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ABSTRACT

The theory of sequential decays of an unstable system is studied. Examples include the sequential emission of two or more photons by an excited atom which reaches its ground state via one or more intermediate levels, and the decay of an unstable particle into other unstable particles. To describe these phenomena, a factorization of the Green's function is introduced. This leads to a simple, and intuitively obvious, description of sequential decays. It also makes possible an assessment of the accuracy of this description.

I. INTRODUCTION

We consider the quantum mechanical description of a system undergoing a sequence of decays. An example of this is provided by the de-excitation of an atom radiating in sequence two or more photons. Other examples include the study of angular correlations in successive nuclear decays and the decay of an unstable daughter in particle physics.

Previous treatments of these phenomena have tended to be heuristic or have introduced approximations at the outset which have obscured many of the subtle features of sequential decays. In this paper we shall apply the Green's function method used by Goldberger and Watson[1] for single-step decays to a general description of multi-step decay processes. Somewhat related techniques have been used by Reff,[2] by Kroll,[3] and by Goldberger and Watson[4] for specific cases of two-step decays. An alternate formulation of the decay problem has been given recently by Mower.[5] His method treats as "closely coupled" all the states involved in a decay. This leads to the algebraic problem of inversion of a matrix whose dimensionality is the number of states considered. Our method takes account from the outset of the time ordering of sequential decays.

The value of the Green's function approach lies in the fact that it gives a rigorous formulation of multistep decays in which the usual description by a product of Breit-Wigner resonance factors is a natural first approximation. This is not true of ordinary (e.g., Rayleigh-Schrödinger) perturbation methods. Correction terms depending

on the ratio of level widths to level spacing may be estimated in a straightforward way. Qualitative statements about the time dependence of the decay may be obtained from the analytic behavior of the Green's function.[4]

We begin with a collection of some relevant results of the Goldberger-Watson[1] formulation of decay processes. A physical system is assumed to be described by a Hamiltonian H. This is written as $H = K + V$, where V is responsible for transitions between eigenstates of K. These eigenstates are written as $g_a, g_b, \cdots$ and satisfy the respective Schrödinger equations

$$K g_a = \epsilon_a g_a ,$$
$$K g_b = \epsilon_b g_b, \cdots . \tag{1.1}$$

Let us now suppose that at time $t = 0$ the system is in a discrete state g_a. We wish to calculate the probability that at time t it will be found in a small domain of continuum states g_b. This probability has the form

$$dP_b(t) = \sum_b' |\psi_b(t)|^2, \tag{1.2}$$

where the sum extends over the domain of states in question. For explicit evaluation we shall write this as

$$\sum_b{}' = d\rho_b \, d\epsilon_b \, , \tag{1.3}$$

in terms of the density $d\rho_b$ of states per unit energy interval. The quantity $\wp_b$ in Eq. (1.2) is defined as

$$\wp_b(t) = \frac{1}{2\pi i} \int_{C_2} d\epsilon \; e^{-i\epsilon t} \, (g_b, \, G(\epsilon) \, g_a), \tag{1.4}$$

where $G(\epsilon)$ is the Green's function

$$G(\epsilon) = (\epsilon - H)^{-1} \tag{1.5}$$

and the contour C_2 extends from $+\infty$ to $-\infty$ and lies above the real ϵ-axis.

The Green's function G can be written in the form

$$G \, g_a = F \, g_a \, G_a \, , \tag{1.6}$$

where F satisfies the equation

$$F = 1 + \frac{1}{\epsilon - K} \; P_a \, V \, F \, , \tag{1.7}$$

with

$$\begin{aligned} P_a &\equiv 1 - \Lambda_a \, , \\ \Lambda_a &\equiv g_a \, g_a{}^{+} \, . \end{aligned} \tag{1.8}$$

That is, Λ_a is the projection operator onto the initial discrete state g_a. The quantity G_a in Eq. (1.6) is

$$\begin{aligned} G_a(\epsilon) &= (g_a, G(\epsilon)\, g_a) \\ &= [\epsilon - \epsilon_a - R_a(\epsilon)]^{-1}, \end{aligned} \tag{1.9}$$

expressed in terms of the diagonal matrix element

$$R_a(\epsilon) = (g_a, R(\epsilon)\, g_a) \tag{1.10}$$

of the <u>transition operator</u>

$$R \equiv V F . \tag{1.11}$$

The matrix element of G appearing in Eq. (1.4) can be expressed in the form[1]

$$\begin{aligned} G_{ba}(\epsilon) &\equiv (g_b, G(\epsilon)\, g_a) \\ &= \frac{1}{\epsilon - \epsilon_b}\; R_{ba}(\epsilon)\; \frac{1}{\epsilon - \epsilon_a - R_a(\epsilon)}, \end{aligned} \tag{1.12}$$

where

$$R_{ba}(\epsilon) \equiv (g_b, R(\epsilon)\, g_a). \tag{1.13}$$

To evaluate the asymptotic transition probability

$$dP_b = \lim_{t\to\infty} dP_b(t) \tag{1.14}$$

we note[1] that the expression (1.12) has no singularities in the upper half ϵ-plane. The contour integral (1.4) may then be evaluated, as explained in Ref. 1, by lowering the contour at $+\infty$ below the real axis onto the second sheet of $G_{ba}(\epsilon)$. In the limit $t \to \infty$ the only significant contribution to $\wp_b$ comes from the pole at $G = \epsilon_b$ and we have

$$\wp_b \equiv \lim_{t\to\infty} \wp_b(t) = \frac{R_{ba}(\epsilon_b)\, e^{-i\epsilon_b t}}{\epsilon_b - \epsilon_a - R_a(\epsilon_b)} \,. \tag{1.15}$$

Equations (1.2) and (1.3) now lead to the result that

$$dP_b = \frac{d\rho_b\, d\epsilon_b\, |R_{ba}(\epsilon)|^2}{|\epsilon_b - \epsilon_a - R_a(\epsilon)|^2} \,. \tag{1.16}$$

The quantity $R_a(\epsilon_b)$ has the form

$$R_a(\epsilon_b) \equiv D_a - (i\Gamma_a/2) \,, \tag{1.17}$$

where D_a and Γ_a are real and $\Gamma_a \geqslant 0$. Indeed, when there are no channels into which the decay can go other than that represented by the g_b, we have[1]

$$\Gamma_a = 2\pi \int d\rho_b \, |R_{ba}(\epsilon_b)|^2 \, . \qquad (1.18)$$

In this case the probability of finding the system within the energy range ϵ_b to $\epsilon_b + d\epsilon_b$ is

$$dP_b = \frac{\Gamma_a}{2\pi} \frac{d\epsilon_b}{|\epsilon_b - \epsilon_a - R_a(\epsilon_b)|^2} \, . \qquad (1.19)$$

It is instructive to check our calculation by integrating Eq. (1.19) over all ϵ_b to show that $\int dP_b = 1$. Using Eq. (1.18) we find that

$$P_b = \int dP_b = \frac{1}{2\pi i} \int d\epsilon_b \left[\frac{1}{\epsilon_b - \epsilon_a - R_a(\epsilon_b)} - \frac{1}{\epsilon_b - \epsilon_a - R_a^*(\epsilon_b)} \right]$$

$$= \lim_{\eta \to 0(+)} \frac{1}{2\pi i} \int d\epsilon \left[g_a, \left(\frac{1}{\epsilon + i\eta - H} - \frac{1}{\epsilon - i\eta - H} \right) g_a \right]$$

$$= \int d\epsilon \left[(g_a, \delta(\epsilon - H) g_a \right]$$

$$= \sum_\lambda \int d\epsilon \, \delta(\epsilon - \epsilon_\lambda) \, |(\psi_\lambda, g_a)|^2 \qquad (1.20)$$

$$= 1,$$

where the ψ_λ and ϵ_λ represent a complete set of eigenfunctions and eigenvalues of H. When the states b are stable, but other states than g_b are accessible to the decay process we obtain instead of

Eq. (1.18) the result that

$$P_b < 1, \tag{1.21}$$

which is physically obvious.

When the states b are themselves unstable, so that further transitions will take place from b to lower states, our discussion leading to Eq. (1.16) is still formally valid, but not very useful because in this case $dP_b = 0$. To see this, let us consider a hydrogen atom which was initially in a $3D$ state, corresponding to g_a. In interpreting g_b to correspond to the $2P$ state plus an emitted photon, we see that $dP_b = 0$. That is, prior to time $t = \infty$, the atom will have undergone a subsequent transition to the $1S$ state. In this case it is clear that

$$R_{ba}(\epsilon_b) = 0. \tag{1.22}$$

We shall be concerned in the remainder of this paper with the adaptation of the above formalism to a study of sequential decays. Although our discussion will be general, it will be helpful on occasion to think of it as applying to the case of an excited atom decaying through a series of radiative transitions until it eventually reaches its stable ground state.

In Section II we shall obtain a formally exact factorization of the Green's function, of which Eq. (1.6) represents the first step. This will be applied in Section III to the description of a "unique

sequence" of decays, by which we mean a sequence for which each intermediate level of the parent system is known. In Section IV we discuss the accuracy of the approximations by which a simple description is possible. In Section V we discuss an "ambiguous sequence" of decays, for which the transitions lead to mixtures of states of the parent system. Finally, in Section VI we introduce generalized "Lee models" for which almost exact solutions to our equations may be obtained.

II. FACTORIZATION OF THE GREEN'S FUNCTION

The description of a two-step decay reviewed in the preceding section will now be generalized. We consider a physical system which decays from some initial state through a sequence of states to a final stable, lowest state. This "system" is supposed to emit some form of radiation with each transition.[6] The states in the sequence will be written as $g_a, g_{b_1}, \cdots g_{b_n}, g_d$. Following the notation of Eq. (1.1), these are assumed to be eigenstates of the Hamiltonian K :

$$\begin{aligned} K g_a &= \epsilon_a g_a , \\ K g_{b_1} &= \epsilon_{b_1} g_{b_1} , \\ &\vdots \\ K g_d &= \epsilon_d g_d . \end{aligned} \tag{2.1}$$

Here g_a is the initial state in which the system is at time $t = 0$ and g_d the final stable state. The complete Hamiltonian for the system is, as in the last section, written as $H = K + V$, where V is responsible for the transitions. Because of the term V in the Hamiltonian, at times $t > 0$ the system will be in a mixture of the states (2.1).

To take explicit account of the radiation emitted, we write

$$
\begin{aligned}
g_a &= \omega_\alpha \lambda_0 \ , \\
g_{b_1} &= \omega_{\beta_1} \lambda_{Q_1} \ , \\
&\vdots \\
g_{b_n} &= \omega_{\beta_n} \lambda_{Q_n} \ , \\
g_d &= \omega_\delta \lambda_Q \ .
\end{aligned} \tag{2.2}
$$

Here we have indicated the discrete internal states of the decaying system by u_α , $u_{\beta_1}, \cdots u_\delta$. Continuum states of the emitted radiation are indicated as λ_{Q_1} , $\lambda_{Q_2} \cdots \lambda_Q$. The "vacuum state" with respect to emitted radiation is λ_0 . The energies of the internal states are written as w_α , $w_1, \cdots w_n$, w_δ . The corresponding energies of the radiation emitted in each transition are u_1, $u_2, \cdots u_n, u$. Thus,[7]

$$
\begin{aligned}
\epsilon_a &= w_\alpha \ , \\
\epsilon_{b_1} &= w_1 + u_1 \ , \\
\epsilon_{b_2} &= w_2 + u_1 + u_2 \ , \\
&\vdots \\
\epsilon_d &= \omega_\delta + u_1 + u_2 + \cdots + u_n + u \ .
\end{aligned} \tag{2.3}
$$

Since the final state g_d is assumed to be stable (steady) we can supplement Eqs. (2.1) with

$$
"H\, g_d = \epsilon_d\, g_d" \ , \tag{2.4}
$$

valid at $t \to \infty$ in the transition amplitude. [Strictly speaking, the eigenstates of H include scattered waves of the emitted radiation

and coupling to many states ω. As we see from Appendix A, the emitted radiation does not overlap the system at late times, so scattering can be neglected.]

We desire the transition probability dP_d that, as $t \to \infty$, the system will be observed in some set of states d. As in Eq. (1.2), we have

$$\begin{aligned} dP_d &= \sum_d{}' \, |\psi_d|^2 \\ &= \sum_Q{}' \, |\psi_d|^2, \end{aligned} \tag{2.5}$$

where the sum over states d is equivalent to a sum over some desired range of the continuum states λ_Q. As in Eq. (1.4), we have

$$\psi_d = \lim_{t\to\infty} \frac{1}{2\pi i} \int_{C_2} d\epsilon \; e^{-i\epsilon t} \, (g_d, \, G(\epsilon) \, g_a), \tag{2.6}$$

where $G(\epsilon)$ is given by Eq. (1.5). Since the states λ_Q form a continuous set, we may generalize Eq. (1.3) to write the sum on Q in Eq. (2.5) as the multiple integral:

$$\sum_Q{}' = \int d\rho_1 \, du_1 \, d\rho_2 \, du_2 \cdots d\rho_n \, du_n \, d\rho \, du \; . \tag{2.7}$$

Here $d\rho_j$ $[j = 1,2,\cdots n]$ is the density of states per unit energy of the radiation emitted in the transition to the state $u_{\beta j}$ and $d\rho$ is the corresponding density for the last transition to u_δ.

To evaluate (2.5) we continue the process of factorization of G begun with Eqs. (1.6) and (1.7). In doing this we must recognize explicitly that with each transition the system may decay into a linear combination of several states ω_β. [For example, an atom may radiate from a pure state α into a mixture of states ω_β, differing in the azimuthal angular momentum quantum number.] We are thus led to supplement the projection operators (1.8) with an additional set into which the decay may go:

$$\begin{aligned} \Lambda_1 &= \sum_{\beta_1}{}' \sum_{Q_1}{}' g_{b_1} g_{b_1}{}^\dagger , \\ &\vdots \\ \Lambda_n &= \sum_{\beta_n}{}' \sum_{Q_n}{}' g_{b_n} g_{b_n}{}^\dagger , \\ \Lambda_d &= \sum_{Q}{}' g_d g_d{}^\dagger . \end{aligned} \tag{2.8}$$

Here the sums over the Q's may be chosen for convenience. We shall suppose for our applications that these sums extend over all directions of emission of the radiation and over those energies well on each side of each resonance line. From our study in Section IV of the accuracy of the resonance approximation we shall see that the range of energies covered by the projection operators should not be too much greater than the line widths. The reason for this is that transitions "off resonance" should be absorbed into the transition operators. The corresponding sums over $\beta_1 \cdots \beta_n$ in Eqs. (2.8) extend over those states of the system into

which transitions may go, as restricted by our assumed observation on the states g_d. We have, appropriately, assumed in (2.8) that the final observed state u_δ is unique. In addition to the projection operators (2.8) we shall find it convenient to define

$$\begin{aligned} E_1 &= 1 - \Lambda_1 , \\ &\vdots \\ E_n &= 1 - \Lambda_n , \\ E_d &= 1 - \Lambda_d , \\ P_a &= 1 - \Lambda_a , \\ P_1 &= E_1 P_a , \\ P_2 &= E_2 P_1 , \\ &\vdots \\ P_n &= E_n P_{n-1} . \end{aligned} \tag{2.9}$$

We note that $\Lambda_j \Lambda_k = \delta_{jk} \Lambda_k$, where Λ_j and Λ_k are any two of the projection operators in Eq. (2.8).

We begin with Eqs. (1.6) and (1.7). The operator F can be written in the form

$$\begin{aligned} F &\equiv F^{(1)} (F_1 + 1), \\ F_1 &\equiv \Lambda_1 (F - 1), \end{aligned} \tag{2.10}$$

where consistency evidently requires that

$$\Lambda_1 F^{(1)} = \Lambda_1 . \tag{2.11}$$

To evaluate F_1 we define the transition operators

$$\begin{aligned} \hat{R}^{(1)} &\equiv V F^{(1)} \\ \mathcal{R}^{(1)} &\equiv \Lambda_1 \hat{R}^{(1)} \Lambda_1 . \end{aligned} \tag{2.12}$$

Then, on inserting the first expression (2.10) into the right-hand side of Eq. (1.6), we find that

$$\begin{aligned} F_1 &= \Lambda_1 \frac{1}{\epsilon - K} P_a V F^{(1)} (F_1 + 1) \\ &= \frac{1}{\epsilon - K} \mathcal{R}^{(1)} F_1 + \Lambda_1 \frac{1}{\epsilon - K} \hat{R}^{(1)} . \end{aligned}$$

Solving this for F_1 gives us

$$\begin{aligned} F_1 &= \frac{1}{\epsilon - K - \mathcal{R}^{(1)}} \Lambda_1 \hat{R}^{(1)} \\ &= \Lambda_1 \frac{1}{\epsilon - K - \mathcal{R}^{(1)}} \hat{R}^{(1)} . \end{aligned} \tag{2.13}$$

The quantity $F^{(1)}$ is the solution of the equation

$$F^{(1)} = 1 + \frac{1}{\epsilon - K} P_1 V F^{(1)} . \tag{2.14}$$

To see this, we first note that $F = E_1 F + \Lambda_1 F$. This relation and the second of Eqs. (2.10) permit us to obtain from Eq. (1.7) the equation

$$E_1 F = E_1 + \frac{1}{E - K} P_1 V F .$$

Substitution of the first of Eqs. (2.10) and use of Eq. (2.14) lead to

$$\begin{aligned} E_1 F &= E_1 + \frac{1}{E - K} P_1 V F^{(1)} (F_1 + 1) \\ &= E_1 + E_1 (F^{(1)} - 1) (F_1 + 1) \\ &= E_1 + E_1 F^{(1)} (F_1 + 1) - E_1 F_1 - E_1 \\ &= E_1 F , \end{aligned} \tag{2.15}$$

since $E_1 F_1 = 0$.

Next, we write

$$\begin{aligned} F^{(1)} &\equiv F^{(2)} (F_2 + 1), \\ F_2 &\equiv \Lambda_2 (F^{(1)} - 1), \\ \hat{R}^{(2)} &\equiv V F^{(2)} , \\ R^{(2)} &\equiv \Lambda_2 \hat{R}^{(2)} \Lambda_2 , \end{aligned} \tag{2.16}$$

etc. Proceeding as before, we find that

$$F_2 = \frac{1}{\epsilon - K - \mathcal{R}^{(2)}} \Lambda_2 \hat{R}^{(2)}, \tag{2.17}$$

and

$$F^{(2)} = 1 + \frac{1}{\epsilon - K} P_2 V F^{(2)}. \tag{2.18}$$

We can evidently continue this factorization until we obtain finally

$$F = F^{(n)} (F_n + 1) (F_{n-1} + 1) \cdots (F_2 + 1) (F_1 + 1),$$

$$F_j = (\epsilon - K - \mathcal{R}^{(j)})^{-1} \Lambda_j \hat{R}^{(j)},$$

$$\hat{R}^{(j)} = V F^{(j)},$$

$$F^{(j)} = 1 + \frac{1}{\epsilon - K} P_j V F^{(j)}, \tag{2.19}$$

for $j = 1,2,\cdots n$.

The operators $\mathcal{R}^{(j)}$ $(j = 1,2,\cdots n)$ are undesirably complicated here, since in general they may contain matrix elements for scattering of radiation emitted prior to or accompanying the jth transition. We can eliminate this as follows. First, we define the "vacuum expectation" of $R^{(j)}$ (that is, vacuum with respect to the emitted radiation) as

$$\mathcal{R}_v^{(j)} = \sum_{\beta'_j}{}' \sum_{\beta_j}{}' \sum_{Q_j}{}' \left(\lambda_0 u_{\beta'_j}, \hat{R}^{(j)} \lambda_0 u_{\beta_j}\right) \times u_{\beta'_j} u_{\beta_j}{}^{\dagger} \lambda_{Q_j} \lambda_{Q_j}{}^{\dagger}, \tag{2.20}$$

where all sums extend over the states of (2.8) defining Λ_j. This corresponds to that part of $\mathcal{R}^{(j)}$ for which the emitted radiation does not interact again with the system (in F_j). Then

$$\mathcal{R}_s^{(j)} = \mathcal{R}^{(j)} - \mathcal{R}_v^{(j)} \tag{2.21}$$

represents the portion of $\mathcal{R}^{(j)}$ which describes scattering of some of the emitted radiation.

Equation (2.21) lets us write

$$F_j = \frac{\Lambda_j}{\epsilon - K - \mathcal{R}^{(j)}} \hat{R}^{(j)} = \frac{\Lambda_j}{\epsilon - K - \mathcal{R}_v^{(j)}} R^{(j)}, \tag{2.22}$$

where

$$R^{(j)} = \hat{R}^{(j)} + \mathcal{R}_s^{(j)} \frac{1}{\epsilon - K - \mathcal{R}^{(j)}} \hat{R}^{(j)}. \tag{2.23}$$

We note from Eqs. (2.20) and (2.23) that

$$\mathcal{R}_v^{(j)} = \sum_{\beta'_j,\beta_j} \sum_{Q_j} (\lambda_0 u_{\beta'_j}, R^{(j)} \lambda_0 u_{\beta_j}) \times u_{\beta'_j} u^{\dagger}_{\beta_j} \lambda_{Q_j} \lambda^{\dagger}_{Q_j}, \tag{2.24}$$

since the second term in Eq. (2.23) vanishes when there is no radiation to be scattered in the final state.

We anticipate that for many applications the second term in Eq. (2.23) is negligible and one can take

$$R^{(j)} \approx \hat{R}^{(j)}. \tag{2.25}$$

The assumption of an interestingly long-lived resonance will often imply a weak perturbation V. In such a case, rescattering of emitted radiation will be unimportant. It is of course to be observed that rescattering of radiation emitted in the jth transition can formally occur in Eq. (2.19) from any R-operator to the left of the F_j that emits it. (An advantage of the wave packet picture is that the tendency for emitted radiation to escape is more clearly seen.) We shall neglect this rescattering of radiation. An estimate of the error involved is made in Section IV.

Some further insight into the significance of the $R^{(j)}$ in Eq. (2.22) may be gained as follows. For the calculation $R^{(j)}$ from Eq. (2.23) the quantity $\mathcal{R}_v^{(j)}$ corresponds to a "self energy." For a long-lived state we might expect $\mathcal{R}_v^{(j)}$ to be negligibly small

everywhere except in the explicitly written propagator $(\epsilon - K - \mathcal{R}_v^{(j)})^{-1}$ in Eq. (2.22). (The operator Λ_j restricts us to energies very close to the energy shell.) If $\mathcal{R}_v^{(j)}$ may be omitted when one is calculating $R^{(j)}$ and $\hat{R}^{(j-1)}$, we have

$$R^{(j)} = \hat{R}^{(j-1)}, \tag{2.26}$$

as is shown in Appendix B. A more complete discussion of relations among the R-operators will be given in Section IV.

Equation (2.19) represents the fundamental result of this paper. In the subsequent sections we shall show how to apply it to the analysis of sequential decays.

In using Eqs. (2.19) to describe sequential decays we shall make three approximations in the calculation of dP_d [Eq. (2.5)]:

(1). The "one" terms in the quantities $(F_1 + 1)$, $(F_2 + 1)\cdots(F_n + 1)$ will be neglected. This neglect forces us to consider only transitions that go through the "resonant states." Since the neglected transitions are not associated with small resonance energy denominators, we expect their amplitudes relative to the resonance transitions to be of order

$$\frac{\Gamma}{\Delta w}, \tag{2.27}$$

where $\Gamma = O(|R|)$ is the width of the resonance transition and Δw is some characteristic energy of the decaying system.

(2) Rescattering of radiation once emitted will be neglected. This approximation is expected to be valid when the lifetime of a state is long compared with flight time of the emitted radiation from the parent system.

(3) The third approximation is not essential to our theory, but a convenience. In this approximation we suppose that relevant matrix elements of the transition operators $R^{(j)}$ can be treated as constants over energy intervals of the order of the level widths. It is anticipated that the relative error resulting from this approximation will be given by an expression of the form (2.27).

These approximations will be discussed farther in Section IV.

III. UNIQUE SEQUENCE OF DECAYS

In this section we discuss the application of the theory of Section II to a "unique sequence" of decays. By unique sequence we mean to imply that the observation made on the state d is sufficient for us to infer that each of the sequence of intermediate states ω_{β_1}, $\omega_{\beta_2}, \cdots \omega_{\beta_n}$ is unique. For such a decay we can then replace Eqs. (2.8) by the set

$$\begin{aligned} \Lambda_a &= g_a\, g_a^\dagger \\ \Lambda_1 &= \sum_{Q_1}{}' g_{b_1}\, g_{b_1}^\dagger, \\ &\vdots \\ \Lambda_n &= \sum_{Q_n}{}' g_{b_n}\, g_{b_n}^\dagger \\ \Lambda_d &= \sum_{Q}{}' g_d\, g_d^\dagger, \end{aligned} \tag{3.1}$$

where there are no sums over internal states. An example of a unique sequence would be the decay of a hydrogen atom from the 3D ($j_z = 0$) to the 2P ($j_z = 0$) to the 1S state. Observation that the polarization vector of the first photon was parallel to the axis of quantization would ensure this.

For a unique sequence the quantity $\mathcal{R}_v^{(j)}$ of Eq. (2.24) has the simple form

$$\mathcal{R}_v^{(j)} = R_j^{(j)} \sum_{Q_j}{}' \lambda_{Q_j}\, \lambda_{Q_j}^\dagger, \tag{3.2}$$

where $R_j^{(j)}$ is the complex number

$$R_j^{(j')}(\epsilon) = (\lambda_0 u_{\beta_j}, R^{(j)} \lambda_0 u_{\beta_j}). \tag{3.3}$$

Equation (2.22) now reads

$$F_j = \Lambda_j \frac{1}{\epsilon - K - R_j^{(j)}} R^{(j)}$$

$$= \sum_{Q_j}{}' g_{b_j} g_{b_j}^{\dagger} \frac{1}{\epsilon - \epsilon_{b_j} - R_j^{(j)}} R^{(j)}. \tag{3.4}$$

To illustrate our results, we consider first the special case of a three-level transition $a \to b_1 \equiv b \to d$, where d is stable. For this case Eq. (2.19) reads

$$F = F^{(1)}(F_1 + 1). \tag{3.5}$$

Use of Eqs. (1.6), (1.9), (2.12), (2.14), and (3.4) gives us

$$G_{ba}(\epsilon) \equiv (g_b, G\, g_a) = \frac{1}{\epsilon - \epsilon_b - R_1^{(1)}} R_{1a}^{(1)} \frac{1}{\epsilon - \epsilon_a - R_a(\epsilon)},$$

$$G_{da}(\epsilon) \equiv (g_d, G\, g_a)$$

$$= \frac{1}{\epsilon - \epsilon_d} R_{d1}^{(1)} \frac{1}{\epsilon - \epsilon_b - R_1^{(1)}} R_{1a}^{(1)} \frac{1}{\epsilon - \epsilon_a - R_a}$$

$$+ \frac{1}{\epsilon - \epsilon_d} R_{da}^{(1)} \frac{1}{\epsilon - \epsilon_a - R_a}. \tag{3.6}$$

Here, $R_{1a}^{(1)}(\epsilon) \equiv \langle g_{b_1}, R^{(1)} g_a \rangle$, etc. In deriving Eq. (3.6) we have set $\hat{R}_{d1} = R_{d1}$. This is correct, since for the final stable state $d(\lambda_Q, \lambda_{Q_1}) = 0$, which implies that $g_d^\dagger \, \mathcal{R}_s^{(1)} = 0$.

The second term in G_{da} above comes from the "1" term in Eq. (3.5). The quantity $R_{da}^{(1)}$ has no virtual states "b" and so describes decays which do not pass through these resonance "states." In accordance with approximation (1) made at the close of Section II, we shall neglect $R_{da}^{(1)}$ in G_{da}. The second approximation made at the close of Section II implies that we neglect in $R_{d1}^{(1)}$ any matrix elements which describe scattering of the radiation "emitted by $R_{1a}^{(1)}$."

The probability of finding the system in a state $g_b = \omega_\beta \lambda_{Q_1}$ at time t is given by Eq. (1.2), where now

$$\begin{aligned} \psi_b(t) &= \frac{1}{2\pi i} \int_{C_2} d\epsilon \; e^{-i\epsilon t} \; G_{ba}(\epsilon) \\ &\approx \frac{e^{-i(\epsilon_b + D_b)t} \; e^{-\Gamma_b t/2}}{\epsilon_b - \epsilon_a + R_1^{(1)} - R_a} \; R_{1a}^{(1)} \\ &\quad - \frac{e^{-i(\epsilon_a + D_a)t} \; e^{-\Gamma_a t/2}}{\epsilon_b - \epsilon_a + R_1^{(1)} - R_a} \; R_{1a}^{(1)} \; . \end{aligned} \tag{3.7}$$

Here we have evaluated the contour integral in a simple two-pole approximation, appropriate for long-lived states, as described in Ref. 1. We have also written

$$R_a(\epsilon_a + R_a) \approx R_a(\epsilon_b + R_b^{(1)}) \approx D_a - i\frac{\Gamma_a}{2} ,$$

$$R_1^{(1)}(\epsilon_a + R_a) \approx R_1^{(1)}(\epsilon_b + R_b^{(1)}) = D_b - i\frac{\Gamma_b}{2} .$$

The quantity $dP_b(t)$ is therefore[8]

$$dP_b(t) = \sum_{Q_1}' \frac{|R_{1a}^{(1)}|^2 e^{-\Gamma_b t}}{[(\epsilon_b - \epsilon_a + D_b - D_a)^2 + \frac{1}{4}(\Gamma_b - \Gamma_a)^2]} \times \left\{1 - 2\cos[(\epsilon_b - \epsilon_a + D_b - D_a)t]\, e^{(\Gamma_b - \Gamma_a)t/2} + e^{(\Gamma_b - \Gamma_a)t}\right\} . \tag{3.8}$$

We see that the expression (3.8) vanishes at $t \to \infty$, as was conjectured in Section I.

The conditions under which

$$2\pi \sum_{Q_1}' |R_{1a}^{(1)}|^2 \approx \Gamma_a \, du_1$$

will be discussed in Section IV.

The probability that as $t \to \infty$ we find the entire system in some range of stable states d is given by Eq. (2.5), where now [recall that we have agreed to set $R_{da}^{(1)} \approx 0$]

$$\gamma_d = \lim_{t\to\infty} \frac{1}{2\pi i} \int_{C_2} d\epsilon \; e^{-i\epsilon t} \, G_{da}(\epsilon)$$

$$= e^{-i\epsilon_d t} \; \frac{R_{dl}^{(1)} \; R_{la}^{(1)}}{(\epsilon_d - \epsilon_b - R_l^{(1)}) \; (\epsilon_d - \epsilon_a - R_a)} \; . \qquad (3.9)$$

Here we have used the second of Eqs. (3.6) and have deformed the contour onto the second sheet of G_{da}, as described in Ref. 1. Our notation here is such that

$$R_{dl}^{(1)} \equiv \lim_{\eta\to 0(+)} R_{dl}^{(1)} \; (\epsilon_d + i\eta),$$

$$R_{la}^{(1)} \equiv \lim_{\eta\to 0(+)} R_{la}^{(1)} \; (\epsilon_d + i\eta),$$

$$R_l^{(1)} \equiv \lim_{\eta\to 0(+)} R_l^{(1)} \; (\epsilon_d + i\eta) \equiv D_l - i \; \frac{\Gamma_l}{2},$$

$$R_a \equiv \lim_{\eta\to 0(+)} R_a(\epsilon_d + i\eta) \equiv D_a - i \; \frac{\Gamma_a}{2}, \qquad (3.10)$$

where D_a, D_l, Γ_a, and Γ_l are real and Γ_a, Γ_l are positive. In evaluating the contour integral above we have noted that Eq. (2.4) implies that $G_{da}(\epsilon)$ has a pole at ϵ_d on the real axis and no other poles on the real axis (barring accidental degeneracies).

On substituting (3.9) into Eq. (2.5), and on using Eq. (2.7), we obtain

$$dP_d = \frac{\Gamma'_1}{2\pi} \frac{\Gamma'_a}{2\pi} \frac{du_1\, du}{|\epsilon_d - \epsilon_b - R_1^{(1)}|^2 \, |\epsilon_d - \epsilon_a - R_a|^2}, \tag{3.11}$$

where

$$\Gamma'_a \equiv 2\pi \int d\rho_1 \; |R_{1a}^{(1)}|^2$$

$$\Gamma'_1 \equiv 2\pi \int d\rho \; |R_{d1}^{(1)}|^2. \tag{3.12}$$

Let us suppose our observation on the state d restricts the energy of the first emitted radiation to the range u_1 to $u_1 + du_1$, but does not restrict u. The probability of this is obtained by integrating (3.11) over all u. When

$$\left| \frac{d\, R_1^{(1)}(\epsilon)}{d\epsilon} \right|_{\epsilon = \epsilon_d} << 1,$$

$$\left| \frac{d\, R_a(\epsilon)}{d\epsilon} \right|_{\epsilon = \epsilon_d} << 1, \tag{3.13}$$

etc., we may treat $R_1^{(1)}$, R_a, Γ'_1, and Γ_a as constants in (3.11). (This is the third approximation mentioned at the close of Section II). Then, using Eqs. (2.3), we obtain

$$dP_d = \frac{du_1}{2\pi} \left(\frac{\Gamma'_1}{\Gamma_1}\right) \left(\frac{\Gamma'_a}{\Gamma_a}\right) (\Gamma_a + \Gamma_1)$$

$$\times \left[(u_1 - w_\alpha - D_a + w_1 + D_1)^2 + \tfrac{1}{4}(\Gamma_a + \Gamma_1)^2\right]^{-1}. \tag{3.14}$$

This has the Lorentz shape of Eq. (1.19), but of course contains level shifts D and widths Γ for states a and b.

In deriving Eq. (3.14) we have made all three of the approximations mentioned in Section II. First, we neglected the "non-resonant" $R_{da}^{(1)}$ term in Eq. (3.6). Second, we have neglected in (3.6) rescattering by $R_{dl}^{(1)}$ of radiation emitted in the first transition. Third, we treated $R_l^{(1)}$ and R_a as constants when integrating over u in Eq. (3.11) to obtain (3.14). The error arising from these approximations will be investigated in Section IV.

Let us now consider the sequence of $(n + 1)$ transitions $a \to b_1 \to b_2 \to \cdots d$ through the states (2.2). The expression (2.5) gives the probability of finding the system in a given set of states d as $t \to \infty$. In this case Eqs. (2.6) and (2.19) lead to the result

$$\gamma_d = \frac{e^{-i\epsilon_d t} R_{dn}^{(n)} R_{nn-1}^{(n)} \cdots R_{21}^{(2)} R_{1a}^{(1)}}{(\epsilon_d - \epsilon_{b_n} - R_n^{(n)})(\epsilon_d - \epsilon_{bn-1} - R_{n-1}^{(n-1)}) \cdots (\epsilon_d - \epsilon_a - R_a)}, \tag{3.15}$$

where we again make the first two approximations mentioned in Section II. The R's here are all evaluated at the energy $\epsilon_d + i\eta$ in the $\lim \eta \to 0(+)$. Following the notation of Eqs. (3.10) we write

$$\begin{aligned} R_j^{(j)} &\equiv D_j - i \frac{\Gamma_j}{2}, \\ R_a &\equiv D_a - i \frac{\Gamma_a}{2}, \end{aligned} \tag{3.16}$$

for $j = 1,2,\cdots n$. Also, as in Eqs. (3.12), we define

$$\Gamma'_a = 2\pi \int d\rho_1 \; |R_{1a}^{(1)}|^2,$$

$$\Gamma'_j = 2\pi \int d\rho_{j+1} \; |R_{j+1j}^{(j+1)}|^2, \quad j = 1,2,\cdots n-1,$$

$$\Gamma'_n = 2\pi \int d\rho \; |R_{dn}^{(n)}|^2, \tag{3.17}$$

where the notation of Eq. (2.7) has been used for the sums over states.

On inserting (3.15) into Eq. (2.5), we obtain the probability dP_d that the emitted radiations are in the intervals u_1 to $u_1 + d\,u_1, \cdots$:[9]

$$d\rho_d = \frac{\frac{\Gamma'_a}{2\pi}\,\frac{\Gamma'_1}{2\pi}\cdots\frac{\Gamma'_n}{2\pi}\; du_1\cdots du_n\, du}{|\epsilon_d - \epsilon_{b_n} - R_n^{(n)}|^2\cdots|\epsilon_d - \epsilon_a - R_a|^2}\,. \tag{3.18}$$

Let us suppose, for example, that only one of the energies, say u_σ, is measured. The probability that this energy lies in the range u_σ to $u_\sigma + d\,u_\sigma$, irrespective of the other energies, is obtained by integrating (3.18) over all the other energies. Again we assume that all the resonances are sufficiently narrow that we may neglect quantities of the order of (3.13) and that $R_a, \cdots R_n^{(n)}$, $\Gamma'_a, \cdots \Gamma_n$ as constants (our third approximation). An elementary integration now gives us the probability

$$dP_d = \frac{du_\sigma}{2\pi}\left(\frac{\Gamma'_n}{\Gamma_n}\right)\cdots\left(\frac{\Gamma'_1}{\Gamma_1}\right)\left(\frac{\Gamma'_a}{\Gamma_a}\right)$$

$$\times \frac{(\Gamma_\sigma + \Gamma_{\sigma-1})}{(u_\sigma + w_\sigma + D_\sigma - w_{\sigma-1} - D_{\sigma-1})^2 + \frac{1}{4}(\Gamma_\sigma + \Gamma_{\sigma-1})^2} \quad . \tag{3.19}$$

Except for normalization, this agrees with Eq. (3.14).

In deriving Eq. (3.19) we have made the same three approximations that were made in the derivation of Eq. (3.14).

IV. DISCUSSION OF APPROXIMATIONS

In this section we shall discuss the accuracy of the results obtained in the last section. We shall also study the relation between the quantities (3.16) and (3.17).

The exact expression for γ_d, for which (3.15) represented our approximation, is

$$\gamma_d = e^{-i\epsilon_d t} (g_d, R^{(n)} (F_n + 1) \cdots (F_1 + 1) g_a) (\epsilon_d - \epsilon_a - R_a)^{-1} . \quad (4.1)$$

Here all functions of ϵ are evaluated at $\epsilon = \epsilon_d + i\eta$ in the limit $\eta \to 0(+)$.

The first approximation is that of replacing (4.1) by

$$\gamma_d = e^{-i\epsilon_d t} (g_d, R^{(n)} F_n F_{n-1} \cdots F_1 g_a) (\epsilon_d - \epsilon_a - R_a)^{-1} . \quad (4.2)$$

This approximation is conceptually straightforward. It corresponds to neglecting transitions which are not resonant and which should not lead to sharply defined energies (to within the widths Γ) for the emitted radiations. Estimates of the relative contributions of non-resonant contributions can sometimes be obtained with dimensional arguments for Δw in (2.27). When this is not the case, a specific calculation must be made. An example of such a calculation is given in Section VI.

Using Eqs. (2.22), we may rewrite (4.2) in the form

$$\psi_d = e^{-i\epsilon_d t} \sum' \frac{R_{d_n}^{(n)} R_{nn-1}^{(n)} \cdots R_{21}^{(2)} R_{1a}^{(1)}}{(\epsilon_d - \epsilon_{b_n} - R_n^{(n)}) \cdots (\epsilon_d - \epsilon_a - R_a)} . \qquad (4.3)$$

The summation here is over intermediate states contained in the projection operators $\Lambda_1 \cdots \Lambda_n$ of Eqs. (2.22), but is restricted by the fact that g_d is a discrete state. If there were no rescattering of once emitted radiation, the intermediate states in (4.3) would be unique and this would reduce to (3.15). Thus, we can write the expression (4.3) as

$$\psi_d = \psi_d\,(3.15) + \psi_d\,(\text{scat}). \qquad (4.4)$$

Here $\psi_d\,(3.15)$ represents the quantity (3.15) and $\psi_d(\text{scat})$ the contribution from rescattering.

For estimating the magnitudes of the two terms in Eq. (4.4) it is convenient to render these dimensionless with multiplication by the factor $D^{1/2}$, where D is the weighting factor (2.7) of final states g_n:

$$D = \delta\rho_1\, \delta u_1 \cdots \delta\rho_n\, \delta u_n\, \delta\rho\, \delta u . \qquad (4.5)$$

To get an order-of-magnitude estimate, we write [see Eqs. (3.17)]

$$\begin{aligned} \delta u_j &\approx \Gamma_j , \\ \Gamma'_j &\approx \delta\rho_j \left| R_{jj-1}^{(j)} \right|^2 , \\ \Gamma_j &\approx \left| \epsilon - \epsilon_j - R_j^{(j)} \right| , \end{aligned} \qquad (4.6)$$

etc. This permits us to estimate the magnitude of (3.15) as

$$|D^{1/2}\ \gamma_d(3.15)| \approx \left[\left(\frac{\Gamma'_n}{\Gamma_n}\right)\cdots\left(\frac{\Gamma'_1}{\Gamma_1}\right)\left(\frac{\Gamma'_a}{\Gamma_a}\right)\right]^{1/2}, \quad (4.7)$$

which is consistent with Eq. (3.19).

Let us estimate $\gamma_d(\text{scat})$ by supposing that the radiation emitted in this transition to state $\omega_{\beta\sigma}$ is rescattered by the quantity $R_{\gamma\gamma}{}^{(\gamma)}_{-1}$. The characteristic magnitude of this can be obtained by supposing it to be the $\mathcal{R}_s{}^{(\gamma)}$ [see Eq. (2.23)]. That is, we suppose $R^{(\sigma)}$ to "emit" radiation of energy u'_σ while $\mathcal{R}_s{}^{(\gamma)}$ scatters this into its final observed state with energy u_σ. Using the arguments which led to the estimate (4.7), we are led to the expression

$$D^{1/2}\ \gamma_d(\text{scat}) \approx \frac{1}{\Gamma_\gamma}\int d\,u'_\sigma\, d\rho'_\sigma\ \ \mathcal{R}_s{}^{(\gamma)}\ \frac{1}{\epsilon - \epsilon'_\gamma - R_\gamma{}^{(\gamma)}}\ \hat{R}^{(\gamma)}$$

$$\cdots\ \frac{1}{\epsilon - \epsilon'_\sigma - R_\sigma{}^{(\sigma)}}\ R^{(\sigma)}\ \delta\rho_\sigma\ \delta u_\sigma \cdots \delta\rho_\gamma\ \delta u_\gamma \left(\frac{\Gamma'_a}{\Gamma_a}\right)^{1/2} \cdots, \quad (4.8)$$

where the integral over u'_σ is of course restricted to the narrow range of energies spanned by the states in Λ_σ. In the denominators above we have the energies

$$\epsilon'_\sigma = w_{\beta\sigma} + u'_\sigma + u_{\sigma-1} + \cdots + u_1 ,$$
$$\vdots$$
$$\epsilon'_\gamma = w_{\beta\gamma} + u_\gamma + \cdots + u'_\sigma + \cdots + u_1 ,$$
$$\epsilon_\gamma = w_{\beta\gamma} + u_\gamma + \cdots + u_\sigma + \cdots + u_1 . \qquad (4.9)$$

There are $\nu + 1 \equiv \gamma - \sigma + 1$ energy denominators in (4.8). In doing the integral over du'_σ we thus encounter $(\nu + 1)$ poles. When the expressions (3.13), etc., are small, we may estimate $\gamma_d(\text{scat})$ by supposing these to coalesce into a single pole of order $(\nu + 1)$. This gives

$$d^{1/2}\ \gamma_d(\text{scat}) \approx \frac{1}{\nu!} \int d\rho'_\sigma \ \frac{d^\nu}{(du_\sigma)^\nu} \left[R_s(\gamma)\, \hat{R}(\gamma) \ldots R(\sigma) \right] \qquad (4.10)$$
$$\times\ \delta\rho_\sigma\, \delta u_\sigma \cdots \delta\rho_\gamma\ \frac{\delta u_\gamma}{\Gamma_\gamma} \left(\frac{\Gamma'_a}{\Gamma_a} \right)^{1/2} \cdots .$$

The only sum over virtual states now is that explicitly indicated by the integral $\int d\rho'_\sigma$. To simplify our expression it is convenient to define a mean width Γ by the equation

$$(\Gamma)^\nu = \Gamma_\sigma\, \Gamma_{\sigma+1} \cdots \Gamma_{\gamma-1} .$$

This lets us write

$$D^{1/2}\ \gamma_d(\text{scat}) \approx [D^{1/2}\ \gamma_d(3.15)] \times \frac{1}{\nu!}\left(\frac{\Gamma}{\Delta w}\right)^{\nu} \int d\rho'_\sigma\ \mathcal{R}_s^{(\gamma)}\,, \qquad (4.11)$$

where Δw is a characteristic energy, such as was introduced in Eq. (2.27), which we take as representing the derivatives in (4.10).

To continue, let us suppose the radiation emitted in the transition to $\omega_{\beta\sigma}$ is a single particle. Then,

$$\int d\rho'_\sigma = \int \frac{d^3k'_\sigma}{du'_\sigma} \approx \frac{k_\sigma^2}{v_\sigma}\,, \qquad (4.12)$$

where k_σ is its momentum and v_σ its velocity. Now, the cross section for scattering this radiation is

$$\sigma \approx \frac{1}{v_\sigma} \int d\rho'_\sigma\ |\mathcal{R}_s^{(\gamma)}|^2 \approx \left(\frac{k_\sigma}{v_\sigma}\right)^2 |\mathcal{R}_s^{(\gamma)}|^2\,.$$

This, along with (4.11), lets us finally express Eq. (4.4) in the form

$$\gamma_d = \gamma_d(3.15)\left\{1 + \left[\frac{1}{\nu!}\left(\frac{\Gamma}{\Delta w}\right)^{\nu}\cdot\left(\frac{\sigma}{\pi_\sigma^2}\right)^{\frac{1}{2}}\right]\right\}, \qquad (4.13)$$

where $\pi_\sigma = k_\sigma^{-1}$.

Emitted radiation may also be scattered by the operator $\hat{R}^{(\gamma)}$ [see Eq. (4.8)]. We estimate this for the largest contribution, which occurs for $\gamma = \sigma + 1$. This is [we use perturbation theory]

$$D^{1/2}\,\gamma_d(\text{scat}) = \frac{1}{\epsilon - \epsilon_\gamma - R_\gamma^{(\gamma)}} \int d\rho'_\sigma \; du'_\sigma$$

$$\times\; V_{\gamma\sigma} \;\; \frac{\langle\sigma|P_\gamma|\sigma\rangle}{\epsilon - \epsilon''_\gamma} \;\; V_{\sigma\sigma'} \;\; \frac{1}{\epsilon - \epsilon'_\sigma - R_\sigma^{(\sigma)}}$$

$$\times\; R_{\sigma'\sigma-1}^{(\sigma)} \;\; \delta\rho_\sigma \;\; \delta u_\sigma \;\; \delta\rho_\gamma \;\; \delta u_\gamma \cdots . \qquad (4.14)$$

Here $R_{\sigma'\sigma-1}^{(\sigma)}$ emits radiation into a state σ'. This is scattered to a state σ by $V_{\sigma\sigma'}$. Because of the projection operator P_γ this must involve a transition to a "distant" state $\omega'_{\beta\sigma}$ of the parent system. This lets us write $\epsilon - \epsilon''_\gamma \approx \Delta w$, as in Eq. (4.11). Finally $V_{\gamma\sigma}$ can be written as $R_{\gamma\sigma}^{(\gamma)}$, since it emits radiation into the state γ. All of this lets us put (4.14) into the form

$$D^{1/2}\,\gamma_d(\text{scat}) \approx \frac{\Gamma}{\Delta w} \int d\rho'_\sigma \; V_{\sigma\sigma'} \; [D^{1/2}\,\gamma_d(3.15)] .$$

On identifying $V_{\sigma\sigma'}$ with $Q_s^{(\gamma)}$, we see that this has just the form of (4.11), with $\nu = 1$. Thus, our estimate of (4.11) is generally valid.

The result (4.13) may be understood qualitatively as follows. Radiation emitted in the transition to state $\omega_{\beta\sigma}$ leaves its source with a "flight time" $\Delta t = \Delta w^{-1}$. The probability that within the time Δt there will be ν other radiations emitted, when they are emitted at random and with a mean rate Γ^{-1}, is $(1/\nu!)\,(\Gamma\,\Delta t)^\nu$

when $\Gamma \Delta t << 1$. The factor $\left(\sigma_\sigma / \pi_\sigma^2\right)^{1/2}$ represents the probability amplitude that scattering will actually occur.

For the special case of dipole radiation emitted by an excited atom we may estimate Δw from the explicit matrix element to be $\Delta w \approx u_\sigma$, the energy of the photon. Since Γ/u_σ is a very small quantity for atomic transitions, we see that the correction term in (4.13) is small indeed.

We have now seen that the three approximations described at the close of Section II and used in our analysis all require the smallness of a ratio of the form $\Gamma/\Delta w$, where Γ is a level width and Δw is a characteristic energy of the system. To estimate the order of this ratio, we must of course consider a specific physical system.

We turn now to a study of the relations between the Γ'_j of Eqs. (3.17) and the Γ_j of Eqs. (3.16). The argument which led to Eq. (85b) of Chapter 8, Ref. 1, permits us to write

$$\Gamma_j = \lim_{\eta \to 0(+)} 2\pi \left(g_{bj} , \hat{R}^{(j)\dagger} (\epsilon_d + i\eta) \right.$$

$$\left. \times \delta(\epsilon_d - K) P_j \hat{R}^{(j)} (\epsilon_d + i\eta) g_{bj} \right). \qquad (4.15)$$

When we know that the transition is a unique sequence, and there are no alternate channels, so the states g_{bj+1} follow the g_{bj}, we might be tempted to write

$$\Gamma_j \rightarrow 2\pi \int d\rho_{j+1} \; |\hat{R}_{j+1,j}^{(j)}|^2 \; \delta(\epsilon_d - \epsilon_{bj+1}) \, d\epsilon_{bj+1} \, . \tag{4.16}$$

To see the relation with Eq. (3.17), we consider

$$\begin{aligned} \hat{R}_{j+1j}^{(j)} &= (g_{bj+1} \; V \, F^{(j)} \, g_{bj}) \\ &= (g_{bj+1} \; V \, F^{(j+1)} \, (F_{j+1} + 1) \, g_{bj}) \\ &= \left[\hat{R}_{j+1}^{(j+1)} \; \frac{1}{\epsilon - \epsilon_{bj+1} - R_{j+1}^{(j+1)}} + 1 \right] \hat{R}_{j+1j}^{(j+1)} \\ &= \frac{\epsilon - \epsilon_{bj+1}}{\epsilon - \epsilon_{bj+1} - R_{j+1}^{(j+1)}} \; \hat{R}_{j+1j}^{(j+1)} \, . \end{aligned} \tag{4.17}$$

It is evident from Eq. (4.17) that the expression (4.16) <u>vanishes</u>. The correct Γ_j does not of course vanish if there is a finite transition rate from the state g_{bj}. The error made in using (4.16) is that the g_{bj+1}'s are not eigenstates of H, so a transition into these states is meaningless, other than as a transient phenomenon.

When the final state is the stable state g_d, then Eq. (4.14) may be used. In this case the level shift $R_d^{(d)}$ vanishes and $\hat{R}_{j+1j}^{(j)} = \hat{R}_{j+1j}^{(j+1)}$. This tells us that

$$\Gamma_n = \Gamma'_n \,. \tag{4.18}$$

[When there are alternate decay channels from the state g_n, Eq. (4.18) is of course no longer true.]

An approximate relation between the Γ_j and the Γ'_j may be easily obtained. We illustrate this for the quantity Γ_a, for which we expect $\Gamma_a > \Gamma'_a$ if there are decay routes through other channels than that of the g_{b_1}. From Eqs. (2.10) and (2.12) we have

$$\begin{aligned} R_a(\epsilon_d) &= (g_a, V F^{(1)} (F_1 + 1) g_a) \\ &= R_a^{(1)}(\epsilon_d) + \sum_{Q_1}{}' \frac{R_{a1}^{(1)} R_{1a}^{(1)}}{\epsilon_d - \epsilon_{b_1} - R_1^{(1)}} \,, \end{aligned} \tag{4.19}$$

where the sum on Q_1 is limited by the projection operator Λ_1. The term $R_a^{(1)}$ contains no virtual states g_{b_1} and thus describes decay through alternate channels. When our "small parameter" $\Gamma/\Delta w$ is indeed negligible, as we have been assuming, $R_{a1}^{(1)} \approx [R_{1a}^{(1)}]^*$. This permits us to obtain from Eq. (4.19)[10]

$$\begin{aligned} \Gamma_a &\equiv -2 \, \mathrm{Im} \, [R_a(\epsilon_d)] \\ &= -2 \, \mathrm{Im} \, [R_a^{(1)}(\epsilon_d)] + 2\pi \sum_{Q_1}{}' \delta(\epsilon_d - \epsilon_{b_1}) \, |R_{1a}^{(1)}|^2 \\ &\quad \times \left[1 + O\left(\frac{\Gamma}{\Delta w}\right) \right]. \end{aligned}$$

On neglecting contributions of order $\Gamma/\Delta w$, we obtain

$$\Gamma_a = -2\ \mathrm{Im}\,[R_a^{(1)}(\epsilon_d)] + \Gamma'_a\ , \tag{4.20}$$

with similar relations between Γ_1 and Γ'_1, etc.

The physical interpretation of Eq. (4.20) is obvious: Γ_a is the total width of the transition from g_a, while Γ'_a is the partial width of the transition through ω_{β_1}. This is completely consistent with Eq. (3.14). On integrating that equation over u_σ we obtain

$$P_d = \left(\frac{\Gamma'_n}{\Gamma_n}\right)\cdots\left(\frac{\Gamma'_1}{\Gamma_1}\right)\left(\frac{\Gamma'_a}{\Gamma_a}\right)\ . \tag{4.21}$$

This is just the probability that the decay goes through the sequence of states $\omega_{\beta_1}\cdots\omega_{\beta_n}$, ω_δ prescribed by our observations on the state g_d.

V. AMBIGUOUS SEQUENCE OF TRANSITIONS

When the three approximations mentioned at the close of Section II are valid, the description of decay through an ambiguous set of states is straightforward. In this case there will be more than one term in the sums over β_n in some of the projection operators (2.8). Then the quantities $\mathcal{R}_v^{(j)}$ in Eq. (2.22) contain matrix elements coupling the states $\omega_{\beta j}$ contained in Λ_j.

Often these non-diagonal elements of $\mathcal{R}_v^{(j)}$ may be neglected, however. (An example is provided by the case of an atom falling through a level consisting of a set of degenerate states differing only in magnetic quantum number.) When this is possible, the treatment given in Section III needs but little modification. Instead of Eq. (3.15), we have

$$\psi_d = e^{-i\epsilon_d t} \sum_{\beta_1 \cdots \beta_n}{}' \frac{R_{d\beta_n}^{(n)} R_{\beta_n \beta_{n-1}} \cdots R_{\beta_1 a}^{(1)}}{(\epsilon_d - \epsilon_{\beta n} - R_{\beta n}^{(n)}) \cdots (\epsilon_d - \epsilon_a - R_a)} . \tag{5.1}$$

Here we have written

$$R_{\beta j \beta j-1}^{(j)} = (g_{bj}, R^{(j)} g_{bj-1}) ,$$
$$R_{\beta j}^{(j)} = (\lambda_0 \omega_{\beta j}, R^{(j)} \omega_{\beta j} \lambda_0) , \tag{5.2}$$

etc.

Let us apply Eq. (5.1) to the case of a three-step transition $a \to b_1 \to d$, where there are two levels $\beta_1 = 1$ and $\beta'_1 = 2$ of the system at the intermediate step. Let us also suppose that we observe

the angle of emission of the particles radiated in the two steps, but not their energy. The probability dP_d is then

$$
\begin{aligned}
dP_d &= \sum_Q{}' \; |\psi_d|^2 \\
&= d\rho \, d\rho_1 \int du_1 \, du_2 \; |\psi_d|^2 \\
&= \frac{(2\pi)^2}{\Gamma_a} \, d\rho \, d\rho_1 \left\{ \frac{|R_{d1}^{(1)} \, R_{1a}^{(1)}|^2}{\Gamma_1} \right. \\
&\quad \left. + \frac{|R_{d2}^{(1)} \, R_{2a}^{(1)}|^2}{\Gamma_2} + \left[\frac{(R_{d1}^{(1)} \, R_{1a}^{(1)})^* \, (R_{d2}^{(1)} \, R_{2a}^{(1)})}{\frac{\Gamma_1 + \Gamma_2}{2} - i \, (D_1 - D_2)} + \text{c.c.} \right] \right\} .
\end{aligned}
\tag{5.3}
$$

Here

$$
\begin{aligned}
R_1^{(1)} &= D_1 - i \, \frac{\Gamma_1}{2} , \\
R_2^{(2)} &= D_2 - i \, \frac{\Gamma_2}{2} ,
\end{aligned}
\tag{5.4}
$$

etc.

When $\Gamma_1 = \Gamma_2$ and $D_1 = D_2$, Eq. (5.3) reduces to the expression

$$
dP_d = \frac{(2\pi)^2}{\Gamma_a \, \Gamma_1} \, d\rho \, d\rho_1 \left| \sum_{\ell=12} R_{d\ell}^{(1)} \, R_{\ell a}^{(1)} \right|^2 . \tag{5.5}
$$

This is of a form familiar in the theory of successive angular correlations.[11]

We illustrate, finally, an example for which off-diagonal matrix elements of $\mathcal{R}_v^{(j)}$ must be kept, using the model just described of a two-state second level. We write

$$R_{1a}^{(1)} = (\omega_1 \lambda_{Q_1}, R^{(1)} g_a) ,$$

$$R_1^{(1)} = (\omega_1 , R^{(1)} \omega_1) ,$$

$$R_{12}^{(1)} = (\omega_1 , R^{(1)} \omega_2) ,$$

etc. We also write $\epsilon_{b_1} = \epsilon_1$, ϵ_2 and

$$F_{11} = (\omega_1 \lambda_{Q_1}, F_1 g_a) ,$$

etc. Then Eq. (2.22) provides the coupled equations

$$\begin{aligned} [\epsilon - \epsilon_1 - R_1^{(1)}] \; F_{11} - R_{12}^{(1)} \; F_{12} &= R_{1a}^{(1)} \\ [\epsilon - \epsilon_2 - R_2^{(1)}] \; F_{12} - R_{21}^{(1)} \; F_{11} &= R_{2a}^{(1)}, \end{aligned} \tag{5.6}$$

which may be solved algebraically for F_{11} and F_{12}. From there we find for $\mathcal{G}_d$ the expression

$$\psi_d = e^{-i\epsilon_d t} \frac{\dfrac{R_{d1}^{(1)} R_{12}^{(1)} R_{1a}^{(1)}}{\epsilon_d - \epsilon_2 - R_2^{(1)}} + R_{d1}^{(1)} R_{1a}^{(1)}}{\left(\epsilon_d - \epsilon_1 - R_1^{(1)} - \dfrac{R_{12}^{(1)} R_{21}^{(1)}}{\epsilon_d - \epsilon_2 - R_2^{(1)}}\right)\left(\epsilon_d - \epsilon_a - R_a\right)}$$

$$+ \text{ same term with "1" and "2" interchanged .} \qquad (5.7)$$

It should be noted that for such decays the simple Lorentz line shape can be replaced by a much more complex line.

VI. GENERALIZED LEE MODEL

To illustrate and clarify the theory presented in the preceding sections, we consider a model analogous to Lee's field theory.[12] In the model, the perturbing potential V connects only successive steps in a sequence of scalar photon emissions, $a \to b_1 \to b_2 \to \cdots \to d$. Thus, we require

$$(g_{b_1}, V g_a) = v(k_1)$$

$$(g_{b'_2}, V g_a) = \delta_{k_{m1}, k'_{m1}} v(k_2) \tag{6.1}$$

$$(g_{b_3}, V g_{b'_2}) = \delta_{k_{m1}, k'_{m1}} \delta_{k_{m2}, k'_{m2}} v(k_3), \text{ etc.}$$

Here g_{b_1}, g_{b_2}, etc. are restricted to a narrow range of photon energies about the resonant value. All matrix elements of V vanish unless they are of type (6.1) or the Hermitean conjugate.

For a three-step cascade we have

$$R_{db}^{(1)} = V_{db}$$

$$R_{bb}^{(1)} = V_{bd} \frac{1}{\epsilon - K} V_{db}$$

$$R_{da}^{(1)} = 0$$

$$R_{ba}^{(1)} = V_{ba} \qquad \text{(Equation 6.2 continued)}$$

$$R_{aa} = V_{ab} \frac{1}{\epsilon - K - R_{bb}^{(1)}} V_{ba} \tag{6.2}$$

and thence

$$G_{da} = \frac{1}{\epsilon - K} V_{db} \frac{1}{\epsilon - K - R_{bb}^{(1)}} V_{ba} \frac{1}{\epsilon - K - R_a} . \tag{6.3}$$

Thus, the "one" terms of (2.19) do not appear, and only the resonant sequence contributes to the decay. It is easy to extend this result for cascades of any number of steps. Of course, the absence of transitions skipping one or more steps follows directly from the construction of V.

The calculation of G_{da} is reduced to quadrature unless $R_{bb}^{(1)}$ contains rescattering terms. Because of our artificial requirement that V vanish except for transitions near the energy shell, rescattering may occur only if the two photons in state g_d have nearly the same energy. Then we see

$$\begin{aligned}(g_b', R\, g_b) &= \delta_{\underset{\sim}{k}'\underset{\sim}{k}} \sum_{\underset{\sim}{q}}{}' \frac{[v(q)]^2}{\epsilon - w_d - k - q + i\eta} \\ &\quad + \frac{\theta(k')\,\theta(k)\,v(k')\,v(k)}{\epsilon - w_c - k - k'} \\ &\equiv \delta_{\underset{\sim}{k}',\underset{\sim}{k}} \mathcal{R}_1^{(1)} + \mathcal{R}_s^{(1)}(\underset{\sim}{k}',\underset{\sim}{k}) ,\end{aligned} \tag{6.4}$$

where $\theta(k)$ is a unit step function which vanishes for $\underline{k}$ outside

the range of $\underset{\sim}{q}$ included in $\sum'_{\underset{\sim}{q}}$.

Following the argument of Section IV, we may estimate the rescattering correction to G_{ba} as

$$(g_b,\ G\ g_a) \approx \frac{1}{\epsilon - w_b - k - \mathcal{R}_1^{(1)}}\ v(k)\ \frac{1}{\epsilon - w_a - \mathcal{R}_a}\ [1 + \theta(k)\ X]. \tag{6.5}$$

The correction is given to lowest order by

$$\begin{aligned} X &= -2\pi i\ \frac{d}{dk'} \int d\rho\ (k')\ [v(k')]^2 \Big|_{k'=k} \\ &= -2i\ \Gamma(b \to d)\ \frac{d}{dk'}\ \ell n\ [k'\ v(k')] \Big|_{k'=k} \\ &= -i\ \Gamma\ T\ . \end{aligned} \tag{6.6}$$

The characteristic time $T \equiv (2/k) + r$ is analogous to the "delay time" which is familiar in discussions of scattering problems.[13] Recalling the discussion in Section IV, we may call T the "escape time" for the photon. The quantity $r = 2\ d\ \ell n\ v/dk$ is a measure of the size of the decaying system. In general, we expect $r \lesssim 1/k$, since k is a characteristic frequency of the system, and the maximum possible frequency, for components traversing a distance r with the velocity of light, would be simply $k_{max} \approx 1/r$. Thus, we conclude,

$$\frac{\delta\ G_{ba}}{G_{ba}}\ \text{rescattering} = O\left(\frac{\Gamma}{k}\right), \tag{6.7}$$

in agreement with the general conclusion of Section IV.

We turn now to the estimation of off-energy-shell effects. To do this we broaden the definition of our Lee model potential, allowing $v(k)$ to be different from zero over a range of energies of the order $w_a - w_b$. Then we have,[14] from (4.20),

$$\Gamma_a = \Gamma_a' - 2\ \mathrm{Im}\ \mathcal{R}_a^{(1)}(w_a) \equiv \Gamma_a' + \Gamma_a'' ,$$

$$\mathcal{R}_a^{(1)} = \int \frac{d^3k}{(2\pi)^3} \quad \frac{v(k)^2\ [1 - \Theta(k)]}{\epsilon - w_b - k - \mathcal{R}_b^{(1)}} , \tag{6.8}$$

$$\mathcal{R}_b^{(1)} = \int \frac{d^3q}{(2\pi)^3} \quad \frac{v(q)^2}{\epsilon - w_d - k - q + i\eta} ,$$

where we neglect rescattering terms in $\mathcal{R}_b^{(1)}$. The decay rate for virtual states far from the energy shell is

$$\Gamma_a''(w_a) = \int \frac{d^3k}{(2\pi)^3} \quad \frac{d^3q}{(2\pi)^3}\ 2\pi\ \delta(w_a - w_d - k - q)\ [1 - \Theta(k)] \cdot \tag{6.9}$$

$$\times \quad \frac{v(k)^2\ v(q)^2}{(w_a - w_b - k - \mathrm{Re}\ \mathcal{R}_a^{(1)})^2 + (\mathrm{Im}\ \mathcal{R}_b^{(1)})^2} .$$

The projection operator $1 - \Theta$ allows us to neglect $\mathcal{R}_b^{(1)}$ in the denominator, so that Γ_a'' is given accurately by ordinary second-order perturbation theory. For simplicity, we have treated the photons $\underset{\sim}{k}$ and $\underset{\sim}{q}$ as distinguishable, but Bose statistics could easily be taken into account.

Neglecting the variation of v with $\underline{k}$, we may estimate the decay rate Γ_a when w_b is much greater than w_a, so that resonant intermediate states are excluded. Neglecting k in the denominator of (6.9), we have

$$\Gamma_a \approx \Gamma_a'' = \frac{16}{15}\left(\frac{1}{w_a - w_b}\right)^2 \frac{(w_a - w_d)}{\pi} \gamma^2 , \qquad (6.10)$$

where γ is the decay rate for the resonant sequence in the case $w_a - w_b = w_b - w_d$. Thus, the decay rate in the absence of resonant intermediate states is suppressed by a factor of order Γ/u, with $u^{-1} = (w_a - w_d)/(w_a - w_b)^2$.

On the other hand, even when w_b lies between w_a and w_d, there will still be a non-resonant contribution Γ_a'' to the total decay rate Γ_a. We estimate Γ_a'' for $w_a - w_b = w_b - w_d$ with $\theta(k)$ taken to vanish when $|w_a - w_b - k|$ exceeds $A\gamma$ $(A \gg 1)$:

$$\Gamma_a'' \approx \frac{\gamma^2}{2\pi} \frac{1}{w_a - w_b}\left(\int_0^{1-\zeta} + \int_{1+\zeta}^{2}\right) dx \frac{x^2(2-x)^2}{(1-x)^2} , \qquad (6.11)$$

$$\zeta = A\gamma/(w_a - w_b) << 1.$$

This gives

$$\Gamma_a'' = \frac{\gamma}{\pi}\left(\frac{1}{A} - \frac{8}{3}\frac{\gamma}{w_a - w_b}\right) + O\left[A\left(\frac{\gamma}{w_a - w_b}\right)^2\right] . \qquad (6.12)$$

The term $\Gamma/\pi A$ simply compensates the effect of truncating the contribution Γ_a' defined as resonant—instead of integrating the resonant energy factor over all intermediate energies. Thus this term should be omitted when Γ_a' is evaluated, as in (4.20), by replacing the resonance factor with a delta-function. The second term, $-8\gamma^2/3\pi(w_a - w_b)$, comes mainly from the reduction in two-photon phase space at the extreme values of k. Combining (6.7) and (6.10), we have verified for an explicit model the result

$$\Gamma_a = \Gamma_a' \left[1 + 0\left(\frac{\Gamma_a'}{u}\right)\right], \tag{6.13}$$

where u is given by a typical photon frequency.

The Lee model permits us to see in a simple example the importance of proper order of operations in the factorization of the Green's function. Consider the problem of inverse decay: Given a state g_a containing one photon, what is the probability as a function of time to observe the state g_b with no photons, and a higher internal energy $(w_b > w_a)$?

Having phrased the question thus, we are tempted to write

$$(g_b,\ G\ g_a) \quad \frac{1}{\epsilon - \omega} \quad \sum_{\underset{\sim}{k'}} v(k')\ G_a(k',\ k), \tag{6.14}$$

with

$$G_a(\underset{\sim}{k'},\ \underset{\sim}{k}) = \left(g_b',\ \frac{1}{\epsilon - K - \mathcal{R}_a}\ g_b\right)$$

$$\mathcal{R}_a(\underset{\sim}{k'},\ \underset{\sim}{k}) = \frac{v(k')\ v(k)}{\epsilon - w_b}\ . \tag{6.15}$$

Note that (6.14) yields a meaningful expression for the transition rate $-2\ \mathrm{Im}\ (g_a,\ R\ g_a)$ only if g_a is taken as a normalized state $\omega_a\ \varphi(\underset{\sim}{k})$, where $\varphi(\underset{\sim}{k})$ is a wave packet for the photon. Otherwise one is speaking of a transition from an unnormalizable state of definite momentum to the normalized state g_b, and the rate for this is undefined. These considerations do not affect the following discussion, but should be kept in mind for applications of the Green's function formalism. Evaluation of (6.14) is difficult, since (6.15) shows that the pole at $\epsilon = w_b$ is spurious, and the inversion required to evaluate G_a is a non-trivial operation. However, we may approach the problem in a different way. It is easy to see that in our model, as in any theory with time-reversal invariance, the Green's function is symmetric,

$$(g_b,\ G\ g_a)\ =\ (g_{\bar{a}},\ G\ g_{\bar{b}})\ =\ (g_a,\ G\ g_b)\,, \tag{6.16}$$

where the states $g_{\bar{a}}$ and $g_{\bar{b}}$ have opposite spins and momenta from g_a and g_b, and the last equality in (6.16) is special to our model. Thus, we may replace (6.14) with

$$(g_b,\ G\ g_a)\ =\ \frac{1}{\epsilon\ -\ w_a\ -\ k\ +\ i\eta}\ v(k)\ \frac{1}{\epsilon\ -\ w_b\ -\ \bar{R}_b}\,, \tag{6.17}$$

with

$$\bar{R}_b\ =\ \sum_{\underset{\sim}{k}}\ \frac{v(k)^2}{\epsilon\ -\ w_a\ -\ k\ +\ i\eta}\ .$$

What we have done is simply to obtain the Green's function for $a \to b$ by transposing the G we already know how to compute, that for the decay $b \to a$. Needless to say, even if time-reversal invariance did not hold, we could carry out the factorization for inverse decay by using transposed F operators obeying

$$F^T = 1 + F^T V \frac{P}{E - K} . \tag{6.18}$$

We conclude that, in general, the factorization of G should proceed from "top" to "bottom" of a cascade, even for inverse processes.

FOOTNOTES AND REFERENCES

* This work was supported in part by the United States Atomic Energy Commission and in part by a grant from the Office of Scientific Research of the United States Air Force.

1. M. L. Goldberger and K. M. Watson, Collision Theory (John Wiley & Sons, Inc., New York, 1964); see Chapter 8.

2. The earliest use of these techniques seems to be that of I. Reff, Phys. Rev. 91, 150 (1953).

3. N. M. Kroll, Quantum Optics and Electronics, ed. by C. DeWitt, A. Blandin, and C. Cohen-Tannoudji (Gordon and Breach, New York, 1964).

4. Reference 1, p. 454.

5. L. Mower, Phys. Rev. 142, 799 (1966).

6. Our terminology is suggested by the example of a radiating atom. For some phenomena involving unstable systems the choice of the "decaying system" may be ambiguous.

7. If it is necessary to take account of the recoil energy of the system, this can be included in the λ's and u's.

8. This expression was given in Ref. 1, p. 454, where it was derived by a different method and somewhat more specialized assumptions.

9. If we desire the probability that the directions of the emitted radiation are restricted to certain angular intervals, we of course do not integrate over all $d\rho_1 \cdots d\rho$.

10. In estimating the error due to the lack of Hermiticity of $R_{al}^{(1)}$ we used Eq. (84), Chapter 8, of Ref. 1. On substituting a δ-function for the imaginary part of $\left(\epsilon_d - \epsilon_{b_1} - R_1^{(1)}\right)^{-1}$, an error of relative order $(\Gamma/\Delta w)$ is again incurred.

11. Decay angular correlations are discussed by R. M. Steffen and H. Frauenfelder in Alpha, Beta and Gamma Ray Spectroscopy, ed. by K. Siegbahn (North-Holland Publishing Co., Amsterdam, 1965), Vol. 2, p. 997; see also M. E. Rose, Elementary Theory of Angular Momentum (John Wiley & Sons, New York, 1957), Chapter 10.

12. T. D. Lee, Phys. Rev. 95, 1329 (1954). Decays in the Lee Model are discussed in detail by M. M. Levy, Nuovo Cimento 13, 115 and 14, 612 (1959). He does not discuss sequential decays, but does treat the problem of renormalization associated with the self-energy of the decaying state. This problem is trivial in our model because of the cutoff imposed on the potential.

13. E. P. Wigner, Phys. Rev. 98, 145 (1955), M. L. Goldberger and K. M. Watson, ibid. 127, 2284 (1962).

14. In obtaining (6.8) we do not have to make the approximation associated with (4.20) that off-diagonal elements of $R^{(1)}$ are Hermitean, since this is an exact statement in the Lee model.

APPENDIX A. DECAY INTO WAVE PACKET STATES

It is sometimes helpful in describing a decay process to specify the state of the emitted radiation with wave packets. For example, the use of wave packets makes it easy to see that Eq. (2.4) is valid at times such that the radiation has escaped from the parent system.

To illustrate, we consider an atom in the initial state ω_α which makes a transition to the state ω_β, emitting a photon of definite polarization. The wave packet state is taken to be

$$L_{\underset{\sim}{p},\underset{\sim}{r}} \equiv \int d^3k\, a(\underset{\sim}{k} - \underset{\sim}{p})\; e^{-i\underset{\sim}{k}\cdot\underset{\sim}{r}}\; \lambda_{\underset{\sim}{k}}\,, \tag{A.1}$$

where $\lambda_{\underset{\sim}{k}}$ is a photon eigenstate of momentum $\underset{\sim}{k}$ and $L_{\underset{\sim}{p},\underset{\sim}{r}}$ corresponds to a photon having components of momentum very close to $\underset{\sim}{p}$, centered at $\underset{\sim}{r}$ in position space. A typical final state is then

$$g_b = \omega_\beta\, L_{\underset{\sim}{p},\underset{\sim}{r}}\,. \tag{A.2}$$

The decay probability is given by (Eq. (1.2), this time with

$$\gamma_b(t) = \frac{1}{2\pi i} \int d^3k\, a^*(\underset{\sim}{k} - \underset{\sim}{p})\, e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}} \int_{C_2} d\epsilon\; e^{-i\epsilon t}(\omega_\beta\, \lambda_{\underset{\sim}{k}},\, G(\epsilon)\, \omega_d)\,. \tag{A.3}$$

As $t \to \infty$, we have

$$\psi_b = \int d^3k \; a^*(\underset{\sim}{k} - \underset{\sim}{p})\, e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}}\, e^{-i\epsilon_k t}\, \frac{R_{ka}(\epsilon_k)}{\epsilon_k - \epsilon_a - \mathcal{R}_a(\epsilon_k)} \quad . \qquad (A.4)$$

Here we have written ϵ_k for the energy of the state $\omega_\beta \lambda_{\underset{\sim}{k}}$ and R_{ka} for the transition matrix element into that state. In the following we shall assume that the R's are slowly varying so that $R_{ka}(\epsilon_k) \approx R_{pa}(\epsilon_p)$ and $\mathcal{R}_a(\epsilon_k) \approx \mathcal{R}_a(\epsilon_p)$.

We now consider two cases. First, suppose the time duration T of the wave packet pulse at a given point in space is long compared to the lifetime of the decay (or, equivalently, $\Delta\epsilon_k << \Gamma$). Then, at times $t >> T$, we may evaluate ψ_b by using the approximation $\epsilon_{\underset{\sim}{k}} \approx \epsilon_p + c\,\hat{p}\cdot(\underset{\sim}{k} - \underset{\sim}{p})$ to obtain

$$\psi_b \approx W(c\,\hat{p}\,t)\; \mathcal{R}_{pa}(\epsilon_p)\; \frac{e^{-i\epsilon_p t}}{\epsilon_p - \epsilon_a - \mathcal{R}_a(\epsilon_p)} \quad . \qquad (A.5)$$

Here

$$W(\underset{\sim}{x}) \equiv \int d^3\underset{\sim}{k}\; a^*(\underset{\sim}{k} - \underset{\sim}{p})\; e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}}\; e^{-i\underset{\sim}{x}\cdot(\underset{\sim}{k}-\underset{\sim}{p})}$$

is the complex conjugate wave packet in position space.

A natural choice of wave packet amplitude for studying decays would be a function obeying

$$W(\underset{\sim}{x}) = 0 \quad x << \mathcal{R}$$

$$W(\underset{\sim}{x}) = 1 \quad x >> \mathcal{R}\,, \tag{A.6}$$

where $\mathcal{R}$ is a distance significantly greater than the size of the atom. With the choice (A.6) the transition amplitude (A.5) is non-vanishing only when the photon has completely escaped from the atom and cannot interact with it again. Thus, (A.5) is equivalent to Eq. (1.15) for $ct >> \mathcal{R}$.

Let us now turn to a second case, appropriate to certain meson decays, in which the wave packet of the photon or other decay product has a narrow time resolution (compared to the decay life-time) but still a broad spatial resolution (compared to the dimensions of the decaying system). To obtain the appropriate limit of (A.4) we use

$$X^{-1} = i \int_0^\infty d\tau\, e^{iX\tau}\,,$$

$$X = \epsilon_k - \epsilon_a - \mathcal{R}_a(\epsilon_k) \equiv \epsilon_k - \epsilon'_a\,, \tag{A.7}$$

and again evaluate the $\mathcal{R}$'s at $\underset{\sim}{k} = \underset{\sim}{p}$ to obtain

$$\psi_b \approx i\, \mathcal{R}_{pa}(\epsilon_p) \int_0^\infty d\tau \int d^3k\; a^*(\underset{\sim}{k} - \underset{\sim}{p})\; e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}}\; e^{-i\epsilon_k(t-\tau)} \times e^{-i\epsilon' a\tau}$$

$$\approx i\, \mathcal{R}_{pa}(\epsilon_p) \int_0^\infty d\tau\, e^{-i\epsilon_a'\tau}\; W(c(t-\tau)\hat{p})$$

$$\approx (i/c)\; \mathcal{R}_{pa}(\epsilon_p)\; e^{-i\epsilon_a t}\; e^{-i(D-i\Gamma/2)(t-r\cdot\hat{p}/c)}\; \theta(t - \underset{\sim}{r}\cdot\hat{p}/c) \times \tilde{W}\left(\frac{\epsilon_a - \epsilon_p}{c},\, 0\right) . \qquad (A.8)$$

Here, $\theta(t)$ is the Heaviside function

$$\theta(t) = 1\,,\ t > 0$$

$$= 0\,,\ t < 0$$

and $\tilde{W}$ is a wave packet of mixed arguments in momentum and position

$$\tilde{W}(k_{\parallel} - p,\, \underset{\sim}{x}_\perp) \equiv 2\pi \int d^2k_\perp\; a^*(\underset{\sim}{k} - \underset{\sim}{p})\; e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}}\; e^{-i\underset{\sim}{k}\cdot\underset{\sim}{x}_\perp}$$

peaked about $k_{\parallel} \equiv \underset{\sim}{k}\cdot\hat{p} = p$ and $\underset{\sim}{x}_\perp \equiv \underset{\sim}{x} - \underset{\sim}{x}\cdot\hat{p}\,\hat{p} = \underset{\sim}{r}_\perp$.

The time dependence of $|\psi_b|^2$ is given by

$$|\psi_b|^2 \sim e^{-\Gamma(t-\underset{\sim}{r}\cdot\hat{p}/c)}\,, \qquad (A.9)$$

in agreement with intuition.

APPENDIX B. DERIVATION OF EQUATION (2.26)

When we can ignore the difference between $\mathcal{R}_s(j)$ and $\mathcal{R}(j)$ in Eq. (2.23), we can write this as

$$R^{(j)} = \hat{R}^{(j)} + \mathcal{R}_s(j) \frac{1}{\epsilon - K - \mathcal{R}_s(j)} \hat{R}^{(j)} . \tag{B.1}$$

Now,

$$\begin{aligned} \hat{R}^{(j-1)} &= V + V \frac{1}{a} P_{j-1} \hat{R}^{(j-1)} \\ &= V + \hat{R}^{(j-1)} \frac{P_{j-1}}{a} V , \end{aligned} \tag{B.2}$$

where $a \equiv \epsilon - K$. If we substitute the right-hand side of (B-1) into this, we have

$$\begin{aligned} \hat{R}^{(j-1)} &= V + \left[1 + \mathcal{R}_s(j) \frac{1}{a - \mathcal{R}_s(j)}\right] \hat{R}^{(j)} \frac{\Lambda_j + E_j}{a} P_{j-1} V \\ &= V + \left[1 + \mathcal{R}_s(j) \frac{1}{a - \mathcal{R}_s(j)}\right] \left[\hat{R}^{(j)} - V + \mathcal{R}_s(j) \frac{1}{a} V\right] \\ &= \left[1 + \mathcal{R}_s(j) \frac{1}{a - \mathcal{R}_s(j)}\right] \hat{R}^{(j)} = R^{(j)} . \end{aligned} \tag{B.3}$$

Here we have used our assumption that $\mathcal{R}_v(j)$ may be neglected in setting

$$\mathcal{R}_s^{(j)} \frac{1}{a - \mathcal{R}_s^{(j)}} \hat{R}^{(j)} \Lambda_j$$

$$= \mathcal{R}_s^{(j)} \frac{1}{a - \mathcal{R}_s^{(j)}} \mathcal{R}_s^{(j)} . \tag{B.4}$$