N Dependence of Coherent Radiation from Crystals

Harry J. Lipkin
Argonne National Laboratory, Argonne, Illinois 60439, and
Department of Nuclear Physics, Weizmann Institute of Science, 76 100 Rehovot, Israel
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The total intensity averaged over all angles of short-wavelength radiation either scattered or spontaneously emitted nearly isotropically from each of a system of N radiators or scatterers is shown to vary linearly with N for large N. Although coherent effects can give intensities proportional to N^2 in certain directions (e.g., Bragg angles), this enhancement is mainly at the expense of radiation otherwise emitted in other directions and does not appear in the overall angular distribution.

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Recently there have been suggestions that coherent radiation from crystals might have an intensity which is proportional to the square of the number N of atoms in the crystal. Effects of this type have been suggested in connection with neutrino scattering[1] and also with superradiance in nuclear γ rays.[2] Recently contradictory arguments[3-6] have been given showing that neutrino scattering is proportional to N rather than N^2. Similar arguments may arise in discussions of possible detectors for other weakly interacting particles (e.g., axions or photinos). It is therefore of interest to provide a simple general argument which can be used to clarify the present situation and be a useful reference for future cases. This Letter shows that the total intensity of radiation with a wavelength much shorter than the interatomic spacings in a crystal, when integrated over all angles, is proportional to N, even though there may be peaks in the angular distribution that are proportional to N^2, for any radiation or scattering process satisfying the following two conditions:

(1) The impulse approximation is valid.

(2) The angular distribution of the elementary process on a single atom has no peaks which are larger than the average intensity by factors of order N.

Any coherent effect mainly rearranges the angular distribution of the radiation; it does not change the overall intensity by large factors.

Consider a system of N radiation sources, each radiating the same amplitude and with a phase that is adjusted to give optimum coherence. The total amplitude observed at a point $\mathbf{R}$ at a large distance from the source is

$$A=\sum_i a e^{i\phi_i} e^{-i\mathbf{k}\cdot\mathbf{r}_i}, \tag{1}$$

where a is the amplitude of radiation from any single source, ϕ_i is the phase of the ith source, which can be adjusted for optimum coherence, $\mathbf{k}$ is the wave vector of the radiation taken in the direction of $\mathbf{R}$. The intensity of the radiation is then

$$|A|^2=\sum_i\sum_j |a|^2 e^{i(\phi_i-\phi_j)} e^{-i\mathbf{k}\cdot(\mathbf{r}_i-\mathbf{r}_j)}. \tag{2}$$

This result also applies to the case of scattering by an assembly of scatterers in the impulse approximation where the scattering amplitude is given by an expression having the form (1) with a being the magnitude of the scattering amplitude from a single scatterer and

$$\phi_i=\mathbf{k}_{\text{in}}\cdot\mathbf{r}_i, \tag{3a}$$

where $\mathbf{k}_{\text{in}}$ is the wave vector of the incoming wave.

Note that the choice

$$\phi_i=\mathbf{k}\cdot\mathbf{r}_i+2n\pi \tag{3b}$$

gives an amplitude proportional to N and an intensity proportional to N^2. This is the case in conventional Bragg scattering, where the relation (3) is just the Bragg condition.

However, this coherence occurs only in the direction of $\mathbf{R}$ or the particular wave vector $\mathbf{k}$. If we assume that the individual sources radiate isotropically, then the total radiation integrated over all angles is given by

$$\int |A|^2 d\Omega=2\pi\sum_i\sum_j |a|^2 e^{i(\phi_i-\phi_j)}\int e^{-ik|r_i-r_j|\cos\theta}d(\cos\theta), \tag{4a}$$

$$\int |A|^2 d\Omega=4\pi\sum_i\sum_j |a|^2 e^{i(\phi_i-\phi_j)}\sin(k|r_i-r_j|)/k|r_i-r_j|, \tag{4b}$$

$$\int |A|^2 d\Omega=2\pi\sum_i\sum_j |a|^2[\sin(k|r_i-r_j|+\phi_i-\phi_j)+\sin(k|r_i-r_j|-\phi_i+\phi_j)]/k|r_i-r_j|. \tag{4c}$$

This expression is seen to be proportional to N, not to N^2, if $k|r_i-r_j|\gg 1$. The factor $k|r_i-r_j|$ in the denominator immediately suppresses all contributions from pairs separated by large distances and ensures than there is no N dependence greater than linear for large N. Furthermore, the appearance of the sine rather than the cosine makes it impossible to achieve constructive interference for a set of sources equally spaced on a line. If the distance between nearest neighbors along this line is d and the overall phase difference between the radiation from nearest neighbors is ϕ, then the total radiation from this line for a large crystal is

$$\int |A|^2 d\Omega = 2\pi N_l |a|^2 \left[1+2\sum_n \frac{\sin(n\phi)}{nkd}\right], \tag{5a}$$

where N_l is the number of atoms in this line. The sum is clearly of order $1/kd$, giving a contribution of order N_l if kd is of order unity. Note that if $\sin\phi=1$, then $\sin(2\phi)=0$, $\sin(3\phi)=-1$, and the sum will be less than $1/kd$. If $\sin\phi$ is small and positive, the sum approaches $\pi/2kd$. A dependence on N^2 is obtained only if $k|r_i-r_j| \ll 1$ for all pairs in the sample.

This argument is easily generalized to the case of a nonisotropic angular distribution, if there are no very large peaks. Since the intensity of radiation at any given angle is positive definite, Eqs. (4)-(5a) can be generalized to become upper bounds for the case of an arbitrary angular distribution by the replacement of a by the amplitude at the peak of the angular distribution, denoted by a_p:

$$\int |A|^2 d\Omega \le 2\pi N_l |a_p|^2 \left[1+2\sum_n \frac{\sin(n\phi)}{nkd}\right]. \tag{5b}$$

Thus as long as the peak intensity is greater than the average intensity only by a factor of order unity and not a factor of order N, the conclusions are unchanged.

This argument applies also to the case of superradiance. Dicke[2] has shown how the rate of a spontaneous radiative transition can be enhanced by a collective effect, in which any one of N atoms may be excited and the many-body wave function is a coherent superposition of states in which a different atom is excited. The transition probability is then enhanced by a factor N and the lifetime of the excited level reduced by a factor N from the lifetime of a single atom. However, this effect depends upon a coherent excitation and a coherent radiation in which all contributions from different atoms are in the same phase. The above treatment shows that this is possible as long as the wavelength of the radiation is large compared to the size of the radiator; i.e., if $k|r_i-r_j| \ll 1$ for all pairs in the sample.

For nuclear γ transitions this effect no longer occurs, because the interatomic distances are no longer negligible in comparison with the wavelength of the radiation. The relative phases of the contributions of radiation from different nuclei depend upon the wavelength of the radiation, the distance between atoms and the angle of the radiation, in a manner similar to the case of x-ray scattering by crystals. Although it is possible to adjust the phases of the radiation to interfere constructively in certain directions in a crystal (e.g., Bragg directions), this enhancement occurs only in a very small solid angle around the Bragg direction, and is mainly at the expense of radiation in other directions. The result of the coherence is primarily to change the angular distribution of the radiation, but not its overall intensity, thus giving little or no change in the lifetime of the excited state.

This argument is easily stated explicitly for the case of superradiance. Consider a system of N atoms or nuclei, in which one is in an excited state and all the rest are in the ground state. The wave function for the superradiant state has an equal probability for any one of the N atoms or nuclei to be in the excited state and is a coherent superposition of such singly excited systems:

$$|S\rangle = (1/\sqrt{N})\sum_i e^{i\phi_i}|g,e_i\rangle, \tag{6}$$

where $|g,e_i\rangle$ denotes the state in which the ith atom is in its excited state and all the remaining atoms are in their ground state and ϕ_i is an arbitrary phase factor chosen to maximize coherence effects. Let J denote the operator describing the radiative transition to the ground state of the entire system, denoted by $|G\rangle$, with the emission of a photon of wave vector $\mathbf{k}$. Then the transition matrix element is given by

$$\langle G|M|S\rangle = (1/\sqrt{N})\sum_i \langle G|J|g,e_i\rangle e^{i\phi_i}e^{-i\mathbf{k}\cdot\mathbf{r}_i}. \tag{7}$$

This has exactly the same form as the relation (1) with

$$a = (1/\sqrt{N})\langle G|J|g,e_i\rangle. \tag{8}$$

The same analysis, Eqs. (2)-(5), then shows that coherence effects can only change the angular distribution of the radiation but cannot give any enhancement in the intensity which goes like a power of N. A judicious choice of phases may give some enhancement from constructive interference with nearest neighbors, but there can be no large effect from large distances.

This shows that for a weak process like neutrino scattering, where the scattering probability is small even for N incoherent scatterers, the total cross section can only be increased by perhaps 1 order of magnitude by coherence effects involving nearest neighbors, but cannot be increased by an additional power of N, unless the wavelengths are much longer than interparticle spacings.

For resonance scattering like Mössbauer scattering, where the individual cross sections are at the unitarity limit, the possibility of coherent excitation of a "superradiant" state can broaden the line to give an enhanced decay rate again by only 1 order of magnitude from coherence effects involving nearest neighbors, but cannot be increased by an additional power of N, unless the wavelengths are much longer than interparticle spacings.

Note that only spontaneous emission is being considered here, not stimulated emission. Spontaneous emission is indeed enhanced by the coherent effect called superradiance with an enhancement factor proportional to N when the wavelength of the radiation is long enough to keep a constant phase for the radiation from all atoms. This enhancement no longer occurs when the wavelength of the radiation is shorter than the average distance between atoms.

This argument holds for any process where the impulse approximation is valid and there are no peaks in the angular distribution of the amplitude from an individual radiator with intensities greater than the average intensity by factors of order N. The impulse approximation (1) which gives the amplitude from N scatterers as the sum of the amplitudes from individual scatterers is automatically valid for the lowest-order contribution to any process described by perturbation theory. It thus holds for the scattering of photons and leptons by atoms and nucleons, including the particular case of neutrino scattering. Angular distributions are generally smooth, described by a spherical harmonic characteristic of the particular relevant multipole. One exception to the smooth angular distribution is the case of stimulated emission, if the number of photons already present with wave vectors in a Bragg direction is of order N. In this case all of the radiation from a single scatterer is already concentrated in the Bragg peak, and the enhancement of the intensity of the peak by a factor of N^2 applies to the entire distribution.

A similar analysis can be applied to multiple-scattering processes which violate the impulse approximation. Consider, for example, a double-scattering process in which radiation emitted from one source α is rescattered by another source i. For this case Eq. (1) is replaced by

$$A=\sum_i a_i' e^{-i\mathbf{k}\cdot\mathbf{r}_i}, \tag{9a}$$

where a_i' is the amplitude of the rescattered radiation from the source i,

$$a_i'=\sum_\alpha a e^{i\phi_\alpha} b e^{-ik|r_\alpha-r_i|}/|r_\alpha-r_i|, \tag{9b}$$

and b is the scattering amplitude for rescattering at the source i. The presence of the double sum suggests at first that with proper phases to give constructive interference everywhere an amplitude proportional to N^2 can be obtained, to give an intensity proportional to N^4. However, such total constructive interference does not occur, as shown by my noting that at large distances the sum (9b) can be estimated by replacing the sum by an integral

$$a_i'\approx a\rho b\int d\Omega\int_0^{R(\theta,\phi)} dr\, r^2\frac{e^{-ikr}}{r}=\frac{a\rho b}{k^2}\int d\Omega\{[ikR(\theta,\phi)+1]e^{-ikR(\theta,\phi)}-1\}, \tag{9c}$$

where ρ is the density of scatterers and $R(\theta,\phi)$ are the coordinates of the boundary of the scatterer in polar coordinates with the origin at r_i. This result (9c) is of order $N^{1/3}$ rather than N. Although this might seem to introduce an additional $N^{2/3}$ dependence in the radiation intensity, the N-dependent factor $R(\theta,\phi)e^{-ikR(\theta,\phi)}$ has many oscillations as a function of the angles and the integral (9c) over the angles is of order unity.

I now show that no strong additional N dependence has been lost by the smearing of the discrete sum by an integral. I consider two cases of discrete sets of terms which have phases that give constructive interference.

The amplitude (9a) has the same form as Eq. (1), with the single source amplitude replaced by a more complicated factor (9b). Thus by analogy with the derivation of Eq. (4a) I assume that the individual sources radiate isotropically and the total radiation integrated over all angles is given by

$$\int |A|^2 d\Omega=2\pi\sum_i\sum_j |a_i'a_j'^*|e^{i(\phi_i-\phi_j)}\int e^{-ik|r_i-r_j|\cos\theta}d(\cos\theta), \tag{10a}$$

where

$$a_i'a_j'^*=\sum_\alpha\sum_\beta |a|^2 e^{i(\phi_\alpha-\phi_\beta)}|b|^2\frac{e^{-ik|r_\alpha-r_i|}}{|r_\alpha-r_i|}\frac{e^{ik|r_\beta-r_j|}}{|r_\beta-r_j|}. \tag{10b}$$

The intensity (10a) has the same form as Eq. (4a), with the single source amplitude replaced by a more complicated factor (10b). The integral in Eq. (10) thus has the same N dependence as Eqs. (4). The only possibility for additional N dependence must come from the summations in Eqs. (9b) and (10b).

The terms with $\alpha=\beta$ and $i=j$ are indeed coherent and interfere constructively to give a contribution

$$(a_i'a_i'^*)_{\alpha=\beta}=\sum_\alpha\frac{|a|^2|b|^2}{|r_\alpha-r_i|^2}=|a|^2\rho|b|^2T=|a|^2KP_1, \tag{11a}$$

where we have replaced the sum by an integral as above, T is the thickness of the crystal, K is a numerical factor of order unity, and

$$P_1=\rho|b|^2T/K \tag{11b}$$

is the probability of single scattering for radiation incident on the crystal. The additional $N^{1/3}$ dependence introduced by the factor T is thus seen to be just the well-known extra N dependence of the ordinary incoherent double scattering. For cases of small single scattering, $P_1\ll 1$, this contribution (11) is negligible in comparison with the single scattering contribution (4).

We now consider the peculiar case where there happens to exist a line in the crystal whose scatterers satisfy the condition

$$e^{i\phi_\alpha}e^{-ik|r_\alpha - r_i|} = 1. \tag{12a}$$

In this case the contributions along the line are indeed coherent and interfere constructively to give a contribution

$$\begin{aligned} a_i' a_j'^* &= \sum_\alpha \sum_\beta \frac{|a|^2 |b|^2}{|r_\alpha - r_i|\,|r_\beta - r_j|} \\ &= \frac{|a|^2|b|^2}{d^2}\left|\sum_{n=1}^{N_l}\frac{1}{n}\right|^2 \\ &= |a|^2 \frac{KP_1}{\rho T d^2}\left|\sum_{n=1}^{N_l}\frac{1}{n}\right|^2. \end{aligned} \tag{12b}$$

This gives an additional weak N dependence like $\ln^2(N)$ but multiplied by a tiny factor of order $P_1 N^{-1/3}$.

We thus see that as long as $P_1 \ll 1$ there is no possibility of gaining additional scattered intensity by double scattering. Thus argument is now easily extended by induction to the case of multiple scattering. The case where $P_1 \approx 1$ requires treatment by other methods, e.g., multiple-scattering theory or definition of an index of refraction. This case is not relevant to detection of weakly interacting particles by their interactions with crystals.

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