

structure is found, but the oscillations do not agree well in phase or in height. For the disagreement in height the above-mentioned uncertainty in the fluorescence yield is a likely cause.

In the semiempirical formulation the energy splitting between the $2p\sigma$ - $1s\sigma$ orbitals is kept constant and is taken as the difference of the K -shell binding energies. These are obtained here in the same way as outlined in Ref. 10. The impact-parameter dependence is introduced by the dependence of the coupling element on the internuclear separation. No better agreement, however, can be obtained by varying the Ar and S binding energies within reasonable limits. For the case of O on Ne a similar discrepancy in $P_{vs}(b)$ is found by Briggs³ between the semiempirical formula and his result of an *ab initio* calculation. In particular a smaller wavelength of the oscillations was found with the *ab initio* calculation. The discrepancy between the present experiment and the semiempirical formula demonstrates the need for *ab initio* calculations on the basis of quasimolecular wave functions for the S-Ar system.

The pronounced interference pattern observed in this experiment shows that all competing processes which occur during such violent heavy-ion collisions, such as outer-shell ionization and capture, and radial and rotational coupling to higher-lying orbitals (especially $2p\pi - 2p\sigma$), do not destroy the well-defined phase relation in the amplitude between the $2p\sigma$ - $1s\sigma$ radial coupling contributions on incoming and outgoing parts of the collision. Also it should be noticed, that the impact velocity (1 MeV/N) is comparable with the $1s$ -electron velocity, in which case the electrons

are not adiabatic; so it may be surprising that the phase relation between the two couplings is conserved.

In spite of the quantitative disagreement of our data with the semiempirical formula we conclude that the experimental result of a strong oscillatory structure shows an interference of the $2p\sigma$ - $1s\sigma$ radial coupling amplitudes. This strong interference effect should provide a test for *ab initio* calculations using quasimolecular wave functions and dynamical couplings.

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Additional Sidebands in Cooperative Resonance Fluorescence

H. J. Carmichael

Center for Statistical Mechanics and Thermodynamics, University of Texas at Austin, Austin, Texas 78712

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Fluorescent spectra for two, three, four, and five atoms in a volume of dimension less than the resonant wavelength have been obtained numerically. For high laser intensities, resonances displaced from line center by twice the Rabi frequency are found in addition to the usual Stark sidebands.

Single-atom resonance fluorescence has been extensively studied in recent years.^{1,2} Considerable interest is now directed towards the fluorescence from cooperative atomic systems.³⁻⁸ In

the present communication we consider two or more identical two-level atoms set in a volume of dimension less than the resonant wavelength. Intense laser radiation coherently drives this

collective atomic system. A dramatic feature of single-atom resonance fluorescence is the AC or dynamic Stark effect,¹ where for sufficiently intense fields the fluorescent spectrum evolves from a single peak to a form exhibiting a central peak plus symmetrically placed sidebands. Where two or more atoms contribute to a cooperative fluorescence there has been discussion regarding the possible existence of additional sidebands.^{4,6,7} In single-atom fluorescence the dynamic Stark effect arises from the splitting of degenerate levels in the composite atom-field system. For the strong-field limit, splitting of a twofold degeneracy yields three distinct transitions for fluorescent photons, and leads to a three-peaked spectrum. For an N -atom system, an $(N+1)$ -fold degeneracy gives correspondingly $(2N+1)$ distinct fluorescent transitions.⁹ On this basis, can we expect a spectrum with additional sidebands? Agarwal *et al.*⁴ have obtained spectra numerically for two and three atoms, but have found no additional peaks. They attribute their result to selection rules which admit only those transitions corresponding to a central peak and a single pair of sidebands. For two atoms this assessment is supported by Amin and Cordes.⁷ A treatment of the two-atom case with single-atom pumping has also failed to detect additional sidebands.⁸ On the other hand, Senitzky⁶ has suggested the existence of these features, basing his calculations on a classical factorization which he expects to be adequate for large N . Such factorizations have been shown, however, to be invalid in other respects.^{10,11} Nevertheless, Senitzky's results are not excluded by the selection rules of Agarwal *et al.* since the additional sidebands obtained by his procedure vanish for intense fields as the inverse of the laser intensity. It is only in this limit that the selection rules become exact. The present Letter reports numerical studies for two, three, four, and five atoms demonstrating that a second pair of sidebands

does appear as a small, yet distinct resonance in the wings of the single-atom spectrum. To observe these features it is necessary to go beyond those intensities which clearly separate the first pair of sidebands since the additional resonances initially remain buried in the wings.

For the description of fluorescence from an N -atom system we adopt the small-volume superradiant master equation¹² with an added coherent driving term. In a frame rotating at the frequency ω_0 of resonant laser light, the reduced density operator ρ satisfies the equation

$$d\rho/dt = \mathcal{L}\rho \quad (1)$$

with

$$\begin{aligned} \mathcal{L}\rho = & -i\Omega[S_+ + S_-, \rho] \\ & + \frac{1}{2}\gamma(2S_- \rho S_+ - S_+ S_- \rho - \rho S_+ S_-). \end{aligned} \quad (2)$$

Here S_+ and S_- are collective atomic operators, 2Ω is the Rabi frequency, and γ is the Einstein A coefficient. This equation gives evolution towards a steady state with density operator ρ^{SS} .^{4,10,11} In this stationary domain the fluorescent spectrum may be written (aside from constant factors)

$$I_{SS}(\omega) = \text{Re} \int_0^\infty d\tau e^{-i\omega\tau} \langle S_+(\tau) S_- \rangle_{SS}. \quad (3)$$

The atomic autocorrelation function is available from the quantum regression theorem,¹³

$$\langle S_+(\tau) S_- \rangle_{SS} = \exp(i\omega_0\tau) \text{tr}[S_+ e^{i\mathcal{L}\tau} (S_- \rho^{SS})]. \quad (4)$$

For numerical implementation of this scheme a reduced basis $|N/2, M\rangle$, $M = -N/2, \dots, +N/2$ has been adopted spanning only those states with maximum Dicke cooperation number $N/2$. In the context of Eqs. (1) and (2) this is adequate since the Dicke cooperation number J is conserved and dynamics are constrained to a single manifold of states determined by initial conditions. An initial ground state has $J = N/2$. Evaluating the trace in Eq. (4) we get

$$\langle S_+(\tau) S_- \rangle_{SS} = \exp(i\omega_0\tau) \sum_{M=-N/2+1}^{N/2} \left[\left(\frac{N}{2} + M \right) \left(\frac{N}{2} + M + 1 \right) \right]^{1/2} S_{M, M-1}(\tau), \quad (5)$$

where

$$dS/d\tau = \mathcal{L}S, \quad (6)$$

$$S_{M, M'}(0) = \left[\left(\frac{N}{2} - M \right) \left(\frac{N}{2} + M + 1 \right) \right]^{1/2} \rho_{M+1, M'}^{SS}. \quad (7)$$

The equations for matrix elements of ρ demonstrate a fortunate division into two independently coupled schemes. Taking advantage of this feature, matrix elements $\rho_{M, M'}^{SS}$ are found solving $(N+1)(N/2+1)$

linear homogeneous equations. For $S_{M,M}(\tau)$ Eq. (6) may be reduced to a set of $(N+1)^2$ real linear equations. We write

$$d\vec{S}/d\tau = Q\vec{S}, \tag{8}$$

where \vec{S} is an $(N+1)^2$ -dimensional vector and Q is an $(N+1)^2 \times (N+1)^2$ matrix. Numerically determining eigenvalues and eigenvectors of Q and its transpose, Eq. (8) may be diagonalized and the incoherent part of the spectrum obtained in the general form

$$I_{SS}^{inc}(\omega) = \sum_{M=-N/2+1}^{N/2} \left[\left(\frac{N}{2} + M \right) \left(\frac{N}{2} - M + 1 \right) \right]^{1/2} \sum_{\substack{k=1 \\ E_k \neq 0}}^{(N+1)^2} \text{Re} \left(\frac{C_{M,k}}{E_k - i(\omega - \omega_0)} \right). \tag{9}$$

Here E_k are eigenvalues of Q , and $C_{M,k}$ are complex constants determined from eigenvectors of Q and Q^t , and initial matrix elements contained in $\vec{S}(0)$. The zero eigenvalue associated with the coherent part of the spectrum is omitted from the sum in Eq. (9).

In the strong-field limit the single-atom spectrum has peaks at $\omega - \omega_0 = 0, \pm 2\Omega$. With $N \geq 2$, fluorescent spectra have been obtained with use of the above numerical approach. Interest lies in the possibility of additional resonances at $\omega - \omega_0 = \pm 4\Omega$. Senitzky's results,⁵ and those of an extended semiclassical calculation,¹⁴ indicate additional resonances whose amplitudes for high intensities behave as $(1/2\Omega)^2$. At $\omega - \omega_0 = \pm 4\Omega$ the background arising from single-atom peaks

will also exhibit a $(1/2\Omega)^2$ behavior, and a significant feature in the wings of the single-atom spectrum is suggested. The criterion for extracting a distinct peak however rests with the derivatives of each contribution. At $\omega - \omega_0 = \pm 4\Omega$ the background slope changes as $\mp (1/2\Omega)^3$, while a new resonance will grow with a slope proportional to its amplitude. It follows that for increasing Ω , the tails of the single-atom spectrum will flatten so that the cooperative resonance, if present, will grow from the background at a high enough intensity. This will be initiated by the appearance of shoulders at $\omega - \omega_0 = \pm 4\Omega$ for the value of Ω at which $dI_{SS}^{inc}(\omega)/d\omega$ vanishes at these frequencies. This requirement is quite

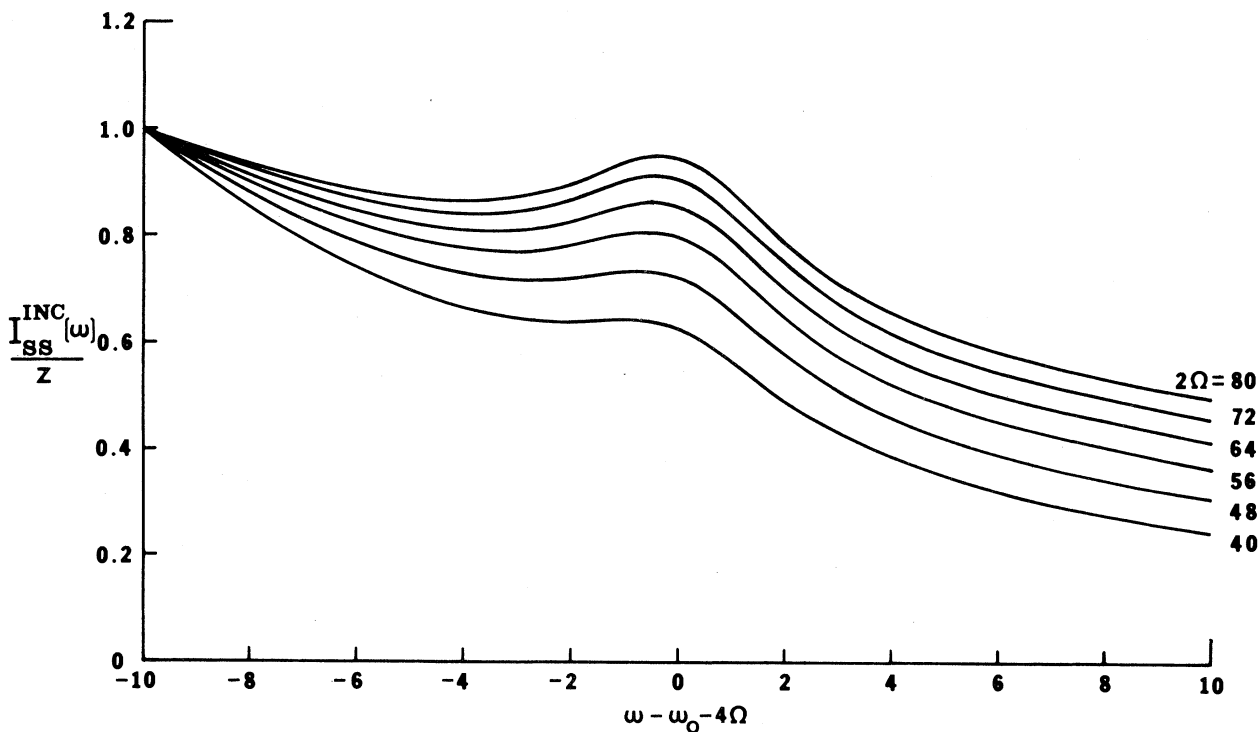


FIG. 1. Fluorescent spectra for two atoms plotted in the frequency range $\omega - (\omega_0 + 4\Omega) = -10$ to $+10$. Each curve corresponds to $\gamma = 1.0$ and is normalized by the factor shown in Table I.

distinct from that assuring separation of the first Stark sidebands. Substantially larger intensities were necessary to satisfy the new condition.

Numerical results for two, three, four, and five atoms clearly demonstrate the existence of cooperative resonances at $\omega - \omega_0 = \pm 4\Omega$. For two atoms, Fig. 1 illustrates the growth of such a feature from the wings of the single-atom spectrum. As Ω varies from $\Omega = 20$ to $\Omega = 40$, a distinct peak emerges from a flattening background. Each curve is normalized by the factor shown in Table I, and peak heights are therefore reduced with increasing laser intensity, but at a rate comparable to the decay in the wings themselves. An enhancement of the cooperative peaks might be expected for larger values of N , and is suggested by Senitzky's calculation.⁵ For $\Omega = 40$, Fig. 2 compares the cases of two, three, four, and five atoms. Substantial increases in peak heights are demonstrated. The values $I_{SS}^{inc}(\omega_0)$ which characterize the central peaks are in good agreement with the form

$$I_{SS}^{inc}(\omega_0) = \frac{1}{6} N(N+2) \quad (10)$$

corresponding to a direct scaling of the single-atom spectrum by the fluorescent intensity.⁶ The cooperative resonances are small features in a spectrum which is otherwise of the single-atom form. A complete discussion of spectral characteristics will be published elsewhere.

The question of further resonances at $\omega - \omega_0 = \pm 6\Omega$, etc., for three, four, and five atoms has not been carefully investigated. However, from the indications of semiclassical calculations¹⁴ we would not expect more peaks (at least for moderate values of N). For fixed N , and in the strong-field limit, the amplitudes of sidebands at $\omega - \omega_0 = \pm 2(n+1)\Omega$ behave as $(1/2\Omega)^{2n}$,¹⁴ while the background contribution from single-atom features is of the order $(1/2n\Omega)^2$. For cases

TABLE I. Normalization factors for the curves of Fig. 1.

Rabi Frequency (2Ω)	Normalization Z (10^{-5})	$I_{SS}^{inc}(\omega_0)$
40	11.60	1.33225
48	6.9777	1.33258
56	4.645	1.33278
64	3.309	1.33291
72	2.475	1.33300
80	1.920	1.33306

other than $n = 1$ the wings of the single-atom spectrum would dominate any resonant contribution.

Experimental detection of cooperative resonances would be a new and interesting demonstration of a collective atomic effect. Of course, the

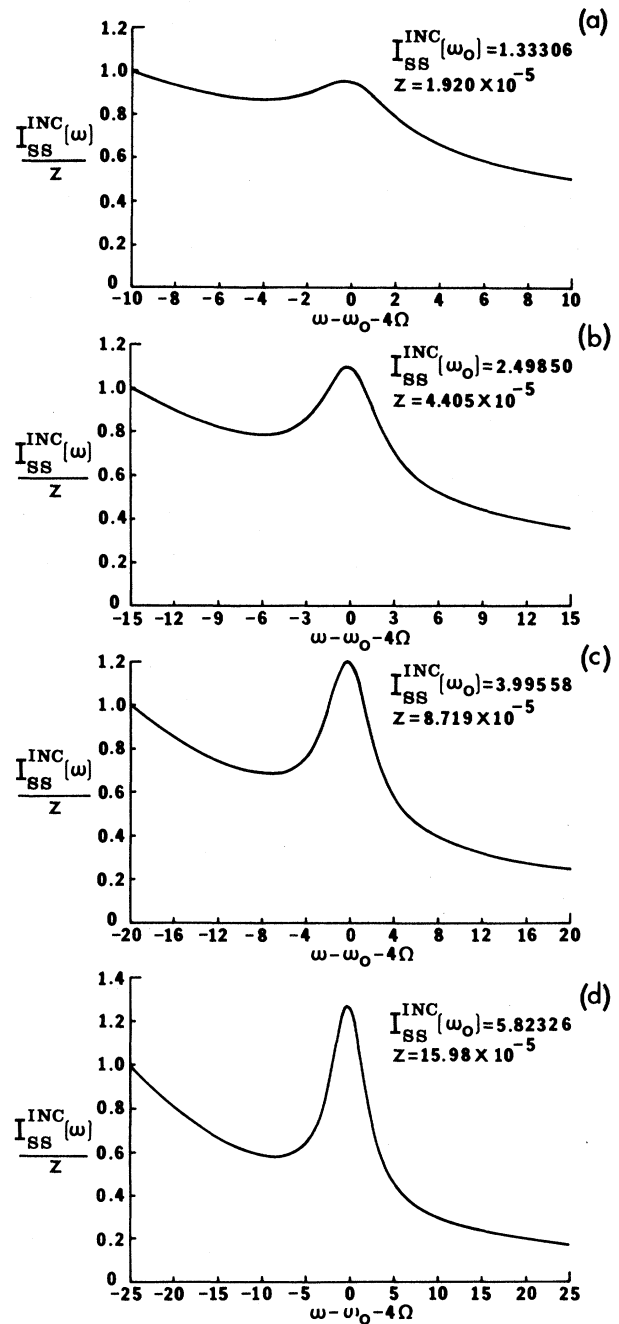


FIG. 2. Fluorescent spectra for (a) two, (b) three, (c) four, and (d) five atoms, each plotted for $\Omega = 40$. $\gamma = 1.0$ in the vicinity of the frequency $\omega_0 + 4\Omega$.

feasibility of such a measurement in the face of very low scattered intensities needs to be carefully assessed. It must also be recognized that Eqs. (1) and (2) have omitted processes which might destroy cooperativity. For example, dipole-dipole interactions¹⁵ will open the path for evolution between states of different Dicke cooperation number J . In the event that spectra are substantially modified in the steady state, cooperative resonances may well remain as transient features.

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Effects of Pumping Process on the Tipping Angle and Quantum Fluctuations in Superfluorescence

C. T. Lee

Department of Physics, Alabama A & M University, Normal, Alabama 35762

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We propose a simple model to incorporate pumping process in the initial evolution of superfluorescence. The calculated initial tipping angle is closest, among various predictions, to the very recently measured value by Vreken and Schuurmans. We also calculate the quantum fluctuations to account for the shot-to-shot variations in experimental results. Additional experiments with varying pumping duration are suggested to further test the validity of our model.

Superfluorescence (SF) is the cooperative spontaneous emission from a system of two-level atoms with complete initial inversion. The possibility of SF was first discussed by Dicke in 1954.¹ It has been under intensive theoretical studies.² However, several recent experimental observations³ have raised some serious questions about the old theory and call for a new one. Since the linear dimensions of the active medium are much larger than the wavelength of the emitted radiation, propagation effects can be important. Such

effects can be easily included in semiclassical calculations based on the coupled Maxwell-Bloch equations.⁴ However, SF can only be initiated by zero-point fluctuations which must be described quantum mechanically. Although a few authors have tried to consider both quantum and propagation effects simultaneously,⁵ the most practical approach remains to be a semiclassical one with the role of the fluctuations simulated by an effective initial tipping angle θ_0 of the so-called Bloch vector. But this mysterious angle has become