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Stimulated absorption and emission of strong monochromatic radiation by a two-level atom

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The time evolution of a two-level system interacting with a classical monochromatic radiation field is calculated by Fourier transforming the equations of motion. This can be done exactly if one uses the convolution of the product of two Fourier transformations. The method produces recurrence relationships for the transforms. The time-dependent amplitudes of the states and the transition probability are evaluated by using two such recurrence relationships. The time-averaged probability shows characteristic resonances at odd multiples of the ratio of the natural frequency of the system to the frequency of the radiation field. The peaks of the resonances are displaced by the Bloch-Siegert shift. At these resonances the system is completely saturated with the flipping frequency dependent on the strength of the radiation field. The time dependence and the frequency spectrum of the dipole moment is determined at the main resonance and at the three-quantum resonance. The spectrum contains the zero-frequency terms which are responsible for the coherence resonances.

I. INTRODUCTION

Recently there has been renewed interest in the solution of the interaction of a strong classical monochromatic radiation field with a two-level system. Muriel[1] has attempted an exact analytical solution, but it was shown by Gupta[2] and Beers and Nickle[3] that his results are not valid for more than half a period of the radiation field. At about the same time Gush and Gush[4] published a method of solution which uses the nonrelativistic Green's-function operator. Their results are expressed in terms of continued fractions. Salzman[5] treats the same problem by integrating the differential equations numerically. He gives an expression for the slowly varying part of the transition probability; a result which is identical with the rotating-wave approximation. Moloney, Ali, and Meath[6] continued the investigation of the time evolution of a two-level atom interacting with a sinusoidal external field. They discuss an iterative power-series solution and give results using as many as 120 terms in the expansion. Some aspects of this problem have also been presented by Shirley,[7] Stenholm,[8] Stenholm and Aminoff,[9] Pegg and Series,[10] Cohen-Tannoudji *et al.*,[11] and Vermani.[12]

We show in this paper that the Schrödinger equations of a two-level atom interacting with a monochromatic wave can be Fourier transformed by using the method of convolution of the product of two Fourier transformations. The resulting recurrence relationships of the transformations can then be solved to any desired accuracy.

We illustrate the method by calculating analytical expressions for the time-dependent amplitudes and probabilities of the two states using the first- and second-order approximation. The approximations are obtained from different recurrence relationships. We also show how these solutions are related to the rotating-wave approximation. In addition we calculate the time-averaged probability of the system. The results show resonances at odd multiples of the ratio ω_0/ω_1 where ω_0 is the natural frequency of the system and ω_1 the frequency of the radiation field. The peaks of the resonances are displaced by the familiar Bloch-Siegert shift.[13]

The time-dependent solutions also allow us to calculate the evolution of the dipole moment of the system. In particular we determine the frequency spectrum of the dipole moment at the main resonance and at the three-quantum resonance, and we point out how the system can have coherence resonances which appear at even multiples of the ratio ω_0/ω_1.

II. DIFFERENTIAL EQUATIONS

We investigate the solution of the Schrödinger equation

$$\hat{H}|\Phi\rangle = i\hbar \frac{\partial}{\partial t}|\Phi\rangle \tag{1}$$

for a two-level atom interacting with a monochromatic wave. The Hamiltonian $\hat{H}$ is written in two parts:

$$\hat{H} = \hat{H}_0 + \hat{H}_1, \tag{2}$$

where $\hat{H}_0$ is the Hamiltonian of the atom and $\hat{H}_1$ represents the interaction of the atom with the radiation field. The states of the atom are quantized, but the radiation field is treated classically. It is convenient to transform the Schrödinger equation to the interaction representation. If we write

$$\hat{H}_1' = \exp(i\hat{H}_0 t/\hbar)\hat{H}_1 \exp(-i\hat{H}_0 t/\hbar) \tag{3}$$

and

$$|\Phi'\rangle = \exp(i\hat{H}_0 t/\hbar)|\Phi\rangle, \tag{4}$$

then Eq. (1) becomes

$$H_1'|\Phi'\rangle = i\hbar \frac{\partial}{\partial t}|\Phi'\rangle. \tag{5}$$

The solution of Eq. (5) is obtained by expanding the wave function $|\Phi'\rangle$ in terms of the eigenstates of the Hamiltonian $\hat{H}_0$:

$$\hat{H}_0|\psi_j\rangle = \hbar\omega_j|\psi_j\rangle. \tag{6}$$

We write

$$|\Phi'\rangle = \sum_j b_j(t)|\psi_j\rangle, \tag{7}$$

and substitution of Eq. (7) into Eq. (5) yields the following set of coupled linear differential equations for the amplitudes $b_k(t)$:

$$i\frac{d}{dt}b_k(t) = \sum_j \exp[i(\omega_k - \omega_j)t]V_{kj}b_j(t) \quad (k = 1, 2, \ldots). \tag{8}$$

In Eq. (8) V_{kj} stands for the matrix element $\hbar^{-1}\langle\psi_k|\hat{H}_1|\psi_j\rangle$.

The wave functions of the two atomic states are $|\psi_m\rangle$ and $|\psi_n\rangle$ with the atom in state $|\psi_m\rangle$ at $t = 0$. The initial conditions $b_m(0) = 1$ and $b_n(0) = 0$ can be incorporated into the differential equations by adding the term $i\delta(t)$ to the equation for $db_m(t)/dt$.

The interaction of the atom with the radiation field is

$$\hat{H}_1 = \hat{V}\cos(\omega_1 t). \tag{9}$$

The operator $\hat{V}$ contains all the space-dependent terms. Thus the appropriate differential equations for a two-level atom interacting with a monochromatic wave are

$$\begin{aligned} i\frac{d}{dt}b_m(t) &= V\cos(\omega_1 t)\exp(-i\omega_0 t)b_n(t) + i\delta(t), \\ i\frac{d}{dt}b_n(t) &= V\cos(\omega_1 t)\exp(i\omega_0 t)b_m(t), \end{aligned} \tag{10}$$

where we set $V = \langle\psi_m|\hat{V}|\psi_n\rangle = \langle\psi_n|\hat{V}|\psi_m\rangle$ and $\omega_0 = \omega_n - \omega_m$.

III. METHODS OF SOLUTION

Before discussing the exact solution of Eq. (10) we present two approximations which are frequently used in the analysis of this system.

A. Short-time solution

One can solve Eq. (10) by assuming that $b_m(t) = 1$ for all time. The amplitude $b_n(t)$ is obtained by integrating the second equation of Eq. (10) with the result

$$b_n(t) = \frac{V}{2}\left(\frac{1 - \exp[i(\omega_0 + \omega_1)t]}{\omega_0 + \omega_1} + \frac{1 - \exp[i(\omega_0 - \omega_1)t]}{\omega_0 - \omega_1}\right). \tag{11}$$

This solution can be substituted into the first equation of Eq. (10) and an integration yields an improved approximation for $b_m(t)$ which in turn can be substituted into the second equation and this way one can obtain series solutions for $b_m(t)$ and $b_n(t)$. For short times and $V \ll \omega_0$ the series converges rapidly. For ω_1 near the resonance frequency ω_0 the second term in Eq. (11) is much larger than the first, and we can write the probability of the state $|\psi_n\rangle$ as

$$|b_n(t)|^2 = V^2 \frac{\sin^2[\frac{1}{2}(\omega_0 - \omega_1)t]}{(\omega_0 - \omega_1)^2} \quad (\omega_1 \simeq \omega_0). \tag{12}$$

At resonance the excitation probability increases quadratically with time. In addition the solution does not conserve the total probability of the system.

B. Rotating-wave approximation

The differential equations of Eq. (10) contain terms which depend on the sum and difference of ω_0 and ω_1. If we neglect the nonresonant high-frequency terms, the equations reduce to

$$\begin{aligned} i\frac{d}{dt}b_m(t) &= \tfrac{1}{2}V\exp(-i\Delta\omega t)b_n(t) + i\delta(t), \\ i\frac{d}{dt}b_n(t) &= \tfrac{1}{2}V\exp(i\Delta\omega t)b_m(t), \end{aligned} \tag{13}$$

where we set $\Delta\omega = \omega_0 - \omega_1$.

One way of solving Eq. (13) is to Fourier transform the amplitudes. We define

$$b_m(t) = -\frac{1}{2\pi i}\int_{-\infty}^{\infty} G_m(\omega)\exp[-i(\omega + \tfrac{1}{2}\Delta\omega)t]d\omega, \tag{14}$$

$$b_n(t) = -\frac{1}{2\pi i}\int_{-\infty}^{\infty} G_n(\omega)\exp[-i(\omega - \tfrac{1}{2}\Delta\omega)t]d\omega, \tag{15}$$

$$i\delta(t) = -\frac{1}{2\pi i}\int_{-\infty}^{\infty}\exp[-i(\omega + \tfrac{1}{2}\Delta\omega)t]d\omega. \tag{16}$$

If we substitute these transformations into Eq. (13), we find that the Fourier transforms must satisfy the equations

$$
\begin{aligned}
(\omega+\tfrac{1}{2}\Delta\omega)G_m(\omega)&=\tfrac{1}{2}V\,G_n(\omega)+1,\\
(\omega-\tfrac{1}{2}\Delta\omega)G_n(\omega)&=\tfrac{1}{2}V\,G_m(\omega),
\end{aligned}
\tag{17}
$$

with the solutions

$$G_m(\omega)=\frac{\omega-\frac{1}{2}\Delta\omega}{(\omega+\frac{1}{2}\Delta\omega)(\omega-\frac{1}{2}\Delta\omega)-(\frac{1}{2}V)^2}\,, \tag{18}$$

$$G_n(\omega)=\frac{\frac{1}{2}V}{(\omega+\frac{1}{2}\Delta\omega)(\omega-\frac{1}{2}\Delta\omega)-(\frac{1}{2}V)^2}\,. \tag{19}$$

The amplitudes in Eqs. (14) and (15) are obtained by extending the integration over ω into the complex plane. The Fourier transforms have poles at

$$\Omega_p=\pm\tfrac{1}{2}[(\Delta\omega)^2+V^2]^{1/2}, \tag{20}$$

and if we displace the poles from the real axis by adding to them a term $-i\epsilon$ we can close the contour by an infinite semicircle in the lower half-plane. The integrand is zero over the semicircle. After the integration we let $\epsilon\to 0$. The amplitudes $b_m(t)$ and $b_n(t)$ are equal to the sum of the residues of the integrands.

Using this procedure we get for the probability $|\psi_n\rangle$ in the rotating-wave approximation

$$|b_n(t)|^2=\frac{V^2}{(\Delta\omega)^2+V^2}\sin^2\{\tfrac{1}{2}[(\Delta\omega)^2+V^2]^{1/2}t\}, \tag{21}$$

which for $V\ll\Delta\omega$ agrees with Eq. (12). This is the familiar Rabi formula. The probability has a periodicity $\tau=2\pi/[(\Delta\omega)^2+V^2]^{1/2}$ and after $t=\tau/2$ the transition probability is

$$\frac{V^2}{(\Delta\omega)^2+V^2}\,.$$

Thus at resonance the transition rate is proportional to V.

The time-averaged excitation probability is

$$
\begin{aligned}
|b_n|^2&=\frac{1}{2}\frac{V^2}{(\Delta\omega)^2+V^2}\\
&=\frac{1}{2}\frac{(V/\omega_1)^2}{(\omega_0/\omega_1-1)^2+(V/\omega_1)^2}\,.
\end{aligned}
\tag{22}
$$

When plotted as a function of ω_0/ω_1 the time-averaged probability is a Lorentzian with a maximum at $\omega_0=\omega_1$ and a width at half-maximum of $2(V/\omega_1)$.

C. Exact solution

It is possible to Fourier transform Eq. (10) without making the rotating-wave approximation. We use the same Fourier transformations for $b_m(t)$, $b_n(t)$, and $\delta(t)$ as in Eqs. (14), (15), and (16), respectively, but we set $\omega_1=0$. We separately transform $\cos(\omega_1 t)$ as

$$\cos(\omega_1 t)=\frac{1}{2}\int_{-\infty}^{\infty}[\delta(\omega-\omega_1)+\delta(\omega+\omega_1)]\exp(-i\omega t)\,d\omega. \tag{23}$$

In the first equation of Eq. (10) we have the product of two Fourier transformations. By using the method of convolution we can write

$$\cos(\omega_1 t)\,b_n(t)=-\frac{1}{4\pi i}\exp(i\tfrac{1}{2}\omega_0 t)\int_{-\infty}^{\infty}\left[\int_{-\infty}^{\infty}G_n(\omega-\rho)\{\delta(\rho-\omega_1)+\delta(\rho+\omega_1)\}\,d\rho\right]\exp(-i\omega t)\,d\omega. \tag{24}$$

The integral in the square brackets of Eq. (24) is equal to $G_n(\omega-\omega_1)+G_n(\omega+\omega_1)$. The remaining integral over ω is substituted together with the Fourier transformations of $b_m(t)$ and $\delta(t)$ into the first equation of Eq. (10). The differential equation is satisfied if

$$(\omega+\tfrac{1}{2}\omega_0)G_m(\omega)=\tfrac{1}{2}V[G_n(\omega-\omega_1)+G_n(\omega+\omega_1)]+1. \tag{25}$$

In a similar fashion we can convolute the product of the Fourier transformations of $\cos(\omega_1 t)$ and $b_m(t)$. The substitution of the result together with the Fourier transformations of $b_n(t)$ into the second equation of Eq. (10) yields the relationship

$$(\omega-\tfrac{1}{2}\omega_0)G_n(\omega)=\tfrac{1}{2}V[G_m(\omega-\omega_1)+G_m(\omega+\omega_1)]. \tag{26}$$

For $\omega_1=0$ Eqs. (25) and (26) can be easily solved, and the results give the probabilities of the atomic states coupled by a static field.

One can obtain the rotating-wave approximation by neglecting $G_n(\omega-\omega_1)$ in Eq. (25) and $G_m(\omega+\omega_1)$ in Eq. (26), respectively. The modified equations are

$$
\begin{aligned}
(\omega+\tfrac{1}{2}\omega_0)G_m(\omega)&=\tfrac{1}{2}V\,G_n(\omega+\omega_1)+1,\\
(\omega-\tfrac{1}{2}\omega_0)G_n(\omega)&=\tfrac{1}{2}V\,G_m(\omega-\omega_1).
\end{aligned}
\tag{27}
$$

In order to get an equation for $G_n(\omega)$, one shifts the frequency ω by an amount $-\omega_1$ in the first equation of Eq. (27), and substitutes the resulting expression for $G_m(\omega-\omega_1)$ into the second equation. This procedure yields

$$G_n(\omega)=\frac{\frac{1}{2}V}{(\omega-\omega_1+\frac{1}{2}\omega_0)(\omega-\frac{1}{2}\omega_0)-(\frac{1}{2}V)^2}\,. \tag{28}$$

The poles of Eq. (28) are shifted from those of Eq. (20) by $+\frac{1}{2}\omega_1$. The amplitude $b_n(t)$, however, is the same as the one obtained from Eq. (19) since the Fourier transformation used here differs from the one of Eq. (15) by the factor $\exp(-i\frac{1}{2}\omega_1 t)$.

If we had neglected $G_n(\omega+\omega_1)$ in Eq. (25) and $G_m(\omega-\omega_1)$ in Eq. (26), respectively, the modified equations would have yielded probabilities for the states which do not show a resonance at $\omega_0=\omega_1$ but show one at $\omega_0=-\omega_1$.

Neglecting one of the Fourier transforms on the right side of Eqs. (25) and (26) allows us to make an appropriate shift of the frequency ω and obtain equations for G_m and G_n with the same argument in all the transforms. This is not possible if we use the exact relationships. A particular shift of the frequency ω does not produce an equation in which all the transforms have the same argument. We can, however, find recurrence relationships for G_m and G_n and use them to calculate the amplitudes $b_m(t)$ and $b_n(t)$ to any desired accuracy.

A recurrence relationship for G_m is obtained by first shifting ω in Eq. (26) by $-\omega_1$. This yields an equation for $G_n(\omega-\omega_1)$ in terms of $G_m(\omega-2\omega_1)$ and $G_m(\omega)$. The result is substituted into Eq. (25). Similarly we shift ω by $+\omega_1$ in Eq. (26). This relates the Fourier transform $G_n(\omega+\omega_1)$ to $G_m(\omega)$ and $G_m(\omega+2\omega_1)$.

These substitutions give us

$$\begin{aligned} X_m(\omega'')G_m(\omega) &= (\omega''+\omega_1)(\omega''-\omega_1) \\ &\quad + (\tfrac{1}{2}V)^2[(\omega''+\omega_1)G_m(\omega-2\omega_1) \\ &\quad + (\omega''-\omega_1)G_m(\omega+2\omega_1)], \end{aligned} \tag{29}$$

where we set $\omega''=\omega-\omega_0/2$ and

$$X_m(\omega'') = (\omega''+\omega_0)(\omega''+\omega_1)(\omega''-\omega_1) - (\tfrac{1}{2}V)^2(2\omega''). \tag{30}$$

We can generate another recurrence relationship by shifting ω by $-2\omega_1$ in Eq. (29) and substituting the expression for $G_m(\omega-2\omega_1)$ into the right side of Eq. (29). Shifting ω by $+2\omega_1$ expresses $G_m(\omega+2\omega_1)$ in terms of $G_m(\omega)$ and $G_m(\omega+4\omega_1)$. This results in a new recurrence relationship between $G_m(\omega)$, $G_m(\omega-4\omega_1)$, and $G_m(\omega+4\omega_1)$. The method can be repeated and the substitutions yield an equation relating $G_m(\omega)$, $G_m(\omega-8\omega_1)$, and $G_m(\omega+8\omega_1)$.[14]

We can obtain approximations for $G_m(\omega)$ by neglecting the Fourier transforms with the shifted arguments in a given recurrence relationship. The exact solution for $G_m(\omega)$ is generated by relating the different recurrence relationships.

In a similar fashion we can get exact recurrence relationships for the Fourier transform of the amplitude b_n. The lowest-order one is

$$\begin{aligned} X_n(\omega')G_n(\omega) &= V\omega' + (\tfrac{1}{2}V)^2[(\omega'+\omega_1)G_n(\omega-2\omega_1) \\ &\quad + (\omega'-\omega_1)G_n(\omega+2\omega_1)], \end{aligned} \tag{31}$$

where we defined $\omega'=\omega+\frac{1}{2}\omega_0$ and

$$X_n(\omega') = (\omega'-\omega_0)(\omega'+\omega_1)(\omega'-\omega_1) - (\tfrac{1}{2}V)^2(2\omega'). \tag{32}$$

We illustrate the power of this method by calculating the first- and second-order approximations and indicate the degree of convergence of the results.

IV. FIRST-ORDER APPROXIMATION

The first-order approximation is obtained by neglecting $G_n(\omega-2\omega_1)$ and $G_n(\omega+2\omega_1)$ in Eq. (31). This yields

$$G_n^{(1)}(\omega) = V\omega'/X_n(\omega'). \tag{33}$$

The poles of $G_n^{(1)}$ are obtained by setting $X_n(\omega')$ equal to zero. The solution of the cubic equation in ω' gives

$$\begin{aligned} \Omega_1' &= \tfrac{2}{3}C\cos(\tfrac{1}{3}\phi) + \tfrac{1}{3}\omega_0, \\ \Omega_2' &= \tfrac{2}{3}C\cos(\tfrac{1}{3}\phi + \tfrac{2}{3}\pi) + \tfrac{1}{3}\omega_0, \\ \Omega_3' &= \tfrac{2}{3}C\cos(\tfrac{1}{3}\phi + \tfrac{4}{3}\pi) + \tfrac{1}{3}\omega_0, \end{aligned} \tag{34}$$

where

$$C = [3\omega_1^2 + 6(\tfrac{1}{2}V)^2 + \omega_0^2]^{1/2}, \tag{35}$$

$$\phi = \cos^{-1}\left(\frac{\omega_0^3 + 9\omega_0(\frac{1}{2}V)^2 - 9\omega_0\omega_1^2}{C^3}\right). \tag{36}$$

For small V/ω_1 the pole Ω_1' is near ω_0 and the poles Ω_2' and Ω_3' are near $-\omega_1$ and $+\omega_1$, respectively. In Fig. 1 we have plotted the poles as a function of ω_0/ω_1 for $V=0.3\omega_1$. The anticrossing of the first and third pole near $\omega_0=\omega_1$ produces a resonance in the probability of the state $|\psi_n\rangle$.

The residues of the poles are given by the expression

$$R_n(\Omega_p') = \{G_n^{(1)}(\omega')(\omega'-\Omega_p')\exp[-i(\omega'-\omega_0)t]\}_{\omega'=\Omega_p'} \quad (p=1,2,3), \tag{37}$$

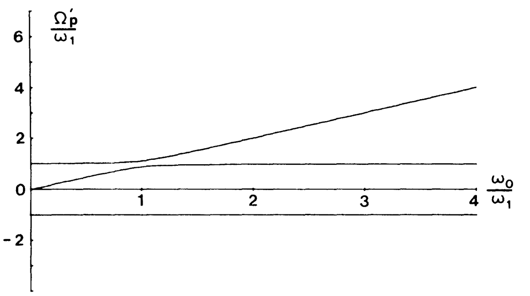

FIG. 1. Poles of $G_n^{(1)}$ as a function of ω_0 for $V=0.3\omega_1$.

and the probability of the atomic state $|\psi_n\rangle$ is

$$|b_n(t)|^2 = \sum_{p,\,q=1}^{3} Q_n(\Omega_p')Q_n(\Omega_q')\cos[(\Omega_p' - \Omega_q')t], \qquad (38)$$

where Q_n is the time-independent part of R_n.

The probability contains constant terms for $p = q$ and oscillatory terms for $p \neq q$. At resonance the probability has a low-frequency variation $(\Omega_1' - \Omega_3')$ which depends strongly on V and two high-frequency contributions with frequencies near $2\omega_0$. The time dependence of $|b_n(t)|^2$ is shown in Fig. 2 for $V = 0.3\omega_1$ and for $\omega_0 = \omega_1$ and $\omega_0 = 1.6\omega_1$, respectively. At resonance the probability reaches a maximum at approximately $t = \pi/V$. This is the same result as was obtained in the rotating-wave approximation. The effect of the pole Ω_2' is evident in the deviation of the curve from the $\sin^2(\frac{1}{2}Vt)$ behavior. The main frequency of oscillation is close to the one given by the rotating-wave approximation. For $\omega_0 = 1.6\omega_1$ the deviation from Eq. (21) is somewhat less pronounced.

If we do not measure the explicit time dependence of the absorption and emission process, we have to average Eq. (38) over time. The terms with $p \neq q$ average to zero and the probability of the excited state becomes

$$|b_n|^2 = \sum_{p=1}^{3} |Q_n(\Omega_p')|^2. \qquad (39)$$

In Fig. 3 we have plotted the probability $|b_n|^2$ as a function of ω_0/ω_1 for $V = 0.3\omega_1$. The curve has a maximum at $\omega_0 \simeq \omega_1$ with a width at half-maximum of roughly $0.6\omega_1$. At the resonance point the average probability is $\frac{1}{2}$, which means that the system is completely saturated.

V. SECOND-ORDER APPROXIMATION

The exact recurrence relationship of Eq. (31) is used as the starting point. We find two new relationships by shifting the frequency ω by $+2\omega_1$ and by $-2\omega_1$, respectively. The first equation relates $G_n(\omega + 2\omega_1)$ to $G_n(\omega)$ and $G_n(\omega + 4\omega_1)$, and the second equation expresses $G_n(\omega - 2\omega_1)$ in terms of $G_n(\omega - 4\omega_1)$ and $G_n(\omega)$. These results are substituted into Eq. (31). The procedure yields a new recurrence relationship between $G_n(\omega)$, $G_n(\omega + 4\omega_1)$, and $G_n(\omega - 4\omega_1)$. Neglecting the Fourier transforms with the shifted frequencies gives us the second-order approximation:

$$G_n^{(2)}(\omega) = \frac{V}{Y_n(\omega')}\{\omega' X_n(\omega' - 2\omega_1)X_n(\omega' + 2\omega_1) + (\tfrac{1}{2}V)^2[(\omega' + \omega_1)(\omega' - 2\omega_1)X_n(\omega' + 2\omega_1) + (\omega' - \omega_1)(\omega' + 2\omega_1)X_n(\omega' - 2\omega_1)]\}. \qquad (40)$$

The function X_n is defined by Eq. (32) and

$$Y_n(\omega') = X_n(\omega')X_n(\omega' - 2\omega_1)X_n(\omega' + 2\omega_1) - (\tfrac{1}{2}V)^4[(\omega' + \omega_1)(\omega' - 3\omega_1)X_n(\omega' + 2\omega_1) + (\omega' - \omega_1)(\omega' + 3\omega_1)X_n(\omega' - 2\omega_1)]. \qquad (41)$$

There are nine poles in Eq. (40) but for two of them the numerator vanishes. The remaining ones are plotted in Fig. 4 as a function of ω_0/ω_1. Comparison with Fig. 1 shows that two of the poles are closely related to the two poles in the first-order approximation which anticross near $\omega_0 = \omega_1$. The third pole in Fig. 1 is replaced in Fig. 4 by two poles which anticross near the main resonance. Two new poles also anticross near $\omega_0 = \omega_1$ and one additional pole is given by the equation $\Omega_p' = -3\omega_1$. In addition to the three anticrossings which are responsible for the main resonance, there are two new anticrossings near $\omega_0 = 3\omega_1$.

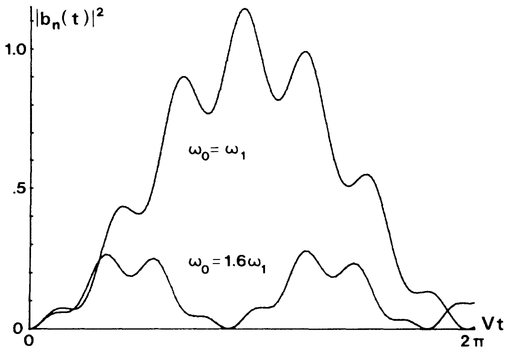

FIG. 2. Probability of state $|\psi_n\rangle$ as a function of time for $\omega_0 = \omega_1$ and $\omega_0 = 1.6\omega_1$ using the first-order approximation. The initial conditions are $b_m(0) = 1$ and $b_n(0) = 0$.

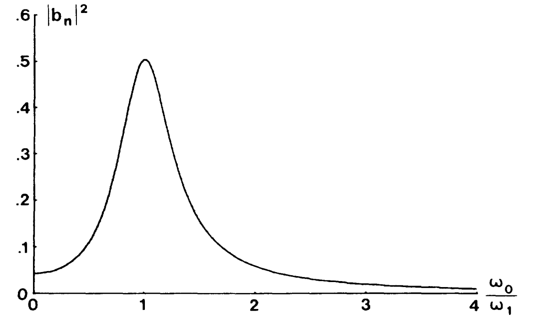

FIG. 3. Time-averaged probability of state $|\psi_n\rangle$ as a function of ω_0 for $V = 0.3\omega_1$ using the first-order approximation.

They produce the three-quantum resonance. There are no anticrossings at $\omega_0 = 2\omega_1$ and thus the system cannot have a two-quantum resonance. It should be noted, however, that at $\omega_0 = 2\omega_1$ the poles have equal separations. The physical significance of this result is discussed in Sec. VI.

The time dependence of the probability of the state $|\psi_n\rangle$ is given by Eq. (38) and is shown in Fig. 5 for $\omega_0 = \omega_1$ and $\omega_0 = 1.6\omega_1$. In order to test the degree of convergence of the first- and second-order approximations, we solved Eq. (10) by a numerical integration. The curves in Fig. 5 are identical with the numerical results.

The time-averaged probability is plotted in Fig. 6 as a function of ω_0/ω_1. The main resonance shows a very small Bloch-Siegert shift. For $V = 0.3\omega_1$ the peak is at $\omega_0 = 0.9998\omega_1$ which should be compared with the result of Ahmad and Bullough.[15] Their calculation gives $\omega_0 = 0.9943\omega_1$. The three-quantum resonance is at $\omega_0 = 2.9717\omega_1$, whereas Ahmad and Bullough find the maximum at $\omega_0 = 2.9916\omega_1$. As can be seen from Fig. 6 both resonances are completely saturated. At the main resonance the probability of the state $|\psi_n\rangle$ is unity at $Vt = \pi$. At the three-quantum resonance the complete exchange of the probability from state $|\psi_m\rangle$ to state $|\psi_n\rangle$ takes about 180 times as long.

VI. DIPOLE MOMENT

Thus far we have examined the behavior of the probability of state $|\psi_n\rangle$. The other quantity of interest in this system is the dipole moment,

$$\begin{aligned} D(t) &= 2b_n(t)\exp(-i\omega_n t)b_m^*(t)\exp(+i\omega_m t) \\ &= 2b_n(t)b_m^*(t)\exp(-i\omega_0 t). \end{aligned} \tag{42}$$

In a magnetic dipole transition, $D(t)$ represents the transverse magnetic moment of the atom. Its x and y components are given, respectively, by the real and imaginary parts of D. In the classical description of an electric dipole transition, $D(t)$ measures the total dipole moment. In the rotating-wave approximation the real and imaginary parts of D can be interpreted as the components in phase and $\pi/2$ out of phase with respect to the applied electric field.

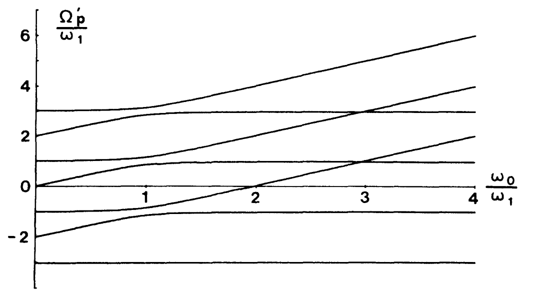

FIG. 4. Poles of $G_n^{(2)}$ as a function of ω_0 for $V = 0.3\omega_1$.

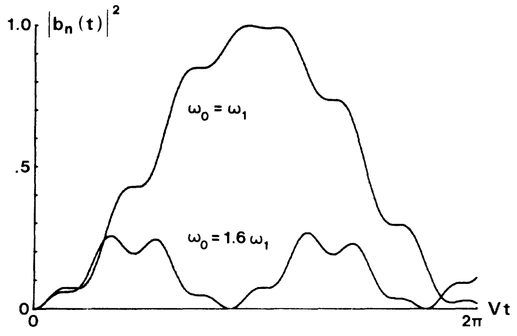

FIG. 5. Probability of state $|\psi_n\rangle$ as a function of time for $\omega_0 = \omega_1$ and $\omega_0 = 1.6\omega_1$ using the second-order approximation. The initial conditions are $b_m(0) = 1$ and $b_n(0) = 0$.

The magnitude of D can be obtained directly from the population of state $|\psi_n\rangle$ using the normalization of the wave function:

$$|D(t)| = 2[|b_n(t)|^2(1 - |b_n(t)|^2)]^{1/2}. \tag{43}$$

The dipole moment vanishes when the system is either in state $|\psi_n\rangle$ or $|\psi_m\rangle$ and reaches a maximum when $|b_n(t)|^2 = |b_m(t)|^2 = 0.5$. The time dependence is essentially an oscillation at twice the flipping frequency (the frequency of population inversion).

Using the notation of Sec. IV we can write the dipole moment as

$$D(t) = 2\sum_{p,q} Q_n(\Omega'_p)Q_m(\Omega''_q)\exp(-i\omega_{pq}t) \tag{44}$$

with

$$\omega_{pq} = \Omega'_p - \Omega''_q - \omega_0. \tag{45}$$

The poles Ω'_p are obtained from Eq. (40) and are plotted in Fig. 4. The poles Ω''_q are those of the

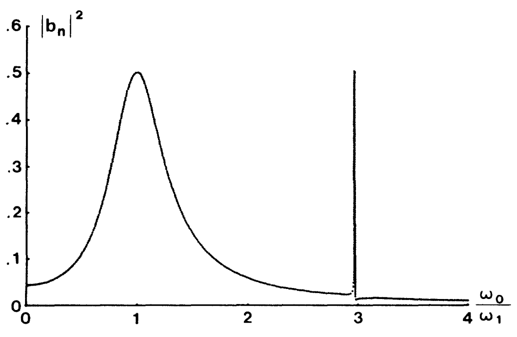

FIG. 6. Time-averaged probability of state $|\psi_n\rangle$ as a function of ω_0 for $V = 0.3\omega_1$ using the second-order approximation.

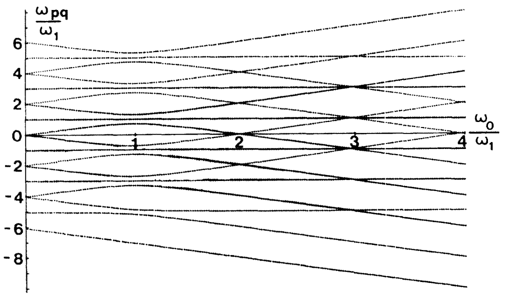

FIG. 7. Evolution frequencies ω_{pq} of the dipole moment as a function of ω_0 for $V=0.3\omega_1$ using the second-order approximation.

Fourier transform $G_m^{(2)}$. They can be derived from the poles of $G_n^{(2)}$ by making the transformation $\omega_0 \rightarrow -\omega_0$. In this form the dipole moment can be analyzed directly in terms of its frequency components. In Fig. 7 the frequencies ω_{pq} are plotted as a function of ω_0/ω_1. The structure is similar to the one shown in Fig. 4.

Anticrossings appear for odd values of ω_0/ω_1. The residues have a resonant behavior around these values and are shown in Fig. 8 for the main resonance and in Fig. 9 for the three-quantum resonance $[D(\omega)=Q_n(\Omega_p')Q_m(\Omega_q'')$ if $\omega=\omega_{pq}$ and $D(\omega)=0$ elsewhere]. At both resonances the dipole moment oscillates at the frequency ω_0. This oscillation is modulated at the flipping frequency. In the main resonance case, the sidebands have opposite signs and almost equal amplitudes. This accounts for the fact that the carrier frequency of the dipole moment is out of phase by about $\pi/2$ with respect to the applied field. In contrast to the results in the rotating-wave approximation the amplitudes are not equal. The counter-rotating part of the field breaks the simple correspondence that occurs in the rotating-wave approximation between the real and imaginary parts of D.

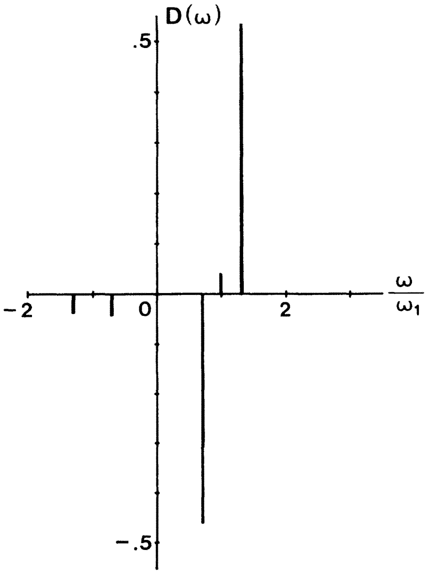

FIG. 8. Frequency spectrum of the dipole moment at the main resonance ($\omega_0=0.9998\omega_1$) for $V=0.3\omega_1$ using the second-order approximation.

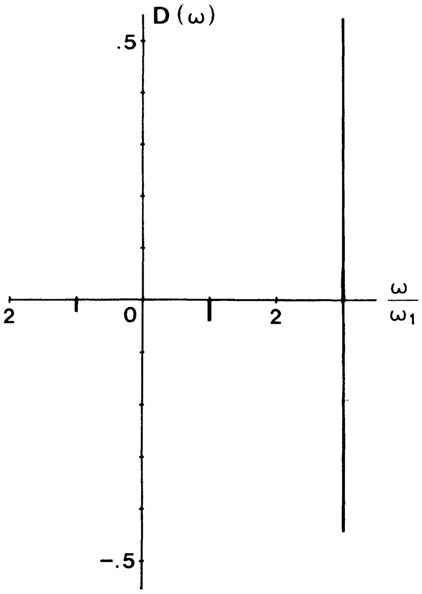

FIG. 9. Frequency spectrum of the dipole moment at the three-quantum resonance ($\omega_0=2.9717\omega_1$) for $V=0.3\omega_1$ using the second-order approximation.

For even values of ω_0/ω_1 the poles Ω_p' and Ω_q'' are equally spaced by the amount ω_0. Equation (45) then implies that one or several frequencies ω_{pq} of the dipole moment are equal to zero. This means that if the system has been initially prepared in a state with $D\neq 0$, part of this dipole moment is conserved for all times. Such a behavior is characteristic of "coherence resonances." These particular resonances are the well-known Haroche resonances.[16]

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