# Frequency-modulated spectra of coherently driven systems

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Frequency-modulated spectra of coherently driven quantum systems are calculated using the density-matrix formulation. The modulated spectra are shown to be related to the two-time correlation functions of the polarization operators of the system. In the limit of modulation frequencies small compared to the width of the spectral features under study, the in-phase signal (quadrature signal) is shown to be related to the derivative of absorption (second derivative of dispersion) in the absence of any modulation. Modulation spectra in strong fields are calculated explicitly for two-level systems; such spectra are shown to yield useful information concerning light shifts and Rabi splittings. The influence of laser temporal fluctuations on such modulation spectra is also discussed.

## I. INTRODUCTION

Frequency-modulation spectroscopy[1-5] has been recently shown to be of great use in probing the spectral features. Relative advantages of using the modulation at frequencies either much smaller or much greater than the width of the spectral features have also been discussed. For example, it has been shown by Bjorklund that both absorption and dispersion associated with a spectral feature can be studied with high degree of sensitivity using frequency-modulation techniques.

In this paper we study theoretically the frequency-modulated spectra of strongly driven systems. We present a density-matrix formulation and obtain analytical expressions for the frequency-modulated spectra. The nature of the frequency-modulation spectra in the limiting cases of large- and small-modulation frequencies is discussed. We show how the Rabi splitting of the energy levels (dynamic Stark effect) could be probed by the frequency-modulation spectroscopy. The treatment we present is fully quantum mechanical and valid generally under the condition of weak modulation. The numerical results for the frequency-modulated spectra are presented in Sec. III. The effect of laser temporal fluctuations on the characteristics of the frequency-modulated spectra is treated in Sec. IV.

## II. GENERAL FORMULATION OF FREQUENCY-MODULATED SPECTRA

In this section we give a general formulation of the frequency-modulated (FM) spectra. We will relate FM spectra to the linear response of the system and to the two-time correlation functions of the system. We present the results in the dipole approximation. The expression for the incident field modulated at frequency $\Omega$ can be written as

$$\vec{E}(t)=\vec{\mathcal{E}}_0^*\sum_{n=-\infty}^{+\infty} J_n(M)\exp[i(\omega_L+n\Omega)t]+\text{c.c.}, \tag{2.1}$$

where $M$ is the modulation index and $J_n$ is the Bessel function. We assume that the modulation is weak so that (2.1) can be approximated by

$$\vec{E}(t)=\vec{E}^{(+)}+\vec{E}^{(-)}, \tag{2.2}$$

where

$$\vec{E}^{(+)}=\vec{\mathcal{E}}_0(1-iM\sin\Omega t)e^{-i\omega_L t}=E^{(-)*}. \tag{2.3}$$

The interaction Hamiltonian in the dipole approximation has the structure

$$H_1=-\vec{P}\cdot\vec{E}(t)\cong-[\vec{P}^{(+)}\cdot\vec{E}^{(-)}(t)+\vec{P}^{(-)}\cdot\vec{E}^{(+)}(t)], \tag{2.4}$$

where $\vec{P}$ is the polarization operator with $\vec{P}^{(\pm)}$ denoting its positive- and negative-frequency components and in writing (2.4) we have used the rotating-wave approximation. The density matrix of the system satisfies

$$\frac{\partial\rho}{\partial t}=L_0\rho-i[H_1(t),\rho], \tag{2.5}$$

where $L_0$ is the unperturbed Liouville operator, giving all other coherent interactions and the incoherent interactions such as radiative decays, collisions, etc. The rate of the absorption of energy from the external field is given by

$$\frac{dW}{dt}=\frac{d\langle\vec{P}\rangle}{dt}\cdot\vec{E}(t). \tag{2.6}$$

Since the modulation index $M$ is weak, we can calculate the response of the polarization to lowest order in $M$. It is clear that the lowest-order response will have the structure

$$\langle\vec{P}(t)\rangle=\vec{A}e^{i(\omega_L+\Omega)t}+\vec{B}e^{i(\omega_L-\Omega)t}+\vec{C}e^{i\omega_L t}+\text{c.c.}, \tag{2.7}$$

where $\vec{A}$ and $\vec{B}$ are linear in $M$ and $\vec{C}$ is independent of $M$. On substituting (2.7), (2.2) in (2.6), carrying out the time averaging over the rapidly oscillating terms such $\exp^{(\pm 2i\omega_L t)}$, $\exp^{[\pm 2i(\omega_L+\Omega)t]}$ and retaining only the terms to lowest order in $M$, we obtain the following result for the rate of absorption of energy $\mathcal{W}$ from the external field:

$$\mathcal{W}=i\omega_L(\vec{C}\cdot\vec{\mathcal{E}}_0-\vec{\mathcal{E}}_0^*\cdot\vec{C}^*)-\omega_L\sin\Omega t[\vec{\mathcal{E}}_0\cdot(\vec{A}-\vec{B}-M\vec{C})+\text{c.c.}]+i\omega_L\cos\Omega t[\vec{\mathcal{E}}_0\cdot(\vec{A}+\vec{B})-\text{c.c.}]$$
$$+i\Omega\cos\Omega t[\vec{\mathcal{E}}_0\cdot(\vec{A}-\vec{B})-\text{c.c.}]-\Omega\sin\Omega t[\vec{\mathcal{E}}_0\cdot(\vec{A}+\vec{B})+\text{c.c.}]. \quad (2.8)$$

Note that the last two terms in (2.8) can be usually ignored as $\omega_L\gg\Omega$. We can thus write for the in-phase ($\cos\Omega t$) component $C$ and the quadrature component $S$, the following:

$$C(\Omega)=i\omega_L[\vec{\mathcal{E}}_0\cdot(\vec{A}+\vec{B})-\text{c.c.}]+i\Omega[\vec{\mathcal{E}}_0\cdot(\vec{A}-\vec{B})-\text{c.c.}],$$
$$S(\Omega)=-\omega_L[\vec{\mathcal{E}}_0\cdot(\vec{A}-\vec{B}-M\vec{C})+\text{c.c.}]-\Omega[\vec{\mathcal{E}}_0\cdot(\vec{A}+\vec{B})+\text{c.c.}]. \quad (2.9)$$

The coefficients $\vec{A}$, $\vec{B}$, and $\vec{C}$ can be calculated from the solution of (2.5). The general characteristics of the spectra depend on the strength of the field $\vec{\mathcal{E}}_0$. We discuss the two cases separately.

### A. Weak external field $\mathcal{E}_0$

For weak external fields it is sufficient to solve (2.5) to lowest order in $\vec{\mathcal{E}}_0$. It is clear that if

$$H_1(t)=\sum_j e^{i\omega_j t}H_j, \quad (2.10)$$

then in the limit of long times, the density matrix will have the structure

$$\rho=\rho^{(0)}+\rho^{(1)}+\cdots,\qquad \rho^{(1)}=\sum_j e^{i\omega_j t}\rho_j^{(1)}, \quad (2.11)$$

with $\rho^{(1)}$ being the solution of

$$\frac{\partial\rho^{(1)}}{\partial t}=L_0\rho^{(1)}-i[H_1(t),\rho^{(0)}(t)]. \quad (2.12)$$

It is easily seen from (2.10)–(2.12) that

$$\rho_j^{(1)}=-i\int_0^\infty d\tau\, e^{L_0\tau-i\omega_j\tau}[H_j,\rho_{\text{st}}^{(0)}], \quad (2.13)$$

where $\rho_{\text{st}}^{(0)}$ is the zeroth-order steady-state solution given by

$$L_0\rho_{\text{st}}^{(0)}=0. \quad (2.14)$$

From (2.13) and (2.11) and using simple properties of quantum mechanical operators, we can write the expectation values of any operator $Q$ in the form

$$\langle Q\rangle=\sum_j ie^{i\omega_j t}\int_0^\infty d\tau\, e^{-i\omega_j\tau}\langle[H_j,Q(\tau)]\rangle, \quad (2.15)$$

where the two-time correlation function $\langle[H_j,Q(\tau)]\rangle$ has to be evaluated in the absence of the external field. On using (2.3), (2.4), and (2.7), we find that the coefficients $\vec{A}$, $\vec{B}$, and $\vec{C}$ are related to the two-time correlation functions of polarization operators in the following way[6]:

$$\vec{C}=-i\int_0^\infty d\tau\, e^{-i\omega_L\tau}\langle[\vec{P}^{(+)}\cdot\vec{\mathcal{E}}_0^*,\vec{P}(\tau)]\rangle, \quad (2.16)$$

$$\vec{A}=\frac{-iM}{2}\int_0^\infty d\tau\, e^{-i(\omega_L+\Omega)\tau}\langle[\vec{P}^{(+)}\cdot\vec{\mathcal{E}}_0^*,\vec{P}(\tau)]\rangle, \quad (2.16a)$$

$$\vec{B}=\frac{+iM}{2}\int_0^\infty d\tau\, e^{-i(\omega_L-\Omega)\tau}\langle[\vec{P}^{(+)}\cdot\vec{\mathcal{E}}_0^*,\vec{P}(\tau)]\rangle. \quad (2.16b)$$

Clearly we have

$$\vec{B}(+\Omega)=-\vec{A}(-\Omega),\quad \frac{\partial\vec{A}}{\partial\Omega}=\frac{\partial\vec{A}}{\partial\omega_L},\quad \frac{\partial\vec{B}}{\partial\Omega}=-\frac{\partial\vec{B}}{\partial\omega_L},$$
$$\vec{A}(\Omega=0)=+\frac{M}{2}\vec{C},\quad \frac{\partial^2\vec{A}}{\partial\Omega^2}=\frac{\partial^2\vec{A}}{\partial\omega_L^2},\quad \frac{\partial^2\vec{B}}{\partial\Omega^2}=\frac{\partial^2\vec{B}}{\partial\omega_L^2}, \quad (2.17)$$

and hence for very small modulation frequencies we get the expansions

$$\vec{A}(\Omega)=\frac{M}{2}\left[\vec{C}+\Omega\frac{\partial\vec{C}}{\partial\omega_L}+\frac{\Omega^2}{2}\frac{\partial^2\vec{C}}{\partial\omega_L^2}+\cdots\right],$$
$$\vec{B}(\Omega)=\frac{-M}{2}\left[\vec{C}-\Omega\frac{\partial\vec{C}}{\partial\omega_L}+\frac{\Omega^2}{2}\frac{\partial^2\vec{C}}{\partial\omega_L^2}\cdots\right]. \quad (2.18)$$

Thus for very small modulation frequencies we get the approximate results

$$C(\Omega)\cong i\omega_L M\Omega\frac{\partial}{\partial\omega_L}(\vec{\mathcal{E}}_0\cdot\vec{C}-\vec{\mathcal{E}}_0^*\cdot\vec{C}^*), \quad (2.19)$$

$$S(\Omega)\cong-\frac{M\Omega^2\omega_L}{2}\frac{\partial^2}{\partial\omega_L^2}(\vec{\mathcal{E}}_0\cdot\vec{C}+\vec{\mathcal{E}}_0^*\cdot\vec{C}^*). \quad (2.20)$$

Note that in the absence of the modulation ($M=0$), the rate of the absorption is related to $\text{Im}\vec{C}\cdot\vec{\mathcal{E}}_0$ [cf. Eq. (2.8)] and the dispersion is related to $\text{Re}\vec{C}\cdot\vec{\mathcal{E}}_0$, since $|C/\mathcal{E}_0|$ effectively gives the complex susceptibility of the system [Eq. (2.7)]. Therefore we have proved *using the density-matrix formulation* that in the limit of very small modulation frequencies, the in-phase signal is related to the derivative of the absorption (in the absence of any modulation) and that the quadrature signal is related to the second derivative of the dispersion.[1,2]

For arbitrary modulation frequencies as is the case discussed in Ref. 4, we have for the two signals

$$C(\Omega)=+iM\omega_L\int_0^\infty d\tau\, e^{-i\omega_L\tau}\sin\Omega\tau \times\langle[\vec{\mathcal{E}}_0\cdot\vec{\mathrm{P}}(\tau),\vec{\mathcal{E}}_0^*\cdot\vec{\mathrm{P}}^{(+)}]\rangle+\text{c.c.}, \tag{2.21}$$

$$S(\Omega)=iM\omega_L\int_0^\infty d\tau\, e^{-i\omega_L\tau}(1-\cos\Omega\tau) \times\langle[\vec{\mathcal{E}}_0\cdot\vec{\mathrm{P}}(\tau),\vec{\mathcal{E}}_0^*\cdot\vec{\mathrm{P}}^{(+)}]\rangle+\text{c.c.} \tag{2.22}$$

The asymmetry appearing in the strucutre of the commutators in (2.21) and (2.22) can be removed if we recall that we have already made the rota ting-wave approximation and hence the commutators in the above can be replaced by

$$\langle[\vec{\mathrm{P}}^{(-)}(\tau)\cdot\vec{\mathcal{E}}_0,\vec{\mathcal{E}}_0^*\cdot\vec{\mathrm{P}}^{(+)}]\rangle,$$

which is the vicinity of a resonant structure at $\omega_0$ with width $\Gamma$, will have the structure

$$\langle[\vec{\mathrm{P}}^{(-)}(\tau)\cdot\vec{\mathcal{E}}_0,\vec{\mathrm{P}}^{(+)}\cdot\vec{\mathcal{E}}_0^*]\rangle=be^{i\omega_0\tau-\Gamma\tau}, \tag{2.23}$$

then the in-phase and quadrature components become

$$C(\Omega)=M\omega_L b\left(\frac{\Gamma}{\Gamma^2+(\omega_0-\omega_L+\Omega)^2}-\frac{\Gamma}{\Gamma^2+(\omega_0-\omega_L-\Omega)^2}\right), \tag{2.24}$$

$$S(\Omega)=-M\omega_L b\left(\frac{2(\omega_0-\omega_L)}{\Gamma^2+(\omega_0-\omega_L)^2}-\frac{(\omega_0-\omega_L+\Omega)}{\Gamma^2+(\omega_0-\omega_L+\Omega)^2}-\frac{(\omega_0-\omega_L-\Omega)}{\Gamma^2+(\omega_0-\omega_L-\Omega)^2}\right). \tag{2.25}$$

If $\Omega\gg\Gamma$, then all the components in the frequency-modulated spectra are well separated and the in-phase signal probes the absorption, whereas the quadrature component probes the dispersion. We have again obtained the result of Bjorklund.[4] Note that our derivation is fully quantum mechanical and the above results are valid for weak external fields only.

### B. Strong external fields

We will now discuss, the nature of the frequency-modulated spectra when $M\ll 1$, but $\vec{\mathcal{E}}_0$ could be strong. In such a case the coefficients $\vec{\mathrm{A}}$, $\vec{\mathrm{B}}$, $\vec{\mathrm{C}}$ [Eq. (2.7)] would be some general functions of $\vec{\mathcal{E}}_0$, and their explicit forms could be calculated provided Eq. (2.5) could be solved in the limit $M=0$. This will be done in the next section for the case of a driven two-level system and it will be shown how the FM spectra[7] could provide us with a convenient method of studying the things like power broadening, dynamic Stark splittings, light shifts, etc. The relations like (2.21) and (2.22) for the spectra can again be obtained by rewriting (2.5) in a frame rotating with the frequency $\omega_L$ of the laser field:

$$\langle\vec{\mathrm{P}}(t)\rangle=\langle\vec{\mathrm{P}}^{(+)}(t)\rangle_f\, e^{-i\omega_L t}+\langle\vec{\mathrm{P}}^{(-)}(t)\rangle_f\, e^{i\omega_L t}, \tag{2.26}$$

$$\frac{\partial\rho_f}{\partial t}=L_f\rho_f-i[H_f(t),\rho_f], \tag{2.27}$$

$$H_f(t)=\frac{M}{2}(e^{i\Omega t}-e^{-i\Omega t})(\vec{\mathrm{P}}^{(-)}\cdot\vec{\mathcal{E}}_0-\vec{\mathrm{P}}^{(+)}\cdot\vec{\mathcal{E}}_0^*), \tag{2.29}$$

where

$$L_f=L_0+i[\vec{\mathrm{P}}^{(-)}\cdot\vec{\mathcal{E}}_0+\vec{\mathrm{P}}^{(+)}\cdot\vec{\mathcal{E}}_0^*,\,]+\cdots, \tag{2.29}$$

where the dots denote detuning terms. On following the procedure which led to (2.15), we find that the first-order terms in the polarization response are

$$\langle\vec{\mathrm{P}}^{(-)}(t)\rangle=ie^{i\omega_L t}\frac{M}{2}\left(e^{i\Omega t}\int_0^\infty d\tau\, e^{-i\Omega\tau}\langle[\vec{\mathrm{P}}^{(-)}\cdot\vec{\mathcal{E}}_0-\vec{\mathrm{P}}^{(+)}\cdot\vec{\mathcal{E}}_0^*,\vec{\mathrm{P}}^{(-)}(\tau)]\rangle-e^{-i\Omega t}\int_0^\infty d\tau\, e^{i\Omega\tau}\langle[\vec{\mathrm{P}}^{(-)}\cdot\vec{\mathcal{E}}_0-\vec{\mathrm{P}}^{(+)}\cdot\vec{\mathcal{E}}_0^*,\vec{\mathrm{P}}^{(-)}(t)]\rangle\right), \tag{2.30}$$

and hence on comparing (2.30) with (2.7) we find that

$$A=i\frac{M}{2}\int_0^\infty d\tau\, e^{-i\Omega\tau}\langle[\vec{\mathrm{P}}^{(-)}\cdot\vec{\mathcal{E}}_0-\vec{\mathrm{P}}^{(+)}\cdot\vec{\mathcal{E}}_0^*,\vec{\mathrm{P}}^{(-)}(\tau)]\rangle, \tag{2.31}$$

$$B=-i\frac{M}{2}\int_0^\infty d\tau\, e^{+i\Omega\tau}\langle[\vec{\mathrm{P}}^{(-)}\cdot\vec{\mathcal{E}}_0-\vec{\mathrm{P}}^{(+)}\cdot\vec{\mathcal{E}}_0^*,\vec{\mathrm{P}}^{(-)}(\tau)]\rangle. \tag{2.32}$$

Thus the in-phase signal will be given by

$$C(\Omega)=+i\omega_L M\int_0^\infty d\tau\sin\Omega\tau\langle[\vec{\mathrm{P}}^{(-)}\cdot\vec{\mathcal{E}}_0-\vec{\mathrm{P}}^{(+)}\cdot\vec{\mathcal{E}}_0^*,\vec{\mathcal{E}}_0\cdot\vec{\mathrm{P}}^{(-)}(\tau)]\rangle+\text{c.c.}. \tag{2.33}$$

## III. FREQUENCY-MODULATED SPECTRA OF A STRONGLY DRIVEN TWO-LEVEL SYSTEM

In this section we investigate the structure of the FM spectra of a strongly driven two-level system of frequency $\omega_0$. The unmodulated spectra associated with such a system have been studied at length both experimentally[12] and theoretically.[13-17] The density-matrix equation (2.5) leads to the following Bloch equations for the expectation values of the dipole-moment operators and the inversion:

$$\frac{\partial\langle S^+\rangle}{\partial t}=\left(i\Delta-\frac{1}{T_2}\right)\langle S^+\rangle-2ig(1+iM\sin\Omega t)\langle S^z\rangle\,,$$

$$\frac{\partial\langle S^-\rangle}{\partial t}=\left(-i\Delta-\frac{1}{T_2}\right)\langle S^-\rangle+2ig(1-iM\sin\Omega t)\langle S^z\rangle\,,$$

$$\frac{\partial}{\partial t}\langle S^z\rangle=-\frac{1}{T_1}(\langle S^z\rangle-\eta)-ig(1-iM\sin\Omega t)\langle S^+\rangle+ig(1+iM\sin\Omega t)\langle S^-\rangle\,,$$

$$g=-\vec{\mathrm{d}}\cdot\vec{\mathscr{E}}_0\,,\quad \Delta=(\omega_0-\omega_L)\,, \tag{3.1}$$

where $\eta$ is the equilibrium value of $\langle S^z\rangle$ in the absence of any external fields. Equations (3.1) are in a frame rotating with frequency $\omega_L$ of the laser field so that the time dependence of the polarization will be

$$\langle\vec{\mathrm{P}}\rangle=\vec{\mathrm{d}}\langle S^+\rangle e^{i\omega_L t}+\vec{\mathrm{d}}\langle S^-\rangle e^{-i\omega_L t}\,, \tag{3.2}$$

where $d$ is the dipole-moment matrix element. In the absence of any modulation ($M=0$) the steady-state solution of (3.1) is given by

$$\langle S^z\rangle=\frac{\eta}{T_1}P^{-1}(0)\left[\left(\frac{1}{T_2}\right)^2+\Delta^2\right]\,,$$

$$\langle S^+\rangle=\frac{\eta}{T_1}P^{-1}(0)(-2ig)\left(\frac{1}{T_2}+i\Delta\right)\,, \tag{3.3}$$

$$P(0)=\frac{4g^2}{T_2}+\frac{1}{T_1}\left[\Delta^2+\left(\frac{1}{T_2}\right)^2\right], \tag{3.4}$$

In the presence of modulation, the first-order corrections to $\langle S^\pm\rangle$, $\langle S^z\rangle$ obey the equation

$$\dot{\psi}^{(1)}=A_0\psi^{(1)}+2gM\sin\Omega t\begin{bmatrix}0&0&1\\0&0&1\\-\frac{1}{2}&-\frac{1}{2}&0\end{bmatrix}\psi^{(0)}\,, \tag{3.5}$$

$$\psi=\begin{bmatrix}\langle S^+\rangle\\\langle S^-\rangle\\\langle S^z\rangle\end{bmatrix}\,,$$

$$A_0=\begin{bmatrix}(i\Delta-1/T_2)&0&-2ig\\0&(-i\Delta-1/T_2)&+2ig\\-ig&ig&-1/T_1\end{bmatrix}\,, \tag{3.6}$$

and hence in steady state

$$\psi^{(1)}(t)=e^{i\Omega t}\psi_+^{(1)}+e^{-i\Omega t}\psi_-^{(1)}\,, \tag{3.7}$$

where

$$\psi_+^{(1)}=-igM(i\Omega-A_0)^{-1}\begin{bmatrix}0&0&1\\0&0&1\\-\frac{1}{2}&-\frac{1}{2}&0\end{bmatrix}\psi^{(0)}(\infty)\,, \tag{3.8}$$

$$\psi_-^{(1)}=-\psi_+^{(1)}(\Omega\to-\Omega)\,. \tag{3.9}$$

On using (3.2)–(3.9) and comparing with (2.7), we find that

$$\vec{\mathrm{A}}=(-igM)\vec{\mathrm{d}}\{[(i\Omega-A_0)^{-1}_{11}+(i\Omega-A_0)^{-1}_{12}]\psi_3^{(0)}(\infty)-\tfrac{1}{2}(i\Omega-A_0)^{-1}_{13}[\psi_1^{(0)}(\infty)+\psi_2^{(0)}(\infty)]\}\,, \tag{3.10}$$

$$\vec{\mathrm{B}}=-A(\Omega\to-\Omega)\,, \tag{3.11}$$

$$\vec{\mathrm{C}}=\frac{\eta}{T_1}P^{-1}(0)(-2ig)d\left(\frac{1}{T_2}+i\Delta\right)\,. \tag{3.12}$$

On substituting (3.10)–(3.12) in (2.9) and on simplification, we find the following results for the in-phase and quadrature components:

$$C(\Omega)=4g^2M\Delta\omega_L\,\mathrm{Im}L(i\Omega)-4g^2M\Omega\,\mathrm{Re}K(i\Omega)\,, \tag{3.13}$$

$$S(\Omega)=4Mg^2\Omega\,\mathrm{Im}K(i\Omega)-4g^2M\Delta\omega_L\left(\frac{\eta}{T_1}P^{-1}(0)-\mathrm{Re}L(i\Omega)\right), \tag{3.14}$$

where

$$K(Z)=P^{-1}(Z)P^{-1}(0)\frac{\eta}{T_1}\left\{4g^2\left(\frac{1}{T_2}\right)^2+\left(Z+\frac{1}{T_2}\right)\left(Z+\frac{1}{T_1}\right)\left[\Delta^2+\left(\frac{1}{T_2}\right)^2\right]\right\},$$

$$L(Z)=P^{-1}(Z)P^{-1}(0)\frac{\eta}{T_1}\left\{4g^2\left(Z+\frac{1}{T_2}\right)+\left(Z+\frac{1}{T_1}\right)\left[\Delta^2+\left(\frac{1}{T_2}\right)^2\right]\right\}, \tag{3.15}$$

$$P(Z)=4g^2\left(Z+\frac{1}{T_2}\right)+\left(Z+\frac{1}{T_1}\right)\left[\Delta^2+\left(Z+\frac{1}{T_2}\right)^2\right].$$

Note that the unmodulated terms $\mathcal{W}^{(0)}$ in the energy absorption [formula (2.8)] are

$$\mathcal{W}^{(0)} = i\omega_L(\vec{C}\cdot\vec{\mathcal{E}}_0) - \vec{\mathcal{E}}_0^{*}\cdot\vec{C}^{*})$$

$$= \left(\frac{-\eta}{T_2}\right)(4g^2\omega_L)\frac{1}{\left[\Delta^2 + \left(\frac{1}{T_2}\right)^2 + 4g^2\frac{T_1}{T_2}\right]}\,, \quad (3.16)$$

which shows the usual power broadening.[18] The analytical results (3.13), (3.14) are valid for arbitrary values of the strength of the field $g$ and $T_1$, $T_2$. For very small values of the modulation frequencies $\Omega \ll 1/T_1, 1/T_2$, it is seen from (3.15) that

$$C(\Omega) \approx 8g^2 M \mathcal{W}_L \Delta(-\eta/T_1^2 T_2)\Omega P^{-2}(0)$$

$$= M\Omega\frac{\partial}{\partial\omega_L}\mathcal{W}^{(0)}(\omega_L)\,. \quad (3.17)$$

Hence the in-phase signal for the case of a two-level system is still proportional to the derivative of the zeroth-order absorption, irrespective of the strengh of the external field. Note further that if the external laser is exactly on resonance $\Delta = 0$, then

$$C(\Omega) = -4g^2 M\Omega\frac{\eta}{T_1 T_2^2}P^{-1}(0)\left(\frac{T_2^{-1}}{\Omega^2 + (1/T_2)^2}\right)\,, \quad (3.18)$$

$$S(\Omega) = -4g^2 M\Omega\frac{\eta}{T_1 T_2^2}\,P^{-1}(0)\frac{\Omega}{\Omega^2 + \left(\frac{1}{T_2}\right)^2}\,. \quad (3.19)$$

Note that the signals $C(\Omega)$ and $S(\Omega)$ for $\Delta = 0$ are in general very small as these are proportional to the modulation frequency $\Omega$. Such signals probe the dressed-atom structure of the atom for $\Delta = 0$, which corresponds to the frequencies $\omega_L \pm 2g$, $\omega_L$ and with widths $\frac{1}{2}(1/T_1 + 1/T_2)$ and $1/T_2$, respectively. The structure of the frequency-modulated spectra is such that only the central frequency $\omega_L$ makes a contribution. It may be interesting to compare this structure with the structure of the fluorescence in the amplitude-modulated fields.

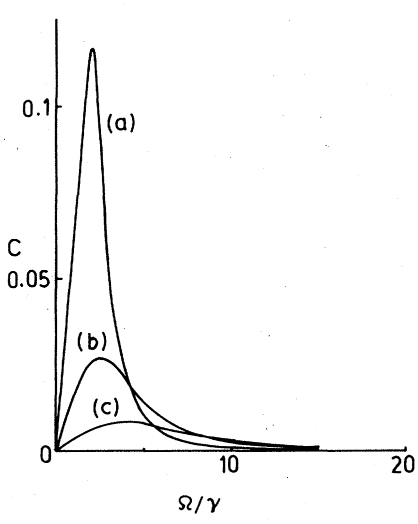


FIG. 1. The in-phase signal as a function of modulation frequency $\Omega$ for weak fields $g = 0.1\gamma$, $1/T_2 = (\gamma + \gamma_c)$, $1/T_1 = 2\gamma$, $\Delta = 2\gamma$ with (a) $\gamma_c = 0$, (b) $\gamma_c = 2\gamma$, (c) $\gamma_c = 5\gamma$; $2\gamma$ being the Einstein $A$ coefficient for the transition.

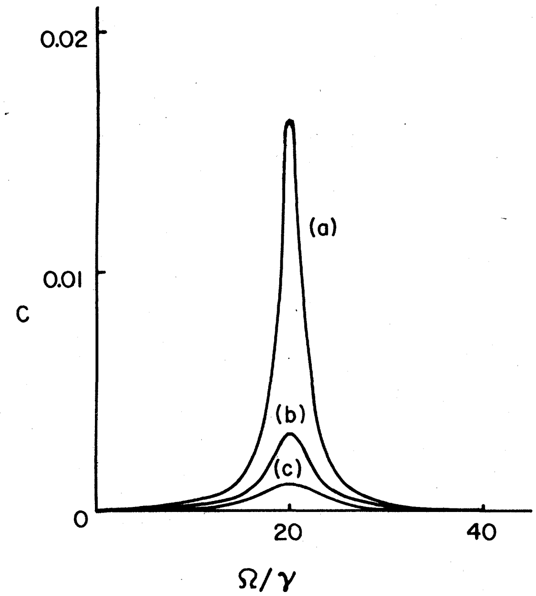


FIG. 2. Same as Fig. 1, but now the external field is strong $g = 10\gamma$.

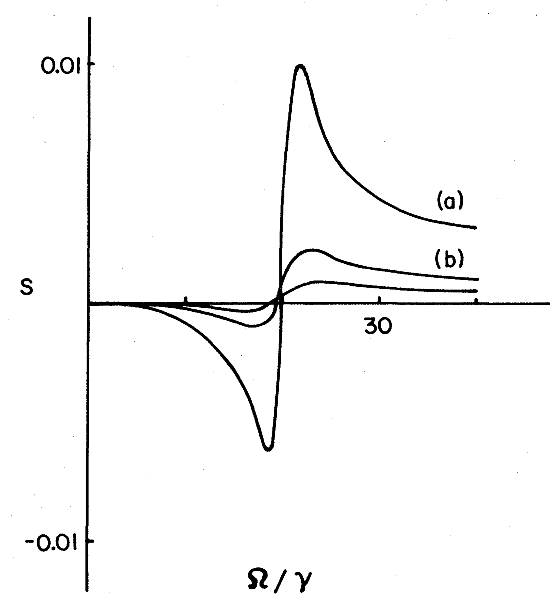


FIG. 3. The quadrature signal as a function of $\Omega$ for strong fields $g = 10\gamma$; other parameters are the same as in Fig. 1.

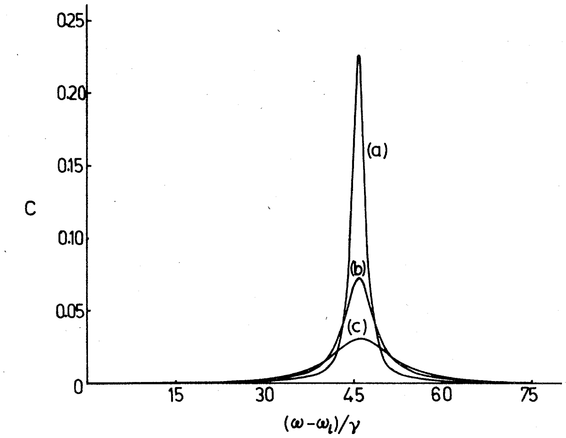


FIG. 4. The in-phase signal as a function of $\omega_L$ for large modulation fequencies $\Omega = 50\gamma$ and for $g = 10\gamma$, $1/T_2 = (\gamma + \gamma_c)$, $1/T_1 = 2\gamma$, and (a) $\gamma_c = 0$, (b) $\gamma_c = 2\gamma$, (c) $\gamma_c = 5\gamma$.

Such a modulated fluorescence is known[11] to have a contribution only" from the sidebands $\omega_L \pm 2g$. The signals $C(\Omega)$ and $S(\Omega)$ are shown in Figs. 1–5 for various values of the relaxation parameters and the field strengths. In Fig. 1–3 we have plotted the dimensionless quantities $C(\Omega)T_1/(4g^2\Delta M\omega_L\eta T_2^3)$, $S(\Omega)T_1/(4g^2\Delta M\omega_L\eta T_2^3)$, whereas in Figs. 4 and 5, we have plotted dimensionless quantities $C(\Omega)T_1/(4g^2M\omega_L\eta T_2^2)$ and $S(\Omega)T_1/(4g^2M\omega_L\eta T_2^2)$. The signals could be studied either as a function of modulation frequency [Figs. 1–3] or as a function of the laser frequency $\omega_L$ (or equivalently $\Delta$) [Figs. 4 and 5]. Figure 1 gives the inphase signal $C(\Omega)$ as a function of $\Omega$ for weak fields $g \ll 1/T_2$ and for various values of $1/T_2$. It is easily seen from (3.15) that the peaks will be at $\Omega \approx \pm\Delta$ with width $1/T_2$. Figures 2 and 3 give the

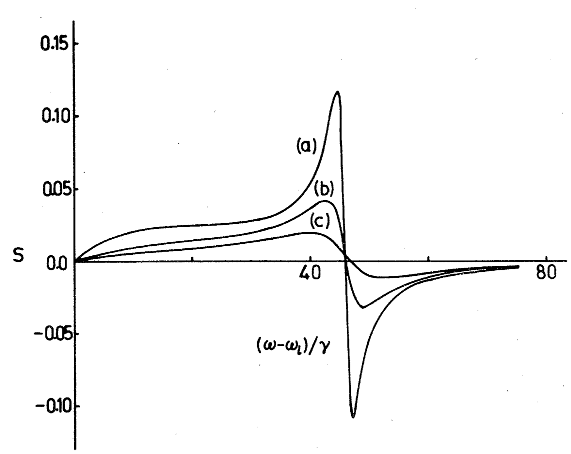


FIG. 5. The quadrature signal as a function of $\omega_L$; parameters are the same as in Fig. 4.

shape of $C(\Omega)$ and $S(\Omega)$ for the strong-field case. $C(\Omega)$ [$S(\Omega)$] has the absorption [dispersion] structure. It is clear from Figs. 2 and 3 that the two signals can be effectively used to study Rabi splittings. Increase of $T_2^{-1}$ reduces the peak heights and considerably broadens the signals. This again could be understood in terms of the roots of $P(z) = 0$, which are approximately given by[19] (for $g \gg 1/T_1, 1/T_2$)

$$-\frac{1}{T_2} + \left(\frac{1}{T_2} - \frac{1}{T_1}\right)\frac{\Delta^2}{\Delta^2 + 4g^2},$$

$$-\frac{1}{T_2} + \frac{1}{2}\left(\frac{1}{T_2} - \frac{1}{T_1}\right)\frac{4g^2}{4g^2 + \Delta^2} \pm i(4g^2 + \Delta^2)^{1/2}.$$

Figures 4 and 5 give the shape of $C(\Omega)$ and $S(\Omega)$, for the strong-field case, as a function of $\omega_L$ or the detuning parameter $\Delta$ for fixed value of $\Omega$. In the spirit of the work by Bjorklund,[4] the modulation frequency has been taken to be very large compared with the width of the structure $1/T_1$, $1/T_2$. The FM spectra again yield information regarding the Rabi splittings, cf. the peaks in Fig. 4 and the position of the dispersionlike structure in Fig. 5. The behavior can again be understood in terms of the roots of $P(i\Omega) = 0$, i.e., the value of $\Delta$ for which $P(i\Omega) = 0$. The $\Delta = 0$ peak in Figs. 4 and 5 is not seen because of the prefactor multiplying the expressions for $S$ and $C$. The other dispersionlike structure in Fig. 5 and the absorptionlike structure in Fig. 4 gives the Rabi splittings. The width of the peak at $\Delta \sim (\Omega^2 - 4g^2)^{1/2}$ in Fig. 4 is of the order of $\Omega/T_2(\Omega^2 - 4g^2)^{1/2} \sim 1/T_2$.

## IV. EFFECT OF LASER TEMPORAL FLUCTUATIONS ON THE FM SPECTRA

We finally discuss the influence of laser temporal fluctuations on the frequency-modulated spectra. The laser temporal fluctuations could be incorporated in the theory by using the phase-diffusion model of the laser,[20] i.e., rewriting the field as

$$\vec{E}^{(+)}(t) = \vec{\mathscr{E}}_0(1 - iM\sin\Omega t)e^{-i\omega_L t - i\Phi(t)},$$
$$\dot{\Phi}(t) = \mu(t), \quad \langle\mu(t_1)\mu(t_2)\rangle = 2\gamma_c\delta(t_1 - t_2), \qquad (4.1)$$
$$\langle\mu(t)\rangle = 0,$$

where $\mu(t)$ is the delta-correlated Gaussian random process. It is evident from the structure of (2.4), (2.6), and (4.1) that we should now calculate the linear response of the operator

$$\vec{P}^{(+)}(t)e^{+i\Phi(t)} + \vec{P}^{(-)}(t)e^{-i\Phi(t)},$$

instead of just the polarization operator $\vec{p}(t)$. In place of (2.7) we now write

$$\langle\langle \vec{P}^{(-)}e^{-i\Phi(t)} + \vec{P}^{(+)}e^{i\Phi(t)} \rangle\rangle = \vec{A}e^{i(\omega_L + \Omega)t} + \vec{B}e^{i(\omega_L - \Omega)t} + \vec{C}e^{i\omega_L T} + \text{c.c.}, \quad (4.2)$$

where the second bracket $\langle\langle\ \rangle\rangle$ now denotes the ensemble averaging over the stochastic distribution of $\Phi$. The rate of absorption of energy from the external field is again given by (2.8). The coefficients $\vec{A}$, $\vec{B}$, and $\vec{C}$ needed in the calculation of the energy absorption could be obtained from (2.12), by following the analysis similar to that which led to (2.21) and (2.22). The final results for the in-phase and quadrature components are

$$C(\Omega)=iM\omega_L\int_0^\infty d\tau\, e^{-i\omega_L\tau-\gamma_c\tau}\sin\Omega\tau\langle[\vec{P}^{(-)}(\tau)\cdot\vec{\mathcal{E}}_0,\vec{P}^{(+)}\cdot\vec{\mathcal{E}}_0^*]\rangle+\text{c.c.}\ , \tag{4.3}$$

$$S(\Omega)=iM\omega_L\int_0^\infty d\tau\, e^{-i\omega_L\tau-\gamma_c\tau}(1-\cos\Omega\tau)\langle[\vec{P}^{(-)}(\tau)\cdot\vec{\mathcal{E}}_0,\vec{P}^{(+)}\cdot\vec{\mathcal{E}}_0^*]\rangle+\text{c.c.} \tag{4.4}$$

In such a case the laser linewidth simply adds to the linewidth of each structure.

The strong-field results could be obtained by using the theory of multiplicative stochastic processes.[20] It can be shown using the same procedure as in Ref. 20, that in place of (3.1), one now has the equations

$$\begin{aligned}
\frac{\partial\langle\langle S^+e^{-i\Phi(t)}\rangle\rangle}{\partial t}&=\left(i\Delta-\frac{1}{T_2'}\right)\langle\langle S^+e^{-i\Phi(t)}\rangle\rangle-2ig(1+iM\sin\Omega t)\langle\langle S^z\rangle\rangle\,,\\
\frac{\partial}{\partial t}\langle\langle S^z\rangle\rangle&=-\frac{1}{T_1}\langle\langle S^z-\eta\rangle\rangle-ig(1-iM\sin\Omega t)\langle\langle S^+e^{-i\Phi(t)}\rangle\rangle+ig(1+iM\sin\Omega t)\langle\langle S^-e^{i\Phi(t)}\rangle\rangle,\quad \frac{1}{T_2'}=\frac{1}{T_2}+\gamma_c\,.
\end{aligned} \tag{4.5}$$

These equations have the same structure as (3.1) but with $1/T_2\rightarrow 1/T_2'=1/T_2+\gamma_c$. In view of this, the results (3.13), (3.14) for the in-phase and quadrature components are valid in the presence of laser temporal fluctuations provided we make the replacement

$$\frac{1}{T_2}\rightarrow\frac{1}{T_2'}=\frac{1}{T_2}+\gamma_c\,. \tag{4.6}$$

The variation of the signals with $\gamma_c$ can be obtained from Figs. 1–5 if we keep in mind (4.6). In general increasing $\gamma_c$ reduces the peak heights and broadens the signals.

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