are also possible. In that case, the results imply that $0.8<|\xi|<2.6$. It is important to note that the experimental point in Fig. 10 is consistent with at least one value of ξ. If the muon weak-coupling strength C_μ were appreciably different from the electron weak coupling strength C_e, the experimental result would not be consistent with any value of ξ. In order that the experimental result be consistent with at least one value of ξ, it is necessary that $0.77<|C_\mu|/|C_e|<1.48$.

The ratio of the differential muon spectrum in $K^+\to\pi^0+\mu^++\nu$ to the total rate of $K^+\to\pi^0+e^++\nu$ has been computed for $\xi=+0.84$. The computation and the experimental results corrected for the detection efficiency are shown in Fig. 11.

The $K^+\to\pi^0+\mu^++\nu$ decay has been investigated in a number of experiments. The results of the present experiment are consistent with the results of the experiments of Gidal *et al.*,[16] Smirnitski and Weissenberg,[15] Jensen *et al.*,[19] and Brown *et al.*[20] Our results are not in agreement with various aspects of the experiments of Dobbs *et al.*[21] and Boyarski *et al.*[22]

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SUMMARY

An analysis assuming a pure vector interaction with constant form factors and equal muon and electron weak-coupling strength has been shown to be consistent with the experimental observations. The vector form-factor ratio ξ can be real or imaginary. The probable solutions for ξ are shown in Fig. 10. There is no evidence on the basis of the present experiment that the form of the leptonic weak-interaction current $J_\lambda{}^l$ in the coupling to the current of the strongly interacting particles which changes strangeness $J_\lambda{}^{s,\Delta s=1}$ is different from the form of $J_\lambda{}^l$ in the coupling to the currents which do not change strangeness.

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Solution of the Schrödinger Equation with a Hamiltonian Periodic in Time*

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The interaction of a quantum system with an oscillating field is studied in a formalism which replaces the semiclassical time-dependent Hamiltonian with a time-independent Hamiltonian represented by an infinite matrix. The formalism is developed as a mathematical equivalent to the semiclassical treatment, and interpreted as a classical approximation to the quantum treatment of the field. Combined with a perturbation theory for two nearly degenerate states, the formalism provides a convenient method for determining resonance transition probabilities including frequency shifts and multiple quantum transitions. The theory is illustrated by a detailed study of the simple case of a two-state system excited by a strong oscillating field.

I. INTRODUCTION

CONSIDER a quantum system with two discrete states α and β. Let the amplitudes for the system to be in these states be $a_\alpha(t)$ and $a_\beta(t)$; the energies, E_α and E_β. Let an oscillating interaction connect these states with a matrix element $2b\cos\omega t$, where b is real. Then the system evolves according to the Schrödinger equation ($\hbar=1$)

$$i\frac{d}{dt}\begin{pmatrix}a_\alpha(t)\\ a_\beta(t)\end{pmatrix}=\begin{pmatrix}E_\alpha & 2b\cos\omega t\\ 2b\cos\omega t & E_\beta\end{pmatrix}\begin{pmatrix}a_\alpha(t)\\ a_\beta(t)\end{pmatrix}. \quad (1)$$

If at some initial time t_0 the system is in state α, then $|a_\beta(t)|^2$ represents the transition probability to state β, and is a function of t, t_0, b, ω, and the energy separation $E_\alpha-E_\beta$. A physical example of this simple system is a spin one-half particle in a static magnetic field with an oscillating magnetic field at frequency ω applied perpendicular to the static field.

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If only one of the two exponentials in the cosine function is retained in the interaction (rotating-field approximation), Eq. (1) can be solved exactly. The purpose of this article is to present some methods and results obtained in an attempt to find the transition probability for Eq. (1) without the rotating-field approximation.

In Sec. II we develop a formal theory relating the solution of a Schrödinger equation with a periodic Hamiltonian to the solution of another Schrödinger equation with a time-independent Hamiltonian represented by an infinite matrix. A physical interpretation of the formalism is obtained by making a correspondence with the theory which treats the interaction as arising from a quantized field.

The formal theory is valid not only for Eq. (1), but for any problem involving the interaction of a quantum system having discrete states with a classical field (well-defined amplitude and phase) of a single frequency. Such problems arise in radio frequency and microwave spectroscopy, especially when done by atomic-beam techniques. For convenience we shall often refer to the quantum system as an atom and the oscillating interactions as arising from an electromagnetic field, although nothing in the formalism restricts it to these cases. The theory covers only the effect of the field on the state of the atom, not the effect of the atom back on the field.

In Sec. III we make a two-state approximation to obtain transition probabilities for isolated resonances in a form similar to the "rotating-field" solution of Eq. (1). The method applies to any time-independent Hamiltonian for discrete states, such as that obtained by the theory of Sec. II. Frequency shifts and multiple quantum transitions are also accounted for by perturbation corrections. The method is illustrated by discussing the resonance transitions of Eq. (1).

In Sec. IV a graphical method for finding resonances in a time-independent Hamiltonian is discussed briefly. Finally, in Sec. V some additional results are presented on the solution of Eq. (1) when the exciting field is strong. Detailed derivations and discussion may be found in the author's thesis.[1]

II. THE FLOQUET THEORY

A. Development from Semiclassical Theory

We are interested in solving the time-dependent Schrödinger equation in matrix form

$$i(d/dt)F(t)=\mathfrak{IC}_C(t)F(t)\,, \tag{2}$$

where $\mathfrak{IC}_C$ is a Hermitian matrix of periodic functions of t with frequency ω or period $T(\omega T=2\pi)$. Our development is in three stages. First we determine the general form of the solution. Second we show the equivalence of finding a solution of this form to solving an eigenvalue-eigenvector problem for an infinite matrix. Lastly we express the time-evolution operator in terms of these eigenvalues and eigenvectors. This reveals the equivalence of Eq. (2) to an alternative Schrödinger equation whose Hamiltonian is a time-independent infinite matrix.

The general form of the solution of a differential equation with periodic coefficients is given by Floquet's theorem.[2] In matrix notation Floquet's theorem asserts the existence of a solution of Eq. (2) in the form

$$F(t)=\Phi(t)e^{-iQt}, \tag{3}$$

where Φ is a matrix of periodic functions of t and Q is a constant diagonal matrix. Since $\mathfrak{IC}_C$ is Hermitian, we can in fact find a solution in which $F(t)$ is unitary. Then

$$U(t;t_0)=F(t)F^{-1}(t_0) \tag{4}$$

is the time-evolution operator, i.e., the solution of (2) obeying $U(t_0;t_0)=\mathbf{1}$. The diagonal elements q_α of Q are called characteristic exponents. From (3) we find $F(t+T)=F(t)e^{-iQT}$. Hence the unitarity of F at all times guarantees that Q is a Hermitian matrix, or that the characteristic exponents are real. This is in contrast with the case of the Mathieu or Hill equation where the characteristic exponents are often complex.

From the formal solution of Eq. (2), one can show that

$$\det U(t_1;t_0)=\exp\left(-i\int_{t_0}^{t_1}\mathrm{Tr}\mathfrak{IC}_C(t)dt\right).$$

By Eqs. (3) and (4), $U(t_0+T;t_0)$ has the same eigenvalues as e^{-iQT}. Combining these results we discover that the sum of the characteristic exponents becomes, within an integral number of ω's, just the time average of the trace of $\mathfrak{IC}_C$:

$$\sum_\alpha q_\alpha=\frac{1}{T}\int_0^T\mathrm{Tr}\mathfrak{IC}_C(t)dt \pmod{\omega}\,. \tag{5}$$

Thus, one relation among the characteristic exponents may be found easily.

The next step after Eq. (3) is to expand the periodic functions of $\Phi(t)$ in Fourier series. We use Greek letters corresponding to atomic states to denote matrix elements in Eqs. (2) and (3), while Roman letters will denote Fourier components. Then the matrix elements of the solution $F(t)$ can be written

$$F_{\alpha\beta}(t)=\sum_n F_{\alpha\beta}{}^n e^{in\omega t}e^{-iq_\beta t}. \tag{6}$$

Similarly the expansion of the semiclassical Hamiltonian is

$$(\mathfrak{IC}_C)_{\alpha\beta}=\sum_n \mathfrak{IC}_{\alpha\beta}{}^n e^{in\omega t}. \tag{7}$$

[1] J. H. Shirley, thesis, California Institute of Technology, 1963 (unpublished).

[2] See, for example, F. R. Moulton, *Differential Equations* (MacMillan Company, New York, 1958) Chap. XVII, or other works on differential equations.

Substituting these expansions into the Schrödinger equation we obtain an infinite set of recursion relations for the $F_{\alpha\beta}{}^n$. They can be rewritten in the form of a matrix eigenvalue equation for the q's:

$$\sum_{\gamma k}(\mathcal{H}_{\alpha\gamma}{}^{n-k}+n\omega\delta_{\alpha\gamma}\delta_{kn})F_{\gamma\beta}{}^k=q_\beta F_{\alpha\beta}{}^n. \tag{8}$$

The operator is an infinite Hermitian matrix with rows identified by the pair of indices α, n, and columns by γ, k. This operator will be denoted by $\mathcal{H}_F$ and called the Floquet Hamiltonian associated with the semiclassical Hamiltonian $\mathcal{H}_C$. We order the components so that α runs over the atomic states before each change in n. In the case of Eq. (1), for example, the Floquet Hamiltonian is

$$\begin{pmatrix}
\cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\
\cdot & E_\beta-2\omega & b & 0 & 0 & 0 & 0 & 0 & 0 & \cdot \\
\cdot & b & E_\alpha-\omega & 0 & 0 & b & 0 & 0 & 0 & \cdot \\
\cdot & 0 & 0 & E_\beta-\omega & b & 0 & 0 & 0 & 0 & \cdot \\
\cdot & 0 & 0 & b & E_\alpha & 0 & 0 & b & 0 & \cdot \\
\cdot & 0 & b & 0 & 0 & E_\beta & b & 0 & 0 & \cdot \\
\cdot & 0 & 0 & 0 & 0 & b & E_\alpha+\omega & 0 & 0 & \cdot \\
\cdot & 0 & 0 & 0 & b & 0 & 0 & E_\beta+\omega & b & \cdot \\
\cdot & 0 & 0 & 0 & 0 & 0 & 0 & b & E_\alpha+2\omega & \cdot \\
\cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot
\end{pmatrix}. \tag{9}$$

Since two indices are required to identify a row or column of the matrix, it is convenient to introduce Dirac notation. Therefore, let $\mathcal{H}_F$ be defined by

$$\langle\alpha n|\mathcal{H}_F|\beta m\rangle=\mathcal{H}_{\alpha\beta}{}^{n-m}+n\omega\delta_{\alpha\beta}\delta_{nm}. \tag{10}$$

The $|\alpha n\rangle$ are an orthonormal basis providing the above matrix representation of $\mathcal{H}_F$. The index α represents an atomic state, but the index n merely represents a Fourier component. For the sake of nomenclature we shall refer to $|\alpha n\rangle$ as a "Floquet state."

From the form of Eq. (10) and the example (9) we see that the Floquet Hamiltonian has a periodic structure with only the number of ω's in the diagonal elements varying from block to block. This structure endows the eigenvalues and eigenvectors of $\mathcal{H}_F$ with periodic properties. The eigenvalues are in principle found from the secular equation $\det(\mathcal{H}_F-\lambda\mathbf{1})=0$. If λ is replaced by $\lambda+p\omega$, the equation is unchanged. Hence, if λ is an eigenvalue, so also is $\lambda+p\omega$ for any integer p. We can thus label the eigenvalues $\lambda_{\alpha n}=q_\alpha+n\omega$, where $q_\alpha=\lambda_{\alpha 0}$ is chosen, for example, as that member of the set having the smallest absolute value.

We denote the normalized eigenvector associated with the eigenvalue $\lambda_{\alpha n}$ by $|\lambda_{\alpha n}\rangle$. By writing the eigenvalue equation for $|\lambda_{\beta\, m+p}\rangle$, utilizing the periodic properties of $\mathcal{H}_F$ and $\lambda_{\beta\, m}$, and making an appropriate choice of phases, we find the following periodicity relation among the components of the eigenvectors:

$$\langle\alpha\, n+p|\lambda_{\beta\, m+p}\rangle=\langle\alpha\, n|\lambda_{\beta\, m}\rangle\,. \tag{11}$$

Since both $F_{\alpha\beta}{}^n$ and $\langle\alpha\, n|\lambda_{\beta\, 0}\rangle$ are the components of eigenvectors of $\mathcal{H}_F$ with the same eigenvalue $q_\beta=\lambda_{\beta\, 0}$, they must be proportional. The unitarity of $F(t)$ forces the proportionality constant to have absolute value one. Hence by an appropriate choice of phases we can identify $F_{\alpha\beta}{}^n$ with $\langle\alpha\, n|\lambda_{\beta\, 0}\rangle$ and rewrite Eq. (6)

$$F_{\alpha\beta}(t)=\sum_n\langle\alpha n|\lambda_{\beta 0}\rangle e^{in\omega t}e^{-iq_\beta t}. \tag{12}$$

We have thus expressed the solution (3) of Eq. (2) in terms of the solution of the eigenvalue problem for the Floquet Hamiltonian.

We now use Eq. (12) for $F(t)$ in Eq. (4) for $U(t;t_0)$. With the aid of the periodicity properties and a change of summation indices, the time evolution operator becomes

$$U_{\beta\alpha}(t;t_0)=\sum_n\sum_{\gamma l}\langle\beta n|\lambda_{\gamma l}\rangle \times\exp[-i\lambda_{\gamma l}(t-t_0)]\langle\lambda_{\gamma l}|\alpha 0\rangle e^{in\omega t}.$$

Since the eigenvectors $|\lambda_{\gamma l}\rangle$ are complete, we can write this more compactly as

$$U_{\beta\alpha}(t;t_0)=\sum_n\langle\beta n|\exp[-i\mathcal{H}_F(t-t_0)]|\alpha 0\rangle e^{in\omega t}. \tag{13}$$

The matrix elements appearing in this relation are the components of a vector $|t\rangle$ which satisfies the Schrödinger equation $i(d/dt)|t\rangle=\mathcal{H}_F|t\rangle$ with the initial condition $|t_0\rangle=|\alpha\, 0\rangle$. Thus $U_{\beta\alpha}(t;t_0)$, which is the amplitude that a system initially in atomic state α at time t_0 evolve to state β by time t according to the time-dependent Hamiltonian $\mathcal{H}_C(t)$, can also be interpreted as the amplitude that a system initially in the Floquet state $|\alpha\, 0\rangle$ at time t_0 evolve to the Floquet state $|\beta n\rangle$ by time t according to the time-independent Floquet Hamiltonian $\mathcal{H}_F$, summed over n with weighting factors $e^{in\omega t}$. The essence of our theory is that by using the latter interpretation of Eq. (13) problems involving Hamiltonians periodic in time may be solved by methods applicable to time-independent Hamiltonians.

B. Relation to Quantized Field Theory

The interpretation of Eq. (13) becomes clearer when it is derived by considering the periodic terms of $\mathcal{H}_C$ as arising from the interaction of the atom with a quantized field. To this end we introduce the Hamiltonian

$$\mathcal{H}_Q=\mathcal{H}_a+\mathcal{H}_f+\mathcal{H}_i,$$

where $\mathcal{H}_a$ is the Hamiltonian of the atom, $\mathcal{H}_f$ is the Hamiltonian of the field, and $\mathcal{H}_i$ is the interaction Hamiltonian, time-independent in the Schrödinger representation. The field is to have a single frequency, hence we use for $\mathcal{H}_f$ just the Hamiltonian of a single quantum harmonic oscillator of frequency ω. We introduce basis states $|\alpha\, n\rangle$, where α refers to the state of the atom and n refers to the excitation state of the oscillator, or the number of photons present. In this basis $\mathcal{H}_a$ and $\mathcal{H}_f$ are diagonal.

If this matrix representation of $\mathcal{H}_Q$ is written out for a specific case, such as that corresponding to Eq. (1), the similarity between it and $\mathcal{H}_F$ will be immediately apparent. There are only two differences. In $\mathcal{H}_Q$, n runs from zero to infinity, while in $\mathcal{H}_F$, n runs from minus infinity to infinity. Also the off-diagonal elements of $\mathcal{H}_Q$ depend on n (e.g., proportional to $\sqrt{n}$ if $\mathcal{H}_i$ is proportional to the annihilation operator), whereas those of $\mathcal{H}_F$ do not. But if we consider $\mathcal{H}_Q$ in the vicinity of some very large photon number N, the variation of matrix elements of $\mathcal{H}_i$ with n will be only of order $1/N$. We are thus led to the approximate identification

$$\mathcal{H}_Q \approx \mathcal{H}_F + N\omega \mathbf{1}, \tag{14}$$

which will be good only for photon numbers near N.

In writing Eq. (14) we are also associating the basis states for the two matrices. We are saying that the quantum state $|\alpha\, N+m\rangle$ is approximately isomorphic to the Floquet state $|\alpha\, m\rangle$ for $m \ll N$. This suggests that in using Floquet states we can interpret them physically as quantum states containing a definite, though very large, number of photons.

This interpretation will be strengthened if by using it we can reconstruct the semiclassical amplitude $U_{\beta\alpha}(t; t_0)$ from the fully quantized theory. In the semiclassical theory the oscillating interaction is considered to have a well-defined amplitude and phase which is unaffected by interaction with an atom. The state of a quantum oscillator which most closely approximates this classical description is the so-called oscillating wave packet,[3] or pure coherent state.[4] In terms of photon number eigenstates it is

$$\sum_{n=0}^{\infty} A_n e^{-in\omega t} |n\rangle ,$$

with the coefficients

$$A_n = \xi_0{}^n \exp(-\tfrac{1}{4}\xi_0{}^2)(2^n n!)^{-1/2},$$

where ξ_0 is the dimensionless classical amplitude of the field. The semiclassical amplitude $U_{\beta\alpha}(t; t_0)$ should correspond to the large photon number limit of the probability amplitude to go from the atomic state α and such an oscillator state at time t_0 to the atomic state β and the same oscillator state at time t:

$$U_{\beta\alpha}(t; t_0) \approx \langle t_f | \exp[-i\mathcal{H}_Q(t-t_0)] | t_0 \rangle , \tag{15}$$

with

$$|t_0\rangle = \textstyle\sum_n A_n e^{-in\omega t_0} |\alpha\, n\rangle$$

and

$$|t_f\rangle = \textstyle\sum_m A_m e^{-im\omega t} |\beta m\rangle .$$

When we go to the classical limit ($\xi_0 \gg 1$), the A_n coefficients become strongly peaked about a very large photon number $N \approx \frac{1}{2}\xi_0{}^2$ and extremely small elsewhere. Then the only important terms in the sums in $|t_0\rangle$ and $|t_f\rangle$ are those close to N. Hence in the limit it should be ligitimate to replace $\mathcal{H}_Q$ in (15) by the Floquet Hamiltonian according to (14). We also replace the quantum states by Floquet states. We can then use the periodicity properties to transform away the explicit appearance of N, leaving

$$U_{\beta\alpha}(t; t_0) \approx \sum_{k,n} A_{n+k}{}^* A_n \times \langle \beta k | \exp[-i\mathcal{H}_F(t-t_0)] | \alpha 0 \rangle e^{ik\omega t}. \tag{16}$$

The sum over n is one plus a term of the order of k^2/N. The matrix elements decrease rapidly enough with k so that Eq. (16) reduces to Eq. (13) in the limit of large N. A derivation of the semiclassical theory from the fully quantized theory is completed by showing that Eq. (13) satisfies Eq. (2) with the semiclassical Hamiltonian. Thus, our Floquet theory, while mathematically equivalent to the semiclassical theory, admits an interpretation in terms of states for a quantized field.

In a similar manner one can start from a quantized field theory including several discrete frequencies with photon numbers for each and evolve a Floquet theory having several Fourier indices, one for each mode. This many-mode Floquet theory has also been derived from the semiclassical theory, the only tricky point being the extension of Floquet's theorem.

C. Transition Probabilities

We can go one step further with the Floquet theory by using it to express the actual probabilities for transitions between atomic states. Using the periodicity properties for a bit of rewriting, we find

$$P_{\alpha\to\beta}(t; t_0) = |U_{\beta\alpha}(t; t_0)|^2 = \sum_{km} \langle \beta k | \exp[-i\mathcal{H}_F(t-t_0)] | \alpha 0 \rangle e^{im\omega t_0} \times \langle \alpha m | \exp[i\mathcal{H}_F(t-t_0)] | \beta k \rangle . \tag{17}$$

We read this as the probability to go from the initial atomic state α and a coherent field state (sum over m), to the final atomic state β, summed over all final field states (sum over k).

In actual experiments the initial time t_0, or equivalently the initial phase of the field seen by the atom, is not well defined, but is determined by a random process, such as the time of a collision or the time of entry of an atom into a radiation field region. Hence the quantity of interest is the transition probability averaged over initial times t_0, while keeping the elapsed time $t-t_0$

[3] L. I. Schiff, *Quantum Mechanics* (McGraw-Hill Book Company, Inc., New York, 1955), 2nd ed., pp. 67–69.
[4] R. J. Glauber, Phys. Rev. **131**, 2766 (1963).

fixed. This average can be readily performed on Eq. (17) yielding

$$P_{\alpha\to\beta}(t-t_0)=\sum_k |\langle\beta k|\exp[-i\mathcal{H}_F(t-t_0)]|\alpha 0\rangle|^2. \quad (18)$$

Equation (18) can be interpreted as the probability to go from a single initial Floquet state $|\alpha\,0\rangle$ (which contains no phase information) to a final Floquet state $|\beta k\rangle$, summed over all final states of the field.

If we also average over $t-t_0$, we obtain the time average transition probability

$$\bar{P}_{\alpha\to\beta}=\sum_k\sum_{\gamma l} |\langle\beta k|\lambda_{\gamma l}\rangle\langle\lambda_{\gamma l}|\alpha 0\rangle|^2=\sum_\gamma T_{\beta\gamma}T_{\alpha\gamma}, \quad (19)$$

where the matrix T is defined to be a partial sum of the squares of the eigenvector components

$$T_{\alpha\beta}=\sum_l |\langle\alpha l|\lambda_{\beta 0}\rangle|^2. \quad (20)$$

If all nonzero differences $\lambda_{\alpha l}-\lambda_{\beta m}$ are large compared to $1/\tau$, where τ is either $t-t_0$, or a relaxation time, whichever is shorter, then $\bar{P}$ is the transition probability needed to compare with experiment. In other cases we may wish to perform a weighted average of Eq. (18).

Regardless of the physical interpretation, Eqs. (18) and (19) provide a method of computing averaged transition probabilities which is superior to previous methods[5–7] that require a complete solution of the Schrödinger equation for $U_{\beta\alpha}(t;t_0)$, squaring, and finally averaging away half of the laboriously computed terms. We have now only to solve the Schrödinger equation with the time-independent Floquet Hamiltonian. The infinite sum over k in Eq. (18) does not cause difficulty, since the matrix elements decrease rapidly as k departs from 0.

III. PERTURBATION SOLUTIONS

A. General Procedure

In the usual case of a weak excitation, when the off-diagonal elements in $\mathcal{H}_F$ are small compared to most differences between diagonal elements [in (9) $b\ll\omega$], an approximate evaluation of the transition probability expression (18) is easy to obtain. We shall not use ordinary perturbation theory but rather a version discussed by Salwen in connection with resonance transitions in molecular beam experiments.[8] Salwen's technique is to identify resonances as occurring between two nearly degenerate diagonal elements of a time-independent Hamiltonian. That portion of the Hamiltonian matrix involving these nearly degenerate levels is separated out as a two-by-two matrix with perturbation corrections to its elements to approximately account for the rest of the matrix. For example, if $E_\alpha\approx E_\beta+n\omega$, the Floquet Hamiltonian would be approximated by

$$\mathcal{H}_2=\begin{pmatrix} E_\alpha+\delta_\alpha & \frac{1}{2}u \\ \frac{1}{2}u^* & E_\beta+\delta_\beta+n\omega \end{pmatrix},$$

where δ_α, δ_β, and part or all of u represent perturbation corrections for the rest of the matrix. The Schrödinger equation with $\mathcal{H}_2$ as Hamiltonian can be solved exactly by elementary techniques. The resulting transition probability is

$$P_{\alpha\to\beta}(t)=|\langle\beta n|\exp(-i\mathcal{H}_2 t)|\alpha 0\rangle|^2=(|u|^2/4q^2)\sin^2 qt, \quad (21)$$

where

$$4q^2=|u|^2+(n\omega-\omega_{\rm res})^2$$

and

$$\omega_{\rm res}=(E_\alpha+\delta_\alpha)-(E_\beta+\delta_\beta).$$

Clearly only one term in the k sum of Eq. (18) can be resonant for a given range of parameters, so we neglect the others and use Eq. (21) as our perturbation approximation to Eq. (18).

Equation (21) is simply a generalization of the Rabi formula long used by workers in atomic and molecular beam resonance spectroscopy.[9,10] It differs from the usual perturbation transition probability[11] by the occurrence of u in q. This simple change makes Eq. (21) valid for large times, whereas the usual perturbation formula is good only so long as it is much less than one. We have no averaging over a continuum as is required for Fermi's "Golden Rule."

The total procedure for finding resonance transition probabilities by the Floquet formalism and perturbation theory then reduces to three steps, each of which can be practically done by inspection: (1) write down the Floquet Hamiltonian, (2) write down $\mathcal{H}_2$, the 2×2 perturbation approximant to the Floquet Hamiltonian covering the transition of interest, and (3) write down Eq. (21) with u and $\omega_{\rm res}$ appropriately evaluated. The usefulness of Eq. (21) breaks down only when resonances appreciably overlap. In such cases a larger than 2×2 matrix must be diagonalized exactly, which is inconvenient to do analytically.

B. Application to Two-State System

To illustrate results of the procedure just outlined we consider the simple two-state problem given in Eq. (1). The Floquet Hamiltonian is given by (9). Suppose that the applied frequency ω is nearly resonant with the energy separation of the two states $E_\alpha\approx E_\beta+\omega$. If we take for $\mathcal{H}_2$ just the two-by-two submatrix involving

[5] F. Bloch and A. Siegert, Phys. Rev. **57**, 522 (1940).
[6] A. F. Stevenson, Phys. Rev. **58**, 1061 (1940).
[7] J. H. Shirley, J. Appl. Phys. **34**, 783 (1963).
[8] H. Salwen, Phys. Rev. **99**, 1274 (1955).

[9] N. F. Ramsey, *Molecular Beams* (Oxford University Press, London, 1956), pp. 115–155.
[10] P. Kusch and V. W. Hughes, in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1959), Vol. 37/1, pp. 54–73.
[11] L. D. Landau and E. M. Lifshitz, *Quantum Mechanics, Non-Relativistic Theory* (Addison-Wesley Publishing Company, Inc., Reading, Massachusetts, 1958), p. 146, Eq. (42. 3).

these levels with no perturbation corrections, we obtain for the transition probability Eq. (21) with $n=1$, $u=2b$, and $\omega_{\rm res}=E_\alpha-E_\beta$, i.e., the usual Rabi line-shape formula.[9,10] This formula is the exact solution of Eq. (1) when the cosine is replaced by one exponential (in magnetic resonance, when the oscillating field is replaced by a rotating field).

With both exponentials of the cosine present we have a nonresonant interaction of the states $|\alpha\,0\rangle$ and $|\beta\,1\rangle$ with the states $|\beta-1\rangle$ and $|\alpha\,2\rangle$ through the "antirotating" component of the oscillating field. We can incorporate this interaction into $\mathcal{H}_2$ by perturbation theory:

$$\mathcal{H}_2\approx\begin{pmatrix} E_\alpha+b^2/2\omega & b \\ b & E_\beta+\omega-b^2/2\omega \end{pmatrix},$$

where $E_\alpha-E_\beta$ has been approximated by ω in the correction terms. Our solution is the same as before except for a shift of the resonance frequency: $\omega_{\rm res}=E_\alpha-E_\beta+b^2/\omega$. This shift was first derived theoretically for magnetic resonance by Bloch and Siegert[5] and is often referred to as the Bloch-Siegert shift. However, the present derivation of it is much easier than previous ones.[5–7, 12]

From the present derivation we see that the Bloch-Siegert shift arises in exactly the same way as the shifts due to couplings with other levels derived by Salwen.[8] They are due to a small intermixing of levels coupled by the interaction. In the language of quantum electrodynamics the shifts arise from virtual (energy-nonconserving) transitions to the additional state and back again. In the Bloch-Siegert shift the additional state just happens to be distinguished by its photon number rather than its atomic state.

In quantum field theory the matrix element $\langle\beta\,1|\mathcal{H}_Q|\alpha\,0\rangle$ is nonzero. An energy shift occurs by a spontaneous virtual transition in the absence of an exciting radiation field. This kind of shift occurs for all possible frequencies of the radiation field and has been the subject of much investigation in the theory of quantum electrodynamics. We are here assuming that all such shifts, like the Lamb shift in hydrogen, have been included in the Hamiltonian of the atom. The shifts we are describing here are increases to such shifts due to the increased probability of the virtual transition when quanta are present in the field, i.e., due to stimulated virtual transitions.

By a careful application of perturbation theory in diagonalizing the Floquet Hamiltonian and the retention of two terms in the k sum of Eq. (18) instead of one, a solution of Eq. (1) can be obtained to second, or even third order in b/ω without excessive labor. We consider the Bloch-Siegert solution to be first order. The second-order solution is

$$P_{\alpha\to\beta}(t)=\frac{b^2}{p^2}\left(1-\frac{b^2}{\omega^2}\right)\sin^2 qt+\frac{b^2}{2\omega^2}-\frac{b^2}{4\omega^2}\left[\left(\frac{b^2}{p^2}+\frac{p^2+\Delta^2}{p^2}\cos 2qt\right)\cos 2\omega t+2\frac{\Delta}{p}\sin 2qt\sin 2\omega t\right], \tag{22}$$

where

$$\Delta=\tfrac{1}{2}(\omega-\omega_0-b^2/\omega)(1-b^2/4\omega^2)\,,\quad \omega_0=E_\alpha-E_\beta\,,$$
$$p^2=b^2+\Delta^2,\quad \text{and}\quad q=(1-b^2/4\omega^2)p\,.$$

When b/ω is small and we are close to resonance, the second line of Eq. (22) oscillates very rapidly compared to the first line and may be disregarded as not observed. The first line is essentially of the form of Eq. (21) except for a small decrease in amplitude and the addition of a constant term representing the nonresonant background transition probability due to overlap with other resonances. The third-order solution includes the next higher correction term to the resonance frequency shift and also some asymmetric distortion from overlap. Similar results are encountered when other problems are solved to higher order. Since the most pronounced effect is the resonance frequency shift calculated in first order, we can use first-order solutions with increased confidence in their usefulness, even when b/ω is as large as $\frac{1}{3}$.

In analogy with Salwen's method[8] we can find transition probabilities for multiple quantum transitions occurring between atomic states not directly connected by matrix elements of the interaction, but indirectly connected through one or more intermediate states. With the Floquet Hamiltonian we are not restricted to distinct atomic states for intermediate states, but can use the same atomic state as the initial or final state, but with a different number of photons present. For example, suppose $E_\alpha\approx E_\beta+3\omega$. The states $|\alpha\,0\rangle$ and $|\beta\,3\rangle$ are not directly connected in the Floquet Hamiltonian (9), but are indirectly connected through $|\beta\,1\rangle$ and $|\alpha\,2\rangle$ as intermediate states. A nonzero matrix element between $|\alpha\,0\rangle$ and $|\beta\,3\rangle$ appears in third order of the perturbation theory. Using it we find by our general procedure a triple quantum transition probability of the form of Eq. (21) with

$$n=3,\quad u=b^3/2\omega^2$$

and

$$\omega_{\rm res}=E_\alpha-E_\beta+3b^2/2\omega.$$

This result has been obtained previously by Winter[13] and the transition has been observed by Marjerie and Brossel.[14] Strong exciting fields are required to observe it, since all three photons must impinge on the atom

[12] S. Autler and C. H. Townes, Phys. Rev. **100**, 703 (1955).

[13] J. Winter, Compt. Rend. **241**, 375, 600 (1955); Ann. Phys. (Paris) 4, 745 (1959).

[14] J. Marjerie and J. Brossel, Compt. Rend. **241**, 373 (1955).

before its virtual intermediate states decay back to the original state. The lifetimes of the virtual intermediate states are very short because of the large amount of energy nonconservation (2ω). Well placed atomic intermediate states may require much less energy nonconservation and lead to multiple quantum transitions which are much easier to observe (larger u for a given value of b).

As Winter[13] noted, the single two-state system described by Eq. (1) shows resonances when $E_\alpha \approx E_\beta + n\omega$ for n any odd integer.[15] Perturbation theory on the Floquet Hamiltonian enables us to easily find a transition probability of the form of Eq. (21) for all of these transitions. The results are (p is an integer)

$$\begin{aligned} &n=2p+1\,,\\ &u=(b^{2p+1})/2^{2p-1}(p!)^2\omega^{2p} \quad (p\geqq 0)\,, \qquad (23)\\ &\omega_{\rm res}=E_\alpha-E_\beta+\{(2p+1)/[p(p+1)]\}b^2/\omega \quad (p>0)\,. \end{aligned}$$

Keeping b and ω fixed and observing the transition probability as a function of $\omega_0=E_\alpha-E_\beta$, we find resonance near $\omega_0=\omega$, 3ω, 5ω, 7ω, $\cdots$. The shifts are all towards smaller ω_0 and decrease with increasing p. The widths u decrease rapidly to the point where the transitions are unobservable, both because of the powers of b/ω, and the factorials in the denominator.

In Salwen's work a time-independent Hamiltonian was obtained by assuming a rotating-field type of interaction and then making a quantum-mechanical transformation to a rotating coordinate system. The Floquet theory makes this transformation unnecessary. On the other hand, the rotating coordinate transformation on Eq. (1) results in a periodic Hamiltonian with frequency 2ω instead of ω, and the Floquet Hamiltonian corresponding to it has a simpler structure than (9), although a somewhat confused correspondence with the quantized field Hamiltonian.

Workers in magnetic resonance have often obtained solutions for spin-flip transition probabilities by solving the corresponding classical problem of a spinning dipole in a magnetic field using a rotating coordinate system.[16] Feynman, Vernon, and Hellwarth[17] have in fact shown that the semiclassical Schrödinger equation for a two-state system is mathematically equivalent to the classical vector equation $d\mathbf{r}/dt=\boldsymbol{\omega}\times\mathbf{r}$. Hence the Bloch-Siegert shift and all the multiple quantum transitions derived above should appear in classical arguments. Ramsey[18] has shown how the Bloch-Siegert shift may be obtained by considering the problem in rotating coordinate frames. The multiple quantum transitions can be derived by several successive transformations to rotating coordinate frames whose axes are slightly tilted with respect to each other. This procedure has been successfully carried out for $n=3$, 5, and 7, giving values of u and $\omega_{\rm res}$ in agreement with (23). But it is clear that this method rapidly becomes much more complicated than perturbation theory on the Floquet Hamiltonian.

The Floquet theory has the additional advantage that it can be used with systems involving several levels where the rotating coordinate transformation fails. For example, suppose we have a three-level system with one level nearly midway between the other two. The oscillating field has nonzero matrix elements only between the middle level and each of the two end levels. For a rotating field interaction Salwen's theory would predict a double quantum resonance between the end levels. For an oscillating field interaction the Floquet theory predicts and gives, solutions for not only the double quantum transition, but also a quadruple quantum transition and in fact a whole series of resonances involving an even number of photons.

IV. INTERPRETATION OF EIGENVALUE PLOTS

When the radiation field is too intense for the perturbation solutions of Sec. III to be adequate, or when resonances overlap, resort must usually be made to some form of numerical computation. It is possible to evaluate Eqs. (18) or (19) numerically since the sums and the infinite matrix can be truncated with negligible error. Much information can be learned, however, just from the characteristic exponents or eigenvalues of the Floquet Hamiltonian. In the first place they represent physical energy levels of the atom-radiation field system in interaction and may be observed experimentally by exciting transitions between them with a second very weak field.[12,19] Also, a plot of the characteristic exponents as a function of a parameter can be very useful in locating resonances and guiding further numerical work.

Suppose in our time-independent Hamiltonian we write the diagonal elements as linear functions of a parameter y: $a_\alpha y+b_\alpha$. Following Salwen[8] we may plot all the diagonal elements as a function of y on the same graph and look for resonances where the lines cross. Alternatively, we may plot the eigenvalues λ_α of the Hamiltonian as a function of y.[20] Besset *et al.*[21] describe resonances as occurring where the curves for two eigenvalues approach each other closely but do not cross. Their description can be verified by considering the first and second derivatives of $\lambda_\alpha(y)$.

By differentiating the eigenvalue equation for a time-

[15] Transitions with n even become allowed if $\cos\omega t$ terms appear in the diagonal elements of the semiclassical Hamiltonian, e.g., if in magnetic resonance the oscillating field has a component parallel to the static field.

[16] I. Rabi, N. F. Ramsey, and J. Schwinger, Rev. Mod. Phys. **26**, 167 (1954).

[17] R. P. Feynman, F. L. Vernon, and R. W. Hellwarth, J. Appl. Phys. **28**, 49 (1957).

[18] N. F. Ramsey, Phys. Rev. **100**, 1191 (1955).

[19] W. Happer, Phys. Rev. **136**, A35 (1964).

[20] Such plots for a five-level system with y proportional to the static magnetic field may be found in Happer's paper (Ref. 19). Figures 1 and 2 of Ref. 12 and Fig. 1 of this paper are also of this type.

[21] C. Besset, J. Horowitz, A. Messiah, and J. Winter, J. Phys. Radium **15**, 251 (1954).

independent Hamiltonian one obtains the relations[22]:

$$\partial\lambda_\alpha/\partial y=\langle\lambda_\alpha|\partial\mathcal{H}/\partial y|\lambda_\alpha\rangle\,, \tag{24}$$

$$\frac{\partial^2\lambda_\alpha}{\partial y^2}=2\sum_{\gamma\neq\alpha}\frac{|\langle\lambda_\alpha|\partial\mathcal{H}/\partial y|\lambda_\gamma\rangle|^2}{\lambda_\alpha-\lambda_\gamma}\,. \tag{25}$$

If we assume that the eigenstate $|\lambda_\alpha\rangle$ is composed almost entirely of one unperturbed state $|\alpha\rangle$, Eqs. (24) and (25) tell us that λ_α is approximately a linear function of y with slope a_α. If we assume that the eigenstate $|\lambda_\alpha\rangle$ is composed almost entirely of two unperturbed states, (24) and (25) tell us that $\lambda_\alpha(y)$ is approximately a hyperbola. Since Eq. (21) was also derived on the basis of a two-state approximation, we can associate with a hyperbolic region of an eigenvalue plot a transition probability between the two states of the form of (21). In fact, the minimum separation of the two branches of the hyperbola is equal to the width parameter u and the position of minimum separation gives the position of the resonance. Thus a perusal of an eigenvalue plot gives information about the location and strength of resonances and whether a one- or two-state approximation can be made or not, independent of perturbation criteria for such approximations.

V. TWO-STATE SYSTEM IN A STRONG OSCILLATING FIELD

In the simple case of only two atomic states in the Floquet Hamiltonian we can determine the time-average transition probability completely from the characteristic exponent. From Eq. (5) we have $q_\alpha+q_\beta=E_\alpha+E_\beta$, hence only one characteristic exponent, which we call q, need be determined. Instead of plotting q as a function of ω, as was done by Autler and Townes,[12] we plot it as a function of $\omega_0\equiv E_\alpha-E_\beta$ using ω as a scaling parameter (see Fig. 1). Let $E_\alpha=\frac{1}{2}\omega_0$ and $E_\beta=-\frac{1}{2}\omega_0$. Then the diagonal elements of the Floquet Hamiltonian (9) are linear functions of ω_0 with slopes $\pm\frac{1}{2}$ determined by the atomic state and independent of

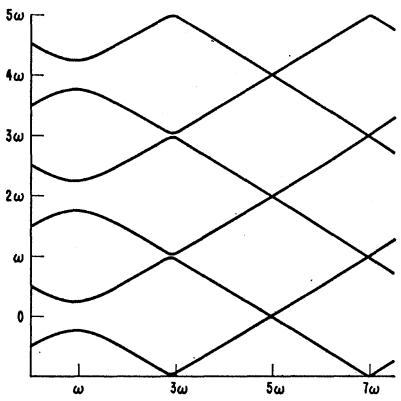

FIG. 1. Six branches of the characteristic exponent q as a function of ω_0 with $b/\omega=\frac{1}{4}$.

[22] Compare with the results of K. Aizu, J. Math. Phys. 4, 762 (1963).

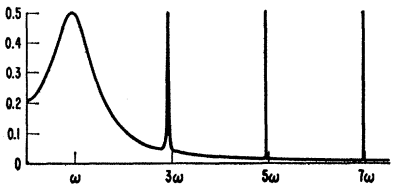

FIG. 2. Time-average transition probability as a function of ω_0 with $b/\omega=\frac{1}{4}$.

photon number. Equation (24) becomes

$$\frac{\partial\lambda_{\alpha 0}}{\partial y}=\frac{\partial q}{\partial\omega_0}=\sum_{\gamma l}a_\gamma|\langle\gamma l|\lambda_{\alpha 0}\rangle|^2=\sum_\gamma a_\gamma T_{\gamma\alpha}\,,$$

where the T matrix was defined by Eq. (20). But the normalization of the eigenvectors gives three other relations among the four components of T. So all four components, and hence the proportion of atomic state α or β in the corresponding eigenvector, can be expressed in terms of the derivative of the eigenvalue $\partial q/\partial\omega_0$. Substituting such expressions for T into Eq. (19) we find for the time-average transition probability

$$\bar{P}_{\alpha\to\beta}=\tfrac{1}{2}[1-4(\partial q/\partial\omega_0)^2]\,. \tag{26}$$

This remarkable result permits us to sketch the time-average transition probability directly from a plot of the characteristic exponent. Unfortunately, no such simple relation exists when more than two atomic states are involved, but to the extent that all but two atomic states can be neglected, Eq. (26) should be approximately true.

Because of the existence of Eq. (26) the characteristic exponent q for Eq. (1) [eigenvalue of (9)] was studied in detail as a function of b and ω_0 with ω fixed as the scale parameter. The following analytical approximations indicate the complexity of the function q. For ω_0 much less than the larger of ω, $(b\omega)^{1/2}$, an expression in terms of a Bessel function was found:

$$q\approx\tfrac{1}{2}[E_\alpha+E_\beta+\omega-\omega_0 J_0(4b/\omega)]\,. \tag{27}$$

For ω_0 much greater than the larger of ω, $(b\omega)^{1/2}$, an expression in terms of a complete elliptic integral was found:

$$q\approx\tfrac{1}{2}(E_\alpha+E_\beta)+(4b/\pi k)E(k)\,, \tag{28}$$

where

$$k^2=16b^2/[16b^2+(\omega-\omega_0)^2]\,.$$

For b small compared to ω and ω_0, a power series with rational functions as coefficients was found:

$$q=\tfrac{1}{2}(\omega-\omega_0)+\frac{2\omega_0 b^2}{\omega^2-\omega_0^2}-\frac{2\omega_0(\omega^2+3\omega_0^2)b^4}{(\omega^2-\omega_0^2)^3}$$
$$+\frac{32\omega_0^3(\omega^2+\omega_0^2)b^6}{(\omega^2-\omega_0^2)^5}+\frac{8\omega_0 b^6}{(\omega^2-\omega_0^2)^2(9\omega^2-\omega_0^2)}+\cdots\,. \tag{29}$$

Near $\omega_0=\omega$ the expansion for q^2 is nonsingular and more rapidly converging. It can be used to find the higher-order

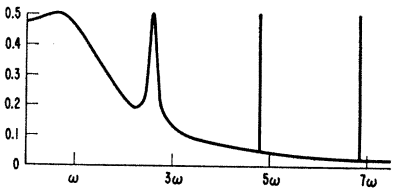

FIG. 3. Time-average transition probability as a function of ω_0 with $b/\omega=\frac{1}{2}$.

terms in the Bloch-Siegert shift. We set $\partial q^2/\partial\omega_0=0$ and solve for ω to find the resonance condition

$$\omega=\omega_0+b^2/\omega_0+b^4/4\omega_0^3-35b^6/32\omega_0^5-\cdots.$$

The asymptotic position of the resonance is found from Eq. (27):

$$\omega\sim 1.663b \quad \text{for} \quad b\gg\omega_0,$$

where the numerical coefficient is 4 divided by the first zero of J_0.

Numerical integration of the Schrödinger equation by a digital computer was performed over a sufficient range of values of b and ω_0 to reveal the general behavior of the characteristic exponent q. In Fig. 1 we have plotted six of the characteristic exponents as a function of ω_0 with $b=\frac{1}{4}\omega$. At the right of Fig. 1 the curves should not actually cross, but their minimum separation is too small to be shown in the figure. The close approach of two branches near $\omega_0=3\omega$, 5ω, and 7ω indicates the presence of resonances which can be adequately described by (21).

For the broad resonance near $\omega_0=\omega$ the time-average transition probability of Eq. (19) gives an adequate description since all frequencies in the time dependence are of the order of ω and not observed. In Fig. 2 we have plotted the time-average transition probability corresponding to Fig. 1 by numerical evaluation of (26). The vertical lines represent resonances whose width is too narrow to be shown in the figure. The relative widths $(2u/\omega)$ of the four peaks shown are 1.2, 1.6×10^{-2}, 6×10^{-5}, and 10^{-7}, respectively. The higher resonances with so little area under them cannot be seen experimentally because of the finite resolving power of the apparatus.

As we increase b each resonance in Fig. 2 broadens and shifts toward smaller values of ω_0 (see Fig. 3). The curve is symmetric about $\omega_0=0$, hence the large transition probability there arises from overlap with the $\omega_0=-\omega$ resonance. As we increase b further the slope of q at $\omega_0=0$ changes sign [see Eq. (27)]. As this happens the twin resonance peaks from $\omega_0=\pm\omega$ coalesce into a large broad background, while the three quantum resonance rides up on this background to appear dominant (see Fig. 4). As we increase b still further each resonance in turn shifts in and broadens until it becomes lost in the ever widening background, but at any given value of b probably only about two resonances are broad enough to be observable.

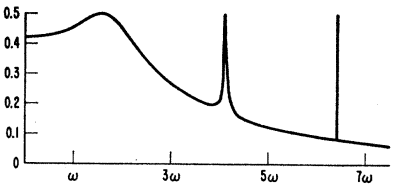

FIG. 4. Time-average transition probability as a function of ω_0 with $b/\omega=1$.

As b increases, the time-average transition probability for fixed ω and ω_0 increases to $\frac{1}{2}$, and then fluctuates closer and closer to $\frac{1}{2}$, hitting it repeatedly. When b is large compared to ω_0 the external oscillating field is much stronger than the static or atomic field producing the energy separation ω_0. Hence the state, defined relative to the weaker field, is no longer significant and merely averages out to about one half.

In Figs. 2–4 we have plotted the transition probability as a function of ω_0 keeping the ratio b/ω fixed. In magnetic resonance observations are often made this way by varying the static magnetic field. But sometimes one records resonances by varying ω and keeping b and ω_0 constant. Figure 5 shows such a plot. As b is increased

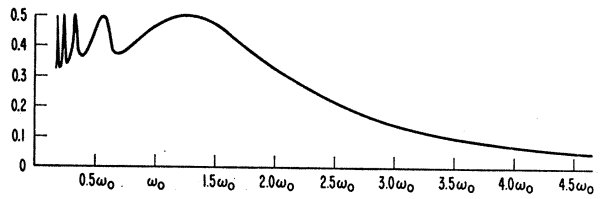

FIG. 5. Time average transition probability as a function of ω with $b/\omega_0=\frac{1}{2}$.

the single quantum resonance in the plot will broaden and shift outward to larger ω indefinitely. The other resonances also broaden and ride along behind.

The fields required to achieve the situations just described are uncommonly strong. To make $b/\omega=1$ we need a peak oscillating field strength of roughly 10^4 V/cm at radio frequencies, 10^6 V/cm at microwave frequencies, and 10^8 V/cm at optical frequencies. Such strong fields can cause ionization or other effects not considered in the preceding theory. Any experimental test would best be done at radio frequencies.

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