Fluorescence in frequency-modulated beams: a probe of the correlation functions of atomic inversion

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A general formulation of the fluorescence signal in frequency-modulated beams is given. Such a signal is shown to probe the two time-correlation functions of atomic-inversion operators. Explicit results for the modulated fluorescence are given for the two-level model under arbitrary conditions.

1. INTRODUCTION

The correlation functions of the dipole-moment operators play a key role[1] in physics, as the absorption line shapes are determined by such correlations. A considerable body of literature has been devoted to the study of dipole–dipole correlations.[2,3] In resonant optics, however, there are other physical variables that characterize the system, for example, atomic inversion. However, atomic inversion does not couple directly with the external field, unlike the case of a spin in a magnetic field, in which the longitudinal field couples with the z component of the spin. Questions that do not seem to have received any attention are: What are the correlations of the atomic-inversion operators, and how can such correlations be probed? One might be tempted to think that the fluorescence spectra[3] might be related to such correlations, since the intensity of fluorescence is proportional to the atomic inversion. However, that is not the case. In this paper we show how the fluorescence in frequency-modulated (FM) beams[4–6] provides answers to the above questions. This is again in contrast to the case of spin in a magnetic field for which the longitudinal susceptibility probes the correlations of the type $\langle [S^z(t), S^z(0)]\rangle$. The organization of this paper is as follows: In Section 2, we present a general formulation and show how the fluorescence in FM beams provides a useful probe of the correlation functions of the atomic-inversion operators.[7] In Section 3, we calculate explicitly the modulated component of the fluorescence for the case of a two-level system interacting with the FM field. Numerical results for the resulting line shapes are also presented in Section 3. Generalizations to other situations are briefly discussed.

2. GENERAL FORMULATION OF FLUORESCENCE IN FREQUENCY-MODULATED BEAMS

Consider a quantum-mechanical system interacting with a FM field:

$$\mathbf{E}(t) = \epsilon_0 \exp[-i\omega t - i\Phi(t)] + \text{c.c.}, \qquad (2.1)$$

where

$$\dot{\Phi}(t) = \Omega M \cos \Omega t. \qquad (2.2)$$

Here, M is the modulation index and Ω is the frequency of the modulation. Assuming arbitrary relaxation (in impact approximation), the density matrix ρ satisfies the equation of motion

$$\partial\rho/\partial t = L_0\rho - i[H_1(t), \rho], \qquad (2.3)$$

where, in the terms of the dipole-moment operator $\mathbf{d}$, one has

$$H_1(t) = -\mathbf{d} \cdot \mathbf{E}(t). \qquad (2.4)$$

In Eq. (2.3), L_0 is the relaxation operator having the structure

$$(L_0\rho)_{ij} = (-i\omega_{ij} - \Gamma_{ij})\rho_{ij} \qquad i \neq j,$$
$$(L_0\rho)_{ii} = -\sum_j \gamma_{ji}\rho_{ii} + \sum_j \gamma_{ij}\rho_{jj}, \qquad (2.5)$$

where various parameters Γ_{ij}, γ_{ij} have the usual meaning. Phase-interrupting collisions are included in Γ_{ij}. We will also make the rotating-wave approximation,[8] so that we approximate Eq. (2.4) by

$$H_1(t) = -\sum_{ij} \mathbf{d}_{ij} \cdot \epsilon_0 |i\rangle\langle j| \exp[-i\omega t - i\Phi(t)] + \text{H.c.}$$
$$= h \exp[-i\omega t - i\Phi(t)] + \text{H.c.}, \qquad \omega_{ij} > 0 \qquad (2.6)$$

where the summation is only over those energy levels such that $|\omega_{ij}| \sim \omega_0$. On making the transformation to the rotating frame

$$\rho_{ij}(t) = \tilde{\rho}_{ij}(t) \exp\{-i[\omega t + \Phi(t)]\} \qquad i \neq j, \omega_{ij} > 0, \qquad (2.7)$$

we obtain from Eq. (2.3)

$$\partial\tilde{\rho}/\partial t = -i[h + h^+, \tilde{\rho}] + L_0\tilde{\rho} + i[\omega + \Omega M \cos \Omega t][B, \tilde{\rho}]. \qquad (2.8)$$

Here, B is a diagonal operator having nonvanishing elements such that

$$[B, \tilde{\rho}]_{ij} = +\tilde{\rho}_{ij} \quad \text{if } \omega_{ij} > 0 \qquad i \neq j,$$
$$= -\tilde{\rho}_{ij} \quad \text{if } \omega_{ij} < 0. \qquad (2.9)$$

We are now in a position to obtain the system's response to

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various orders in the modulation index M. For this purpose we rewrite Eq. (2.8) as

$$\partial\tilde{\rho}/\partial t = \alpha\tilde{\rho} + i\Omega M \cos \Omega t[B, \tilde{\rho}]. \quad (2.10)$$

Hence, in the long-time limit, we will have to first order in M

$$\tilde{\rho}(t) \rightarrow \tilde{\rho}^{(0)} + \frac{i\Omega M}{2}\{\exp(i\Omega t)(i\Omega - \alpha)^{-1}[B, \tilde{\rho}^{(0)}] + \exp(-i\Omega t)(-i\Omega - \alpha)^{-1}[B, \tilde{\rho}^{(0)}]\}. \quad (2.11)$$

Therefore, to first order in the modulation index M, the population of the level $|i\rangle$ is given by

$$\rho_{ii}(t) = M \cos \Omega t C_i(\Omega) + M \sin \Omega t S_i(\Omega), \quad (2.12)$$

where

$$C_i(\Omega) = -\Omega\, \mathrm{Im}(\langle i|\{(-i\Omega - \alpha)^{-1}[B, \tilde{\rho}^{(0)}]\}|i\rangle), \quad (2.13)$$

$$S_i(\Omega) = +\Omega\, \mathrm{Re}(\langle i|\{(-i\Omega - \alpha)^{-1}[B, \tilde{\rho}^{(0)}]\}|i\rangle). \quad (2.14)$$

The expectation values on the right-hand sides of the above equations can be expressed as the time-correlation functions of the inversion operators. One obviously has

$$\begin{aligned}\langle i|(-i\Omega - \alpha)^{-1}[B, \tilde{\rho}^{(0)}]|i\rangle &= \int_0^\infty d\tau \langle i|\{\exp(+i\Omega\tau + \alpha\tau)[B, \tilde{\rho}^{(0)}]\}|i\rangle \\ &= \int_0^\infty d\tau\, \mathrm{Tr}\,(\{\exp(i\Omega\tau + \alpha\tau)[B, \tilde{\rho}^{(0)}]\}|i\rangle\langle i|) \\ &= \int_0^\infty d\tau \exp(i\Omega\tau)\langle[(|i\rangle\langle i|)_\tau, B]\rangle. \end{aligned} \quad (2.15)$$

Note that the fluorescence from the ith level will be proportional to $\rho_{ii}(t)$. We have thus shown the relationship of the modulated component of fluorescence in FM beams to the correlation functions of the operators corresponding to the population of the various levels

$$C_i(\Omega) = -\Omega\, \mathrm{Im} \int_0^\infty d\tau \exp(i\Omega\tau)\langle[|i\rangle\langle i|_\tau, B]\rangle, \quad (2.16)$$

$$S_i(\Omega) = +\Omega\, \mathrm{Re} \int_0^\infty d\tau \exp(i\Omega\tau)\langle[|i\rangle\langle i|_\tau, B]\rangle. \quad (2.17)$$

We now discuss some special cases.

A. Two-Level Transition

Consider a two-level system with states $|1\rangle$ and $|2\rangle$ with energy separation ω_0 interacting with a FM beam. In such a case one obviously has

$$B = |1\rangle\langle 1|, \qquad |i\rangle \equiv |1\rangle, \quad (2.18)$$

and hence

$$C_i(\Omega) = -\Omega\, \mathrm{Im} \int_0^\infty d\tau \exp(i\Omega\tau)\langle[(|1\rangle\langle 1|)_\tau, (|1\rangle\langle 1|)_0]\rangle,$$

$$S_i(\Omega) = +\Omega\, \mathrm{Re} \int_0^\infty d\tau \exp(i\Omega\tau)\langle[(|1\rangle\langle 1|)_\tau, (|1\rangle\langle 1|)_0]\rangle. \quad (2.19)$$

The correlation functions of Eqs. (2.19) can also be expressed in terms of the inversion operator

$$S^z \equiv \tfrac{1}{2}(|1\rangle\langle 1| - |2\rangle\langle 2|) \quad (2.20)$$

with the result that

$$C_i(\Omega) = -\Omega\, \mathrm{Im} \int_0^\infty d\tau \exp(i\Omega\tau) \langle[S^z(\tau), S^z(0)]\rangle,$$

$$S_i(\Omega) = +\Omega\, \mathrm{Re} \int_0^\infty d\tau \exp(i\Omega\tau) \langle[S^z(\tau), S^z(0)]\rangle. \quad (2.21)$$

We thus obtain one of the key results of this paper: The fluorescence in the FM beams probes the autocorrelation function of the inversion operators of the system.

B. Three-Level (Λ) System

As another example of the modulated fluorescence in FM beams, we now consider a Λ system with energy levels $|1\rangle$, $|2\rangle$, and $|3\rangle$ and with allowed transitions $|1\rangle \leftrightarrow |2\rangle$ and $|1\rangle \leftrightarrow |3\rangle$. Here $|1\rangle$ represents the topmost state. For this Λ system, one again obtains the result [Eqs. (2.19)] for the modulated fluorescence. The correlation function in the present case will have contributions from both decay channels $|1\rangle \rightarrow |2\rangle$ and $|1\rangle \rightarrow |3\rangle$.

3. MODULATED FLUORESCENCE FROM A TWO-LEVEL SYSTEM IN FREQUENCY-MODULATED BEAMS

In this section we calculate the modulated fluorescence from a two-level system interacting with a field of arbitrary strength. Bjorklund *et al.*[5] have already demonstrated the usefulness of the modulated fluorescence in such a situation. This calculation can be easily done using the Bloch equations and Eqs. (2.21). The Bloch equations read

$$\frac{d}{dt}\begin{bmatrix}\langle S^+\rangle \\ \langle S^-\rangle \\ \langle S^z\rangle\end{bmatrix} = \begin{bmatrix} i\Delta - \dfrac{1}{T_2} & 0 & -2ig \\ 0 & -i\Delta - \dfrac{1}{T_2} & +2ig \\ -ig & +ig & -\dfrac{1}{T_1}\end{bmatrix}\begin{bmatrix}\langle S^+\rangle \\ \langle S^-\rangle \\ \langle S^z\rangle\end{bmatrix} + \begin{bmatrix}0 \\ 0 \\ \dfrac{\eta}{T_1}\end{bmatrix}, \quad (3.1)$$

$$\Delta = \omega_0 - \omega.$$

If one uses Eqs. (3.1) and, say, the regression theorem, one can show that[9]

$$\hat{R}(z) \equiv \int_0^\infty d\tau \exp(-z\tau)\langle[S^z(\tau), S^z(0)]\rangle$$
$$= \frac{-4i\Delta g^2\eta}{T_1} P^{-1}(0)P^{-1}(z)\left(z + \frac{2}{T_2}\right), \quad (3.2)$$

where

$$P(z) = 4g^2\left(z + \frac{1}{T_2}\right) + \left(z + \frac{1}{T_1}\right)\left[\Delta^2 + \left(z + \frac{1}{T_2}\right)^2\right]. \quad (3.3)$$

The in-phase and the quadrature components of the signal are then

$$C_i(\Omega) = -\Omega\, \mathrm{Im}\, \hat{R}(-i\Omega),$$
$$S_i(\Omega) = +\Omega\, \mathrm{Re}\, \hat{R}(-i\Omega). \quad (3.4)$$

In Figs. 1 and 2 we display the behavior of $C_i(\Omega)$ and $S_i(\Omega)$ as a function of the modulation frequency Ω and the detuning Δ. The results are given for both weak and strong fields. T_1, T_2, and η are taken to correspond to the radiative relaxation. Figure 1 shows the usual absorption- and dispersion-shaped resonances around $\Omega = 0$ for weak fields, whereas for strong fields we find that the resonant structures correspond to the dynamical Stark splitting. Figure 2 gives the scan as a function of detuning. The signals are odd functions of Δ, as is evident from Eq. (3.2). The resonances correspond to $\Omega^2 = \Delta^2 + 4g^2$.

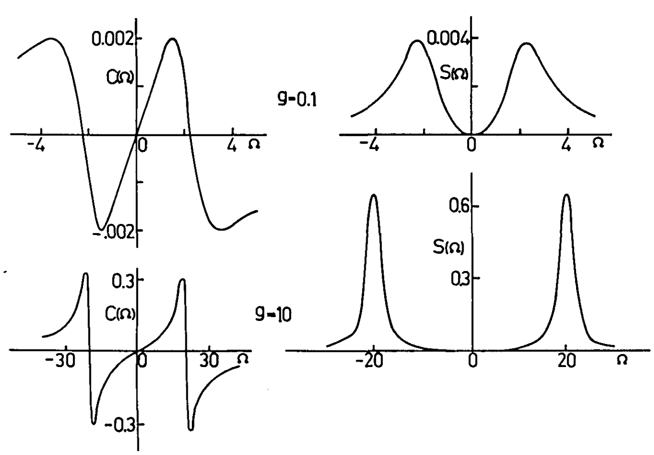

Fig. 1. The in-phase $C(\Omega)$ and out-of-phase $S(\Omega)$ components of the modulated fluorescence as a function of the modulation frequency for the radiative relaxation for weak fields ($g = 0.1$) and for strong fields ($g = 10.0$) with detuning $\Delta = 2$. All parameters are in units of $1/T_2$.

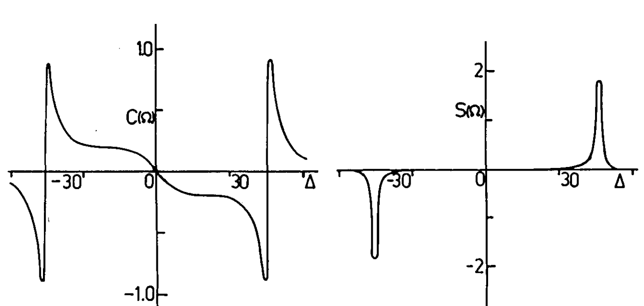

Fig. 2. C and S as a function of the detuning for large modulation frequency $\Omega = 50\ \lambda$ and for $g = 10$.

Similar results can be obtained for the Λ system by using the dynamical equations of Ref. 10. Finally, we mention that the fluorescence from a two-level system with the two levels connected by a two-photon transition can also be studied by similar methods. It is well known[11] that the dynamics in this situation can also be described by Bloch equations with

$$\epsilon(t) \rightarrow \epsilon^2(t) = \epsilon_0^2 \exp[-2i\omega_l t - 2i\Phi(t)] \quad (3.5)$$

and with the inclusion of the Stark shift, which depends on $|\epsilon(t)|^2$. The Stark-shift term is independent of the modulation. Hence the FM fluorescence that probes two-photon transitions of a two-level system in FM beams will be given by Eqs. (3.4) with $\Delta \rightarrow \omega_0 - 2\omega_l -$ (Stark shift), $M \rightarrow 2M$, and $g \rightarrow 2$ photon matrix element.

In conclusion, we have shown how the correlation functions of the atomic inversion can be probed by using fluorescence in FM beams. This result is especially important as the atomic-inversion operator does not couple directly with the external field.

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7. It should be borne in mind that what we are proposing is an interesting application of the known technique rather than a new spectroscopic technique.
8. Note that ω is the optical frequency, Ω is typically in the megahertz-to-gigahertz range,[6] and hence $\Omega \ll \omega$. Modulation is taken to be weak, $M \ll 1$. Similarly, the Rabi frequency $\sim(\mathbf{d}\cdot\mathbf{E}/\hbar)$ is taken to be much smaller than ω. Under these conditions the rotating-wave approximation holds.
9. Note that in the absence of relaxation effects $(d/dt)S^z \propto S^y$, and hence correlations of inversion operators are simply related to those of dipole operators. In presence of relaxation we have $(d/dt)S^z = 2gS^y - (1/T_1)(S^z - \eta)$ + random operator force. The situation is thus much more complex and requires more effort although in principle one can calculate the correlation function by using this equation.
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