COHERENT EFFECTS IN TRANSITIONS BETWEEN STATES CONTAINING SEVERAL NUCLEAR EXCITONS

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When ten separated identical nuclei are all excited to the same first excited state they decay independently with the natural line width and lifetime. But a standard Wigner-Weisskopf treatment of the initial state as a pure quantum state says that it decays exponentially to a state with nine excited nuclei with ten times the natural width and ten times the exponent for a single decay. The resolution of this paradox would have amused and interested Misha Marinov. We explore the possibility that a synchrotron radiation beam may be sufficiently intense to excite more than a single Mössbauer nuclear resonance in the same sample in the same pulse; i.e. to excite a coherent state of several nuclear excitons. We describe multi-exciton states using a quasispin description of the states of the nucleus. We then consider the decay and production of a two-exciton state and find that the two-exciton decay is twice as fast as a single exciton decay and therefore that the width for exciting a second exciton when one exciton is already present is twice the width for exciting a single exciton. The implications of this factor of two are examined and the treatment is then generalized to the three-exciton and multi-exciton cases. The conclusion that the width for the excitation of an additional nuclear exciton is enhanced by a factor $n+1$ when n excitons are already present is interesting and deserves further investigation.

Contents

1 Introduction. A paradox in the quantum mechanics of multiply-excited systems

1.1 Simple coherence effects in excitation transition probabilities

The possibility that synchrotron radiation sources[1] may be available with sufficient intensity to allow the excitation of several nuclei in a sample[2] by a single pulse leads to investigation of multiple excitations in solids. New coherence phenomena arise which first lead to an apparent paradox. The essentials can be seen in the simple example of a system with two nuclei, denoted by A and B which can be excited from the ground state by a synchrotron radiation pulse. Let $|0\rangle$, $|A\rangle$, $|B\rangle$ and $|AB\rangle$ denote respectively the states where neither nucleus is excited, nucleus A is excited, nucleus B is excited and where both nuclei are excited. Let the transition operator denoted by T describing the excitation of a nucleus by a synchrotron radiation pulse be equal for the two nuclei and independent of whether the other nucleus is excited. Then the matrix elements of the transition operator satisfy the relation

$$\langle A| T |0\rangle = \langle B| T |0\rangle = \langle AB| T |A\rangle = \langle AB| T |B\rangle \equiv T . \tag{1}$$

A state with a single excitation produced when a pulse of synchrotron radiation strikes both nuclei equally is described as

$$|1\rangle \equiv \frac{1}{\sqrt{2}} \cdot \{|A\rangle + e^{i\phi} \cdot |B\rangle\}; \qquad |\langle AB| T |1\rangle|^2 = |T|^2 \cdot \{1 + \cos\phi\} , \tag{2}$$

where ϕ denotes the relative phase of the two excitations which depends upon the production mechanism.

The transition probability $|\langle AB| T |1\rangle|^2$ for producing a double excitation from the singly excited state $|1\rangle$ depends upon the relative phase ϕ between the excitations of nucleus A and nucleus B. For the case where $\phi = 0$ we see that the transition probability is $2|T|^2$ or double the transition probability for a single excitation. This enhancement factor of two arises when the relative phase ϕ has just the value to produce constructive interference between the two single excitations.

We shall see in detail below that the condition for this constructive interference is exactly produced in realistic systems with many nuclei and a single excitation in the case where the first excitation is a nuclear exciton[1,3-5] produced by the same synchrotron radiation pulse that produces the second excitation. However, in all other cases the relative phase ϕ is random and the there is no enhancement.

We now extend this to the case of three nuclei with the third nucleon denoted by C. We define the coherent double excitation state where any pair of the three nuclei is excited, and the phase produces constructive interference for the third excitation,

$$|2_{coh}\rangle \equiv \frac{1}{\sqrt{3}} \cdot \{|AB\rangle + |BC\rangle + |CA\rangle\}; \quad |\langle ABC| T |2_{coh}\rangle|^2 = 3 \cdot |T|^2 . \quad (3)$$

Extending this treatment to the case of an initial state where (n-1) coherent excitations are already present; e.g. a state containing (n-1) nuclear excitons, leads to the result that the transition probability for producing a state with n-excitations from this state is enhanced by a factor n. This suggests a picture with a collective excitation which behaves like a boson, where the creation of such a boson is enhanced by the same factor as the stimulated emission of any boson when other bosons are already present.

In the remainder of this paper we consider the production and decay of nuclear excitations in detail. The result will show that this "boson-enhancement" picture has a general validity for the case of multiple production of nuclear excitons, and that it is possible to define operators which create and annihilate nuclear excitons which obey boson commutation rules to a very good approximation.

1.2 Simple coherence effects in decays of multiply excited states

The decays of the states $|AB\rangle$ and $|ABC\rangle$ with double and triple excitations respectively are simply described by the Fermi Golden Rule of time-dependent perturbation theory as proportional to the square of the transition matrix element (1) of the operator T. This gives the well-known result that each excited nucleus decays independently with the standard exponential time dependence $e^{-\lambda t}$ where λ is the decay constant proportional to $|T|^2$ given by the Fermi Golden rule for a single decay.

However, we can also describe the decay as a cascade of several decays using a basis of the coherent states,

$$|ABC\rangle \rightarrow |2^{\gamma}_{coh}\rangle \rightarrow |1\rangle \rightarrow |0\rangle \; , \quad (4)$$

where the coherent doubly-excited state of three nuclei is now redefined to explicitly include the photon emitted by the third nucleus,

$$|2^{\gamma}_{coh}\rangle \equiv \frac{1}{\sqrt{3}} \cdot \{|AB\gamma_C\rangle + |BC\gamma_A\rangle + |CA\gamma_B\rangle\}; \quad |\langle ABC| T |2^{\gamma}_{coh}\rangle|^2 = 3 \cdot |T|^2 , \quad (5)$$

where γ_A, γ_B and γ_C denote that nuclei A, B and C respectively are now in their ground states but a photon has been emitted in their transition from the excited state.

We immediately find that the first decay of the cascade, $|ABC\rangle \rightarrow |2^{\gamma}_{coh}\rangle$ is described by the squared transition matrix element (3), $|\langle ABC|\,T\,|2^{\gamma}_{coh}\rangle|^2 = 3 \cdot |T|^2$, and that the time dependence of this decay is $e^{-3\lambda t}$. If the photons emitted in the three terms in the state $|2^{\gamma}_{coh}\rangle$ are identical, then these three terms can contribute coherently to the subsequent decay as in the decay of a nuclear exciton and its decay can be speeded up in the standard nuclear exciton manner.[5,6] This does not occur in a normal decay without exciton correlations and the decays of the three terms are incoherent. We therefore assume here that the subsequent decays are incoherent in order to examine the additional speedup resulting from the simultaneous presence of several excitations. The singly-excited state $|1\rangle$ in the cascade (5) is therefore a complicated state with terms containing different photons. Each term decays individually with the normal time dependence $e^{-\lambda t}$.

The second decay of the cascade is described by the incoherent decays of each doubly-excited term into a coherent singly-excited state , with a squared transition matrix element given by Eq. (2) with $\phi = 0$. $|\langle AB|\,T\,|1_{coh}\rangle|^2 = 2 \cdot |T|^2$. The time dependence of this decay is $e^{-2\lambda t}$.

At first this appears inconsistent with the well-known result that all decays have the time dependence $e^{-\lambda t}$. However, we shall now see that the two descriptions are completely equivalent.

1.3 The cascade decay of a multi-excited state

The time behavior of the radiation from the decay of a multi-excited state where each transition has a different lifetime would be expected to be a linear combination of several exponentials, one corresponding to each lifetime. However, for the case where the decay width of the $n \rightarrow n-1$ transition is proportional to n the expression simplifies in a surprising manner and the observed radiation is just a single exponential, exactly as if there were no cascade and there were n independent nuclei decaying. We now examine in detail how this surprising result arises.

Consider the cascade decay (4) of a state $|ABC\rangle$ containing three nuclear excitations. Let $P_{3X}(t)$, $P_{2X}(t)$ and $P_{1X}(t)$ denote the probabilities respectively for the system to be in the triply-excited, doubly-excited and singly-excited states. The decay rates from these states are respectively 3λ, 2λ and λ and the probabilities $P_{3X}(t)$, $P_{2X}(t)$ and $P_{1X}(t)$ satisfy the differential equations and initial conditions:

$$\frac{dP_{3X}}{dt} = -3\lambda P_{3X}\,; \quad P_{3X}(0) = 1\,, \quad P_{3X}(t) = e^{-3\lambda t}\,, \tag{6}$$

$$\frac{dP_{2X}}{dt} = -2\lambda P_{2X} - \frac{dP_{3X}}{dt}; \quad P_{2X}(0) = 0, \tag{7}$$

$$\frac{dP_{1X}}{dt} = -\lambda P_{1X} - \frac{dP_{2X}}{dt} - \frac{dP_{3X}}{dt}; \quad P_{1X}(0) = 0, \tag{8}$$

where we have also given the trivial solution of the equation for $P_{3X}(t)$. These can be rewritten in a compact general form in terms of the number N of initial nuclear excitations, where $N = 3$ in this case, but the results are easily extended to hold for all values of N.

$$\frac{dP_{kX}}{dt} = -k\lambda P_{kX} - \sum_{j=k+1}^{N} \frac{dP_{jX}}{dt}; \quad P_{kX}(0) = \delta_{kN}. \tag{9}$$

The total radiation emitted from all nuclei is given by

$$R(t) = 3\lambda P_{3X}(t) + 2\lambda P_{2X}(t) + \lambda P_{1X}(t) = \sum_{k=1}^{N} k\lambda P_{kX}(t); \quad R(0) = N\lambda. \tag{10}$$

Substituting Eqs. (6-8) gives the differential equation for $R(t)$ which is easily solved,

$$\frac{dR}{dt} = -\lambda R; \quad R(t) = Ne^{-\lambda t}. \tag{11}$$

Thus, although the individual probabilities $P_{kX}(t)$ contain several exponentials, the total radiation decays exponentially with the single exponent of the decay rate for a single nucleus. All other exponential terms drop out of the total radiation.

To show that this treatment applies to the cascade decay of a general N-fold excited state we note that Eqs. (9) and (10) will hold for all values of N if we generalize the assumption that the decay rate for the transition $kX \to (k-1)X$ is $k\lambda$ to hold for all values of k. Eq. (9) can be rearranged to the form

$$\sum_{j\geq k}^{N} \frac{dP_{jX}}{dt} = -k\lambda P_{kX}; \quad P_{kX}(0) = \delta_{kN}. \tag{12}$$

This simply states that the change in the total probability for the system to be in a state of k or more excitations is given by the decay of the k-fold excited state with the initial condition that all N nuclei are excited at $t = 0$. The general result that Eq. (11) holds for all values of N follows from summing this over the index k, rearranging and evaluating the sums,

$$-R = -\sum_{k=1}^{N} k\lambda P_{kX} = \sum_{k=1}^{N}\sum_{j\geq k}^{N} \frac{dP_{jX}}{dt} = \sum_{j=1}^{N}\sum_{k=1}^{j} \frac{dP_{jX}}{dt} = \sum_{j=1}^{N} j\frac{dP_{jX}}{dt} = \frac{1}{\lambda}\cdot\frac{dR}{dt}. \tag{13}$$

We have therefore resolved the apparent contradiction about different exponentials. The radiation for the cascade decay of a state with k excitations via coherent states is therefore the same as for k independent decays with the normal lifetime. Thus the radiation does not show the presence of a cascade decay with a speeded-up first decay.

However, one apparently new phenomenon remains. The natural line width for the excitation of the state with k excitations from a state with $k-1$ *coherent* excitations is k times the normal line width. For ordinary decays of systems with n excited nuclei this analysis merely confirms what we already know. That coherent states like $|2_{coh}\rangle$ and $|1_{coh}\rangle$ are produced in the decays of states with multiple excitations is only of academic interest since such coherence is never observed in real experiments. The one case where the coherence is of interest is in the production and decay of nuclear excitons where the coherent state is actually created experimentally. In this case the enhancement of the width for excitation from a state which contains coherent excitations can be very important for excitation from a broad-band synchrotron radiation source with an energy band width much larger than the natural line width of the nuclear resonance.

We now show in detail how these same results hold for systems of coherently excited nuclear excitons and their cascade decays.

2 Simple properties of multiphoton excitation

2.1 What exactly is a nuclear exciton?

Coherent scattering and Bragg scattering of Mössbauer resonance radiation are now well understood and well established experimentally. [1,3–5,7–10] But this is all at the one-photon level, where the coherent excitation of many nuclei has been described as a nuclear exciton.[3–5] Only one nucleus is excited, but it can be one of many, and the amplitudes for the excitations of different nuclei add coherently to produce a forward peak in the outgoing radiation, a speedup of the nuclear lifetime, and peaks at Bragg angles.[7,8]

Synchrotron radiation beams of sufficient intensity may be available to enable the excitation of several nuclei simultaneously in the same synchrotron pulse. The question arises what happens when several nuclear excitons are present in a lattice as a result of excitation by several photons. How does the presence of several excitons affect the cross section for additional excitation by another photon? How does the presence of several excitons affect the decay lifetime? Do nuclear excitons behave like bosons? One can immediately see a number of interesting analogies which are not quite right: Dicke superradiance,[11] atomic excitons, spin waves, and gauge fields.

We are still a long way from coherent γ-ray light and γ-ray lasers. The energy difference between a state with one nuclear excitation and a state with two nuclear excitations is much too large to maintain any phase coherence between these two states. There is a factor of 10^{12} between the resonance energy of the order of tens of kilovolts and the natural line widths which are of order of tens of nanovolts. Thus the relative phase between a one-exciton and a two-exciton state reverses 10^{12} times during the lifetime of the nuclear excitation. There is little hope of maintaining any phase coherence over such a long period.

The presence of several excited nuclei which can decay coherently suggests Dicke superradiance.[11] But the distances between nearest neighbors are comparable to the wave length of the radiated photons. This makes the physics very different and more complicated than simple superradiance.[12]

One can follow the Dicke example and define a quasispin for each nucleus which is "up" when the nucleus is excited and "down" when the nucleus is in its ground state. The nuclear exciton then appears as a kind of "quasispin wave". The wave function differs from the Dicke case because the quasispins are located at definite lattice points and there is a definite phase between the amplitudes at different lattice points. But the wave does not move; there are no quasispin-quasispin interactions that can flip quasispins. The same is true for the atomic exciton analogy. The nuclear exciton is created and decays, but does not move in space.

One can also describe the relative phases of the quasispins at different points on the lattice as a kind of gauge field. Transformations of these phases resemble gauge transformations since they do not change the lattice energy, which depends only upon the number of excited nuclei; i.e. upon the z-component of the total quasispin. Quasispin rotations about the z-axis can be described as SU(1) gauge transformations under which the Hamiltonian of the lattice is "gauge invariant". But the relevant physics of emission and absorption of photons is not invariant under these gauge transformations. The coherence properties of excitation and decay are very sensitive to these phases.

So far no consistent framework has been developed to enable the inclusion of these various coherence effects in the calculation of the production and decay of these multi-exciton states.

2.2 Problems with a classical approach

We now try to pinpoint the differences between conventional treatments of the Mössbauer effect and what is needed to treat multiphoton processes and multiple excitations in the crystal.

The first treatment by Hamermesh *et al*[13] using a classical radiation field obtains all the correct results for one photon processes. The extensive development of this approach is now summarized in an excellent review by Hannon and Trammell.[4] One might expect that a classical description of the electromagnetic field which works for even one photon should work even better for many photons. However, the problem here is not in the classical description of the photon; it is the quantum effects not considered in the classical description of the lattice.

This problem is simply seen in the example of a photon incident on a thick crystal where so may photons have already passed through the first part of the crystal that all the Mössbauer nuclei in this part of the crystal have already been excited. The photon will pass through this part of the crystal without seeing any Mössbauer nuclei that can be excited and therefore without seeing anything like the dielectric constant and index of refraction that was seen by the first photon. Clearly the classical formulation does not work here. The nuclear excitation changes the properties of the crystal.

The classical picture describes the photon propagation through a medium in terms of parameters like a refractive index and a dielectric constant. To determine these parameters from theory requires a microscopic picture in which the medium consists of nuclei having very sharp resonances. The presence of different nuclei with different hyperfine splittings produces a complicated structure in the frequency spectrum of the dielectric constant and index of refraction.

All this is still well defined when there is only one photon and the treatments of Hamermesh *et al*[13] and all those following are valid. But nuclear excitations in the medium can produce a variation in the dielectric constant and index of refraction of a crystal as a function of the number of excited nuclei present which changes with time during the nuclear lifetime when photons are emitted and the number of nuclear excitations is changing. Perhaps there is a classical way to describe this with the properties of the medium being affected by the strength of the electromagnetic field in the sample.

This now suggests an anology with lasers and nonlinear optics. But here the Mössbauer situation introduces two important differences: (1) a very long lifetime; (2) a very short wave length of the same order as the distance between neighboring atoms. The parameters describing these lifetimes and distances differ by many orders of magnitude from the values relevant to conventional lasers and nonlinear optics. Thus extrapolations from this other physics are highly questionable. This is already seen in the lifetime speedup at the one photon level. There the speedup does not go as the number of excited atoms, N, but as $N^{(1/3)}$ because the wave length is not very much longer than the distances between atoms.[5,6]

We attempt here to avoid the complications of nonlinear optics by using the microscopic quantum picture of the lattice with nuclear excitons. This solves one problem, and may offer some insight. But it raises another.

The simple classical method with a medium described by bulk properties like a dielectric constant and index of refraction describes the passage of a radiation field through a medium and includes all the effects of multiple scattering which must be taken into account in the microscopic photon picture.

Multiple scattering of nuclear excitons is not simple. In a thick sample a nuclear exciton exists with amplitudes for nuclear excitation over a finite range in the sample. When it decays and progresses through the sample, it produces a new nuclear exciton further forward in the sample with a different spatial distribution. Meanwhile, if there were several nuclear excitations initially present there are still other nuclear excitations present with the initial spatial distribution.

The question of how to take all these complications into account is not clear. Perhaps it can be handled with classical nonlinear optics with the properties of the medium simply being functions of the electromagnetic field. Perhaps it is necessary to describe the medium itself by a quantum-mechanical wave function in which there are different probability amplitudes for each state of the medium with each having different dielectric constants and different indices of refraction.

Before attacking these problems we try to start in a simple way, with simple toy models of states having a small number of nuclear excitons in a thin sample with no multiple scattering. We consider the case where the number N of nuclei which can be excited to make a single nuclear exciton is large, while the number N_e of simultaneously excited nuclei; i.e. the number of nuclear excitons is small but finite, $N \gg N_e > 1$.

3 A quasispin description of nuclear excitons

3.1 The quasispin description of two-level systems

It is convenient to introduce a quasispin formalism using the $SU(2)$ Lie algebra approach to collective excitations in 2-level systems first developed for the nuclear shell model.[14] A similar formalism was also used by Dicke to treat superradiance.[11]

Consider a crystal containing N atoms, in which each nucleus can be either in an excited state or the ground state. We describe the state of each nucleus by a two-component spinor, and define Pauli spin matrices denoted by σ_μ^i where $i = 1, 2, 3$ and $\mu = 1......N$ and σ_μ^3 has the eigenvalue $+1$ if nucleus number

μ is excited and has the eigenvalue -1 if nucleus number μ is in its ground state. The interaction Hamiltonian describing transitions between the excited and ground state of a nucleus by the emission or absorption of a photon with wave number $\vec{k}$ is

$$H_{int}(\vec{k}) = g \cdot \left(\sum_{\mu=1}^{N} a^{\dagger}_{\vec{k}} e^{-i\vec{k}\cdot(\vec{r}_\mu + \vec{\xi}_\mu)} \sigma^{-}_{\mu} + \sum_{\mu=1}^{N} a_{\vec{k}} e^{i\vec{k}\cdot(\vec{r}_\mu + \vec{\xi}_\mu)} \sigma^{+}_{\mu} \right), \qquad (14)$$

where $\vec{r}_\mu$ denotes the equilibrium position of nucleus μ, $\vec{\xi}_\mu$ the displacement of the nucleus from its equilibrium position, and $\vec{p}_\mu$ its momentum, $a^{\dagger}_{\vec{k}}$ is a creation operator for a photon with momentum $\vec{k}$, we neglect spin, $\sigma^{\pm}_{\mu} = ((\sigma^1_\mu + i\sigma^2_\mu)/2$ is a quasispin raising or lowering operator and g is a constant specifying the strength of the interaction which multiplies all transition matrix elements and determines absolute decay rates. The unperturbed Hamiltonian for the system including the lattice dynamics can be written:

$$H = \frac{\epsilon}{2} \sum_{\mu=1}^{N} \sigma^3_\mu + \frac{\vec{p}^2_\mu}{2M} + \sum_{\mu,\nu=1}^{N} V_{\mu\nu} \,, \qquad (15)$$

where ϵ is the nuclear excitation energy, M is the mass of the atom or ion and $V_{\mu\nu}$ is some two-body interaction potential depending only upon the coordinates of the atoms and not on their momenta.

The transition rate for elastic (Mösssbauer) photon emission between some initial state denoted by $|\psi_i\rangle$ and a final state denoted by $|\psi_f\rangle$ in which the state of the lattice is unchanged is given by the Fermi Golden Rule of time-dependent perturbation theory as proportional to the square of the transition matrix element of the operator Eq. (14),

$$W^{el}_{i\to f}(\vec{k}) = g^2 f_{LM} \cdot \mid \langle\psi_f| \sum_{\mu=1}^{N} e^{-i\vec{k}\cdot\vec{r}_\mu} \sigma^{-}_{\mu} \, |\psi_i\rangle \mid^2 , \qquad (16)$$

where f_{LM} denotes the Lamb-Mössbauer factor

$$f_{LM} = \mid \langle\psi_i| \, e^{-i\vec{k}\cdot\vec{\xi}_\mu} \, |\psi_i\rangle \mid^2 , \qquad (17)$$

which gives the probability that the transition is elastic and we assume that f_{LM} is the same for all nuclei μ. This must be corrected in the case where the Mössbauer nuclei occupy different types of lattice sites and have different values for f_{LM}, but does not affect our conclusions regarding the effects of multiple excitation coherence.

Thus the sum of the elastic transition rates over all final states is proportional to the sum of Eq. (16) and a common phase space factor , which we disregard. Evaluating this sum by closure gives

$$W_i(\vec{k}) = \sum_f W^{el}_{i\to f}(\vec{k}) = g^2 f_{LM} \cdot \langle\psi_i| \sum_{\mu,\nu=1}^{N} e^{i\vec{k}\cdot\vec{r}_\nu}\sigma_\nu^+ e^{-i\vec{k}\cdot\vec{r}_\mu}\sigma_\mu^- \,|\psi_i\rangle \ . \qquad (18)$$

The direct term in the sum with $\mu = \nu$ is can be trivially evaluated to give

$$W_i(\vec{k}) = g^2 f_{LM} \cdot N_e + g^2 f_{LM}\delta W_i(\vec{k}) \qquad (19)$$

where N_e denotes the number of excited nuclei in the initial state and the interference term $\delta W_i(\vec{k})$ is given by

$$\delta W_i(\vec{k}) = \langle\psi_i| \sum_{\nu=1}^{N} \sum_{\substack{\mu=1\\ \mu\neq\nu}}^{N} e^{i\vec{k}\cdot\vec{r}_\nu}\sigma_\nu^+ e^{-i\vec{k}\cdot\vec{r}_\mu}\sigma_\mu^- \,|\psi_i\rangle \ . \qquad (20)$$

The direct term arises from the independent emission from each excited atom and is proportional to the number N_e of excited nuclei. The interference term expresses the quantum mechanics of the coherence between the excitations of a nucleus at the point $\vec{r}_\nu$ and a nucleus at the point $\vec{r}_\mu$. This is the direct analog of the famous two slit experiment in which the amplitudes of the waves passing through the two slits must be added if the slit through which the particle passed is not known. Here the amplitudes from the excitations of two nuclei must be added if it is not known which nucleus was excited. The relative phase of the two excitations is determined by the phase factor $e^{i\vec{k}\cdot\vec{r}_\nu} \cdot e^{-i\vec{k}\cdot\vec{r}_\mu}$ appearing in the interaction operator and by the relative phase in the initial state $|\psi_i\rangle$; e.g. by the phase in a nuclear exciton determined by the way it was produced. When this produces constructive interference the resulting enhancement has sometimes been called superradiance, but this has been confused in the literature.

There are two different sources for enhancement. One source is the interference between amplitudes for the emission of a photon by different nuclei from a state when only one nucleus is excited; i.e. $N_e = 1$, when the state is described as the state of a single nuclear exciton. The enhancement factor here is a function of the total number of nuclei N which can be excited. This coherent emission is sometimes also called superradiance, but this designation is controversial. The other source for enhancement arises from the presence of several excitations; i.e. $N_e > 1$. This introduces a combinatorial factor proportional to N_e which is generally called superradiance. This is just the enhancement factor that arose in the simple examples (2) and (3) and can be considered as analogous to stimulated emission of excitons considered as bosons.

3.2 The wave functions of states with one or two nuclear excitons

We now construct explicitly the wave functions for states with one or two nuclear excitons and examine the coherence effects discussed in the simple model with several independent excitations.

Let $|\psi_{0i}\rangle$ denote the initial state with no nuclei excited and the lattice in a state denoted by i. The elastic transition in the lattice induced by the absorption of a photon with wave vector $\vec{k}$ is described by the interaction Hamiltonian (14) and leads to a final state of the lattice with one nuclear excitation

$$|\psi_{f1}\rangle = H_{int}(\vec{k})\,|\psi_{0i}\rangle = g\sqrt{f_{LM}} \cdot \sum_{\mu=1}^{N} e^{i\vec{k}\cdot\vec{r}_\mu}\sigma_\mu^+\,|\psi_{0i}\rangle\ . \tag{21}$$

This is just the nuclear exciton state. The norm of this state is

$$\langle\psi_{f1}\,|\psi_{f1}\rangle = g^2 f_{LM} N\,. \tag{22}$$

Thus the normalized state with one excitation (the one-exciton wave function) is

$$|\psi_1\rangle = \frac{1}{\sqrt{N}} \cdot \sum_{\mu=1}^{N} e^{i\vec{k}\cdot\vec{r}_\mu}\sigma_\mu^+\,|\psi_{0i}\rangle = \frac{1}{g\cdot\sqrt{f_{LM}N}} \cdot H_{int}(\vec{k})\,|\psi_{0i}\rangle\ . \tag{23}$$

The transition rate given by the Fermi Golden Rule is proportional to the square of the transition matrix element

$$|\,\langle\psi_1|\,H_{int}(\vec{k})\,|\psi_{0i}\rangle\,|^2 = g^2 N f_{LM}\,. \tag{24}$$

This proportionality to $g^2 N f_{LM}$ is exactly the same as for the transition rate in the incoherent excitation of N nuclei.

We now consider the transition in the lattice induced by the absorption of two photons with wave vector $\vec{k}$ and described by the interaction Hamiltonian (14) as a double excitation. This leads to a two-exciton final state of the lattice:

$$|\psi_{f2}\rangle = [H_{int}(\vec{k})]^2\,|\psi_{0i}\rangle = g^2 f_{LM} \cdot \sum_{\mu,\nu=1}^{N} e^{i\vec{k}\cdot\vec{r}_\mu}\sigma_\mu^+ e^{i\vec{k}\cdot\vec{r}_\nu}\sigma_\nu^+\,|\psi_{0i}\rangle\ , \tag{25}$$

$$|\psi_{f2}\rangle = g\cdot\sqrt{f_{LM}N}\cdot[H_{int}(\vec{k})]\,|\psi_1\rangle\ . \tag{26}$$

The normalized two-exciton wave function is easily obtained by using some quasispin algebra

$$\sigma_\tau^- \sigma_\rho^- \sigma_\mu^+ \sigma_\nu^+ \left|\psi_{0i}\right\rangle = \delta_{\tau\mu}\delta_{\rho\nu} + \delta_{\tau\nu}\delta_{\rho\mu} - 2\delta_{\tau\mu}\delta_{\rho\nu}\delta_{\mu\nu} \left|\psi_{0i}\right\rangle , \tag{27}$$

$$\left\langle\psi_{f2}\right|\psi_{f2}\rangle = g^4 \cdot 2N(N-1) f_{LM} , \tag{28}$$

$$\left|\psi_2\right\rangle = \frac{1}{\sqrt{2N(N-1)}} \cdot \sum_{\mu,\nu=1}^{N} e^{i\vec{k}\cdot\vec{r}_\mu}\sigma_\mu^+ e^{i\vec{k}\cdot\vec{r}_\nu}\sigma_\nu^+ \left|\psi_{0i}\right\rangle , \tag{29}$$

$$\left|\psi_2\right\rangle = \frac{1}{g\cdot\sqrt{2(N-1)f_{LM}}} \cdot H_{int}(\vec{k}) \left|\psi_1\right\rangle . \tag{30}$$

The transition rate for the excitation of the second exciton from a one-exciton state is given by the Fermi Golden Rule as proportional to the square of the transition matrix element

$$\left| \left\langle\psi_2\right| H_{int}(\vec{k}) \left|\psi_1\right\rangle \right|^2 = 2g^2(N-1) f_{LM} . \tag{31}$$

This is double that for the transition rate in incoherent double excitation of N nuclei. It appears that there is an enhancement of the cross section by a factor of two because of the coherence between the two nuclear excitons. This is the same factor of two that appeared in the simple example (4-13). The Lamb-Mössbauer factor appears here just as in the normal case of double excitation.

3.3 Decays of states with one or two nuclear excitons

The cross section calculated above by first-order perturbation theory may not be valid for the case of a resonance. For a more reliable estimate, we follow the procedure used in the simple example described in Eqs. (4-13) and examine the decays of states with one or two nuclear excitons. We note that the wave vector $\vec{k}'$ of the emitted photon can be different from wave vector $\vec{k}$ of the photon that created the exciton. The transition matrix element for such a decay from a state with a single nuclear exciton (23) is

$$\left\langle\psi_{0i}\right| T(\vec{k}') \left|\psi_1\right\rangle = \left\langle\psi_{0i}\right| H_{int}(\vec{k}') \left|\psi_1\right\rangle = g \cdot \left\langle\psi_{0i}\right| \sum_{\rho=1}^{N} e^{-i\vec{k}'\cdot\vec{r}_\rho}\sigma_\rho^- \left|\psi_1\right\rangle , \tag{32}$$

$$\left\langle\psi_{0i}\right| T(\vec{k}') \left|\psi_1\right\rangle = \frac{g}{\sqrt{N}} \cdot \left\langle\psi_{0i}\right| \sum_{\mu,\rho=1}^{N} e^{-i\vec{k}'\cdot\vec{r}_\rho}\sigma_\rho^- e^{i\vec{k}\cdot\vec{r}_\mu}\sigma_\mu^+ \left|\psi_{0i}\right\rangle , \tag{33}$$

$$\langle\psi_{0i}|\, T(\vec{k}')\, |\psi_1\rangle = \frac{g}{\sqrt{N}} \cdot \langle\psi_{0i}| \sum_{\rho=1}^{N} e^{i(\vec{k}-\vec{k}')\cdot\vec{r}_\rho}\, |\psi_{0i}\rangle \ . \tag{34}$$

The transition for photon emission from a two-exciton state (29) is described as a decay in two steps : first by emission of a photon with wave vector $\vec{k}'$ to a singly excited state and subsequently to a state with no excitation.

The transition matrix element between this two-exciton state $|\psi_2\rangle$ and a final one-exciton state by emission of a photon with wave vector $\vec{k}'$ is

$$\langle\psi_1|\, T(\vec{k}')\, |\psi_2\rangle = g \cdot \langle\psi_1| \sum_{\rho=1}^{N} e^{-i\vec{k}'\cdot\vec{r}_\rho}\sigma_\rho^-\, |\psi_2\rangle \tag{35}$$

$$\langle\psi_1|\, T(\vec{k}')\, |\psi_2\rangle =$$

$$= \frac{g}{N\sqrt{2(N-1)}} \cdot \sum_{\mu,\nu,\rho,\tau=1}^{N} \langle\psi_{0i}|\, e^{-i\vec{k}\cdot\vec{r}_\tau}\sigma_\tau^-\, e^{-i\vec{k}'\cdot\vec{r}_\rho}\sigma_\rho^-\, e^{i\vec{k}\cdot\vec{r}_\mu}\sigma_\mu^+ e^{i\vec{k}\cdot\vec{r}_\nu}\sigma_\nu^+\, |\psi_{0i}\rangle \ , \tag{36}$$

$$\langle\psi_1|\, T(\vec{k}')\, |\psi_2\rangle = g \cdot \frac{\sqrt{2(N-1)}}{N} \cdot \sum_{\rho=1}^{N} \langle\psi_{0i}|\, e^{i(\vec{k}-\vec{k}')\cdot\vec{r}_\rho}\, |\psi_{0i}\rangle \ , \tag{37}$$

$$\langle\psi_1|\, T(\vec{k}')\, |\psi_2\rangle = \frac{\sqrt{2(N-1)}}{\sqrt{N}} \cdot \langle\psi_{0i}|\, T(\vec{k}')\, |\psi_1\rangle \ , \tag{38}$$

where we have used Eq. (27).

Comparing this result with the transition matrix element for a one-exciton decay, we see that the double excitation enhances the decay rate by a factor $2(N-1)/N \approx 2$. This is exactly the same enhancement that appeared in the simple example (4-13) and (31) observed for the excitation of a two-exciton state from a one-exciton state where we have used the Fermi Golden Rule to describe the excitation. While the Golden Rule may not be adequate to describe a resonance excitation, it is definitely adequate to describe the decay of an excited state into the continuum by the emission of a photon, and the same combinatorial factor describing the effects of exciton coherence appears in both excitation and decay.

The same factor of two applies both to inelastic photon emission and internal conversion. For these cases the transition matrix element for inelastic emission or internal conversion has the same form as in Eqs. (35-37) except that there is no sum over the index ρ. Since it is known by the nuclear recoil and/or the vacancy produced by the conversion electron which nucleus emitted the photon, the transitions in different nuclei are incoherent, and the total transition rate is obtained by squaring the transition matrix element for each nucleus and then summing over all nuclei. The result gives the same factor of two.

Regardless of whether the decay of the first excitation is coherent or incoherent, the system remains in a nuclear exciton state with a single excited nucleus in the lattice. However there is also a single photon present. The lattice transition matrix element is now exactly the same as for the single nuclear exciton. But there is now an additional possible enhancement due to stimulated emission.

This suggests that if a synchrotron radiation pulse is incident on a crystal which contains one nuclear exciton, the width of the resonance for excitation producing a second nuclear exciton is double the width for the case where no nuclear exciton is initially present. This can be a very important effect if the number of excitons present is appreciable.

3.4 Decays via internal conversion and inelastic scattering

To consider the case of a decay via internal conversion, we write the normalized wave functions for the final states after singly and doubly excited states have decayed via internal conversion in an atom at $\vec{r}_\mu$ leaving a hole in an electron shell,

$$|\psi_{0h\mu}\rangle = h^\dagger(\vec{r}_\mu)\,|\psi_{0i}\rangle \tag{39}$$

$$|\psi_{1h\mu}\rangle = \frac{1}{\sqrt{N-1}} \cdot \sum_{\nu=1;\nu\neq\mu}^{N} h^\dagger(\vec{r}_\mu) e^{i\vec{k}\cdot\vec{r}_\nu} \sigma_\nu^+ \,|\psi_{0i}\rangle \ , \tag{40}$$

where $h^\dagger(\vec{r}_\mu)$ denotes an operator that creates a hole in an electron shell of the atom at $\vec{r}_\mu$.

The matrix element for an internal conversion transition is

$$\langle\psi_{1h\mu}|\, T^\dagger(h)\,|\psi_2\rangle \quad = \quad g' \cdot \langle\psi_{1h\mu}|\, h^\dagger(\vec{r}_\mu)\sigma_\mu^- \,|\psi_2\rangle \ , \tag{41}$$

$$\begin{aligned} \langle\psi_{1h\mu}|\, T^\dagger(h)\,|\psi_2\rangle \quad &= \quad \frac{g'}{(N-1)\sqrt{2N}} \\ \times \sum_{\nu\neq\mu,\rho,\tau,=1}^{N} &\langle\psi_{0i}|\, e^{-i\vec{k}\cdot\vec{r}_\nu}\sigma_\nu^- \, h(\vec{r}_\mu) h^\dagger(\vec{r}_\mu)\sigma_\mu^- e^{i\vec{k}\cdot\vec{r}_\rho}\sigma_\rho^+ e^{i\vec{k}\cdot\vec{r}_\tau}\sigma_\tau^+ \,|\psi_{0i}\rangle \ . \end{aligned} \tag{42}$$

Thus,

$$\langle\psi_{1h\mu}|\, T^\dagger(h)\,|\psi_2\rangle = \sqrt{2} \cdot \langle\psi_{0h\mu}|\, T^\dagger(h)\,|\psi_1\rangle \ . \tag{43}$$

Inelastic emission is treated in the same way as internal conversion. Since it is known by the nuclear recoil as in the vacancy produced by the conversion electron which nucleus emitted the photon, the transitions in different nuclei are incoherent, and the total transition rate is obtained by squaring the transition matrix element for each nucleus and then summing over all nuclei. The result gives the same factor of two.

3.5 The difference between coherent and incoherent cascade decays

We now show by the use of explicit nuclear exciton wave functions how the conclusions from the simple example (4-13) apply to the exciton case, and the excitation widths are different for coherent and incoherent excitations. The two-exciton cascade decay can be written:

$$|\psi_2\rangle \to \sqrt{2}\cdot\langle\psi_{0i}|\,T(\vec{k}')\,|\psi_1\rangle\cdot|\psi_1,\gamma\rangle \to \sqrt{2}\cdot[\langle\psi_{0i}|\,T(\vec{k}')\,|\psi_1\rangle]^2\cdot|\psi_0,2\gamma\rangle\ . \quad (44)$$

The analogous incoherent cascade beginning with two well-defined excited nuclei at lattice points μ and ν is described by the simple model in Eqs. (4) and (5) for the more general case of a triple cascade.

Both the coherent and incoherent transitions have the same doubling of the decay rate for the first transition from the doubly excited state to the singly excited state in comparison with the decay rate for the singly excited state. In the time reversed transitions the excitation of a second nucleus from the intermediate state with one excitation has double the width of the resonance for the excitation of a single nucleus. But the state describing the coherent single independent excitation at two well defined lattice sites cannot be produced in any realistic excitation experiment. However, the single nuclear exciton excitation which occurs in the decay (44) is easily achieved in an excitation experiment by exciting the crystal with a synchrotron radiation photon.

In the coherent case, the transition occurs between a coherent state with two excitations and a coherent state with a single excitation, accompanied by a single photon state. The factor of two implies a speedup of the decay of a single transition and a doubling of the natural line width. There then follows the decay of the single coherent state, now with the normal speeded up lifetime for the single transition.

The coherent two-step transition is therefore a cascade, whose time behavior is exactly the same as in the simple example (4-13).

Regardless of whether the decay of the first excitation is coherent or incoherent, the system remains in a coherent state with a single excited nucleus in the lattice. The lattice transition matrix element is now exactly the same as for the singly excited coherent case. However in the coherent case there is also a single photon present and therefore an additional possible enhancement due to stimulated emission. However, probability of such stimulated emission is probably small.

4 The quasispin description of states with many nuclear excitons

4.1 Explicit description of multiple exciton states

We construct the whole set of nuclear exciton states explicitly using our quasispin algebra. We immediately encounter a crucial difference between the nuclear exciton and Dicke superradiance.[11] Because each excited nucleus is at a different well-defined point on a lattice or in an amorphous solid, an additional degree of freedom arises here which does not exist in Dicke superradiance.

The physics of the difference is seen by noting that in any system of N nuclei, located at different points in a solid, there are N independent states of the system in which one nucleus is excited and all the others are in their ground state. The nuclear exciton created by the absorption of a single photon from a synchrotron radiation source is a well-defined linear combination of these N states with well defined phases between them determined by the production mechanism. With these phases, the amplitudes for the decay by emission of a photon in the forward direction all interfere constructively, while the amplitude for emission in the forward direction vanishes for all the other $N-1$ states of the N nucleon system which are orthogonal to the nuclear exciton state.

This physics is conveniently described formally in our quasispin description by noting that an arbitrary phase arises in defining the total quasispin of the system, and that this phase can be chosen to pick out the particular states for which interference between different forward emission amplitudes is constructive. We begin by defining the quasispin operators for each nucleus as objects with quasispin 1/2 defined in terms of the Pauli spin operators $\vec{\sigma}_\mu$ with the conventional factor (1/2),

$$\vec{s}_\mu = (1/2)\vec{\sigma}_\mu; \qquad s_\mu^{\pm} = \sigma_\mu^{\pm}\,. \tag{45}$$

We now define the total quasispin of the system by choosing appropriate phase factors as follows:

$$S_{\vec{k}}^{\pm} = \sum_{\mu=1}^{N} e^{\pm i\vec{k}\cdot\vec{r}_\mu} s_\mu^{\pm} = \sum_{\mu=1}^{N} e^{\pm i\vec{k}\cdot\vec{r}_\mu} \sigma_\mu^{\pm}; \qquad S^3 = \sum_{\mu=1}^{N} s_\mu^3 = (1/2)\sum_{\mu=1}^{N} \sigma_\mu^3\,, \tag{46}$$

$$(S_{\vec{k}})^2 \equiv (1/2)(S_{\vec{k}}^{+}S_{\vec{k}}^{-} + S_{\vec{k}}^{-}S_{\vec{k}}^{+}) + (S^3)^2\,. \tag{47}$$

The total quasispin operators satisfy angular momentum commutation rules by construction,

$$[S^3, S^{\pm}_{\vec{k}}] = \pm S^{\pm}_{\vec{k}};\ [S^{+}_{\vec{k}}, S^{-}_{\vec{k}}] = 2S^3;\ [(S_{\vec{k}})^2, (S^{+}_{\vec{k}}] = [(S_{\vec{k}})^2, S^{-}_{\vec{k}}] = [(S_{\vec{k}})^2, S^3] = 0\,. \tag{48}$$

Any arbitrary choice of phases with opposite signs for the raising and lowering quasispin operators will give total quasispin operators that satisfy the angular momentum commutation rules. Changing these phases by adding an additional space-dependent phase gives a unitary transformation which resembles a gauge transformation. However, the interaction Hamiltonian (14) is not invariant under such an apparent gauge transformation. The choice of phases (46) chooses a "gauge" in which the interaction Hamiltonian (14) assumes the very simple form:

$$H_{int}(\vec{k}) = g \cdot (a^{\dagger}_{\vec{k}} S^{-}_{\vec{k}} + a_{\vec{k}} S^{+}_{\vec{k}})\,, \tag{49}$$

and the total Hamiltonian (14-15) which includes the part of the interaction that describes emission and absorption of photons of wave vector $\vec{k}$ can be written

$$H_{tot}(\vec{k}) = H + H_{int}(\vec{k}) = \frac{\epsilon}{2} S^3 + \frac{\vec{p}^2_{\mu}}{2M} + \sum_{\mu,\nu=1}^{N} V_{\mu\nu} + g \cdot (a^{\dagger}_{\vec{k}} S^{-}_{\vec{k}} + a_{\vec{k}} S^{+}_{\vec{k}})\,. \tag{50}$$

We now note that this piece of the total Hamiltonian commutes with the total quasispin

$$[(S_{\vec{k}})^2, H_{tot}(\vec{k})] = 0\,. \tag{51}$$

The operators $S^{\pm}_{\vec{k}}$ are seen to be operators which excite and de-excite nuclear exciton states. The operator S^3 simply counts the number N_e of excited nuclei. Its eigenvalues are $N_e-(N/2)$ and varies from $-N/2$ when no nuclei are excited to $+N/2$ when all nuclei are excited.

The states of the system can be classified into quasispin multiplets labeled by the eigenvalue of the operator $(S_{\vec{k}})^2$ denoted in the common notation by $S_{\vec{k}}(S_{\vec{k}}+1)$. The individual states in the multiplet can be labeled by the eigenvalues of S^3 denoted by m. We immediately note that the lowest state in any given multiplet with $m = -S_{\vec{k}}$ is annihilated by the interaction operator (49) if there are no photons initially present,

$$H_{int}(\vec{k})\,|S_{\vec{k}}, -S_{\vec{k}}\rangle = g \cdot a^{\dagger}_{\vec{k}} S^{-}_{\vec{k}}\,|S_{\vec{k}}, -S_{\vec{k}}\rangle = 0\,. \tag{52}$$

Thus these lowest states in the quasispin multiplet cannot radiate photons in the forward direction; i.e. in the direction of $\vec{k}$. This is trivially obvious for the case $S_{\vec{k}} = N/2$ where the lowest state in the multiplet contains no excited nuclei. However, for other values of $S_{\vec{k}}$ the lowest state contains $N_e = (N/2) - S_{\vec{k}}$ excited nuclei which from Eq. (52) are not allowed to decay by emitting forward photons. These excited states of the system are thus called "subradiant"[5] in the forward direction, in contrast to the coherent nuclear exciton states which are sometimes called "superradiant" in the forward direction.

The states in a given multiplet can be characterized by a "seniority number" $v \equiv (N/2) - S_{\vec{k}}$ which is just the number of subradiant nuclear excitations in the system which cannot radiate in the forward direction. The states in the multiplet are created by adding nuclear excitons.

For the case where the number of excitations N_e is small it is convenient to define "normalized exciton creation and destruction operators", and verify that in the approximation where the number of excited nuclei is small compared to the total number of nuclei, $2N_e/N \ll 1$ they indeed satisfy boson commutation rules.

$$X^\dagger = \frac{1}{\sqrt{N}} \cdot S^+_{\vec{k}}; \qquad X = \frac{1}{\sqrt{N}} \cdot S^-_{\vec{k}} \,, \tag{53}$$

$$[X^\dagger, X] = \frac{1}{N} \cdot [S^+_{\vec{k}}, S^-_{\vec{k}}] = \frac{2}{N} \cdot S^3 = -1 + \frac{2N_e}{N} = -1 + O\left(\frac{N_e}{N}\right) \approx -1 \,, \tag{54}$$

$$[\sigma^-_\mu, X^\dagger] = \frac{-2}{\sqrt{N}} \cdot e^{i\vec{k}\cdot\vec{r}_\mu} s^3_\mu = \frac{1}{\sqrt{N}} \cdot e^{i\vec{k}\cdot\vec{r}_\mu} \left[1 + O\left(\frac{N_e}{N}\right)\right] \approx \frac{1}{\sqrt{N}} \cdot e^{i\vec{k}\cdot\vec{r}_\mu} \,, \tag{55}$$

where we have noted that the operator s^3_μ has the eigenvalue $-1/2$ for all terms in the multiexciton wave function in which the nucleus at the point $\vec{r}_\mu$ is not excited.

The Hamiltonian (50) is simply expressed in terms of these exciton operators,

$$H_{tot}(\vec{k}) = H + H_{int}(\vec{k}) = \frac{\epsilon}{2} S^3 + \frac{\vec{p}^{\,2}_\mu}{2M} + \sum_{\mu,\nu=1}^{N} V_{\mu\nu} + g\sqrt{N} \cdot (a^\dagger_{\vec{k}} X + a_{\vec{k}} X^\dagger) \,. \tag{56}$$

The transition matrix elements for the absorption or emission of a photon from a state where there are already n excitons present and no subradiant excitations; i.e. $S_{\vec{k}} = N/2$, is seen to be

$$\langle n+1|\, H_{int}(\vec{k}) \left| n, \vec{k} \right\rangle = g \,\langle m+1|\, S^+_{\vec{k}} \,|m\rangle = g\sqrt{(S-m)(S+m+1)} \,, \tag{57}$$

$$\langle n+1|\, H_{int}(\vec{k}) \left| n, \vec{k} \right\rangle = g\sqrt{(n+1)(N-n)} \approx g\sqrt{nN} \,, \tag{58}$$

$$\left\langle n-1, \vec{k} \right|\, H_{int}(\vec{k}) \,|n\rangle = g \,\langle m-1|\, S^-_{\vec{k}} \,|m\rangle = g\sqrt{(S-m+1)(S+m)} \,, \tag{59}$$

$$\left\langle n-1,\vec{k}\right| H_{int}(\vec{k})\left|n\right\rangle = g\sqrt{n(N-n+1)} \approx g\sqrt{nN}\,. \tag{60}$$

4.2 Explicit description of the two enhancements arising in a multiple exciton state

We now generalize the treatment of two-exciton decays in Eqs. (35- 37) to the general multi-exciton case using the formalism of Eqs. (18 - 20) and show the interplay of the two enhancements.

We write the initial state with N_e excitons

$$|\psi(N_e)\rangle \equiv \frac{(X^\dagger)^{N_e}\,|\psi_{0i}\rangle}{\sqrt{N_e!}}\,. \tag{61}$$

Then

$$\sigma_\mu^-\,|\psi(N_e)\rangle \approx \sigma_\mu^- \cdot \frac{(X^\dagger)^{N_e}\,|\psi_{0i}\rangle}{\sqrt{N_e!}} = \sqrt{\frac{N_e}{N}} \cdot e^{i\vec{k}\cdot\vec{r}_\mu} \cdot \frac{(X^\dagger)^{(N_e-1)}}{(N_e-1)!}\,|\psi_{0i}\rangle\,. \tag{62}$$

Substituting Eqs. (61) and (62) into Eq. (20) gives

$$\delta W_{N_e}(\vec{k}') \approx \sum_{\nu=1}^{N}\sum_{\substack{\mu=1\\ \mu\neq\nu}}^{N} \frac{N_e}{N} \cdot \frac{\langle\psi_{0i}|\,e^{i(\vec{k}'-\vec{k})\cdot(\vec{r}_\nu-\vec{r}_\mu)}(X)^{(N_e-1)}(X^\dagger)^{(N_e-1)}\,|\psi_{0i}\rangle}{\sqrt{(N_e-1)!}}\,, \tag{63}$$

$$\delta W_{N_e}(\vec{k}') \approx \sum_{\nu=1}^{N}\sum_{\substack{\mu=1\\ \mu\neq\nu}}^{N} \frac{N_e}{N} \cdot e^{i(\vec{k}'-\vec{k})\cdot(\vec{r}_\nu-\vec{r}_\mu)} \approx N_e \cdot \delta W_{N_e=1}(\vec{k}')\,. \tag{64}$$

The factor N_e is the enhancement due to superradiance. The lattice sum over the indices μ and ν determine the enhancement due to the coherence from the nuclear exciton. This enhancement factor depends upon the geometry and is not proportional to the number of nuclei N that can be excited. For a three dimensional system this enhancement or speedup is proportional to $N^{1/3}$.

The enhancement factor N_e can also be seen as a "boson enhancement" factor as in Eqs. (2) and (3) or as a simple combinatorial factor. A one-exciton state in which any one of N nuclei can be excited with equal amplitude is the sum of N equal terms each normalized by a factor $1/\sqrt{N}$. A two-exciton state in which any pair of the $N(N-1)/2$ possible pairs of nuclei can be excited with equal amplitude is the sum of $N(N-1)/2$ equal terms each normalized by a factor $\sqrt{2}/[N(N-1)$. Each term in the two-exciton state can decay into either of two terms in the one-exciton state. Thus there are a total of $N(N-1)$ equal terms contributing to the transition matrix element between the two-exciton and the one-exciton state, and the product of the normalization factors for the two states produce an overall normalization factor of $\sqrt{2/[N^2(N-1)]}$. This gives an enhancement factor for the transition rate of $2 \times (N-1)$. The factor $N-1$ arises from the coherence of the exciton decay in the exact forward direction. The factor 2 arises because there are two excitons in the initial state, $N_e = 2$. A similar combinatorial factor of N_e arises for all numbers of excitations.

5 Conclusions

We find that nuclear excitons behave like a collective boson excitation, where production of a nuclear exciton can be enhanced by a "stimulated emission factor" if there are already nuclear excitons present. Thus the excitation of the n-th nuclear exciton in a system which already contains $n-1$ nuclear excitons is enhanced by a factor n over the excitation of a single exciton when no others are present. This implies that the partial width for the excitation of the n-th exciton is enhanced by a factor n over the normal width for the decay and excitation of a single exciton.

Since the excitation probability from a broad-band synchrotron beam with a width much larger than the natural line width of the nuclear resonance depends upon the integral of the cross section over the energy width of the beam, the increased partial width of the resonance produces an increased excitation probability from the broad beam.

The decay of an n-exciton state is the sum of exponentials corresponding to the enhanced decay widths of each state containing more than a single nuclear exciton. However, the time dependence of the radiation emitted is exactly the same as for n independent excitons, and contains only a single exponential which is the same as that for a single exciton.

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