INHOMOGENEOUS BROADENING IN THE THEORY OF SUPERRADIANT EMISSION*

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Using the theory of superradiant spontaneous emission as formulated by Eberly and Rehler, it is shown how to incorporate the effect of inhomogeneous broadening of the atomic transition. Specifically, an explicit expression is given for the intensity of emission as a functional of the atomic line shape. In the simplest case, that of Lorentzian line shape, the evaluation of the functional leads to the appearance of the transverse relaxation time T_2^* in a natural way.

The subject of cooperative spontaneous emission, or superradiance, was first discussed in a famous paper by Dicke (1954). The phenomenon itself is intrinsically interesting because it very subtly blends the quantum and the classical, or the spontaneous and the stimulated, aspects of atomic emission. It has attracted the attention of a large number of workers in quantum optics recently (see, for example, Ernst and Stehle 1968, Arecchi *et al.* 1969, Eberly and Rehler 1969 and 1970, Lehmberg 1969, Agarwal 1970, Allen and Peters 1970, Bonifacio *et al.* 1970), and will almost certainly become the subject of even more widespread discussion.

Until the very recent past most theoretical work on superradiant phenomena was confined to systems of atoms contained in regions of the order of the optical wavelength, or smaller, in size. From an experimental point of view, this is an extremly unrealistic restriction. A detailed examination, both analytical and numerical, of superradiant emission from macroscopic objects is now available (see, for example, Rehler and Eberly 1971, and other references therein).

However, another equally serious restriction remains in all work on superradiance to date. There is no discussion, of a quantitative nature, of the influence of inhomogeneous broadening on the emission process. All workers assume that each atom cooperating in the emission process has the same transition frequency ω. However, atomic linewidths of the order of 10^{12} sec^{-1} are not uncommon in materials of interest. In such materials the phase relationships between the wavefunctions of the constituent atoms are destroyed in less

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than a picosecond. Since it is these phase relationships which are at the heart of cooperative emission, the quantitative consideration of finite, even large, atomic linewidths demands attention.

In this note we intend to show that the formulation of the theory of superradiant emission from macroscopic systems given by Rehler and the present author (Rehler and Eberly 1971) is ideally suited for the inclusion of inhomogeneous broadening.

We may begin by reviewing briefly the important points of our theory (see Eberly and Rehler 1970 for another capsule summary and another application, of the full theory presented in Rehler and Eberly 1971). The quantity of interest is the expectation of the Poynting vector of the emitted radiation, which we will call the "intensity" of the radiation. To compute this quantity we work in quantum electrodynamics, and first compute the electric and magnetic field strengths from the Hertz dipole operator of the atomic system. The Poynting vector, and thus the intensity, are then given by the usual combinations of electric and magnetic field strengths. The important element of this approach is that it shows, in quantum electrodynamics, how the field intensity is based on sums of atomic dipole operators.

It is sufficient to look at the case of N atoms with a $\Delta m = 0$ electric dipole transition. The atoms are assumed identical except for their transition frequencies ν_m which are distributed in some way about a central frequency ω and their positions $\boldsymbol{r}_m$, which are distributed randomly throughout the region the atoms occupy. If this region is small enough to allow retardation effects within it to be neglected, then the leading contribution to the Hertz vector operator for the system is given by:

$$\pi^{(\pm)}(\boldsymbol{r}, t) = \frac{\hat{z}p}{r}\, u^{-1}(T, 0)\left[\sum_{m=1}^{N} R_{m\pm} e^{\pm i\left[\nu_m\left(T+\frac{\hat{\boldsymbol{r}}\cdot\boldsymbol{r}_m}{c}\right)+\varphi_m\right]}\right] u(T, 0). \tag{1}$$

Here $\boldsymbol{r}$ and t are the position and time of the observation point, and T is the retarded time, $T = t - \frac{r}{c}$, all measured with respect to the place and time of the emission. The dipole matrix element is denoted p, and for simplicity we assume the atoms to have a common magnetic axis $\hat{\boldsymbol{z}}$.

The operators $R_{m\pm}$ are the usual raising and lowering operators for the m^{th} atom, defined for example by Dicke (1954); the phases φ_m are determined by initial conditions and will be specified later; and the unitary operators u and u^{-1} are those which connect the Heisenberg and interaction pictures. Note that we have given the positive and negative frequency parts of the Hertz vector, $\boldsymbol{\pi}^{(\pm)}(\boldsymbol{r}, t)$. The vector itself, which is given by the sum $\boldsymbol{\pi} = \boldsymbol{\pi}^{(+)} + \boldsymbol{\pi}^{(-)}$, is not needed.

It is convenient now to introduce a "de-tuning" frequency γ_m for each atom:

$$\gamma_m = \nu_m - \omega \tag{2}$$

and furthermore, to adopt the notation $S_{k\pm}$ for the following sum of atomic operators:

$$\omega^2 S_{k\pm} = \sum_{m=1}^{N} \nu_m^2 R_{m\pm} e^{\pm i(\gamma_m T + \boldsymbol{k}_m \cdot \boldsymbol{r}_m + \varphi_m)}. \tag{3}$$

We have defined the wave vector $\boldsymbol{k}_m = (\nu_m/c)\hat{\boldsymbol{r}}$, which bears the same relation to the "central" wave vector $\boldsymbol{k} = (\omega/c)\hat{\boldsymbol{r}}$ that ν_m does to ω. Note that, when twice differentiated with respect to either space or time, the bracketed sum of operators in (1) will give an expression very closely related to $\omega^2 S_{k\pm} \exp(\pm i\omega T)$. Since the Hertz vector must be twice differentiated to yield the electric and magnetic fields, we can anticipate that the operators $S_{k\pm}$ will be significant.

In fact, in the manner described in the fifth paragraph, we may easily establish that the radiated intensity is given by the simple expression:

$$I[\boldsymbol{k}, t] = I_0(\boldsymbol{k}) \langle \psi_H | u^{-1}(T, 0)\, S_{k+} S_{k-} u(T, 0) | \psi_H \rangle, \tag{4}$$

where $|\psi_H>$ is the appropriate Heisenberg state, and $I_0(\boldsymbol{k})$ is the intensity of emission from a single atom of the kind we are considering. That is, it is given by the usual dipole formula:

$$I_0(\boldsymbol{k}) = \frac{p^2\omega^4}{2\pi c^3} [1-(\hat{\boldsymbol{k}} \cdot \hat{\boldsymbol{z}})^2]. \tag{5}$$

Except for the appearance of the non-zero de-tuning frequencies γ_m in the definition of the $S_{k\pm}$ operators in (3), the intensity formula (4) is identical with that given previously (Rehler and Eberly 1971), and the same approximations were used here in its derivation. The only approximation used, but not already mentioned explicitly here, is a slowly-varying envelope approximation in which derivatives of u and u^{-1} are assumed much smaller than derivatives of $\exp(\pm i\omega T)$. This approximation may bevalidated a posteriori.

We now turn our attention to the evaluation of the intensity expectation in (4). Let us first of all abbreviate some of the notation. It suffices to introduce the state $|\chi>$ and the position-dependent phase Φ_m *via* the definitions:

$$|\chi> = u(T, 0)|\psi_H> \tag{6}$$

$$\Phi_m(\boldsymbol{r}_m) = \boldsymbol{k}_m \cdot \boldsymbol{r}_m + \varphi_m \tag{7}$$

Aided by definitions (6) and (7), as well as the expression (3) for $S_{k\pm}$, and by making a kind of stationary phase approximations in which we ignore the small de-tuning frequencies γ_m except when they appear in exponents, we are able to reduce (4) to the expression:

$$I(\boldsymbol{k}, t) = I_0(\boldsymbol{k}) \sum_{m,l} <\chi|\, R_{m+} R_{l-} |\chi>\, e^{i[(\gamma_m - \gamma_l)T + \Phi_m - \Phi_l]}. \tag{8}$$

The next step is to seperate the one-atom from the two-atom contributions to $I(\boldsymbol{k}, t)$. We are then left with the unwieldy formula:

$$I(\boldsymbol{k}, t) = I_0(\boldsymbol{k}) \sum_{m=1}^{N} \langle\chi|R_{m+}R_{m-}|\chi\rangle +$$

$$+ I_0(\boldsymbol{k}) \sum_{l}^{N} \sum_{m\neq l}^{N} \langle\chi|R_{m+}R_{l-}|\chi\rangle e^{i[(\gamma_m - \gamma_l)T + \Phi_m - \Phi_l]} \tag{9}$$

But the atoms themselves are identical, by assumption, except for their different positions and different amounts of detuning. If the state $|\chi>$ does not itself introduce any distinctions between the atoms (and all theories of superradiance assume as much) then the expectations to be computed in (9) are actually independent of m and l and may as well be evaluated for atoms 1 and 2 as any other. This identity of the atoms produces a great simplification, of course. Instead of (9) we may write

$$I(\boldsymbol{k}, t) = NI_0(\boldsymbol{k}) \langle\chi|R_{1+} R_{1-}|\chi\rangle + \tag{10}$$

$$+I_0(\boldsymbol{k}) \langle\chi|R_{1+}R_{2-}|\chi\rangle \sum_m \sum_{l\neq m} e^{i[(\gamma_m-\gamma_l)T+\Phi_m-\Phi_l]}.$$

As a final clarification we rewrite the double sum remaining in (10). If the missing elements $l = m$ are restored, the double sum factors into the product of two sums which are complex conjugates of each other. Since the contribution of the missing elements (the sum along the "diagonal") is just N, we may write:

$$\sum_l \sum_{m\neq l} e^{i[(\gamma_m-\gamma_l)T+\Phi_m-\Phi_l]} = |\sum_m e^{i(\gamma_m T+\Phi_m)}|^2 - N. \tag{11}$$

We may interpret the lone remaining summation as N times the average value of exp $(i\gamma_m T+ +i\Phi_m)$ taken over all N atoms. Let us denote this average by $\Gamma(\boldsymbol{k}, t)$ in an obvious generalization of a notation introduced earlier by Eberly and Rehler (1969). It is then clear that the full expression for the expectation of the intensity takes the simple form:

$$I(\boldsymbol{k}, t) = NI_0(\boldsymbol{k}) \left[\langle[1]\rangle + N\langle[2]\rangle \left\{\Gamma(\boldsymbol{k}, t) - \frac{1}{N}\right\}\right] \tag{12}$$

where $\langle[1]\rangle$ and $\langle[2]\rangle$ obviously stand for the one-atom and two-atom expectations in (10).

Before evaluating $\Gamma(\boldsymbol{k}, t)$ let us make a remark or two in general. The expression for $I(\boldsymbol{k}, t)$ in (12) shows how few restrictions there actually are on the occurence of superradiance. There will be a term in $I(\boldsymbol{k}, t)$ proportional to N^2, *i. e.*, there will be superradiance, whenever $\Gamma(\boldsymbol{k}, t)$ and $\langle[2]\rangle$ exist, making the reasonable assumption that they will be independent of N. The one-atom expectations are entirely irrelevant to superradiance, as are all p-atom expectations, for $p > 2$.

The evaluation of $\Gamma(\boldsymbol{k}, t)$ is straightforward in principle, and in some cases can be carried out explicitly. According to our definition, we are interested in an average

$$\frac{1}{N} \sum_m e^{i[\gamma_m T+\Phi_m(r_m)]} \equiv \{e^{i(\gamma T+\Phi)}\}_{\text{avg.}} \tag{13}$$

to be taken over all of the atoms present. We may first re-apply our earlier assumption that retardation effects within the region of the atoms can be neglected. In this case the position and de-tuning dependences of the exponential are separated. By asserting that the distribution of de-tunings among the atoms is statistically independent of the distribution of positions, which is only reasonable, we are free to compute the averages over de-tuning and posi-

tion separately. The absolute square of the position average leads again to the quantity denoted $\Gamma(\boldsymbol{k}, \boldsymbol{k}_1)$ for plane-wave-excited atoms by Eberly and Rehler (1969).

The de-tuning average may be simplified by introducing the inhomogeneously broadened atomic line shape function $g(\gamma)$ under the assumption that the de-tunings are densely distributed in some range of frequencies. We assume $g(\gamma)$ is normalized to unity, in which case

$$\{e^{i\gamma T}\}_{\text{avg.}} = \int_{-\infty}^{\infty} e^{i\gamma T} g(\gamma)\, d\gamma. \tag{14}$$

Thus the absolute value of the average in (13), *i. e.* $\Gamma(\boldsymbol{k}, t)$, may be written $\Gamma(\boldsymbol{k}, t) = \Gamma(t)\, \Gamma(\boldsymbol{k}, \boldsymbol{k}_1)$, assuming initial excitation by a plane wave with vector $\boldsymbol{k}_1$, following Dicke (1954) and Eberly and Rehler (1969), where we define

$$\Gamma(t) = |\int e^{i\gamma T} g(\gamma) d\gamma|^2 \tag{15}$$

in accordance with our present results. Therefore we may say that the effect of inhomogeneous broadening (*i. e.*, of a spread of atomic resonant frequencies) is accounted for by an integral functional of the atomic line shape function.

There are two very simple special cases which we may comment on separately. In the case of a Lorentzian atomic line, the Fourier transform given in (14) leads to exponential damping, of course. The form of the superradiant term in the intensity expression (12) is then:

$$I(\boldsymbol{k}, t)\Big|_{\substack{\text{superrad.}\\ \text{part}}} = N^2\, I_0(\boldsymbol{k})\, \langle[2]\rangle\, \Gamma(\boldsymbol{k}, \boldsymbol{k}_1)\, e^{-2|T|/T_2^*} \tag{16}$$

where T_2^* is, roughly speaking, the inverse of the width of the Lorentzian, and is the usual reversible transverse moment decay time. That this interpretation is consistent with (16) is apparent in all specific models of superradiance, because in them $\langle[2]\rangle$ is related to the square of the single-atom dipole moment. Note that in (16) one sees that the intensity will decay as an explicit function of time, independently of the time dependence of $\langle[2]\rangle$. For this reason it will be difficult to extend the recent analytic solutions given for the time behaviour of $I(\boldsymbol{k}, t)$ (see Rehler and Eberly 1971, and references therein) to the case of inhomogeneously broadened systems (see, however, Agarwal 1971.)

The second very simple special case is that of a symmetric double-peaked atomic line shape function $g(\gamma)$. It is clear from (14) and (15) that the superradiant part of $I(\boldsymbol{k}, t)$ in this case will exhibit oscillations at the beat frequency between the two peaks of the line. We mention this obvious result only because the laser transition in ruby exhibits just such a doubly peaked line. (For extended remarks, see Eberly 1971.)

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