PHYSICAL REVIEW A VOLUME 15, NUMBER 3 MARCH 1977

Floquet theory and applications

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(Received 15 July 1974; revised manuscript received 12 July 1976)

Floquet's theorem and a corresponding perturbation theory are derived for Hamiltonians periodic in time. The resulting theory is then applied to the interaction of stationary and moving, neutral two-level atomic or molecular systems with monochromatic linearly and circularly polarized electromagnetic radiation.

I. INTRODUCTION

A. Origin and statement of the problem

A recently developed spectroscopic technique[1] uses an optical field ($\omega \approx 10^{14}$ rad/sec) to interact with a group of slowly moving atoms whose velocity components parallel to the direction of the field are small. Reflecting on the limits of resolution of this technique leads to considering the effect of an applied field on the orbital motion of an atom.[2] Of particular interest are the effects of (i) the resonant interaction of the field with the internal levels of the atom on the evolution of the wave packet describing the atomic center-of-mass motion and (ii) the recoil effect associated with the absorption or emission of a photon by the atom.[3] An understanding of this problem would lead to additional insight in a number of other areas: the theory of gas lasers; frequency standards which use a gaseous absorption cell as reference; such applications as discussed by Ashkin[4] "to separate, velocity analyze or trap neutral atoms of specific isotropic species or hyperfine level"; and in the recently observed deflection of an atomic beam by laser light.[5,6]

To study the problem mentioned above, it is necessary to consider solutions of Schrödinger's equation with a temporally periodic Hamiltonian. The main purpose of this paper is to discuss Floquet's theorem for general temporally periodic Hamiltonians, derive a perturbation theory from Floquet's theorem, and apply this theory to the interaction of stationary and moving neutral two-level atomic or molecular systems with monochromatic linearly and circularly polarized electromagnetic radiation.

B. Background

Bloch and Siegert,[7] in one of the earliest discussions of the interaction of atoms with periodic fields, considered the interaction of a stationary spin-$\frac{1}{2}$ atom in a static magnetic field B_0 with an applied radio-frequency field $2B_1\cos(\omega t)$ of small intensity and which was aligned perpendicular to B_0. They concluded that increasing the oscillating field strength causes a shift in the resonance frequency from $\omega = \omega_0$ to ω defined by $\omega_0 = \omega - (\gamma B_1)^2/4\omega$, where $\gamma = -\omega_0/B_0$ and ω_0 is the Rabi frequency. Magerie and Brossel[8] experimentally demonstrated that a sufficient increase in field strength causes multiple transitions, thus showing that the next B_1^4 term in the series for the resonance frequency eventually becomes significant. Further work aimed directly at approximating the Bloch and Siegert shift has been done by Ahmad and Bullough[9] and by S. Swain.[10]

Shirley[11] took an approach to the Bloch and Siegert problem which is close in spirit to the general approach used in this paper. By means of Floquet theory, he derived expressions for the higher-order terms in the Bloch and Siegert shift. He related the solution of a Schrödinger equation with a temporally periodic Hamiltonian represented by an infinite matrix. In this way, he determined resonance transition probabilities including higher-order frequency shifts as well as multiple quantum transitions for the case of a stationary two-state system excited by a strong oscillating field. Pegg and Series[12,13] obtained similar results by viewing the atomic Hamiltonian from a reference frame in which it appeared to be nearly static, thus obtaining a time-dependent Schrödinger equation in an approximately integrable form. Quantum transitions and higher-order terms in the Bloch-Siegert shift were described as transitions induced by field harmonics in the appropriate frame. Other approaches to the problem of a time-dependent Schrödinger equation have been developed,[14-17] but they will not be discussed here.

All of the methods mentioned above are powerful in that they can be applied to a broad range of time-dependent quantum-mechanical problems. A common limitation is, however, that they do not easily generalize to systems with Hamiltonians which possess operators with continuous spectrums. A simple example occurs in the case of a slowly moving two-state atom in a z-directed static magnetic field B_0 interacting with a plane wave, classical field propagating in the z direction with components aligned perpendicular to B_0. The total Hamiltonian in this case is given by a 2×2 matrix, periodic in time, containing both position and momentum operators. The methods which will be introduced here are capable of dealing with such cases.

C. Organization and summary

Let the total Hamiltonian for a system be of the form $H(t)=H_0+H_1(t)$, where H_0 denotes the unperturbed static Hamiltonian, $H_1(t)$ a temporally periodic perturbation of H_0, and λ a coupling parameter. We will first consider what modification of the finite-dimensional matrix version of Floquet theory will be necessary to include cases in which H_0 or $H_1(t)$ possess continuous or discrete or both types of spectrums. A version of time-dependent perturbation theory appropriate to this case will be presented and discussed in Sec. II.

In Sec. III, we consider the Bloch-Siegert problem. Using the version of Floquet theory developed in Sec. II, we determine resonance transition probabilities, the higher-order Bloch-Siegert frequency shift, and also discuss multiple quantum transitions from a mathematical point of view.

A generalization of the Bloch-Siegert problem is presented early in Sec. IV. We allow the two-state atom to move slowly through a certain prescribed, classical standing wave. A transformation of the relevant Schrödinger equation then enables us to relate the solution of this problem to the solution of the corresponding "stationary-atom" problem worked out in Sec. III. We note the appearance of Doppler frequency shifts and recoil terms due to the motion of the atom through the standing wave. Our third example is concerned with an exactly soluble model: a single two-state neutral atom in a monochromatic, circularly polarized plane wave of arbitrary intensity. We study the coupling between the center of mass and internal degrees of freedom from the Floquet point of view.

II. FLOQUET'S THEOREM AND HAMILTONIANS PERIODIC IN TIME

Floquet's theorem has become a well known and much utilized tool of solid-state physics. Here it has been used in connection with the spatial periodicity of the effective potential for a single electron. According to Floquet's theorem, the spatial periodicity implies that the solutions of Schrödinger's equation for the electron wave function can be chosen as the product of a spatially periodic function and an exponential function of position. These so called Bloch waves have played a very important role in solid-state physics. However, in connection with Hamiltonians periodic in time it appears that Floquet's theorem has seldom been exploited.[18-21] It is of course a rather simple mathematical point that Floquet's theorem applies to systems periodic in time as well as in position. But it is still reasonable to expect that new physical insights will emerge when time periodic Hamiltonians are considered from this point of view.

A. Elementary example

In order to motivate our later discussion consider a stationary two-state dipole in a linearly polarized, single frequency, classical field. The Hamiltonian is

$$H(t)=\tfrac{1}{2}\hbar\,\omega_0\sigma_z+\tfrac{1}{2}\hbar\,\omega_0\lambda\sigma_x\sin\omega t\ , \qquad (2.1)$$

where ω_0 denotes the transition frequency, ω is the applied field frequency, and λ is a coupling parameter proportional to the amplitude of the oscillating field. (More precisely, λ is the ratio of the Rabi frequency to ω_0.) σ_x and σ_z are the Pauli spin operators. Schrödinger's equation can be written

$$i\hbar\dot{U}(t)=\tfrac{1}{2}\hbar\,\omega_0(\sigma_z+\lambda\sigma_x\sin\omega t)U(t)\ , \qquad (2.2)$$

where U is the time displacement operator for the system. Differentiating this equation with respect to time gives

$$i\hbar\ddot{U}=(\tfrac{1}{2}\hbar\,\omega_0\omega\lambda\sigma_x\cos\omega t)U+\tfrac{1}{2}\hbar\,\omega_0(\sigma_z+\lambda\sigma_x\sin\omega t)\dot{U}\ .$$

Using Eq. (2.2) to eliminate $\dot{U}$ yields an operator version of Hill's equation, viz.,

$$\ddot{U}+[\tfrac{1}{2}i\omega_0\omega\,\lambda\cos\omega t\,\sigma_x+\tfrac{1}{4}\omega_0^2(1+\lambda^2\sin^2\omega t)]U=0. \qquad (2.3)$$

Equation (2.3) can be put into diagonal form by passing from U to U_R where

$$U_R(t)\equiv e^{i\pi\sigma_y/4}\,U(t)\ . \qquad (2.4)$$

The transformed version of Eq. (2.3) reads

$$\ddot{U}_R+[\tfrac{1}{2}\,i\omega_0\omega\lambda\sigma_z\cos\omega t+\tfrac{1}{4}\omega_0^2(1+\lambda^2\sin^2\omega t)]U_R=0\ . \qquad (2.5)$$

Let $|0\rangle$ denote the state vector for the dipole at $t=0$, $|s\rangle$ $(s=\pm 1)$ denote the eigenvectors of σ_z, and $\psi_R(t,s)\equiv\langle s|U_R(t)|0\rangle$. The wave-function equivalent of Eq. (2.5) is

$$\ddot{\psi}_R(t,s)+[\tfrac{1}{2}i\omega_0\omega\lambda s\cos\omega t+\tfrac{1}{4}\omega_0^2(1+\lambda^2\sin^2\omega t)]\psi_R(t,s)=0\ . \qquad (2.6)$$

According to Floquet's theorem, Eq. (2.6) has solutions of the form $e^{\pm i\mu t}\phi_s^{\pm}(\lambda,t)$, where $\phi_s^{\pm}(\lambda,t)$ are periodic in time with period $2\pi/\omega$ and μ is time independent. μ must be entirely real in order for $\psi_R(t, s)$ to be normalizable for all time. The most general solution to Eq. (2.6) is

$$\psi_R(t, s) = a_s e^{i\mu t}\phi_s^+(\lambda, t) + b_s e^{-i\mu t}\phi_s^-(\lambda, t) \ , \qquad (2.7)$$

where a_s and b_s are constants determined by the initial conditions. Replacing t by $-t$ in Eq. (2.6) shows that $e^{-i\mu t}\phi_s^+(\lambda, -t)$ is a solution. This means that $\phi_s^-(\lambda, t)$ is proportional to $\phi_s^+(\lambda, -t)$. As a consequence we need only to calculate one of these functions.

Several methods have been developed to determine the Floquet constant $\mu(\lambda)$ and the periodic functions $\phi_s^\pm(\lambda, t)$.[22] We employ the standard approach of expanding μ and $\phi_s^+(\lambda, t)$ into power series in λ, i.e.,

$$\mu = \sum_{n=0}^{\infty} \mu_n \lambda^n, \quad \phi_s^+(\lambda, t) = \sum_{n=0}^{\infty} \phi_n(s, t)\lambda^n \ . \qquad (2.8)$$

We then substitute these expansions into Eq. (2.6).

For each power of λ we obtain a linear second-order differential equation for the time-dependent coefficients. The condition that the solution has no secular time dependence determines the values of the expansion coefficients for $\mu(\lambda)$. Using this procedure we can, in principle, calculate $\mu(\lambda)$ and ϕ_s^+ to any order in λ. The function $\phi_s^+(\lambda, t)$ is of the form

$$\phi_s^+ = \sum_{n=0}^{\infty} \lambda^n \sum_{m=0}^{n} (a_{nm}^{(s)} \cos m\omega t + b_{nm}^{(s)} \sin m\omega t). \qquad (2.9)$$

Also for the Hamiltonian of Eq. (2.1) it is convenient to expand $(\mu - \omega/2)^2$ as a power series in λ, i.e., set

$$\mu(\lambda) = \tfrac{1}{2}\omega + \tfrac{1}{2}[(\omega_0 - \omega)^2 + \mu_1'^2\lambda^2 + \mu_2'^2\lambda^4 + \cdots]^{1/2} \ . \qquad (2.10)$$

In the important case that $\omega = \omega_0$, the series in Eq. (2.10) has a positive radius of convergence. The representation for $\mu(\lambda)$ given in Eq. (2.10) thus remains valid in this case. Contrast this with the series for $\mu(\lambda)$ given in Eq. (2.8): for $\omega = \omega_0$, each term is infinite! Coefficients for the power series of $(\mu - \omega/2)^2$, as well as the first few a, b coefficients, are listed in Appendix A. The a and b coefficients exhibit infinite resonances whenever the applied field frequency ω is an odd subharmonic of the transition frequency ω_0. The existence of such resonances has been demonstrated both theoretically[11-13] and experimentally[8] in connection with optical pumping experiments. We will return to this example in the next section.

B. General remarks

The central idea in the previous example and the features which allow generalization are (a) Floquet's theorem introduces a characteristic constant $\mu(\lambda)$ for the system, (b) periodic functions $\phi^\pm(\lambda, t)$ with the period of the Hamiltonian from which the general solution can be constructed, and (c) there is a systematic procedure for calculating these quantities to each order in λ. In the general case of an arbitrary but periodic Hamiltonian these three features persist.

If the Hamiltonian $H(t)$ is periodic with period τ the solution of Schrödinger's equation for the time evolution operator $U(t)$ can be written as the product of a periodic operator $P(t)$ with period τ and an exponential operator $e^{i\mu t}$, where μ is a time-independent operator, i.e.,

$$U(t) = P(t)e^{-i\mu t} \ . \qquad (2.11)$$

In addition μ must be Hermitian and $P(t)$ unitary as well as periodic. These properties follow from only the assumption that the time evolution operator of Schrödinger's equation is unitary for all time. To prove this we use the spectral theorem for unitary operators,[23] i.e., there exists a Hermitian operator μ such that $U(t=\tau) = e^{-i\mu t}$. We define a new operator $P(t) = U(t)e^{i\mu t}$ and note that, because $\mu = \mu^+$ and $U(t)$ is unitary, $P(t)$ must be unitary. In addition we can conclude that the operator $P(t)$ is periodic with period τ. In order to verify this, set $V(t) = U(t+\tau)$. Since by assumption $H(t+\tau) = H(t)$, it follows that $V(t)$ must be a solution to Schrödinger's equation with the initial condition $V(t=0) = U(\tau)$. Next set $W(t) = U(t)U(\tau)$. By operating with $U(\tau)$ from the left on both sides of Schrödinger's equation for $U(t)$ it follows that $W(t)$ is also a solution with the initial condition $W(t=0) = U(\tau)$. From the uniqueness of the solution we must then have $W(t) = V(t)$ or $U(t+\tau) = U(t)U(\tau)$. From the definition of the operator $P(t)$ we have

$$P(t+\tau) = U(t+\tau)e^{i(t+\tau)\mu} \ .$$

Using the previous result we obtain

$$P(t+\tau) = U(t)U(\tau)e^{i(t+\tau)\mu} \ .$$

Because $U(\tau) = e^{-i\mu\tau}$ the above two time-independent factors cancel leaving $U(t)e^{i\mu t}$ which is just $P(t)$. Thus $P(t+\tau) = P(t)$ completing the proof.

Before we leave the proof, we remark that the order of the factors on the right of Eq. (2.11) is important. If these factors are reversed, the resulting $P(t)$ will not be periodic unless $U(\tau)$ commutes with $U(t)$ for all t.

Notice that in a sense μ is an effective constant Hamiltonian for the system. More precisely if the time scale for secular changes is large compared to the period τ, the time-averaged state vector, averaged over a period τ, evolves as if μ were the Hamiltonian.

C. Second example

As an illustration of the idea of μ playing the role of an effective constant Hamiltonian for time periodic systems, consider the following problem. Suppose that the Hamiltonian for a system can be written as the sum of a time-independent part H_0 and a time-dependent part $H_1 f(t)$, where H_1 is constant and $f(t)$ is shown in Fig. 1:

$$H(t) = H_0 + H_1 f(t) \ . \tag{2.12}$$

For the time range $(0, \tau_0)$ the Hamiltonian is H_0 and the time evolution operator is

$$U(t) = e^{-itH_0/\hbar}, \quad 0 < t < \tau_0 \ . \tag{2.13}$$

In the time range (τ_0, τ) the Hamiltonian is $H_0 + H_1$. In this time range the time evolution operator is the product of the exponential $e^{-it(H_0+H_1)/\hbar}$ and a constant operator. This constant operator is determined by the condition that $U(t)$ must be continuous at $t = \tau_0$. Thus

$$U(t) = e^{-it(H_0+H_1)/\hbar} e^{i\tau_0 H_1/\hbar}, \quad \tau_0 < t < \tau \ . \tag{2.14}$$

But by Floquet's theorem the time evolution operator $U(t)$ can also be written as $P(t)e^{-i\mu t}$, where μ is a constant Hermitian operator and $P(t)$ is a unitary periodic operator with period τ. The periodicity of $P(t)$ can be used to obtain μ. In order to do this first note that $P(0) = I$, the identity operator, so $P(\tau) = I$. Then by making use of the Floquet form expressed in Eq. (2.11) it follows that $U(\tau) = e^{-i\mu\tau}$. But $U(\tau)$ is known from Eq. (2.14) and therefore

$$\begin{aligned} e^{-i\mu\tau} &= e^{-i\tau(H_0+H_1)/\hbar} e^{i\tau_0 H_1/\hbar}, \\ \mu &= (i/\tau)\ln(e^{-i\tau(H_0+H_1)/\hbar} e^{i\tau_0 H_1/\hbar}). \end{aligned} \tag{2.15}$$

In the special case that H_0 and H_1 commute,

$$\mu = H_0/\hbar + (1 - \tau_0/\tau)(H_1/\hbar). \tag{2.16}$$

The determination of μ is connected with the determination of $P(t)$ in the following way. Since $P(t)$ is periodic in time with period τ Eq. (2.11) implies that only a knowledge of $U(t)$ over one cycle is necessary to determine $P(t)$ for all time. Of course the truth of this last statement also relies on the fact that μ has already been calculated. Because of Eq. (2.11) $P(t)$ can be written as the

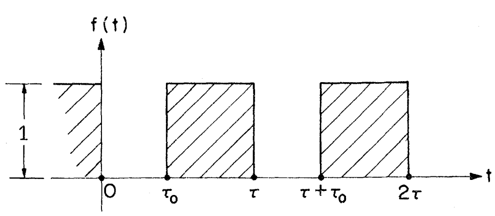

FIG. 1. The Periodic function $f(t)$ [see Eq. (2.12)].

product $U(t)e^{i\mu t}$, and since μ is known from Eq. (2.16) above this determines the exponential factor. Also Eq. (2.13) yields the expression for $U(t)$ in the time range $(0, \tau_0)$ and Eq. (2.14) yields the corresponding result for the range (τ_0, τ). In this way we obtain

$$P(t) = \exp\left(\frac{it}{\tau}(\tau - \tau_0)\frac{H_1}{\hbar}\right), \quad 0 < t < \tau_0, \tag{2.17}$$

$$P(t) = \exp\left(\frac{i\tau_0}{\tau}(\tau - t)\frac{H_1}{\hbar}\right), \quad \tau_0 < t < \tau \ . \tag{2.18}$$

Notice that the constant operator μ of Eq. (2.16) appears as a weighted average between the two constant parts H_0 and H_1 of the original Hamiltonian. While this feature applies only to this problem it does serve to emphasize the idea that μ acts like an effective constant Hamiltonian for periodic systems. Also it is interesting and worthwhile to note how with the aid of Floquet's theorem, $U(t)$, the time evolution operator, has been determined for all values of time through only a knowledge of its value over one time cycle. Actually this feature is always present for any time periodic Hamiltonian. Suppose $U(t)$ is known on the interval $(0, \tau)$, where τ is the period of the given Hamiltonian. Since $U(\tau) = e^{-i\mu\tau}$ this determines μ at least in principle and from the Floquet form Eq. (2.11) we can determine $P(t)$ on the interval $(0, \tau)$. This completes the determination of $P(t)$ because of the periodicity requirement on this operator and hence $U(t)$ has also been determined for all values of time.

D. Perturbation theory

Previously reference was made to three general features concerning systems with time periodic Hamiltonians. Two of these have been introduced and discussed briefly with respect to the periodic square-wave Hamiltonian. The first was related to the appearance of μ, the effective constant Hamiltonian for time periodic systems, and the second feature had to do with the existence of the periodic operator $P(t)$ for such systems. From a knowledge of these two operators it turned out that one could determine the time evolution operator $U(t)$ for the system. The third feature connected with time periodic Hamiltonians deals with the question of determining the two operators μ and $P(t)$ of Floquet's theorem through an appropriate version of perturbation theory.

Suppose that the given Hamiltonian can be written as the sum of a time-independent part H_0 and a time-dependent part $\lambda H_1(t)$:

$$H(t) = H_0 + \lambda H_1(t) \ . \tag{2.19}$$

It is assumed also that the operator $H_1(t)$ is periodic in time with period τ and λ is a coupling parameter. With the Floquet solution in mind the natural perturbation approach is to expand the operator μ into a time-independent power series in λ and to expand the periodic operator $P(t)$ into a time-dependent power series in λ, i.e.,

$$\mu = \frac{H_0}{\hbar} + \sum_{n=1}^{\infty} \lambda^n \mu_n . \tag{2.20}$$

$$P(t) = I + \sum_{m=1}^{\infty} \lambda^m P_m(t). \tag{2.21}$$

Notice that the leading term in the series for μ is proportional to the constant part of the Hamiltonian H_0 while the corresponding term for $P(t)$ is the unit operator. When such expansions exist the following three equations form a set of recursion relations from which the operators μ_n and $P_n(t)$ can be obtained:

$$\Gamma_n(t) = H_1(t) P_{n-1}(t) - \sum_{m=1}^{n} \hbar P_{n-m}(t) \mu_m ,$$

$$P_0(t) = I , \tag{2.22}$$

$$i\hbar P_n(t) = \int_0^t dt'\, e^{-(i/\hbar)(t-t')H_0} \Gamma_n(t')\, e^{(i/\hbar)(t-t')H_0} , \tag{2.23}$$

$$0 = P_n(0) = P_n(\tau), \quad n = 1, 2, \ldots . \tag{2.24}$$

The recursion calculation begins with a determination of μ_1. Setting $n = 1$ in (2.22) and (2.23), we have

$$\Gamma_1(t) = H_1(t) - \mu_1 , \tag{2.25}$$

and

$$i\hbar P_1(t) = \int_0^t dt'\, e^{-(i/\hbar)(t-t')H_0} \Gamma_1(t')\, e^{(i/\hbar)(t-t')H_0} . \tag{2.26}$$

The operator μ_1 is determined by (2.26) and by the boundary condition, $P_1(\tau) = 0$, given in (2.24). An application of this boundary condition to (2.26), followed by cancellations of exponentials on the right and left of the resulting equation, gives

$$0 = \int_0^\tau dt'\, e^{(i/\hbar)t'H_0} \Gamma_1(t') e^{-it'H_0/\hbar} . \tag{2.27}$$

Substitution of (2.25) into (2.27) finally yields

$$\frac{1}{\hbar} \int_0^\tau dt'\, e^{(it'/\hbar)H_0} H_1(t') e^{-it'H_0/\hbar} = \int_0^\tau dt'\, e^{it'H_0/\hbar} \mu_1 e^{-it'H_0/\hbar} . \tag{2.28}$$

It is this last equation which must be inverted to obtain μ_1. If it is possible to carry this out, then (2.25) and (2.26) determine $\Gamma_1(t)$ and $P_1(t)$, respectively. To obtain the next order, set $n = 2$ in (2.22). For $\Gamma_2(t)$ the result is

$$\Gamma_2(t) = H_1(t) P_1(t) - P_1(t) \mu_1 - \mu_2 , \tag{2.29}$$

while $P_2(t)$ is obtained by setting $n = 2$ in (2.23). Repeating the previous argument, μ_2 is determined by

$$\frac{1}{\hbar} \int_0^\tau dt'\, e^{it'H_0/\hbar} [H_1(t') P_1(t') - P_1(t') \mu_1] e^{-(it'/\hbar)} H_0 = \int_0^\tau dt'\, e^{it'H_0/\hbar} \mu_2 e^{-it'H_0/\hbar} . \tag{2.30}$$

Again, assuming this equation can be solved for μ_2, both $P_2(t)$ and $\Gamma_2(t)$ are determined. The general case, which is a bit messy, is quite similar: At each step an equation of the form,

$$B = \int_0^1 e^{iCs} G e^{-iCs}\, ds, \tag{2.31}$$

where B and C are known operators, is to be solved for the unknown operator G. The operator C is assumed self-adjoint.

Under the assumption that the spectrum of C is contained entirely in an interval of length less than 2π, Eq. (2.31) has the solution

$$G = \int_{-\infty}^{\infty} g(\alpha) \exp(\tfrac{1}{2} c(\alpha - i)) B \times \exp(-\tfrac{1}{2} c(\alpha - i))\, d\alpha , \tag{2.32}$$

where

$$g(\alpha) = \pi e^{\pi\alpha} (1 + e^{\pi\alpha})^{-2} . \tag{2.33}$$

To establish (2.32), first substitute the integral given there into (2.31), apply the spectral theorem to the self-adjoint operator C, and interchange the integrals in the resultant equation. We have

$$\tilde{B} = \int_{\mathrm{sp}(C)} dE(x) B \int_{\mathrm{sp}(C)} dE(y) \int_{-\infty}^{\infty} d\alpha\, g(\alpha) \times \int_0^1 ds \exp\left(\frac{x-y}{2} (\alpha - i + 2is)\right), \tag{2.34}$$

where $dE(\cdot)$ is the spectral resolution of C and $\mathrm{sp}(C)$ is the spectrum of C. We want to prove that $\tilde{B} = B$. Note that the integral over s in (2.34) has the value

$$\int_0^1 (\cdot)\, ds = \exp(\tfrac{1}{2}(x-y)\alpha) \frac{\sin(\tfrac{1}{2}(x-y))}{(\tfrac{1}{2}(x-y))} , \tag{2.35}$$

while standard contour integration shows that

$$\int_{-\infty}^{\infty} g(\alpha) e^{\alpha\beta}\, d\alpha = \frac{\beta}{\sin\beta} , \tag{2.36}$$

if $-\pi < \beta < \pi$. Since the spectrum of C is assumed to be contained in an interval of length less than

2π, it is clear that

$$|\tfrac{1}{2}(x-y)|<\pi ,$$

and, hence, (2.36) is applicable. Combining (2.34), (2.35), and (2.36) gives

$$\tilde{B}=\int_{\text{sp}(C)} dE(x)B\int_{\text{sp}(C)} dE(y) . \tag{2.37}$$

However, both integrals are equal to the identity operator because $dE(\cdot)$, as a spectral resolution, is also a resolution of the identity. Thus $\tilde{B}=B$.

Several remarks are in order. Note that no assumptions were made on the spectrum of C, other than that it must be contained in an interval of length less than 2π. In particular, the spectrum may contain both discrete and continuous parts. Finally, even when the spectrum does not satisfy the length restriction, the operator C may be scaled and a solution to (2.31) can then be obtained from (2.32). Such a solution is often valid even when the scale factor is removed.

Let us now focus attention on the properties of μ_n, $P_n(t)$, and the series defined by these operators.

Because of the boundary condition (2.24), it is not obvious that expansions for μ and $P(t)$, when derived from a solution of the recurrence relations (2.22)–(2.24), give the appropriate Floquet solution to Schrödinger's equation with the Hamiltonian given in Eq. (2.19). To see that this is the case if we assume the recurrence relations (2.22)–(2.24) have a solution, differentiate Eq. (2.23) with respect to time, eliminate $\Gamma_n(t)$ by means of Eq. (2.22), multiply the resultant equation by λ^n, sum from $n=1$ to ∞, and use the expansion for $P(t)$ given in Eq. (2.21). The result is

$$i\hbar\frac{\partial P}{\partial t}+[P(t),H_0]=\lambda H_1(t)P(t)-\sum_{n=1}^{\infty}\sum_{m=1}^{n}\lambda^n\hbar P_{n-m}(t)\mu_m .$$

The double sum on the right is the product, $P(t)\hbar(\mu-\mu_0)$. This follows from the general form of the product of two power series together with Eqs. (2.20) and (2.21). The equation for $P(t)$ is thus

$$i\hbar\dot{P}(t)+P(t)\hbar\mu=[H_0+\lambda H_1(t)]P(t) .$$

Operating from the right with $e^{-i\mu t}$ gives,

$$i\hbar\frac{\partial}{\partial t}P(t)e^{-i\mu t}=[H_0+\lambda H_1(t)]P(t) ,$$

which is Schrödinger's equation. But is $P(t)$ periodic? By Eqs. (2.24) and (2.21), $P(\tau)=P(0)=1$. A repetition of the argument used to prove Floquet's theorem in Sec. II B then implies that $P(t)$ is periodic. The periodicity can also be obtained from the recurrence relations. The operators $P_n(t)$ are first shown to be periodic and, by (2.21), it follows that $P(t)$ is also periodic.

III. STATIONARY TWO-STATE ATOMS IN LINEARLY AND CIRCULARLY POLARIZED FIELDS

In this section we consider in detail the problem of a stationary two-state dipole in a linearly polarized field. The time evolution operator $U(t)$ is calculated using the perturbation theory associated with Floquet's theorem. From this operator we then obtain wave functions and transition probabilities for the system. As noted previously the expansions for these quantities exhibit resonances whenever the applied field frequency is an odd subharmonic of the atomic transition frequency. Alternative forms of these expansions are presented which suggest that these resonances can be interpreted as branch points of certain functions. This interpretation is supported by a comparison of (i) the expression for the frequency with which the atom alternates between the upper and lower states and (ii) the expression for the transition probability with similar quantities calculated by Bloch and Siegert.[7]

Consider a stationary two-state neutral system interacting with a monochromatic field that can be either circularly or linearly polarized and detuned from the transition frequency ω_0. The Hamiltonian describing this situation is

$$H(t)=\tfrac{1}{2}\hbar\omega_0\sigma_z+\hbar\omega_0\lambda\sigma_x f(t) , \tag{3.1}$$

where λ is a dimensionless coupling parameter which measures the ratio of the oscillating field component to the constant z-directed component, and

$$f(t)=\tfrac{1}{2}\sin\omega t,\quad \text{linear polarization,}$$
$$f(t)=-e^{i\omega t\sigma_z},\quad \text{circular polarization.} \tag{3.2}$$

A. Linear polarization

Following the method outlined in Sec. II, the perturbation calculation begins with the determination of μ_1. Putting $s=t/\tau$ in Eq. (2.28) gives

$$\frac{1}{\hbar}\int_0^1 ds\, e^{i\tau sH_0/\hbar}H_1(\tau s)e^{-i\tau sH_0/\hbar}=\int_0^1 ds\, e^{i\tau sH_0/\hbar}\mu_1 e^{-i\tau sH_0/\hbar} , \tag{3.3}$$

where H_0 and $H_1(t)$ are defined by Eqs. (3.1) and (3.2). In this example, $\tau=2\pi/\omega$. Equation (3.3) can be cast into the form of Eq. (2.31) by setting

$$C=\tfrac{1}{2}\omega_0\tau\sigma_z$$

and

$$B=\frac{1}{\hbar}\int_0^1 ds\, e^{i\tau s H_0/\hbar}H_1(\tau s)e^{-i\tau s H_0/\hbar}$$
$$=\frac{\omega_0\pi/\tau^2}{\omega_0^2-\omega^2}(e^{i\omega_0\tau\sigma_z}-I)\sigma_x \quad . \qquad (3.4)$$

An application of Eq. (2.32) yields

$$\mu_1=[i\omega\omega_0^2/2(\omega^2-\omega_0^2)]\sigma_x\sigma_z \quad , \qquad (3.5)$$

which is valid for $\omega_0<\omega$. Alternately, Eq. (3.5) can be thought of as an ansatz for μ_1. From Eq. (3.3) and the commutation rules for the Pauli spin matrices, we then obtain that Eq. (3.5) is valid under the condition that ω_0 is not an integer multiple of ω. This is the only restriction necessary to invert Eq. (2.31). Substituting Eq. (3.5) into Eq. (2.22) we have

$$\Gamma_1(t)=\tfrac{1}{2}\hbar\,\omega_0\sigma_x\sin\omega t-\frac{\hbar i\omega\omega_0^2/2}{\omega^2-\omega_0^2}\sigma_x\sigma_z \quad . \qquad (3.6)$$

With the determination of $\Gamma_1(t)$ the periodic operator $P_1(t)$ can be evaluated by substituting Eq. (3.6) into Eq. (2.23). One finds

$$P_1(t)=\frac{\sigma_x}{2i}\int_0^t dt'\, e^{i(t-t')\omega_0\sigma_z}\times\left(\omega_0\sin\omega t'-\frac{i\omega\omega_0^2}{\omega^2-\omega_0^2}\sigma_z\right) ,$$
$$=\frac{i}{4}\omega_0\sigma_x\left(-\frac{2\omega}{\omega^2-\omega_0^2}+\frac{e^{i\omega t\sigma_z}}{\omega-\omega_0}+\frac{e^{-i\omega t\sigma_z}}{\omega+\omega_0}\right) . \qquad (3.7)$$

Notice that the initial conditon for $P_1(t)$ is correct since $P_1(t=0)=0$. Next observe that $P_1(t)$ of Eq. (3.7) is also periodic with period τ.

The higher-order coefficients μ_n and $P_n(t)$ are obtained by the same procedure. In this case the form for μ_n is $\nu_n\sigma_x^n\sigma_z$, where ν_n is a scalar which is determined from the condition that $P_n(\tau)=0$. It is assumed that all lower-order coefficients have been evaluated so that each term of $\Gamma_n(t)$ is known except for the μ_n term. Once μ_n has been calculated $\Gamma_n(t)$ can then be determined from Eq. (2.22). Finally, $P_n(t)$ is obtained using $\Gamma_n(t)$ together with Eq. (2.23). When this procedure is carried out for each order n one finds that μ and $P(t)$ have the following general forms

$$\mu=\sum_{n=0}^{\infty}\lambda^n\sigma_x^n\nu_n\sigma_z \quad ,$$
$$P(t)=\sum_{n=0}^{\infty}\lambda^n\sigma_x^n\sum_{m=0}^{n}(A_{nm}e^{im\omega t\sigma_z}+B_{nm}e^{-im\omega t\sigma_z}) . \qquad (3.8)$$

The coefficients ν_n, A_{nm}, and B_{nm} are listed in Appendix B for $n\leqslant 4$. They exhibit infinite resonances whenever ω is an odd subharmonic of ω_0. Also exhibiting this resonant property are the scalar coefficients ν_n of Eq. (3.8) which serve to define the μ expansion. Of course the reason for these infinite resonances is that for these values of ω the solution is not analytic at $\lambda=0$.

B. Circular polarization

In the case of circular polarization closed-form expressions for μ and $P(t)$ can be found. In particular,

$$\mu=(1+\omega/D)(\tfrac{1}{2}\Delta\sigma_z-\omega_0\lambda\sigma_x) ,$$
$$P(t)=I+(I-e^{-i\omega t\sigma_z})\left[\frac{\omega_0\lambda}{iD}\sigma_y+\frac{1}{2}\left(\frac{\Delta}{D}-1\right)\right] , \qquad (3.9)$$

where $\Delta=\omega_0-\omega$ and $D=(\Delta^2+4\omega_0^2\lambda^2)^{1/2}$. The solution for $U(t)$ most often quoted is

$$U(t)=e^{-(i/2)\omega t\sigma_z}\exp\{-it[\tfrac{1}{2}(\omega_0-\omega)\sigma_z-\omega_0\lambda\sigma_x]\}. \qquad (3.10)$$

It can be shown that the Floquet solution $P(t)e^{-i\mu t}$ is equivalent to this familiar result.[24,25] The importance of form (3.10) for the time evolution operator is that it enables us to readily calculate the induced transition probability. By expanding the exponential of Eq. (3.10) into trigonometric functions and by taking matrix elements we obtain for the upper-state wave function $\psi_1(t)$

$$e^{(i/2)\omega t}\psi_1(t)=\left[\cos\left(\frac{t}{2}D\right)+\frac{\Delta}{iD}\sin\left(\frac{t}{2}D\right)\right]\psi_1(0)+\frac{2i\omega_0\lambda}{D}\sin\left(\frac{t}{2}D\right)\psi_2(0) ,$$

where $\Delta=\omega_0-\omega$ and $D=(\Delta^2+4\omega_0^2\lambda^2)^{1/2}$. $W(t)$, the transition probability, is obtained from this last equation by setting $\psi_1(0)=0$ and calculating $|\psi_1(t)|^2$. The result is

$$W(t)=\left(\frac{4\omega_0^2\lambda^2}{(\omega_0-\omega)^2+4\omega_0^2\lambda^2}\right)\times\sin^2\left(\frac{t}{2}[(\omega_0-\omega)^2+4\omega_0^2\lambda^2]^{1/2}\right). \qquad (3.11)$$

The same expression for the transition probability can be derived using the Floquet form for $U(t)$. While rederiving Eq. (3.11) from this point of view requires more work than the previous method, it is important in that it will act as a guide in reducing the more complicated expression for linear polarization to something similar to Eq. (3.11).

C. Floquet operators μ, $e^{-i\mu t}$, and $P(t)$

A very important operator in this calculation for both the circularly and linearly polarized cases is the exponential $e^{-i\mu t}$. The trigonometric expansion of this ultimately gives rise to the slowly varying part of the transition probability. It is obtained using the expansion

$$e^{-i\mu t} = \cos(\Omega)t - (i\mu/\Omega)\sin(\Omega t) , \qquad (3.12)$$

valid for any operator μ that has associated with it a scalar Ω with the property that $\mu^2 = \Omega^2 I$. For the case of circular polarization

$$\Omega = \tfrac{1}{2}(\omega + D) . \qquad (3.13)$$

The corresponding expression for Ω in the linearly polarized case has the same form as that given by Eq. (3.13) except that D is replaced by

$$D = \left((\omega_0 - \omega)^2 + \frac{\omega_0^3 \lambda^2}{2(\omega + \omega_0)} - \frac{\omega_0^5 \lambda^4}{32(\omega + \omega_0)^3} + \cdots\right)^{1/2} . \qquad (3.14)$$

In view of the infinite resonances (whenever ω is an odd subharmonic of ω_0) that are exhibited by the ν_n coefficients that define the operator μ, Eq. (3.14) is rather remarkable for it implies that none of these resonances are present in μ^2. The absence of infinite resonances in the expression for D is consistent with the interpretation of $D/2$ as the frequency of the slowly varying part of the induced transition probability. The basis for this interpretation will become clear with the determination of the expression for the induced-transition probability. The derivation of Eq. (3.14) can be found in Appendix C.

An alternative expression for the frequency D given by Eq. (3.14) can be obtained by expanding the coefficients of λ^2 and λ^4 about the frequency difference $\omega_0 - \omega$. This yields

$$D = \left\{\left[\left(1 + \frac{\lambda^2}{16} - \frac{3\lambda^4}{16.64} + \cdots\right)\omega_0 - \omega\right]^2 + \left(\frac{\omega_0}{2}\lambda\right)^2\left(1 - \frac{\lambda^2}{32} + \cdots\right) + \cdots\right\}^{1/2} . \qquad (3.15)$$

A check of this result can now be obtained by comparing $D/2$ above with the frequency of the slowly varying part of the transition probability for linear polarization as calculated by Bloch and Siegert. For this frequency they obtain

$$B = \frac{\mu H_1^*}{\hbar}\left[1 + \left(\frac{H_0 - H_r^*}{H_1^*}\right)^2\right]^{1/2} ,$$

$$H_1^* = \tfrac{1}{2}H_1, \quad \text{and } H_r^* \approx H_r(1 - H_1^2/16H_r^2), \qquad (3.16a)$$

where H_1 denotes the amplitude of the oscillating field taken to be in the x direction, and H_r denotes the resonance value of the constant z-directed field H_0. In our notation

$$\mu H_1 = \tfrac{1}{2}\hbar\omega_0\lambda, \quad \mu H_0 = \tfrac{1}{2}\hbar\omega_0, \quad \mu H_r = \tfrac{1}{2}\hbar\omega . \qquad (3.17)$$

A substitution of Eq. (3.17) into Eq. (3.16a) yields ($\omega \approx \omega_0$)

$$B \approx \tfrac{1}{2}\{[(1 + \tfrac{1}{16}\lambda^2)\omega_0 - \omega]^2 + (\tfrac{1}{2}\omega_0\lambda)^2\}^{1/2} . \qquad (3.16b)$$

A comparison of this result with the frequency $D/2$ of Eq. (3.15) reveals an agreement to terms of the order λ^2. However, since the calculation of Bloch and Siegert included only λ^2 dependence no higher-order comparison could be made.

Returning to the calculation of induced transition probability for the fixed dipole, and in particular to the expansion of the exponential $e^{-i\mu t}$, Eqs. (3.12), (3.13), and (3.14) imply the following:

$$\begin{aligned} &e^{-i\mu t} = \cos\tfrac{1}{2}t(\omega + D) - [i\mu/\tfrac{1}{2}(\omega + D)]\sin\tfrac{1}{2}t(\omega + D), \\ &D = \left((\omega_0 - \omega)^2 + \frac{\omega_0^3\lambda^2}{2(\omega_0 + \omega)} - \frac{\omega_0^5\lambda^4}{32(\omega_0 + \omega)^3} + \cdots\right)^{1/2}, \quad \text{(linear)} \\ &D = [(\omega_0 - \omega)^2 + 4\omega_0^2\lambda^2]^{1/2}, \quad \text{(circular)}. \end{aligned} \qquad (3.18)$$

The exponential $e^{-i\mu t}$ is not the only quantity that exhibits a similarity of form between the linearly and circularly polarized cases. We find that even μ has a form common to both cases. In particular

$$\mu = \tfrac{1}{2}(\omega + D)\left(\frac{a}{D}\sigma_z - \frac{b\lambda}{D}\sigma\right)$$

where

$$\left.\begin{aligned} &a = \omega_0 - \omega + \frac{(\omega_0\lambda)^2}{4(\omega + \omega_0)^2}\left(2\omega_0 + \omega - \frac{3(\omega_0\lambda)^2(8\omega^3 - 3\omega^2\omega_0 - 4\omega\omega_0^2 - \omega_0^3)}{16(\omega + \omega_0)^2(9\omega^2 - \omega_0^2)} + \cdots\right), \\ &b = \frac{\omega\omega_0}{\omega + \omega_0}\left(1 - \frac{(\omega_0\lambda)^2(6\omega^2 + 3\omega\omega_0 + \omega_0^2)}{4(\omega + \omega_0)^2(9\omega^2 - \omega_0^2)} + \cdots\right), \end{aligned}\right\} \text{(linear)}$$

$$\sigma = \sigma_y \qquad (3.19)$$

$$\left.\begin{aligned} &a = \omega_0 - \omega, \\ &b = 2\omega_0, \\ &\sigma = \sigma_x. \end{aligned}\right\} \text{(circular)}.$$

It is true that Eq. (3.19) for μ simply represents a replacement of the operator series of Eq. (3.8) with the $a(\lambda)$ and $b(\lambda)$ series. However, we can see that Eq. (3.19) contains no infinite resonances at $\omega=\omega_0$ up to and including the λ^4 terms. This feature by itself makes form (3.19) for μ preferable to form (3.8). The functions $a(\lambda)$ and $b(\lambda)$ for the linear cases were determined by equating expressions (3.8) and (3.19) for the operator μ.

A rather important feature of μ in the linear case is the dependence of the radius of convergence on ω and ω_0. From Eq. (3.19) we can see that at $3\omega=\omega_0$ the radius of convergence for both of the series defining $a(\lambda)$ and $b(\lambda)$ is zero. One possible explanation for this resonant behavior is that of a branch point at $3\omega=\omega_0$. Two functions $a(\lambda)$ and $b(\lambda)$ consistent with this interpretation and having the proper expansions can be constructed.

The determination of μ simplifies the calculation of $e^{-i\mu t}$. Equation (3.18) can be written as

$$(\omega+D)e^{-i\mu t}=[\tfrac{1}{2}(\omega+D)-\mu\sigma_z]e^{(it/2)(\omega+D)\sigma_z}+[\tfrac{1}{2}(\omega+D)-\mu\sigma_z]e^{-(it/2)(\omega+D)\sigma_z}.$$

By substituting Eq. (3.19) into this equation we get

$$\begin{aligned}e^{-i\mu t}&=(\Phi_1-i\lambda\Phi_2\sigma)e^{(it/2)(\omega+D)\sigma_z}+(\Phi_3+i\lambda\Phi_2\sigma)e^{-(it/2)(\omega+D)\sigma_z};\\ \Phi_1&=\tfrac{1}{2}(1-a/D),\\ \Phi_2&=-b/2D,\\ \Phi_3&=\tfrac{1}{2}(1+a/D).\end{aligned}\tag{3.20}$$

As in the case of μ, the exponential $e^{-i\mu t}$ also contains no infinite resonances at $\omega=\omega_0$ up to and including λ^4 terms.

The next step on the way to determining the transition probability $W(t)$ is the calculation of the periodic operator $P(t)$. Since the expression for $P(t)$ in the linear case involves an infinite harmonic series the much simpler and closed form circular-case expression for $P(t)$ given by Eq. (3.9) will be examined first. It is expected that this expression can be used as a guide to tell us what harmonics to retain and what resonant denominators are present in the linear case. From Eq. (3.9) we have

$$P(t)=\theta_1+i\lambda\theta_2\sigma_y+e^{-i\omega t\sigma_z}(\theta_3+i\lambda\theta_4\sigma_y),$$

where

$$\theta_2=-\theta_4=\omega_0/D\tag{3.21}$$

and

$$\begin{aligned}\theta_3&=\tfrac{1}{2}(1-\Delta/D)=\frac{2(\omega_0\lambda)^2}{D(D+\Delta)},\\ \theta_1&=\tfrac{1}{2}(1+\Delta/D)=1-\frac{2(\omega_0\lambda)^2}{D(D+\Delta)}.\end{aligned}\tag{3.22}$$

Equation (3.22) reveals that the λ^2 terms of the expansion of θ_1 or θ_3 would contain double pole at $\omega=\omega_0$ due to the presence of the denominator $D(D+\Delta)$. It will be seen shortly that a similar resonant behavior exists in the corresponding terms of the linear-case expansion for $P(t)$ and that the introduction of $D(D+\Delta)$ as a denominator for these terms suggests that the resonances at $\omega=\omega_0$ and $3\omega=\omega_0$ should be interpreted as branch points. Based on Eq. (3.8) we can write $P(t)$ for the linear case as follows:

$$P(t)=\theta_1+i\lambda\theta_2\sigma_x+e^{-i\omega t\sigma_z}(\theta_3+i\lambda\theta_4\sigma_x)+\cdots,$$

where

$$\begin{aligned}\theta_1&=\frac{1}{2}+\frac{1}{2D}\left(\Delta+\frac{\omega\omega_0(\omega_0\lambda)^2}{4\Delta(\omega+\omega_0)^2}+\cdots\right)\\&\quad-\frac{(\omega_0\lambda)^2}{8D(D+\Delta)(\omega+\omega_0)^2}\left(3\omega^2-\omega_0^2-\frac{(\omega_0\lambda)^2(-\omega^5+71\omega^4\omega_0+82\omega^3\omega_0^2+58\omega^2\omega_0^3-\omega\omega_0^4-\omega_0^5)}{64\omega^2(\omega+\omega_0)^2\Delta}\right),\\ \theta_2&=\frac{\omega\omega_0}{2(\omega+\omega_0)D}\left(1-\frac{(\omega_0\lambda)^2(-15\omega^3+21\omega^2\omega_0+7\omega\omega_0^2+3\omega_0^3)}{16\Delta(\omega+\omega_0)^2(9\omega^2-\omega_0^2)}+\cdots\right),\\ \theta_3&=\frac{\omega(\omega_0\lambda)^2}{4(\omega+\omega_0)D(D+\Delta)}\left(1-\frac{(\omega_0\lambda)^2(-21\omega^4+60\omega^3\omega_0+22\omega^2\omega_0^2+4\omega\omega_0^3-\omega_0^4)}{32\omega\Delta(\omega+\omega_0)^2(9\omega^2-\omega_0^2)}+\cdots\right),\\ \theta_4&=-\frac{\omega_0}{4D}\left(1-\frac{(\omega_0\lambda)^2(\omega_0^2-5\omega^2)}{32\omega\Delta(\omega+\omega_0)^2}+\cdots\right).\end{aligned}\tag{3.23}$$

In Eq. (3.23) notice that $\omega=\omega_0$ and $3\omega=\omega_0$ appear as single-order poles.

The expressions given below in (3.24) are constructed by analogy with the circularly polarized case. They agree with those given in (3.23) to order λ^4 and make explicit the desired branch-point behavior. This feature suggests that these resonances could be interpreted as branch points. Using partial fraction decompositions we have

$$
\begin{aligned}
\theta_1 &= \frac{1}{2} + \frac{1}{2D}\left((\omega_0-\omega)^2 + \frac{\omega\omega_0(\omega_0\lambda)^2}{2(\omega+\omega_0)^2} + \cdots\right)^{1/2} \\
&\quad + \frac{(\omega_0\lambda)^2}{8D(D+\Delta)(\omega+\omega_0)}\left((\omega_0-\omega)^2 + \frac{(5\omega\omega_0^4+8\omega_0^5)(\omega_0\lambda)^2}{2\omega^2(\omega+\omega_0)^3} + \cdots\right)^{1/2} \\
&\quad - \frac{(\omega_0\lambda)^2}{8D(D+\Delta)(\omega+\omega_0)^2}\left(2\omega^2 + \frac{(\omega_0\lambda)^2(-\omega^3+71\omega^2\omega_0+81\omega\omega_0^2+129\omega_0^3)}{64\omega^2(\omega+\omega_0)} + \cdots\right), \\
\theta_2 &= \frac{\omega\omega_0}{2(\omega+\omega_0)^2(3\omega+\omega_0)D}\left[3\omega^2+8\omega\omega_0+\omega_0^2+\tfrac{1}{16}(\omega_0\lambda)^2\right] \\
&\quad - \frac{\omega\omega_0}{2(\omega+\omega_0)D}\left[\frac{1}{(\omega+\omega_0)}\left((\omega_0-\omega)^2 + \frac{\omega\omega_0(\omega_0\lambda)^2}{(\omega+\omega_0)(3\omega+\omega_0)} + \cdots\right)^{1/2}\right. \\
&\qquad \left. + \frac{1}{(3\omega+\omega_0)}\left((3\omega-\omega_0)^2 + \frac{(9\omega^2+10\omega\omega_0+\omega_0^2)(\omega_0\lambda)^2}{4(\omega+\omega_0)^2} + \cdots\right)^{1/2}\right], \\
\theta_3 &= \frac{\omega(\omega_0\lambda)^2}{4(\omega+\omega_0)^2(3\omega+\omega_0)D(D+\Delta)}\left(3\omega^2+8\omega\omega_0+\omega_0^2 - \frac{(3\omega+\omega)}{32\omega}(\omega_0\lambda)^2\right) \\
&\quad - \frac{\omega(\omega_0\lambda)^2}{4(\omega+\omega_0)D(D+\Delta)}\left[\frac{1}{(\omega+\omega_0)}\left((\omega_0-\omega)^2 + \frac{\omega_0(\omega_0\lambda)^2}{2(\omega+\omega_0)} + \cdots\right)^{1/2}\right. \\
&\qquad \left. + \frac{1}{3\omega+\omega_0}\left[(3\omega-\omega_0)^2 + \tfrac{3}{8}(\omega_0\lambda)^2 + \cdots + \cdots\right]^{1/2}\right], \\
\theta_4 &= -\frac{\omega_0}{4(\omega+\omega_0)D}\left[2\omega - \frac{(\omega_0\lambda)^2}{32\omega} + \left((\omega_0-\omega)^2 + \frac{\omega(\omega_0\lambda)^2}{4(\omega+\omega_0)} + \cdots\right)^{1/2}\right].
\end{aligned}
\tag{3.24}
$$

This completes the discussion of the resonant behavior of the first few terms of $P(t)$. As was the case for μ and the exponential $e^{-i\mu t}$ an expression for $P(t)$ could be found which was devoid of any infinite resonances at either $\omega=\omega_0$ or $3\omega=\omega_0$.

D. Transition probability for linear polarization

With the determination of both $P(t)$ and the operator $e^{-i\mu t}$ we are now in a position to calculate the time evolution operator for both cases. The product $P(t)^{-i\mu t}$ can be obtained using Eqs. (3.20) and (3.23). With $\psi_1(t=0)=0$ and $|\psi_2(t=0)|^2=1$ wave function $\psi_1(t)$ can be obtained by taking matrix elements of the product $P(t)e^{-i\mu t}$: we obtain

$$
\begin{aligned}
(\omega+D)\psi_1(t)e^{-(i/2)\omega t} &= A\psi_2(0)\lambda(\theta_2\Phi_1-\theta_1\Phi_2)e^{(it/2)D} \\
&\quad + A\psi_2(0)\lambda(\theta_4\Phi_3+\theta_3\Phi_2)e^{-(it/2)D} + \cdots
\end{aligned}
$$

with $A=\langle 2|i\sigma|1\rangle$. Taking the square of the modulus

$$
\begin{aligned}
(\omega+D)^2|\psi_1(t)|^2 &= \lambda^2[\theta_2\Phi_1+\theta_4\Phi_3 \\
&\quad -(\theta_1-\theta_3)\Phi_2]^2\cos^2(\tfrac{1}{2}tD) \\
&\quad + \lambda^2[\theta_2\Phi_1-\theta_4\Phi_3 \\
&\quad -(\theta_1+\theta_3)\Phi_2]^2\sin^2(\tfrac{1}{2}tD) + \cdots .
\end{aligned}
\tag{3.25}
$$

We obtain for the linear-case probability $W(t)$

$$
W(t) = \frac{(\omega_0\lambda)^2}{16(\omega+\omega_0)^2}\left(\frac{\omega_0^2 f_1^2}{(\omega+D)^2}\cos^2(\tfrac{1}{2}tD) + \frac{f_2^2}{D^2}\sin^2(\tfrac{1}{2}tD)\right) + \cdots, \tag{3.26}
$$

where

$$
f_1 = 1 - \frac{(\omega_0\lambda)^2(87\omega^4+30\omega^3\omega_0+8\omega^2\omega_0^2+2\omega\omega_0^3+\omega_0^4)}{32\omega(\omega+\omega_0)^3(9\omega^2-\omega_0^2)} + \cdots,
$$

$$
f_2 = 3\omega+\omega_0 - \frac{(\omega_0\lambda)^2(105\omega^4+66\omega^3\omega_0+24\omega^2\omega_0^2-2\omega\omega_0^3-\omega_0^4)}{32\omega(\omega+\omega_0)^2(9\omega^2-\omega_0^2)} + \cdots .
$$

For $\omega\approx\omega_0$ the omitted terms in Eq. (3.26) all oscillate with frequencies ω_0 or greater. For $\omega\to\omega_0/3$ the series for $f_{1,2}$ obviously diverges. Possibly $\lambda=0$ is a branch point of $W(t)$ for $\omega=\omega_0/3$ similar to the branch point at $\lambda=0$ for $\omega=\omega_0$.

In an earlier discussion of the frequency term D given by Eq. (3.14) it was noted that Bloch and Siegert also obtained an expression for the transition probability $W(t)$ for the linearly polarized case which involved an equivalent frequency. This calculation was carried out from a different point of view. Agreement to terms of order λ^2 was obtained between the frequency $D/2$ and the equivalent expression presented by Bloch and Siegert. This was accomplished by expanding certain terms of Eq. (3.14) about the frequency difference $\omega_0 - \omega$. The result of this expansion was Eq. (3.15). In order to compare the full expression for $W(t)$ with the Bloch-Siegert result write, for $\omega \approx \omega_0$,

$$W(t) = \frac{(\omega_0\lambda/8)^2(1-\lambda^2/16+\cdots)^2}{(\omega_0+D)^2}\cos^2\left(\frac{t}{2}D\right) + \frac{(\omega_0\lambda/2)^2(1-3\lambda^2/64+\cdots)^2}{D^2}\sin^2\left(\frac{t}{2}D\right)+\cdots,$$

where

$$D = \left\{\left[\left(1+\frac{\lambda^2}{16}-\frac{3\lambda^4}{16.64}+\cdots\right)\omega_0 - \omega\right]^2 + \left(\frac{\omega_0}{2}\lambda\right)^2\left(1-\frac{\lambda^2}{32}\right)+\cdots\right\}^{1/2}. \quad (3.27)$$

For the transition probability Bloch and Siegert give

$$\overline{P}_{-1/2}(t) = \frac{(\frac{1}{2}\omega_0\lambda)^2\sin^2(\frac{1}{2}t\{[(1+\frac{1}{16}\lambda^2)\omega_0-\omega]^2+(\frac{1}{2}\omega_0\lambda)^2\}^{1/2})}{[(1+\frac{1}{16}\lambda^2)\omega_0-\omega]^2+(\frac{1}{2}\omega_0\lambda)^2}. \quad (3.28)$$

Equations (3.27) and (3.28) agree to order λ^2 except for the cosine term of Eq. (3.27). This term is perhaps more properly grouped with the higher-frequency terms in $W(t)$ since it serves to match these terms to the initial condition at $t=0$.

IV. MOVABLE TWO-STATE ATOM IN TWO FIELD CONFIGURATIONS

A neutral atomic system in an optical field nearly resonant with an internal transition periodically exchanges energy with the field. The associated linear momentum transfer alters the motion of the atomic center of mass. This section is concerned with several features of this phenomenon which appears in connection with two models. The first model to be considered is that of a movable two-state atom in a prescribed, single-frequency, standing wave. The Floquet solution to Schrödinger's equation for this model will be compared with the Floquet solution to Schrödinger's equation for the corresponding situation in which a stationary two-state atom interacts with a prescribed, linearly polarized, single-frequency field. (See Sec. III.) The second model, viz., a two-state atom in a prescribed, single-frequency, circularly polarized, plane wave, is exactly soluble. In this case particular emphasis will be given to the modulation of wave packets and to the time dependence of level populations.

For slowly moving atoms we use the Hamiltonian

$$H(t) = p^2/2M + \tfrac{1}{2}\hbar\omega_0\sigma_z - \hbar\omega_0\lambda f(\vec{r},t), \quad (4.1)$$

where M is the atomic mass, ω_0 the internal transition frequency, ω $(=kc)$ is the applied field frequency, and $\vec{k}$ is in the z direction. The coupling constant λ is proportional to the atomic dipole moment and the field strength. We study two special field configurations

$$f(\vec{r},t) = \tfrac{1}{2}\sigma_x \sin\omega t\cos\vec{k}\cdot\vec{r} - \tfrac{1}{2}\sigma_y\sin\omega t\sin\vec{k}\cdot\vec{r}$$

and (4.2)

$$f(\vec{r},t) = \sigma_x\cos(\omega t - \vec{k}\cdot\vec{r}) + \sigma_y\sin(\omega t-\vec{k}\cdot\vec{r}).$$

The center-of-mass position $\vec{r}$ and momentum $\vec{p}$ operators are assumed to satisfy the usual commutation rules. In addition the internal coordinates $\vec{\sigma}$ commute with the translational coordinates $(\vec{p},\vec{r})$. The above Hamiltonian is the Pauli Hamiltonian for a neutral, spin-$\frac{1}{2}$ particle with a nonvanishing magnetic moment in combined static magnetic and transverse plane wave fields.[26]

A. Standing wave

In order to use the Floquet form of perturbation theory the Hamiltonian of Eq. (4.1) must be written in terms of a time-independent part H_0 and a time-dependent part $H_1(t)$. For the standing wave H_0 is given by

$$H_0 = p^2/2M + \tfrac{1}{2}\hbar\omega_0\sigma_z, \quad (4.3)$$

while for $H_1(t)$ we have

$$H_1(t) = -\tfrac{1}{2}\hbar\omega_0\sin(\omega t)(\sigma_x\cos\vec{k}\cdot\vec{r} - \sigma_y\sin\vec{k}\cdot\vec{r}), \\ = -\tfrac{1}{2}\hbar\omega_0\sin(\omega t)\,\sigma_x e^{-i\vec{k}\cdot\vec{r}\sigma_z}. \quad (4.4)$$

The connection between this example and that of the previous fixed dipole case becomes clear after performing a certain unitary transformation of the Schrödinger equation corresponding to the Hamiltonian defined by Eqs. (4.3) and (4.4). For the purpose of defining this transformation the following notation will be useful

$$\theta = \frac{p^2+(\hbar k/2)^2}{2M}, \quad \omega_1 = \omega_0 + \frac{\vec{k}\cdot\vec{p}}{M}. \quad (4.5)$$

Instead of considering the Schrödinger equation for the time evolution operator $U(t)$ consider the transformed version of this equation for the operator $U_T(t)$ defined below

$$U_T(t) = e^{(it/\hbar)\theta(\vec{p})}e^{-(i/2)\vec{k}\cdot\vec{r}\sigma_z}U(t). \quad (4.6)$$

For the transformed Hamiltonian one finds

$$H_T(t) = \tfrac{1}{2}\hbar\omega_1(\vec{p})\sigma_z + \tfrac{1}{2}\hbar\omega_0\lambda\sigma_x \sin\omega t \,. \qquad (4.7)$$

A comparison of the Hamiltonian $H_T(t)$ above with the Hamiltonian of the previous example defined by Eqs. (3.1) and (3.2) reveals that the only difference is the presence of the additional $\vec{k}\cdot\vec{p}/M$ momentum term in the time-independent part of $H_T(t)$. Consequently the algebra of the Floquet perturbation theory for the present problem is quite similar to that of the previous case. The reason for this being that since the operator $\vec{k}\cdot\vec{p}/M$ commutes with any component of the Pauli spin operator $\vec{\sigma}$ the operator $\omega_1(\vec{p})$ defined by Eq. (4.5) acts just like a scalar. However, the $\vec{k}\cdot\vec{p}/M$ term is important physically since this term is responsible for the appearance of Doppler shifts of the applied field frequency ω to the atomic transition frequency ω_0, i.e.,

$$\omega_1 - \omega = \omega_0 - \omega + kp_z/M = \omega_0 - \omega(1 + p_z/Mc) \,.$$

Having transformed the problem to something very similar to the previous stationary dipole example mathematically means that the solution for $U_T(t)$ will closely resemble that of the previous case and should be identical in the $\vec{k}=\vec{0}$ limit. Applying the Floquet theorem to the Hamiltonian defined by Eq. (4.7) leads to the conclusion that there exists a Hermitian time-independent operator μ_T and a time-dependent, unitary operator $P_T(t)$ having period τ $(=2\pi/\omega)$ such that $U_T(t) = P_T(t)\,e^{-it\mu_T}$. But due to the similarity discussed above the expansion coefficients for these operators are given by Eq. (3.8) with the difference being that in the present case instead of ν_n, A_{nm}, and B_{nm} representing scalars they now represent functions of the operator $\vec{k}\cdot\vec{p}/M$. The significant difference between these coefficients is that the momentum operator $\omega_1(\vec{p})$ replaces ω_0 in all resonant denominators and also in certain numerators. With respect to the algebra of the perturbation calculation this is a minor point and in fact the outline for calculating μ_n and $P_n(t)$ for the fixed-dipole example holds here as well. Based on this outline the expansion coefficients up to and including μ_4 and $P_4(t)$ have been calculated and recorded in Appendix B.

In addition to the Doppler shifting of the field frequency ω another feature associated with this model is the recoil effect. Mathematically it is contained in the position exponentials of Eq. (4.6). Recalling the transformation defined by this equation note that at $t=0$

$$U_T(t=0) = e^{-(i/2)\vec{k}\cdot\vec{r}\,\sigma_z} = P_T(t=0) \,. \qquad (4.8)$$

[Having $P_T(t=0) = e^{-(i/2)\vec{k}\cdot\vec{r}\,\sigma_z}$ instead of $P_T(t=0) = I$ was ignored in the above perturbation calculation since it played a nonessential role.] Equations (4.6) and (4.8) imply

$$U(t) = e^{(i/2)\vec{k}\cdot\vec{r}\,\sigma_z}\, e^{-(it/\hbar)\,\theta(\vec{p})}\,[P_T(t)\,e^{-i\mu_T t}]\,e^{-(i/2)\vec{k}\cdot\vec{r}\,\sigma_z} \,. \qquad (4.9)$$

Noting that

$$e^{(i/2)\vec{k}\cdot\vec{r}\,\sigma_z}\,\vec{p}\,e^{-(i/2)\vec{k}\cdot\vec{r}\,\sigma_z} = \vec{p} - \tfrac{1}{2}\hbar\vec{k}\sigma_z \,, \qquad (4.10)$$

yields

$$U(t) = e^{-(it/\hbar)\,\theta(\vec{p} - (\hbar/2)\vec{k}\sigma_z)}\, e^{(i/2)\vec{k}\cdot\vec{r}\,\sigma_z} \times [P_T(t)\,e^{-i\mu_T t}]\,e^{-(i/2)\vec{k}\cdot\vec{r}\,\sigma_z} \,. \qquad (4.11)$$

We found that many of the expansion coefficients defining the operators μ_T and $P_T(t)$ contained the momentum-dependent resonant term $\omega_1 - \omega$. An application of Eq. (4.10) gives $(n = 1, 2, \ldots)$

$$e^{(i/2)\vec{k}\cdot\vec{r}\,\sigma_z}\,\frac{1}{(\omega_1-\omega)^n}\,e^{-(i/2)\vec{k}\cdot\vec{r}\,\sigma_z} = \frac{1}{[\omega_0 - \omega + (\vec{k}/M)\cdot(\vec{p} - \frac{1}{2}\hbar\vec{k}\sigma_z)]^n} \,, \qquad (4.12)$$

which clarifies the origin of the atomic recoil effect.

B. Circularly polarized plane wave

Consider the Hamiltonian of Eqs. (4.1) and (4.2) with the circularly polarized oscillating field $f(\vec{r},t)$. The time-independent part of this Hamiltonian H_0 is given by Eq. (4.3) while the time-dependent part may be written as

$$H_1(t) = -\hbar\omega_0\sigma_x\,e^{i(\omega t - \vec{k}\cdot\vec{r})\sigma_z} \,. \qquad (4.13)$$

Although this Hamiltonian differs from that of the previous case, Doppler shifts and recoil effects will again be present in the solution.

As in the previous two examples the perturbation theory begins with the determination of the operator μ_1. Based on the technique of Sec. II we obtain for μ

$$\mu_1 = e^{(i/2)\vec{k}\cdot\vec{r}\,\sigma_z}\sigma_y\sigma_z f_1(\vec{p})\,e^{-(i/2)\vec{k}\cdot\vec{r}\,\sigma_z} \,, \qquad (4.14)$$

and for $\Gamma_1(t)$

$$\Gamma_1(t) = -\hbar\sigma_x\,e^{-(i/2)\vec{k}\cdot\vec{r}\,\sigma_z} \times [\omega_0 e^{i\omega t\sigma_z} + i f_1(\vec{p})]\,e^{-(i/2)\vec{k}\cdot\vec{r}\,\sigma_z} \,, \qquad (4.15)$$

where

$$f_1(\vec{p}) = i\omega_0\,\frac{\omega_0 + \vec{k}\cdot\vec{p}/M}{\omega_0 - \omega + \vec{k}\cdot\vec{p}/M} \,. \qquad (4.16)$$

The evaluation of $P_1(t)$ follows the determination of μ_1 and $\Gamma_1(t)$. This is accomplished by substituting Eqs. (4.13)–(4.16) into Eq. (2.20). This gives

$$P_1(t) = e^{(i/2)\vec{k}\cdot\vec{r}\sigma_z} i\omega_0\sigma_y \frac{e^{i\omega\sigma_z t} - I}{\omega_0 - \omega + \vec{k}\cdot\vec{p}/M} \times e^{-(i/2)\vec{k}\cdot\vec{r}\sigma_z}. \quad (4.17)$$

In this type of computation it occasionally happens that the first-order coefficients such as μ_1 and $P_1(t)$ reveal a general pattern for the higher-order expansion coefficients. Expecting that there will be a certain degree of similarity between the first-order and higher-order coefficients we propose the ansatz

$$P_n(t) = e^{(i/2)\vec{k}\cdot\vec{r}\sigma_z} b_n(\vec{p}) \times (I - e^{-i\omega t\sigma_z})\sigma_y^n e^{-(i/2)\vec{k}\cdot\vec{r}\sigma_z}, \quad (4.18)$$
$$\mu_n = e^{(i/2)\vec{k}\cdot\vec{r}\sigma_z} a_n(\vec{p})\sigma_y^n e^{-(i/2)\vec{k}\cdot\vec{r}\sigma_z},$$

where for each integer n the coefficients $a_n(\vec{p})$ and $b_n(\vec{p})$ are functions of only the linear momentum operator $\vec{p}$. This assumption means that both a_n and b_n commute with any component of the Pauli spin operator $\vec{\sigma}$. With $n=1$ Eqs. (4.16), (4.17), and (4.18) then imply

$$a_1 = i\omega_0 \frac{\omega_0 + \vec{k}\cdot\vec{p}/M}{\omega_0 - \omega + \vec{k}\cdot\vec{p}/M}, \quad b_1 = -\frac{i\omega_0}{\omega_0 - \omega + \vec{k}\cdot\vec{p}/M}. \quad (4.19)$$

The higher-order coefficients a_n and b_n will be determined in the process of establishing the validity of the above ansatz. Specifically by inserting Eq. (4.18) into the general recursion relations (2.22), (2.23), and (2.24) two basic equations relating a_n and b_n to lower-order terms will be obtained. One of these equations will then be used to eliminate the b_n terms from the other equation leaving a_n as a function of only the lower-order a_n coefficients. The solution to this relation will then yield the general expression for a_n. In turn the b_n coefficients will be obtained by inserting the a_n expression into the relation originally used to eliminate b_n from the calculation. The consistency of the original ansatz will then be apparent from the a_n and b_n expressions.

Employing the above procedure yields

$$a_n = \frac{\omega}{\omega_n - \omega}\left(i\omega_0(-1)^{n-1} b_{n-1} + \sum_{m=1}^{n-1} a_m b_{n-m}\right), \quad (4.20)$$

where

$$\omega_n = \tfrac{1}{2}[1 - (-1)^n](\omega_0 + \vec{k}\cdot\vec{p}/M), \quad (n = 1, 2, \ldots).$$

If Eq. (4.20) is used to eliminate the $b_n(\vec{p})$ coefficients from $\Gamma_n(t)$, then the operator $P_n(t)$ can be rewritten as

$$P_n(t) = e^{(i/2)\vec{k}\cdot\vec{r}\sigma_z}[(-1)^n/\omega] \times a_n(I - e^{-i\omega t\sigma_z})\sigma_y^n e^{-(i/2)\vec{k}\cdot\vec{r}\sigma_z}. \quad (4.21)$$

A comparison of Eq. (4.18) with Eq. (4.21) reveals that

$$b_n = (-1)^n a_n/\omega, \quad n = 2, 3, \ldots. \quad (4.22)$$

This expression can now be used to obtain a recursion relation for a_n. However, before doing this it is more convenient to introduce a slight notational change. For $n = 2, 3, \ldots$ set $A_n = a_n$ and take $A_1 = a_1 - i\omega_0$. With $B_n = b_n$ for $n = 1, 2, \ldots$ Eq. (4.22) becomes

$$B_n = (-1)^n A_n/\omega, \quad n = 1, 2, \ldots. \quad (4.23)$$

By rewriting Eq. (4.20) in terms of the A_n and B_n coefficients and then using Eq. (4.23) to eliminate all of the B_n coefficients we get

$$A_n = -\frac{1}{\omega}\sum_{m=1}^{n-1} A_m A_{n-m}(-1)^m, \quad n = 2, 4, \ldots,$$
$$A_n = \frac{2i\omega_0 A_{n-1} - \sum_{m=1}^{n-1} A_m A_{n-m}(-1)^m}{\omega_0 - \omega + \vec{k}\cdot\vec{p}/M}, \quad n = 3, 5, \ldots. \quad (4.24)$$

Based on the form of the first few values of A_n let us assume that there exists a set of scalars $\{f_n\}$ with $f_1 = 1$ such that for $n = 1, 2, \ldots$

$$A_n = \frac{\omega(i\omega_0)^n f_n}{(\omega_0 - \omega + \vec{k}\cdot\vec{p}/M)^n}. \quad (4.25)$$

Fortunately it turns out that such a set of scalars does exist.[27] These scalars are related to the binomial coefficients in the following way:

$$f_{2k} = \binom{2k-1}{k}, \quad f_{2k+1} = 2f_{2k}, \quad k = 1, 2, \ldots. \quad (4.26)$$

The determination of the set $\{f_n\}$ completes the solution since Eq. (4.25) can be used to evaluate each A_n coefficient. Having determined the A_n coefficients Eq. (4.23) determines the associated value of B_n. A list of these coefficients follows.

$$A_1 = i\omega X, \quad B_1 = -iX;$$
$$A_{2k} = \omega(-1)^k X^{2k}\binom{2k-1}{k},$$
$$B_{2k} = (-1)^k X^{2k}\binom{2k-1}{k};$$
$$A_{2k+1} = 2i\omega(-1)^k X^{2k+1}\binom{2k-1}{k},$$
$$B_{2k+1} = -2i(-1)^k X^{2k+1}\binom{2k-1}{k}, \quad k = 1, 2, \ldots$$

where

$$X = \frac{\omega_0}{\omega_0 - \omega + \vec{k}\cdot\vec{p}/M}\,. \tag{4.27}$$

C. Alternate forms of the time development operator for circular polarization

Naturally the next question that comes to mind is: Can the perturbation series for the operators μ and $P(t)$ be summed for certain values of λ? The answer to this question is yes! A closed-form expression for both μ and $P(t)$ does exist and moreover it is valid for all real λ. Let us consider the series for μ first. By inserting the A_n coefficients of Eq. (4.27) into Eq. (4.18) we get

$$\mu = p^2/2\hbar M + e^{(i/2)\vec{k}\cdot\vec{r}\,\sigma_z} S(\lambda)\, e^{-(i/2)\vec{k}\cdot\vec{r}\,\sigma_z}\,,$$

where

$$\begin{aligned} S(\lambda) &= \tfrac{1}{2}\omega_0\sigma_z + \lambda(i\omega_0 + A_1)\sigma_y\sigma_z + \sum_{n=2}^{\infty}\lambda^n A_n \sigma_y^n \sigma_z\,, \\ &= \tfrac{1}{2}\omega_0\sigma_z + i\lambda(\omega_0 + \omega X)\sigma_y\sigma_z \\ &\quad + \sum_{k=1}^{\infty}(-1)^k(\lambda X)^{2k}\binom{2k-1}{k}(\omega + 2i\omega\lambda X\sigma_y)\sigma_z\,. \end{aligned} \tag{4.28}$$

For the purpose of evaluating the above expression the momentum operator $\vec{p}$ can be visualized as being replaced by its transform variable $\vec{p}'$. With this replacement in mind we have

$$\sum_{k=1}^{\infty}(-1)^k[\lambda X(\vec{p})]^{2k}\binom{2k-1}{k} = \frac{1}{2}\left(\frac{\Delta}{D} - 1\right), \tag{4.29}$$

where

$$\Delta = \omega_0 - \omega + \vec{k}\cdot\vec{p}/M,\quad D = (\Delta^2 + 4\lambda^2\omega_0^2)^{1/2}.$$

The ultimate justification for this operator replacement is that the final expression for the time evolution operator must be a solution to Schrödinger's equation for all real values of λ. A substitution of Eq. (4.29) into Eq. (4.28) then yields

$$\mu = e^{(i/2)\vec{k}\cdot\vec{r}\,\sigma_z}\,\Gamma_1(\vec{p})\, e^{-(i/2)\vec{k}\cdot\vec{r}\,\sigma_z}\,, \tag{4.30}$$

$$\Gamma_1(\vec{p}) = \frac{p^2 + (\hbar k/2)^2}{2\hbar M} + \left(1 + \frac{\omega}{D}\right)\left(\frac{\Delta}{2}\sigma_z - \omega_0\lambda\sigma_x\right),$$

where again

$$\Delta = \omega_0 - \omega + \vec{k}\cdot\vec{p}/M,\quad D = (\Delta^2 + 4\omega_0^2\lambda^2)^{1/2}.$$

A similar calculation based on Eq. (4.29) yields

$$P(t) = I + (I - e^{-i\omega t\sigma_z})\, e^{(i/2)\vec{k}\cdot\vec{r}\,\sigma_z}\,\Gamma_2(\vec{p})\, e^{-(i/2)\vec{k}\cdot\vec{r}\,\sigma_z}\,, \tag{4.31}$$

$$\Gamma_2(\vec{p}) = -(i\omega_0\lambda/D)\sigma_y + \tfrac{1}{2}(\Delta/D - 1)\,.$$

Notice that the time-independent operator μ defined by Eq. (4.30) is Hermitian and that $P(t)$ as given by Eq. (4.31) is periodic with period $\tau = 2\pi/\omega$. It can be verified that $P(t)$ is unitary. The previously mentioned recoil effects are contained in the position exponentials while the Doppler shifts are contained in the $\vec{k}\cdot\vec{p}/M$ terms which are found hidden in the Δ and D operators. Finally the Floquet solution $P(t)e^{-i\mu t}$ with μ and $P(t)$ defined above for the time evolution operator satisfies Schrödinger's equation with the Hamiltonian of Eq. (4.12) for all real λ! This can be verified by a rather long but straightforward calculation. This calculation also shows $P(t)$ is unitary for all t.

If the Schrödinger equation

$$i\hbar\frac{\partial}{\partial t}U(t) = H(t)\,U(t)\,,$$

for the Hamiltonian of Eq. (4.12) is cast into the form

$$i\hbar\frac{\partial}{\partial t}[e^{(i/2)(\omega t - \vec{k}\cdot\vec{r})\sigma_z}U(t)] = H'[e^{(i/2)(\omega t - \vec{k}\cdot\vec{r})\sigma_z}U(t)]\,,$$

one finds

$$H' = \frac{1}{2M}\left[p^2 + \left(\frac{\hbar k}{2}\right)^2\right] + \frac{\hbar}{2}\left(\omega_0 - \omega + \frac{\vec{k}\cdot\vec{p}}{M}\right)\sigma_z - \hbar\omega_0\lambda\sigma_x\,. \tag{4.32}$$

This form emphasizes the relationship of the present calculation to the problem of a stationary two-state atom in a circularly polarized field. The middle term in H' shows again that new effects encountered for a movable (finite M) dipole are to be interpreted in terms of the Doppler effect. The solution for the time evolution operator $U(t)$ may be written

$$U(t) = e^{-(i/2)(\omega t - \vec{k}\cdot\vec{r})\sigma_z}\, e^{-(it/\hbar)H'}\, e^{-(i/2)\vec{k}\cdot\vec{r}\,\sigma_z}\,. \tag{4.33}$$

The equivalence of this solution and the Floquet solution $P(t)e^{-i\mu t}$ is another long but straightforward calculation.

V. CONCLUDING REMARKS

In summary, we have given a general form of Floquet's theorem (see also Salzman's discussion[17]), derived a perturbation theory aimed at obtaining the Floquet operators, and applied this theory to the interaction of stationary and moving neutral two-level atoms or molecular systems with monochromatic linearly and circularly polarized electromagnetic radiation.

Floquet's theorem is derived under quite general assumptions, but it should be noted that the operator μ is not unique. The perturbation theory given in Sec. II is designed to give the "physical" μ: When the coupling parameter vanishes, this choice of μ reduces to a constant multiple of the unperturbed Hamiltonian. While conditions for the physical μ to exist were given in Sec. IID, general necessary and sufficient conditions are not known.

ACKNOWLEDGMENTS

One of us (F. J. N.) wishes to acknowledge helpful conversations with Professor G. D. Allen and Professor J. Ward.

APPENDIX A

The coefficients μ_1^2 and μ_2^2 of Eq. (2.22) are given by the following relations:

$$\mu_1'^2 = \frac{\omega_0^3}{2(\omega+\omega_0)}, \quad \mu_1'^2 = -\frac{\omega_0^5}{32(\omega+\omega_0)^3}$$

while the harmonic coefficients a_{nm} and b_{nm} of Eq. (2.21) are

$$a_{00} = 1,$$

$$a_{10} = -\frac{is\omega\omega_0}{2(\omega^2-\omega_0^2)},$$

$$a_{11} = \frac{is\omega\omega_0}{2(\omega^2-\omega_0^2)}, \quad b_{11} = \frac{s\omega_0^2}{2(\omega^2-\omega_0^2)},$$

$$a_{20} = -\frac{\omega_0^2(3\omega^2+\omega_0^2)}{16(\omega^2-\omega_0^2)^2},$$

$$a_{21} = \frac{\omega^2\omega_0^2}{4(\omega^2-\omega_0^2)^2}, \quad b_{21} = -\frac{i\omega\omega_0^3}{4(\omega^2-\omega_0^2)^2},$$

$$a_{22} = -\frac{\omega_0^2}{16(\omega^2-\omega_0^2)}, \quad b_{22} = \frac{i\omega_0^3}{16\omega(\omega^2-\omega_0^2)},$$

$$a_{30} = \frac{is\omega\omega_0^3(15\omega^4+50\omega^2\omega_0^2-\omega_0^4)}{32(\omega^2-\omega_0^2)^3(9\omega^2-\omega_0^2)},$$

$$a_{31} = -\frac{is\omega_0^3(5\omega^3+10\omega^2\omega_0^2+\omega_0^4)}{64\omega(\omega^2-\omega_0^2)^3},$$

$$b_{31} = -\frac{s\omega_0^4(\omega^2+\omega_0^2)}{8(\omega^2-\omega_0^2)^3},$$

$$a_{32} = \frac{is\omega\omega_0^3}{32(\omega^2-\omega_0^2)^2}, \quad b_{32} = \frac{s\omega_0^4}{32(\omega^2-\omega_0^2)^2},$$

$$a_{33} = -\frac{is\omega_0^3(3\omega^2+\omega_0^2)}{64\omega(\omega^2-\omega_0^2)(9\omega^2-\omega_0^2)},$$

$$b_{33} = -\frac{s\omega_0^4}{16(\omega^2-\omega_0^2)(9\omega^2-\omega_0^2)}.$$

APPENDIX B

Here we give the coefficients ν_n, A_{nm}, and B_{nm} for $n, m \leq 4$ [see Eq. (3.8)]:

$$\nu_0 = \omega_1/2,$$

$$\nu_1 = \frac{i\omega\omega_1\omega_0}{2(\omega^2-\omega_1^2)},$$

$$\nu_2 = -\frac{\omega_1\omega_0^2(3\omega^2-\omega_1^2)}{8(\omega^2-\omega_1^2)^2},$$

$$\nu_3 = -\frac{i\omega\omega_1\omega_0^3(15\omega^4+\omega_1^4)}{8(\omega^2-\omega_1^2)^3(9\omega^2-\omega_1^2)},$$

$$\nu_4 = \frac{\omega_1\omega_0^4(105\omega^6+105\omega^4\omega_1^2-21\omega^2\omega_1^4+3\omega_1^6)}{128(\omega^2-\omega_1^2)^4(9\omega^2-\omega_1^2)},$$

where $\omega_1 = \omega_0$ for the fixed dipole and for the moving dipole $\omega_1 = \omega_0 + \vec{k}\cdot\vec{p}/M$. Also,

$$A_{00} = B_{00} = \tfrac{1}{2},$$

$$A_{10} + B_{10} = -\frac{i\omega\omega_0}{2(\omega^2-\omega_1^2)},$$

$$A_{11} = \frac{i\omega_0}{4(\omega-\omega_1)}, \quad B_{11} = \frac{i\omega_0}{4(\omega+\omega_1)},$$

$$A_{20} + B_{20} = -\frac{\omega_0^2(3\omega^2+\omega_1^2)}{16(\omega^2-\omega_1^2)^2},$$

$$A_{21} = \frac{\omega\omega_0^2}{8(\omega+\omega_1)(\omega^2-\omega_1^2)}, \quad B_{21} = \frac{\omega\omega_0^2}{8(\omega-\omega_1)(\omega^2-\omega_1^2)},$$

$$A_{22} = -\frac{\omega_0^2}{32\omega(\omega-\omega_1)}, \quad B_{22} = -\frac{\omega_0^2}{32\omega(\omega+\omega_1)},$$

$$A_{30} + B_{30} = \frac{i\omega\omega_0^3(15\omega^4+50\omega^2\omega_1^2-\omega_1^4)}{32(\omega^2-\omega_1^2)^3(9\omega^2-\omega_1^2)},$$

$$A_{31} = -\frac{i\omega_0^3(5\omega^3+3\omega^2\omega_1+7\omega\omega_1^2+\omega_1^3)}{128\omega(\omega^2-\omega_1^2)^2(\omega-\omega_1)},$$

$$B_{31} = -\frac{i\omega_0^3(5\omega^3-3\omega^2\omega_1+7\omega\omega_1^2-\omega_1^3)}{128\omega(\omega^2-\omega_1^2)^2(\omega+\omega_1)},$$

$$A_{32} = \frac{i\omega_0^3}{64(\omega^2-\omega_1^2)(\omega+\omega_1)},$$

$$B_{32} = \frac{i\omega_0^3}{64(\omega^2-\omega_1^2)(\omega-\omega_1)},$$

$$A_{33} = -\frac{i\omega_0^3}{128\omega(\omega-\omega_1)(3\omega-\omega_1)},$$

$$B_{33} = -\frac{i\omega_0^3}{128\omega(\omega+\omega_1)(3\omega+\omega_1)},$$

$$A_{40} + B_{40} = \frac{\omega_0^4(9\omega^8+548\omega^6\omega_1^2+254\omega^4\omega_1^4-44\omega^2\omega_1^6+\omega_1^8)}{8\times128\omega^2(\omega^2-\omega_1^2)^4(9\omega^2-\omega_1^2)},$$

$$-A_{41} = \frac{\omega_0^4(21\omega^5-27\omega^4\omega_1+146\omega^3\omega_1^2-6\omega^2\omega_1^3-7\omega\omega_1^4+\omega_1^5)}{4\times64(\omega^2-\omega_1^2)^3(\omega+\omega_1)(9\omega^2-\omega_1^2)},$$

$$-B_{41} = \frac{\omega_0^4(21\omega^5+27\omega^4\omega_1+146\omega^3\omega_1^2+6\omega^2\omega_1^3-7\omega\omega_1^4-\omega_1^5)}{4\times64(\omega^2-\omega_1^2)^3(\omega-\omega_1)(9\omega^2-\omega_1^2)},$$

$$A_{42} = \frac{\omega_0^4(7\omega^3+5\omega^2\omega_1+9\omega\omega_1^2-5\omega_1^3)}{8\times64\omega(\omega^2-\omega_1^2)^2(3\omega-\omega_1)(\omega-\omega_1)},$$

$$B_{42} = \frac{\omega_0^4(7\omega^3-5\omega^2\omega_1+9\omega\omega_1^2+5\omega_1^3)}{8\times64\omega(\omega^2-\omega_1^2)^2(3\omega+\omega_1)(\omega+\omega_1)},$$

$$A_{43}=-\frac{\omega_0^4}{4\times 64(\omega^2-\omega_1^2)(\omega+\omega_1)(3\omega+\omega_1)},$$

$$B_{43}=-\frac{\omega_0^4}{4\times 64(\omega^2-\omega_1^2)(\omega-\omega_1)(3\omega-\omega_1)},$$

$$A_{44}=\frac{\omega_0^4}{16\times 128\omega^2(\omega-\omega_1)(3\omega-\omega_1)},$$

$$B_{44}=\frac{\omega_0^4}{16\times 128\omega^2(\omega+\omega_1)(3\omega+\omega_1)}.$$

APPENDIX C

The problem of a moving two-state dipole interacting with two monochromatic and oppositely traveling circularly polarized fields was considered from the Floquet point of view in Sec. IV. Here it was shown that the form of Schrödinger's equation corresponding to the Hamiltonian $H_0+\lambda H_1(t)$, where H_0 was given by Eq. (4.3) and $H_1(t)$ by Eq. (4.4) could be simplified by a certain transformation. The result of this transformation was

$$i\frac{\partial U_T}{\partial t}=[\tfrac{1}{2}\omega_1(\vec{p})\sigma_z+\tfrac{1}{2}\omega_0\lambda\sigma_x\sin(\omega t)]U_T,$$

where $\omega_1(\vec{p})=\omega_0+\vec{k}\cdot\vec{p}/M$. Floquet's theorem was then applied to this equation and a solution of the form $P_T(t)e^{-i\mu_T t}$ was obtained for $U_T(t)$. The object of this Appendix is to prove that the operator μ_T has the property

$$\mu_T^2=\tfrac{1}{4}(\omega+D)^2,$$

where

$$D=\left((\omega_1-\omega)^2+\frac{\omega_1\omega_0^2\lambda^2}{2(\omega_1+\omega)}-\frac{\omega_1\omega_0^4\lambda^4}{32(\omega_1+\omega)^3}+\cdots\right)^{1/2},$$

$$\omega_1=\omega_0+\vec{k}\cdot\vec{p}/M. \quad \text{(C1)}$$

We obtain the expansion coefficients (ν_n) for μ_T up to fifth order in λ from Appendix B. Thus

$$\mu_T=(\nu_0+\lambda^2\nu_2+\lambda^4\nu_4+\cdots)\sigma_z+\lambda(\nu_1+\lambda^2\nu_3+\cdots)\sigma_y.$$

A combination of Eq. (C2) and the relation $\sigma_z\sigma_y+\sigma_y\sigma_z=0$ yields

$$\mu_T^2=\nu_0^2+(2\nu_0\nu_2+\nu_1^2)\lambda^2+(2\nu_0\nu_4+\nu_2^2+2\nu_1\nu_3)\lambda^4+\cdots. \quad \text{(C2)}$$

From Appendix B,

$$2\nu_0\nu_2+\nu_1^2=-\frac{\omega_1^2\omega_0^2(3\omega^2-\omega_1^2)}{8(\omega^2-\omega_1^2)^2}+\frac{2\omega_1^2\omega^2\omega_0^2}{8(\omega^2-\omega_1^2)^2}=-\frac{\omega_1^2\omega_0^2}{8(\omega^2-\omega_1^2)}, \quad \text{(C3)}$$

$$2\nu_0\nu_4=\frac{\omega_1^2\omega_0^4(105\omega^6+105\omega^4\omega_1^2-21\omega^2\omega_1^4+3\omega_1^6)}{128(\omega^2-\omega_1^2)^4(9\omega^2-\omega_1^2)},$$

$$2\nu_1\nu_3=-\frac{\omega_1^2\omega_0^4(240\omega^6+16\omega^2\omega_1^4)}{128(\omega^2-\omega_1^2)^4(9\omega^2-\omega_1^2)},$$

$$\nu_2^2=\frac{\omega_1^2\omega_0^4(162\omega^6-126\omega^4\omega_1^2+30\omega^2\omega_1^4-2\omega_1^6)}{128(\omega^2-\omega_1^2)^4(9\omega^2-\omega_1^2)},$$

so that

$$2\nu_0\nu_4+\nu_2^2+2\nu_1\nu_3=\frac{\omega_1^2\omega_0^4(27\omega^6-21\omega^4\omega_1^2-7\omega^2\omega_1^4+\omega_1^6)}{128(\omega^2-\omega_1^2)^4(9\omega^2-\omega_1^2)},$$

$$=\frac{\omega_1^2\omega_0^4(3\omega^4-2\omega^2\omega_1^2-\omega_1^4)}{128(\omega^2-\omega_1^2)^4},$$

$$=\frac{\omega_1^2\omega_0^4(3\omega^2+\omega_1^2)}{128(\omega^2-\omega_1^2)^3}. \quad \text{(C4)}$$

Substituting Eqs. (C3) and (C4) into Eq. (C2) gives

$$\mu_T^2=\frac{\omega_1^2}{4}-\frac{\omega_1^2\omega_0^2\lambda^2}{(8\omega^2-\omega_1^2)}+\frac{\omega_1^2\omega_0^4(3\omega^2+\omega_1^2)\lambda^4}{128(\omega^2-\omega_1^2)^3}-\cdots. \quad \text{(C5)}$$

Thinking of operator $\vec{k}\cdot\vec{p}/M$ as being replaced by its scalar transform variable we can take the square root of the right-hand side of Eq. (C5). Denoting this expression by Ω_T

$$\Omega_T=\tfrac{1}{2}\omega_1(1+X)^{1/2},$$

where

$$X=-\frac{\omega_0^2\lambda^2}{2(\omega^2-\omega_1^2)}+\frac{\omega_0^4\lambda^4(3\omega^2+\omega_1^2)}{32(\omega^2-\omega_1^2)^3}-\cdots, \quad \text{(C6)}$$

and recalling that $(1+X)^{1/2}=1+X/2-X^2/8+\cdots$ yields

$$\Omega_T=\frac{\omega_1}{2}-\frac{\omega_1\omega_0^2\lambda^2}{8(\omega^2-\omega_1^2)}+\frac{\omega_1\omega_0^4(\omega^2+3\omega_1^2)\lambda^4}{128(\omega^2-\omega_1^2)^3}-\cdots,$$

$$=\frac{\omega}{2}+\frac{\omega_1-\omega}{2}+\left(\frac{\omega_1\omega_0^2}{2(\omega+\omega_1)}\right)\left(\frac{\lambda^2}{4(\omega_1-\omega)}\right)+\left(\frac{-\omega_1\omega_0^4}{32(\omega_1+\omega)^3}\right)\left(\frac{\lambda}{4(\omega_1-\omega)}\right)-\left(\frac{\omega_1\omega_0^2}{2(\omega_1+\omega)}\right)^2\left(\frac{\lambda^4}{16(\omega_1-\omega)^3}\right)+\cdots,$$

$$=\frac{\omega}{2}+\frac{\omega_1-\omega}{2}+\frac{\alpha\lambda^2}{4(\omega_1-\omega)}+\frac{\beta\lambda^4}{4(\omega_1-\omega)}-\frac{\alpha^2\lambda^4}{16(\omega_1-\omega)^3}+\cdots,$$

where

$$\alpha = \frac{\omega_1 \omega_0^2}{2(\omega_1 + \omega)}, \quad \beta = -\frac{\omega_1 \omega_0^4}{32(\omega_1 + \omega)^3}.$$

An examination of the expansion for Ω_T above reveals that it is identical to order λ^4 with the expansion for

$$\tfrac{1}{2}\omega + \tfrac{1}{2}[(\omega_1 - \omega)^2 + \alpha\lambda^2 + \beta\lambda^4 + \cdots]^{1/2}.$$

Thus

$$\Omega_T = \frac{\omega}{2} + \frac{1}{2}\left((\omega_1 - \omega)^2 + \frac{\omega_1 \omega_0^2 \lambda^2}{2(\omega_1 + \omega)} - \frac{\omega_1 \omega_0^4 \lambda^4}{32(\omega_1 + \omega)^3} + \cdots\right)^{1/2},$$

completing the verification of the first claim.

Schrödinger's equation describing the case of a stationary two-state dipole interacting with a linearly polarized field can be obtained from Eq. (C1) by setting either $M = \infty$ or $\vec{k} = \vec{0}$. This implies $\omega_1 = \omega_0$ in all expressions. For the stationary dipole we obtain

$$\mu^2 = \tfrac{1}{4}(\omega + D)^2,$$

where

$$D = \left((\omega_0 - \omega)^2 + \frac{\omega_0^3 \lambda^2}{2(\omega_0 + \omega)} - \frac{\omega_0^5 \lambda^4}{32(\omega_0 + \omega)^3} + \cdots\right)^{1/2}.$$

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*Taken from a dissertation submitted to the Faculty of the Polytechnic Institute of Brooklyn (now New York) in partial fulfillment of the requirements for the degree of Doctor of Philosophy, 1973.

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