \alpha s &\rightarrow \beta s + 1 \rightarrow \alpha s \\ \alpha s &\rightarrow \beta s - 1 \rightarrow \alpha s \end{aligned} \right\} \tag{46}$$

$$\beta s + n \rightarrow \alpha s + n + 1 \rightarrow \beta s + n \tag{47}$$

$$\alpha s + n \rightarrow \beta s + n + 1 \rightarrow \alpha s + n. \tag{48}$$

To see that these are the only processes, consider example (47). The only alternatives are of the type

$$\beta s + n \rightarrow \alpha s + n - 1 \rightarrow \beta s + n \tag{49}$$

and

$$\beta s + n \rightarrow \alpha s + n + 1 \rightarrow \beta s + n + 2 \rightarrow \alpha s + n + 1 \rightarrow \beta s + n. \tag{50}$$

Process (49) is not allowed because it violates the exclusive condition and (50) is not allowed because the process is reducible. One may argue similarly for (46) and (48). Thus (46), (47) and (48) are the only processes which contribute to $\mathcal{D}_{\alpha s}$, $\mathcal{D}_{\beta s+n}^{\alpha s \dots \alpha s+n-1}$ and $\mathcal{D}_{\alpha s+n}^{\alpha s \dots \beta s+n-1}$ and we may therefore write down the exact contributions following rule 2 as:

$$\mathcal{D}_{\alpha s} = E - E_{\alpha} - s\omega - |g|^2(s+1)/\mathcal{D}_{\beta s+1}^{\alpha s} - |g|^2 s/\mathcal{D}_{\beta s-1}^{\alpha s} \tag{51}$$

$$\mathcal{D}_{\beta s+n}^{\alpha s \dots \alpha s+n-1} = E - E_{\beta} - (s+n)\omega - |g|^2(s+n+1)/\mathcal{D}_{\alpha s+n+1}^{\alpha s \dots \beta s+n} \tag{52}$$

$$\mathcal{D}_{\alpha s+n}^{\alpha s \dots \beta s+n-1} = E - E_{\alpha} - (s+n)\omega - |g|^2(s+n+1)/\mathcal{D}_{\beta s+n+1}^{\alpha s \dots \alpha s+n} \tag{53}$$

(We have dropped the superscript λ from g as this is superfluous in the single-mode case.) It is clear that (52) and (53) may be used repeatedly to generate continued fraction expressions for $\mathcal{D}_{\alpha s+n}^{\alpha s \dots \beta s+n-1}$ and $\mathcal{D}_{\beta s+n}^{\alpha s \dots \alpha s+n-1}$.

One may show in an entirely analogous manner that

$$\mathcal{D}_{\beta s-n}^{\alpha s, \beta s-1 \dots \alpha, s-n+1} = E - E_{\beta} - (s-n)\omega - |g|^2(s-n)/\mathcal{D}_{\alpha s-n-1}^{\alpha s \dots \beta s-n} \tag{54}$$

$$\mathcal{D}_{\alpha s-n}^{\alpha s, \beta s-1 \dots \beta s-n-1} = E - E_{\alpha} - (s-n)\omega - |g|^2(s-n)/\mathcal{D}_{\beta s-n-1}^{\alpha s \dots \alpha s-n}. \tag{55}$$

Consider rule 1 applied to the calculation of $L_{\beta s+n}^{\alpha s}$ where n is odd. The lowest-order process which contributes is the n th, and higher-order processes than the n th are not allowed because then αs or $\beta s + n$ becomes an intermediate state. Therefore the only contributing process is

$$\alpha s \rightarrow \beta s + 1 \rightarrow \alpha s + 2 \rightarrow \dots \rightarrow \alpha s + n - 1 \rightarrow \beta s + n$$

and $L_{\beta s+n}^{\alpha s}$ is consequently given simply by

$$L_{\beta s+n}^{\alpha s} = \frac{g(s+1)^{1/2} g(s+2)^{1/2} \dots g(s+n)^{1/2}}{\mathcal{D}_{\alpha s} \mathcal{D}_{\beta s+1}^{\alpha s} \mathcal{D}_{\alpha s+2}^{\alpha s, \beta s+1} \dots \mathcal{D}_{\beta s+n}^{\alpha s \dots \alpha s+n-1}} \tag{56}$$

For $n = 1$ this expression is consistent with equation (43) applied to the single-mode case. Similarly one finds

$$L_{\beta s-n}^{\alpha s} = \frac{g^* s^{1/2} g^*(s-1)^{1/2} \dots g^*(s-n+1)^{1/2}}{\mathcal{D}_{\alpha s-n} \mathcal{D}_{\beta s-1}^{\alpha s} \mathcal{D}_{\alpha s-2}^{\alpha s, \beta s-1} \dots \mathcal{D}_{\beta s-n}^{\alpha s \dots \alpha s-n+1}} \tag{57}$$

Expressions identical to (56) and (57) may be obtained for $L_{\alpha s \pm n}^{\alpha s}$ where n is now even, except that the final propagator in (56) must become $\mathcal{D}_{\alpha s+n}^{\alpha s \dots \beta s+n-1}$ to give $L_{\alpha s+n}^{\alpha s}$, and the final propagator in (57) must become $\mathcal{D}_{\alpha s-n}^{\alpha s \dots \beta s-n+1}$ to give $L_{\alpha s-n}^{\alpha s}$.

These results are identical with the ones obtained in I if one makes the following correspondence in notation:

$$\begin{aligned} \mathcal{D}_{\alpha s+n}^{\alpha s \dots \beta s+n-1} &\equiv \lambda_{s+n}; & \mathcal{D}_{\alpha s-n}^{\alpha s \dots \alpha s-n+1} &\equiv l_{s-n}; & L_{\alpha s \pm n}^{\alpha s} &\equiv A_{s \pm n}^s(e) \\ \mathcal{D}_{\beta s+n}^{\alpha s \dots \alpha s+n-1} &\equiv \mu_{s+n}; & \mathcal{D}_{\beta s-n}^{\alpha s \dots \beta s-n+1} &\equiv m_{s-n}; & L_{\beta s \pm n}^{\alpha s} &\equiv B_{s \pm n}^s(e). \end{aligned} \tag{58}$$

The notation used here is more transparent than that used in I.

One may repeat the calculations with the roles of α and β interchanged, and so recover the remainder of the results given in I.

Consider one of the simplest expressions

$$L_{\alpha s}^{\alpha s} = \left(E - E_x - s\omega - \frac{|g|^2(s+1)}{E - E_\beta - (s+1)\omega - \frac{|g|^2(s+2)}{E - E_\alpha - (s+2)\omega \dots}} - \frac{|g|^2 s}{E - E_\beta - (s-1)\omega - \frac{|g|^2(s-1)}{E - E_\alpha - (s-2)\omega \dots}} \right)^{-1} \quad (59)$$

The continued fractions in (59) have a simple physical interpretation. Consider the first. The system may undergo emission from the state $|\alpha s\rangle$ to the state $|\beta s+1\rangle$ and then back again. The probability for this is proportional to $|g|^2(s+1)$. Thus the free propagator $(E - E_x - s\omega)^{-1}$ is modified by transitions to the state $|\beta s+1\rangle$. However, the system may propagate whilst it is in the intermediate state $|\beta s+1\rangle$, the free propagator for this state being $[E - E_\beta - (s+1)\omega]^{-1}$. This is itself modified by transitions to the state $|\alpha s+2\rangle$, and thus the term $|g|^2(s+2)/[E - E_\alpha - (s-2)\omega]$ has to be subtracted. Hence the continued fraction structure reflects the modification of the free propagators due to transitions to intermediate states. The free propagator $[E - E_\beta - (s+1)\omega]^{-1}$ is not modified by the transition $|\beta s+1\rangle \rightarrow |\alpha s\rangle$ because this process has been taken into account at an earlier stage in the continued fraction. Similarly, the state $|\alpha s\rangle$ may undergo transitions to the state $|\beta s-1\rangle$ and back again (ie absorption). The resulting sequence of transitions gives rise to the second continued fraction in (49). Thus the first continued fraction describes purely emissive processes, the second purely absorptive.

The interpretation given here may be extended, with appropriate generalizations, to the multilevel, multimode case.

5. Stimulated and spontaneous emission

There has been a recent revival of interest in the quantum theory of stimulated and spontaneous emission (Bullough and Caudrey 1971, Knight 1972, Ackerhalt *et al* 1973, Bullough *et al* 1974). These papers deal with the generalization of the Lamb shift and the damping constants to include contributions from stimulated processes. Bullough and Caudrey, Ackerhalt, Knight and Eberly, and Bullough, Caudrey and Obada use reaction field theory together with the two-level atomic model. However, as Knight (1972) has emphasized, the two-level atom is *not* a good approximation to the multilevel atom as far as self-energy effects are concerned, and care must be taken in interpreting the results obtained from this model. Knight uses the $A \cdot p$ forms of the interaction, and discusses only the electromagnetic shifts, but considers the multilevel case.

In this section we apply the continued fraction method to this situation. We treat the multilevel atom model, but unlike Knight use the dipole approximation for the interaction. The expressions we obtain for the shifts are in agreement with those found by the previously quoted authors, but in addition we give the generalization of the damping constant to include stimulated processes. (The appropriate generalization for the two-level model has been given by Bullough *et al* 1974). Our method gives an expression for the lineshape directly, and furthermore allows the interpretation that each level E_i has a shift Δ_i and a damping constant Γ_i associated with it. However, in determining the

lineshape for transitions from level i to level j we show that it is the *difference* of the shifts $\Delta_i - \Delta_j$ and the *sum* of the damping constants $\Gamma_i + \Gamma_j$ which are important. Our method has the further advantage that it may be straightforwardly extended to treat fourth-order contributions.

We suppose that at time $t = 0$ the atom is in the state $|a\rangle$ and there are n photons present (ie the field is in a pure number states). To obtain expressions for the lineshape we first ask for the probability that the atom will be found in the state $|b\rangle$ at the later time t with an additional photon present in the mode λ (λ need not be one of the modes initially occupied). To calculate this we need $L_{bn\lambda}^{an}$. For the present purposes it is sufficient to consider only the first-order contribution, which derives from the process $a_n \rightarrow b_n \lambda$. Rule 1 gives straightforwardly

$$L_{bn\lambda}^{an} = \frac{g_{ab}^\lambda (n_\lambda + 1)^{1/2}}{\mathcal{D}_{an} \mathcal{D}_{bn\lambda}^{an}}$$

We assume $n_\lambda \lesssim 1$, all λ , so that complications due to the dynamic Stark effect may be neglected (eg Stroud 1973). Then it is sufficient to take the second-order approximations to \mathcal{D}_{an} and $\mathcal{D}_{bn\lambda}^{an}$, and these are given by equations (42a) and (42b) respectively. We take the zeroth approximation to the propagators which appear in the denominators of these expressions. Explicitly then, we have

$$L_{bn\lambda}^{an} = g_{ab}^\lambda (n_\lambda + 1)^{1/2} \left[z - E_a - \sum_{\lambda_1} \sum_c |g_{ac}^{\lambda_1}|^2 \left(\frac{n_{\lambda_1}}{z - E_c + \omega_{\lambda_1}} + \frac{n_{\lambda_1} + 1}{z - E_c - \omega_{\lambda_1}} \right) \right]^{-1} \\ \times \left[z - E_b - \omega_\lambda - \sum_{\lambda_1} \sum_c |g_{bc}^{\lambda_1}|^2 \left(\frac{n_{\lambda_1}(1 - \delta_{\lambda\lambda_1}\delta_{ca})}{z - E_c - \omega_\lambda + \omega_{\lambda_1}} + \frac{n_{\lambda_1} + 1 + \delta_{\lambda\lambda_1}}{z - E_c - \omega_\lambda - \omega_{\lambda_1}} \right) \right]^{-1} \quad (60)$$

where $z \equiv E - \sum_\lambda n_\lambda \omega_\lambda$. Bearing in mind that in the formula for the inverse Laplace transform (4) the contour lies above the real axis (a rigorous method of evaluating these integrals is described by Goldberger and Watson 1964) we set

$$z \equiv x + i\varepsilon, \quad \varepsilon \rightarrow 0^+ \quad (61)$$

when it is clear that the poles of (60) occur at

$$x - E_a - \sum_{\lambda_1} \sum_c |g_{ac}^{\lambda_1}|^2 \left(\frac{n_{\lambda_1}}{x - E_c + \omega_{\lambda_1} + i\varepsilon} + \frac{n_{\lambda_1} + 1}{x - E_c - \omega_{\lambda_1} + i\varepsilon} \right) = 0 \quad (62)$$

and

$$x - E_b - \omega_\lambda - \sum_{\lambda_1} \sum_c |g_{bc}^{\lambda_1}|^2 \left(\frac{n_{\lambda_1}(1 - \delta_{\lambda\lambda_1}\delta_{ca})}{x - E_c - \omega_\lambda + \omega_{\lambda_1} + i\varepsilon} + \frac{n_{\lambda_1} + 1 + \delta_{\lambda\lambda_1}}{x - E_c - \omega_\lambda - \omega_{\lambda_1} + i\varepsilon} \right) = 0. \quad (63)$$

It seems adequate to order $|g|^2$ to set $x = E_a$ and $x = E_b + \omega_\lambda$ in the denominators of (62) and (63) respectively, when we obtain

$$x - E_a = \sum_{\lambda_1} |g_{ac}^{\lambda_1}|^2 \left(\frac{n_{\lambda_1}}{E_{a,c} + \omega_{\lambda_1} + i\varepsilon} + \frac{n_{\lambda_1} + 1}{E_{a,c} - \omega_{\lambda_1} + i\varepsilon} \right) \quad (64)$$

$$x - E_b - \omega_\lambda = \sum_{\lambda_1} |g_{bc}^{\lambda_1}|^2 \left(\frac{n_{\lambda_1}(1 - \delta_{\lambda\lambda_1}\delta_{ca})}{E_{b,c} + \omega_{\lambda_1} + i\varepsilon} + \frac{n_{\lambda_1} + 1 + \delta_{\lambda\lambda_1}}{E_{b,c} - \omega_{\lambda_1} + i\varepsilon} \right) \quad (65)$$

where $E_{i,j} \equiv E_i - E_j$. We assume for simplicity that the incident photons are unpolarized, and we replace the sums over modes by integrals over wavevectors in the usual way:

$$\sum_{\lambda} \rightarrow \frac{V}{4\pi^3} \int k^2 dk \int d\Omega \tag{66}$$

(Ω represents the solid angle.) Using

$$\frac{1}{x \pm i\epsilon} = \frac{P}{x} \mp i\pi \delta(x) \tag{67}$$

where P denotes the principal value of the integral, we find that the poles may be written

$$x = E_a + \Delta_a - i\Gamma_a; \quad x = E_b + \omega_{\lambda} + \Delta_b - i\Gamma_b. \tag{68}$$

The shifts Δ_i and halfwidths Γ_i of the level E_i are given explicitly by

$$\Delta_i = \frac{V}{4\pi^2 c} \sum_c \int d\Omega \int k^2 dk |g_{c,i}^k|^2 P \left(\frac{n_k}{k - k_{c,i}} - \frac{n_k + 1}{k_{i,c} + k} \right) \tag{69}$$

and

$$\Gamma_i = \frac{V}{4\pi^2 c} \sum_c \int d\Omega k_c^2 [|g_{c,i}^k|^2 n_k |k_{c,i}| \theta(k_{c,i}) + |g_{i,c}^k|^2 (n_k + 1) |k_{i,c}| \theta(k_{i,c})] \tag{70}$$

where $k_i = E_i/c$ and $\theta(x) = 1$ if $x > 0$ and 0 if $x \leq 0$. Note that (70) is the generalization of the usual Wigner–Weisskopf type of result to the multilevel case and to include stimulated processes. We have assumed that the n_{λ} are sufficiently dense for the condition $(1 - \delta_{\lambda\lambda_1} \delta_{c\lambda})$ in the first term of (65) to be replaced by unity. The $\delta_{\lambda\lambda_1}$ term in the final part of (65) makes zero contribution when the sum is replaced by an integral.

Our expressions for the electromagnetic shifts Δ_i and radiative damping constants Γ_i are in line with the usual interpretation that real transitions (ie energy conserving or resonant ones) give rise to lifetime effects, and virtual transitions (ie energy non-conserving or non-resonant ones) give rise to frequency shifts. Thus the contributions to Γ_i arise from transitions from the energy level E_i to levels E_c with *higher* energy with the corresponding *absorption* of photons of wavevector $k = k_{c,i}$, and from transitions to levels of *lower* energy with the spontaneous and stimulated *emission* of photons of wavevector $k = k_{i,c}$. Because the principal parts of the integrals in (69) have to be taken, it is clear that the photons of wavevector $k = |k_{i,c}|$ make zero contribution so that the shifts Δ_i arise from all transitions except the resonant ones.

Thus we may write

$$L_{ba}^{an} = \frac{g_{ab}^{\lambda} (n_{\lambda} + 1)^{1/2}}{(x - E_a - \Delta_a + i\Gamma_a)(x - E_b - \omega_{\lambda} - \Delta_b + i\Gamma_b)} \tag{71}$$

and the inverse Laplace transform is easily performed. From (4) one finds

$$P_{ba}^{an}(t) = \frac{|g_{ab}^{\lambda}|^2 (n_{\lambda} + 1)}{(\omega_{\lambda} - E_{a,b} - \Delta_{a,b})^2 + \Gamma_{a,b}^2} [e^{-2\Gamma_a t} + e^{-2\Gamma_b t} - 2e^{-(\Gamma_a + \Gamma_b)t} \cos(\omega_{\lambda} - E_{a,b} - \Delta_{a,b})t] \tag{72}$$

where, as before, a comma between two subscripts indicates that the difference is to be taken between the corresponding quantities, eg $\Gamma_{a,b} \equiv \Gamma_a - \Gamma_b$. If we assume that our

detector responds to the average probability of a photon being present over a period very much greater than the atomic lifetimes, then the response will be proportional to

$$\sum_b \int_0^\infty dt P_{b\bar{n}\lambda}^{an}(t) = \sum_b \frac{|g_{ab}^\lambda|^2 (n_\lambda + 1)}{2\Gamma_a \Gamma_b} \frac{\Gamma_a + \Gamma_b}{(\omega_\lambda - E_{a,b} - \Delta_{a,b})^2 + (\Gamma_a + \Gamma_b)^2} \quad (73)$$

so that although the difference of the atomic lifetimes appears in (72), it is their sum only which appears in the physically more important result (73). If ω_λ is close to resonance for a particular atomic level, ie $\omega_\lambda \simeq E_{a,b}$ for just one state $|b\rangle$, then this term in the sum over b will dominate the other contributions.

Now expression (73) refers to the observation of photons due to transitions between two excited states. Obviously, the shutter of the photon detector must be open from time $t = 0$ when the system is in the state a with \bar{n} photons present; at times $t \gg \Gamma_a^{-1}, \Gamma_b^{-1}$ the occupation probability of the levels a and b will be approximately zero, as the atom will have decayed to lower levels. If we now take b to be the ground state of the system and consider spontaneous emission from level a we have the situation discussed by Weisskopf and Wigner (1930). It follows from (70) that $\Gamma_b = 0$, and so for times $t \gg \Gamma_a^{-1}$ the probability of the system being in the ground state is close to unity. It is then sufficient, following Weisskopf and Wigner, to assume that the probability of observing a photon is proportional to $P_{b\bar{n}\lambda}^{an}(\infty)$ where from (72),

$$P_{b\bar{n}\lambda}^{an}(\infty) = \frac{|g_{ab}^\lambda|^2}{(\omega_\lambda - E_{a,b} - \Delta_{a,b})^2 + \Gamma_a^2}.$$

Our previous criterion for observing a photon also leads to this result as the decaying terms in (72) are negligible compared with the time-independent term if the shutter of the photon detector is open for an interval much greater than Γ_a^{-1} .

To proceed further in the discussion of (73) it is convenient to consider a definite experimental situation: we assume a unidirectional beam of photons in the state $|\bar{n}\rangle$ is incident on the atomic system. Choosing a direction away from the incident beam, one seeks to detect a photon in the mode λ_1 . (Obviously, λ_1 is not one of the modes occupied at $t = 0$ ie $n_{\lambda_1} = 0$.) The lineshape observed is obtained from (73) on multiplying by the density of states factor

$$\frac{V}{4\pi^3} k_1^2 \delta k_1 \delta \Omega \quad (74)$$

where δk_1 and $\delta \Omega$ are determined by the resolution of the photon detector, which we have assumed does not differentiate polarization. Using expression (2) for g_{ba}^k , we find

$$P(k_1) \delta k_1 \delta \Omega = \frac{ck_1^3 \cos^2 \theta_{k_1}}{16\pi^3 \epsilon_0} \sum_b \frac{\Gamma_a + \Gamma_b}{\Gamma_a \Gamma_b} \frac{|\mu_{ab}|^2 \delta k_1 \delta \Omega}{(\omega_{k_1} - E_{a,b} - \Delta_{a,b})^2 + (\Gamma_a + \Gamma_b)^2}. \quad (75)$$

Our derivation of (75) demonstrates that it is meaningful to associate shifts and lifetimes with each atomic level, and that it is the sums of these lifetimes for the two states concerned which determine the linewidth, whereas it is the difference of the shifts which determines the resonance condition.

In the Wigner-Weisskopf approach one obtains a factor $k_{a,b}^3$ in the numerator instead of the k_1^3 of (75). Equation (75) therefore shows small but significant deviations from the

Lorentzian shape, and the maximum occurs, not at $\omega_{\mathbf{k}_1} = E_{a,b} + \Delta_{a,b}$ but at

$$\omega_{\mathbf{k}_1} = E_{a,b} + \Delta_{a,b} + 3(\Gamma_a + \Gamma_b)^2 / 2E_{a,b} \tag{76}$$

(see Power and Zienau 1959, for detailed discussion).

The next step is to obtain explicit expressions for the shifts and linewidths, and it is convenient to consider the case of pure spontaneous emission first. Accordingly, we set $n_{\mathbf{k}} = 0$ in (69) and (70), and using expression (2) for $g_{ij}^{\mathbf{k}}$, we obtain

$$\Delta_i^{(0)} = -\frac{1}{6\pi^2\epsilon_0} \sum_c |\mu_{c,i}|^2 \mathbf{P} \int \frac{k^3 dk}{k + k_{c,i}} \tag{77}$$

$$\Gamma_i^{(0)} = \frac{1}{6\pi\epsilon_0} \sum_c |\mu_{c,i}|^2 k_{i,c}^3 \theta(k_{i,c}). \tag{78}$$

The angular integrals in (77) and (78) have been performed, and we use the superscript (0) to indicate that we are considering the case of pure spontaneous emission. Equation (77) is close to the expression of Bethe (1947) for the non-relativistic shift, but still contains divergent terms. Equation (78) indicates that the lifetime of level i is determined by spontaneous emission of resonant photons from level i to all levels lower than E_i in energy which are connected by the dipole matrix elements, in agreement with the results of Weisskopf and Wigner (1930a, b).

We very briefly discuss the divergent terms in (77): evaluating the integral we obtain

$$\Delta_i^{(0)} = -\frac{1}{6\pi^2\epsilon_0} \sum_c |\mu_{c,i}|^2 \left(\int k^2 dk - k_{c,i} \int k dk + k_{c,i}^2 \int dk - k_{c,i}^3 \mathbf{P} \int \frac{dk}{k + k_{c,i}} \right). \tag{79}$$

Now the first term in (79), which is cubically divergent, is exactly cancelled by $H_P^{(d)}$, as may be seen by comparing (79) with (3e) and (3f). This cancellation was first demonstrated by Power and Zienau (1959). The Thomas-Reiche-Kuhn sum rule

$$\sum_c |\mu_{c,i}|^2 k_{c,i} = \frac{e^2}{2mc} \tag{80}$$

may be used to write the second term of (79) as

$$(\Delta_i^{(0)})_2 = -\frac{1}{6\pi^2\epsilon_0} \frac{e^2}{2mc} \int k dk. \tag{81}$$

This expression is independent of the level quantum numbers i , and so it shifts each level equally. It may be removed therefore by redefining the zero of energy. The third term may be removed by mass renormalization, following Bethe (1947). Consequently, we are left with

$$\Delta_i^{(0)} = \frac{1}{6\pi^2\epsilon_0} \sum_c |\mu_{c,i}|^2 k_{c,i}^3 \mathbf{P} \int \frac{dk}{k + k_{c,i}}. \tag{82}$$

This expression is now identical to Bethe's for the non-relativistic Lamb shift. It is still divergent, but if we impose a cut-off at the Compton wavenumber, $K_C = mc$, we obtain

$$\Delta_i^{(0)} = \frac{1}{6\pi^2\epsilon_0} \sum_c |\mu_{c,i}|^2 k_{c,i}^3 \ln \left| \frac{mc}{k_{c,i}} \right|. \tag{83}$$

Consider now the stimulated contributions to Δ_i , which we write as $\Delta_i^{(n)}$. One easily obtains

$$\Delta_i^{(n)} = \frac{1}{8\pi^3 \epsilon_0} \sum_c |\mu_{c,i}|^2 \int d\Omega \cos^2\theta \int dk k^3 n(k, \Omega) P \left(\frac{1}{k - k_{c,i}} - \frac{1}{k + k_{c,i}} \right) \quad (84)$$

$$= \frac{1}{8\pi^3 \epsilon_0} \sum_c |\mu_{c,i}|^2 \int d\Omega \cos^2\theta \left[2k_{c,i} \int dk n(k, \Omega) k + k_{c,i}^3 P \int dk n(k, \Omega) \left(\frac{1}{k - k_{c,i}} + \frac{1}{k + k_{c,i}} \right) \right]. \quad (85)$$

The sum rule (80) may again be used to show that we may disregard the first term of (85). We note that expressions analogous to the first and third terms of (79) do not appear in the stimulated contribution; such terms which arise from the $(k + k_{i,c})^{-1}$ factor in (84) are in fact cancelled out by similar terms from the $-(k - k_{i,c})^{-1}$ factor. One is thus left with only the final term in (85). Combining (85) with (77) we obtain for the generalized Lamb shift

$$\Delta_i = \Delta_i^{(0)} + \Delta_i^{(n)} = \frac{1}{8\pi^3 \epsilon_0} \sum_c |\mu_{c,i}|^2 k_{c,i}^3 \int d\Omega \cos^2\theta P \left(\frac{n(k, \Omega)}{k - k_{c,i}} + \frac{n(k, \Omega) + 1}{k + k_{c,i}} \right). \quad (86)$$

This is the generalization of Bethe's formula to include stimulated processes.

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