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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

The transition from incoherence to coherence emission in superfluorescence (SF) was elucidated. The exact solution to the superradiance master equation was reported. Analytic solution to a non-Markovian master equation for oscillatory SF was obtained. Frequency shift and decay time of radiation in a cavity were calculated. The effects of pumping process on the tipping angle and quantum fluctuations in SF were determined.

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COOPERATIVE PHENOMENA IN THE INTERACTION OF MULTIMODE RADIATION WITH EXTENDED RESONANT MEDIUM

AFOSR-76-2899

I. RESEARCH OBJECTIVES

In 1954 Dicke[1] pointed out the possibility of cooperative spontaneous emission of radiation from an excited system of N two-level atoms at a rate proportional to N^2 rather than N which is the case for non-cooperative emission. This phenomenon is called superradiance or superfluorescence (SF). It can be utilized to generate coherent short pulses of high intensity from far infrared to, perhaps, x-ray region. It has been under very intensive theoretical studies since 1967[2] and also very intensive experimental studies since 1973.[3] These recent experimental observations have raised some serious questions about old theory and call for new approaches.

The general purpose of this research project has been to try to answer some of these questions and to gain better understanding, through analytical studies, the following aspects of SF:

A. Initial Evolution:

The initial stage is the most intriguing and most interesting part of SF for the following reasons:

1. It starts as incoherent emission and spontaneously evoles into coherent emission, a good example of self-organization which is the subject of a new discipline "Synergetics" advocated by Haken.[4]

Some authors have gone so far as to declare that the way this transition is made is the only interesting feature of SF.

2. Superfluorescence is initiated by zero-point fluctuations which can only be understood quantum mechanically. But in the semiclassical approach, the role of this fluctuation is simulated by an effective initial tipping angle θ_o of the so-called Bloch vector. This angle has become a hot issue of current debate because widely different values have been predicted.

3. In experimental observations, the shot-to-shot fluctuations are very large. It is believed that this tremendous fluctuations arise from the random initiation rather than from long-term stochastic development.

B. Overall Behavior:

All the previous calculations resulted in single-pulse SF. But the very first experiment by Skribanowitz et al.[3] in 1973 gave multipulse oscillating emission. This so-called ringing is due to the effects of radiation reaction ignored previously. Inclusion of these effects change the governing master equation from Markovian to non-Markovian. Satisfactory solution of such non-Markovian master equation is a formidable task.

ACCOMPLISHMENTS OF THE RESEARCH

A. Incoherence-to-Coherence Transition

The quantum number that plays the central role in the description of superradiance is the so-called cooperation number r. When the emitting atoms either are confined to a region smaller than the wavelength of the emitted radiation or interact with only one radiation mode, r is conserved. However, in the interaction of multimode radiation with extended medium, r may or may not be conserved in the process of emitting a photon. So we have both r-conserving (rC) and r-nonconserving (rNC) processes to consider. Radiation emitted through rC process is coherent while that through rNC process is incoherent.

Assuming the excited atomic system to be in a Dicke state $|r,m\rangle$, we can calculate the emission rates through rC and rNC processes, respectively, as follows:

$$T^{rC}(r,m) = (r+m)(r-m+1)\gamma\mu = (N-2\ell-n)(n+1)\gamma\mu, \tag{1}$$

$$\begin{aligned} T^{rNC}(r,m) &= \frac{(r+m)(r+m-1)}{2r}\gamma(1-\mu) \\ &= \frac{(N-2\ell-n)(N-2\ell-n-1)}{N-2}\gamma(1-\mu), \end{aligned} \tag{2}$$

where m is 1/2 of the difference of the number of atoms in the excited state and the ground state, γ is the initial spontaneous emission rate of a single excited atom, μ is the geometric parameter, ℓ and n are the numbers of photons emitted through rNC and rC processes, respectively; namely,

$$\ell \equiv N/2 - r, \qquad n \equiv r - m. \tag{3}$$

For multimode radiation interacting with extended system, we have typically $\mu << 1$. Under such condition, we can see from Eqs. (1) and (2) that at the beginning, when both ℓ and n are very small, photons are emitted predominantly through rNC processes and undirectionally. Later on, as n increases, rNC processes, which emit photons directionally, become more and more important. Therefor, there is a transition from incoherence to coherence in the spontaneous emission which occurs when both ℓ and n are still much smaller than N. This transition is the first subject of our investigation.

1. Numerical Study

Let $P(\ell,n)$ be the probabilty that when $\ell+n$ photons have been emitted, ℓ of them are through rNC processes and n of them through rC processes. We can write down a partial difference equation for $P(\ell,n)$ as follows:

$$P(\ell,n) = \frac{n\sigma}{1+n\sigma} P(\ell,n-1) + \frac{1}{1+(n+1)\sigma} P(\ell-1,n) \tag{4}$$

where $\sigma \equiv \mu/(1-\mu)$. We have obtain the solution to Eq. (4):

$$P(\ell,n) = \left[\sum_{\beta} \prod_{i=1}^{n+1} (1+i\sigma)^{-\beta_i}\right] \left[\prod_{i=1}^{n} (1+i\sigma)^{-1}\right] n!\,\sigma^n \tag{5}$$

where $\beta_i = 0, 1, 2, \ldots$, and the summation is over all possible choices of β_i such that $\sum_{i=1}^{n+1} \beta_i = \ell$.

Equation (5) has been evaluated numerically by using the so-called steepest-descent method. The result has been reported in Publication No. 1.

2. Analytical Study

Let $P_{\ell,n}(t)$ be the probability that, at time t, ℓ photons have been emitted through rNC processes and n photons through rC processes. The differential equation for $P_{\ell,n}(t)$ is

$$\frac{d}{dt} P_{\ell,n}(t) = N\gamma(1-\mu)P_{\ell-1,n}(t) + Nn\gamma\mu P_{\ell,n-1}(t)$$
$$- \{N\gamma(1-\mu) + N(n+1)\gamma\mu\}P_{\ell,n}(t) \tag{6}$$

We have obtained the solution to Eq. (6) as follows:

$$P_{\ell,n}(t) = e^{-N\gamma t}\{1-e^{-N\gamma\mu t}\}^n\{N\gamma(1-\mu)t\}^\ell/\ell! \tag{7}$$

From this solution we have calculated the expectation values of the photon numbers

$$\langle\ell\rangle = N\gamma(1-\mu)t, \qquad \langle n\rangle = e^{N\gamma\mu t} - 1 \tag{8}$$

and the emission intensities in terms of photon number

$$I^{rNC} = N\gamma(1-\mu), \qquad I^{rC} = N\gamma\mu e^{N\gamma\mu t} \tag{9}$$

Let the transition point be determined by the condition $I^{rNC} = I^{rC}$; then we have

$$t_c = \frac{1}{N\gamma\mu} \ell n\{\frac{1-\mu}{\mu}\} \tag{10}$$

The result of this analytical study has been reported in Publication No. 2.

B. Exact Solution of Superradiance Master Equation

The overall behavior is predominantly determined by rC processes. If the active region is small enough that we can assume that a photon escape out of the system as soon as it is emitted. Under these conditions, the evolution is described by the so-called superradiance master equation (SME):

$$\frac{d}{dt} P(n,t) = (N-n+1)\, n\, P(n-1,t) - (N-n)(n+1)\, P(n,t), \qquad n = 0, 1, 2, \ldots, N \; (=2r) \tag{11}$$

where P(n,t) is the probability that the atomic system is in the Dicke state $|r,m>$ such that $r-m = n$. We have obtained the exact solutions to Eq. (11) under various initial conditions.

1. Complete Initial Excitation

In this case we have the initial condition $P(n,0) = \delta_{n,0}$; and the exact solution to the SME has been found to be:

$$\begin{aligned} P(n<N/2,t) &= \sum_{i=0}^{n} A_{n,i} e^{-(N-i)(i+1)t} \\ P(N/2 \leq n<N,t) &= \sum_{i=0}^{N-n-2} A_{n,i} e^{-(N-i)(i+1)t} \\ &\quad + \sum_{i=N-n-1}^{N/2-1} \{B_{n,i} + C_{n,i} t\} e^{-(N-i)(i+1)t} \\ P(N,t) &= 1 + \sum_{i=0}^{N/2-1} \{B_{N,i} + C_{N,i} t\} e^{-(N-i)(i+1)t} \end{aligned} \tag{12}$$

where we have defined

$$A_{n,i} \equiv (-1)^i \frac{(N-2i-1)N!n!(N-n-i-2)!}{(N-n)!(n-i)!i!(N-i-1)!} \tag{13}$$

$$B_{n,i} \equiv \frac{(-1)^{N-n-1}\; N!\; n!}{(N-n)!(n-i)!i!(N-i-1)!(i+n-N+1)!}$$

$$\times\left[2 - (N-2i-1)\sum_{k=i+1}^{N-i-1}\frac{1}{k(n-k+1)}\right] \tag{14}$$

$$C_{n,i} \equiv \frac{(-1)^{N-n-1}(N-2i-1)^2\; N!\; n!}{(N-n)!(n-i)!i!(N-i-1)!(i+n-N+1)!} \tag{15}$$

From this solution we have calculated various expectation values of physical quantities. The most important one is that of the photon number

$$\frac{\langle n\rangle}{N} = 1 - \sum_{i=0}^{N/2-1}\frac{(N-1)!}{(N-i-1)!i!}\; e^{-(N-i)(i+1)t}$$

$$\times\left[2 - (N-21-1)\sum_{k=i+1}^{N-i-1}\frac{1}{k} + (N-2i-1)^2 t\right] \tag{16}$$

For practical purpose, we have also obtained the following approximate expression

$$\frac{\langle n\rangle}{N} \simeq e^{Nt}/N - 2!(e^{Nt}/N)^2 + 3!(e^{Nt}/N)^3 + \ldots\ldots..$$

$$= \int_0^\infty \frac{xe^{-x}}{x+Ne^{-Nt}}\,dx \tag{17}$$

As by-products, we have also discovered many identities involving binomial coefficients and Stirling numbers of the second kind. These identities are new, as far as we know, and might be of pure mathematical interest.

These results have been reported in Publication No. 3.

2. Arbitrary Initial Excitation

The evolution of the probability distribution of the atomic system is considered as a Markov process described by a time-evolution matrix (TEM). The exact analytic expressions for the elements of this TEM have been obtained by solving Eq. (11). It has been proved that the TEM satisfies the normalization condition, the initial condition, and the Chapman-Kolmogorov equation as required. The exact general formulas for the expectation values of various physical quantities, expressed in terms of an arbitrry initial distribution, have been obtained. For example, the emission intensity as a function of time has been found to be

$$I = \sum_{i=0}^{r-1} e^{-(N-i)(i+1)t} \Bigg\{ (-1)^i (N-i)(i+1)(N-2i-1) \times \sum_{\ell=i+1}^{N-i-1} P_\ell(0) \frac{(N-\ell)!(\ell-i-1)!}{\ell!(N-\ell-i-1)!} + \sum_{\ell=0}^{i} \frac{P_\ell(0)(-1)^\ell (N-\ell)!}{\ell!(N-\ell-i-1)!(i-\ell)!} \times \Bigg[(N-i)(i+1)\{2-(N-2i-1)\sum_{k=i+1}^{N-i-1}\frac{1}{k-\ell}\} + (N-2i-1)^2\{(N-i)(i+1)t-1\} \Bigg] \Bigg\} \tag{18}$$

where $P_\ell(0)$ is the initial probability distribution.

The general formulas have also been applied to the specific case of superradiant initial state. The most noteworthy result is the normally ordered intensity fluctuation

$$\sigma_n^2(I) = \frac{1}{r}\{\tau \tanh(\tau/2) - 2\} \tanh(\tau/2) \tag{19}$$

These results have been reported in Publication No. 4.

C. Oscillatory Superfluorescence

For an extended system, an emitted photon cannot escape out of the system immediately. So photon reaction must be considered and the radiation process becomes non-Markovian. The basic equation for such oscillatory superfluorescence and superradiance is:

$$\frac{dW(t)}{dt} = \Lambda_F W(t) - ige^{-t/2T_2^*}\left[(aR^+ + a^\dagger R^-), e^{\Lambda_F t} W(0)\right]$$
$$- g^2 \int_0^t \left[(aR^+ + a^\dagger R^-), e^{\Lambda_F (t-t')}\left[(aR^+ + a^\dagger R^-), W(t')\right]\right]$$
$$\times e^{-(t+t')/2T_2^*} dt' \qquad (20)$$

where (a) $W(t)$ is the density matrix of the system composed of N two-level atoms and a single axial mode of the internal field (i.e., the field inside the active volume); (b) $a^\dagger$ and a are the photon creation and annihilation operators; (c) R^+ and R^- are the collective atomic excitation and deexcitation operators; (d) $\Lambda_F W(t) = K\left\{[aW(t), a^\dagger] + \text{H.c.}\right\}$ whre $K = c/2L$ (L being the length of the active region) is the damping constant, so Λ_F describes the irreversible escape of photons from the active region; (e) g is the atom-field coupling constant and T_2^* is the smallest atomic relaxation time which will be assumed to be infinite in the time scale of our problem.

All order of atom-field correlations have been included in Eq. (20). A complete solution of this equation seems to be impossible. We have obtained solutions that include zeroth order and first order correlations. From these solutions we can calculate the expection values of various physical observables. For example, the radiation intensity as a function of time have been found to be

$$I_o(t) = \sum_{i=0}^{N/2-1} \Bigg\{ (N-i)(i+1)\left[2 - (N-2i-1)\sum_{k=i+1}^{N-i-1} \frac{1}{k}\right] X_o(i,t)$$

$$- (N-2i-1)^2 \Big[X_o(i,t) - (N-i)(i+1)Y_o(i,t)\Big] \Bigg\}$$

$$\times \frac{N!}{(N-i-1)!\,i!} \tag{21}$$

for the case of zeroth order correlation; where

$$X_o(i,t) = e^{-Kt/2}\left[\cos\omega_i t + \frac{k}{2\omega_i}\sin\omega_i t\right] \tag{22}$$

$$Y_o(i,t) = (K/2\omega_i)^2 I\, e^{-Kt/2}\left[(1 + 2\omega_i t/K)\sin\omega_i t - \omega_i t\cos\omega_i t\right] \tag{23}$$

and where

$$\omega_i = \left[KI(N-i)(i+1) - K^2/4\right]^{\frac{1}{2}} \tag{24}$$

To obtain the radiation intensity $I_1(t)$ for the case of first order correlation, we need only to replace the $X_o(i,t)$ and $Y_o(i,t)$ in Eq. (21) by $X_1(i,t)$ and $Y_1(i,t)$ which are the inverse Laplace transforms of $\tilde{X}_1(i,s)$ and $\tilde{Y}_1(i,s)$, respectively:

$$\tilde{X}_1(i,s) = KI/\Big\{s(s+K)(s+2K) + KI(N-i)(i+1)(3s+2K)\Big\} \tag{25}$$

$$\tilde{Y}_1(i,s) = (3s+2K)\{X_1(i,s)\}^2 \tag{26}$$

The details of these results have been reported in Publication No. 5.

D. Superradiance in a Cavity

With possible applications to the problem of optical bistability[4] in mind, we study the cooperative effects in the interaction of matter with radiation in a cavity which is partially open, resulting in the broadening and frequency shift of cavity modes.

1. Frequency Shift and Decay Time of Radiation in a Cavity

We consider a small Fabry-Perot cavity imbedded in a larger one with perfectly reflecting walls, receding to infinity eventually to represent the universe. This is a generalization of the Lang-Scully-Lamb model[5] for single-quasimode laser operation in the sense that both mirrors, instead of just one, of the inside cavity are semitransparent. We have obtained a complete set of orthonormal modes of the universe. Using these mode functions, we have studied the leakage of a monochromatic radiation out of the inside cavity. We have found that this leakage leads to a frequency shift

$$\Delta\Omega = \frac{c}{2\ell}\tan^{-1}\sqrt{(1-R)/R} \tag{27}$$

a decay time

$$\tau = -2\ell/c \ln R \tag{28}$$

and an enhancement factor, defined as the ratio of the amplitude of the mode function inside the cavity to that of outside,

$$\beta = \sqrt{(1+3R)/(1-R)} \tag{29}$$

where 2ℓ is the length of the cavity and R is the reflectance of the mirrors.

The details of the derivation have been reported in Publication No. 6.

2. Transition Probabilities between Mixed Dicke States

The cooperative phenomena in the interaction of radiation with matter are usually discussed in terms of the so-called Dicke state $|r,m,\alpha>$ which is a pure state in the terminology of density matrix. Such a pure state is specified by a degeneracy index α in addition to the angular momentum eigenvalues r and m. Since the physical meaning of α is quite obscure, it is more realistic to consider a mixed Dicke state $|r,m>$ to which each $|r,m,\alpha>$ contributes with equal weight and random phase. Using perturbation approach and the diagrammatic technique developed by us, we calculated the various transition probabilities in the interaction of multimode radiation with resonant atoms in a Fabry-Perot cavity of dimensions much larger than the wavelength of the radiation.

a. r-Conserving Processes:

$$P(|r,m> \rightarrow |r,m-1>) = A\mu\beta^2(r+m)(r-m+1) \quad (30)$$

where A is the Einstein coefficient for spontaneous emission, μ is the geometric parameter, and β is the enhancement factor given in Eq. (29).

b. r-Increasing Processes:

$$P(|r,m> \rightarrow |r+1,m-1>) = A(1-\mu)(r-m+1)(r-m+2)(N/2-r)/4r^2 \quad (31)$$

c. r-Decreasing Processes:

$$P(|r,m> \rightarrow |r-1,m-1>) = A(1-\mu)(r+m)(r+m-1)(N/2-r)/4r^2 \quad (32)$$

The preliminary results of this calculation have been reported at the APS meeting in Washington, D.C., 24-27 April 1980.

E. Initiation of Superfluorescence during Pumping

In all the theoretical sudies on superfluorescence, the complete initial excitation is assumed to be achieved instantaneously. But this is impossible realistically. So we have proposed the following master equation to describe the initiation of superfluorescence which allows finite duration for pumping:

$$\frac{d}{dt} P(N,n,t) = \rho P(N-1,n,t) - \rho P(N,n,t) + \mu\gamma(N-n+1)nP(N,n-1,t) - \mu\gamma(N-n)(n+1)P(N,n,t) \tag{33}$$

where $P(N,n,t)$ is the probability that N atoms have been excited and n photons have been emitted at time t. The first two terms on the right hand side of Eq. (33) describe the pumping as a Poisson process at a constant rate ρ; and the last two terms describe the superfluorescence with varying cooperation number, where μ is the geometric parameter and γ is the single atom emission rate.

Under the condition that $n \ll N$, we have found the solution to Eq. (33) to be

$$P(N,n,t) = \frac{(\rho/\mu\gamma)^N}{N!} e^{-\mu\gamma t} \sum_{i=0}^{n} (-1)^i \binom{n}{i} \left[\frac{1 - e^{-(i+1)\mu\gamma t}}{i+1}\right]^N \tag{34}$$

From this solution, we have calculated the expectation value of the initial tipping angle to be

$$\theta_o = (2/N)^{\frac{1}{2}} \exp(\tau_p/4\tau_R) \tag{35}$$

with quantum fluctuation in the range

$$\theta_o^{\pm} = \theta_o\{1 \pm (1 - e^{-\tau_p/2\tau_R})\}^{\frac{1}{2}} \tag{36}$$

where τ_p is the pumping duration and $\tau_R = 1/(\mu\gamma N)$ is the superradiance time.

Using the experimental conditions of Vrehen and Schuurmans:[6] $N \simeq 2 \times 10^8$, $\tau_p \simeq 2$ ns, $\tau_R \simeq 0.4$ ns, we can calculate $\theta_o \simeq 3.5 \times 10^{-4}$ and $\theta_o^{\pm} \simeq 0.7 \times 10^{-4}$ to 4.9×10^{-4}; which compare favorably with the measured values:[6] $\theta_o \simeq 5 \times 10^{-4}$ and $\theta_o^{\pm} \simeq 10^{-4}$ to 2.5×10^{-3}.

The details of the derivation have been reported in Publication No. 7.

III. CUMULATIVE LIST OF PUBLICATIONS

1. C. T. Lee, "Transition from Incoherence to Coherence in the Spontaneous Emission of Extended Systems", Phys. Rev. A 13, 1657 (1976).
2. C. T. Lee, "Transition Point of Superradiance", Phys. Rev. A 14, 1926 (1976).
3. C. T. Lee, "Exact Solution of Superradiance Master Equation. I. Complete Initial Excitation", Phys. Rev. A 15, 2019 (1977).
4. C. T. Lee, "Exact Solution of Superradiance Master Equation. II. Arbitrary Initial Excitation", Phys. Rev. A 16, 301 (1977).
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6. C. T. Lee, Frequency Shift and Decay Time of Radiation in a Partially Open Cavity", Opt. Communi. 27, 277 (1978).
7. C. T. Lee, "Effects of Pumping Process on the Tipping Angle and Quantum Fluctuations in Superfluorescence", Phys. Rev. Lett. 43, 1110 (1979).

IV. INTERACTIONS

1. Seminar lecture on "Rate Equation for Superradiance" at the Physics Department, University of Alabama in Huntsville, 13 April 1976.
2. Participation in the Nineth International Quantum Electronics Conference, Amsterdam, Netherlands, 14-18 June 1976.
3. Presentation of a paper on "Exact Analysis of the Spontaneous Emission from Dicke's Superradiant State" at the American Physical Society Meeting in Washington, D.C., 25-28 April 1977.
4. Seminar lecture on "Incoherence-to-Coherence Transition in the Spontaneous Emission of Large Systems", at Columbia Radiation Laboratory, Columbia University, 2 June 1977.
5. Presentation of a paper on Analytic Solutions of Non-Markovian Master Equation for Oscillatory Superfluorescence" at the Fourth Rochester Conference on Coherence and Quantum Optics, Rochester University, 8-10 June 1977.
6. Seminar lecture on "Optical Bistability" at the Marshall Space Flight Center, Huntsville, Alabama, 31 January 1978.
7. Presentation of a paper on "Transition Probabilities between Mixed Dicke States" at the American Physical Society Meeting in Washington, D.C., 24-27 April 1978.
8. Participation in the 1979 Conference on Laser Engineering and Applications in Washington, D.C., 30 May-1 June 1979.
9. Presentation of a paper on "Initiation of Superfluorescence during Pumping at the International Conference on Lasers '79, Orlando, Florida, 17-21 December 1979.

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