

6.2 Superradiance

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6.2.1 Definitions and historical layout

SuperRadiance (SR) is the *cooperative* spontaneous emission of radiation by a collection of atoms, molecules or nuclei. The spontaneous transition from an excited state to the ground state is the result of interaction with the vacuum fluctuations of the electromagnetic field. The theory of spontaneous emission from a single atom was first proposed by Dirac [27Dir] and then developed by Wigner and Weisskopf [30Wei]. It yields the usual exponential decay with the natural radiation lifetime γ^{-1} , see (5.1.66) and (6.2.1). In most cases one observes this type of radiation also from a system of several atoms, because the spontaneous decays of the sources are not correlated. In 1954 Dicke [54Dic] recognized that if the phases of the atomic states do not suffer random changes, then the interaction of the atoms with each other through their common radiation electromagnetic field results in a correlation between the atomic dipole moments. In this way a macroscopic polarization is created which is proportional to the number of atoms N , the multi-atomic system radiates collectively, and the radiation is coherent. The decay time of this collective emission is proportional to $1/N$, and the *radiation intensity is proportional to the square of the number of atoms* N^2 . This is the most characteristic property of SR. Furthermore, the shape of the emitted electromagnetic pulse is no longer exponential, but has a peak (or some ringing) after a certain delay. These are the reasons why SR is one of the efficient methods used to generate intensive and short electromagnetic pulses.

SR is effective only if other interactions, such as collisions, thermal noise etc., which are usually also present besides the interaction with the radiation electromagnetic field, do not disturb the phases of the atomic states during emission. If this condition is not fulfilled, one has ordinary spontaneous emission, or amplification of the latter: *Amplified Spontaneous Emission* (ASE), an incoherent effect which is sometimes also called *superluminescence*. We use here the term *super-radiance* for any cooperative emission process, although the cooperative spontaneous emission of a macroscopic system of atoms being initially in a fully excited but uncorrelated state is termed often by the special name *SuperFluorescence* (SF).

The first experimental observation of SR followed not until nearly two decades after the prediction by Dicke. In 1973 Skribanowitz et al. [73Skr] were able to realize experimental conditions where the phase memory time of the radiation centers was longer than the cooperative spontaneous emission time. Their experiment showed the effect in a pencil-shaped extended sample consisting of HF molecules.

Several reviews and books [74Aga, 75All, 81Sch, 82Gro, 82Vre, 85Har, 86Leo, 93And, 96Ben] treat SR in great detail from different points of view, mainly theoretically. Experimental investigations on extended systems are reviewed in [82Vre]. The work [85Har] gives a detailed description of microwave SR experiments in a cavity, not considered here. In the book [96Ben] one of the chapters is devoted exclusively to the discussion of the experiments performed with superradiant systems, and in several other chapters emphasis is laid on the experimental justification of the different aspects of the effect. It contains also a rather comprehensive bibliography of both experimental and theoretical works in the field.

6.2.2 Superradiance theory

6.2.2.1 Superradiance of a system with dimensions smaller than the radiation wavelength

A two-level atom is a simple and effective model of a real atom interacting resonantly with the electromagnetic field. The upper and lower states, $|e\rangle$ and $|g\rangle$, are supposed to be separated by an energy interval $\hbar\omega_0$. If the atom is initially in its upper state, (and the field in the vacuum state), then the probability P_e of finding the atom in the state $|e\rangle$ obeys the equation [27Dir, 30Wei]:

$$\frac{dP_e}{dt} = -\gamma P_e, \quad \text{with} \quad \gamma = \frac{1}{3\pi\epsilon_0} \frac{|\mathbf{d}|^2 \omega_0^3}{\hbar c^3}, \quad (6.2.1)$$

where $\mathbf{d} = \langle e|\hat{\mathbf{D}}|g\rangle$ is the off-diagonal matrix element of the dipole operator $\hat{\mathbf{D}}$, see below. Equation (6.2.1) gives the usual exponential law of spontaneous emission. It is valid for an arbitrary quantum system provided its dimensions are less than the emission wavelength.

In order to treat the problem of several atoms one introduces an effective quantum-mechanical energy operator, $\hat{H}_0 = \hbar\omega_0 \hat{\mathcal{R}}_3^i$, and the raising and lowering operators $\hat{\mathcal{R}}_+^i$ and $\hat{\mathcal{R}}_-^i$ for the i -th atom with:

$$\hat{\mathcal{R}}_3^i := \frac{1}{2}(|e\rangle\langle e| - |g\rangle\langle g|)^i, \quad \hat{\mathcal{R}}_+^i := (|e\rangle\langle g|)^i, \quad \hat{\mathcal{R}}_-^i := (|g\rangle\langle e|)^i, \quad (6.2.2)$$

which are seen to obey the commutation relations of angular momentum operators:

$$[\hat{\mathcal{R}}_3^i, \hat{\mathcal{R}}_\pm^j] = \pm \delta_{ij} \hat{\mathcal{R}}_\pm^i, \quad [\hat{\mathcal{R}}_+^i, \hat{\mathcal{R}}_-^j] = 2\delta_{ij} \hat{\mathcal{R}}_3^i. \quad (6.2.3)$$

The corresponding operators for the total system are:

$$\hat{\mathcal{R}}_\alpha = \sum_i^N \hat{\mathcal{R}}_\alpha^i \quad (\alpha = +, -, 3). \quad (6.2.4)$$

The total dipole moment operator can be expressed as: $\hat{\mathbf{D}} = \sum_i \hat{\mathbf{D}}^i = \mathbf{d}(\hat{\mathcal{R}}_+ + \hat{\mathcal{R}}_-)$. The energy operator of the N identical two-level atoms is $\hat{H}_0 = \hbar\omega_0 \hat{\mathcal{R}}_3$. As it is easily seen, it has $N+1$ different eigenvalues:

$$E_M = M\hbar\omega_0 \quad \text{with} \quad M = N/2, N/2 - 1, \dots, -N/2, \quad (6.2.5)$$

where $M = (N_+ - N_-)/2$ is half of the difference of the number of atoms in the upper and lower levels, N_+ and N_- . In each subspace determined by a degenerate energy value there is exactly one linear combination that is *symmetric* with respect to the permutation of the atoms:

$$\begin{aligned} & |e, e, e, \dots, e\rangle & M = N/2, \\ & \frac{1}{\sqrt{N}}(|g, e, \dots, e\rangle + |e, g, \dots, e\rangle + \dots + |e, e, \dots, g\rangle) & M = N/2 - 1, \\ & \binom{N}{2}^{-\frac{1}{2}}(|g, g, e, \dots, e\rangle + |g, e, g, \dots, e\rangle + \dots + |e, e, \dots, g, g\rangle) & M = N/2 - 2, \\ & \vdots \\ & |g, g, g, \dots, g\rangle & M = -N/2. \end{aligned} \quad (6.2.6)$$

These states all belong to the highest eigenvalue, $\frac{N}{2}(\frac{N}{2} + 1)$, of the operator $\hat{\mathcal{R}}^2 = \left(\frac{1}{2}(\hat{\mathcal{R}}_- \hat{\mathcal{R}}_+ + \hat{\mathcal{R}}_+ \hat{\mathcal{R}}_-) + (\hat{\mathcal{R}}_3)^2\right)$. $\hat{\mathcal{R}}^2$ commutes with both the energy operator of the unperturbed system of atoms \hat{H}_0 and the dipole interaction operator

$$\hat{H}_{\text{int}} = -\hat{\mathcal{E}}\hat{\mathbf{D}}, \quad (6.2.7)$$

where

$$\hat{\mathcal{E}} = i \sum_{\mathbf{k}\sigma} \epsilon_{\mathbf{k}\sigma} \sqrt{\hbar\omega_{\mathbf{k}}/2\epsilon_0 V} (\hat{a}_{\mathbf{k}\sigma} e^{i\mathbf{k}\cdot\mathbf{r}} - \text{h.c.}) = \hat{\mathcal{E}}^+ + \hat{\mathcal{E}}^- \quad (6.2.8)$$

is the electric field operator, written as the sum of its positive and negative frequency parts as in (5.1.14), and which is to be taken at the position of the atoms. Therefore, if the system starts from the symmetric state, where all the atoms are inverted ($M = \frac{1}{2}N$), then it will remain during the evolution in the totally symmetric $N+1$ -dimensional subspace given by (6.2.6). Starting from the uppermost state with $M = N/2$, the system first emits one photon and gets into the state with $M = N/2 - 1$, where all atoms but one are excited, then to the state with $M = N/2 - 2$, where all atoms but two are excited, etc. Because of the symmetric superposition of the individual atomic states, we cannot say which of the atoms has emitted the photon, we can only state that the number of excited atoms has become less by one. The states given by (6.2.6) are typical examples of entangled states. It is important to note that besides those given in (6.2.6) there exist other common eigenstates of $\hat{\mathcal{R}}^2$ and $\hat{\mathcal{R}}_3$, belonging to smaller eigenvalues of $\hat{\mathcal{R}}^2$, but these are not totally symmetric and the transitions between levels belonging to different eigenvalues of $\hat{\mathcal{R}}^2$ are forbidden. This latter effect is called *subradiance* [85Pav, 96Ben, 96DeV].

Calculating the matrix elements of the total dipole moment operator $\hat{\mathbf{D}}$ between the states given in (6.2.6), we can find the transition probabilities for the allowed transitions and we obtain [71Bon, 74Aga, 96Ben] a master equation for the probability P_M of observing the ensemble of atoms in the symmetric state of energy E_M :

$$\frac{dP_M}{dt} = \gamma(\tfrac{1}{2}N + M + 1)(\tfrac{1}{2}N - M)P_{M+1} - \gamma(\tfrac{1}{2}N + M)(\tfrac{1}{2}N - M + 1)P_M. \quad (6.2.9)$$

Superradiance corresponds to the initial condition: $P_{\frac{N}{2}}(t=0) = 1$, and $P_M(t=0) = 0$, $M \neq \frac{1}{2}N$, and the evolution of the atomic system appears as a cascade emission down the “ladder” of the $N+1$ equidistant levels given in (6.2.6). While the system reaches level M , it emits $n = N/2 - M$ photons, therefore the solution of (6.2.9) gives information about the number of the emitted photons. As it is seen from the coefficients in (6.2.9) the emission of the photons is getting exponentially faster and faster while M decreases from $N/2$ till around 0, and then the emission slows down again while M goes to $-N/2$. An approximate analytic solution [71Bon, 74Aga, 96Ben] of (6.2.9) yields the following expression for the radiated intensity given as the number of emitted photons per unit time:

$$I(t) = \gamma \frac{1}{4} N^2 \operatorname{sech}^2[\gamma \frac{1}{2} N(t - t_D)], \quad (6.2.10)$$

where $t_D = (\gamma N)^{-1} \ln N$. From (6.2.10) the main features of the superradiant pulse are readily seen:

1. The initial intensity of the pulse is the same as that of ordinary spontaneous emission of N atoms: $I(0) = \gamma N$.
2. Its duration measured at half maximum is of the order $(\gamma N)^{-1}$, i.e. N times *shorter* than the radiation decay time of a single atom.
3. The maximum of the intensity is proportional to the *square* of the total number of atoms N , and appears after a *delay time* of t_D .

These properties keep their validity for a macroscopically extended system, as well, with the exception that the value of the delay time is modified, see (6.2.19) below. From the solution of (6.2.9) one can also find the photon statistics of the emitted pulse. In the central part of the SR pulse – when $M = 0$ – it is nearly Poissonian, and hence the radiation is coherent, while at the beginning and at the end it shows larger fluctuations in photon number [71Bon, 96Ben].

The picture outlined above for the small system is an idealized one. In reality, the near-field dipole–dipole coupling between the atoms will lead to a site-dependent perturbation of the relevant

atomic levels. This effect destroys the coherence of the identical atomic emitters, and the radiated intensity remains proportional only to N , even in the absence of other dephasing effects [72Fri].

There are two ways to circumvent the problem above. The first one is to place the atomic system into a resonant cavity. Spontaneous emission of atoms is a consequence of the interaction with fluctuations of the huge number of modes of the electromagnetic vacuum surrounding the atoms. A cavity of appropriate dimension modifies the mode structure of the field around the atoms so that only a few or just one resonant mode is present, and therefore the density of the possible final states is considerably reduced. The dynamics of the system is then described by a modified master equation. Cooperative emission of a system of Rydberg atoms in a cavity has been observed by Haroche and coworkers, for a review see [85Har]. The same effect has been demonstrated for a system of nuclear spins in a magnetic field [90Baz, 96Ben]. This enhancing effect of a cavity had been first considered in [54Blo].

The other possibility to retain the cooperativity of the atoms is to use an extended system, which is discussed in the following Sect. 6.2.2.2.

6.2.2.2 Superradiation of an extended multiatomic system

In a cylindrical, pencil-shaped extended system the collective interaction will be effective only with the photon modes, for which the wave vectors are nearly parallel with the axis of the cylinder. The atoms have symmetric positions with respect to this mode, and then the influence of near-field dipole–dipole coupling is negligible. In this case, however, only these modes will give a contribution to the probability of the spontaneous decay, and their density is proportional to the diffraction solid-angle λ^2/A , where λ is the wavelength and A is the cross-section of the sample. Therefore, the emission will go in this narrow cone in both directions, and the decay rate – being reduced by the same factor – will be of the order of $\gamma N \lambda^2/A$. The time scale of superradiance for an extended pencil-shaped sample is the inverse of this collective rate. It is customary to introduce an additional numerical factor and to define *superradiation time* as:

$$T_R = \left(\frac{3}{8\pi} \gamma N \lambda^2/A \right)^{-1} = \left(\frac{3}{8\pi} \gamma N_0 \lambda^2 L \right)^{-1} = 2 \varepsilon_0 \frac{\hbar c}{N_0 d^2 \omega_0 L}, \quad (6.2.11)$$

where N_0 is the number of inverted atoms per unit volume in the sample of length L . Note, however, that some authors use a different factor, and define superradiation time as $T'_R = 3T_R$! We see that for an extended system the duration of the superradiant pulse decreases by a factor proportional to the number of atoms in the volume $\lambda^2 L$, rather than by a factor equal to the total number of atoms N which is the case in a small sample.

In order to describe SR in an extended system in more detail one has to take into account the spatial positions of the atoms, as well as the propagation of the emitted electromagnetic field. One can derive the relevant equations by a quantum-electrodynamical calculation, see e.g. [79Haa, 79Pol, 81Sch, 82Gro, 96Ben]. To have a tractable description, one introduces *smoothly varying* fields by averaging the microscopic operators over a volume much smaller than the relevant wavelength, but still containing several atoms: $\hat{\mathcal{R}}_\alpha(\mathbf{r}) = \frac{1}{\Delta V N_0} \sum_{\mathbf{r}_i \in \Delta V} \hat{\mathcal{R}}_\alpha^i$. One considers a one-dimensional problem: a cylinder of length $L \gg \sqrt{A} \gg \lambda$, by treating propagation only along the axis of the cylinder, the x axis, and neglecting the transversal variation of the fields. Then the standard approximations – used also in laser theory – can be exploited. Slowly varying, scalar electric field operator amplitudes \hat{E}^\pm and atomic operators \hat{R}_\pm and \hat{Z} are introduced, so that factors of the form $e^{\pm i(kx - \omega_0 t)}$ are separated from the variables:

$$\hat{\mathcal{E}}^\pm(x, t) = \frac{\hbar}{2dT_R} \hat{E}^\pm(x, t) e^{\pm i(kx - \omega t)}, \quad \hat{\mathcal{R}}_\pm(x, t) = \frac{1}{2} \hat{R}_\pm(x, t) e^{\mp i(kx - \omega t)}, \quad \hat{\mathcal{Z}} = 2\hat{\mathcal{R}}_3. \quad (6.2.12)$$

The coefficient of \hat{E}^\pm , containing T_R – the superradiation time introduced in (6.2.11) – has been chosen so that the slowly varying electric field amplitude is dimensionless, as well as \hat{R}_\pm , and \hat{Z} . Then using the Rotating Wave Approximation (RWA) and the Slowly Varying Envelope Approximation (SVEA) one arrives at the following system:

$$\begin{aligned} \left(\frac{\partial}{\partial x} + \frac{1}{c} \frac{\partial}{\partial t} \right) \hat{E}^\pm &= \pm \frac{i}{L} \hat{R}_\mp, \\ \frac{\partial}{\partial t} \hat{R}_\pm &= \frac{i}{T_R} \hat{E}^\mp \hat{Z}, \quad \frac{\partial}{\partial t} \hat{Z} = \frac{i}{2T_R} (\hat{R}_+ \hat{E}^+ - \hat{E}^- \hat{R}_-). \end{aligned} \quad (6.2.13)$$

It is impossible to solve these nonlinear Quantum Maxwell–Bloch (QMB) equations. In order to obtain results to be compared with experiments, one has to simplify the problem to a semiclassical treatment, which amounts to replace the operators in equations (6.2.13) by corresponding classical complex variables. With the convention: $\hat{E}^+ \rightarrow E^*$, $\hat{E}^- \rightarrow E$, $\hat{R}_+ \rightarrow R$, $\hat{R}_- \rightarrow R^*$, $\hat{Z} \rightarrow Z$, one obtains the undamped semiclassical Maxwell–Bloch (MB) equations, which are essentially the same as those in (1.1.45) but for the dimensionless classical quantities used in the present section. The *necessity of retaining* at least some *elements of a quantum treatment* is seen, however, from the following fact: If the variables in (6.2.13) are replaced by their classical counterparts, and the initial conditions correspond to total inversion density: $Z = 1$, zero polarization: $R = R^* = 0$, and no field present along the whole sample, then the system remains in that state, and does not start to radiate, as initially all the derivatives are equal to zero. This shows that quantum fluctuations are especially important in the initial stage of the process. *This quantum effect manifests itself in the experiments*, as well, the characteristic properties of the emitted pulses show *fluctuations* in repeated observations. The quantum nature of the initial stage can be incorporated into the semiclassical theory by a method worked out by Glauber and Haake, and by Polder and Schuurmans [79Haa, 79Pol, 81Haa, 81Sch]. The quantum correlation function of the atomic polarization operators in the initial state $|\{e\}\rangle$ (i.e. all the atoms in the upper state $|e\rangle$) gives:

$$\langle \{e\} | \hat{R}_+(x, t=0) \hat{R}_-(x', t=0) | \{e\} \rangle = \frac{4L}{N} \delta(x - x'), \quad (6.2.14)$$

which is the consequence of the relation $\langle \{e\} | \hat{R}_+^i \hat{R}_-^j | \{e\} \rangle = \delta_{ij}$ and the smoothing procedure. Considering the variables in (6.2.13) as classical stochastic quantities one solves the system for a large number of random initial conditions with correlation functions corresponding to (6.2.14). The average over the realizations is to be compared with the average experimental pulse forms.

Two comments on the one-dimensional approximation leading to (6.2.13) are in order:

1. As there are no end mirrors like in a laser, diffraction effects at the output of the long cylinder cannot be neglected. The reduction of diffraction would necessitate a sample with a large Fresnel number $\mathcal{F} = \mathcal{A}/\lambda L$. This, on the other hand, would contradict to the one-dimensional model. The optimal choice for the geometry is therefore to choose a Fresnel number close to 1 [79Pol, 82Gro, 96Ben]. Theories that take into account diffraction in more detail have been considered in [81Mat, 89Ave, 89Sch, 91Ave, 96Ben].
2. The form of SVEA in (6.2.12) assumes the propagation only in the positive x direction. There should be other terms with phase $(\omega t + kx)$ describing waves propagating in the opposite direction. In the observations of SR both pulses appear, but according to the experiments [81Vre] the two pulses can be considered to be independent, at least in gases. In solids with a greater density of active atoms there seems to be a correlation between the counterpropagating pulses, for an experiment see [84Flo]. Theoretical proposals explaining this correlation can be found in [92Jan, 94Tri, 94Mal, 96Ben].

In order to take into account relaxation effects, one can introduce phenomenological damping terms with time constants T_1 and T_2 describing population decay and polarization dephasing, respectively. The distribution of the atoms according to their resonant frequencies – inhomogeneous broadening – can be also included by a line shape function $g(\nu) = g(\omega_0 - \omega_c)$, where ω_c is a central

frequency assumed to be identical with the carrier frequency of the field. $g(\nu)$ is usually a Gaussian of the form $g(\nu) = (T_2^*/\sqrt{\pi}) \exp(-\nu^2 T_2^{*2})$. The resulting equations are then the same as the ones of semiclassical laser theory. We stress, however, that *superradiance in the pure sense is possible only if all these additional dephasing effects are absent*, i.e. the process is faster than T_1 , T_2 and T_2^* .

In SR the atoms of the system are initially in their excited states. Therefore, an extended superradiant sample is a coherent amplifier, because during propagation the field amplitude is growing. Owing to its principal similarity to superradiance, we also consider briefly the coherent attenuator which differs from SR in the initial conditions. In the attenuator all atoms are in the ground state in the beginning, and there is an injected external pulse.

In the undamped MB equations (see Sect. 1.1.7.3) the variables allow the following parameterization: $Z = \cos \theta$, $|R| = \sin \theta$. This means, that the values of the quasiclassical atomic variables can be illustrated by a point on the “Bloch sphere”. Then, in the absence of phase modulation (when E is real), the classical counterparts of (6.2.13) yield $\theta(x, t) = \frac{1}{T_R} \int_{-\infty}^t E(x, t') dt'$, which means that a pulse with electric field E generates a polarization and depopulates the upper level of the amplifier. Important information on coherent propagation of light pulses – in particular that of a superradiant pulse – is provided by a general result, the area theorem of McCall and Hahn [69McC, 75All]. The pulse area is defined as:

$$\mathcal{A}(x) := \theta(x, t = \infty) = \frac{1}{T_R} \int_{-\infty}^{\infty} E(x, t') dt' . \quad (6.2.15)$$

This dimensionless quantity obeys a simple differential equation:

$$\frac{d\mathcal{A}}{dx} = \pm \frac{\alpha}{2} \sin \mathcal{A} , \quad \text{with} \quad \alpha = \frac{2\pi g(0)}{LT_R} , \quad (6.2.16)$$

where the + and – signs correspond to the amplifier and attenuator, and α is the gain or the absorption coefficient, respectively. Equation (6.2.16) admits the following solution:

$$\tan(\mathcal{A}/2) = \tan(\mathcal{A}_0/2) \exp[\pm(\alpha/2)(x - x_0)] . \quad (6.2.17)$$

In the case of an attenuator the asymptotic value $\mathcal{A}(\infty)$ is necessarily $2n\pi$, $n = 0, 1, 2, \dots$, where n is determined by \mathcal{A}_0 . In the case of an amplifier the asymptotic value is $(2n + 1)\pi$ depending again on \mathcal{A}_0 . In SR, where initially no field is present, the asymptotic value of the pulse area is π .

The undamped Maxwell–Bloch system has analytical solutions for the attenuator. The simplest one is the particularly stable “ 2π secanthyperbolic pulse” [69McC, 75All]:

$$E = \frac{2T_R}{T} \operatorname{sech} \left[\frac{1}{T} \left(t - \frac{x}{v} \right) \right] . \quad (6.2.18)$$

The duration T and the velocity v of this pulse are in the relation: $c/v = 1 + \alpha cT/2$. This ratio can be of the order of 10^2 , the velocity of the coherent pulse is reduced by two orders of magnitude if compared with the incoherent propagation. The stability of the secanthyperbolic solution is explained in the mathematical theory of solitons. Experiments as well as theory show that all incoming pulses for which the area at the input is $\pi < \mathcal{A}_0 < 3\pi$ are transformed into this type of pulse after sufficiently long propagation in an attenuator. McCall and Hahn have coined the name Self-Induced Transparency (SIT) to this effect, and demonstrated its remarkable properties in ruby [69McC]. Several other works on SIT are cited in [75All].

For superradiance, i.e. for an amplifier with the initial condition $E(x, t = 0) = 0$, there is no analytic solution of the MB equations even in the absence of the relaxation terms. We know, however, that during the propagation the *intensity* – the area below the square of the amplitude – must grow, while the *area* below the slowly varying field amplitude *cannot be larger than π* , if initially the field was zero. Therefore the slowly varying *field envelope* must change its sign several times. The sign change of the envelope means that the field oscillates in opposite phase than before, and therefore a series of pulses occur in the output radiation. This effect, called ringing, has been first predicted in [69Bur].

A characteristic property of an initially completely inverted superradiant system is that the evolution of the process begins after a certain delay time. In the experiments this delay of the radiated pulses shows fluctuations, which is the consequence of the intrinsic quantum nature of the effect. The mean value of the time instants when the first peak of the SR pulse appears after the excitation is denoted by T_D . The analysis of the MB equations allows one to calculate its value, as well as the width of this first peak, T_W . The calculation based on the correlation function (6.2.14) yields also the relative standard deviation $\Delta T_D = (\langle T_D^2 \rangle - \langle T_D \rangle^2)^{1/2} / \langle T_D \rangle$ of the delay time. One has [76Mac, 79Haa, 79Pol, 81Mac, 81Sch]:

$$T_D = \frac{1}{4} T_R \left[\ln \frac{R_0}{2\pi} \right]^2, \quad T_W = T_R \ln \left| \frac{R_0}{2\pi} \right|, \quad \Delta T_D = 2.3 / (\ln N), \quad (6.2.19)$$

where R_0 is the average initial value of $|R|$ in the MB equations, which – according to (6.2.14) – is given by $R_0 = 2/\sqrt{N}$. The initial fluctuations of $|R| = \sin \theta$ can be considered as the fluctuations of θ , and the small initial mean value is $\theta_0 \approx \sin \theta_0 = 2/\sqrt{N}$, while the initial phase of R is completely random. The experiment confirming this value of the initial “tipping angle” θ_0 [79Vre] will be described below.

We summarize now the conditions to observe superradiance in an extended system. The relaxation times must be longer than the cooperative emission time, T_R , and if the atoms are to emit cooperatively, they must act as a single system, therefore the so-called Arecchi–Courtens cooperation time $T_c := T_R|_{L=cT_c} = \sqrt{T_R L}/c$ must be shorter than T_R , otherwise the system breaks into a number of independently radiating segments. Accordingly the characteristic times have to satisfy:

$$L/c < T_c < T_R < T_D < T_1, T_2, T_2^*. \quad (6.2.20)$$

The duration of the excitation T_p must also be shorter than the evolution of the process T_D .

Sometimes the relation $T_R < T_1, T_2, T_2^*$ is satisfied, but T_D is longer than the relaxation times. Then by injecting a small area pulse into the sample one can increase the initial polarization (tipping angle), and thus reduce T_D to make it shorter than the characteristic times of damping. In that case one speaks of *induced* or *triggered* superradiation [86Var, 87Mal].

If the time constant of the phase relaxation is shorter than the process: T_2 is smaller than T_R (this is the usual case), then the dominating term in the time derivative of R will be $-\frac{1}{T_2} R$, which has been omitted so far. Then, after a very short transition period the polarization becomes proportional to the field amplitude. In this way we arrive at the rate equation model of the atomic subsystem [75All, 96Ben], and the result is the incoherent process called Amplified Spontaneous Emission (ASE). It can be shown [96Ben] that inhomogeneous broadening leads to the same effect.

6.2.3 Superradiance experiments

The observations of SR can be classified according to the size of the sample compared with the emitted wavelength. Small sample experiments with Rydberg atoms in cavities [85Har], as well as with nuclear spins [90Baz] belong to one group. The other, much larger group consists of observations in extended pencil-shaped samples. A selective list of SR experiments is given in Table 6.2.1.

Table 6.2.1. Selected superradiance experiments in the infrared and visible.

Material	Wavelength	Remarks	Ref.
HF	252, 126, 84, 63, 50 μm	first demonstration	[73Skr]
Na vapor	3.41, 2.21, 9.10 μm	first near infrared	[76Gro]
Tl vapor	1.3 μm	SR suppressed by inhomogeneous broadening	[76Flu]
Cs vapor	2.931 μm	single pulses	[77Gib]
Rb atomic beam	2.79 ... 1.3 μm (7 lines)	polarization characteristics	[78Cru]
Cs vapor	3.1 μm	Doppler beats between different groups of atoms	[78Gro]
Eu vapor	557.7, 545.3, 1759.6 nm	first visible SR	[79Bre]
Eu vapor	605.7 nm (dominant)	20 lines	[79Cah]
Rb	2.79 μm	beats from different isotopes	[79Mar]
Cs vapor	2.931 μm	measurement of tipping angle	[79Vre]
Cs vapor	2.931 μm	triggered SR	[80Car]
Cs vapor	2.931 μm	delay time dependence on Fresnel number	[81Vre]
Cs vapor	3.01 μm	hyperfine structure quantum beats	[81Rys]
CH ₃ F	496 μm	single pulse, wide range of lengths, cross sections, pressures	[81Ros]
C ₁₂ H ₁₀ in diphenyl	373.9 nm	SR in an organic material	[83Zin]
O ₂ centers in KCl	592.8, 629.1 nm	two-color SR in a solid	[84Flo]
Cr: ruby, Nd:YAG at 100 K	694, 1061 nm	phase changes in ringing	[84Var]
Ga vapor	5.75, 1.21 μm	SR and the first observation of subradiance	[85Pav]
Cr: ruby, Nd:YAG at 100 K	694, 1061 nm	triggered SR, transition to ASE	[86Var]
O ₂ in KCl	629.1 nm	transition to ASE	[87Mal]
O ₂ centers in KCl	629.1 nm	two-dimensional SR from a lamellar sample	[87Sch]
O ₂ centers in KCl	629.04 nm	spatial coherence of SR	[88Sch]
CH ₃ F	496 μm		[91Sch]
CH ₃ CN	373 μm		[91Sch]
NH ₃	291 μm	two-photon noise initiated SR	[92Bak]
Pair of Ba ions	493 nm	microscopically resolved SR and subradiance	[96DeV]
Na BE-condensate	589 nm	superradiant Rayleigh scattering accompanied by emission of directed atomic beams	[99Ino]

6.2.3.1 A microscopic observation of superradiance and subradiance

In 1996 DeVoe and Brewer [96DeV] observed directly the collective spontaneous emission of trapped $^{138}\text{Ba}^+$ ions as a function of the ion-ion separation r . The ions were confined to fixed positions in a radio-frequency Paul trap. For two atoms (ions) the decay from the fully excited state $|e, e\rangle$ to the ground state $|g, g\rangle$ may go through the symmetric state $|s\rangle := (|e, g\rangle + |g, e\rangle)/\sqrt{2}$, as well as through the antisymmetric state $|a\rangle := (|e, g\rangle - |g, e\rangle)/\sqrt{2}$. If the distance between the atoms r is much less than the wavelength of the emission, $kr \ll 1$, then the decay rate through the symmetric channel is twice that of the single atom rate (superradiance), see (6.2.9), while the transition probability through the antisymmetric channel is approximately zero. The latter is a special case of subradiance. However, if the correct r dependence of the electromagnetic interaction between the two atoms is taken into account, then both the symmetric, and the antisym-

metric channels have a nonvanishing decay rate. In the experiment, so far, only $kr \gg 1$ could be realized, and then the following approximate formula is valid for the collective decay rates: $\Gamma_{\pm}(r) = \gamma(1 \pm \beta \sin(kr)/kr)$, corresponding to the symmetric (+) and the antisymmetric (−) channels. For an ideal case with nondegenerate levels the value of the constant is $\beta = 1.5$. The experimental conditions of [96DeV] corresponded to $\beta = 0.33$. The collective decay rate depends also on the relative orientations of the atomic dipoles. The formula for Γ_{\pm} above is for the case that the dipoles are parallel to each other and perpendicular to the vector \mathbf{r} joining them. This orientation could be achieved by a polarized exciting laser beam with polarization vector \mathbf{e}_1 perpendicular to \mathbf{r} . For collinear dipoles the collective effect is suppressed. (For the general dependence on the distance and relative orientation see [95Bre, 96Ben]). In the experiment the antisymmetric state $|a\rangle$ was excited and the decay rate was measured. The relevant part of the curve together with the experimental results are shown in Fig. 6.2.1.

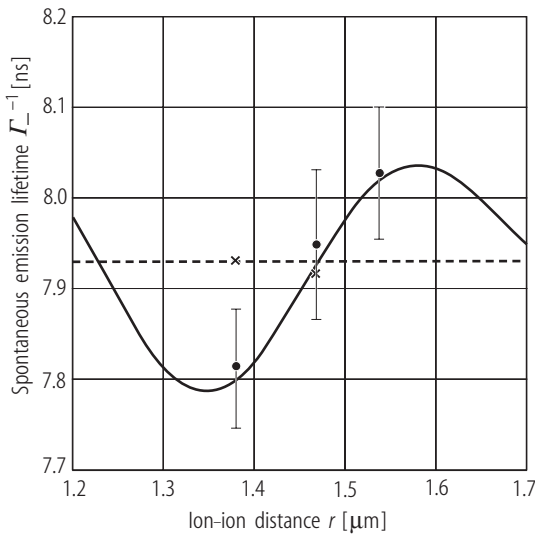


Fig. 6.2.1. Theoretical (solid curve) and experimental values of collective lifetimes ($1/\Gamma_-$) at $r = 1380, 1470$, and 1540 nm. The single-ion lifetime in the same trap is shown by the dashed horizontal line. Full circles with error bars: exciting laser polarization $\mathbf{e}_1 \perp \mathbf{r}$, the dipoles are parallel. Crosses (error bars omitted): $\mathbf{e}_1 \parallel \mathbf{r}$, the dipoles are collinear, a collective effect is not expected. The point for which the lifetime is smaller than the single-atom value is a signature of superradiance, while the point above that value corresponds to subradiance [96DeV].

6.2.3.2 Superradiance experiments in pencil-shaped macroscopic samples

These experiments can be divided again into two classes with respect to the time scale of the relaxation processes. In gases the relaxation times are typically in the nanosecond range whereas in solids these processes are faster, by at least one or two orders of magnitude. Therefore to avoid phase relaxation the solid samples must be kept at low temperature.

In order to observe SR, one has to achieve a possibly complete inversion on a nondegenerate two-level transition, without competing transitions. This is usually accomplished via a third higher level, or by a cascade decay through several levels. In some of the experiments magnetic fields have been applied to remove degeneracy. The excitation goes either along the axis of the cylinder or with uniform lateral pumping. The typical setup and level scheme is shown in Fig. 6.2.2 [73Skr, 76Mac]. The geometry of the sample corresponds usually to a Fresnel number $\mathcal{F} = 1$. The resulting series of SR pulses of the experiment [73Skr] are shown in Fig. 6.2.3. The multiple peaks are attributed to Burnham–Chiao ringing [69Bur, 96Ben]. According to an interference experiment in ruby [84Var], the consecutive peaks are in opposite phases, as required by the one-dimensional propagation theory (see the explanation in the second paragraph after (6.2.18)).

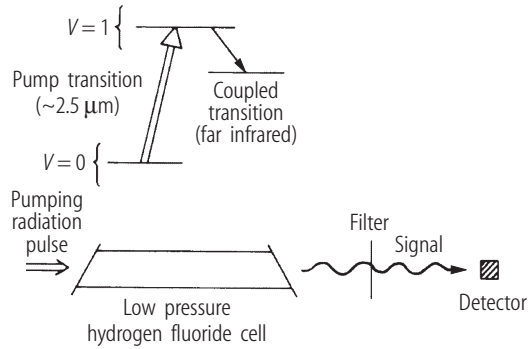


Fig. 6.2.2. Level scheme for hydrogen fluoride, HF, and principle of the experimental arrangement of the first observation of SR [73Skr, 76Mac]. The windows of the sample cell are tilted to prevent multiple reflections.

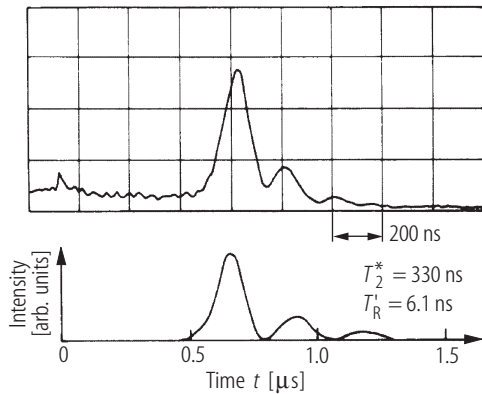


Fig. 6.2.3. Oscilloscope trace of an SR pulse observed in HF gas at 84 μm on the $J = 3 \rightarrow 2$ rotational transition, and theoretical fit [73Skr]. The parameters are: pump intensity $I = 1 \text{ kW/cm}^2$, gas pressure $p = 1.3 \text{ mtorr}$, cell length $L = 1 \text{ m}$. The small peak on the oscilloscope trace at $t = 0$ is the 3 μm pump pulse, highly attenuated [73Skr].

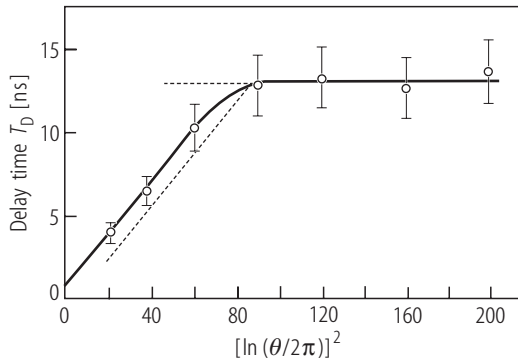


Fig. 6.2.4. Delay time T_D of the output pulse vs. $[\ln(\theta/2\pi)]^2$. The dashed line is used to correct for the delay of the injection pulse with respect to the pump pulse [77Gib].

An important step in resolving the question of quantum initiation of SR was the experiment of Vrehen and Schuurmans [79Vre]. They injected small area pulses, so-called tipping pulses, at the relevant wavelength into the sample immediately after it had been completely inverted by a short pump pulse. As long as the area of the tipping pulse θ was smaller than θ_0 (see the note after (6.2.19)), the delay time of the output pulse would not be affected. However, when $\theta > \theta_0$ the delay was reduced. Therefore, by measuring the delay time as a function of the area of the injected pulse, the magnitude of θ_0 could be found. This plot is shown in Fig. 6.2.4. As expected, T_D increased linearly for large injection pulses. For small injection pulses the initiation of SR was dominated by quantum fluctuations, and T_D was constant. At the cross-over $\theta = \theta_0$ by definition. The results confirm the theoretical value $\theta_0 \simeq 2/\sqrt{N}$.

Theoretical [81Mat, 83Wat, 89Ave, 91Ave] and experimental [81Vre] investigations confirm that transverse effects can significantly influence the pulse shape. The excitation generates a Gaussian profile in the inversion and this in turn leads to a radial dependence of the intensity peaks, delay

and ringing. According to the analysis of [81Mat], detectors not resolving this transversal dependence average out the ringing, this is how single pulses have been observed in [77Gib]. In the experiment [73Skr] the detector viewed a small area in the near field of the beam, and the central portion of the output pulse exhibited ringing (Fig. 6.2.3). According to another experiment, [81Vre], the observed results for the delay times were in reasonable agreement with the one-dimensional approach for Fresnel numbers $\mathcal{F} = 0.8 \dots 1$. For larger values of \mathcal{F} , both the delay time and its fluctuations were reduced. According to the estimations of [81Vre]: $\langle T_D \rangle \sim |\ln 2\pi N / \mathcal{F}|^2$, and $\Delta(T_D) \simeq 2.3 / [\mathcal{F} \ln(N/\mathcal{F})]$. The measured reduction is the consequence of the above mentioned averaging detection method.

6.2.3.3 Superradiant-type Rayleigh scattering from a Bose–Einstein condensate

An effect which is not strictly SR, but very similar to it has been observed [99Ino] in a Bose–Einstein (BE) condensate, which is an ideal cooperative system of atoms. The sample, which was $L = 200 \mu\text{m}$ in length and $20 \mu\text{m}$ in diameter, contained several million sodium atoms. It was exposed to laser pulses that intersected the condensate perpendicular to its long axis with pulse durations between 10 and $800 \mu\text{s}$. This excitation was slightly red detuned from the $3S_{1/2}, F = 1 \rightarrow 3P_{3/2}, F = 0, 1, 2$ transition. Pulses of intensity above $1 \text{ mW}/\text{cm}^2$ triggered collimated emission of light beams from both ends of the sample along its long axis. Typical images of the emitted pulses consisted of a few bright spots with angular widths equal to the diffraction limit for a source with a diameter of $14 \mu\text{m}$.

An entirely new feature of this experiment was that the atoms recoiled by the interaction with the exciting photons could be directly seen as highly directional beams flying out from the sample, when the confining magnetic trap was turned off. As required by momentum conservation, these beams propagated at an angle of 45° with the long axis of the condensate.

The observed effect was actually cooperative Rayleigh scattering with the characteristic properties of superradiance. Time-resolved measurements showed that by increasing the intensity of the excitation the evolution of the emitted pulses was shortened and became more highly peaked. The emission was also accompanied by a faster than normal exponential decay of the number of atoms remaining in the condensate at rest after excitation. For sufficiently intense excitation more peaks appeared in the emission, and several bunches of recoiled atoms were observed, as a result of sequential scattering. The radiated intensity is expected to be proportional to the *square* of the contrast of the matter wave interference pattern, although in the experiment [99Ino] the density of the condensate has not been varied.

This strong cooperative effect could be observed only when the polarization of the exciting laser was perpendicular to the long axis of the condensate, so that dipole radiation from the atoms could be coherently amplified along this axis. To the contrary, excitation with polarization parallel to the long axis resulted in ordinary weak isotropic Rayleigh scattering. The superradiant features also disappeared when the temperature was raised above the condensation value, mainly because the collective phase memory time T_2^* was then reduced by Doppler broadening with a factor of 30. Therefore the appearance of the superradiant emission indicated sensitively the appearance of the Bose–Einstein phase transition.

6.2.4 Outlook

The fast development of experimental techniques for trapping atoms and ions will allow to extend SR experiments in the microscopic domain. As a further development of the works described in [85Har] and [96DeV] one expects to observe SR from a few atoms (well-defined number) confined in a trap in a possibly symmetrical arrangement. This technique is promising also in modeling and probably realizing information processing with quantum-optical methods (quantum informatics), see also Sect. 5.1.10.3. At present the main obstacle in the way of making a quantum computer is decoherence, the same effect that usually destroys superradiance.

As for the extended systems, the wavelength can be further reduced. A strong stimulation of superradiance research is the fact that it can be an intensive source of X rays and γ rays from nuclear transitions, where no easily handled mirrors are present (Vol. VIII/1B, Chap. 7.1: “X-ray lasers”). Note also that the $\sim N^2$ dependence of the intensity has already been realized in Free Electron Lasers (FEL). In this sense a FEL can also be considered as a source of superradiation (Vol. VIII/1B, Chap. 6.1: “Free-electron lasers”). Possible other sources of SR could be the electron–hole plasma and/or excitons in semiconductors. In these cases the characteristic quadratic dependence of the radiated intensity on the number of the emitting centers must be proven. One also expects new experiments demonstrating SR from Bose–Einstein condensates. In connection with lasing without inversion, also a scheme of superradiance without inversion has been considered [98Mal].

Several theoretical works predict special effects in photon statistics if superradiance is initiated by squeezed vacuum instead of true vacuum, for references see [96Ben]. Even a “superradiant laser” has been proposed by Haake et al. which would produce intensive squeezed light [93Haa]. It is interesting, however, that experimental investigations of the photon statistics of any superradiant light are completely missing.

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