

Stimulated and spontaneous radiative frequency shifts of a two-level system

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The quantum-electrodynamic frequency shifts of a two-level system in spontaneous emission and in an intense quantized radiation field are given up to eighth order in the coupling parameter, using the recursive perturbation method developed previously by one of the authors. The first six orders of the latter agree with the semiclassical result of Shirley and thus disagree with Chang and Stehle's calculation. The eighth-order term agrees with a recent semiclassical calculation by Ahmad and Bullough. The transition from the quantum mechanical case (the number of photons small) to the classical case (the number of photons very large) is also exhibited in our approach.

I. INTRODUCTION

A new perturbation method, which we shall call the recursive perturbation method, was developed recently by one of the authors.¹ The successive terms of a perturbation series for the energies of a quantum system are obtained by an algebraic recurrence relation rather than by iterations or by diagrammatic methods. The method is not only a powerful numerical method particularly suited for the computer, but it also provides a new line of analytical approach to the study of quantum systems by means of difference equations. In the original papers,^{1,2} the type of Hamiltonians considered is a general one consisting of sums of products of boson and spin operators. The method has recently been extended³ to apply to a general type of Hamiltonian consisting of elements of the $SU(n)$ algebras.

In this paper, a Hamiltonian of considerable interest in quantum optics involving only the boson and spin operators will be considered. The recursive perturbation method is used not only to derive the first few terms of the energy series but is also used in such a way that it is suggestive, we hope, in leading to a possible closed-form expression for the energies of the system.

Several papers have recently appeared which were concerned with the higher-order terms in the so-called Bloch-Siegert shift.⁴ The renewal of interest in the Bloch-Siegert shift was partly stimulated by a paper of Chang and Stehle,⁵ who derived the shift from a purportedly quantum-electrodynamic calculation which disagreed with an earlier result of Shirley⁶ who derived the shift from a semiclassical theory. Several other authors⁷ have since performed calculations using various approaches and all of their results (up to the sixth-order term) agree with the result of Shirley. The estimate of the eighth-order term by Hannaford *et al.*⁸ differs from our exact result by about 10%. Significantly, however, the recent

result of Ahmad and Bullough⁹ from a semiclassical theory for the eighth-order term agrees with our fully quantum-mechanical result.

The quantum-mechanical treatment of the Bloch-Siegert shift involves the determination of the eigenvalues of the following Hamiltonian:

$$H = \omega_0 S^z + \omega a^\dagger a + \lambda [(a^\dagger S^- + a S^+) + (a^\dagger S^+ + a S^-)], \quad (1.1)$$

where the S 's are the usual spin operators and a^\dagger and a are the photon creation and annihilation operators for the quantized radiation field with frequency ω . For a spin- $\frac{1}{2}$ system, the unperturbed Hamiltonian

$$H_0 = \omega_0 S^z + \omega a^\dagger a \quad (1.2)$$

has energies $\pm \frac{1}{2} \omega_0 + n\omega$, $n=0, 1, 2, \dots$, where the spin is in the static field \vec{B}_0 parallel to the z axis, ω_0 is the Larmor frequency in \vec{B}_0 , and n is the number of photons of frequency ω . The quantized field has a linear polarization parallel to the x axis and its coupling with the spin is given by

$$H_1 = \lambda (S^+ + S^-)(a^\dagger + a). \quad (1.3)$$

The coupling constant λ , in the limit of very large n , is related to the Rabi frequency ω_1 associated with the radiation field by

$$\lambda = \omega_1 / 4\sqrt{n}. \quad (1.4)$$

The terms $(a^\dagger S + a S^\dagger)$ in (1.1) are usually referred to as the rotating terms and the terms $(a^\dagger S^+ + a S^-)$ as the counter-rotating terms.

Hamiltonians of the form (1.1) or its equivalent,

$$H = \omega_0 S^z + \omega a^\dagger a + i\lambda [(a^\dagger S^- - a S^+) + (a^\dagger S^+ - a S^-)], \quad (1.5)$$

have long been the basis of one of the most useful models in quantum optics. Recent results include the elucidation of the phase-transition properties of a system of N two-level atoms interacting with a radiation field.^{10,11} The importance of the

counter-rotating terms in the theory of spontaneous radiative frequency shifts was pointed out by Ackerhalt, Knight, and Eberly.¹²

This paper is devoted to the study of the eigenvalues of H . The eigenvalues of H can be expressed in closed form if the perturbing terms of H contain only the rotating terms or the counter-rotating terms. With both the rotating and counter-rotating terms present, however, a closed-form expression for the eigenvalues of H has not been found, and one therefore resorts to perturbation methods. We have used the recursive perturbation method to derive the correction terms to the energies $\pm \frac{1}{2}\omega_0 + n\omega$ up to eighth order in the coupling parameter as functions of ω_0 , ω , n and have deduced an expression for the correction terms for the frequency shift of a two-level atom in spontaneous emission by putting $n=0$ and an expression for the correction terms of the Bloch-Siegert shift by letting n be very large. The recursive perturbation method also gives a simple recurrence relation by which higher-order terms can be quite readily computed, and can be used to show how the perturbation series can be summed into a closed-form expression when only the rotating or the counter-rotating terms are present.

II. RECURSIVE PERTURBATION METHOD

The recursive perturbation method given by Hioe¹ consists essentially of three steps:

(i) Use the Bargmann analytic function representation¹³ of the field and spin operators.

(ii) At each order of the perturbation calculation, the eigenfunction of the Hamiltonian is taken to be a finite combination of polynomials in the Bargmann analytic function variables, the number of terms being dependent on the order of the perturbation being calculated and on the form of the perturbing Hamiltonian.

(iii) Comparison of like powers of the expansion variables gives a set of recurrence relations by which terms of the energy expansion can be derived recursively in terms of the known quantities of the previous orders.

The Bargmann representation of the field and spin operators is given by

$$a^\dagger \rightarrow z, \quad a \rightarrow \frac{\partial}{\partial z}, \quad (2.1)$$

$$S^+ \rightarrow u \frac{\partial}{\partial v}, \quad S^- \rightarrow v \frac{\partial}{\partial u}, \quad S^z \rightarrow \frac{1}{2} \left(u \frac{\partial}{\partial u} - v \frac{\partial}{\partial v} \right),$$

where u , v , and z are arbitrary complex variables (the superscript z in S^z denotes, of course, the z component). In the Bargmann representation the eigenfunction of a Hamiltonian consisting of boson and spin operators is of the form^{1,2}

$$f(u, v, z) = \sum_{k=0}^{\infty} \sum_{i=0}^{2S} c_{i,k} u^i v^{2S-i} z^k, \quad (2.2)$$

$[S(S+1)]^{1/2}$ being the total spin angular momentum. If the Hamiltonian H can be written as the sum of H_0 , the unperturbed part, and λH_1 , the perturbing part, then the eigenvalue and eigenfunction of H will be written as

$$E^{\{K\}}(\lambda) = \sum_{p=0}^{\infty} A_p^{\{K\}} \lambda^p \quad (2.3)$$

and

$$f^{\{K\}}(\lambda; u, v, z) = \sum_{p=0}^{\infty} B_p^{\{K\}}(u, v, z) \lambda^p, \quad (2.4)$$

where $\{K\}$ denotes a set of quantum numbers used to designate the particular energy considered, and $A_0^{\{K\}}$ and $B_0^{\{K\}}(u, v, z)$ are the unperturbed eigenvalue and eigenfunction, the higher-order A 's and B 's being the quantities to be determined. If the unperturbed eigenfunction $B_0^{\{K\}}(u, v, z)$ is given by $u^{\sigma} v^{2S-\sigma} z^n$ and if the highest powers of z and $\partial/\partial z$ appearing in the perturbing Hamiltonian are I and J , respectively, and the greater of the highest powers of u and v and the greater of the highest powers of $\partial/\partial u$ and $\partial/\partial v$ appearing in the perturbing Hamiltonian are P and Q , respectively, then following Ref. 1, we may derive the A 's in (2.3) recursively in a systematic and consistent manner by letting

$$f^{\sigma,n}(\lambda; u, v, z) = B_0^{\sigma,n}(u, v, z) \sum_{p=0}^{\infty} \beta_p^{\sigma,n}(u, v, z) \lambda^p, \quad (2.5)$$

where $\beta_0^{\sigma,n}(u, v, z) = 1$ and for $p \geq 1$,

$$\beta_p^{\sigma,n}(u, v, z) = \sum_{k=-Jp}^{Ip} \sum_{i=-Qp}^{Pp} b_{p;i,k}^{\sigma,n} u^i v^{-i} z^k, \quad (2.6)$$

the prime in the summation denoting the exclusion of the term $k=i=0$. The crucial part of the recursive perturbation method is expressed by Eq. (2.6). Substitutions of Eqs. (2.5) and (2.3) into the eigenvalue equation for H and comparisons of the coefficients of the like powers of u , v , and z lead us to a recurrence relation involving A 's and b 's, thus enabling us to determine these quantities recursively. It is interesting to note that (see the following sections) the recurrence relation automatically gives $b_{p;i,k}^{\sigma,n} = 0$ for $|i| > 2S$ or $k < -n$, consistent with the correct form of the eigenfunction (2.2).

III. HAMILTONIAN WITH BOTH ROTATING AND COUNTER-ROTATING TERMS

Consider a spin- $\frac{1}{2}$ system with the Hamiltonian (1.1). In the Bargmann representation, the Hamil-

tonian is given by

$$H = \frac{1}{2}\omega_0\left(u\frac{\partial}{\partial u} - v\frac{\partial}{\partial v}\right) + \omega z\frac{\partial}{\partial z} + \lambda\left[\left(\frac{\partial}{\partial z}u\frac{\partial}{\partial v} + zv\frac{\partial}{\partial u}\right) + \left(zu\frac{\partial}{\partial v} + \frac{\partial}{\partial z}v\frac{\partial}{\partial u}\right)\right]. \tag{3.1}$$

The eigenvalue and eigenfunction of the unperturbed Hamiltonian corresponding to the quantum numbers σ and n are given by

$$A_0^{\sigma,n} = (\sigma - \frac{1}{2})\omega_0 + n\omega, \quad \sigma = 0, 1 \text{ and } n = 0, 1, 2, \dots \tag{3.2}$$

and

$$B_0^{\sigma,n}(u, v, z) = u^\sigma v^{1-\sigma} z^n. \tag{3.3}$$

$$\sum_p \lambda^p \left\{ \left[\frac{1}{2}\omega_0\left(u\frac{\partial}{\partial u} - v\frac{\partial}{\partial v}\right) + \omega z\frac{\partial}{\partial z} \right] \beta_p^{\sigma,n}(u, v, z) + \left[\left((1-\sigma)nuv^{-1}z^{-1} + nu\frac{\partial}{\partial v}z^{-1} + (1-\sigma)uv^{-1}\frac{\partial}{\partial z} + u\frac{\partial^2}{\partial v\partial z} + \sigma u^{-1}vz + \frac{\partial}{\partial u}vz \right) + \left((1-\sigma)uv^{-1}z + u\frac{\partial}{\partial v}z + \sigma nu^{-1}vz^{-1} + n\frac{\partial}{\partial u}vz^{-1} + \sigma u^{-1}v\frac{\partial}{\partial z} + v\frac{\partial^2}{\partial u\partial z} \right) \right] \beta_{p-1}^{\sigma,n}(u, v, z) \right\} = \sum_p \lambda^p \sum_{q=0}^{p-1} A_{p-q}^{\sigma,n} \beta_q^{\sigma,n}(u, v, z). \tag{3.6}$$

The crucial part of the recursive perturbation method is to let

$$\beta_p^{\sigma,n}(u, v, z) = \sum_{k=-p}^p \sum_{i=-p}^p {}'b_{p;i,k}^{\sigma,n} u^i v^{-i} z^k, \tag{3.6a}$$

where the prime in the summation denotes the exclusion of the term $k = i = 0$. Now comparing the coefficients of $u^i v^{-i} z^k$ on both sides of (3.6) gives the following recurrence relation (the superscripts σ and n on A 's and b 's are omitted for convenience):

$$(i\omega_0 + k\omega)b_{p;i,k} + [(-\sigma - i + 2)(n + k + 1)b_{p-1;i-1,k+1} + (\sigma + i + 1)b_{p-1;i+1,k-1}] + [(-\sigma - i + 2)b_{p-1;i-1,k-1} + (\sigma + i + 1)(n + k + 1)b_{p-1;i+1,k+1}] = \sum_{q=0}^{p-1} A_{p-q} b_{q;i,k}, \tag{3.7}$$

with

$$b_{0;i,k} = \delta_{i0}\delta_{k0} \tag{3.7a}$$

and

$$b_{p;i,k} = 0 \text{ if } i = k = 0, \ p > 0 \text{ or if } |i| \text{ or } |k| > p.$$

For a particular order p considered, the recurrence relation (3.7) formally implies a set of simultaneous equations in $(2p + 1)^2$ unknowns, $b_{p;i,k}$, $i, k = -p, -p + 1, \dots, p$ (excluding $i = k = 0$) and A_p which are given in terms of the A 's and b 's of lower orders than p .

However, we have a number of simplifying fac-

In the presence of interaction, let the same eigenvalue and eigenfunction now become

$$E^{\sigma,n}(\lambda) = \sum_{p=0}^{\infty} A_p^{\sigma,n} \lambda^p, \tag{3.4}$$

$$f^{\sigma,n}(\lambda; u, v, z) = \sum_{p=0}^{\infty} B_p^{\sigma,n}(u, v, z) \lambda^p = B_0^{\sigma,n}(u, v, z) \sum_{p=0}^{\infty} \beta_p^{\sigma,n}(u, v, z) \lambda^p, \tag{3.5}$$

with $\beta_0^{\sigma,n}(u, v, z) = 1$. Substituting (3.5) and (3.4) into the eigenvalue equation for H , comparing the coefficients of λ^n on both sides and remembering $H_0 B_0^{\sigma,n} = A_0^{\sigma,n} B_0^{\sigma,n}$, we get

tors in practice. First, the $b_{p;i,k}$ can be determined *individually* as there is only one b of order p appearing in each equation (3.7). Second, $b_{p;i,k}$ is equal to zero for $|i| > 2S$ or $k < -n$ in addition to its being equal to zero as given by (3.7a). Thus for a spin- $\frac{1}{2}$ atom, the only nonzero $b_{p;i,k}$ are those for which $i = 0, \pm 1$ and $k > -n$. Further simplification results for the Hamiltonian (1.1), as it turns out that if $\omega \neq \omega_0$, the only nonzero $b_{p;i,k}^{\sigma,n}$ are those for which (i) when p is even, $i = 0, k = \text{even integer} > -n$ and (ii) when p is odd, $i = 1, k = \text{odd integer} > -n$ for $\sigma = 0$, and $i = -1, k = \text{odd integer} > -n$ for $\sigma = 1$. Thus, without much trouble, we derive the following expression for the frequency shift:

$$E^{1,n}(\lambda) - E^{0,n}(\lambda) = \omega_0 - \frac{2\omega_0(2n+1)}{\omega^2 - \omega_0^2} \lambda^2 + \frac{2\omega_0(\omega^2 + 3\omega_0^2)(2n^2 + 2n + 1)}{(\omega^2 - \omega_0^2)^3} \lambda^4 + \dots \quad (3.8)$$

For $n=0$, we have obtained the following expression for the frequency shift of a two-level atom undergoing "spontaneous emission"¹⁴ up to eighth order in the coupling parameter:

$$E^{1,0}(\lambda) - E^{0,0}(\lambda) = \omega_0 - \frac{2\omega_0}{\omega^2 - \omega_0^2} \lambda^2 + \frac{2\omega_0(\omega^2 + 3\omega_0^2)}{(\omega^2 - \omega_0^2)^3} \lambda^4 - \frac{4\omega_0(3\omega^8 + 64\omega^6\omega_0^2 + 70\omega^4\omega_0^4 - 8\omega^2\omega_0^6 - \omega_0^8)}{\omega^2(\omega^2 - \omega_0^2)^5(9\omega^2 - \omega_0^2)} \lambda^6 \\ + \frac{2\omega_0(27\omega^{12} + 2577\omega^{10}\omega_0^2 + 13070\omega^8\omega_0^4 + 7026\omega^6\omega_0^6 - 2217\omega^4\omega_0^8 - 51\omega^2\omega_0^{10} + 48\omega_0^{12})}{\omega^2(\omega^2 - \omega_0^2)^7(9\omega^2 - \omega_0^2)^2} \lambda^8. \quad (3.9)$$

For very large n , we have obtained the following expression for the Bloch-Siegert shift up to eighth order in the coupling parameter:

$$E^{1,n}(\lambda) - E^{0,n}(\lambda) = \omega_0 - \frac{2\omega_0}{\omega^2 - \omega_0^2} 2b^2 + \frac{2\omega_0(\omega^2 + 3\omega_0^2)}{(\omega^2 - \omega_0^2)^3} 2b^4 - \left(\frac{32\omega_0^3(\omega^2 + \omega_0^2)}{(\omega^2 - \omega_0^2)^5} + \frac{8\omega_0}{(\omega^2 - \omega_0^2)^2(9\omega^2 - \omega_0^2)} \right) 2b^6 \\ + \left(\frac{-2\omega_0(\omega^6 - 39\omega^4\omega_0^2 - 101\omega^2\omega_0^4 - 53\omega_0^6)}{(\omega^2 - \omega_0^2)^7} + \frac{16\omega_0(\omega^2 + \omega_0^2)}{(\omega^2 - \omega_0^2)^4(9\omega^2 - \omega_0^2)} \right. \\ \left. + \frac{4\omega_0(3\omega^8 + 14\omega^6\omega_0^2 + 388\omega^4\omega_0^4 + 130\omega^2\omega_0^6 - 23\omega_0^8)}{(\omega^2 - \omega_0^2)^7(9\omega^2 - \omega_0^2)} \right. \\ \left. - \frac{8\omega_0(9\omega^8 + 252\omega^6\omega_0^2 + 382\omega^4\omega_0^4 - 148\omega^2\omega_0^6 + 17\omega_0^8)}{(\omega^2 - \omega_0^2)^6(9\omega^2 - \omega_0^2)^2} \right) 2b^8, \quad (3.10)$$

where $b \equiv \sqrt{n}\lambda$ is the notation used by Shirley⁶ and the terms up to b^8 in (3.10) agree with those given by him.

It will be observed that in Eq. (3.5), since $B_0^{\sigma,n}(\mu, \nu, z)$ is cancelled on both sides of the eigenvalue equation for H , it is the function $\beta_p^{\sigma,n}(\mu, \nu, z)$ and not the function $B_p^{\sigma,n}(\mu, \nu, z)$ which plays the crucial role and it will be noted that the power of z in $\sum_{p=0}^{\infty} \beta_p^{\sigma,n}(\mu, \nu, z)\lambda^p$ ranges from $-n$ to $+\infty$. Thus in the fully quantum-mechanical case $n=0$, we have only positive powers of z while in the fully classical case $n=\infty$, we have the powers of z

ranging from $-\infty$ to $+\infty$. Shirley's formulation of the problem using the Floquet Hamiltonian is closely related to this point.¹⁵

For the Bloch-Siegert shift, define

$$q \equiv \frac{1}{2} \{ \omega - [E^{1,n}(\lambda) - E^{0,n}(\lambda)] \}. \quad (3.11)$$

Shirley⁶ showed that q is related to the spin transition probability through its derivative:

$$P = \frac{1}{2} [1 - 4(\partial q / \partial \omega_0)^2]. \quad (3.11a)$$

The quantity q^2 is a more useful quantity as it is nonsingular at $\omega = \omega_0$. We find from (3.10),

$$q^2 = \frac{1}{4}(\omega - \omega_0)^2 + \frac{2\omega_0 b^2}{\omega + \omega_0} - \frac{2\omega_0 b^4}{(\omega + \omega_0)^3} + \frac{8\omega_0(\omega^2 - 5\omega\omega_0 - 2\omega_0^2)b^6}{(\omega + \omega_0)^5(9\omega^2 - \omega_0^2)} - \frac{2\omega_0(9\omega^4 - 252\omega^3\omega_0 + 346\omega^2\omega_0^2 + 364\omega\omega_0^3 + 77\omega_0^4)b^8}{(\omega + \omega_0)^7(9\omega^2 - \omega_0^2)^2} + \dots \quad (3.12)$$

Setting $\partial q^2 / \partial \omega_0 = 0$ to obtain a maximum¹⁶ for P , we get

$$\omega_0 = \omega - \frac{4\omega b^2}{(\omega + \omega_0)^2} + \frac{4(\omega - 2\omega_0)b^4}{(\omega + \omega_0)^4} - \frac{16(9\omega^5 - 126\omega^4\omega_0 + 82\omega^3\omega_0^2 + 42\omega^2\omega_0^3 - 23\omega\omega_0^4 - 8\omega_0^5)b^6}{(\omega + \omega_0)^6(9\omega^2 - \omega_0^2)^2} \\ + \frac{4(81\omega^7 - 5022\omega^6\omega_0 + 20709\omega^5\omega_0^2 + 234\omega^4\omega_0^3 - 8285\omega^3\omega_0^4 + 1382\omega^2\omega_0^5 + 2471\omega\omega_0^6 + 462\omega_0^7)b^8}{(\omega + \omega_0)^8(9\omega^2 - \omega_0^2)^3} + \dots \quad (3.13)$$

The position of the main resonance can be determined iteratively from (3.13). It is usually more convenient (but not more accurate) to express ω_0 explicitly in terms of b and ω or to express ω explicitly in terms of b and ω_0 . From (3.13), we find

$$\omega_0 = \omega - \frac{b^2}{\omega} - \frac{5}{4} \frac{b^4}{\omega^3} - \frac{61}{32} \frac{b^6}{\omega^5} - \frac{407}{128} \frac{b^8}{\omega^7} - \dots, \quad (3.14)$$

$$\omega = \omega_0 + \frac{b^2}{\omega_0} + \frac{1}{4} \frac{b^4}{\omega_0^3} - \frac{35}{32} \frac{b^6}{\omega_0^5} + \frac{103}{128} \frac{b^8}{\omega_0^7} + \dots \quad (3.15)$$

IV. COMPARISON WITH THE RESULTS OF OTHER AUTHORS

For the spontaneous shift, the terms of order higher than the second in (3.9) are new as far as we know.

For the Bloch-Siegert shift, the terms up to b^6 in (3.14) agree with the semiclassical result of Shirley.⁶ Our result thus disagrees with the conclusion of Chang and Stehle⁵ who claimed that the semiclassical treatment leads to a frequency shift at variance with their quantum-electrodynamic result. Our result is of course fully quantum electrodynamic, but in agreement with the results of Shirley and other authors⁷ rather than with the result of Chang and Stehle. The eighth-order term in (3.14) was estimated semiclassically by Hannaford *et al.*⁸ as approximately equal to $-227b^8/64\omega^7$ and this differs from our exact result by about 10%. It is significant, however, that our eighth-order term in (3.14) agrees with the semiclassical result of Ahmad and Bullough⁹ which was given very recently. The eighth-order term in (3.15) has not been given previously although it can be easily obtained by inverting (3.14).

The expressions (3.12) and (3.13) are independently useful, and it will be noted that while (3.14) and (3.15) can be obtained from them, the converse is not true. The coefficients up to the terms in b^6 in (3.12) were given previously by Pegg¹⁷ from a semiclassical theory and the terms up to b^6 in (3.13) were given previously by Hannaford *et al.*⁸ also from a semiclassical theory. The eighth-order terms in (3.12) and (3.13) are new and have never been given previously from either the semiclassical or quantum theory.

It should be mentioned that Stenholm¹⁸ derived a continued-fraction expression for the Bloch-Siegert shift from a semiclassical theory which he showed to give a power-series expression for the shift in agreement with the result of Shirley and the quantum-electrodynamic result of Cohen-Tannoudji *et al.* (up to sixth order). It would obviously be of value to have the quantum-electrodynamic Bloch-Siegert shift also expressed as a continued fraction and this has been done and will be published elsewhere.¹⁹ It should also be mentioned that for small values of ω_0/ω , Hannaford *et al.*⁸ obtained a semiclassical expression which expresses the shift in terms of Bessel functions and powers of ω_0/ω , and these expressions were said to give a better fit than (3.14) or (3.15) in that region.

V. HAMILTONIAN WITH ONLY THE ROTATING OR COUNTER-ROTATING TERMS

The eigenvalues of a spin- $\frac{1}{2}$ system with Hamiltonian (1.1) excluding the counter-rotating terms,

i.e.,

$$H = \omega_0 S^z + \omega a^\dagger a + \lambda (a^\dagger S^- + a S^+) \quad (5.1)$$

have been given by Tavis and Cummings²⁰ and Mallory.²¹ With a view to understanding the solution of Eq. (3.7), however, it is instructive to see how Eq. (3.7) simplifies in this case, resulting in the possibility of summing the perturbation series into a simple closed-form expression.

In the absence of the counter-rotating terms, the recurrence relation (3.7) reduces, for $\sigma = 1$, to

$$(i\omega_0 + k\omega)b_{p,i,k} + [(-i+1)(n+k+1)b_{p-1;i-1,k+1} + (i+2)b_{p-1;i+1,k-1}] = \sum_{q=0}^{p-1} A_{p-q} b_{q;i,k} \quad (5.2)$$

Remembering that only certain $b_{p,i,k}$ are nonzero from Sec. IV, it is easy to see that all $b_{p,i,k} = 0$ except those with $i = -1$, $k = 1$, and $p = \text{odd integer}$, and the odd-numbered A_p (i.e., $p = \text{odd number}$) vanish. Thus the recurrence relation (5.2) becomes (omitting the subscripts i and k in b 's)

$$(\omega - \omega_0)b_{2p-1} = \sum_{q=1}^{2p-2} A_{2p-1-q} b_q \quad (5.3)$$

and

$$A_{2p} = (n+1)b_{2p-1}. \quad (5.4)$$

We thus get

$$A_{2p} = \frac{1}{\omega - \omega_0} \sum_{q=1}^{p-1} A_{2p-2q} A_{2q} \quad \text{for } p \geq 2 \quad (5.5)$$

with

$$A_2 = -(n+1)/(\omega - \omega_0). \quad (5.6)$$

The nonlinear equation (5.5) turns out to have a simple solution, because if we assume the series

$$-\frac{1}{2}(\omega - \omega_0) + A_2 \lambda^2 + A_4 \lambda^4 + \dots \quad (5.7)$$

to be an expansion of the expression

$$-\frac{1}{2}(\omega - \omega_0) \left(1 + \frac{4(n+1)\lambda^2}{(\omega - \omega_0)^2} \right)^t, \quad (5.8)$$

then we have

$$A_{2p} = (-1)^{p-1} \left(\frac{2}{\omega - \omega_0} \right)^{2p-1} (n+1)^p \binom{t}{p}. \quad (5.9)$$

But Eq. (5.5) requires that for $p \geq 2$,

$$(-1)^{p-1} \left(\frac{2}{\omega - \omega_0} \right)^{2p-1} (n+1)^p \binom{t}{p} = \frac{1}{\omega - \omega_0} \sum_{q=1}^{p-1} (-1)^{p-q-1} \left(\frac{2}{\omega - \omega_0} \right)^{2p-2q-1} \\ \times (n+1)^{p-q} \binom{t}{p-q} (-1)^{q-1} \left(\frac{2}{\omega - \omega_0} \right)^{2q-1} (n+1)^q \binom{t}{q}, \quad (5.10)$$

or

$$\binom{t}{p} = -\frac{1}{2} \sum_{q=1}^{p-1} \binom{t}{p-q} \binom{t}{q} = -\frac{1}{2} \left[\binom{2t}{p} - 2 \binom{t}{p} \right], \quad (5.11)$$

i.e., $\binom{2t}{p}$ must be equal to zero for $p \geq 2$, and this has a nontrivial solution $t = \frac{1}{2}$. Thus, we obtain for the case $\sigma = 1$ the following exact eigenvalues of H given by (5.1):

$$E^{1,n}(\lambda) = n\omega + \frac{1}{2}\omega_0 + \frac{1}{2}(\omega - \omega_0) \\ - \frac{1}{2}(\omega - \omega_0) \left(1 + \frac{4(n+1)\lambda^2}{(\omega - \omega_0)^2} \right)^{1/2} \\ = (n + \frac{1}{2})\omega - \frac{1}{2}[(\omega - \omega_0)^2 + 4(n+1)\lambda^2]^{1/2}, \\ n = 0, 1, 2, \dots \quad (5.12)$$

The remaining eigenvalues of H (the case $\sigma = 0$) can be obtained in a similar way and are found to be

$$E^{0,n}(\lambda) = (n - \frac{1}{2})\omega + \frac{1}{2}[(\omega - \omega_0)^2 + 4n\lambda^2]^{1/2}, \\ n = 0, 1, 2, \dots \quad (5.13)$$

Expressions (5.12) and (5.13) are in agreement with the results given by Tavis and Cummings, and Mallory using different methods. In our approach, the removal of the singularities at $\omega = \omega_0$ of $1/(\omega - \omega_0)^{2p-1}$ following the summation of the perturbation series is clearly exhibited.

For a spin- $\frac{1}{2}$ system with the Hamiltonian consisting only of the counter-rotating terms, i.e.,

$$H = \omega_0 S^z + \omega a^\dagger a + \lambda (a^\dagger S^+ + a S^-), \quad (5.14)$$

the eigenvalues of H can also be obtained in a similar way, and they are found to be

$$E^{1,n}(\lambda) = (n - \frac{1}{2})\omega + \frac{1}{2}[(\omega + \omega_0)^2 + 4n\lambda^2]^{1/2} \quad (5.15)$$

and

$$E^{0,n}(\lambda) = (n + \frac{1}{2})\omega - \frac{1}{2}[(\omega + \omega_0)^2 + 4(n+1)\lambda^2]^{1/2}, \\ n = 0, 1, 2, \dots \quad (5.16)$$

As mentioned earlier, when both the rotating and the counter-rotating terms are present, we have not been able to obtain a closed-form expression for the eigenvalues of H from the recurrence relation (3.7). It is hoped that the approach we used leading to the expressions (5.12), (5.13), (5.15), (5.16), (3.9), and (3.10) would shed more light on this problem which might lead us to obtain the required closed-form expression.

VI. SUMMARY

We have presented a new fully quantum-mechanical method for calculating the stimulated and spontaneous radiative frequency shifts of a two-level system and we have presented a recurrence relation (3.7) by which the successive perturbation terms can be obtained with considerable speed. Expressions for the spontaneous and stimulated frequency shifts (3.9) and (3.10) up to eighth order in the coupling constant are given as examples but these are not representative of the power of our recursive method. The power will be clearly exhibited in the actual numerical computation with a computer. Generalizations of the method to the study of multilevel atoms in a radiation field are obvious.

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¹F. T. Hioe, J. Math. Phys. **15**, 445 (1974).

²F. T. Hioe, J. Math. Phys. **15**, 1174 (1974).

³F. T. Hioe, J. Math. Phys. **15**, 1615 (1974).

⁴F. Bloch and A. Siegert, Phys. Rev. **57**, 522 (1940).

⁵C. S. Chang and P. Stehle, Phys. Rev. A **4**, 641 (1971), see especially Sec. VI and Eq. (87).

⁶J. H. Shirley, Phys. Rev. **138**, B979 (1965).

⁷C. Cohen-Tanoudji, J. Dupont-Roc, and C. Fabre, J. Phys. B **6**, L214 (1973); P. Hannaford, D. T. Pegg, and G. W. Series, J. Phys. B **6**, L222 (1973); S. Stenholm, J. Phys. B **5**, 890 (1972).

⁸P. Hannaford, D. T. Pegg, and G. W. Series, J. Phys. B **6**, L222 (1973).

- ⁹F. Ahmad and R. K. Bullough, *J. Phys. B* 7, L147 (1974).
- ¹⁰F. T. Hioe, *Phys. Rev. A* 8, 1440 (1973); Y. K. Wang and F. T. Hioe, *Phys. Rev. A* 7, 831 (1973).
- ¹¹K. Hepp and E. H. Lieb, *Phys. Rev. A* 8, 2517 (1973); *Ann. Phys. (N. Y.)* 76, 360 (1973).
- ¹²J. R. Ackerhalt, P. L. Knight, and J. H. Eberly, *Phys. Rev. Lett.* 30, 456 (1973). See also L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1974), Secs. 7.4 and 7.5.
- ¹³V. Bargmann, *Commun. Pure Appl. Math.* 20, 1 (1967); *Rev. Mod. Phys.* 34, 829 (1962).
- ¹⁴The model interaction of one field mode with a two-level system is, of course, too simple to represent adequately many features of real spontaneous emission. It is amusing, however, that the real spontaneous-emission second-order frequency shift for a two-level system (see Ref. 12) is *exactly* reproduced by our Eq. (3.9) if the second-order term is simply summed over all frequencies.
- ¹⁵See also S. Swain, *J. Phys. A* 6, L169 (1973).
- ¹⁶Although it is generally accepted that setting $\partial q/\partial\omega_0 = 0$ is equivalent to maximizing the transition probability, a fully quantum-mechanical proof has never been given. The relation (3.11a) was derived semiclassically.
- ¹⁷D. T. Pegg, *J. Phys. B* 6, 246 (1973).
- ¹⁸S. Stenholm, *J. Phys. B* 6, L240 (1973).
- ¹⁹F. T. Hioe and E. W. Montroll, *J. Math. Phys.* (to be published).
- ²⁰M. Tavis and F. W. Cummings, *Phys. Rev.* 170, 379 (1968).
- ²¹W. R. Mallory, *Phys. Rev.* 188, 1976 (1969).