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Decay Theory of Closely Coupled Unstable States

LYMAN MOWER*
Joint Institute for Laboratory Astrophysics, Boulder, Colorado
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A treatment of decay theory is presented which generalizes existing treatments so as to apply to closely coupled unstable states. The treatment is based upon a Green's function formulation for the transition amplitude in which the states of interest are selected by suitable projection operators. As an indication of the practicality of the resulting formulation a number of examples are given involving the stimulated transition between closely coupled unstable states.

1. INTRODUCTION

THE quantum theory of radiation is by now a familiar subject that has been ably presented by Heitler on the basis of time-dependent perturbation theory.[1] Recently a number of treatments have appeared that present the subject from a different point of view.[2,3] In particular, that of Goldberger and Watson parallels the formal treatment of time-independent scattering theory based on a Green's function formulation. The power of this approach is seen by noting that this formulation can be made exact and is easily able to demonstrate the nonexponential decay properties of excited states—a subject missing from the perturbation-theory approach of Heitler. The nonexponential decay illustrated by the Green's-function technique shows a power-series dependence on time, the leading term of which depends on the energy dependence of the density of final states.

Common to these treatments are a number of drawbacks. To begin with, the initial and final states are treated on an unequal footing, thus making the consideration of transitions between unstable levels unnecessarily complicated. Secondly, radiative transitions between unstable states are inaccurately treated in that contributions to the natural lifetime may be inadvertently left out. This omission also shows up in the treatment of resonance fluorescence of coupled levels under anticrossing conditions.[4] Thirdly, a more practical, but perhaps a more important, consideration is that the formalism rapidly becomes unwieldy when other than isolated levels are concerned.

A method to remove these objections is suggested by Feshbach's treatment of nuclear reaction theory.[5] This involves the introduction of suitable projection operators that select out the isolated levels as well as the sets of coupled levels of interest. It is the purpose of this note to extend this treatment so that it will apply to transition probabilities. The derivation given is based on the Green's function and the determination of its matrix elements in the unperturbed energy representation when many levels are strongly coupled by an interaction energy. The directness and the simplicity of this approach, however, are obtained at the cost of the introduction of the matrix elements of the level shift operator, the properties of which have been described in detail by Goldberger and Watson.[3] For the case of a single excited state our derivation is equivalent to that of Goldberger and Watson and represents a more direct approach. The advantage of this approach is realized when several closely coupled states are involved.

As an illustration of the practical utility of this formulation, we give in the last section a number of

* Visiting Fellow during 1964–65; present address: Physics Department, University of New Hampshire, Durham, New Hampshire.

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[5] H. Feshbach, Ann. Phys. **19**, 287 (1962).

applications that represent different aspects of stimulated transitions between unstable states.

2. FORMAL DERIVATION

We consider to begin with a system that is initially in a given state. The system is made unstable by the presence of a perturbation that induces a transition from the original state. The probability that at time t the system has made a transition from one state to another may be calculated if we can follow the system as it evolves in time. If originally the system was in the ath eigenstate of the Hamiltonian K, then through the perturbation V, at time t it will have evolved according to the expression ($\hbar=1$)

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

where

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

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

The transition amplitude at time t between the states $|a\rangle$ and $|b\rangle$ belonging to the spectrum of K is given by

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

which may be expressed in terms of the Green's function as

$$I_{ba}(t)=\frac{1}{2\pi i}\int_c dE\, e^{-iEt}\langle b|G(E)|a\rangle. \tag{5}$$

The contour c runs from $+\infty$ to $-\infty$ above the real axis above the singularities of $G(E)$.

To calculate the transition amplitude and hence the transition probability we need to know the energy dependence of the Green's function in the representation appropriate to the unperturbed spectrum. If the levels a and b are isolated from the remaining spectrum of K, then the problem is relatively simple. However, if the levels are not isolated, the evaluation of $I_{ba}(t)$ is not simple. To reduce the complexity somewhat and to make clear the states of the system to be included in the evaluation of the matrix elements of the Green's function, we introduce the language of projection operators: we introduce the relations

$$P+Q=1,\quad QP=PQ=0,\quad P^2=P, \tag{6}$$

where P is the projection operator onto a particular set of levels which we wish to treat on an equal footing. It takes the form

$$P\equiv\sum_{i,\alpha}|i\alpha\rangle\langle i\alpha|, \tag{7}$$

where

$$K|i\alpha\rangle=E_{i,\alpha}|i\alpha\rangle. \tag{8}$$

The variables i and α as well as their ranges depend on the type of problem to be considered.

Using the projection operator language it is clear that we need to know the two projections of the Green's function: $QG(E)P$ and $PG(E)P$. The first projection gives us the matrix element of G between a state outside the set of levels and a state inside the set of levels which we wish to treat on an equal basis. The second projection operator has matrix elements only between members of the set of levels in question. To determine the above projections of the Green's function we start with the identity

$$(E-H)G(E)=1. \tag{9}$$

Two coupled operator equations may be obtained for the projections QGP and PGP by first multiplying this identity from the right by P:

$$(E-H)PG(E)P+(E-H)QG(E)P=P \tag{10}$$

and then taking the left projection of this equation onto first P and then Q:

$$P(E-H)P(PGP)-PHQ(QGP)=P, \tag{11}$$

$$-QHP(PGP)+Q(E-H)Q(QGP)=0. \tag{12}$$

Here we have introduced the identities

$$Q^2=Q,\quad P^2=P, \tag{13}$$

to clarify the nature of the operators. On the assumption that the operator $Q(E-H)Q$ possesses an inverse in the space spanned by Q, we may solve for QGP in terms of PGP:

$$QGP=(E-QHQ)^{-1}QHP(PGP). \tag{14}$$

Hence, the substitution of this expression into the first of the above pair of equations leads to

$$[(E-PHP)-PHQ(E-QHQ)^{-1}QHP]PGP=P, \tag{15}$$

which may now be inverted to read

$$PG(E)P=[E-PKP-PRP]^{-1}P. \tag{16}$$

Thus, the second projection operator of interest is given by

$$QG(E)P=(E-QHQ)^{-1}QHP(E-PKP-PRP)^{-1}, \tag{17}$$

where the integral operator R, known as the level shift operator, is given by

$$R=V+VQ(E-QHQ)^{-1}QV. \tag{18}$$

For the case of a single isolated state, the projection operator P is simply

$$P\equiv|a\rangle\langle a|, \tag{19}$$

and the above formulation is equivalent to that of Goldberger and Watson.[3]

In order to write down the actual matrix element $\langle b|G(E)|a\rangle$, it is necessary to specify whether or not the state $|b\rangle$ is to be included in the set of closely coupled states. If it is, then the matrix element in question is

given by

$$\langle b|G(E)|a\rangle=\langle b|PGP|a\rangle$$
$$=\langle b|(E-PKP-PRP)^{-1}|a\rangle. \quad (20)$$

For the case in which the set of coupled levels is a finite and small number, the matrix elements of the inverse of the operator $(E-PKP-PRP)$ may be easily calculated. In general the above matrix element may be written formally as

$$\langle b|G(E)|a\rangle=\frac{[\text{cofactor}(E-PKP-PRP)]ab}{\text{Det}[E-PKP-PRP]}. \quad (21)$$

For the interesting case of two coupled levels, this expression reduces to

$$\langle a|G(E)|a\rangle=\frac{E-E_b-R_{bb}(E)}{[E-E_a-R_{aa}(E)][E-E_b-R_{bb}(E)]-R_{ab}(E)R_{ba}(E)},$$
$$\langle b|G(E)|a\rangle=-\frac{R_{ba}(E)}{[E-E_a-R_{aa}(E)][E-E_b-R_{bb}(E)]-R_{ab}(E)R_{ba}(E)}. \quad (22)$$

For a greater number of coupled states the matrix elements of PGP are considerably more complicated.

If, on the other hand, the state $|b\rangle$ does not belong to the set of levels belonging to the projection operator P, then the matrix element of G is given by

$$\langle b|G(E)|a\rangle=\langle b|QGP|a\rangle=\sum_{1,c}\langle b|(E-QHQ)^{-1}|1\rangle\langle 1|QHP|c\rangle\langle c|(E-PKP-PRP)^{-1}|a\rangle. \quad (23)$$

Here, the states $|1\rangle$ represent a complete set of states, while $|c\rangle$ represents the set of levels belonging to P. If, we now make use of the algebraic relationship

$$(E-QHQ)^{-1}=(E-QKQ)^{-1}+(E-QKQ)^{-1}QVQ(E-QHQ)^{-1},$$

then we see that for those perturbations V that couple only the state $|b\rangle$ with the members of the closely coupled set the above expression for the matrix element of QGP simplifies to

$$\langle b|G(E)|a\rangle=(E-E_b)^{-1}\sum_c\langle b|V|c\rangle\langle c|(E-PKP-PRP)^{-1}|a\rangle. \quad (24)$$

In arriving at this expression we have introduced for the complete set $|1\rangle$ that set of states belonging to K. If, in addition to the above form for the perturbation V, we have only two closely coupled levels $|a\rangle$ and $|c\rangle$, then the above matrix element of QGP becomes

$$\langle b|G(E)|a\rangle=\frac{[\langle b|V|a\rangle(E-E_c-R_{cc})-\langle b|V|c\rangle R_{ca}]}{(E-E_b)\{[E-E_c-R_{cc}(E)][E-E_a-R_{aa}(E)]-R_{ac}(E)R_{ca}(E)\}}. \quad (25)$$

The form given in (17) for QGP includes the possibility of the interaction of the final state of the system with the perturber via intermediate states. When this is possible it is sometimes desirable to replace $(E-QHQ)^{-1}$ by $(E-QKQ)^{-1}$. To do this is an exact fashion, we make use of the relationship

$$(E-QKQ)^{-1}QRP=(E-QHQ)^{-1}QVP=(E-QHQ)^{-1}QHP, \quad (26)$$

which follows from the definition for QHP:

$$QRP=QVP+QVQ(E-QHQ)^{-1}QVP=(E-QKQ)(E-QHQ)^{-1}QVP. \quad (27)$$

Thus, an alternate form for QGP is

$$QG(E)P=(E-QKQ)^{-1}QR(E)P(E-PKP-PRP)^{-1}. \quad (28)$$

Using this form, we see that the expression for the matrix element of G between one member $|a\rangle$ of a set of closely couples states and a state $|b\rangle$ not a member of such a set is given by

$$\langle b|G(E)|a\rangle=\sum_c(E-E_b)^{-1}\langle b|R|c\rangle\langle c|(E-PKP-PRP)^{-1}|a\rangle, \quad (29)$$

where the sum is over the set of closely coupled states. If the number of closely coupled states is *two*, then the above matrix element reduces to

$$\langle b|G(E)|a\rangle=\frac{\langle b|R|a\rangle[E-E_c-R_{cc}(E)]-\langle b|R|c\rangle\langle c|R|a\rangle}{(E-E_b)\{[E-E_a-R_{aa}(E)][E-E_c-R_{cc}(E)]-R_{ac}(E)R_{ca}(E)\}}. \quad (30)$$

Here the matrix element of QGP is given exactly and no assumption need be made concerning the form for V. It is now necessary, however, to relate the matrix elements of R to those for V via the definition (18) of the level shift operator.

In the evaluation of the contour integral in (5) it is important to note the analytical properties of the level shift operator $R(E)$ as a function of complex E. For the case of a single isolated state with a simple projection operator P:

$$P=|a\rangle\langle a|,$$

the analytical properties of R have been presented at length by Goldberger and Watson.[3] As the complications introduced by considering other than single isolated states do not change the analytical properties of the level shift operator, we summarize, briefly, their results.

When all the states connected to the initial state by the perturbation are discrete and have finite norm, then the singularities of R as a function of E are all discrete and lie along the real axis. The resulting transition amplitude is oscillatory in nature and describes a reversible situation. In the limit that some of the states connected to the initial state have a nonfinite norm and describe the possibility of a nonreversible decay of the initial state, then the discrete spectra of the level shift operator merge to form a discrete spectrum superimposed onto a continuous spectrum along the real axis. This condition is realized, for instance, when the dimensions of the box relative to the dimensions of the "atomic system" contained therein become infinite. When this occurs the level shift operator is no longer Hermitian and as a function of complex E is analytic everywhere save on the real axis where it has a number of discrete poles superimposed onto a continuum. From the nature of the singularities the form of the level shift operator as a function of complex E is determined to be

$$R_{aa}{}^{\pm}(E)=D_{aa}(E)\mp iI_{aa}(E),$$

where

$$\mathrm{Re}R_{aa}{}^{\pm}(E)=D_{aa}(E),\quad \mathrm{Im}R_{aa}{}^{\pm}(E)=\mp I_{aa}(E).$$

Here the $\pm$ superscript refers, respectively, to the upper and lower region of the complex E plane. (To make this statement concerning the form of R it is necessary to assume that $\langle a|VQV|a\rangle<\infty$.) The insertion of this form for the diagonal matrix elements of R into the expressions for the matrix element of the Green's function $\langle b|G(E)|a\rangle$ serve to show that as a function of E, $\langle b|G(E)|a\rangle$ is analytic everywhere off the real axis.

Along the real axis, $I_{aa}(E)$ is different from zero only along the cut $E>E_m$, where E_m is the lowest energy value in the continuum. Hence $\langle b|G(E)|a\rangle$ is analytic everywhere in the complex plane save on the cut along the real axis $E>E_m$. To evaluate the contour integral in (5) by contour analysis it is necessary to continue analytically the matrix elements of the level shift operator from the first Riemann sheet through the cut onto the second sheet. This may be done by defining the value of the level shift operator on the second sheet in the vicinity of the cut as

$$R_{\mathrm{II}}(\epsilon\pm i\eta)=R_{\mathrm{I}}(\epsilon\mp i\eta),$$

where the complex energy near the cut has been written as

$$E=\epsilon\pm i\eta,\quad \eta>0.$$

With this definition the contributions to the transition amplitude in (5) include the residues at the poles on the second sheet plus a contribution along a new branch cut. Further details may be found in Goldberger and Watson.[3]

In extending the above discussion to the more general form for R given by (18), we note that the formalism is such as to render the off-diagonal matrix elements of R as Hermitian and to remove the rapid energy variations from all matrix elements of R. Hence we may apply directly the above considerations concerning the analytical properties of R for the simple projection operator to the more general case. In writing down the form for the various diagonal matrix elements of the level shift operator we shall refer to Goldberger and Watson for details where necessary.

3. APPLICATIONS OF FORMULATION

The applications in the following section have been chosen primarily as simple illustrations of the theory. Several applications represent different aspects of the stimulated transition between unstable levels. The results may be of some interest in spectroscopy for discussing both beam and swarm experiments. While the examples chosen are mainly from atomic physics, the theory developed in the previous section is not so limited. As an illustration of this we also consider the possibility of a model possessing a double pole and discuss briefly the nature of its decay.

3.1 Stimulated Transitions between Two Unstable States

As a first illustration we consider the stimulated transition between two unstable levels. We assume for simplicity that the two levels are isolated in the sense that the only stimulated transitions possible are between the two levels of interest. The amplitude for the transition is given by (5):

$$I_{ba}(t)=(1/2\pi i)\int_c e^{-iEt}dE\langle b|G(E)|a\rangle, \qquad (5)$$

where for a two-level system the matrix element of the Green's function is given by (22). The perturbation energy for this illustration is assumed to have two parts:

$$V=V_r+H_1, \qquad (31)$$

where V_r refers to the coupling of the states to the radiation field and H_1 refers to the coupling with the external field inducing the transitions between the two states. With this decomposition the matrix elements of the level shift operator may be written down directly if we first note that the projection operator Q is given by

$$Q=1-|a\rangle\langle a|-|b\rangle\langle b|$$

in this application. Hence, the matrix elements of the level shift operator on the second Riemann sheet $\mathrm{Im}E<0$ are given by[3]

$$\begin{aligned} R_{aa}&=\langle a|V_r(E-K-QV_rQ)^{-1}V_r|a\rangle \\ &\equiv D_a-\tfrac{1}{2}i\Gamma_a, \\ R_{bb}&=D_b-\tfrac{1}{2}i\Gamma_b, \\ R_{ab}&=\langle a|H_1|b\rangle\equiv H_{1ab}=H_{ab}. \end{aligned} \tag{32}$$

In writing these matrix elements we have assumed that the external perturbation couples the states a and b only and that the interaction energy with the radiation field V_r has nonvanishing matrix elements between the states a or b and the continuum ("ground" states) only. In addition, we have tacitly recognized that the level shift operator has no nondiagonal matrix elements involving V_r. Hence, the transition amplitude is given by

$$\begin{aligned} I_{ba}(t)&=\frac{-1}{2\pi i}\int_c \frac{dE\, e^{-iEt}R_{ba}(E)}{[(E-E_a-R_{aa})(E-E_b-R_{bb})-R_{ab}R_{ba}]} \\ &=\frac{-1}{2\pi i}\int_c dE\, e^{-iEt}\frac{H_{ba}}{(E-E_+)(E-E_-)}, \end{aligned} \tag{33}$$

where on the second Riemann sheet E_+ and E_- are the two complex roots of

$$(E-\bar{E}_a+\tfrac{1}{2}i\Gamma_a)(E-\bar{E}_b+\tfrac{1}{2}i\Gamma_b)-|H_{ab}|^2=0 \tag{34}$$

or

$$\begin{aligned} E_\pm=&\tfrac{1}{2}(\bar{E}_a+\bar{E}_b)-\tfrac{1}{4}i(\Gamma_a+\Gamma_b) \\ &\pm\tfrac{1}{2}[(\bar{E}_a-\bar{E}_b+\tfrac{1}{2}i\Gamma_b-\tfrac{1}{2}i\Gamma_a)^2+4|H_{ab}|^2]^{1/2}. \end{aligned}$$

In writing this expression, we have absorbed the level shifts D_a and D_b in the terms $\bar{E}_a\equiv E_a+D_a$, $\bar{E}_b\equiv E_b+D_b$. Here, the contour c lies above the two poles at E_+ and E_-.

If for the moment we ignore the contribution to the transition amplitude that shows nonexponential decay, then the transition amplitude is found simply by completing the contour in the lower half-plane and evaluating the integral at the two singularities given by (34). We find that $I_{ba}(t)$ is given by

$$I_{ba}(t)=-[H_{ba}/(E_+-E_-)](e^{-iE_+t}-e^{-iE_-t}). \tag{35}$$

To exhibit the dependence of $I_{ba}(t)$ on the levels $\bar{E}_a$ and $\bar{E}_b$ we write out explicitly the real and imaginary parts of $E_\pm$:

$$E_+=(a+b)+i(-c+d),\quad E_-=(a-b)+i(-c-d), \tag{36}$$

where

$$\begin{aligned} a&=\tfrac{1}{2}(\bar{E}_a+\bar{E}_b), \\ b&=\tfrac{1}{4}\sqrt{2}[(A^2+B^2)^{1/2}+A]^{1/2}, \\ c&=(\Gamma_a+\Gamma_b)/4, \\ d&=\tfrac{1}{4}\sqrt{2}[(A^2+B^2)^{1/2}-A]^{1/2}\,\mathrm{sgn}B, \end{aligned}$$

and

$$\begin{aligned} A&=(\bar{E}_a-\bar{E}_b)^2-\tfrac{1}{4}(\Gamma_a-\Gamma_b)^2+4|H_{ab}|^2, \\ B&=(\bar{E}_a-\bar{E}_b)(\Gamma_b-\Gamma_a). \end{aligned}$$

Using the above expression for E_+ and E_-, we see that the transition amplitude takes the following form:

$$I_{ba}(t)=-[H_{ba}/2(b+id)](e^{-ibt+dt}-e^{ibt-dt})e^{-iat-ct}. \tag{37}$$

The transition probability is given by

$$\begin{aligned} P_{ba}(t)&\equiv|I_{ba}(t)|^2 \\ &=[|H_{ba}|^2/4(b^2+d^2)]e^{-2ct}(e^{2dt}+e^{-2dt}-e^{2ibt}-e^{2ibt}) \\ &=[|H_{ba}|^2/2(b^2+d^2)]e^{-2ct}(\cosh 2dt-\cos 2bt). \end{aligned} \tag{38}$$

In the limit that $|H_{ba}|^2$ goes to zero,

$$d=\tfrac{1}{4}\sqrt{2}[(A^2+B^2)^{1/2}-A]^{1/2}\to\tfrac{1}{4}|\Gamma_a-\Gamma_b|<c. \tag{39}$$

This inequality is still satisfied for $H_{ba}\neq 0$; hence, the above probability is finite for all time.

Usually what is required is the total probability that a transition has occurred. This may be expressed as

$$\begin{aligned} P_T&\equiv\Gamma_b\int_0^\infty P_{ba}(t)dt \\ &=\frac{\Gamma_b|H_{ba}|^2}{2(b^2+d^2)}\int_0^\infty e^{-2ct}(\cosh 2dt-\cos 2bt)dt. \end{aligned} \tag{40}$$

For $c>d$ the integral is finite, and we find

$$P_T=\frac{\Gamma_b|H_{ba}|^2c}{4(c^2-d^2)(c^2+b^2)}. \tag{41}$$

In terms of the above expressions for b, c, and d, we see that

$$\begin{aligned} (c^2-d^2)(c^2+b^2)&=(\tfrac{1}{16})^2\{[(\Gamma_a+\Gamma_b)^2+2A]^2-4(A^2+B^2)\} \\ &=(\tfrac{1}{16})^2\{(\Gamma_a+\Gamma_b)^4+4A(\Gamma_a+\Gamma_b)^2-4B^2\}. \end{aligned}$$

Hence, in terms of the energy values $\bar{E}_a$ and $\bar{E}_b$,

$$\begin{aligned} (c^2-d^2)(c^2+b^2)=&(\tfrac{1}{8})^2\{4\Gamma_a\Gamma_b(\bar{E}_a-\bar{E}_b)^2 \\ &+(\Gamma_a+\Gamma_b)^2(4|H_{ab}|^2+\Gamma_a\Gamma_b)\}. \end{aligned}$$

Thus, the total probability that such a transition does occur is given by

$$P_T=\frac{|2H_{ba}|^2\Gamma_b(\Gamma_a+\Gamma_b)}{4\Gamma_a\Gamma_b(\bar{E}_a-\bar{E}_b)^2+(\Gamma_a+\Gamma_b)^2(|2H_{ab}|^2+\Gamma_a\Gamma_b)}. \tag{42}$$

The identification of this expression with that usually given is seen by noting that in the unperturbed

representation

$$K\equiv H_0+H_{\rm rf}, \tag{43}$$

where H_0 denotes the Hamiltonian of the system in the absence of the rf field, while $H_{\rm rf}$ denotes the Hamiltonian of the isolated field. Hence the two energy levels $\bar{E}_a$ and $\bar{E}_b$ are given by ($\hbar=1$)

$$\bar{E}_a\equiv E_a+(n)\omega, \quad \bar{E}_b\equiv E_b+(n+1)\omega, \tag{44}$$

where the level shifts are included in E_a, E_b. Hence, in terms of the frequency of the level separation,

$$E_a-E_b=\omega_{ab},$$

the total transition probability is given by

$$P_T=\frac{|2H_{ab}|^2\Gamma_b(\Gamma_a+\Gamma_b)}{4\Gamma_a\Gamma_b(\omega_{ab}-\omega)^2+(\Gamma_a+\Gamma_b)^2(|2H_{ab}|^2+\Gamma_a\Gamma_b)}, \tag{45}$$

which agrees with the expression found by Bennett.[6]

(a) *Nonexponential Decay Associated with Stimulated Transitions*

The contribution to the transition probability just found displays an exponential decay and is valid for times such that $(1/tE)\ll 1$ provided that $\Gamma/E\ll 1$ also. Inasmuch as the transition probability exhibits a resonance phenomenon, it is of some interest to obtain an estimate of the contribution to the transition probability that displays a nonexponential decay character. To this end we now re-examine the problem in detail.

As noted above, the transition amplitude between the two closely coupled unstable states $|a\rangle$ and $|b\rangle$ is given by

$$I_{ba}(t)=\frac{-1}{2\pi i}\int_c \frac{e^{-iEt}R_{ba}(E)dE}{[E-E_a-R_{aa}(E)][E-E_b-R_{bb}(E)]-R_{ab}R_{ba}}, \tag{46}$$

where the contour runs from $+\infty$ to $-\infty$ above the real axis along which the singularities of $R(E)$ lie. For purposes of clarity, we repeat our remarks made in Sec. 2 concerning the analytic properties of $R(E)$. In doing so we follow closely the approach given by Goldberger and Watson.[3]

As a function of complex E the level shift operator is analytic everywhere in the complex plane save for a cut along the real axis for $E>E_m$ plus a number of discrete poles embedded in the cut. The first constitutes the spectrum of H belonging to the continuum while the second are the eigenvalues of the discrete bound states. (Again, this statement of analyticity depends on the inequality $\langle a|VQV|a\rangle<\infty$, which is violated in an unrenormalized, nonrelativistic quantum electrodynamics. This difficulty may be removed by introducing a suitable upper-limit cutoff on any integrals over real E.) To utilize the method of contour analysis it is necessary to continue $R(E)$ through the cut from the first Riemann sheet onto the second. This is accomplished by relating the matrix elements of the level shift operator on the two sheets near the cut through the definition

$$R_a^{\rm II}(E-i\eta)=R_a^{\rm I}(E+\eta),$$

where η is an infinitesimal and greater than zero. As before, the form of the matrix element of the level shift operator on the first sheet is given by

$$(R_a^{\rm I})^{\pm}(E)=D_a(E)\mp iI_a(E),$$

where the $\pm$ denotes, respectively, the upper and lower half-plane of complex E.

Having introduced the analytical continuation of the diagonal matrix elements of the level shift operator, we may now deform the contour by bending it down at $E=E_m$ along the first sheet to $E=E_m-i\infty$. The contour to the right of $E=E_m$ may be deformed in a similar fashion provided we pass through the cut onto the second sheet. In doing so we also must pick up any contributions arising from isolated poles of the integrand resulting from the analytical continuation of $R_{aa}(E)$ and $R_{bb}(E)$. The resulting expression for $I_{ba}(t)$ consists of two contour integrals, one about the new cut extending from E_m to $E_m-i\infty$ as well as the contour about any singularities of the integrand:

$$I_{ba}(t)=\frac{-1}{2\pi i}\left\{\int_{c_0}+\int_{c_1}\right\}\times\frac{e^{-iEt}dE\,R_{ba}(E)}{[E-E_a-R_a(E)][E-E_b-R_b(E)]-R_{ab}R_{ba}}. \tag{47}$$

The contours are indicated in Fig. 1.

If we equate terms of the form $1-dR_a/dE_a$ to unity, then the contribution arising from the contour c_0 is just that we have previously calculated. By equating terms of the form $1-dR_a/dE_a$ to unity we are in effect ignoring

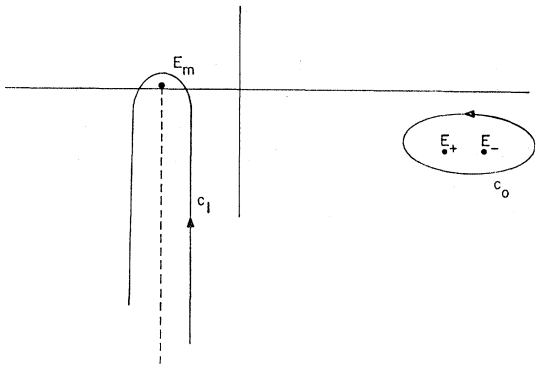

FIG. 1. Contour of integration for Eq. (47).

[6] W. R. Bennett, Jr., Phys. Rev. **126**, 580 (1962).

shifts of the energy levels E_a and E_b of the order of $D_a\Gamma_a/E_a$. For a resonance to be resolved, Γ_a/E_a must be much less than unity. Hence, this correction is negligible with respect to D_a which in itself is small. (This statement is made on the assumption that a phenomenological energy spectrum of H_0 has been introduced.)

The remaining contribution to $I_{ba}(t)$ which gives rise to the nonexponential decay may be written as

$$\begin{aligned} I_1(t)\equiv&\frac{-1}{2\pi i}\int_{c_1}\frac{R_{ba}e^{-iEt}dE}{[E-E_a-R_{aa}(E)][E-E_b-R_{bb}(E)]-R_{ab}R_{ba}}\\ =&\frac{-1}{2\pi i}\int_{E_m}^{E_m-i\infty}\frac{e^{-iEt}H_{ba}dE}{(E-E_+^{\mathrm{I}})(E-E_-^{\mathrm{I}})}+\frac{-1}{2\pi i}\int_{E_m-i\infty}^{E_m}\frac{e^{-iEt}H_{ba}dE}{(E-E_+^{\mathrm{II}})(E-E_-^{\mathrm{II}})}\\ =&\frac{-1}{2\pi i}\int_{E_m}^{E_m-i\infty}dE\,\frac{e^{-iEt}H_{ba}[(E-E_+^{\mathrm{II}})(E-E_-^{\mathrm{II}})-(E-E_+^{\mathrm{I}})(E-E_-^{\mathrm{I}})]}{(E-E_+^{\mathrm{II}})(E-E_-^{\mathrm{II}})(E-E_+^{\mathrm{I}})(E-E_-^{\mathrm{I}})}, \end{aligned}\tag{48}$$

where H_{ba} is assumed to be independent of the total energy E and where

$$\begin{aligned} E_\pm\equiv&\tfrac{1}{2}[E_a+E_b+R_a(E)+R_b(E)]\\ &\pm\tfrac{1}{2}\{[E_a-E_b+R_a(E)-R_b(E)]^2+4|H_{ba}|^2\}^{1/2}. \end{aligned}\tag{49}$$

The superscripts on $E_\pm$ denote the sheet on which the level shift operator is to be evaluated.[7] An inspection of the integral shows that for t large only contributions of the integrand near $E=0$ occur. Hence, we proceed with this in mind. The term in the bracket in the integral may be written as

$$\begin{aligned} [\ ]=&(E_a-E)[R_b^{\mathrm{II}}(E)-R_b^{\mathrm{I}}(E)]\\ &+(E_b-E)[R_a^{\mathrm{II}}(E)-R_a^{\mathrm{I}}(E)]+R_a^{\mathrm{II}}R_b^{\mathrm{II}}-R_a^{\mathrm{I}}R_b^{\mathrm{I}}. \end{aligned}$$

The introduction of the form of R_a and R_b near the real axis shows that the major contribution to the numerator near $E=0$ is $\mathrm{Lim}_{E\to 0}[\ ]=-2iE_aE_b(E)-2iE_bI_a(E)$, where we have tacitly ignored the real part of R_a, R_b in this estimate.

If we expand the integrand in a power series in $1/t$ by integration by parts, then the first nonzero contribution arises from the above limiting form of the numerator. To proceed further we introduce the following phenomenological form for $I(E)$:

$$I_a(E)=\tfrac{1}{2}\Gamma(E/E_a)^n,$$

with a similar form for $I_b(E)$. (The exponent n is an integer for radiative decay processes.) With this form, the first term in the expansion of $I_3(t)$ in powers of t is given by

$$I_1(t)\underset{t\to\infty}{\sim}-\frac{H_{ba}}{2\pi}\left(\frac{-i}{t}\right)^{n+1}n!\frac{[(E_a\Gamma_b/E_b{}^n)+E_b\Gamma_a/E_a{}^n]}{[E_aE_b-|H_{ab}|^2]^2}.\tag{50}$$

For $n=3$, appropriate to radiative decay, the nonexponential contribution to the transition amplitude is

$$\begin{aligned} I_1(t)\sim&+\frac{-6H_{ba}}{2}\frac{(\Gamma_bE_a{}^4+\Gamma_aE_b{}^4)}{(E_aE_b-|H_{ba}|^2)^2t^4E_a{}^3E_b{}^3}\\ \sim&+\frac{-3H_{ba}}{\pi E_aE_b}\left(\frac{\Gamma_b}{E_b{}^4t^4}+\frac{\Gamma_a}{E_a{}^4t^4}\right). \end{aligned}\tag{51}$$

The nonexponential contribution to I_3 does not display any resonance behavior near $E_a\cong E_b$ and, while the principal contribution for $\Gamma t\gg 1$, is in itself of negligible magnitude. Hence, we may safely ignore its effect on the total probability for the stimulated transition between two unstable levels.

(b) *Energy Variation of the Matrix Elements of the Level Shift Operator*

As a final point in our discussion of the transition probability for the stimulated transition between two unstable levels, we note that we have tacitly ignored the variation of the matrix elements of R as a function of the total energy E. If, in our treatment, we had utilized the formulation of Goldberger and Watson appropriate for isolated, unstable levels, the neglect of the energy dependence of R would have been catastrophic. Only by noting the strong energy dependence of both the diagonal and off-diagonal matrix elements in their formulation is it possible to obtain the results given in the first part of the section. We note that to treat the above problem using their formulation an order of magnitude more care and algebra are required than in the use of the generalization presented here. In effect, then, the largest contribution to the energy variation of the level shift operator has been removed by our generalization of Goldberger and Watson's formulation. What energy variation is left is largely retained in the diagonal matrix elements of R; the variation of the off-diagonal matrix elements of R with respect to the total energy E is relegated to a higher order effect. To show the effect of the residual energy variation of the

[7] To simplify the evaluation of the integral, we choose the energy scale such that E_m equal zero.

diagonal matrix elements of the level shift operator, we re-express the transition amplitude with this in mind. Again, we write the transition amplitude as

$$I_{ba}(t)=-\frac{1}{2\pi i}\times\int_c \frac{dE\, e^{-iEt}R_{ba}}{[E-E_a-R_a(E)][E-E_b-R_b(E)]-R_{ab}R_{ba}}, \tag{52}$$

where we have noted explicitly the energy dependence of the diagonal matrix elements of R only. If the energy variation of these elements in the vicinity of the value they assume at E_a or E_b is slight, then we may expand both $R_a(E)$ and $R_b(E)$ in a truncated Taylor series about these values; for instance,

$$R_a(E)\sim R_a(E_a)+(E-E_a)dR_a/dE_a.$$

Hence, the transition amplitude may be rewritten as

$$I_{ba}(t)=\frac{-1}{2\pi i}\times\int_c \frac{dE\, e^{-iEt}R_{ba}'(E_b)/(1-dR_a/dE_a)}{(E-E_a-R_a')(E-E_b-R_b')-R_{ab}'R_{ba}'}, \tag{53}$$

where for E on the second Riemann surface with $\mathrm{Im}E<0$ the prime has the following significance:

$$R_a'\equiv R_a(E_a)/(1-dR_a/dE_a),$$
$$R_b'\equiv R_b(E_b)/(1-dR_b/dE_b),$$
$$R_{ab}'=R_{ab}/(1-dR_a/dE_a).$$

The first apparent effect such a slight energy dependence then has is to alter the magnitude of the transition probability. The most interesting effect is the presence of additional contributions to the level shift as well as the natural linewidth of the state:

$$\begin{aligned}R_a'&\equiv R_a(E_a)/(1-dR_a/dE_a)\\&=(D_a-\tfrac{1}{2}i\Gamma_a)/(1-dD_a/dE_a+\tfrac{1}{2}id\Gamma_a/dE_a)\\&=D_a'-\tfrac{1}{2}i\Gamma_a',\end{aligned}$$

where

$$D_a'=\frac{[D_a(1-dD_a/dE_a)-\tfrac{1}{4}\Gamma_a d\Gamma_a/dE_a]}{(1-dD_a/dE_a)^2+\tfrac{1}{4}(d\Gamma_a/dE)^2}$$

and

$$\Gamma_a'=\frac{\Gamma_a(1-dD_a/dE_a)+D_a d\Gamma_a/dE_a}{(1-dD_a/dE_a)^2+\tfrac{1}{4}(d\Gamma_a/dE_a)^2},$$

with a similar expression for R_b. Provided that we know both the level shift as well as the linewidth as a function of the energy, the calculation of these corrections is straightforward.

As an illustration of the magnitude of these corrections, when applied to the $2S_{1/2}$ state of hydrogen the additional shift is

$$-D_a dD_a/dE_a\sim -D_a^2/E_a\sim\tfrac{1}{4}\text{ kc/sec},$$

a shift too small to be observable using present techniques.

3.2 Double Poles

An example of a decay mode that differs in character from those just treated is associated with the possibility of a double pole in the spectrum of the Green's function. Whenever this occurs the decay is no longer of the previous form but contains a term depending linearly on time modulated by the exponential decay. The presence of this type of decay may be interpreted as an isolated case included in the previous treatment or, as we prefer to do here, as a result of accidental degeneracy in the chosen model.

To illustrate this particular decay mode we consider a three-level system—two closely coupled excited states, $|a\rangle$ and $|b\rangle$, and a ground state. The decay mode to be observed depends upon the preparation of the initial state of the system which we choose here as the higher, $|a\rangle$. The decay of the higher state will deviate from an exponential behavior if

(1) transitions between the two excited states $|a\rangle$ and $|b\rangle$ are allowed;
(2) the values of the energies involved are suitably chosen.

To verify these statements we write the amplitude for the probability that at time t the system, originally in state $|a\rangle$, is still in that state:

$$I_{aa}(t)=\frac{1}{2\pi i}\times\int_c \frac{dE\, e^{-iEt}(E-E_b-R_b')(1-dR_a/dE_a)^{-1}}{(E-E_b-R_b')(E-E_a-R_a')-R_{ab}'R_{ba}'}, \tag{54}$$

where we have used the notation introduced in the previous section. Further, if we write

$$R_a'\equiv D_a'-\tfrac{1}{2}i\Gamma_a',\quad R_b'\equiv D_b'-\tfrac{1}{2}i\Gamma_b',$$

then the transition amplitude may be expressed as

$$I_{aa}(t)=\frac{1}{2\pi i}\int_c \frac{dE\, e^{-iEt}(E-\bar{E}_b+\tfrac{1}{2}i\Gamma_b)(1-dR_a/dE_a)^{-1}}{(E-E_+)(E-E_-)}, \tag{55}$$

where

$$E_\pm\equiv\tfrac{1}{2}\bar{E}_a+\bar{E}_b-\tfrac{1}{4}i(\Gamma_a'+\Gamma_b')\pm\tfrac{1}{2}\{[\bar{E}_a-\bar{E}_b+\tfrac{1}{2}i(\Gamma_b'-\Gamma_a')]^2+4R_{ab}'R_{ba}'\}^{1/2}. \tag{56}$$

Here we have absorbed the real shifts in the definition

$$\bar{E}_a\equiv E_a+D_a',\text{ etc.}$$

A double root in the integrand and hence a deviation from a purely exponential decay occurs for those values of the parameters of the problem that satisfy

$$\begin{aligned}(\bar{E}_a-\bar{E}_b)^2-\tfrac{1}{4}(\Gamma_a'-\Gamma_b')^2+\mathrm{Re}(4R_{ab}'R_{ba}')=0,\\ (\bar{E}_a-\bar{E}_b)(\Gamma_b'-\Gamma_a')+\mathrm{Im}(4R_{ab}'R_{ba}')=0.\end{aligned} \tag{57}$$

Whenever these conditions are satisfied, then the probability amplitude that the system is still in the state $|a\rangle$ at time t is given by

$$\begin{aligned}I_{aa}(t)\simeq&[1-it(E_0-\bar{E}_b+\tfrac{1}{2}i\Gamma_b')]\\ &\times(1-dR_a/dE_a)^{-1}\exp(-iE_0t),\end{aligned} \tag{58}$$

where

$$E_0=\tfrac{1}{2}(\bar{E}_a+\bar{E}_b)-\tfrac{1}{4}i(\Gamma_a'+\Gamma_b').$$

Thus

$$\begin{aligned}I_{aa}(t)\simeq&\{1-\tfrac{1}{2}it[\bar{E}_a-\bar{E}_b+\tfrac{1}{2}i(\Gamma_b'-\Gamma_a')]\}\\ &\times\exp[-\tfrac{1}{2}i(\bar{E}_a+\bar{E}_b)t]\exp[-\tfrac{1}{4}(\Gamma_a'+\Gamma_b')t].\end{aligned} \tag{59}$$

Provided $R_{ab}\neq 0$, the model chosen will exhibit deviations from a purely exponential decay. We note that this deviation occurs for small t and is of an essentially different character from the nonexponential decay valid for large t discussed earlier.

A model of this type has recently been discussed as a possible example of an elementary particle.[8] In the example the following parameters were chosen: $\Gamma_a'=D_a'=0$ and $R_{ab}=\alpha$. For α real, the second condition can be satisfied if a degeneracy exists:

$$\bar{E}_a=\bar{E}_b.$$

Under these conditions the probability that the system is still in the initial state $|a\rangle$ at time t is given by

$$P_{aa}(t)\sim(1+\tfrac{1}{4}\Gamma_b't)^2\exp(-\tfrac{1}{2}\Gamma_b't). \tag{60}$$

Within the context of the Schrödinger equation as usually formulated, then, nonexponential decays of the above type can be expected as a result of accidental degeneracies. Nonexponential decays can also be expected if the energy spectrum of the total Hamiltonian has intrinsic degeneracies.[9]

3.3 Transitions between Two Members of Degenerate States

As a perturbation on the first example treated, we consider the stimulated transition between two degenerate states. The physical situation giving rise to this case might be a fine-structure transition between two states of an atomic system in the absence of the complete resolution of hyperfine structure.

The projection operators for the two degenerate levels are given by

$$\begin{aligned}P_i&\equiv\sum_{\alpha=1}^{g_i}|i\alpha\rangle\langle i\alpha|,\\ P_j&=\sum_{\mu=1}^{g_j}|j\mu\rangle\langle\mu j|,\end{aligned} \tag{61}$$

where the states in question are eigenstates of K:

$$\begin{aligned}K|i\alpha\rangle&=E_i|i\alpha\rangle;\quad\alpha=1,\cdots g_i,\\ K|j\mu\rangle&=E_j|j\mu\rangle;\quad\mu=1,\cdots g_j.\end{aligned} \tag{62}$$

The two degenerate levels i and j may be assumed to be in relative isolation from the rest of the atomic spectrum even in the presence of a perturbation inducing transitions between the two levels. This may be realized by introducing a unitary transformation to remove all virtual transitions.[10] The result of this isolation procedure, however, is to modify the perturbation energy in the space spanned by the two levels in question. The major effect of the decoupling procedure is to introduce diagonal matrix elements of the perturbation energy that give rise to transition frequencies between the two levels that are different from the usual Bohr frequencies. With this in mind, we assume that the perturbation energy V has nonzero matrix elements in the space spanned by $|i\alpha\rangle$ and $|j\mu\rangle$ only.

The transition amplitude for an induced transition between the degenerate states $|i\alpha\rangle$ and $|j\mu\rangle$ depends on the matrix element of the Green's function between these states. We may write it as

$$\begin{aligned}&\langle\alpha i|G(E)|j\mu\rangle\\ &\quad\equiv\langle\alpha i|(E-Q_jHQ_j)^{-1}Q_jHP_j(P_jGP_j)|j\mu\rangle.\end{aligned} \tag{63}$$

With the above choice of states as eigenstates of K,

$$Q_jHP_j=Q_jVP_j.$$

To simplify this result further, we make use of the degree of freedom made available to us by the degeneracies of the two levels. We choose the degenerate sets $|i\alpha\rangle$ and $|j\mu\rangle$ to simplify the above matrix element. Thus, if we choose the degenerate sets to diagonalize the two Green's functions, we may achieve the desired simplification: i.e., we choose the set $|j\mu\rangle$ such that

$$\langle j\mu|R|j\mu'\rangle=R_{j,\mu\mu}\delta_{\mu\mu'}, \tag{64}$$

where

$$P_jRP_j=P_jVP_j+P_jVQ_j(E-Q_jHQ_j)^{-1}Q_jVP_j,$$

and the set $|i\alpha\rangle$ such that $(E-Q_jHQ_j)^{-1}$ is diagonal in $|i,\alpha\rangle$ space. The matrix elements of $(E-Q_jHQ_j)^{-1}$ may be calculated by repeating our original decomposition of the Green's function using Q_jHQ_j instead of H. We find

$$\begin{aligned}P_iG'P_i&\equiv P_i(E-Q_jHQ_j)^{-1}P_i\\ &=(E-P_iKP_i-P_iR'P_i)^{-1},\end{aligned} \tag{65}$$

[8] J. S. Bell and C. J. Goebel, Phys. Rev. **138**, B1198 (1965).
[9] M. L. Goldberger and K. M. Watson, Phys. Rev. **136**, B1472 (1964).
[10] J.-M. Winter, Ann. Phys. **4**, 745 (1959).

where

$$R' \equiv Q_j V Q_j + (Q_j V Q_j) Q_i [E - Q_i (Q_j H Q_j) Q_i]^{-1} Q_i (Q_j V Q_j).$$

By virtue of the assumed properties of V, the only nonzero matrix elements of R' are given by

$$\langle \alpha i | R' | i\alpha' \rangle = \langle \alpha i | V | i\alpha' \rangle. \tag{66}$$

Hence, $P_i G' P_i$ may be diagonalized by choosing the set $|i\alpha\rangle$ to diagonalize R':

$$\langle \alpha i | R' | i\alpha \rangle = \langle \alpha i | V | i\alpha \rangle \delta_{\alpha\alpha}' \equiv V_{i,\alpha\alpha} \delta_{\alpha\alpha}'. \tag{67}$$

With this choice in mind, we also see that the diagonal matrix element of R is given by

$$\langle j\mu | R | j\mu \rangle \equiv R_{j,\mu\mu} = V_{j,\mu\mu} + \sum_{\alpha,\alpha'} \langle j\mu | V | i\alpha \rangle \langle i\alpha | P_i G' P_i | i\alpha' \rangle \langle i\alpha' | V | j\mu \rangle$$
$$= V_{j,\mu\mu} + \sum_{\alpha} |\langle j\mu | V | i\alpha \rangle|^2 / (E - E_i - V_{i,\alpha\alpha}), \tag{68}$$

where we have written the matrix elements of $P_i G' P_i$ as

$$\langle i\alpha | P_i G' P_i | i\alpha' \rangle = (E - E_i - V_{i,\alpha\alpha})^{-1} \delta_{\alpha\alpha}'.$$

By collecting the above results, we see that the matrix element of the Green's function for the induced transition between the degenerate states $|i\alpha\rangle$ and $|j\mu\rangle$ is given by

$$\langle \alpha i | G(E) | j\mu \rangle = \sum_{\alpha',\mu'} \langle \alpha i | (E - Q_j H Q_j)^{-1} | i\alpha' \rangle \langle \alpha' i | V | j\mu' \rangle \langle j\mu' | (E - E_j - P_j R P_j)^{-1} | j\mu \rangle$$
$$= \frac{\langle \alpha i | V | j\mu \rangle}{(E - E_i - V_{i,\alpha\alpha})[E - E_j - V_{j,\mu\mu} - \sum_\beta |\langle j\mu | V | i\beta \rangle|^2 / (E - E_i - V_{i,\beta\beta})]}. \tag{69}$$

In general, the diagonal matrix elements of $V_{i,\alpha\alpha}$ will depend upon α. However, in the anticipation that the diagonal matrix elements will be small compared to the off-diagonal matrix elements we rewrite the above expression as

$$\langle \alpha i | G(E) | j\mu \rangle = \frac{\langle \alpha i | V | j\mu \rangle}{(E - E_{i,\alpha})(E - E_{j,\mu}) - \sum_\beta |\langle j\mu | V | i\beta \rangle|^2 - \Delta_{\alpha\mu}(E)}, \tag{70}$$

where

$$\Delta_{\alpha\mu} \equiv \sum_\beta \frac{(V_{i,\beta\beta} - V_{i,\alpha\alpha})}{E - E_{i,\beta}} |\langle j\mu | V | i\beta \rangle|^2,$$

and we have introduced the notation

$$E_{i,\alpha} \equiv E_i + V_{i,\alpha\alpha}.$$

In the absence of the term $\Delta_{\alpha\mu}(E)$, the contribution of the above matrix element to the transition amplitude occurs at $E = E_\pm$, where

$$E_\pm \equiv \tfrac{1}{2}(E_{i,\alpha} + E_{j,\mu}) \pm \tfrac{1}{2}[(E_{i,\alpha} - E_{j,\mu})^2 + 4 \textstyle\sum_\beta |\langle j\mu | V | i\beta \rangle|^2]^{1/2}. \tag{71}$$

With this value of $E_\pm$, we may estimate the magnitude of $\Delta_{\alpha\mu}$

$$\Delta_{\alpha\mu}(E_\pm) = 2 \sum_\beta \frac{(V_{i,\beta\beta} - V_{i,\alpha\alpha}) |\langle j\mu | V | i\beta \rangle|^2}{\{(E_{j,\mu} - E_{i,\alpha}) + 2(V_{i,\alpha\alpha} - V_{i,\beta\beta}) \pm [(E_{i,\alpha} - E_{j,\mu})^2 + 4 \textstyle\sum_\gamma |\langle j\mu | V | i\gamma \rangle|^2]^{1/2}\}}. \tag{72}$$

As a particular illustration of the theory we consider the transitions of an atomic system stimulated by the presence of an external electromagnetic field. If the language of second quantization is used to describe the external field and if an emission process is studied, then the energies of the initial and the final states are given by

$$E_j = W_j + (n+1)\hbar\omega, \quad E_i = W_i + n\hbar\omega,$$

where $W_{i,j}$ are the energy levels of the atomic system in the absence of the external rf field and $n'\hbar\omega$ is the energy of the electromagnetic field with n' photons of frequency ω present. Here, for purposes of this illustration we have introduced $\hbar$. If magnetic dipole transitions only are allowed, then the matrix element of interaction is given by[11]

$$\langle \alpha i | V | j\mu \rangle = \langle \alpha i | H_1 | j\mu \rangle$$
$$= -i(\hat{e}_\lambda \times \nabla G) \cdot \langle \alpha i | \mathbf{M}_m | j\mu \rangle \times [\hbar(n+1)/2\omega\epsilon_0 V]^{1/2}. \tag{73}$$

Here, $G(r)$ is the scalar amplitude of the mode of the external field in the cavity of volume V. It is so normalized that

$$\int_V |G(r)|^2 d\tau = V.$$

The factor $\langle \alpha i | \mathbf{M}_m | j\mu \rangle$ is the magnetic dipole moment

[11] M. Mizushima, Phys. Rev. 133, A414 (1964).

of the isolated atom for the two states of interest, $\hat{e}_\lambda$ denotes the direction of polarization of the rf magnetic field, and ϵ_0 is the permittivity of free space in mks units.

The unitary transformation required to isolate the initial and final states from the remainder of the atomic spectra introduces shifts of the following amounts (Refs. 10 and 11):

$$\begin{aligned}\Omega_m &\equiv V_{j,\mu\mu}-V_{i,\alpha\alpha}\\ &=\tfrac{1}{2}\mu_0 H^2\textstyle\sum_\gamma{}'|\langle j\mu|\mathbf{M}|\gamma\rangle|^2\\ &\quad\times[(W_j-W_\gamma-\hbar\omega)^{-1}+(W_j-W_\gamma+\hbar\omega)^{-1}]\\ &-\tfrac{1}{2}\mu_0 H^2\textstyle\sum_\gamma{}'|\langle\alpha i|\mathbf{M}|\gamma\rangle|^2\\ &\quad\times[(W_i-W_\gamma-\hbar\omega)^{-1}+(W_i-W_\gamma+\hbar\omega)^{-1}]. \quad (74)\end{aligned}$$

Here, the prime on the sum indicates that these energy corrections differ from the usual results of Rayleigh perturbation theory by the omission of both the initial and final states in both summations. In writing this result we have made the following identification:

$$\tfrac{1}{2}\mu_0 H^2=\sum_\lambda^2(nhc^2/2\omega V)|\hat{e}_\lambda\times\nabla G|^2, \quad n\gg 1 \quad (75)$$

where H and μ_0 are the magnetic-field strength and permeability in mks units, respectively.

In the absence of the correction $\Delta\mu\alpha$, the transition probability for the induced emission of a photon of frequency ω is given by

$$P_{ij}(t)=(4/g_j)\sum_{\alpha,\mu}[\ ]^{-1}|\langle\alpha i|V|j\mu\rangle|^2\sin^2\{[\ ]^{1/2}t/2\}, \quad (76)$$

where

$$[\ ]\equiv(E_{i,\alpha}-E_{j,\mu})^2+4\textstyle\sum_\beta|\langle j\mu|V|i\beta\rangle|^2,$$

and g_j is the degeneracy of the initial state. As a function of the external rf frequency the transition probability may be maximized by the choice

$$E_{i,\alpha}-E_{j,\mu}\equiv W_i-W_j-\hbar\omega-\Omega_m=0$$

or

$$\hbar\omega=W_i-W_j-\Omega_m.$$

For many precision measurements, $\Omega_m\ll|W_i-W_j|$. Under these conditions, the correction

$$[\Delta\alpha\mu/\textstyle\sum_\beta|\langle j\mu|V|i\beta\rangle|^2]$$

is at most of order $|\Omega_m|^{1/2}/|W_j-W_l|^{1/2}$, where W_l (with $l\neq i$ or j) is the energy level closest to either W_i or W_j included in the sum defining Ω_m. Hence, under these conditions, we may safely ignore $\Delta\mu\alpha$.

The frequency shifts in the magnetic dipole transitional between the hyperfine levels of $^2P_{3/2}$ states of Cs^{133} have been measured by Faist, *et al.*[12] and compared with a theory of the lineshape similar to that presented here. They calculated the average frequency shift

$$\bar{\Omega}\equiv\frac{\sum_{\alpha\mu}|\langle i\alpha|H_1|j\mu\rangle|^2(V_{i,\alpha\alpha}-V_{j,\mu\mu})}{\sum_{\alpha,\mu}|\langle i\alpha|H_1|j\mu\rangle|} \quad (77)$$

and found that the ratio $\bar{\Omega}/|W_i-W_j|$ was of the order $4\times10^{-3}H^2$ where H is the magnitude of the external rf magnetic field in oersteds. Hence, for fields in oersteds of strength $1<H<5$ we may safely ignore the correction $\Delta\mu\alpha$ and use the simple theory.

3.4 Rf Quenching of a Metastable Level

As a final example, we treat the problem of the radiative decay of a metastable level induced by a strong electromagnetic field. The stimulation processes will markedly change the decay rate if near the metastable level an unstable level exists that may be connected with the metastable level by a dipole transition. Thus, initially we assume that the atomic system is in a metastable state and, because of the interaction of the system with an rf field, the system makes a transition to an unstable level that quickly radiates to the ground state. If we then measure the number of emitted photons as a function of the frequency of the rf field, then a relative maximum number of photons will be counted if the frequency of the rf field just equals the Bohr frequency for the two excited levels in question.

Our energy spectrum as formulated consists of two excited states with different lifetimes plus a ground state. Actually, the transition in the presence of a strong rf field may be made either by absorbing or emitting a single photon; depending on the circumstances, one of these processes will be virtual while the other will be real. Hence, we should really consider the problem of an atomic system initially in one of several closely coupled excited states—in that one that has a relatively long lifetime. The rf field then induces a decay via either of the nearby unstable states. The states are assumed to be in relative isolation from the remaining states of the atomic system.

For simplicity, we divide the discussion into two parts. For intensities of the external rf field sufficiently low, we may ignore all transitions save the resonant absorption between the initial metastable state and the intermediate unstable state. Even for this case complexities occur. By using the language of quantized fields we must recognize that two exit channels exist for the strongly coupled states: one from the metastable state and one from the unstable state. As the number of rf photons differ in each of these two channels, the ground state themselves for these two decay processes differ.

As the intensity of the external rf field increases, additional excited states play an important role and additional decay channels become available. For simplicity, in this case we shall treat only the induced one-photon absorption or emission transition.

[12] A. Faist, E. Geneux, and S. Koide, J. Phys. Soc. Japan **19**, 2299 (1964).

(a) *Moderately Intense External Fields*

The states and the associated energies of the unperturbed system are given by

$$\begin{aligned} K|W_a;n\rangle&=(W_a+n\hbar\omega)|W_a;n\rangle\equiv E_{a,n}|W_a;n\rangle,\\ K|W_b;n-1\rangle&=[W_b+(n-1)\hbar\omega]|W_b;n-1\rangle\\ &\equiv E_{b,n-1}|W_b;n-1\rangle, \quad (78)\\ K|W_0;n';\mathbf{k}_\sigma\rangle&=(W_0+n'\hbar\omega+\hbar\omega_\sigma)|W_0;n';\mathbf{k}_\sigma\rangle\\ &\equiv E_{0,n'}|W_0;n';\mathbf{k}_\sigma\rangle, \end{aligned}$$

where K denotes that part of the Hamiltonian that describes the system in the absence of the atom-rf field interaction. Here n denotes the number of rf photons present, while $\mathbf{k}_\sigma$ denotes the wave number of the photon emitted in the decay process. The presence of the interaction between the external rf field and the atomic system strongly couples the two states $|W_a;n\rangle$ and $|W_b;n-1\rangle$, which we represent as $|a\rangle$ and $|b\rangle$, respectively. The matrix element of the interaction energy between these states is given by

$$\begin{aligned}\langle W_a;n|V|W_b;n-1\rangle&\\ =\langle W_a;n|H_1|W_b;n-1\rangle&\equiv H_{ab}. \quad (79)\end{aligned}$$

The transition amplitude for the decay of the state $|a\rangle$ in the presence of the external rf field is

$$\begin{aligned} I_{0a}(t)=&\frac{1}{2\pi i}\int_c e^{-iEt}\langle W_0;n;\mathbf{k}_\sigma|G(E)|a\rangle dE\\ &+\frac{1}{2\pi i}\int_c e^{-iEt}\langle W_0;n-1;\mathbf{k}_\lambda|G(E)|a\rangle dE, \quad (80)\end{aligned}$$

where the two terms represent the two possible modes of decay: one, directly to ground, and the second, indirectly to ground via state $|b\rangle$. (In this statement and in what follows we again set $\hbar=1$.) We may re-express the above in terms of matrix elements of the level shift operator as

$$\begin{aligned} I_{0a}(t)=&\frac{1}{2\pi i}\int_c e^{-iEt}(E-E_{0,n})^{-1}\langle W_0;n;\mathbf{k}_\sigma|R|a\rangle g_{aa}dE\\ &+\frac{1}{2\pi i}\int_c e^{-iEt}(E-E_{0,n-1})^{-1}\\ &\times\langle W_0;n-1;\mathbf{k}_\lambda|R|b\rangle g_{ba}dE, \quad (81)\end{aligned}$$

where the matrix elements of the Green's function are given by (22) if we replace E_a by $E_{a,n}$ and E_b by $E_{b,n-1}$. The matrix elements of the level shift operator are given by (32). The remaining matrix elements of R between the ground states and the excited states represent the coupling to the radiation field giving rise to the decay and will be retained as is in the subsequent calculation.

Hence, the transition amplitude for the stimulated decay of an atomic system initially in one of two closely coupled states is given by

$$\begin{aligned} I_{0a}(t)=&\frac{1}{2\pi i}\int_c\frac{dE\langle W_0;n;\mathbf{k}_\sigma|R|a\rangle(E-\bar{E}_{b,n-1}+\frac{1}{2}i\Gamma_b)e^{-iEt}}{(E-E_{0,n})(E-E_+)(E-E_-)}\\ &-\frac{1}{2\pi i}\int_c\frac{dE\langle W_0;n-1;\mathbf{k}_\lambda|R|a\rangle H_{ba}e^{-iEt}}{(E-E_{0,n-1})(E-E_+)(E-E_-)}, \quad (82)\end{aligned}$$

where

$$\begin{aligned} E_\pm\equiv&\tfrac{1}{2}(\bar{E}_{a,n}+\bar{E}_{b,n-1})-\tfrac{1}{4}i(\Gamma_a+\Gamma_b)\\ &\pm\tfrac{1}{2}[(\bar{E}_{a,n}-\bar{E}_{b,n-1}+\tfrac{1}{2}i\Gamma_b-\tfrac{1}{2}i\Gamma_a)^2+4|H_{ab}|^2]^{1/2} \quad (83)\end{aligned}$$

and we have absorbed the real part of the diagonal matrix of R in the definition

$$E_{a,n}\equiv\bar{E}_{a,n}+D_a.$$

We assume that both the matrix elements

$$\langle W_0;n;\mathbf{k}_\sigma|R|a\rangle, \quad \langle W_0;n-1;\mathbf{k}_\lambda|R|b\rangle$$

are independent of the total energy E such that we may evaluate them at $E=E_{0,n}$ and $E=E_{0,n-1}$, respectively. (The variation with energy may be included in a more precise calculation.) With this in mind, then, the transition amplitude for times such that $\Gamma_a t$, $\Gamma_b t$ are not too large and for a system in which

$$\Gamma_a/E_{a,n}\ll 1; \quad \Gamma_b/E_{b,n-1}\ll 1,$$

is given by

$$\begin{aligned} I_{0a}(t)\simeq&\langle W_0;n;\mathbf{k}_\sigma|R|a\rangle\left\{\frac{(E_{0,n}-\bar{E}_{b,n-1}+\frac{1}{2}i\Gamma_b)e^{-iE_{0,n}t}}{(E_{0,n}-E_+)(E_{0,n}-E_-)}+\frac{(E_+-\bar{E}_{b,n-1}+\frac{1}{2}i\Gamma_b)e^{-iE_+t}}{(E_+-E_{0,n})(E_+-E_-)}-\frac{(E_--\bar{E}_{b,n-1}+\frac{1}{2}i\Gamma_b)e^{-iE_-t}}{(E_--E_{0,n})(E_+-E_-)}\right\}\\ &-\langle W_0;n-1;\mathbf{k}_\lambda|R|b\rangle H_{ba}\left\{\frac{e^{-iE_{0,n-1}t}}{(E_{0,n-1}-E_+)(E_{0,n-1}-E_-)}+\frac{e^{-iE_+t}}{(E_+-E_{0,n-1})(E_+-E_-)}-\frac{e^{-iE_-t}}{(E_--E_{0,n-1})(E_+-E_-)}\right\}. \quad (84)\end{aligned}$$

For $\Gamma_a t$, $\Gamma_b t\gg 0$, only the first term in each bracket is appreciable. Hence, if we restrict ourselves to the nonvanishing contribution for large values of t, then the transition probability per unit energy range of the emitted

photon in the induced decay of the metastable state $|a\rangle$ is given by

$$P_{0a}\underset{\Gamma_b t\gg 1}{\rightarrow}\left(\frac{1}{2\pi}\right)\frac{\Gamma_a[(E_{0,n}-\bar{E}_{b,n-1})^2+\frac{1}{4}\Gamma_b^2]}{[(E_{0,n}-\bar{E}_{a,n})(E_{0,n}-\bar{E}_{b,n-1})-|H_{ab}|^2-\frac{1}{4}\Gamma_a\Gamma_b]^2+\frac{1}{4}[\Gamma_a(E_{0,n}-\bar{E}_{b,n-1})+\Gamma_b(E_{0,n}-\bar{E}_{a,n}]^2}$$
$$+\left(\frac{1}{2\pi}\right)\frac{\Gamma_b|H_{ab}|^2}{[(E_{0,n-1}-\bar{E}_{a,n})(E_{0,n-1}-\bar{E}_{b,n-1})-|H_{ab}|^2-\frac{1}{4}\Gamma_a\Gamma_b]^2+\frac{1}{4}[\Gamma_a(E_{0,n-1}-\bar{E}_{b,n-1})+\Gamma_b(E_{0,n-1}-\bar{E}_{a,n})]^2}. \quad (85)$$

In writing this expression we have noted that the two decay photons are independent, and hence, the two decay channels are independent. In doing so we have utilized the relationship[13]

$$2\pi\sum_{\ell_\lambda}\int d\rho_f\langle a|R|W_0;n;\mathbf{k}_\sigma\rangle\langle W_0;n';\mathbf{k}_\lambda|R|b\rangle = \Gamma_a\delta_{ab}\delta_{nn'}\delta_{\mathbf{k}_\sigma\mathbf{k}\lambda'}. \quad (86)$$

The first term in the above expression for the stimulated transition probability describes the modification of the direct decay of the state $|a\rangle$ by the presence of the external rf field. The second term describes the indirect decay of the state $|a\rangle$ available by the presence of the interaction energy $H_{ab}\equiv H_1$. Similar expressions have been given by Hack and Hamermesh.[14] To see this more clearly, we introduce the following frequencies in terms of the energy differences involved in the above expression:

$$\begin{aligned}E_{0,n}-\bar{E}_{a,n}&\equiv\omega_\sigma-\omega_0',\\ E_{0,n}-\bar{E}_{b,n-1}&\equiv\omega_\sigma-\omega_0'-\omega_{ba}+\omega,\\ E_{0,n-1}-\bar{E}_{b,n-1}&\equiv\omega_\lambda-\omega_0,\\ E_{0,n-1}-\bar{E}_{a,n}&\equiv\omega_\lambda-\omega_0+\omega_{ba}-\omega.\end{aligned} \quad (87)$$

In the absence of the interaction, the first term in the expression for the transition probability, which we refer to as $P_{0a,D}$, reduces to

$$P_{0a,D}=\frac{\Gamma_a/2\pi}{(\omega_\sigma-\omega_0')^2+\frac{1}{4}\Gamma_a^2}, \quad (88)$$

a Lorentz-shape line with a width Γ_a. By assumption $\Gamma_a\ll\Gamma_b$; hence, we may ignore this contribution for $|H_1|\gg\Gamma_a$. (The presence of a strong interaction $|H_1|>\Gamma_b$ serves only to shift the resonance and broaden the line somewhat; in addition, it substantially decreases the probability at resonance by the ratio Γ_a^2/Γ_b^2.)

The major contribution to the induced transition probability for $\Gamma_a\ll\Gamma_b$ is given by the second term in the above expression, which we refer to as $P_{0a,I}$, where

$$P_{0a,I}=\frac{\Gamma_b|H_1|^2/2\pi}{[(\omega_\lambda-\omega_0)(\omega_\lambda-\omega_0+\omega_{ba}-\omega)-|H_1|^2-\frac{1}{4}\Gamma_a\Gamma_b]^2+\frac{1}{4}[\Gamma_a(\omega_\lambda-\omega_0)+\Gamma_b(\omega_\lambda-\omega_0+\omega_{ba}-\omega)]^2}. \quad (89)$$

At rf resonance, $\omega=\omega_{ba}$, this expression simplifies to

$$P_{0a,I\,\mathrm{res}}=\frac{\Gamma_b|H_1|^2/2\pi}{[(\omega_\lambda-\omega_0)^2-|H_1|^2-\frac{1}{4}\Gamma_a\Gamma_b]^2+\frac{1}{4}(\omega_\lambda-\omega_0)^2(\Gamma_a+\Gamma_b)^2}, \quad (90)$$

which, as a function of the frequency ω_λ of the emitted photon, has extrema at

$$\omega_\lambda=\omega_0,\quad (\omega_\lambda-\omega_0)^2=|H_1|^2-(\Gamma_a^2+\Gamma_b^2)/8. \quad (91)$$

For intensities of the external field such that the matrix element coupling the states $|a\rangle$ and $|b\rangle$ is less than the decay rate Γ_b, then a single line exists and is located at the natural line center. The maximum transition probability for this case is given by

$$P_{0a\,\max}\simeq\Gamma_b/2\pi|H_1|^2;\quad \Gamma_a\ll|H_1|<\Gamma_b \quad (92)$$

with a half-width given by

$$\Delta\sim 4|H_1|^2/\Gamma_b,$$

which is narrower than the natural line. For somewhat larger intensities, $|H_1|>\Gamma_b$, the line splits and has a relative minimum at $\omega_\lambda=\omega_0$. The maxima are located at

$$\omega_\lambda=\omega_0\pm|H_1|(1-\Gamma_b^2/8|H_1|^2)^{1/2}, \quad (93)$$

with a probability

$$P_{0a\,\max}\simeq(2/\pi\Gamma_b)(1-\Gamma_b^2/16|H_1|^2)^{-1}, \quad (94)$$

somewhat larger than the probability for the natural line. The width of this line is

$$\Delta\sim\Gamma_b/\sqrt{2},$$

which is comparable to the natural line.

[13] M. E. Rose and R. L. Carovillano, Phys. Rev. **122**, 1185 (1961).
[14] M. N. Hack and M. Hamermesh, Nuovo Cimento **19**, 546 (1961).

For values of the time t that do not satisfy the inequality $(\Gamma_a+\Gamma_b)t\gg 0$, we must treat the time dependence of the transition probability directly. In the previous case we examined the transition probability at rf resonance ($\omega=\omega_{ba}$) as a function of the frequency of the decay photon. In general, it is easier experimentally to count the emitted photons instead of scanning them with a monochromatic filter. Hence, we should deal with the rf frequency dependence of the total transition probability found from the transition probability per unit energy range of the emitted photon by summing over the frequency range of the emitted photon. As there are two different types of photons emitted, one for each decay channel, this must be taken into account. Thus we have

$$P_{\text{total}}(t)=\int_0^\infty d\omega_\sigma P_{0a,D}(t)+\int_0^\infty d\omega_\lambda P_{0a,I}(t), \quad (95)$$

where

$$P_{0a,D}(t)=\frac{\Gamma_a}{2\pi}\left|\frac{(E_{0,n}-\bar{E}_{b,n-1}+\frac{1}{2}i\Gamma_b)}{(E_{0,n}-E_+)(E_{0,n}-E_-)}e^{-iE_{0,n}t}+\frac{(E_+-\bar{E}_{b,n-1}+\frac{1}{2}i\Gamma_b)}{(E_+-E_{0,n})(E_+-E_-)}e^{-iE+t}-\frac{(E_--\bar{E}_{b,n-1}+\frac{1}{2}i\Gamma_b)}{(E_--E_{0,n})(E_+-E_-)}e^{-iE-t}\right|^2, \quad (96)$$

$$P_{0a,I}(t)=\frac{\Gamma_b}{2\pi}|H_{1ab}|^2\left|\frac{e^{-iE_{0,n-1}t}}{(E_{0,n-1}-E_+)(E_{0,n-1}-E_-)}+\frac{e^{-iE+t}}{(E_+-E_{0,n-1})(E_+-E_-)}-\frac{e^{-iE-t}}{(E_--E_{0,n-1})(E_+-E_-)}\right|^2. \quad (97)$$

In writing these transition probabilities we have made use of Eq. (84). We may replace ω_σ to $E_{0,n}$ and ω_λ to $E_{0,n-1}$ by noting that for $\hbar=1$,

$$d\omega_\sigma=dE_{0,n}, \quad d\omega_\lambda=dE_{0,n-1}.$$

Finally, the lower limits on the integrals may be extended to $-\infty$ by noting that the antiresonances so included contribute very little to the integral. The integrals may then be evaluated by contour integration. To simplify the resulting expressions, we introduce the following notation:

$$E_+=(a+b)-i(c-d), \quad E_-=(a-b)-i(c+d), \quad (98)$$

where

$$a=\tfrac{1}{2}(\bar{E}_{a,n}+\bar{E}_{b,n-1}), \quad b=\tfrac{1}{4}\sqrt{2}[(A^2+B^2)^{1/2}+A]^{1/2}, \quad c=\tfrac{1}{4}(\Gamma_a+\Gamma_b), \quad d=\tfrac{1}{4}\sqrt{2}[(A^2+B^2)^{1/2}-A]^{1/2}\,\text{sgn}B,$$

and

$$A\equiv(\bar{E}_{a,n}-\bar{E}_{b,n-1})^2+4|H_{1ab}|^2-\tfrac{1}{4}(\Gamma_a-\Gamma_b)^2, \quad B\equiv(\bar{E}_{a,n}-\bar{E}_{b,n-1})(\Gamma_b-\Gamma_a).$$

In terms of this notation, we find

$$\begin{aligned}\int_{-\infty}^{\infty}P_{0a,D}(t)dE_{0,n}=&\frac{\Gamma_a\{c[(a-\bar{E}_{b,n-1})^2+\Gamma_b^2/4+b^2+c^2-d^2]+2bd(a-\bar{E}_{b,n-1})\}}{4(b^2+c^2)(c^2-d^2)}\\&-\Gamma_a\frac{\{c[(a-\bar{E}_{b,n-1})^2+(c-\Gamma_b/2)^2+b^2+d^2]+2d[b(a-\bar{E}_{b,n-1})-d(c-\Gamma_b/2)]\}}{4(b^2+d^2)(c^2-d^2)}e^{-2ct}\cosh 2dt\\&-\Gamma_a\frac{\{d[(a-\bar{E}_{b,n-1})^2+(c-\Gamma_b/2)^2+b^2+d^2]+2c[b(a-\bar{E}_{b,n-1})-d(c-\Gamma_b/2)]\}}{4(b^2+d^2)(c^2-d^2)}e^{-2ct}\sinh 2dt\\&+\Gamma_a\frac{\{2b[b(c-\Gamma_b/2)+d(a-\bar{E}_{b,n-1})]+c[(a-\bar{E}_{b,n-1})^2+(c-\Gamma_b/2)^2-b^2-d^2]\}}{4(b^2+d^2)(b^2+c^2)}e^{-2ct}\cos 2bt\\&+\Gamma_a\frac{\{2c[b(c-\Gamma_b/2)+d(a-\bar{E}_{b,n-1})]-b[(a-\bar{E}_{b,n-1})^2+(c-\Gamma_b/2)^2-b^2-d^2]\}}{4(b^2+d^2)(b^2+c^2)}e^{-2ct}\sin 2bt, \quad (99)\end{aligned}$$

and

$$\begin{aligned}\int_{-\infty}^{\infty}P_{0a,I}(t)dE_{0,n-1}=&\frac{\Gamma_b|H_1|^2c}{4(b^2+c^2)(c^2-d^2)}-\frac{\Gamma_b|H_1|^2e^{-2ct}}{4(b^2+d^2)(c^2-d^2)}[c\cosh 2dt+d\sinh 2dt]\\&+\frac{\Gamma_b|H_1|^2e^{-2ct}}{4(b^2+d^2)(b^2+c^2)}[c\cos 2bt-b\sin 2bt]. \quad (100)\end{aligned}$$

In terms of consistency we note that each of the two contributions to the total transition probability vanish

at $t=0$:

$$\int_{-\infty}^{\infty} P_{0a,D}(t=0)dE_{0,n}=0, \quad \int_{-\infty}^{\infty} P_{0a,I}(t=0)dE_{0,n-1}=0, \tag{101}$$

hence, the total transition probability vanishes at $t=0$. Furthermore, if we introduce the definitions for a, b, c, and d, we see that the time-independent contributions to the total transition probability are given by

$$\frac{\Gamma_b|H_1|^2c}{4(b^2+c^2)(c^2-d^2)}=\frac{4\Gamma_b|H_1|^2(\Gamma_a+\Gamma_b)}{(\Gamma_a+\Gamma_b)^2(\Gamma_a\Gamma_b+4|H_1|^2)+4\Gamma_a\Gamma_b(\bar{E}_{a,n}-\bar{E}_{b,n-1})^2}$$

and

$$\frac{\Gamma_a\{c[(a-\bar{E}_{b,n-1})^2+\Gamma_b^2/4+b^2+c^2-d^2]+2bd(a-\bar{E}_{b,n-1})\}}{4(b^2+c^2)(c^2-d^2)}$$
$$=\frac{4\Gamma_a\Gamma_b(\bar{E}_{a,n}-\bar{E}_{b,n-1})^2+(\Gamma_a+\Gamma_b)[\Gamma_a\Gamma_b(\Gamma_a+\Gamma_b)+4\Gamma_a|H_1|^2]}{(\Gamma_a+\Gamma_b)^2(\Gamma_a\Gamma_b+4|H_1|^2)+4\Gamma_a\Gamma_b(\bar{E}_{a,n}-\bar{E}_{b,n-1})^2},$$

which sum to unity. Hence, in the limit of large time the total transition probability approaches unity. That is, if one waits long enough the system, originally in state $|a\rangle$, will eventually be found in the ground state with a decay photon.

Of some interest from a practical point of view is the decay rate. Using the above expressions for the total transition probability we find that the decay rate is

$$\frac{dP_T(t)}{dt}\equiv\frac{d}{dt}\int P_{0a,I}d\omega_\lambda+\frac{d}{dt}\int P_{0a,D}d\omega_\sigma=\frac{\Gamma_b|H_1|^2e^{-2ct}}{2(b^2+d^2)}[\cosh 2dt-\cos 2bt]$$
$$+\frac{\Gamma_a e^{-2ct}}{2(b^2+d^2)}\{[(a-\bar{E}_{b,n-1})^2+(c-\Gamma_b/2)^2+b^2+d^2]\cosh 2dt+[2b(a-\bar{E}_{b,n-1})-2d(c-\Gamma_b/2)]\sinh 2dt$$
$$-[(a-\bar{E}_{b,n-1})^2+(c-\Gamma_b/2)^2-b^2-d^2]\cos 2bt-[2b(c-\Gamma_b/2)+2d(a-\bar{E}_{b,n-1})]\sin 2bt\}. \tag{102}$$

The initial decay rate is given simply by

$$(dP_T(t)/dt)\,|_{t=0}=\Gamma_a; \tag{103}$$

the contribution from the induced decay of state $|a\rangle$ via state $|b\rangle$ varies as t^3 for small t. The decay rate via state $|b\rangle$ for $t\neq 0$,

$$\frac{d}{dt}\int P_{0a,I}d\omega_\lambda=\frac{\Gamma_b|H_1|^2e^{-2ct}}{2(b^2+d^2)}[\cosh 2dt-\cos 2bt], \tag{104}$$

has a simple interpretation. It is simply the rate of decay of state b to ground, Γ_b, times the probability that after a time t the system has made the transition from $|a\rangle$ to $|b\rangle$. For $\Gamma_b\gg\Gamma_a$ this is the major contribution to the decay rate of the system, hence we take a brief look at this rate. If we introduce the definitions of b and d, we find

$$2(b^2+d^2)=\tfrac{1}{2}(A^2+B^2)^{1/2}$$
$$=\tfrac{1}{2}[(\bar{E}_{a,n}-E_{b,n-1})^2+\tfrac{1}{4}(\Gamma_a-\Gamma_b)^2$$
$$+4|H_1|^2]^2-2|H_1|^2(\Gamma_a-\Gamma_b)^2$$

which becomes small at rf resonance,

$$\bar{E}_{a,n}-\bar{E}_{b,n-1}=\omega-\omega_{ba}=0.$$

Under this condition, $d=0$, and the above decay rate becomes

$$\frac{d}{dt}\int P_{0a,I}d\omega_\lambda\underset{\omega=\omega_{ba}}{=}\frac{\Gamma_b|H_1|^2e^{-2ct}}{b^2}\sin^2 bt, \tag{105}$$

where

$$b=\tfrac{1}{2}[4|H_1|^2-\tfrac{1}{4}(\Gamma_a-\Gamma_b)^2]^{1/2},$$
$$2c=\tfrac{1}{2}(\Gamma_a+\Gamma_b).$$

(b) *Strong rf Fields*

At higher intensities of the external inducing field it is possible to cause a transition from the metastable state via stimulated emission as well as by absorption. Not only does the stimulated emission process couple more states, but it also introduces new decay channels. Virtual processes exist that couple not only with the initial state but with the final state as well. To illustrate this coupling process, we redefine the states as

$$|a_n'\rangle\equiv|W_a;n'\rangle, \quad |b_n'\rangle\equiv W_b;n'\rangle.$$

In terms of this notation, we assume as initial conditions that the state of the system is represented by

$$\psi|t=0\rangle=|a_n\rangle,$$

that is, initially the system is in the metastable state $|a\rangle$

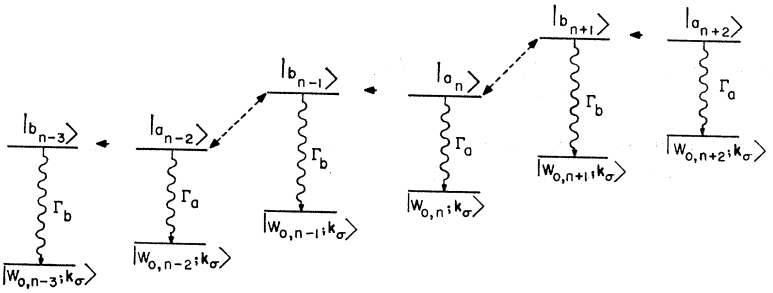

FIG. 2. Energy-level diagram for the relevant states involved in the rf-induced decay of a metastable state. Absorption of real rf photons is denoted by horizontal arrows. Virtual absorption or emission of a rf photon is denoted by a diagonal arrow. Decay photons are denoted by vertical arrows.

with n rf photons of frequency ω. The specific states closely coupled by the rf interaction are the following:

$$|a_{n-2}\rangle,\ |b_{n-3}\rangle;\quad |a_n\rangle,\ |b_{n-1}\rangle;\quad |a_{n+2}\rangle,\ |b_{n+1}\rangle.$$

This coupling is schematized in Fig. 2.

The presence of the additional states as well as the additional exit channels requires a modification of the initial formulation. For simplicity, we limit oursevles to the induced process and ignore the decay through the direct exit channels. With this in mind, a study of the above energy-level diagram leads to the following expression for the transition amplitude of the rf-induced decay of the metastable state $|a\rangle$:

$$I_{0a}(t)=\frac{1}{2\pi i}\int_c dE\, e^{-iEt}\sum_{n'}(E-E_{0,n'})^{-1}\times\langle W_{0,n'}\mathbf{k}_\sigma|R_r|b_{n'}\rangle\langle b_{n'}|g|a_n\rangle, \quad (106)$$

where the sum over n' includes the three decay channels $|b_{n-3}\rangle$, $|b_{n-1}\rangle$, and $|b_{n+1}\rangle$.

The level-shift operator in the space spanned by the coupled states is unchanged in form from that given before. The projection operator Q, however, is given by

$$Q\equiv 1-P,$$
$$P=\sum_{n'}|a_{n'}\rangle\langle a_{n'}|+\sum_{n'}|b_{n'}\rangle\langle b_{n'}|,$$

where the sum over n' includes just the coupled states.

In the space spanned by the coupled states the nonzero matrix elements of the level-shift operator are given by

$$\langle b_{n-1}|R|a_n\rangle=\langle b_{n-1}|H_1|a_n\rangle\equiv H_{-,n},$$
$$\langle b_{n+1}|R|a_n\rangle=\langle b_{n+1}|H_1|a_n\rangle\equiv H_{+,n},$$
$$\langle b_{n-1}|R|a_{n-2}\rangle=\langle b_{n-1}|H_1|a_{n-2}\rangle\equiv H_{+,n-2},$$
$$\langle b_{n-3}|R|a_{n-2}\rangle=\langle b_{n-3}|H_1|a_{n-2}\rangle\equiv H_{-,n-2},$$
$$\langle b_{n+1}|R|a_{n+2}\rangle=\langle b_{n+1}|H_1|a_{n+2}\rangle\equiv H_{-,n+2}.$$

Here, only one-photon processes have been included. The subscript $(-)$ refers to an absorption process, while the $(+)$ refers to a stimulated emission process. In the limit of the number of photons n being large compared to one, all these matrix elements have the same magnitude. The remaining nonzero matrix elements of R take a familiar form for $\mathrm{Im}E<0$ on the second Riemann sheet.

$$\langle a_{n'}|R|a_{n'}\rangle\equiv R_{a_{n'},a_{n'}}=D_a-i\Gamma_a/2,$$
$$\langle b_{n'}|R|b_{n'}\rangle\equiv R_{b_{n'},b_{n'}}=D_b-i\Gamma_b/2,$$

where we have assumed that the individual matrix elements were insensitive to the differences between the number of rf photons in the states considered.

From the form of these matrix elements we see that on the second Riemann sheet for $\mathrm{Im}E<0$ the array for the matrix elements of g^{-1}, the inverse Green's function, in the space spanned by the closely coupled states is given by

	b_{n-3}	a_{n-2}	b_{n-1}	a_n	b_{n+1}	a_{n+2}
b_{n-3}	$E-\bar{E}_{b,n-3}+i\Gamma_b/2$	$-H_{-,n-2}$	0	0	0	0
a_{n-2}	$-H_{-,n-2}^*$	$E-\bar{E}_{a,n-2}+i\Gamma_b/2$	$-H_{+,n-2}^*$	0	0	0
b_{n-1}	0	$-H_{+,n-2}$	$E-\bar{E}_{b,n-1}+i\Gamma_b/2$	$-H_{-,n}$	0	0
a_n	0	0	$-H_{-,n}^*$	$E-\bar{E}_{a,n}+i\Gamma_a/2$	$-H_{+,n}^*$	0
b_{n+1}	0	0	0	$-H_{+,n}$	$E-\bar{E}_{b,n+1}+i\Gamma_b/2$	$-H_{-,n+2}$
a_{n+2}	0	0	0	0	$-H_{-,n+2}^*$	$E-\bar{E}_{a,n+2}+i\Gamma_a/2$

The states of this array consist of pairs coupled by an absorption process that are in turn coupled to one another via stimulated emission processes. In writing this array we have absorbed the level shift of each state through the definition $\bar{E}_i=D_i+E_i$.

The matrix elements of the Green's functions that appear in the expression for the transition amplitude may be calculated from the above array by means of the relation

$$\langle b_{n'}|g|a_n\rangle=(\text{cofactor } g^{-1})_{an,bn'}/\mathrm{Det},$$

where Det is the determinant formed from the above array. We find the following cofactors of interest in this

calculation.

$$(\text{cofactor})_{an,bn-3}=-(H_{-,n-2})(H_{+,n-2}{}^*)(H_{-,n})[(E-\bar{E}_{b,n+1}+i\Gamma_b/2)(E-\bar{E}_{a,n+2}+i\Gamma_a/2)-|H_{-,n+2}|^2],$$

$$(\text{cofactor})_{an,bn-1}=-(H_{-,n})[(E-\bar{E}_{b,n-3}+i\Gamma_b/2)(E-\bar{E}_{a,n-2}+i\Gamma_a/2)-|H_{-,n-2}|^2]\times[(E-\bar{E}_{b,n+1}+i\Gamma_b/2)(E-\bar{E}_{a,n+2}+i\Gamma_a/2)-|H_{-,n+2}|^2],$$

$$(\text{cofactor})_{an,bn+1}=(H_{+,n})|H_{+,n-2}|^2(E-\bar{E}_{b,n-3}+i\Gamma_b/2)(E-\bar{E}_{a,n+2}+i\Gamma_a/2)-(H_{+,n})(E-\bar{E}_{a,n+2}+i\Gamma_a/2)\times(E-\bar{E}_{b,n-1}+i\Gamma_b/2)[(E-\bar{E}_{b,n-3}+i\Gamma_b/2)(E-\bar{E}_{a,n-2}+i\Gamma_a/2)-|H_{-,n-2}|^2],$$

$$(\text{cofactor})_{an,an}=-|H_{+,n-2}|^2(E-\bar{E}_{b,n-3}+i\Gamma_b/2)[(E-\bar{E}_{b,n+1}+i\Gamma_b/2)(E-\bar{E}_{a,n+2}+i\Gamma_a/2)-|H_{-,n+2}|^2]+(E-\bar{E}_{b,n-1}+i\Gamma_b/2)[(E-\bar{E}_{b,n-3}+i\Gamma_b/2)(E-\bar{E}_{a,n-2}+i\Gamma_a/2)-|H_{-,n-2}|^2]\times[(E-\bar{E}_{b,n+1}+i\Gamma_b/2)(E-\bar{E}_{a,n+2}+i\Gamma_a/2)-|H_{-,n+2}|^2].$$

The determinant of the above array for the elements of g^{-1} may be calculated using the above cofactors. If we expand the determinant in terms of the a_n row, we find

$$\text{Det}(E)=+H_{-,n}{}^*(\text{cofactor})_{an,bn-1}+(E-\bar{E}_{an}+i\Gamma_a/2)(\text{cofactor})_{an,an}+H_{+,n}{}^*(\text{cofactor})_{an,bn+1}.$$

After a certain amount of algebra to render the fat from this expression, we find

$$\text{Det}=[n-2][n][n+2]-|H_{+,n-2}|^2(E-\bar{E}_{an}+i\Gamma_a/2)(E-\bar{E}_{b,n-3}+i\Gamma_b/2)[n+2]-|H_{+,n}|^2(E-\bar{E}_{a,n+2}+i\Gamma_a/2)(E-\bar{E}_{b,n-1}+i\Gamma_b/2)[n-2]+|H_{+,n}|^2|H_{+,n-2}|^2(E-\bar{E}_{a,n+2}+i\Gamma_a/2)(E-\bar{E}_{b,n-3}+i\Gamma_b/2),$$

where we have introduced the abbreviation

$$[n']\equiv[(E-\bar{E}_{b,n'-1}+i\Gamma_b/2)(E-\bar{E}_{a,n'}+i\Gamma_a/2)-|H_{-,n'}|^2].$$

To proceed further we limit our attention to that component of the transition amplitude that is not damped in time. Hence, if we make use of the relationship:

$$2\pi\sum_{\mathbf{k}_\lambda}\int d\rho_f\langle b_n|R_r|W_{0,n};\mathbf{k}_\sigma\rangle\langle W_{0,n'};\mathbf{k}_\lambda|R_r|b_{n'}\rangle=\Gamma_b\delta_{nn'}\delta_{\mathbf{k}_\sigma,\mathbf{k}_\lambda},\tag{107}$$

then we find that the probability for the rf-induced transition from a metastable state is given by

$$P_{0a}=\frac{\Gamma_b}{2\pi}\{|\langle b_{n-1}|g(E=E_{0,n-1})|a_n\rangle|^2+|\langle b_{n-3}|g(E=E_{0,n-3})|a_n\rangle|^2+|\langle b_{n+1}|g(E=E_{0,n+1})|a_n\rangle|^2\}$$

$$=\frac{\Gamma_b}{2\pi}\left\{\left|\frac{(\text{cofactor})_{an,bn-1}}{\text{Det}(E)}\right|^2_{E=E_{0,n-1}}+\left|\frac{(\text{cofactor})_{an,bn-3}}{\text{Det}(E)}\right|^2_{E=E_{0,n-3}}+\left|\frac{(\text{cofactor})_{an,bn+1}}{\text{Det}(E)}\right|^2_{E=E_{0,n+1}}\right\}.\tag{108}$$

An examination of the cofactors involved suffices to show the asymmetry of the decay of the metastable state $|a\rangle$.

To simplify these expressions, we introduce the following definitions

$$\bar{E}_b-\bar{E}_a=\omega_{ba},\quad \bar{E}_0-\bar{E}_b=\omega_\sigma-\omega_0,$$

where ω_σ is the frequency of the decay photon in radians and $\omega_0\equiv\omega_{b0}$. In terms of these frequencies, we find that the various terms in the expression for the transition probability may be written as

$$\left|\frac{(\text{cofactor})_{an,bn-1}}{\text{Det}(D)}\right|^2_{E=E_{0,n-1}}=+|(H_{-,n})|^2|(n-1,n-2)|^2|(n-1,n+2)|^2\div|(n-1,n-2)(n-1,n)(n-1,n+2)$$
$$-|H_{+,n}|^2(n-1,n-2)(\omega_\sigma-\omega_0+\omega_{ba}-3\omega+i\Gamma_a/2)(\omega_\sigma-\omega_0+i\Gamma_b/2)$$
$$-|H_{+,n-2}|^2(n-1,n+2)(\omega_\sigma-\omega_0+\omega_{ba}-\omega+i\Gamma_a/2)(\omega_\sigma-\omega_0+2\omega+i\Gamma_b/2)$$
$$+|H_{+,n}|^2|H_{+,n-2}|^2(\omega_\sigma-\omega_0+\omega_{ba}-3\omega+i\Gamma_a/2)(\omega_\sigma-\omega_0+2\omega+i\Gamma_b/2)|^2,$$

$$\left|\frac{(\text{cofactor})_{an,bn-3}}{\text{Det}(E)}\right|^2_{E=E_{0,n-3}}=|(H_{-,n-2})(H_{+,n-2})(H_{-,n})(n-3,n+2)|^2\div|(n-3,n-2)(n-3,n)(n-3,n+2)$$
$$-|H_{+,n-2}|^2(n-3,n+2)(\omega_\sigma-\omega_0+\omega_{ba}-3\omega+i\Gamma_a/2)(\omega_\sigma-\omega_0+i\Gamma_b/2)$$
$$-|H_{+,n}|^2(n-3,n-2)(\omega_\sigma-\omega_0+\omega_{ba}-5\omega+i\Gamma_a/2)(\omega_\sigma-\omega_0-2\omega+i\Gamma_b/2)$$
$$+|H_{+,n}|^2|H_{+,n-2}|^2(\omega_\sigma-\omega_0+\omega_{ba}-5\omega+i\Gamma_a/2)(\omega_\sigma-\omega_0+i\Gamma_b/2)|^2,$$

and

$$\left|\frac{(\text{cofactor})_{an,bn+1}}{\text{Det}(E)}\right|^2\Bigg|_{E=E_{0,n+1}}$$
$$=|(H_{+,n})|H_{+,n-2}|^2(\omega_\sigma-\omega_0+4\omega+i\Gamma_b/2)(\omega_\sigma-\omega_0+\omega_{ba}-\omega+i\Gamma_a/2)$$
$$-(H_{+,n})(n+1,n-2)(\omega_\sigma-\omega_0+\omega_{ba}-\omega+i\Gamma_a/2)(\omega_\sigma-\omega_0+2\omega+i\Gamma_b/2)|^2\div|(n+1,n-2)(n+1,n)(n+1,n+2)$$
$$-|H_{+,n-2}|^2(\omega_\sigma-\omega_0+\omega_{ba}+\omega+i\Gamma_a/2)(\omega_\sigma-\omega_0+4\omega+i\Gamma_b/2)(n+1,n+2)$$
$$-|H_{+,n}|^2(\omega_\sigma-\omega_0+\omega_{ba}-\omega+i\Gamma_a/2)(\omega_\sigma-\omega_0+2\omega+i\Gamma_b/2)(n+1,n-2)$$
$$+|H_{+,n}|^2|H_{+,n-2}|^2(\omega_\sigma-\omega_0+\omega_{ba}-\omega+i\Gamma_a/2)(\omega_\sigma-\omega_0+4\omega+i\Gamma_b/2)|^2.$$

Here

$$(n',n'')\equiv[\omega_\sigma-\omega_0+(n'-n''+1)\omega+i\Gamma_b/2][\omega_\sigma-\omega_0+\omega_{ba}+(n'-n'')\omega+i\Gamma_a/2]-|H_{-,n''}|^2.$$

To locate the maxima in the transition probability as functions of $\omega_\sigma-\omega_0$, we assume

(1) $\Gamma_a\ll\Gamma_b<|H_\pm|$;

(2) that we may safely ignore the off-diagonal coupling terms $|H_{+,n'}|$ that occur in the denominators. The resonant contributions then all arise from the first term in each of the denominators involving the product of three terms of the form (n',n''). To justify this approach, a subsequent calculation to include the remaining terms of the denominator would show that the locations of the relative maxima are shifted by terms of the order of magnitude of

$$|H_{+,n'}|^2/2\omega_{ba}.$$

The major contribution to the transition probability comes, as one might expect, from the term involving the decay channel via the state $|b_{n-1}\rangle$. The smallest contribution arises from the term that decays via the state $|b_{n-3}\rangle$. Without going into the details of the algebra we list the location of the resonances as well as the value of the transition probability at these resonances.

Location of resonance	Magnitude of P_{0a}
$\omega_\sigma=\omega_0\pm\|H_-\|$	$P_{0a}=(2/\pi\Gamma_b)[1+\theta(\|H_-\|^2/\omega_{ba}^2)]$
$\omega_\sigma=\omega_0+2\omega_{ba}\pm\|H_-\|$	$P_{0a}=(2/\pi\Gamma_b)(\|H_-\|^2/4\omega_{ba}^2)$
$\omega_\sigma=\omega_0-2\omega_{ba}\pm\|H_-\|$	$P_{0a}=(2/\pi\Gamma_b)(\|H_-\|^2/4\omega_{ba}^2)(\Gamma_b/4\omega_{ba})^2$
$\omega_\sigma=\omega_0-4\omega_{ba}\pm\|H_-\|$	$P_{0a}=(2/\pi\Gamma_b)(\|H_-\|^2/4\omega_{ba}^2)(\Gamma_b/8\omega_{ba})^2$

Other resonances exist at $\omega_\sigma=\omega_0+n\omega_{ba}\pm|H_-|^2$ with even smaller magnitudes of P_{0a}.

The perturbation of the strong resonance at $\omega_\sigma=\omega_0\pm|H_-|$ may be estimated by determining the zero of the real part of the denominator of $g_{bn-1,an}$, that is $\text{Det}(E_{0,n-1})$, in the limit $\Gamma_b\to 0$. If we assume that

$$|H_{\pm,n'}|=|H_-|,$$

which is valid in the limit of intense rf fields, then the substitution of

$$\omega_\sigma=\omega_0+|H_-|+\delta$$

into the equation

$$\text{Real}[\text{Det}(E_{0,n-1})]=0$$

yields the following equation for δ:

$$\delta^2+2|H_-|\delta-|H_-|^4/4\omega_{ba}^2=0,$$

where we have ignored terms of order δ/ω_{ba} compared to one. Hence, the main resonance of the transition probability occurs at

$$\omega_\sigma=\omega_0\pm|H_-|(1+|H_-|^2/8\omega_{ba}^2).$$

The shift here arises from the coupling between the states arising from stimulated emission and takes the expected Bloch-Siegert form.[15]

Finally, to take a specific example, if the two states in question are the $2S_{1/2}$ and $2P_{3/2}$ states in hydrogen, then as pointed out by von Roos,[16] for external beam intensities of $\sim$7.2 W/cm^2 the interaction energy $|H_-|$ is about 1/10 the $2S_{1/2}-2P_{3/2}$ separation ω_{ba}. Hence, the intensities of the satellites are less than 1/100 that of the two main lines. The frequency split of the main line, $2|H_-|$, is about equal to three times the natural linewidth of the $2P-1S$ transition (Lyman α line). However, the broad band tuning requirements placed on the rf source are so great that the observation is most likely precluded.

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[15] F. Bloch and A. Siegert, Phys. Rev. **57**, 522 (1940).
[16] O. von Roos, Phys. Rev. **137**, A358 (1965).