Directed Spontaneous Emission from $N$-atom Extended Ensemble

HARRY J. LIPKIN

Department of Particle Physics Weizmann Institute of Science, Rehovot 76100, Israel
School of Physics and Astronomy, Raymond and Beverly Sackler Faculty of Exact Sciences, Tel Aviv University, Tel Aviv, Israel
Advanced Photon Source, Argonne National Laboratory, Argonne, IL 60439-4815, USA

Abstract

Coherence and interference play crucial roles in emission and absorption of photons to and from large systems with many atoms. Confusion has arisen because nuclear X-ray physicists and atomic quantum-optics physicists do not understand one another’s individual descriptions of related phenomena. Basic physics same for all wave lengths from optical to nuclear gamma ray photons. But different languages are used to describe this physics in different domains. Crucial parameters vary over many orders of magnitude and what is intuitive or counterintuitive varies widely. Differences in parameters arising from differences between coherent emission effects in different domains produce very different results. Unified general treatment of the entire photon spectrum makes basic physics intelligible to all. In the optical region the mean distance between the scattering atoms is much longer than the photon wave length, Dicke superradiant scattering is isotropic and multiple scattering, Fano couplings are important and the lifetimes of intermediate stats are sufficiently short to be negligible. In X-ray scattering the mean distance between atoms is comparable to the photon wave length, Dicke superradiance is concentrated in a forward peak, multiple scattering and Fano effects are negligible, lifetimes are measurably long, speedup shortening the lifetime is important and most of the radiation is not elastically scattered but lost to absorption. Explicit calculations for a one-dimensional array shows the great difference between the case where the photon wave length is much shorter or comparable to the distance between nearest neighbors. A full investigation of the angular distribution and speedup of the intensity for two and three-dimensional scatterers give very different results for the two cases.
I. INTRODUCTION

The paper [1] “Directed Spontaneous Emission from an Extended Ensemble of $N$ atoms: Timing is Everything” considers a state prepared by absorbing a photon of wave vector $\vec{k}_0$ in which one atom is excited and we don’t know which one. It asks “When the absorbed photon is spontaneously emitted will it go in $4\pi$ sr. or will it be directionally correlated with $\vec{k}_0$? The perhaps counterintuitive answer is the latter.”

Such states have been prepared with photons of any wavelength from optical photons to nuclear gamma ray photons. The basic physics is the same for all, but different languages are used to describe this physics in different domains. Crucial parameters vary over many orders of magnitude and what is intuitive and what is counterintuitive vary widely. Confusion has arisen because nuclear X-ray physicists and atomic quantum-optics physicists do not understand one another’s individual descriptions of related phenomena. As Maurice Goldhaber [8] remarked about the discovery of the Mössbauer effect, “This will teach many old things to new people.”

The purpose of this paper is to provide a unified description which makes the basic physics intelligible to all, while pointing out some important differences between coherent emission effects in different domains and showing how differences in parameters produce very different results.

The first time this question was considered and called “superradiance” by Dicke [6] the emitted photon did go in $4\pi$ sr. In the nuclear gamma ray case this question has been investigated in detail both theoretically [2,3] and experimentally [4,5]. And the latter answer above is not counterintuitive at all. And in other domains Dicke superradiance [7] gave different answers.

The general situation can be summarized as follows:

1. The angular distribution of the emitted photon depends upon upon the parameters of the source. These parameters can vary by many orders of magnitude between optical and nuclear $\gamma$ ray sources. The angular distribution of the emitted photon can vary from isotropic to strongly forward peaked.

2. The photon emission is always speeded up for a three-dimensional system. The lifetime of this coherent or superradiant state is always much shorter than the lifetime of an isolated atom or nucleus in the given excited state. But the degree of speedup; i.e. the ratio of the lifetime of the superradiant state to the single atom lifetime depends upon the parameters of the source.

3. The probability that the initial excitation will produce a photon with the same wave length and energy as the initially absorbed photon varies widely over the different domains. In quantum optics, this probability is of order unity; in nuclear gamma radiation it is at best only a few percent and maybe considerably smaller.

4. The probability that a nuclear excitation will produce a photon with the same wave length and energy as the initially absorbed photon is very small for the following reasons:
The dominant process for energy transfer from an excited nucleus is internal conversion, in which no photon is emitted and an electron is ejected from one of the atomic shells. The ratio of the probability of this electron ejection to the probability that a photon is emitted is called the internal conversion coefficient and is greater than ten for most transitions relevant here.

In photon emission from a nucleus that is free and at rest, energy is lost in the recoil of the nucleus which balances the momentum of the emitted photon and there is no photon emitted with the energy of the initially absorbed photon.

In photon emission from a nucleus that is in thermal equilibrium at a sufficiently high temperature, the energy spectrum includes nuclei with sufficient energy to compensate for the recoil energy. Only these can be absorbed resonantly by another nucleus.

In photon emission from a nucleus bound in a crystal the recoil energy can be exchanged with emission and absorption of lattice phonons. The probability of elastic emission with no energy transfer to the lattice is called the Debye-Waller or Lamb-Mössbauer factor. Only these elastically emitted photons can participate in Dicke superradiance.

5. If the source dimension is not sufficiently small to allow the emitted photon to escape without further interactions complicated rescattering effects can occur which depend strongly on the source parameters and can vary wildly between different systems and different wavelengths. These are very different for optical photons and nuclear γ rays as listed below. There is no further discussion of multiple scattering in this paper.

- Continuous rescattering of optical photons is called “Fano coupling” and plays an important role on optical superradiance.
- Rescattering of superradiant nuclear γ rays can occur as the γ ray proceeds forward in a thick crystal. However because the speeded up superradiant photons have a much wider energy spectrum than the natural line width the absorption through the crystal distorts the energy spectrum, first making a hole as photons within the natural line width are absorbed. These effects are not discussed in this paper.

II. COMPARISON OF FORWARD AND $90^\circ$ EMISSION FROM AN ORDERED ARRAY

The essential differences are seen in comparing forward and $90^\circ$ emission from an ordered array of $N$ atoms equally spaced by a distance $\vec{D}$. Consider a state prepared by absorbing a photon of wave vector $\vec{k}_0$ in which one atom is excited and we don’t know which one.

In forward emission the path difference $\vec{D}$ between initial excitations of neighboring atoms in the array is exactly compensated by the path difference of the emitted photon. Thus all the forward amplitudes are in phase and the total forward amplitude is proportional to $N$.

$$A(0^\circ) = \sqrt{Na}; \quad I(0^\circ) = Na^2 \quad (2.1)$$
In 90° emission there is no path difference between emitted photons. The phase difference
\( \vec{k}_0 \cdot \vec{D} \) between the initial excitations of nearest neighbors thus remains in the outgoing radiation. The amplitudes with this phase difference are easily summed in a geometric series to give

\[
A(90^\circ) = \frac{a}{\sqrt{N}} \sum_{n=0}^{N-1} e^{in\vec{k}_0 \cdot \vec{D}} = \frac{a}{\sqrt{N}} \cdot e^{iN\vec{k}_0 \cdot \vec{D}} - 1
\]  

(2.2)

where \( a \) denotes the amplitude emitted from a single atom and is assumed to have an isotropic angular distribution

\[
\frac{A(90^\circ)}{A(0^\circ)} \approx \frac{1}{N} \cdot \frac{e^{iN\vec{k}_0 \cdot \vec{D}} - 1}{e^{i\vec{k}_0 \cdot \vec{D}} - 1}
\]  

(2.3)

When \( \vec{k}_0 \cdot \vec{D} \approx 0 \), \( e^{iN\vec{k}_0 \cdot \vec{D}} \approx 1 \), \( A(90^\circ) \approx A(0^\circ) \) and the angular distribution is approximately isotropic.

But when \( \vec{k}_0 \cdot \vec{D} \) is of order unity, the amplitude at 90° is strongly suppressed.

\[
\frac{A(90^\circ)}{A(0^\circ)} \approx \frac{1}{N^2}; \quad \frac{I(90^\circ)}{I(0^\circ)} \approx \frac{1}{N^2}
\]  

(2.4)

In a disordered linear array the intensity is the sum of intensities from \( N \) nuclei with random phases.

\[
I_{dis}(90^\circ) = a^2; \quad \frac{I_{dis}(90^\circ)}{I(0^\circ)} \approx \frac{1}{N}
\]  

(2.5)

Three different domains of values of the parameter \( \vec{k}_0 \cdot \vec{D} \) lead to very different conclusions.

1. When \( N\vec{k}_0 \cdot \vec{D} \approx 0 \), \( e^{iN\vec{k}_0 \cdot \vec{D}} \approx 1 \), \( A(90^\circ) \approx A(0^\circ) \) and the angular distribution is approximately isotropic. This is simple Dicke superradiance.

2. When \( \vec{k}_0 \cdot \vec{D} \) is of order unity, \( A(90^\circ) \approx (1/N) \cdot A(0^\circ) \). The emission at 90° is strongly suppressed. This is the case in nuclear resonance scattering.

3. When \( 0 \leq n\vec{k}_0 \cdot \vec{D} \leq 1 \) for small but finite values of \( n \) much less than \( N \) and \( n\vec{k}_0 \cdot \vec{D} \) reaches unity for a finite value of \( n \ll N \). Then \( 0 \ll A(90^\circ) \ll A(0^\circ) \). There may be interesting physics in this intermediate domain which may be considered counterintuitive in quantum optics. It goes beyond simple isotropic Dicke superradiance and does not exist in the X-ray region.

### III. EXPLICIT CALCULATION OF THE INTENSITY SUMMED OVER THE ANGULAR DISTRIBUTION

Forward coherence occurs only in the exact \( \vec{k}_0 \) direction. That the amplitude for the emitted photon to be enhanced by a factor \( N \) in the exact forward direction can be shown to be true in general for all sources. However, the exact forward direction has zero solid angle. The forward intensity observed in an experiment is an integral over a finite solid angle.
If individual sources of a system of $N$ atoms are at positions denoted by $r_\mu$ and radiate isotropically the total radiation intensity $I$ integrated over all angles can be shown to be

$$I_{\text{tot}} \equiv \int I d\Omega = \frac{4\pi}{N} \cdot \sum_\mu \sum_\nu i_1 e^{i(\phi_\mu - \phi_\nu)} \cdot \frac{\sin(k|r_\mu - r_\nu|)}{k|r_\mu - r_\nu|} \quad (3.1)$$

where $i_1$ is intensity per unit solid angle for the transition from a single excited nucleus and the phases $\phi_\mu$ and $\phi_\nu$ depend upon the preparation of the source. For simplicity we drop the subscript $0$ and define $\vec{k} \equiv \vec{k}_0$ If the source is concentrated in a very small volume, $k|r_\mu - r_\nu| \ll 1$, $I_{\text{tot}} \propto N$ and typical Dicke superradiance is observed.

If, however, $k|r_\mu - r_\nu| \gg 1$, the factor $k|r_\mu - r_\nu|$ in denominator suppresses contributions from pairs separated by large distances and $I$ is no longer $\propto N$.

Explicit $N$ dependence is seen by assuming optimum phase, introducing cylindrical coordinates $(r, z)$ for an axially symmetric cylindrical system with radius $a$ and length $L$, evaluating the double sum by converting one sum to an integral, and neglecting differences between the distances to boundaries of different atoms.

$$I_{\text{tot}} = 4\pi i_1 \int_0^L dz \int_0^a 2\pi \rho \, r \, dr \, \cos(kz) \cdot \frac{\sin kR}{kR} \quad (3.2)$$

where $\rho$ denotes density of atoms and we define

$$R \equiv \sqrt{r^2 + z^2}; \quad R_{\text{max}} \equiv \sqrt{a^2 + z^2} \quad (3.3)$$

Changing variables for the integration over $r$ and noting that $rdr = RdR$ gives

$$I_{\text{tot}} = 8\pi^2 i_1 \rho \int_0^L dz \int_0^{R_{\text{max}}} dR \, \cos(kz) \cdot \frac{\sin kR}{k} \quad (3.4)$$

$$I_{\text{tot}} = 8\pi^2 i_1 \rho \int_0^L dz \cos(kz) \cdot \frac{\cos kz - \cos kR_{\text{max}}}{k^2} \approx 8\pi^2 i_1 \rho \int_0^L dz \frac{\cos^2 kz}{k^2} \approx 4\pi^2 i_1 \rho L \frac{\cos^2 kL}{k^2} \quad (3.5)$$

where we have neglected the oscillating part of the integral over $z$.

This result can be compared with the exact forward intensity which is always enhanced by a factor $N = \pi a^2 L \rho$. This would give a total intensity of $4\pi^2 i_1 a^2 L \rho$ if this intensity remained constant over the full $4\pi$ solid angle. We can therefore define an “effective solid angle” for the forward beam as

$$\Omega_{\text{eff}} = \frac{I_{\text{tot}}}{4\pi^2 i_1 a^2 L \rho} = \frac{1}{k^2 a^2} \quad (3.6)$$

The result shows enhancement or “speedup” of intensity compared to decay of a single nucleus by a factor $2\pi \rho L / k^2$ and a narrowing of the solid angle of the forward peak by a factor of $k^2 a^2$. This shows that increasing the length of the source in the direction of the incident photon increases the speedup as well as increasing the forward intensity, while increasing the size of the source in the direction normal to the incident photon direction does not increase the speedup; it only increases the forward intensity at the expense of narrowing the angular distribution.
One particularly interesting case is the “end fire mode” for a long thin needle [7] of length \( L \) and diameter \( a \). The enhancement is seen to be given is given by the length of the needle \( L \).

Another interesting case is a flat disc with a thickness of only a few atomic layers. The speedup is only due to the number of atomic layers and independent of the size of the disc. There is no enhancement for a two-dimensional disc. Thus the enhancement is seen to arise only for a three-dimensional source. No additional enhancement is found for one or two dimensional sources.

This can also be seen by explicit examination of the summation (3.1) for the one and two dimensional cases. In one dimension the sum vanishes for large values of \( |r_\mu - r_\nu| \) and the periodic variation of the integrand further prevents any enhancement at large distances. The summation in two dimensions does not vanish for large values of \( |r_\mu - r_\nu| \) because the \( |r_\mu - r_\nu| \) denominator is canceled by a similar factor in the two-dimensional integrand. However the periodic variation of the integrand remains and still prevents any enhancement at large distances.

The enhancement can be seen as resulting from competing factors of enhancement by a factor \( N \) in the exact forward direction and reduction in the angular width of the forward peak.

The forward enhancement can be expressed as a factor proportional to \( R^d \) where \( R \) is the linear size of the source and \( d \) is the dimension. If the width scales like \( 1/R \), we find that there is an overall enhancement by a factor \( R \) or \( N^{(1/3)} \) for a three dimensional spherical source and no similar enhancement for linear or planar sources.

### IV. SOME FURTHER QUESTIONS

The question has been asked whether the quantum-optical analysis in the present paper [1] adds any new knowledge in a different language to what has been long known in X-ray physics. In this example we note that X-ray physics is defined by the region in which \( \vec{k}_0 \cdot \vec{D} \geq 1 \). The region where \( \vec{k}_0 \cdot \vec{D} \approx 0 \) gives nearly isotropic Dicke superradiance and is outside the domain of X-ray physics.

Another region explored in this paper [1] does not exist in X-ray physics; namely where \( \vec{k}_0 \cdot \vec{D} \) is small but finite and \( nk_0 \cdot \vec{D} \) reaches unity for a finite value of \( n \ll N \). There may be interesting physics in this domain which goes beyond simple isotropic Dicke superradiance and does not exist in the X-ray region. If so, it should be stated in a way that pinpoints the new knowledge and can be understood by all physicists and not only by a group which speaks in its own jargon.

Some of the different aspects of coherence from ensembles with a single excitation can be summarized as follows:

1. In forward scattering the phase difference between the excitations of different nuclei is exactly canceled by the phase produced by the path difference in forward propagation. This is independent of the structure and the order of the sample.

2. In an ordered crystal lattice, the excitation phases can be exactly compensated at Bragg angles to give constructive interference and an enhanced peak. However, at
other angles the scattering is still coherent and the interference is destructive, giving a smaller peak than would be obtained if the scattering were incoherent.

3. In a completely disordered crystal, the coherence of the incident X-ray excitation is completely lost, since the outgoing phases are completely random. Therefore the 90° elastic peak will be stronger in a disordered sample that in an ordered sample where there is destructive interference.

4. In samples where the distance between nearest neighbors is much smaller than the photon wave length a coherent isotropic emission can occur. How much of this emission is isotropic and how much is forward depends on the conditions and parameters of the experiment. This includes the region explored [1] where \( \mathbf{k}_0 \cdot \mathbf{D} \) is small but finite and \( n\mathbf{k}_0 \cdot \mathbf{D} \) reaches unity for a finite value of \( n \ll N \). This can occur in quantum optics experiments but not in nuclear X-rays.

There seems to be a crucial qualitative difference between the directed spontaneous emission of optical and X-ray photons from an extended ensemble of \( N \) atoms. This has been shown in the discussion following eq.(2.2) The first paragraph of ref.( [1]) raises two questions whose answers may be very different in different areas.

1. Why don’t we know which atom is excited? Simple ignorance will not give coherence. There must be a quantum mechanical reason which prevents the production mechanism and the emission of the emitted photon from leaving a trail identifying the active atom [9]. This reason can be very different in different areas of physics. A photon carries momentum and the momentum transfer identifies the active atom. The momentum transfer when a free nucleus emits or absorbs a nuclear resonance photon is enormous; the recoil energy transfer is many orders of magnitude larger than the natural line width. The momentum transfer can be absorbed without a trace by a nucleus only if it is localized to an extent where the Heisenberg momentum uncertainty is as great as the momentum transfer. The physics of atomic optics is clearly very different.

2. Why is directional correlation counterintuitive? Atomic physicists may know that a Dicke superradiant state will always emit a photon in \( 4\pi \) sr. if the atoms are all in a spatial region which is very much smaller than the photon wave length. But X-ray crystallographers know that the photons in coherent X-ray scattering are highly correlated and that there is coherent and incoherent scattering.

V. PREVIOUS HISTORY FOR X-RAY SCATTERING

This issue has been already considered in detail on the pages of Physical Review Letters [2–5] for the case of resonant nuclear scattering. The correct answer is certainly not counterintuitive.

The basic physics needed to understand directed spontaneous emission from an extended ensemble of \( N \) atoms goes back much further even than Dicke’s seminal paper on superradiance [6] or Ott’s 1935 X-ray paper [10], Lamb’s 1949 paper [11] and Van Hove’s 1954 paper [12]. It is a transition from an initial state of a system with many atoms in which one atom
is excited and we don’t know which one and which has no photon to a final state in which all the atoms are in their ground state with no excitation and there is one emitted photon with wave vector $\vec{k}_0$.

This transition is described to lowest order in a perturbation series in the fine structure constant $\alpha$ by the Fermi “golden rule” as proportional to the square of the transition matrix element. There is no point to going back to the elementary quantum mechanics needed to derive the Fermi golden rule. The only problem is to define properly the initial and final states and express the experimental observation in terms of sums of squares of the relevant matrix elements.

The relation between Lamb’s treatment [11] of neutron capture in crystals and Ott’s X-ray treatment [10] was first pointed out by Kaufman [13,14] and reported in detail in a history of these developments [15]. A general formulation including these and other processes of momentum transfer to bound systems is given in a quantum mechanics book [16] which shows the relation of the dual wave-particle descriptions of similar phenomena.

That scattered photons go in the direction of the absorbed photon is classical wave optics and Huygens’ principle, which states that radiation scattered from an assembly of scatterers or emitted coherently from an assembly of sources are described by combining the waves of all the scatterers or sources with the proper phases. That the transmission of light through a medium is directionally correlated with the direction of the incident beam is not counterintuitive at all.

The N-atom state in a crystal prepared by the absorption of a single photon has been well known in nuclear resonance physics and called a “nuclear exciton” by Hannon and Trammell [17]. However its application is always in cases where the distance between nearest neighbors is always at least of the order of the photon wave length. A crystal never has a large number of nuclei confined to a region much smaller than the wave length.

VI. SOME FURTHER DETAILS

Angular Distributions - Basic physics often missed

1. Mössbauer scattering from single nucleus essentially isotropic. Variations involve only lowest spherical harmonics.

2. Angular distribution of coherent forward scattering sharply forward peaked.

3. Forward peaking from constructive interference between amplitudes from different nuclei.

4. Combination of constructive and destructive interference at other angles

- Crucial to understanding of all experiments and not generally understood.
- Interference at angles far from forward; e.g. 90° generally destructive.
- Exact forward amplitude has zero solid angle. Experiments measure integral of intensity over finite width of forward peak.
- Forward peak intensity and width both vary with thickness of sample, and both must be considered.
ACKNOWLEDGMENTS

Some of these issues were clarified in discussions at the April, 2000 workshop at HASY-LAB [19]. It is a pleasure to thank E. E. Alp, A.Q.R. Baron, U. van Buerck, R. Coussement H. Franz, E. Gerdau, W. Potzel, R. Roehlsberger, W. Sturhahn and T. Toellner for helpful discussions and comments.

This work was supported in part by the U.S. Department of Energy, Basic Energy Sciences, Office of Science, under Contract No. W-31-109-Eng-38.

REFERENCES

[1] Marlan O. Scully et al Phys. Rev. Lett. 96, 01051 (2006)
[2] J. P. Hannon and G. T. Trammell, Phys. Rev. Lett. 61, 653 (1988)
[3] Harry J. Lipkin, Phys. Rev. Lett. 58, 1176 (1987), 60, 2227 (1988) and 61, 654 (1988)
[4] R. Rüffer, E. Gerdau, R. Hollatz and J. P. Hannon Phys. Rev. Lett. 58, 2359 (1987)
[5] U. van Bürck et al, Phys. Rev. Lett. 59, 355 (1987)
[6] R. H. Dicke, Phys. Rev. 93, 99 (1954)
[7] C. T. Lee, Phys. Rev. A13, 1657 (1976)
[8] Maurice Goldhaber, private communication
[9] Harry J. Lipkin, hep-ph/9907551 Physics Letters B 477 (2000) 195
[10] H. Ott, Ann. Physik, 23, 169 (1935)
[11] W. E. Lamb, Jr. Phys. Rev. 55, 190 (1939)
[12] L. Van Hove, Phys. Rev. 95, 249 (1954)
[13] B. Kaufman, private communication to W. E. Lamb, unpublished (1939)
[14] B. Kaufman and Harry J. Lipkin, Ann. Phys. 18, 294 (1962)
[15] Harry J. Lipkin, Hyperfine Interactions 72 3 (1992)
[16] Harry J. Lipkin, Quantum Mechanics, North-Holland Publishing Co. Amsterdam (1973) pp.33-110
[17] J. P. Hannon and G. T. Trammell, in Resonant Anomalous X-Ray Scattering, Proceedings of the ICAS Conference, August 17-21, 1992 Malente/Hamburg, Germany, Edited by G. Materlik, C. J. Sparks and K. Fischer, Elsevier Publishers, Amsterdam (1992), and Hyperfine Interactions 123 (1999) 12
[18] Wolfgang Sturhahn, private communication
[19] Proceedings of the Workshop on ”Nuclear Resonance Scattering at the Free-Electron-Laser Facility (X-FEL)” April 27/28, 2000, HASYLAB, Hamburg; eds. H. Franz and U. van Bürck. available as report ”TESLA-FEL 2000-19” from DESY
[20] For a review of nuclear resonance excitation by synchrotron sources, see E. Gerdau, Hyperfine Interactions, vol. 123 (1999) and the papers therein
[21] G. T. Trammell, in “Chemical Effects of Nuclear Transformations”, International Atomic Energy Agency, Vienna (1961) p. 75