Sequential Decay Theory and Sequential Transitions*

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A theory of sequential decay of an unstable system is presented using the projection-operator algebra suggested by the recent projection-operator approach to reaction theory. The formulation is an attempt to provide an alternative to perturbation theory. While exact, the theory is cast in a phenomenological form that is suggestive in its interpretation and may be extended so as to apply to sequential transitions in general. Examples treated include the transition between unstable states of an atom induced by an intense electromagnetic field as well as a problem in resonance fluorescence. Higher-order corrections to the more familiar theory are a natural consequence of the description of sequential decay and sequential transitions presented here.

1. INTRODUCTION

THE theory of the decay of unstable states has recently been presented on the basis of the resolvent operator and has been applied to a number of simple models with some success.[1] The formulation itself has been modified and somewhat simplified[2] through the use of the language of nuclear reaction theory in the form presented by Feshbach.[3] As a step in the further development of the techniques, we apply it to the problem of sequential decays as well as to sequential transitions.

As an extension of the Goldberger-Watson operator formulation of decay theory, the theory of sequential decays has recently been presented by Goldhaber and Watson and applied to simple cascade models as well as to a modification of the Lee model.[4] In this paper, we simplify the formulation and illustrate its applicability to types of problems not normally included in the concept of sequential decay. The value of the formulation, of course, is that while it is exact, it still may be cast in a phenomenological form that is suggestive in interpretation. As a first illustration of the extended concept of sequential decay theory, we treat the problem of the transition between unstable states induced by an intense electromagnetic field. Here the additional broadening due to the induced transitions follows naturally from our formulation and does not require a separate transformation to decouple the states of interest from the remaining background spectrum of states. The sequential decay theory presented here effectively removes the virtual transitions and leaves only real transitions present. The resulting formulation clarifies a discussion of the intensity shifts in atomic beam experiments.

As a further illustration of the versatility of the concept of sequential decay, we treat the problem of resonance fluorescence under double resonance conditions. Here again, the additional broadening as well as shifts due to the induced transitions follows naturally from our formalism. By using the language of projection operators, we retain sufficient generality to treat induced hyperfine transitions in either the excited or ground state manifold of states.

In the following section we review briefly the theory of decay theory based on the projection theory language of Feshbach. For details concerning the handling of the poles of the Green's function, reference is made to either Goldberger and Watson[1] or to the author's work where additional references will be found. Following this brief review, the concept of sequential decay is introduced and treated using a sequence of projection operators. For use in the actual applications of this method, a number of formal relations between the level shift operators associated with this sequence are derived.

2. FORMAL DERIVATION

As a brief review, we consider a system that is initially in a given state. In the presence of a perturbation, the system may become unstable and make a transition from the original state; the transition probability may be calculated if we can follow the evolution of the system in time. If initially the system was in the ath eigenstate of the Hamiltonian K, then through the presence of the perturbation V, at time t the system will have evolved to the state

$$|\psi(t)\rangle = e^{-iHt}|a\rangle, \tag{1}$$

where the Hamiltonian of the system in the presence of the perturbation is given by H,

$$H \equiv K+V \tag{2}$$

and

$$K|a\rangle = E_a|a\rangle, \tag{3}$$

and we have set $\hbar=1$. The transition amplitude at time t between the states $|a\rangle$ and $|b\rangle$ of the spectrum of K is given by

$$I_{ba}(t) \equiv \langle b|\psi(t)\rangle = \langle b|e^{-iHt}|a\rangle, \tag{4}$$

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[1] M. L. Goldberger and K. M. Watson, *Collision Theory* (John Wiley & Sons, Inc., New York, 1964), Chap. 8.
[2] L. Mower, Phys. Rev. 142, 799 (1966).
[3] H. Feshbach, Ann. Phys. (N. Y.) 19, 287 (1962).
[4] A. S. Goldhaber and K. M. Watson, Phys. Rev. 160, 1151 (1967).

which may be expressed in terms of the resolvent operator or Green's function for the system as

$$I_{ba}(t)=(2\pi i)^{-1}\int_c dE\, e^{-iEt}G_{ba}(E)\,, \tag{5}$$

where

$$G_{ba}(E)\equiv\langle b|\,(E-H)^{-1}\,|a\rangle\,.$$

Here, the contour c runs from $+\infty$ to $-\infty$ above the singularities of $G(E)$ on the real axis. For the problem of interest, the spectrum of H will not be discrete, but will be a mixture of a discrete and a continuum with several discrete states in the continuum. Hence, the evaluation of $I_{ba}(t)$ by contour methods requires the method of analytical continuation to separate out the singularities from the continuum. For the techniques used in this separation, reference is made to Goldberger and Watson.[1] To facilitate this separation, we introduce the following projection operators of the unperturbed Hamiltonian K:

$$\Lambda_a+P_a=1;\quad \Lambda_aP_a=P_a\Lambda_a=0,\quad \Lambda_a{}^2=\Lambda_a\,, \tag{6}$$

where Λ_a is the projection operator for the initial state. If the initial state is an isolated state, then Λ_a is a simple projection operator,

$$\Lambda_a=|a\rangle\langle a|\,. \tag{7}$$

However, if the initial state is in a manifold of closely separated or closely coupled states, then Λ_a may be the sum of simple projection operators projecting onto the manifold of initial state.

In terms of these projection operators, it is clear that for the calculation of the transition amplitudes we need to known two projections of the Green's function: $\Lambda_aG(E)\Lambda_a$ and $P_aG(E)\Lambda_a$. The first projection includes only those matrix elements within the initial state manifold, while the second projection gives the matrix elements between the initial states and any other state of the spectrum of the unperturbed Hamiltonian K. To determine these projections, we start with the identity

$$(E-H)G(E)=1 \tag{8}$$

and first multiply from the right by Λ_a, and then from the left successively by Λ_a and P_a. The two operator equations that result,

$$\begin{aligned}&\Lambda_a(E-\Lambda_aH\Lambda_a)\Lambda_aG(E)\Lambda_a-\Lambda_aHP_aG(E)\Lambda_a=\Lambda_a\,,\\&-P_aH\Lambda_aG(E)\Lambda_a+P_a(E-P_aHP_a)P_aG(E)\Lambda_a=0\,,\end{aligned} \tag{9}$$

may be formally solved for the two projections desired. On the assumption that $P_a(E-P_aHP_a)P_a$ has an inverse, the two projections are given by

$$\begin{aligned}&P_aG(E)\Lambda_a=P_a(E-P_aHP_a)^{-1}P_aH\Lambda_aG(E)\Lambda_a\,,\\&\Lambda_aG(E)\Lambda_a=\Lambda_a(E-K-\Lambda_aR(E)\Lambda_a)^{-1}\,,\end{aligned} \tag{10}$$

where we have introduced the level shift operator $R(E)$ defined by

$$R(E)=V+VP_a(E-P_aHP_a)^{-1}P_aV\,. \tag{11}$$

To facilitate the subsequent analysis, we note that the following identity holds:

$$P_a(E-P_aHP_a)^{-1}P_aV=P_a(E-K)^{-1}P_aR(E)\,, \tag{12}$$

which follows from a simple expansion of $R(E)$ given by (11). Thus, we rewrite (10) as

$$P_aG(E)\Lambda_a=P_a(E-K)^{-1}P_aR(E)\Lambda_aG_{aa}(E)\,,$$

where

$$G_{aa}(E)\equiv\Lambda_aG(E)\Lambda_a\,. \tag{13}$$

If the initial state is isolated, then the subsequent analysis for the determination of $I_{ba}(t)$ or $I_{aa}(t)$ follows that of Goldberger and Watson.[1] On the other hand, if transitions between coupled unstable states are desired, then the analysis follows that of the author.[2] Here, however, we are interested in developing a formalism for the transition amplitude when the system is known to pass through a number of states (possibly unstable) in making the transition from the initial state $|a\rangle$ to the final state $|b\rangle$. One method of taking these steps into account would be simply to increase the size of the original manifold of states spanned by Λ_a. Such a method would treat all the states in the transition sequence on an equal basis and would eventually involve the inversion of a matrix whose rank would depend upon the number of states in the sequence. As an alternative procedure, in the next section we introduce a modification of the method of Goldhaber and Watson based on a sequence of partial projections.[4] This method seems to be a natural extension of the partitioning technique.[5]

Theory of Sequential Decay

If in the transition between the initial and final state the system has a possibility of passing through a number of intermediate states, then this fact must be made evident. If the expressions for the projections of $G(E)$ as given by (13) are exact, then the information desired must reside in the level shift operator given by (11). To make these statements transparent, we introduce a sequence of projection operators Λ_i corresponding to each of the i intermediate states. Again, the states may be isolated, in which case the associated projection operator is simple, or they may be degenerate or closely coupled, in which case the associated projection operator spans a manifold of states. We will also find it useful to introduce the following projection operators related to the Λ_i:

$$\begin{aligned}&P_a=1-\Lambda_a\,,\\&P_1=(1-\Lambda_a)(1-\Lambda_1)=1-\Lambda_a-\Lambda_1\,,\\&\vdots\\&P_i=(1-\Lambda_a)\prod_{j=1}^{i}(1-\Lambda_j)=1-\Lambda_a-\sum_{j=1}^{i}\Lambda_j\,.\end{aligned} \tag{14}$$

[5] P.-O. Löwdin, J. Math. Phys. 3, 969 (1962); 6, 1341 (1965); and the earlier references listed there.

What is now necessary is the explicit factorization of $R(E)$ to make clear the steps involved in the transition sequence. To realize this, a step-by-step analysis will be followed.

If in going from the initial state $|a\rangle$ the system may pass through an intermediate state, then we are actually interested only in a part of the projection $P_aG(E)\Lambda_a$, namely,

$$P_1G(E)\Lambda_a=P_1(E-K)^{-1}P_1R(E)\Lambda_aG_{aa}(E). \quad (15)$$

The remaining part, namely, $\Lambda_1G(E)\Lambda_a$, is a measure of the transition amplitude between the initial state $|a\rangle$ and $|1\rangle$ *as a final state.* Hence, we need the projection of $R(E)$,

$$P_1R(E)\Lambda_a=P_1V\Lambda_a+P_1VP_a(E-P_aHP_a)^{-1}P_aV\Lambda_a. \quad (16)$$

The subsequent analysis will be considerably simplified if we write out the various projections of

$$P_a(E-P_aHP_a)^{-1}$$

onto the two subspaces spanned by Λ_1 and P_1 making up the total space spanned by P_a:

$$P_1+\Lambda_1=P_a. \quad (17)$$

The algebra necessary to find the four projections of $P_a(E-P_aHP_a)^{-1}$ desired is similar to that used to derive (9) and is a special case of a more general derivation to be given shortly. We shall find that the level shift operator may be reexpressed as

$$\begin{aligned}R(E)&=R^{(1)}(E)+R^{(1)}(E)\Lambda_1G^{(1)}(E)\Lambda_1R^{(1)}(E)\\&=R^{(1)}(E)[1+\Lambda_1G^{(1)}(E)\Lambda_1R^{(1)}(E)],\end{aligned} \quad (18)$$

where $R^1(E)$ is defined by

$$R^{(1)}(E)\equiv V+VP_1(E-P_1HP_1)^{-1}P_1V. \quad (19)$$

The new Green's function $G^{(1)}(E)$ is defined by the expression

$$\Lambda_1G^{(1)}(E)\Lambda_1\equiv\Lambda_1(E-\Lambda_1K-\Lambda_1R^{(1)}(E)\Lambda_1)^{-1}. \quad (20)$$

An inspection of the definitions of both $R^{(1)}(E)$ and $\Lambda_1G^{(1)}(E)\Lambda_1$, shows that in the relationship between $R(E)$ and $R^{(1)}(E)$ as given by (18) we have achieved the desired factorization: The dependence on the two projectors P_1 and Λ_1 is both explicit and separate (that is, not in the form P_1+V_1). Thus, the desired projection of the Green's function is given by

$$\begin{aligned}P_1G(E)\Lambda_a=&P_1(E-K)P_1R^{(1)}(E)\\&\times[1+\Lambda_1G^{(1)}(E)\Lambda_1R^{(1)}(E)]\Lambda_aG_{aa}(E).\end{aligned} \quad (21)$$

We see that the first term on the right describes the direction transition between the initial state and the final state, while the second includes $|1\rangle$ as an intermediate state. This interpretation follows from an inspection of (19) where we see that the matrix elements in $P_1R^{(1)}\Lambda_a$ do not include any contributions from state $|1\rangle$ as an intermediate state.

Now, if in going from the initial to the final state the system must pass not through one intermediate state but through two intermediate states, then we simply repeat the above analysis. Here the projection $P_1G(E)\Lambda_a$ contains two contributions,

$$P_1G(E)\Lambda_a=P_2G(E)\Lambda_a+\Lambda_2G(E)\Lambda_a, \quad (22)$$

where the first refers to that portion of $G(E)$ that has matrix elements between the initial and the final states while the second has matrix elements between the initial state and the second intermediate state only. The above expression follows from the relation

$$P_1=P_2+\Lambda_2 \quad (23)$$

between the projection operators. On noting the explicit form of Eq. (21) and the interpretation we gave for it, we see that we need to focus attention on the first factor $R^{(1)}(E)$ which includes the interaction energy between the first intermediate state and all subsequent states. Hence, we proceed to factor $R^{(1)}(E)$ in much the same way that we did with $R(E)$. Again, in the factorization procedure of the level shift operator $R^{(1)}(E)$ as given by (19) we need the four projections of $P_1(E-P_1HP_1)^{-1}P$ with P_1 related to P_2 through (23). We find that $R^{(1)}(E)$ may be factored as

$$R^{(1)}(E)=R^{(2)}(E)\Lambda_2G^{(2)}(E)\Lambda_2R^{(2)}(E) \quad (24)$$

$$=R^{(2)}(E)[1+\Lambda_2G^{(2)}(E)\Lambda_2R^{(2)}(E)], \quad (25)$$

with the level shift operator $R^{(2)}$ defined as

$$R^{(2)}(E)\equiv V+VP_2(E-P_2HP_2)^{-1}P_2V, \quad (26)$$

and the Green's function $G^{(2)}(E)$ defined by

$$\Lambda_2G^{(2)}(E)\Lambda_2=\Lambda_2[E-\Lambda_2K-\Lambda_2R^{(2)}(E)\Lambda_2]^{-1}. \quad (27)$$

Hence, the desired projection of the complete Green's function describing transitions between the initial state $|a\rangle$ to a final state with the possibility of passing through two intermediate states is given by

$$\begin{aligned}P_2G(E)\Lambda_a=&P_2(E-K)^{-1}P_2R^{(2)}(E)\\&\times[1+\Lambda_2G^{(2)}(E)\Lambda_2R^{(2)}(E)]\\&\times[1+\Lambda_1G^{(1)}(E)\Lambda_1R^{(1)}(E)]\Lambda_aG_{aa}.\end{aligned} \quad (28)$$

To provide some measure of physical interpretation we expand the above expression:

$$\begin{aligned}P_2G(E)\Lambda_a=&P_2(E-K)^{-1}P_2\\&\times[R^{(2)}(E)+R^{(2)}(E)\Lambda_2G^{(2)}(E)\Lambda_2R^{(2)}(E)\\&+R^{(2)}(E)\Lambda_1G^{(1)}(E)\Lambda_1R^{(1)}(E)\\&+R^{(2)}(E)\Lambda_2G^{(2)}(E)\Lambda_2R^{(2)}(E)\Lambda_1G^{(1)}(E)\\&\times\Lambda_1R^{(1)}(E)]\Lambda_aG_{aa}(E).\end{aligned} \quad (29)$$

An inspection of the form of both $R^{(1)}(E)$ and $R^{(2)}(E)$ allows for the following interpretation. In (29), the first term in the square brackets has matrix elements between the initial and the final state for direct transitions only. The second term describes transitions that pass

through the second but not the first intermediate state; the third term describes transitions that pass through the first intermediate state and possibly the second. Finally, the last term describes a transition passing through both intermediate states in the sequence 1, 2. Clearly, if we replaced the remaining two factors of $R^{(1)}(E)$ in terms of $R^{(2)}$ as given by (25), the extension to (29) would include a term describing a transition that passed through the first intermediate state, but not the second, as well as a term describing a transition passing through both intermediate states in the sequence 2, 1. In addition there would be a term describing a transition passing first through state 2, then through state 1, and finally passing through state 2 again. The presence of this last term destroys the otherwise symmetric character of Eq. (29). However, one should note that the Green's functions $G^{(1)}$ and $G_{aa}(E)$ contain $R^{(1)}(E)$ and $R(E)$ explicitly and are therefore implicit functions of $R^{(2)}(E)$. If the two states were discrete, then the further factoring of the Green's functions and subsequent manipulations would result in a transition probability exhibiting a symmetry in the paths involving states 1 and 2. However, if continua are associated with the states 1) and 2), then the further manipulations must be handled with *care.*

Clearly, in attempting to generalize the above factorization so as to include the possibility of i intermediate states or steps, we want to avoid the complications introduced by the presence of alternative paths. Hence, we turn our attention to Eq. (28), which is a statement of the projection of the complete Green's function for transitions between an initial state $|a\rangle$ and a final state with the possibility of passing through at most two intermediate states. In this factored form, the generalization so as to include the possibility of i intermediate states or steps in the transition process is straightforward. The pertinent projection of the Green's function is

$$P_iG(E)\Lambda_a=P_i(E-K)^{-1}P_iR^{(i)}(E)\times\prod_{j=1}^{i}[1+\Lambda_jG^{(j)}(E)\Lambda_jR^{(j)}(E)]\Lambda_aG_{aa}(E). \quad (30)$$

This result is found by simply repeating the steps leading up to Eq. (28). The expression for the projection of the Green's function includes not only the contribution arising from the sequence of i steps ordered from 1 to i, but also the contribution from all other nonordered sequential transitions as well. The former consists of the single term remaining after dropping the 1 in each of the i factors in the above expression. Here, the Green's functions $G^{(j)}(E)$ are given by

$$\Lambda_jG^{(j)}(E)\Lambda_j\equiv\Lambda_j(E-\Lambda_jK-\Lambda R^{(j)}(E)\Lambda_j)^{-1}, \quad (31)$$

with the associated level shift operator given by

$$R^{(j)}(E)\equiv V+VP_j(E-P_jHP_j)^{-1}P_jV. \quad (32)$$

The general relationship between $R^{(j-1)}$ and $R^{(j)}$ is derived in the Appendix and is given by

$$R^{(j-1)}(E)=R^{(j)}(E)+R^{(j)}(E)\Lambda_jG^{(j)}(E)\Lambda_jR^{(j)}(E). \quad (33)$$

As pointed out earlier, (18) and (25) are special cases of the above general expression.

The transition amplitude follows from the insertion of the above expression for $P_iG(E)\Lambda_a$ into (5) and the subsequent evaluation of the integral over energy. If the final state is stable, then for t sufficiently large we may ignore the possibility of complex poles and pick up only the contribution from the final-state pole at $E=E_b$. For this case the transition amplitude is given by

$$I_{ba}(t)\approx e^{-iE_bt}\langle b|R^{(i)}(E_b)\times\prod_{j=1}^{i}[1+\Lambda_jG^{(j)}(E_b)\Lambda_jR^{(j)}(E_b)]\Lambda_aG(E_b)|a\rangle, \quad (34)$$

where i intermediate steps are assumed. Here, we have ignored terms which damp out exponentially with time. Apart from slight differences in notation, this is the result of Goldhaber and Watson arrived at by a different factorization method.[4] If, however, the final state is not stable, then the pole at $E=E_b$ is spurious and *does not* occur. Indeed, in this case it is necessary to keep all contributions from the poles in the continuum in order to find the correct transition probability.[2] In the first application of this formalism we shall see that the pole at $E=E_b$ is removable.

We note that in the general form for the projection operator given by Eq. (30), we have made use of the idea of sequential transitions. Nowhere, however, did we make explicit use of a decay of the initial state as popularly understood. Thus, we have neither postulated a specific form for the Hamiltonian nor have we specified the form of the spectrum of H. Hence, the results we have found are quite general and may be applied to a cascade-decay problem,[4] for instance, or may be applied to problems in which a number of sequences are suggested by physical observation. It is with this latter point of view in mind that we have chosen our illustrations. In each illustration we shall find that higher-order corrections in the perturbative sense appear easily. Indeed, the value of the present formulation seems to be the ease in which higher-order perturbations may be obtained.

3. TRANSITIONS BETWEEN UNSTABLE STATES

As a first illustration of the formalism just developed, we consider a particularly simple problem, namely, the transition between two unstable states. These states may be isolated, they may be members of two different sets or manifolds of states, or they may be states of the same manifold. The specific form of the projection operators actually used will depend upon our inter-

pretation of the location of these states. One of the purposes of this illustration is to show that care must be exercised in evaluating the expression for the transition amplitude occurring in (34). In the case to be considered, the pole on the real axis at $E=E_b$ is removable and hence does not contribute to the transition amplitude. In all cases mentioned above, the transition amplitude is given by

$$I_{ba}(t)=(2\pi i)^{-1}\int_c \langle b|G(E)|a\rangle e^{-iEt}dE$$
$$=(2\pi i)^{-1}\int_c \langle b|\Lambda_b(E-K)^{-1}\times\Lambda_b R(E)\Lambda_a G_{aa}(E)|a\rangle e^{-iEt}dt. \quad (35)$$

Now, the level shift operator $R(E)$ has complicated matrix elements in and between both manifolds spanned by Λ_a and Λ_b. We may simplify this situation by writing $R(E)$ in terms of $R^{(b)}$ through (19) with state $|1\rangle$ representing $|b\rangle$. Thus we have the expressions

$$R(E)=R^{(b)}+R^{(b)}\Lambda_b G^{(b)}(E)\Lambda_b R^{(b)}, \quad (36)$$

with

$$G_{bb}{}^{(b)}(E)=\Lambda_b(E-K-\Lambda_b R^{(b)}\Lambda_b)^{-1},$$
$$R^{(b)}(E)=V+VP_b(E-P_bHP_b)^{-1}P_bV,$$
$$P_b=(1-\Lambda_a)(1-\Lambda_b)=1-\Lambda_a-\Lambda_b. \quad (37)$$

If we combine these expressions to form the integrand in (35), we have the formidable expression

$$\Lambda_b(E-K)^{-1}\Lambda_b R(E)\Lambda_a G_{aa}(E)\Lambda_a$$
$$=\Lambda_b(E-K)^{-1}\Lambda_b[R^{(b)}(E)+R^{(b)}(E)\times\Lambda_b(E-K-\Lambda_b R^{(b)}\Lambda_b)^{-1}\Lambda_b R^{(b)}(E)]$$
$$\times\Lambda_a[E-K-\Lambda_a R^{(b)}\Lambda_a-\Lambda_a R^{(b)}\times\Lambda_b(E-K-\Lambda_b R^{(b)}\Lambda_b)^{-1}\Lambda_b R^{(b)}\Lambda_a]^{-1}, \quad (38)$$

which serves to point out the importance of noting the energy dependence of the diagonal and off-diagonal matrix elements of $R(E)$. In the case that states $|a\rangle$ and $|b\rangle$ are otherwise *isolated* or are in the same manifold of states the above expression reduces simply to

$$|b\rangle\{R_{ba}{}^{(b)}[(E-E_a-R_{aa}{}^{(b)})(E-E_b-R_{bb}{}^{(b)})-R_{ab}{}^{(b)}R_{ba}{}^{(b)}]^{-1}\}\langle a|, \quad (39)$$

where we note the disappearance of the pole at $E=E_b$. Hence, for this case, the transition amplitude reduces to

$$I_{ba}(t)=(2\pi i)^{-1}\int_c e^{-iEt}R_{ba}{}^{(b)}(E)\times[(E-E_a-R_{aa}{}^{(b)})(E-E_b-R_{bb}{}^{(b)})-R_{ab}{}^{(b)}R_{ba}{}^{(b)}]dE, \quad (40)$$

which apart from an error in sign is identical to the first of Eqs. (33) in Ref. 2. The integration over time may be performed as in (2) with the result that

$$I_{ba}(t)=-[R_{ba}{}^{(b)}/2(b+id)](e^{-ibt+dt}-e^{ibt-dt})e^{+iat-ct}, \quad (41)$$

where

$$a=\tfrac{1}{2}(E_a+E_b+\Re_e R_{aa}{}^{(b)}+\Re_e R_{bb}{}^{(b)}),$$
$$b=\tfrac{1}{4}\sqrt{2}[(A^2+B^2)^{1/2}+A]^{1/2},$$
$$c=-\tfrac{1}{2}\,\mathrm{Im}(R_{aa}{}^{(b)}+R_{bb}{}^{(b)}),$$
$$d=\tfrac{1}{4}\sqrt{2}[(A^2+B^2)^{1/2}-A]^{1/2}\,\mathrm{sgn}B, \quad (42)$$

and

$$A=(E_a-E_b+\Re_e R_{aa}{}^{(b)}-\Re_e R_{bb}{}^{(b)})^2-\tfrac{1}{2}(\mathrm{Im}R_{aa}{}^{(b)}-\mathrm{Im}R_{bb}{}^{(b)})^2+4|R_{ab}{}^{(b)}|^2,$$
$$B=2(E_a-E_b+\Re_e R_{aa}{}^{(b)}-\Re_e R_{bb}{}^{(b)})\times(\mathrm{Im}R_{aa}{}^{(b)}-\mathrm{Im}R_{bb}{}^{(b)}).$$

Here we have assumed that the energy variation of $R^{(b)}(E)$ was so slight that little difference would be realized by evaluating $R_{aa}{}^{(b)}(E)$ at $E=E_a$ and $R_{bb}{}^{(b)}(E)$ at $E=E_b$. It is important to note also that both states $|a\rangle$ and $|b\rangle$ are essentially discrete; neither has a continuum of states associated with it. Through the coupling via V *only* does the continuum arise.

As a specific example of the above simple problem we consider the shift of the transition frequency as observed (possibly) in an atomic beam experiment. For simplicity, we assume that the two states are members of the same hyperfine multiplet and hence have the same lifetime. The atoms in the beam are assumed to be prepared in a given state, $|a\rangle$ and then passed through a cavity where an rf field is allowed to induce transitions from the state $|a\rangle$ to the state $|b\rangle$, say. The particular state of the atom is detected after the atoms in the beam pass through the field region. The states $|a\rangle$ and $|b\rangle$ are particular states of the unperturbed Hamiltonian K, where K is

$$K=H_{\mathrm{rf}}+H_{\mathrm{at}}+H_{\mathrm{rad}}. \quad (43)$$

Here H_{rf} is the Hamiltonian of the rf electromagnetic field, H_{at} is the Hamiltonian of the unperturbed atom, and H_{rad} is the Hamiltonian of the radiation field (decay photon). In the number representation for both H_{rf} and H_{rad} the eigenvalues of K are

$$K|a;n;0\rangle=(n\omega+W_a)|a;n;0\rangle,$$
$$K|b;n+1;0\rangle=[(n+1)\omega+W_b]|b;n+1;0\rangle, \quad (44)$$

where n is the number of photons with frequency ω in the radiation field, W_a is the energy of the ath unperturbed state of the atom, and there are 0 decay photons assumed present in state a.

The energy of interaction between the atom and the radiation fields may be simplified when the dipole approximation is assumed.[6] For simplicity we assume that V has the form

$$V=H_1+H_2, \quad (45)$$

[6] E. A. Power and S. Zienau, Phil. Trans. Roy. Soc. (London) A251, 427 (1959).

where H_1 is the energy of interaction between the atom and the rf field and H_2 is the energy of interaction between the atom and the radiation (decay photon) field. To lowest order in the interaction energies we write the level shift operator given in (37) as

$$R^{(b)}(E)=R_1{}^{(b)}(E)+R_2{}^{(b)}(E), \tag{46}$$

where the first term depends solely on the rf field. The second term has diagonal matrix elements only and describes the shift and width due to the vacuum radiation field:

$$R_{2aa}{}^{(b)}=R_{2bb}{}^{(b)}=D-i\tfrac{1}{2}\Gamma. \tag{46'}$$

The diagonal matrix elements of $R_2{}^{(b)}$ are taken to be equal as the two states are in the same manifold. For simplicity the matrix elements are also assumed to be insensitive to a variation in energy and, hence, are evaluated at

$$E\approx W_b\approx W_a.$$

To lowest order in H_1 the diagonal matrix elements of $R_1{}^{(b)}(E)$ are real; as a result of this observation and the form of the matrix elements of R_2, we may simplify the constants in Eq. (42). In particular, both B and d vanish, and the transition amplitude reduces to

$$I_{ba}(t)=-i(R_{ba}{}^{(b)}/b)\sin bte^{+iat-ct}. \tag{47}$$

Hence, the transition probability is the familiar expression

$$P_{ba}(t)=|R_{1ba}{}^{(b)}/b|^2\sin^2 bte^{-2ct}. \tag{48}$$

Here, in terms of the matrix elements of $R^{(b)}$,

$$2c=\Gamma$$

$$b^2=\tfrac{1}{4}[(W_a-W_b+\Re_e R_{1aa}{}^{(b)}-\Re_e R_{1bb}{}^{(b)})^2+4|R_{1ab}{}^{(b)}|^2]. \tag{49}$$

We turn now to the evaluation of the matrix elements of $R^{(b)}$ involving H_1, the interaction energy between the rf radiation field and the unperturbed atom. For ease of comparison, we introduce the notation used by Mizushima.[7] The matrix elements of H_1 are nondiagonal in the rf photon number and diagonal in the decay photon number and are given by ($\hbar=1$)

$$\langle n;a|H_1|b;n+1\rangle=-i(\hat{e}_\lambda X\nabla G)\cdot\langle a|\mathbf{M}_m|b\rangle\times[(n+1)/2\omega\epsilon_0 V]^{1/2},$$

$$\langle n;a|H_1|c;n+1\rangle=i\omega G\langle a|\mathbf{e}_\lambda\cdot\mathbf{M}_e|c\rangle\times[(n+1)/2\omega\epsilon_0 V]^{1/2}. \tag{50}$$

Here, a and b may be the same electronic state, but c must differ from a. The $\mathbf{M}$'s are the total magnetic and electric dipole moment of the atom while $\hat{e}_\lambda$ is the direction of polarization of the rf field. $G(r)$ is essentially the amplitude of the single mode field at the location of the atom. It is normalized such that the integral over the volume V satisfies

$$(1/V)\int_V|G(r)|^2dV=1. \tag{51}$$

We use mks units throughout.

An inspection of form of $R_{1ab}{}^{(b)}$ shows that to the lowest order in the rf field, the matrix element is given simply by

$$R_{1ab}{}^{(b)}\sim\langle n;a|H_1|b;n+1\rangle\equiv H_{1ab}. \tag{52}$$

Similarly, an inspection of the form of the diagonal matrix elements of $R_1{}^{(b)}$ shows that the contributions of H_1 to $R_1{}^{(b)}$ to lowest order are given by

$$R_{1aa}{}^{(b)}=\sum_{c\neq a,b}\left\{\frac{|\langle a;n|H_1|c;n+1\rangle|^2}{W_a-W_c-\omega}+\frac{|\langle a;n|H_1|c;n-1\rangle|^2}{W_a-W_c+\omega}\right\},$$

$$R_{1bb}{}^{(b)}=\sum_{c\neq a,b}\left\{\frac{|\langle b;n+1|H_1|c;n\rangle|^2}{W_b-W_c+\omega}+\frac{|\langle b;n+1|H_1|c;n+2\rangle|^2}{W_b-W_c-\omega}\right\}. \tag{53}$$

Here the above energy corrections differ from the usual expressions for the second-order correction to the energy found by the usual perturbation theory in that the projection operator P_b has restricted the sum to states *other* than $|a\rangle$ or $|b\rangle$.[7,8] Hence, the energy denominators do not vanish except in the case of accidental degeneracy.

The time-dependent transition probability, which can now be written as

$$P_{ba}(t)=\{|H_{1ab}|^2/[\ ]\}\sin^2[\ ]te^{-\Gamma t}, \tag{54}$$

where

$$[\ ]=\tfrac{1}{4}[(W_a-W_b-\omega+\Omega)^2+4|H_{1ab}|^2],$$

no longer has a maximum at the Bohr frequency for a given time t, but at the frequency

$$\omega=W_a-W_b+\Omega, \tag{55}$$

where the selection rules for dipole transitions allow us to decompose the shift Ω into an electric and a magnetic contribution:

$$\Omega=\Omega_e+\Omega_m. \tag{56}$$

Here

$$\Omega_e\equiv(n\omega G^2/2V\epsilon_0)\sum_{c\neq a,b}|\langle a|\mathbf{e}_\lambda\cdot\mathbf{M}_e|c\rangle|^2\times[(W_a-W_c-\omega)^{-1}+(W_a-W_c+\omega)^{-1}]-(n\omega G^2/2V\epsilon_0)\sum_{c\neq a,b}|\langle b|\mathbf{e}_\lambda\cdot\mathbf{M}_e|c\rangle|^2\times[(W_b-W_c+\omega)^{-1}+(W_b-W_c-\omega)^{-1}]. \tag{57}$$

[7] See, for example, K. Gottfried, *Quantum Mechanics* (W. A. Benjamin, Inc., N. Y., 1966), Sec. 45.2.

[8] A. Dalgarno in *Quantum Theory*, edited by D. R. Bates (Academic Press Inc., N. Y., 1961), Vol. I, pp. 171–209.

If we now identify $(n\omega G^2/2V\epsilon_0)$ with the energy density of the electric field at the atom, $\rho_e \equiv \frac{1}{2}\epsilon_0 \mathbf{E}^2$, then we may write the electric-field shift as

$$\Omega_e = (\tfrac{1}{2}\epsilon_0 \mathbf{E}^2) \sum_{c\neq a,b} |\langle a|\mathbf{e}_\lambda \cdot \mathbf{M}_e|c\rangle|^2 \times [(W_a - W_c - \omega)^{-1} + (W_a - W_c + \omega)^{-1}] - (\tfrac{1}{2}\epsilon_0 \mathbf{E}^2) \sum_{c\neq a,b} |\langle b|\mathbf{e}_\lambda \cdot \mathbf{M}_e|c\rangle|^2 \times [(W_b - W_c + \omega)^{-1} + (W_b - W_c - \omega)^{-1}]. \quad (58)$$

Similarly, the shift in frequency arising from the magnetic dipole coupling may be written as

$$\Omega_m = (\tfrac{1}{2}\mu_0 \mathbf{H}^2) \sum_{c\neq a,b} |\langle a|\mathbf{e}_\lambda \cdot \mathbf{M}_m|c\rangle|^2 \times [(W_a - W_c - \omega)^{-1} + (W_a - W_c + \omega)^{-1}] - (\tfrac{1}{2}\mu_0 \mathbf{H}^2) \sum_{c\neq a,b} |\langle b|\mathbf{e}_\lambda \cdot \mathbf{M}_m|c\rangle|^2 \times [(W_b - W_c + \omega)^{-1} + (W_b - W_c - \omega)^{-1}]. \quad (59)$$

We note that if either the rf electric or the magnetic field may be measured accurately, the induced shifts provide a means for measuring the electric and magnetic dipole moments. Thus, if the hyperfine manifold containing $|a\rangle$ and $|b\rangle$ is reasonably separated from nearby electronic levels, the number of terms contributing to Ω_m is finite and small. By measuring the shift and the rf magnetic field intensity, the magnetic dipole moment may be calculated directly.

These expressions, apart from notational differences, are identical to those found by Mizushima.[9] We differ, however, in our approach as well as in our interpretation. If we had not used either the decay theory outlined here, or that in Ref. 2, in order to treat this problem it would have been necessary to introduce a unitary transformation to decouple the states of interest from the remaining spectrum of states.[10–12] The use of the projection-operator technique effectively removed the virtual transitions with an accompanying redefinition of the energy levels involved.

It should also be pointed out that the results of Mizushima follow directly from a general perturbation theory developed by Salwen.[13] While developed specifically for transitions between stable states induced by rotating fields, his approach itself may be extended so as to apply to arbitrary electromagnetic fields. It ignores, however, the finite lifetime of the states and assumes that the states of interest are separable. The projection-operator approach presented here recognizes the instability of the states due to the presence of the vacuum radiation field, and hence, is inherently more powerful than the perturbation theory approach.

4. RESONANCE FLUORESCENCE

As an illustration of the step-by-step process of sequential transitions, we consider the problem of resonance fluorescence of an atomic system.[11] We assume that initially the atom is in a sublevel of the ground state and is exposed to a beam of photons with a spectral energy distribution that is peaked about the energy separation between the ground state and the first excited state. The width of the spectral energy distribution of the beam is taken to be large compared to the width of the excited states. Once the atomic system is excited, it may decay by either spontaneous or stimulated emission. To add to the interest of the process, we assume that a second field or bath of the photons is present. (To distinguish this from the first, we shall refer to the beam as optical photons and the second bath as rf photons.) The rf photons induce a transition from one excited state to another. As a result of such a transition, the angular properties of the emitted optical radiation will differ from the initial beam of optical photons, thus facilitating detection.

The energy of the noninteracting system of atom plus fields is described by the Hamiltonian

$$K = H_{0\,\mathrm{at}} + H_{0\,\mathrm{rad}}, \quad (60)$$

where the first term on the right is the Hamiltonian of the atomic system unperturbed by the fields. To distinguish between the various sublevels of the ground state it may include, for example, a dc magnetic field. The remaining term is the Hamiltonian of the radiation fields, including the optical, the rf, as well as the vacuum radiation field.

We describe the states of the unperturbed Hamiltonian that are of interest to us as follows. The initial state of the system consists of the atom in the ath sublevel of the ground state in the presence of n rf photons of a given single mode of frequency ω. There are in addition n_1 optical photons with wave number k_1, n_2 with wave number k_2, etc. Finally, the initial state is assumed to be in the ground state of the vacuum or decay radiation field. The initial state is specified by the ket

$$|a_n\rangle \equiv |a; n_1, n_2, n_3, \cdots; 0; n\rangle. \quad (61)$$

The excited states of the system differ from the ground state with respect to the atomic state, the number of rf photons present, and also with respect to the number of optical photons present. For simplicity, we shall consider among the states of interest the two states

$$|b_n; -k_i\rangle \equiv |b; n_1, n_2, \cdots n_{i-1}, n_i - 1, n_{i+1}, \cdots; 0; n\rangle,$$
$$|c_{n'}; -k_i\rangle \equiv |c; n_1, n_2, \cdots n_{i-1}, n_i - 1, n_{i+1}, \cdots; 0; n'\rangle. \quad (62)$$

Here, the number of rf photons in the first state is the

[9] M. Mizushima, Phys. Rev. **133**, A414 (1964).
[10] J.-M. Winter, Ann. Phys. (Paris) **4**, 745 (1959).
[11] H. H. Nielsen, Rev. Mod. Phys. **23**, 90 (1951).
[12] W. Heitler, *Quantum Theory of Radiation* (Oxford University Press, London, 1954), Secs. 15 and 16.
[13] H. Salwen, Phys. Rev. **99**, 1274 (1955).

same as in the initial state. Both states differ from the initial state in that a single optical photon of wave number $\mathbf{k}_i$ has been absorbed from the beam.

The final state of our system to be considered explicitly differs from the initial state in being in a different sublevel of the ground state. In addition, the number of rf photons differs from the initial state. Finally, we assume that an optical photon has been absorbed in the excitation process, while an optical (i.e., decay) photon with wave number $\mathbf{k}_\sigma$ and polarization σ has been emitted in the decay process. The state is specified by the ket

$$|a_{n'}'; -\mathbf{k}_i; \mathbf{k}_\sigma\rangle \equiv |a'; n_1, n_2, \cdots n_{i-1}, n_i-1, \times n_{i+1}, \cdots; \mathbf{k}_\sigma; n'\rangle. \quad (63)$$

As eigenstates of K, the energies of these unperturbed states are given by ($\hbar=c=1$)

$$\begin{aligned} K|a_n\rangle &= E_{a,n}|a_n\rangle \equiv [W_a+\textstyle\sum_j n_jk_j+n\omega]|a_n\rangle, \\ K|b_n; -\mathbf{k}_i\rangle &= E_{b,n;-\mathbf{k}_i}|b_n; -\mathbf{k}_i\rangle \\ &\equiv [W_b+\textstyle\sum_j{}' n_jk_j+n\omega]|b_n; -\mathbf{k}_i\rangle, \\ K|c_n; -\mathbf{k}_i\rangle &= E_{c,n';-\mathbf{k}_i}|c_{n'}; -\mathbf{k}_i\rangle \\ &\equiv [W_c+\textstyle\sum_j{}' n_jk_j+n'\omega]|c_{n'}; -\mathbf{k}_i\rangle, \\ K|a'_{n'}; -\mathbf{k}_i; \mathbf{k}_\sigma\rangle &= E_{a',n';-\mathbf{k}_i;\mathbf{k}_\sigma}|a'_{n'}; -\mathbf{k}_i; \mathbf{k}_\sigma\rangle \\ &\equiv [W_{a'}+\textstyle\sum_j{}' n_jk_j+k_\sigma+n'\omega]|a'_{n'}; -\mathbf{k}_j; \mathbf{k}_\sigma\rangle. \end{aligned} \quad (64)$$

Here, the prime on $\sum$ designates the omission of one photon with wave number $\mathbf{k}_i$. The number of photons is assumed to be so large that we may replace the sum $\sum$ over the photon number by the integral

$$\int_{-\infty}^{\infty} U(k)dk,$$

where $U(k)$ is the spectral energy distribution with width $\Delta(k)$. The description of the optical photons as a beam implies that the vector wave numbers $\mathbf{k}_j$ are peaked in a particular direction, say $\mathbf{k}_i$. We shall ignore the spread about this direction.

The presence of the interaction between the atom and the optical and rf photons introduces virtual as well as real transitions: the ground state of the atomic system is no longer stable as the optical photons may induce the atom to jump to one or more of its excited states. Of these transitions, the most likely to occur are the real transitions satisfying energy conservation conditions. The virtual transitions which may occur between the ground state and an excited state are a result of an indirect path involving several optical transitions. Similarly, the presence of the rf radiation field induces transitions among the various intermediate states and may also involve real as well as virtual transitions. The virtual processes again differ from the energy conserving, real processes by introducing additional, indirect processes involving several rf transitions. In both cases of optical and rf radiation-induced transitions, the virtual transitions may be removed by a canonical transformation.[10–12] The net result of this transformation, in addition to restricting all transitions to energy-conserving direct transitions, is to decouple the states of interest effectively from the remaining states of the energy spectrum of the unperturbed system. This decoupling process results in the appearanee of a shift in the energy of the states of interest. In thee prsence of induced optical and rf transitions, these shifts would be proportional to the intensity of the optical and rf magnetic fields, respectively. The transition theory presented in Sec. 2, by embodying the concept of a sequence, effectively provides the above decoupling. This decoupling is achieved through the introduction of the projection operators P_i which limit the area of Hilbert space available for transitions. Thus, while the states of interest are only four in number, in principle through the projection operators P_i we are able to take into account the contributions from the interactions of these states with the remaining states of the system.

The interaction energy may be represented simply as

$$V=H_1+H_2, \quad (65)$$

where H_1 refers to the energy of interaction between the atomic system and the optical and radiation field, while H_2 refers to the corresponding interaction energy involving the rf field. We restrict attention to single photon transitions and treat both H_1 and H_2 in the dipole approximation only.[6]

The transition amplitude between the initial and the final state is given by

$$\begin{aligned} I_{a'n';\,-\mathbf{k}_i;\,\mathbf{k}\sigma,a_n}(t) &\equiv I_{a'n'\sigma,a_n}(t) \\ &= (2\pi i)^{-1}\int_c \langle a'_{n'}; -\mathbf{k}_i; \mathbf{k}_\sigma|G(E)|a_n\rangle e^{-iEt}dE. \end{aligned} \quad (66)$$

While we may treat the transitions through the two excited states as two steps in a sequence in the sense of Sec. 2, it is somewhat simpler to consider a sequence passing through a single manifold of states. Thus, the matrix element in the above transition amplitude is given by

$$\begin{aligned} &\langle a'_{n'}; -\mathbf{k}_i; +\mathbf{k}_\sigma|G(E)|a_n\rangle \\ &= (E-E_{a',n';-\mathbf{k}_i;\mathbf{k}_\sigma})^{-1}\langle a'_{n'}; -\mathbf{k}_i; \mathbf{k}_\sigma| \\ &\qquad \times R(E)\Lambda_{a_n}G(E)\Lambda_{a_n}|a_n\rangle \\ &= (E-E_{a',n';-\mathbf{k}_i;\mathbf{k}_\sigma})^{-1}\langle a'_{n'}; -\mathbf{k}_i; \mathbf{k}_\sigma|[R^{(1)}(E) \\ &\quad +R^{(1)}(E)\Lambda_1G^{(1)}(E)\Lambda_1R^{(1)}(E)]\Lambda_{a_n}G\Lambda_{a_n}|a_n\rangle, \end{aligned} \quad (67)$$

where we have used (18). Here $G^{(1)}(E)$ is given by (20) and the level shift operator $R^{(1)}$ is given by (19) with the specific projection operators:

$$\begin{aligned} P_1 &= 1-\Lambda_1-\Lambda_{a_n}, \\ \Lambda_{a_n} &\equiv |a_n\rangle\langle a_n|, \\ \Lambda_1 &\equiv \sum_i |b_n; -\mathbf{k}_i\rangle\langle -\mathbf{k}_i; b_i| \\ &\qquad +\sum_i |c_{n'}; -\mathbf{k}_i\rangle\langle -\mathbf{k}_i; c_{n'}|. \end{aligned} \quad (68)$$

From the form of the level shift operator $R^{(1)}$ as given by (19) with the above projection operator, we see that the first term in (67) involving $R^{(1)}(E)$ describes direct transitions only and not transitions passing through the intermediate states. As such, it vanishes unless $n'=n$, and is otherwise insensitive to the properties of the rf field; hence, we may safely ignore it. The remaining component,

$$\langle a'_{n'};-\mathbf{k}_i;\mathbf{k}_\sigma|G(E)|a_n\rangle = (E-E_{a',n';-\mathbf{k}_i;\mathbf{k}_\sigma})^{-1}\langle a'_{n'};-\mathbf{k}_i;\mathbf{k}_\sigma|R^{(1)}(E) \times\Lambda_1 G^{(1)}(E)\Lambda_1 R^{(1)}(E)\Lambda_{a_n}G(E)|a_n\rangle, \quad (69)$$

has the following physical interpretation: The atomic system while in the initial state interacts with the radiation fields, and is excited to the states spanned by Λ_1 by absorbing a photon through $R^{(1)}$. While in this manifold it interacts with the radiation fields, primarily the rf field, and then drops down to the ground state at the same time emitting a photon through $R^{(1)}$. If we limit ourselves to one-photon processes, the above matrix element reduces to

$$\langle a'_{n'};-\mathbf{k}_i;\mathbf{k}_\sigma|G(E)|a_n\rangle = (E-E_{a'n';-\mathbf{k}_i;\mathbf{k}_\sigma})^{-1}\langle a'_{n'};-\mathbf{k}_i;\mathbf{k}_\sigma|H_1\Lambda_1 G^{(1)}(E) \times\Lambda_1 H_1\Lambda_{a_n}G(E)|a_n\rangle. \quad (70)$$

By restricting ourselves to one-photon processes, we see that $n'=n\pm1$ *if* the resulting amplitude is to be sensitive to the intensity of the rf field. In writing (70) we have apparently considered real processes only. Thus $n'=n-1$ would then apply to the case $W_c>W_b$, etc. The presence of the projection operator P_1 in *both* $G^{(1)}(E)$ and $G(E)$ ensures us, however, of retaining the effects of the virtual processes.

Calculation of $G^{(1)}(E)$

The contributions to the matrix elements of $\Lambda_1 G^{(1)}(E)\Lambda_1$ involving K we may write down immediately from (64); the contributions from $R^{(1)}$ require more care. If we recall the statement made earlier concerning the effect of the vacuum radiation field, we see that we may write the contribution $\Lambda_1 R^{(1)}\Lambda_1$ as

$$\Lambda_1 R^{(1)}(E)\Lambda_1=\Lambda_1 R_r{}^{(1)}(E)\Lambda_1+\Lambda_1 R_1{}^{(1)}(E)\Lambda_1, \quad (71)$$

where the first term has diagonal elements only which represent a level shift and width:

$$\langle b_n;-\mathbf{k}_i;|R_r{}^{(1)}(E)|b_n;-\mathbf{k}_i'\rangle\equiv(D_b-i\Gamma_b/2)\delta_{\mathbf{k}_i,\mathbf{k}_i'},$$
$$\langle c_{n'};-\mathbf{k}_i|R_r{}^{(1)}(E)|c_{n'};-\mathbf{k}_i'\rangle\equiv(D_c-i\Gamma_c/2)\delta_{\mathbf{k}_i,\mathbf{k}_i'}. \quad (72)$$

We have assumed that the matrix elements are relatively insensitive to the energy of the system and have been evaluated at the energy E_b, E_c of the excited states. (In a more careful calculation, this energy variation must be included. It would appear as a renormalization factor.[2]) The contribution to $\Lambda_1 R_1{}^{(1)}\Lambda$, to lowest order in H_2 and nondiagonal in n, is simply

$$\langle b_n;-\mathbf{k}_i|R_1{}^{(1)}(E)|c_{n'};-\mathbf{k}_i'\rangle = \langle b_n|H_2|c_{n'}\rangle\delta_{n',n\pm1}\delta_{\mathbf{k}_i,\mathbf{k}_i'}, \quad (73)$$

while the contributions to lowest order in H_1 and H_2 and diagonal in n are given by

$$\langle b_n;-\mathbf{k}_i|R_1{}^{(1)}(E)|b_n;-\mathbf{k}_i'\rangle = \sum_{\substack{d\neq a,b,c\\ n'=n\pm1}}\frac{|\langle b_n|H_2|d_{n'}\rangle|^2}{E-E_{dn'}}\delta_{\mathbf{k}_i,\mathbf{k}_i'} + \sum_{\gamma\neq a,b,c}\frac{|\langle b_n;-\mathbf{k}_i|H_1|\gamma_n\rangle|^2}{E-E_{\gamma n}}\delta_{\mathbf{k}_i,\mathbf{k}_i'}, \quad (74)$$

with a similar contribution from $|c_{n'};-\mathbf{k}_i\rangle$. Here, the off-diagonal contribution (73) describes direct transitions induced by the rf field. The contributions to $R^{(1)}$ diagonal in n, (74), arise from virtual transitions induced by the rf and optical fields, respectively.

On closer inspection, we see that we should write the matrix element in (70) as

$$\langle a'_{n'};-\mathbf{k}_i;\mathbf{k}_\sigma|G(E)|a_n\rangle = (E-E_{a',n';-\mathbf{k}_i;\mathbf{k}_\sigma})^{-1}\langle a'_{n'};-\mathbf{k}_i;\mathbf{k}_\sigma|H_1\Lambda_{1,n'} \times G^{(1)}(E)\Lambda_{1,n}H_1\Lambda_{a_n}G(E)|a_n\rangle, \quad (70')$$

where we have noted by the additional subscripts on Λ_1 that H_1 does not involve the creation or destruction of an rf photon. The form of $[\Lambda_{1,n'}G^{(1)}(E)\Lambda_{1,n})^{-1}$ is given below:

	c_{n-1}	b_n	c_{n+1}
c_{n-1}	$E-\tilde{E}_{c,n-1;-\mathbf{k}_i}-\Delta_{c,n-1}+i\frac{1}{2}\Gamma_c$	$-\langle c_{n-1}\|H_2\|b_n\rangle$	0
b_n	$-\langle b_n\|H_2\|c_{n-1}\rangle$	$E-\tilde{E}_{b,n;-\mathbf{k}_i}-\Delta_{b,n}+i\frac{1}{2}\Gamma_b$	$-\langle b_n\|H_2\|c_{n+1}\rangle$
c_{n+1}	0	$-\langle c_{n+1}\|H_2\|b_n\rangle$	$E-\tilde{E}_{c,n+1;-\mathbf{k}_i}-\Delta_{c,n+1}+i\frac{1}{2}\Gamma_c$

Here the tilde indicates the natural level shift has been included in the energy of the atomic level:

$$\tilde{W}_c\equiv W_c+D_c. \quad (75)$$

The energy-dependent shifts $\Delta_{b,n}$ and $\Delta_{c,n'}$ are given by (74). From the form of the sequence assumed, the matrix elements of $\Lambda^1 G^{(1)}\Lambda_1$ of interest are given by

$$\langle c_{n'};-\mathbf{k}_i|G^{(1)}(E)|b_n;-\mathbf{k}_i'\rangle = \frac{\text{cofactor}(\Lambda_1 G^{(1)}\Lambda_1)^{-1}{}_{bn,cn'}\delta_{\mathbf{k}_i,\mathbf{k}_i'}}{\text{Det}[(\Lambda_1 G^{(1)}\Lambda_1)^{-1}]}, \quad (76)$$

with

$$\langle c_{n\pm1};-\mathbf{k}_i|G^{(1)}(E)|b_n;-\mathbf{k}_i'\rangle = \frac{\langle c_{n\pm1}|H_2|b_n\rangle(E-\bar{E}_{c,n\mp1}-\Delta_{c,n\mp1}+i\tfrac{1}{2}\Gamma_c)\delta_{\mathbf{k}_i,\mathbf{k}_i'}}{\det[(\Lambda_1 G^{(1)}\Lambda_1)^{-1}]} \quad (77)$$

with the determinant given by

$$\begin{aligned}\det[(\Lambda_1 G^{(1)}\Lambda_1)^{-1}] &= (E-\bar{E}_{c,n-1;-\mathbf{k}_i}-\Delta_{c,n-1}+i\tfrac{1}{2}\Gamma_c)(E-\bar{E}_{b,n;-\mathbf{k}_i}\\ &\quad -\Lambda_{b,n}+i\tfrac{1}{2}\Gamma_b)(E-\bar{E}_{c,n+1;-\mathbf{k}_i}-\Delta_{c,n+1}+i\tfrac{1}{2}\Gamma_c)\\ &\quad -|\langle b_n|H_2|c_{n-1}\rangle|^2(E-\bar{E}_{c,n+1;-\mathbf{k}_i}-\Delta_{c,n+1}+i\tfrac{1}{2}\Gamma_c)\\ &\quad -|\langle b_n|H_2|c_{n+1}\rangle|^2(E-\bar{E}_{c,n-1;-\mathbf{k}_i}-\Delta_{c,n-1}+i\tfrac{1}{2}\Gamma_c). \end{aligned} \quad (78)$$

In writing the matrix elements of $\Lambda_{1,n'}G^1\Lambda_{1,n}$ in the form given by (76) we have allowed for a coupling through the state $|b_n;-\mathbf{k}_i\rangle$ of the processes of absorption and stimulated emission of rf photons. From an analysis based upon perturbation theory,[2] the inclusion of this coupling will result in an additional resonance yielding a transition probability of order $|H_2|^2/\omega_{bc}^2$ times the primary contribution. Here $|H_2|$ represents the matrix element of the rf interaction energy between states $|b_n;-\mathbf{k}_i\rangle$ and $|c_{n'};-\mathbf{k}_i\rangle$, while ω_{bc} is the energy difference. By limiting ourselves to the two simple decay channels involving $n'=n\pm1$ rf photons emanating from the initial state with n rf photons, we have already ignored terms of this magnitude. Hence, to be consistent, we consider the inverse of $(\Lambda_{1,n'}G^1(E)\Lambda_{1,n})$ to have the simpler form

	$c_{n'}$	b_n
$c_{n'}$	$E-\bar{E}_{c,n';-\mathbf{k}_i}-\Delta_{c,n'}+i\tfrac{1}{2}\Gamma_c$	$-\langle c_{n'}\|H_2\|b_n\rangle$
b_n	$-\langle b_n\|H_2\|c_{n'}\rangle$	$E-\bar{E}_{b,n;-\mathbf{k}_i}-\Delta_{b,n}+i\tfrac{1}{2}\Gamma_b$

The matrix element of $\Lambda_{1,n'}G^{(1)}(E)\Lambda_{1,n}$ for the two decay channels considered then has the simple form

$$\langle c_{n'};-\mathbf{k}_i|G^{(1)}(E)|b_n;-\mathbf{k}_i'\rangle = \langle c_{n'}|H_2|b_n\rangle\delta_{n',n\pm1}\delta_{\mathbf{k}_i,\mathbf{k}_i'}/\det, \quad (76')$$

with

$$\det\equiv(E-\bar{E}_{c,n';-\mathbf{k}_i}-\Delta_{c,n'}+i\tfrac{1}{2}\Gamma_c)(E-\bar{E}_{b,n;-\mathbf{k}_i} -\Delta_{b,n}+i\tfrac{1}{2}\Gamma_b)-|\langle c_{n'}|H_2|b_n\rangle|^2. \quad (78')$$

The decoupling that we have introduced may be made *exact* by choosing the sense of the rf field such that only one of the two matrix elements $\langle c_{n'}|H_2|b_n\rangle$ with $n'=n\pm1$ differs from zero at a time. When this is not possible, the double resonance should be treated using a coherent representation for the rf field.

The Green's Function $G_{an,an}$

The matrix element of interest is diagonal in the rf photon number and is given simply as

$$G_{an,an}(E)=[E-E_{a,n}-\langle a_n|R(E)|a_n\rangle]^{-1}. \quad (79)$$

But the level shift operator contains a contribution from the excited state through the sequence

$$\langle a_n|R(E)|a_n\rangle=\langle a_n|R^{(1)}|a_n\rangle + \langle a_n|R^{(1)}\Lambda_{1,n}G^{(1)}(E)\Lambda_{1,n}R^{(1)}|a_n\rangle, \quad (80)$$

where the first matrix element vanishes, as $|a_n\rangle$ is assumed to be stable in the absence of the optical field. [It should be noted that the optical photons also contribute to $\langle a_n|R^{(1)}|a_n\rangle$. However, for $U(k)$ peaked about a wave number given by the energy difference W_b-W_a, it should be negligible compared to (81).] The second term may be calculated by noting first that

$$\langle a_n|R^{(1)}\Lambda_{1,n}G^{(1)}(E)\Lambda_{1,n}R^{(1)}|a_n\rangle = \langle a_n|H_1\Lambda_{1,n}G^{(1)}(E)\Lambda_{1,n}H_1|a_n\rangle \quad (81)$$

or

$$\langle a_n|R^{(1)}\Lambda_{1,n}G^{(1)}(E)\Lambda_{1,n}R^{(1)}|a_n\rangle = \sum_{i,i'}\langle a_n|H_1|b_n;-\mathbf{k}_i\rangle\langle-\mathbf{k}_i;b_n|G^{(1)}(E) \times|b_n;-\mathbf{k}_i'\rangle\langle-\mathbf{k}_i';b_n|H_1|a_n\rangle. \quad (82)$$

But

$$\langle-\mathbf{k}_i;b_n|G^{(1)}(E)|b_n;-\mathbf{k}_i'\rangle = \frac{(E-\bar{E}_{c,n';-\mathbf{k}_i}-\Delta_{c,n'}+i\tfrac{1}{2}\Gamma_c)\delta_{\mathbf{k}_i,\mathbf{k}_i'}}{\det}, \quad (83)$$

where det is given by (78'). Hence, the matrix element in question is given by

$$\langle a_n|R(E)|a_n\rangle=\sum_i\frac{|\langle a_n|H_1|b_n;-\mathbf{k}_i\rangle|^2(E-\bar{E}_{c,n';-\mathbf{k}_i}-\Delta_{c,n'}+i\tfrac{1}{2}\Gamma_c)}{\det} \cong\int_{-\infty}^{\infty}\frac{U(k_i)dk_i|\langle a_n|H_1|b_n;-\mathbf{k}_i\rangle|^2(E-\bar{E}_{c,n';-\mathbf{k}_i}-\Delta_{c,n'}+i\tfrac{1}{2}\Gamma_c)}{\det}. \quad (84)$$

The integration here must be performed with care as E may involve k_i implicitly. In the limit that we can ignore $|\langle c_{n'}|H_2|b_n\rangle|^2$ in (78'), we see that the above matrix element reduces to

$$\langle a_n|R(E)|a_n\rangle\cong\int_{-\infty}^{\infty}\frac{U(k_i)dk_i|\langle a|H_1|b;-\mathbf{k}_i\rangle|^2}{E-\bar{E}_{b,n;-\mathbf{k}_i}-\Delta_{b,n}+i\tfrac{1}{2}\Gamma_b} \quad (85)$$

or

$$\langle a_n|R(E)|a_n\rangle\equiv\Delta_a(E)-i\tfrac{1}{2}\Gamma_a(E), \quad (86)$$

where for $|\langle c_{n'}|H_2|b_n\rangle|^2$ negligible as compared to $\frac{1}{4}\Gamma_c\Gamma_b$

$$\Delta_a \cong \int_{-\infty}^{\infty} \frac{U(k_i)dk_i|\langle a|H_1|b;-\mathbf{k}_i\rangle|^2(E-\bar{E}_{b,n;-\mathbf{k}_i}-\Delta_{b,n})}{(E-\bar{E}_{b,n;-\mathbf{k}_i}-\Delta_{b,n})^2+\frac{1}{4}\Gamma_b{}^2},$$

$$\Gamma_a \cong \int_{-\infty}^{\infty} \frac{U(k_i)dk_i|\langle a|H_1|b;-\mathbf{k}_i\rangle|^2\Gamma_b}{(E-\bar{E}_{b,n;-\mathbf{k}_i}-\Delta_{b,n})^2+\frac{1}{4}\Gamma_b{}^2}. \tag{87}$$

The expression (86) for $\langle a_n|R(E)|a_n\rangle$ serves also to define Δ_a and Γ_a for the case $|\langle c_{n'}|H_2|b_n\rangle|^2$ non-negligible. In the form given by (87) above we recognize the intensity-dependent shift and width discussed by Heitler[14] as well as by Barrat and Cohen-Tannoudji.[15] Using (86) we see that the matrix element of the Green's function in (79) is given by

$$G_{an,an}=[E-E_{a,n}-\Delta_a(E)+i\tfrac{1}{2}\Gamma_a(E)]^{-1}. \tag{88}$$

Transition Amplitude

Before calculating the transition amplitude,

$$I_{a'n'\sigma,a_n}(t)=(2\pi i)^{-1}\int_c dEe^{-iEt}\langle a'_{n'};-\mathbf{k}_i;\mathbf{k}_\sigma|G(E)|a_n\rangle, \tag{66}$$

we note first that the matrix element

$$\langle a'_{n'};-\mathbf{k}_i;\mathbf{k}_\sigma|G(E)|a_n\rangle=\frac{\langle a'_{n'};-\mathbf{k}_i;\mathbf{k}_\sigma|H_1\Lambda_{1,n'}G^{(1)}(E)\Lambda_{1,n}H_1|a_n\rangle G_{an,an}}{E-E_{a',n';-\mathbf{k}_i;\mathbf{k}_\sigma}} \tag{70}$$

must be rewritten using the explicit form for the projection operators $\Lambda_{1,n'}$ given by (68). Thus, we find

$$\begin{aligned}\langle a'_{n'};-\mathbf{k}_i;\mathbf{k}_\sigma|G(E)|a_n\rangle=&\sum_{i',i''}\langle a'_{n'};-\mathbf{k}_i;\mathbf{k}_\sigma|H_1|c_{n'};-\mathbf{k}_i'\rangle\langle c_{n'};-\mathbf{k}_i'|G^{(1)}(E)|b_n;-\mathbf{k}_i''\rangle\\&\times\langle b_n;-\mathbf{k}_i''|H_1|a_n\rangle G_{an,an}(E)(E-E_{a',n';-\mathbf{k}_i;\mathbf{k}_\sigma})^{-1}\\=&\sum_{i''}\langle a'_{n'};-\mathbf{k}_i;\mathbf{k}_\sigma|H_1|c_{n'};-\mathbf{k}_i\rangle\langle c_{n'};-\mathbf{k}_i|G^{(1)}(E)|b_n;-\mathbf{k}_i''\rangle\\&\times\langle b_n;-\mathbf{k}_i''|H_1|a_n\rangle G_{an,an}(E)(E-E_{a',n;-\mathbf{k}_i;\mathbf{k}_\sigma})^{-1},\end{aligned} \tag{89}$$

where we have used the relation

$$\langle a'_{n'};-\mathbf{k}_i;\mathbf{k}_\sigma|H_1|c_{n'};-\mathbf{k}_i'\rangle=\langle a'_{n'};-\mathbf{k}_i;\mathbf{k}_\sigma|H_1|c_{n'};-\mathbf{k}_i\rangle\delta_{\mathbf{k}_i,\mathbf{k}_i'}.$$

If we now introduce the matrix elements of the two Green's functions given in (76′) and (88), we find, on performing the sum over i'', that the transition amplitude is given by

$$I_{a'n'\sigma,a_n}(t)=\int_c \frac{dEe^{-iEt}\langle a'_{n'};-\mathbf{k}_i;\mathbf{k}_\sigma|H_1|c_{n'};-\mathbf{k}_i\rangle\langle c_{n'}|H_2|b_n\rangle\langle b_n;-\mathbf{k}_i|H_1|a_n\rangle}{(2\pi i)(E-E_{a',n';-\mathbf{k}_i;+\mathbf{k}_\sigma})(E-E_{a,n}-\Delta_a(E)+i\frac{1}{2}\Gamma_a(E))\det}, \tag{90}$$

with det given by (78′). Here n' is restricted to $n\pm1$. The expression for det is in the form $(E-E_+)(E-E_-)$, where $\mathrm{Im}E_\pm<0$. Hence, for times long compared to Γ_a^{-1} and $(\mathrm{Im}E_\pm)^{-1}$, the major contribution to the above integral occurs at $E=E_{a',n';-\mathbf{k}_i;\mathbf{k}_\sigma}$. (Here, we ignore the contribution to $I_{a'n'\sigma,a_n}$ that shows nonexponential dependence.) In ignoring the contributions to the above integral that damp out in time, it must be noted that the time scale used is in terms of the periods of the pertinent frequencies, here the optical and rf frequencies. With this in mind, the transition amplitude, for times long compared to Γ_a^{-1} and $\mathrm{Im}(E_\pm)^{-1}$, is given by

$$I_{a'n'\sigma,a_n}(t)\sim\frac{\langle a'_{n'};-\mathbf{k}_i;\mathbf{k}_\sigma|H_1|c_{n'};-\mathbf{k}_i\rangle\langle c_{n'}|H_2|b_n\rangle\langle b_n;-\mathbf{k}_i|H_1|a_n\rangle e^{-iE_{a',n';-\mathbf{k}_i;\mathbf{k}_\sigma}t}}{(E_{a',n';-\mathbf{k}_i;\mathbf{k}_\sigma}-E_{a,n}-\Delta_a'+i\frac{1}{2}\Gamma_a')\det'}, \tag{91}$$

with det′ given by

$$\det'\equiv(E_{a',n';-\mathbf{k}_i;\mathbf{k}_\sigma}-\bar{E}_{c,n';-\mathbf{k}_i}-\Delta_{c,n'}+i\tfrac{1}{2}\Gamma_c)(E_{a',n';-\mathbf{k}_i;\mathbf{k}_\sigma}-\bar{E}_{b,n;-\mathbf{k}_i}-\Delta_{b,n}+i\tfrac{1}{2}b)-|\langle c_{n'}|H_2|b_n\rangle|^2. \tag{92}$$

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[14] See Ref. 12, Sec. 20.
[15] J. P. Barrat and C. Cohen-Tannoudji, J. Phys. Radium **22**, 329 (1961).

The energy differences are given by

$$E_{a',n\pm1;-\mathbf{k}_i;\mathbf{k}_\sigma}-\tilde{E}_{c,n\pm1;-\mathbf{k}_i}=E_{a'}-\tilde{E}_c+k_\sigma,$$
$$E_{a',n\pm1;-\mathbf{k}_i;\mathbf{k}_\sigma}-E_{b,n;-\mathbf{k}_i}=E_{a'}-\tilde{E}_b+k_\sigma\pm\omega,$$
$$E_{a',n\pm1;-\mathbf{k}_i;\mathbf{k}_\sigma}-E_{a,n}=E_{a'}-E_a+k_\sigma-k_i\pm\omega. \quad (93)$$

The transition amplitude as given by (91) may serve, for instance, as a first step in a more complete description of the magnetic resonance in an excited state observed using optical pumping techniques.[16] The major changes that would result from this approach would be in the appearance of an intensity-dependent shift of the resonance frequency. Under near saturation conditions, this effect could be important.

When the spectral energy distribution of the beam of photons is quite broad, the shape of the emission line becomes independent of this distribution. To see this we sum the probability for the transition $a_n \rightarrow a'_{n,\sigma}$ over the distribution of absorbed photon:

$$P_{a'n'\sigma,b_n}(t)\equiv\sum_i |I_{a'n'\sigma,a_n}(t)|^2$$
$$\cong\int_{-\infty}^{\infty} dk_i U(k_i)|I_{a'n'\sigma,a_n}(t)|^2. \quad (94)$$

Here we have anticipated the similarity to the transition probability for stimulated emission from the state $|b_n\rangle$ by changing the subscript. If we drop the extraneous factors in the description of the states in the first matrix element in (91), we find

$$P_{a'n'\sigma,b_n}(t)=|\langle a'_{n'};\mathbf{k}_\sigma|H_1|c_{n'}\rangle\langle c_{n'}|H_2|b_n\rangle/\det'|^2$$
$$\times\int_{-\infty}^{\infty} U(k_i)dk_i|\langle b_n;-\mathbf{k}_i|H_1|a_n\rangle|^2/$$
$$[(E_{a'}-E_a+k_\sigma-k_i+(n'-n)\omega-\Delta_{a'})^2+\tfrac{1}{4}\Gamma_{a'}{}^2]. \quad (95)$$

By assumption, $U(k_1)$ varies slowly in the vicinityof

$$k_i=k_0\equiv(k_\sigma+E_{a'}+n'\omega)-(E_a+n\omega+\Delta_{a'}), \quad (96)$$

where k_0 is equal to the difference in energy between the final state and the initial state. In addition, in the dipole approximation the matrix element $|\langle b_n;-\mathbf{k}_i|H_1|a_n\rangle|^2$ varies as $k_i^{\pm1}$ [see (50)], which is essentially constant in the immediate vicinity of $k_i=k_0$. We assume that $U(k_i)$ falls sufficiently fast near $k_i=0$ to avoid any singularities at zero energy. The major contribution to the transition probability occurs for those photons satisfying the energy conservation condition, (96), and is given by

$$P_{a'n'\sigma,b_n}=|\langle a'_{n'};\mathbf{k}_\sigma|H_1|c_{n'}\rangle\langle c_{n'}|H_2|b_n\rangle/\det'|^2$$
$$\times[2\pi U(k_i)/\Gamma_a'(k_i)]|\langle b_n;-\mathbf{k}_i|H_1|a_n\rangle|^2|_{k_i=k_\sigma}. \quad (97)$$

For a final photon $\mathbf{k}_\sigma$ satisfying the energy conservation condition (96), the transition probability per unit time $\Gamma_a'(k_i)$ for absorption of a photon from the initial state is determined by (86):

$$\Gamma_a'\equiv\Gamma_a(E=E_{a',n';-\mathbf{k}_i,\mathbf{k}_\sigma})$$
$$=-\tfrac{1}{2}\,\mathrm{Im}\langle a_n|R(E=E_{a',n';-\mathbf{k}_i;\mathbf{k}_\sigma})|a_n\rangle. \quad (98)$$

An evaluation of the above matrix element as given by (84) involves an integration over the spectral energy distribution of the initial photons. The integrand in (84) depends on k_i only implicitly through the final photon k_σ. This dependence is made explicit by the energy conservation condition (96). By repeating the analysis leading up to (97) we find that

$$\Gamma_a'(k_i)=2\pi U(k_i)|\langle a_n|H_1|b_n;-\mathbf{k}_i\rangle|^2, \quad (99)$$

where $k_i=k_0$ by (96). Finally, if we integrate over the angle of the emitted photon $\mathbf{k}_\sigma$, we find that the transition probability per unit energy range for the stimulated emission of a photon k_σ, is given by

$$P_{a'n'b_n}=(\Gamma_c/2\pi)|\langle c_{n\pm1}|H_2|b_n\rangle/\det'|^2, \quad (100)$$

where

$$|\det'|^2=[(E_{a'}-\tilde{E}_c+k_\sigma-\Delta_{c,n\pm1})$$
$$\times(E_{a'}-\tilde{E}_b+k_\sigma\pm\omega-\Delta_{b,n})$$
$$-\tfrac{1}{4}\Gamma_b\Gamma_c-|\langle c_{n\pm1}|H_2|b_n\rangle|^2]^2$$
$$+\tfrac{1}{4}[\Gamma_c(E_{a'}-\tilde{E}_b+k_\sigma\pm\omega-\Delta_{b,n})$$
$$+\Gamma_b(E_{a'}-E_c+k_\sigma-\Delta_{c,n\pm1})]^2.$$

Apart from notational differences, this result is in close agreement with the second part of Eq. (85) or Ref. 2. The subsequent analysis given there can be followed *if* we note that the rf resonance no longer occurs at the Bohr frequency, but is shifted by the rf and optical intensity-dependent terms included in $\Delta_{b,n}$ and $\Delta_{c,n\pm1}$.

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APPENDIX

The relationship between the level shift operators that appears in the step-by-step analysis given in the text is easy to come by. We start out with the definition of the level shift operator for the $j-1$th step in the decay process:

$$R^{(j-1)}(E)\equiv V+VP_{j-1}(E-P_{j-1}HP_{j-1})^{-1}P_{j-1}V. \quad (A1)$$

The projection operator P_{j-1} acting on any of the previous j states ($|a\rangle$, $|1\rangle$, $\cdots$, $|j-1\rangle$) yields no contribution—it in effect couples to the remaining portion of the spectrum. From its definition we see that

$$P_{j-1}=P_j+\Lambda_j. \quad (A2)$$

[16] J. P. Barrat and C. Cohen-Tannoudji, J. Phys. Radium **22**, 443 (1961).

Hence, if we take the four projections of

$$P_{j-1}(E-P_{j-1}HP_{j-1})^{-1}$$

onto the various subspaces spanned by the combinations of P_j and Λ_j, we will be able to relate $R^{j-1}(E)$ to $R^j(E)$. We start with the identity

$$(E-P_{j-1}HP_{j-1})P_{j-1}(E-P_{j-1}HP_{j-1})^{-1}P_{j-1}=P_{j-1} \quad \text{(A3)}$$

and multiply from the left first by P_j and then by Λ_j. The two operator equations that result,

$$\begin{aligned}&P_j(E-P_jHP_j)P_j(E-P_{j-1}HP_{j-1})^{-1}P_{j-1}\\&\qquad -P_jH\Lambda_j(E-P_{j-1}HP_{j-1})^{-1}P_{j-1}=P_j,\\&-\Lambda_jHP_j(E-P_{j-1}HP_{j-1})^{-1}P_{j-1}+\Lambda_j(E-\Lambda_jH\Lambda_j)\\&\qquad\times\Lambda_j(E-P_{j-1}HP_{j-1})^{-1}P_{j-1}=\Lambda_j,\end{aligned} \quad \text{(A4)}$$

may be formally solved for the projections desired. If we assume that the inverses of $\Lambda_j(E-\Lambda_jH\Lambda_j)$ and $P_j(E-P_jHP_j)$ exist, we find

$$\begin{aligned}&\Lambda_j(E-P_{j-1}HP_{j-1})^{-1}P_{j-1}\\&\qquad=\Lambda_jG^{(j)}(E)[\Lambda_j+\Lambda_jHP_j(E-P_jHP_j)^{-1}P_j],\\&P_j(E-P_{j-1}HP_{j-1})^{-1}P_{j-1}=P_j(E-P_jHP_j)^{-1}P_j\\&\qquad+P_j(E-P_jHP_j)^{-1}P_jH\Lambda_jG^{(j)}(E)\\&\qquad\times[\Lambda_j+\Lambda_jHP_j(E-P_jHP_j)^{-1}P_j],\end{aligned} \quad \text{(A5)}$$

where we have introduced the definitions

$$\begin{aligned}\Lambda_jG^{(j)}(E)\Lambda_j&\equiv\Lambda_j[E-K-\Lambda_jR^{(j)}(E)\Lambda_j]^{-1},\\R^{(j)}(E)&\equiv V+VP_j(E-P_jHP_j)^{-1}P_jV.\end{aligned} \quad \text{(A6)}$$

If we combine these two expressions, we find

$$\begin{aligned}&P_{j-1}(E-P_{j-1}HP_{j-1})^{-1}P_{j-1}\\&\quad\equiv(P_j+\Lambda_j)(E-P_{j-1}HP_{j-1})^{-1}P_{j-1}\\&\quad=P_j(E-P_jHP_j)^{-1}P_j\\&\qquad+[P_j(E-P_jHP_j)^{-1}P_jH\Lambda_j+\Lambda_j]G^{(j)}(E)\\&\qquad\times[\Lambda_j+\Lambda_jHP_j(E-P_jHP_j)^{-1}P_j].\end{aligned} \quad \text{(A7)}$$

The substitution of this relation into (A1) yields the following expression for $R^{j-1}(E)$:

$$\begin{aligned}R^{(j-1)}(E)&=R^{(j)}(E)\\&\quad+V[P_j(E-P_jHP_j)^{-1}P_jH\Lambda_j+\Lambda_j]G^{(j)}(E)\\&\qquad\times[\Lambda_j+\Lambda_jHP_j(E-P_jHP_j)^{-1}P_j]V\end{aligned} \quad \text{(A8)}$$

or

$$R^{(j-1)}(E)=R^j(E)+R^{(j)}(E)\Lambda_jG^{(j)}(E)\Lambda_jR^{(j)}(E), \quad \text{(A9)}$$

which is seen to be identical with the expression (33) given in the text. In arriving at (A9) we have made use of the definition of $R^{(j)}(E)$ given in (A6) and the fact that by definition K has no matrix elements between the subspaces spanned by Λ_j and P_j.