

Multiple sidebands in cooperative resonance fluorescence: Exact semiclassical results

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We report on the semiclassical solution of the Bloch equations for cooperative resonance fluorescence with collective damping. These equations are shown to be equivalent to the relevant master equation, in the limit of large cooperation number and time scales faster than the one-atom lifetime (reciprocal Einstein A coefficient). The resulting spectrum is derived for a coherent driving field of arbitrary intensity. Additional spectral peaks, as well as the central triplet, are predicted above threshold. A physical explanation of the extra peaks is proposed.

I. INTRODUCTION

Since the first theoretical work of Newstein¹ and of Mollow¹ on resonance fluorescence for a strongly driven two-level atom, and the first observation of the resulting triplet spectrum,² a great deal of attention has been given to the subject of resonance fluorescence.³ In particular it is now well-established that a threshold field strength exists. Above this threshold, an additional pair of sidebands occurs in addition to the central component, in the fluorescent radiation. The ratio of the height of the central line to the sideband is 3:1, while the linewidth ratio is 1:1.5, in the strong-field limit.

In addition, cooperative atomic emission has become a very interesting subject⁴ since it was first proposed by Dicke.⁵ The question of the behavior of a group of two-level atoms driven by a resonant coherent laser field, with a collective decay mechanism, has become of interest as it combines both resonance fluorescence and cooperative emission. It has become common to describe this situation using the approximation of N atoms coupled identically to a single radiation mode. While this description does not include physical features such as spatial fluctuations and dipole-dipole interactions, it is nevertheless of interest, as the simplest model of a group of atoms with a cooperative decay.

This model of a coherently driven group of atoms with a cooperative decay was discussed by Senitzky, who obtained results valid in the case of either a very low or a very-high-intensity driving field.⁶ More recently a calculation of the spectrum in the high-intensity limit was obtained.⁷ This calculation gave a spectral triplet similar to the Mollow result, with additional sidebands in the fluorescent

radiation at spacings equal to the Rabi frequency. However, the extra sidebands were predicted to vanish asymptotically in the strong-field limit.

The question of additional sidebands can be directly related to the dressed-atom picture of atomic fluorescence.⁸ The number of different possible transitions between dressed-atom levels depends on the cooperation number J . Here J is defined as in super-radiance,^{9,5} and is equal to $(N/2)$ for N atoms initially in the ground state. For noninteracting atoms, there are $(2J+1)$ equally spaced energy levels of cooperation number J . Transitions between states of different J do not occur within the cooperative decay Hamiltonian. Including the interaction with a field mode splits the energy levels, so that up to $(4J+1)$ different possible transitions occur. Thus one would expect a triplet with $J=\frac{1}{2}$ and a five-peak spectrum with $J=1$.

Whether the extra sidebands would be observable as distinct peaks depends on the transition matrix elements, and it has been demonstrated¹⁰ that the matrix elements have selection rules that give vanishing extra sideband intensities in the intense-driving-field limit. This prediction agrees with Senitzky's work and also with other results of the dressed-atom picture.¹¹ Thus it is clear that unlike the case of the first sidebands, the high-intensity limit is not the optimal limit for observation of extra sidebands.

The earliest numerical treatment of the model with ($J=1$) was given by Agarwal *et al.*¹⁰ This work showed the existence of cooperative effects, but no extra spectral peaks. More recent numerical work¹² has shown that extra sidebands only occur when there is a sufficiently large Rabi frequency to resolve the extra sidebands from the central triplet. This resolution problem is worse

at low J values. At larger values of the cooperation number there is a relatively larger spectral intensity in the extra sidebands, for Rabi frequencies well above threshold.

For low J values it also becomes theoretically possible to solve the master equation exactly for the spectrum. This was achieved by Agarwal *et al.*,¹³ who obtained results for the case of $J = 1$. In this calculation, it was demonstrated that the exact result (for $J = 1$ only) does give extra sidebands, with five spectral peaks at a large enough, but not infinite, Rabi frequency.

In the case of superfluorescence, where a similar master equation is used without the coherent driving field, progress has been made by using an atomic coherent-state representation.¹⁴ This allows the superfluorescent master equation to be transformed into an exact Fokker-Planck equation.¹⁵ In a recent paper,¹⁶ cooperative resonance fluorescence was analyzed using a Fokker-Planck equation method, by including a coherent driving field in the Fokker-Planck equation treatment that was developed originally for superfluorescence. It was demonstrated that a critical-point transition occurs in the thermodynamic limit of a large number of atoms (or a large cooperation number). This is a nonequilibrium transition which occurs at a threshold field strength. It was also shown that above the nonequilibrium critical point, a "dissipative structure" forms similar to Lotka-Volterra cycles and the cyclic structures that form in other nonequilibrium transitions.¹⁷

In the present paper, the atomic coherent-state representation is used to calculate the fluorescent spectrum in the limit of a large number of atoms. For simplicity, the calculation is made with a semiclassical factorization of the quantum operators. This is an approximation which gives a zero linewidth. It is shown to be valid for a large number of atoms (large cooperation number) and on time scales faster than the one-atom lifetime (reciprocal Einstein A coefficient), which is a similar physical regime to that in which superfluorescence is observed. Thus the semiclassical result is a transient spectrum.

The spectral calculation is carried out with the same factorization used previously,⁷ but is exact relative to this factorization and holds for an arbitrary input intensity. The results depend on the initial state chosen (due to the neglect of quantum fluctuations implicit in the factorization). For atoms initially in the ground state, a particularly simple result obtains. An optimal driving field which maximizes the extra sidebands is calculated, and this is predicted to occur for a finite intensity relative to the oscillation threshold, not

in the intense-field limit.

II. MASTER EQUATION AND BLOCH EQUATIONS

The equations of motion used to describe the system can be equally derived from the master equation approach, or from the Heisenberg equations of motion. Here we choose to use the master equation. We suppose there are N two-level atoms equally coupled to the radiation field. This can be either a point system (without dipole-dipole coupling) of dimensions less than a wavelength, or an extended system interacting with a single radiation mode. In the extended system, the equations are obtained by adiabatically eliminating the radiation field for times $t \ll T_1, T_2$ (Ref. 15) giving a region of validity comparable to that of single-pulse superfluorescence. (Just as in superfluorescence, modifications would be expected due to dipole-dipole interactions¹⁸ and geometric or propagation effects.¹⁹⁻²¹)

The relevant interaction picture master equation in the dipole and rotating frame approximations is,^{10,16} for a coherent input at frequency ω_0 ,

$$\begin{aligned} \frac{\partial \hat{\rho}}{\partial t} = & -i\frac{1}{2}(\Omega)(\hat{J}^+ + \hat{J}^-, \hat{\rho}) \\ & + \frac{1}{2}\gamma(2\hat{J}^-\hat{\rho}\hat{J}^+ - \hat{J}^+\hat{J}^-\hat{\rho} - \hat{\rho}\hat{J}^+\hat{J}^-). \end{aligned} \quad (1)$$

Here Ω is the Rabi frequency, γ is the Einstein A coefficient, and \hat{J}^\pm are collective atomic raising and lowering operators. These are defined as

$$\begin{aligned} \hat{J}^\pm &= [\hat{J}^x \pm i\hat{J}^y], \\ \hat{J}^z &= [\sum_i \hat{J}_i^z], \end{aligned} \quad (2)$$

where \hat{J}_i^α are the usual atomic operators with commutation relations

$$(\hat{J}^x, \hat{J}^y) = i\hat{J}^z \quad (3)$$

The atomic coherent-state representation¹⁴ can be utilized by defining

$$\begin{aligned} |J, z\rangle &= (1 + zz^*)^{-J} \sum_{\nu=0}^{2J} z^\nu \left[\binom{2J}{\nu} \right]^{1/2} |J, J - \nu\rangle, \\ \hat{\rho} &= \int d^2z P(z, z^*, t) |J, z\rangle \langle J, z|. \end{aligned} \quad (4)$$

As Eq. (1) is invariant with respect to \hat{J}^2 , attention can be focused on states of fixed J when the initial value of $\hat{\rho}$ is diagonal relative to \hat{J}^2 . Hence using standard operator techniques an equivalent Fokker-Planck equation is derived:

$$\begin{aligned} \frac{\partial}{\partial t} P(z, z^*, t) = & \left\{ -\frac{\partial}{\partial z} \left[\frac{1}{2}i\Omega(z^2 - 1) + \gamma(J+1)z \right] + \text{c.c.} \right. \\ & \left. + \frac{\gamma}{2} \left(\frac{\partial^2}{\partial z^2} z^2 + \frac{\partial^2}{\partial z \partial z^*} \right) + \text{c.c.} \right\} \\ & \times P(z, z^*, t). \end{aligned} \quad (5)$$

The diffusion matrix in this equation is non-positive definite, so that solutions to the present Fokker-Planck equation would only exist as generalized functions. However, the equation is equivalent to a four-dimensional Fokker-Planck equation with positive-definite diffusion,²² and generates the following Itô stochastic differential equation:

$$\frac{\partial}{\partial t} \begin{pmatrix} z_1 \\ z_2 \end{pmatrix} = \begin{pmatrix} i\Omega(z_1^2 - 1)/2 + \gamma(J+1)z_1 \\ -i\Omega(z_2^2 - 1)/2 + \gamma(J+1)z_2 \end{pmatrix} + \sqrt{\gamma} \begin{bmatrix} z_1^2 & 1 \\ 1 & z_2^2 \end{bmatrix}^{1/2} \begin{pmatrix} \xi_1(t) \\ \xi_2(t) \end{pmatrix}. \quad (6)$$

Here ξ_1, ξ_2 are delta-correlated Gaussian random processes, and

$$\langle \xi_i(t) \xi_j(t') \rangle = \delta_{ij} \delta(t - t'), \quad \langle z_i \rangle = \langle z_i^* \rangle. \quad (7)$$

These equations have been analyzed in the limit $J \rightarrow \infty$,¹⁶ and it is known that the quantum fluctuation terms are only significant on time scales of order $t \geq \gamma^{-1}$. Hence the approximation of neglecting the quantum fluctuation terms is reasonable on time scales less than γ^{-1} . The threshold Rabi frequency for oscillations to occur is $(J\gamma)$; thus many Rabi oscillations will occur for large J , even on this time scale.

An alternate procedure would be to make a semiclassical factorization, with resulting Bloch equations:

$$\begin{aligned} (\partial/\partial t) \langle \hat{J}^+ \rangle &= -i\Omega \langle \hat{J}^z \rangle + \gamma \langle \hat{J}^+ \rangle \langle \hat{J}^z \rangle, \\ (\partial/\partial t) \langle \hat{J}^- \rangle &= (\partial/\partial t) \langle \hat{J}^+ \rangle^*, \\ (\partial/\partial t) \langle \hat{J}^z \rangle &= -\frac{1}{2} i\Omega \langle \hat{J}^+ - \hat{J}^- \rangle - \gamma \langle \hat{J}^+ \rangle \langle \hat{J}^- \rangle. \end{aligned} \quad (8)$$

These equations are invariant with respect to $\langle \hat{J}^z \rangle$, which allows them to be solved exactly for an initial value of $\langle \hat{J}^z \rangle = J^2$. For an initial ground state, $J = N/2$. A similar transformation can be used to that in Eq. (4) to define a semiclassical, nonfluctuating atomic coherent-state variable z :

$$\begin{aligned} \langle \hat{J}^+(t) \rangle &= 2Jz(t)/[1 + z(t)z^*(t)], \\ \langle \hat{J}^-(t) \rangle &= \langle \hat{J}^+(t) \rangle^*, \\ \langle \hat{J}^z(t) \rangle &= J[1 - z(t)z^*(t)]/[1 + z(t)z^*(t)]. \end{aligned} \quad (9)$$

The resulting complex differential equation is

$$(\partial/\partial t)z = i\Omega(z^2 - 1)/2 + \gamma Jz. \quad (10)$$

This is identical to Eq. (6) apart from terms of order $(1/J)$ that are relevant on time scales of order γ^{-1} or greater. Thus the semiclassical factorization for large J is equivalent to neglecting the quantum-fluctuation terms in Eq. (6).

The solutions depend on the value of the driving

field relative to the threshold point $\Omega_0 = \gamma J$. Defining the parameters

$$\begin{aligned} y &= \Omega_0/\Omega, \\ x &= \begin{cases} (y^2 - 1)^{1/2}/i & (\Omega < \Omega_0), \\ (1 - y^2)^{1/2} & (\Omega > \Omega_0) \end{cases}, \\ \gamma^\pm &= iy \pm x, \end{aligned} \quad (11)$$

one obtains the solution

$$\begin{aligned} z(t) &= [\alpha(t)r^- - r^+]/[\alpha(t) - 1], \\ \alpha(t) &= \sqrt{C} \exp(i\Omega x t - i|\Omega x|t_0). \end{aligned} \quad (12)$$

Here (C, t_0) define the initial state of the atomic system at $(t=0)$. The above equations are a complete solution to the semiclassical equations for arbitrary C, t_0, J and arbitrary driving field Ω .^{16,23} For $\Omega < \Omega_0$ there is a rapid decay to the global attractor at $z = r^-$, giving a (sech²) pulse^{4,5} in the case $\Omega = 0$ (for initial inversion). This can be demonstrated, on noting that $i\Omega x$ is positive so that $\alpha(t) \rightarrow \infty$ as $t \rightarrow \infty$. Hence in Eq. (12), the long-time limit is dominated by terms proportional to $\alpha(t)$. For $\Omega > \Omega_0$ the solutions behave like a family of Lotka-Volterra cycles, with frequency $W = \Omega x$. The existence of a threshold was also noted by Senitzky,⁶ who obtained solutions valid in the limits $\Omega \rightarrow 0$ or $\Omega \rightarrow \infty$.

III. SPECTRAL CALCULATIONS

The behavior of the atomic system is very different above and below threshold. Below threshold, both the semiclassical and quantum treatments give similar results in the large- J limit, for inversion, polarization, and spectrum, respectively:

$$\begin{aligned} \langle \hat{J}^z / J_0 \rangle &= -(1 - \Omega^2/\Omega_0^2)^{1/2}, \\ \langle \hat{J}^y / J_0 \rangle &= \Omega/\Omega_0, \\ S(\omega) &= \delta(\omega - \omega_0) J^2 (\Omega/\Omega_0)^2. \end{aligned} \quad (13)$$

These results are the leading terms in an asymptotic expansion in $(1/J)$, and the factorization is correct to leading order, below threshold.

In physical terms, the system behaves similarly to a classical harmonic oscillator, with all the atoms in phase. It must be recognized that in an extended atomic system, the fluorescent radiation would not be all in phase. The reason for this is that the adiabatic elimination of the radiation field means that only the high-intensity "end-fire" mode has been included. Perpendicular, low-intensity fluorescence from an extended medium would also occur, but is neglected in the approximations leading to the J^2 -invariant master equation.

Above threshold, the semiclassical equations have a set of cyclic solutions of the same frequency. An analysis using the cyclic averaging approach²⁴ shows that for times long compared to γ^{-1} the quantum fluctuations cause cycle phase and amplitude diffusion.¹⁶ This results in a steady-state distribution extending over the entire complex z plane (which will be included in Sec. IV). For shorter time scales, the spectrum depends on the initial value of the cycle parameters.

Each cyclic solution is nonsinusoidal, and can physically be represented in the rotating frame, as a damped, driven "pendulum" solution. As the damping is nonlinear in origin, the cycles are nonsinusoidal. The Fourier transform of the semiclassical cyclic polarization has components of all harmonics of the usual sideband frequencies, giving rise to additional sidebands. As the semiclassical equations hold asymptotically for $J \rightarrow \infty$,

the result is compatible with the dressed atom picture of increasing numbers of "dressed" transitions in the large- J limit.

For a driving field $\Omega > \Omega_0$, above threshold, the spectrum depends on C , the initial cycle parameter. To calculate the spectrum, the integration over t is transformed to an integration over the cycle path

$$S(\omega + \omega_0) = \sum_n \delta(\omega - nW) \times \left| \int_C \frac{2Jz}{(1+zz^*)} \left[\frac{(z-r^-)\sqrt{C}}{(z-r^+)} \right]^n d\tau_c(z) \right|^2. \quad (14)$$

Here ($W = \Omega x$) is defined as in Sec. III, and $d\tau_c(z)$ is the time differential for the cycle C , where $d\tau_c(z) = xdz / [\pi i(z-r^+)(z-r^-)]$. The integrals can be evaluated using Cauchy's theorem as in the Appendix:

$$S(\omega + \omega_0) = (J^2/y^2) \left[\delta(\omega) |1 - v(C)x(r^+ - Cr^-)|^2 + \sum_{n \neq 0} \delta(\omega - nW) x^2 \left(\frac{[1 \mp v(C)(2x^2 + C - 1)]C}{1 \pm v(C)(2Cx^2 + 1 - C)} \right)^n \right]. \quad (15)$$

The auxiliary function $v(C)$ equals $[(1-C)^2 + 4Cx^2]^{-1/2}$ and the upper sign of (\pm) is for $n > 0$.

The most interesting case is that for an initial ground state, where $C = 1$ and the cycle passes through the poles of the Bloch sphere. In this case the expression simplifies to give the following result:

$$S(\omega + \omega_0) = \left(\frac{J^2}{y^2} \right) \left[\delta(\omega)(1-x)^2 + \sum_{n \neq 0} \delta(\omega - nW) x^2 \left(\frac{1-x}{1+x} \right)^{|n|} \right]. \quad (16)$$

Several features are of interest in this spectrum. Firstly, the central peak shows a decrease in intensity as $\Omega \rightarrow \infty$, while the first pair of sidebands increase to an asymptotic value, as noted by Senitzky.⁷ The next and succeeding pairs of sidebands are zero both at threshold and in the high-intensity limit, reaching a maximum intensity at a finite driving field relative to Ω_0 as shown in Fig. 1. The optimum driving field for the observation of a five peaked spectrum in the large J limit is for a driving field above threshold, with $\Omega = (\frac{4}{3})^{1/2} \Omega_0$.

It should be noted that quantum fluctuation corrections of order $(1/J)$ have the effect of causing finite spectral widths in the peaks. It is possible for relatively large peak widths to occur at low- J values. The finite peak width effects are of greater importance in the outer sidebands as these have a much lower intensity relative to the central

triplet. In fact it is clear from the dressed-atom picture that for finite J values, there are at most $(4J+1)$ Lorentzian spectral components of different frequencies. Sidebands of greater detuning than $N\Omega$ do not occur, owing to quantum fluctuation effects, which are not included in a semiclassical calculation.

The decrease in intensity of the central peak is distinctly different from the one-atom spectral result, in which the central peak is invariant for $\Omega \rightarrow \infty$. However, this semiclassical result (which was also obtained in the calculation of Senitzky⁷) is essentially a transient result due to the initial

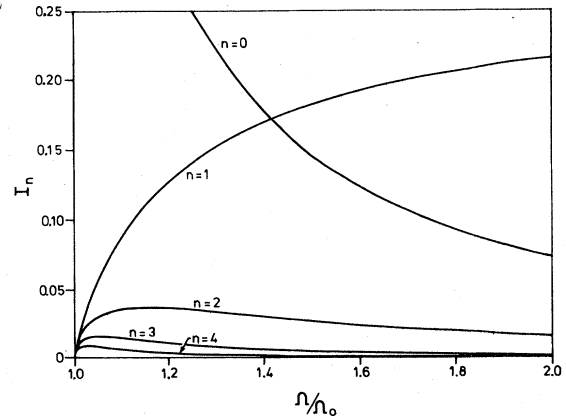


FIG. 1. Sideband intensity vs driving field in the semiclassical factorization for an initial ground state ($C = 1$).

ground state. In the next section, it is shown that on longer time scales, the inclusion of quantum fluctuations results in a distribution over all cycle parameters (C), which causes a steady-state central-peak intensity that is comparable to the one-atom result (apart from a scaling factor).

IV. ENSEMBLE AVERAGES

In the high- J limit, the steady-state distribution function can be readily calculated including quantum fluctuations.¹⁶ The result in the atomic coherent state representation is

$$P(z) \propto \begin{cases} \delta^2(z - r^-) & (\Omega < \Omega_0), \\ |z - r_+|^4 + (2 - 4y^2) |z - r^-|^2 |z - r^+|^2 \\ + |z - r^-|^4 & (\Omega > \Omega_0). \end{cases} \quad (17)$$

Here y, r^\pm are defined as previously.

The spectral calculation in the steady-state limit is given by

$$S(\omega) \equiv \frac{1}{\pi} \operatorname{Re} \int_0^\infty dt e^{-i\omega t} \langle \hat{J}^+(t) \hat{J}^-(0) \rangle, \quad (18)$$

where¹⁵

$$\begin{aligned} \langle \hat{J}^+(t) \hat{J}^-(0) \rangle &= \int \int \langle \hat{J}^+ \rangle_z P(z't | z 0) \\ &\times \left(\frac{2Jz^*}{1+zz^*} - \frac{\partial}{\partial z} \right) P(z) d^2z d^2z'. \end{aligned} \quad (19)$$

Here $\langle \hat{J}^+ \rangle_z = 2Jz'/(1+|z'|^2)$. In general as $P(z)$

is known, it is only necessary to evaluate the propagator. As previously, the propagator is evaluated without fluctuation terms, and for simplicity the large- Ω limit is utilized for $P(z)$:

$$\begin{aligned} \lim_{(\Omega/J) \rightarrow \infty} P(z) &= \frac{1}{\pi} \left(\frac{1}{1+zz^*} \right)^2 \left(\frac{2Jz^*}{1+zz^*} - \frac{\partial}{\partial z} \right) P(z) \\ &= \left(\frac{2(J+1)z^*}{1+zz^*} \right) P(z). \end{aligned} \quad (20)$$

Thus the only effect of the differential operator in z is to change the J -dependent weighting factor.

As the propagator is evaluated to first order in an expansion in $(1/\Omega)$, the deterministic result from Sec. III is utilized. The integration over $P(z)$ is transformed to an integration over C , the cycle parameter, where

$$C \equiv \left| \frac{z - r^+}{z - r^-} \right|^2. \quad (21)$$

The corresponding distribution is

$$\begin{aligned} f(C) &= \left(\frac{xy}{\tan^{-1}(y/x)} \right) [(1+C)^2 - 4y^2C]^{-1}, \\ \lim_{\Omega \rightarrow \infty} f(C) &= [1+C]^{-2}. \end{aligned} \quad (22)$$

Spectral results in the steady state now correspond to an integration of the spectrum for each cycle over the relevant distribution function

$$\begin{aligned} \lim_{J, \Omega/J \rightarrow \infty} S(\omega + \omega_0) &= \frac{J(J+1)}{y^2} \int \left[\delta(\omega) |1 - v(C)x(r^+ - Cr^-)|^2 \right. \\ &\left. + \sum_{n \neq 0} \delta(\omega - nW) x^2 C^n \left(\frac{1 \mp v(C)(2x^2 + C - 1)}{1 \pm v(C)(1 - C + 2Cx^2)} \right)^n \right] \frac{dC}{(1+C)^2}. \end{aligned} \quad (23)$$

Carrying out the integration in the appropriate limit, one obtains

$$\begin{aligned} \lim_{J, \Omega/J \rightarrow \infty} S(\omega + \omega_0) &= \frac{J(J+1)}{y^2} \int \left[\delta(\omega) \left(\frac{1-C}{1+C} \right)^2 y^2 + \sum_{n \neq 0} \delta(\omega - nW) \left(\frac{y^2 C}{(1+C)^2} \right)^{|n|} \right] \frac{dC}{(1+C)^2} \\ &= \frac{N(N+2)}{4} \left[\frac{\delta(\omega)}{3} + \sum_{n \neq 0} \delta(\omega - \Omega n) \left(\frac{\Omega_0}{\Omega} \right)^{2(|n|-1)} \frac{(|n|!)^2}{(2|n|+1)!} \right]. \end{aligned} \quad (24)$$

The asymptotic ratio of peak areas (delta-function weights) is

$$2 : 1 : \frac{1}{5} (\Omega_0/\Omega)^2 : \dots \quad (25)$$

This ratio is identical to the usual ratio for the central triplet peak areas. However, there are additional spectral components corresponding to the appearance of multiple sidebands in the spectrum. These additional components have a weight of $(\Omega_0/\Omega)^{2(|n|-1)}$ for the n th pair of sidebands, and so vanish in the limit of an intense driving field.

This calculation gives an approximate stationary result, relative to the time development of Eq. (1). The result includes the effects of quantum fluctuations on the steady-state atomic coherent-state distribution function, in the large- J -value case. Thus the spectrum is not transient, in the sense that it does not depend on the initial state except for the time-invariant value of $\langle \hat{J}^2 \rangle$. However, the peak widths have been neglected in the high-intensity limit of $\Omega \gg J\gamma$, so that a zero-width approximation is used to calculate the propagator $P(z't' | zt)$. In this sense the calculation is for the total peak areas rather than the spectral line shape.

V. SUMMARY

A calculation of the fluorescent spectrum of a driven, collectively damped atomic system is given in the semiclassical factorization. The semiclassical result is shown to be valid on time scales of order $(t < \gamma^{-1})$ and with a large cooperation number J . Additional sidebands occur in addition to those of the central triplet. The Rabi frequency which maximizes the intensity of the first pair of additional sidebands is $\Omega = (\frac{4}{3})^{1/2} \gamma J$ for a cooperation number J .

A simple physical model for the atomic polarization vector is that of a nonuniformly damped driven pendulum. This causes a nonsinusoidal oscillation above threshold, so that additional fluorescent sidebands occur. Similar types of limit cycle behavior also occur in many other nonequilibrium phase transitions.

In the high-intensity limit, the nonuniform damping is small relative to the driving field, and only a triplet spectrum remains, with intensity proportional to J^2 . In this limit, it is shown that the steady-state triplet peak areas have the same relative size as in the one-atom case. However the direct observation of the J^2 -invariant steady-state spectrum would be difficult in an atomic system due to J^2 -breaking effects that occur on longer time scales. (In the calculation of Carmichael and Walls,²⁵ J^2 breaking is simulated by an independent-atom approximation.²⁶)

The observation of the multiple-peak high-intensity transient spectrum reported here would be of great interest in demonstrating atomic cooperativity in a driven atomic system.

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APPENDIX

The details of the calculation of the spectrum involve an integral over a cycle path for the polarization. This calculation will be studied in detail to illustrate how the spectral calculation proceeds. Any observable (including the polarization) involves a function $O(z, z^*)$, so that

$$\langle O \rangle_c = \oint_c O(z, z^*) d\tau_c(z). \quad (\text{A1})$$

Although this function is nonanalytic, the cycle equation can be used to define z^* in terms of z and thus obtain a meromorphic function which can be integrated by residues. The cycle equation is simply obtained, for cycles of radius ρ , center δ :

$$z^* = \delta^* + \rho^2(z - \delta)^{-1}, \quad (\text{A2})$$

where

$$\delta = iy + x \left(\frac{1+C}{1-C} \right), \quad \rho^2 = (\delta \delta^* - 1).$$

Hence we write the Volterra cycle integral in the following form:

$$\langle O \rangle_c = \oint_c \frac{O(z, \delta^* + \rho^2/(z - \delta)) x dz}{\pi i (z - r^*)(z - r^*)}. \quad (\text{A3})$$

This clearly has poles at $z = r^*$ and it is straightforward to verify that cycles with $c < 1$ enclose r_+ while cycles with $c > 1$ enclose r_- . In fact it is only necessary to make the calculation for $0 < c < 1$, as the results for $c > 1$ are obtained using the symmetry transformation $(c \rightarrow 1/c, z \rightarrow -z^*)$.

The function O will usually have a pole structure, which we now determine. In general, the denominator of O has a factor of form

$$\begin{aligned} (1 + zz^*) &= [(z - \delta)(1 + z\delta^*) + z\rho^2]/(z - \delta) \\ &= \delta^*(z^2 - \epsilon^2)/(z - \delta). \end{aligned} \quad (\text{A4})$$

Here we define $\epsilon = \delta/|\delta|$ and utilize the identity (A2). It is now necessary to determine which if any of the poles $\pm \epsilon$ are included in the residue theorem. This is straightforward, as the cycle is a circle of radius ρ around the centerpoint δ . Thus we merely have to determine the distance $|\pm \epsilon - \delta|$:

$$|\pm \epsilon - \delta| = |\delta| \mp 1. \quad (\text{A5})$$

Noting that $|\delta| = (1 + \rho^2)^{1/2}$ the following relation holds:

$$(|\delta| + 1)^2 > \rho^2 > (|\delta| - 1)^2. \quad (\text{A6})$$

It therefore follows that the cycle encloses the pole at ϵ only. The cycles never enclose the $-\epsilon$ pole.

The calculation for the polarization is obtained on using Cauchy's integral theorem

$$\langle J^+ \rangle_c = \left(\frac{x}{\pi i} \right) \oint_c \frac{2z(z-\delta)J dz}{\delta^*(z^2-\epsilon^2)(z-r^*)(z-r^-)} \quad (\text{A7})$$

$$= 2Jx \sum_{\text{Res}} \frac{2z(z-\delta)}{\delta^*(z^2-\epsilon^2)(z-r^*)(z-r^-)}.$$

The residue at $z=r^+$ is evaluated at first with the following identities:

$$\begin{aligned} (r^+ - \delta) &= -2Cx/(1-C), \\ (r^{*2} - \epsilon^2) &= 4iCxyr^+/\delta^*(1-C). \end{aligned} \quad (\text{A8})$$

Hence the residue reduces to the following simple result:

$$\text{Res}(r^+) = i/(2xy). \quad (\text{A9})$$

The residue at $z=\epsilon$ is evaluated in a similar way with the following identities:

$$\begin{aligned} (\epsilon - \delta) &= \frac{\delta}{|\delta|} (1 - |\delta|), \\ \delta^*(\epsilon - r^+) &= 2iy(1 - |\delta|). \end{aligned} \quad (\text{A10})$$

Hence once again there is a simple result

$$\text{Res}(\epsilon) = \epsilon/(2iy), \quad (\text{A11})$$

$$\langle \hat{J}^+ \rangle_c = Jx \left(\frac{i}{xy} + \frac{(1-C) + x(1+C)/(iy)}{[1+C^2+2C(x^2-y^2)]^{1/2}} \right).$$

The pendulum result is the situation where the inversion goes through both the north and south poles of the Bloch sphere. In this case we obtain ($\epsilon = C = 1$)

$$\langle \hat{J}^+ \rangle = iJ[\Omega/\Omega_0 - (\Omega^2/\Omega_0^2 - 1)^{1/2}]. \quad (\text{A12})$$

This solution displays a lot of the character expected above threshold, with a gradually decreasing polarization as the driving field increases and the system is close to the poles of the Bloch sphere for longer times on the average.

The time-average results can be summarized as follows, for inversion, polarization, and dispersion, respectively:

$$\begin{aligned} \langle \hat{J}^z/J \rangle_c &= 0, \\ \langle \hat{J}^y/J \rangle_c &= (\Omega/\Omega_0)[1 - v(C)x^2(1+C)], \\ \langle \hat{J}^x/J \rangle_c &= v(C)x(1-C). \end{aligned} \quad (\text{A13})$$

All these results were obtained with a cycle parameter $c < 1$. However, the results are invariant under the transformation ($C \rightarrow 1/C, x \rightarrow -x$) and therefore hold equally for $C > 1$.

In the limit of $\Omega \rightarrow \infty$, it is interesting to note that for the central cycle ($C=1$) both the mean polarization and dispersion vanish. However, for the cycles with $C \neq 1$, the dispersive term tends

to a finite value of $(1-C)/(1+C)$, whose sign depends on the cycle parameter. This implies the existence of a central spectral peak for those cycles with $C \neq 1$ in the limit of a large driving field.

The correlation function above threshold is of interest as it determines the fluorescent spectrum. A general expression for the spectrum is

$$\begin{aligned} S(\omega + \omega_0) &= \lim_{t \rightarrow \infty} \left(\frac{1}{2\pi t} \right) \int_0^t \int_0^t dt_1 dt_2 e^{i\omega(t_1-t_2)} \\ &\quad \times \langle \hat{J}^+(t_2) \hat{J}^-(t_1) \rangle. \end{aligned} \quad (\text{A14})$$

This will be calculated semiclassically to give a simple expression for the peak areas and frequencies while neglecting quantum noise effects. The system is cyclic of period $2\pi T$, so the spectrum will be a sum of delta functions in the semiclassical decorrelation, where $W = T^{-1} = \Omega x$:

$$\begin{aligned} S(\omega + \omega_0) &= \sum_n \delta(\omega - nW) \left(\frac{1}{4\pi^2 T^2} \right) \\ &\quad \times \int_0^{2\pi T} \int_0^{2\pi T} e^{iWn(t_1-t_2)} \langle \hat{J}^+(t_2) \rangle \langle \hat{J}^-(t_1) \rangle dt_1 dt_2. \end{aligned} \quad (\text{A15})$$

Here the mean values cannot be steady-state values, but must be defined relative to a specified initial state in order to determine a spectrum. In particular it will be of interest to determine the spectrum relative to the initial state $2J=N, C=1$. This corresponds to all the atoms initially in the ground state which is likely to be the simplest possible experiment. From Eq. (12)

$$\begin{aligned} e^{i\omega(t-t_0)} &= \left(\frac{z-r^+}{\sqrt{C}(z-r^-)} \right)^{\omega T} \\ &= \left(\frac{z^*+r^-}{\sqrt{C}(z^*+r^+)} \right)^{-\omega T}. \end{aligned} \quad (\text{A16})$$

Thus the spectrum corresponding to initial values of c, J, t_0 can be obtained on transforming to integration over z :

$$\begin{aligned} S(\omega + \omega_0) &= \sum_n \delta(\omega - nW) \left| \oint_c \frac{2Jz}{(1+zz^*)} \left(\frac{\sqrt{C}(z-r^-)}{(z-r^+)} \right)^n \right. \\ &\quad \left. \times d\tau_c(z) \right|^2. \end{aligned} \quad (\text{A17})$$

This integral is another Cauchy theorem application. The path of integration is a cycle path in the complex z plane. Therefore on integrating we have a term corresponding to each pole inside the path of integration. Define the relevant integral as follows:

$$J(C, m) = \oint_C \frac{2Jxz(z-\delta)}{\delta^*(z^2-\epsilon^2)} \left(\frac{\sqrt{C}(z-r^-)}{(z-r^+)} \right)^m \times \frac{dz}{(\pi i)(z-r^+)(z-r^-)}. \quad (\text{A18})$$

Now for $C < 1$ the poles inside the path of integration are at $z = \epsilon$, $z = r^+$. Hence the result depends on the value of m as follows:

(i) $m < 0$. In this case only the pole at $z = \epsilon$ is to be included, so one obtains

$$J(C, m) = \left(\frac{J\epsilon x}{iy} \right) \left(\frac{(\epsilon - r^-)\sqrt{C}}{(\epsilon - r^+)} \right)^m. \quad (\text{A19})$$

(ii) This case reduces to Eq. (A11) for the polarization:

$$J(C, 0) = (iJ/y)(1 - \epsilon x). \quad (\text{A20})$$

(iii) $m > 0$. In this integration we take advantage of a theorem of integral calculus that states that if a uniform function has a finite number of singularities then the sum of the residues (including the one at infinity) is zero. In this case the residue at infinity is zero, so the result is (minus the residue at $z = -\epsilon$) because this residue is always outside the cycle path:

$$J(C, m) = \left(\frac{J\epsilon x}{iy} \right) \left(\frac{(\epsilon + r^-)\sqrt{C}}{(\epsilon + r^+)} \right)^m. \quad (\text{A21})$$

In order to simplify the results, it is useful to have the following identities:

$$\begin{aligned} |\epsilon \pm r^-|^2 &= 2[1 \mp v(C)(2x^2 + C - 1)], \\ |\epsilon \pm r^+|^2 &= 2[1 \pm v(C)(1 - C + 2Cx^2)]. \end{aligned} \quad (\text{A22})$$

Hence we obtain

$$|J(C, m)|^2 = \begin{cases} \left(\frac{J^2 x^2 C^m}{y^2} \right) \left(\frac{1 + v(C)(2x^2 + C - 1)}{1 - v(C)(1 - C + 2Cx^2)} \right)^m, & m < 0 \\ \left(\frac{J^2 x^2 C^m}{y^2} \right) \left(\frac{1 - v(C)(2x^2 + C - 1)}{1 + v(C)(1 - C + 2Cx^2)} \right)^m, & m > 0 \end{cases} \quad (\text{A23})$$

In summary the spectrum is as follows, for $C < 1$:

$$\begin{aligned} S_c(\omega + \omega_0) &= \left(\frac{J^2}{y^2} \right) \left[\delta(\omega) |1 - v(C)x(r^+ - Cr^-)|^2 \right. \\ &+ \sum_{m \neq 0} \delta(\omega - mW) x^2 C^m \\ &\times \left. \left(\frac{1 \mp v(C)(2x^2 + C - 1)}{1 \pm v(C)(1 - C + 2Cx^2)} \right)^m \right], \end{aligned} \quad (\text{A24})$$

where the uppermost sign of (\pm) refers to $m > 0$.

To clarify the steady-state calculations, we now calculate the asymptotic form of the spectrum for large $\Omega/J\gamma$. In this case

$$\begin{aligned} v(C) &= \frac{1}{1+C} \left(1 + \frac{2Cy^2}{(1+C)^2} \right), \\ |1 - v(C)x(r^+ - Cr^-)|^2 &= \left| iy \left(\frac{1-C}{1+C} \right) \right|^2, \\ C \left| \frac{1 \mp v(C)(2x^2 + C - 1)}{1 \pm v(C)(1 - C + 2Cx^2)} \right| &= \left(\frac{y^2 C}{(1+C)^2} \right)^{\pm 1}, \\ S_c(\omega + \omega_0) &= \delta(\omega) \left(\frac{1-C}{1+C} \right)^2 y^2 \\ &+ \sum_{n \neq 0} \delta(\omega - n\Omega) \left(\frac{y^2 C}{(1+C)^2} \right)^{|n|}. \end{aligned} \quad (\text{A25})$$

The above spectrum is the asymptotic one (for $\Omega \rightarrow \infty$ or $y \rightarrow 0$) which is utilized in the steady-state calculation, Eq. (24).

The spectrum for $C > 1$ is related to the one for < 1 by the symmetry relation referred to previously; that is, the spectrum for $C' = 1/C$ is obtained as follows:

$$S_c(\omega + \omega_0) = S_c(-\omega + \omega_0). \quad (\text{A26})$$

Both spectra are geometric progressions, with one the mirror image of the other when reflected about the central peak at ω_0 . In general it is also clear that the fluorescence is asymmetric given a value of $C \neq 1$. This is essentially a transient feature due to the particular initial value chosen. In the true (quantum) steady state, there is a distribution over the cycle parameter which produces a spectrum with both wings.

Of greatest interest is the semiclassical result for $C = 1$, which corresponds to an initial ground state. In this case the following completely symmetric result is obtained:

$$\begin{aligned} S_1(\omega + \omega_0) &= \left(\frac{J^2}{y^2} \right) \left[\delta(\omega)(1-x)^2 \right. \\ &+ \sum_{n \neq 0} \delta(\omega - nW) x^2 \left. \left(\frac{1-x}{1+x} \right)^{|n|} \right]. \end{aligned} \quad (\text{A27})$$

The total power radiated is proportional to the sum of the geometric series

$$\begin{aligned} P_{\text{tot}} &\propto (J^2/y^2)(1-x) \\ &\propto (J\Omega/\Omega_0)^2 [1 - [1 - (\Omega_0/\Omega)^2]^{1/2}]. \end{aligned} \quad (\text{A28})$$

In the strong-field limit, the spectrum to order $(1/\Omega^2)$ is of form

$$\begin{aligned} \lim_{\Omega/J\gamma \rightarrow \infty} S_1(\omega + \omega_0) &= J^2 \left[\delta(\omega) \left(\frac{\Omega_0}{2\Omega} \right)^2 \right. \\ &+ \left. \frac{1}{4} \sum_{n \neq 0} \delta(\omega - n\Omega) \left(\frac{\Omega_0}{n\Omega} \right) \right]. \end{aligned} \quad (\text{A29})$$

This agrees with the asymptotic result for

$n = \pm 1, \pm 2$ obtained by Senitzky⁷ noting that our $C=1$ corresponds to his $x=1$.

The interesting feature of this limit is the vanishing central peak and vanishing sidebands for $|n| > 1$. In reality the vanishing central peak is due to neglect of quantum fluctuations: Even for an initial ground state, the distribution in C will

broaden on a time scale of γ^{-1} due to spontaneous emission,¹⁶ resulting in a final steady state with a nonvanishing central peak in the strong-field limit. In fact, the above result corresponds to a classical sinusoidal oscillator in the rotating frame, with frequency Ω , in the strong-field limit.

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