Superradiance and anomalous hyperfine splitting in inhomogeneous ensembles

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Collective effects in the interaction of light with ensembles of identical scatterers play an important role in many fields of physics. However, often the term “identical” is not accurate due to the presence of hyperfine fields which induce inhomogeneous transition shifts and splittings. Here we develop a formalism based on the Green function method to model the linear response of such inhomogeneous ensembles in one-dimensional waveguides. We obtain a compact formula for the superradiant response of multilevel systems in one-dimensional waveguides, where in the same time the emitters are inhomogeneous.

One-dimensional waveguides play a special role for superradiance because they facilitate the otherwise challenging uniform illumination of the scatterer ensemble. Superradiance relies on permutation invariance, i.e., the invariance of the system under the exchange of any two scatterers in the ensemble, which in turn relies on uniform illumination of the ensemble. In the single-photon regime, incident pulses are re-emitted in a highly directional manner [10, 17], preserving the incident wavevector. As such, it has been found that arranging the atoms in quasi one dimensional arrangements such as pencil geometries can enhance superradiance [38]. Also, uniform illumination can be easily achieved if the atoms are placed in a one dimensional waveguide like structure [39, 41], or x-ray grazing incidence reflection from thin films [42, 49]. The atomic excitations propagate through the waveguide as a polariton, and the waveguide structure restricts the propagation of the scattered light to one dimensional, plane wave propagation. This results in uniform illumination, with translational symmetry playing the role of permutation symmetry, achieving superradiance without requiring the wavelength to be much larger than the atomic spacing.

In this work, we investigate the superradiant response of multilevel and inhomogeneous scatterers, e.g., atoms or nuclei, in one-dimensional waveguides. If the length scale of the environmental variation is much larger than the emitter spacing, the ensemble can be partitioned into approximately uniform sub-ensembles. Thus, the collective interaction can still play a significant role, however the interplay between the spectral inhomogeneity and the collective scattering results in non-trivial structure of the fields, Doppler broadening due to thermal interactions, or simply by fabrication in the case of quantum dots. Inhomogeneous broadening effects have been also investigated in the related process of superfluorescence [35-37]. It is the purpose of this work to present a versatile formalism which allows to theoretically model the superradiant response of multilevel systems in one-dimensional waveguides, where in the same time the emitters are inhomogeneous.

I. INTRODUCTION

When an ensemble of identical atoms interacts with light of wavelength much larger than the size of the ensemble, the atoms absorb and emit radiation collectively, resulting in the phenomenon of superradiance. This has been first investigated theoretically by Dicke for the case of two-level systems in 1954 [1]. Provided the average coherence of the ensemble absorbing and emitting radiation collectively, with a factor of \( N \) enhancement of the decay rate compared to the single atom value [1-4]. Following the pioneering experiment by Skribanowitz and co-workers [5], superradiance has been demonstrated and studied in many systems, such as Bose-Einstein condensates [6, 7], quantum dots [8], color centers [9], cold atoms [10, 11], Mössbauer nuclei [12, 14] and trapped atoms coupled to a cavity [15, 16], to name a few. Thus, the concept of superradiance has implications in many fields, such as quantum information [15, 16], cavity quantum electrodynamics [17, 18], astrophysics [19] or advanced light source [20].

The mechanism behind superradiance and collective emission also applies to multilevel systems, and extensions to the Dicke model for multilevel atoms have been explored [21, 27]. Like the Dicke model, these assume completely uniform illumination of the ensemble of atoms, and that the latter are all completely identical. In parallel, since the 1970s theoretical works have been addressing the effect of inhomogeneous broadening on superradiance [28-34]. This is pertinent to many physical systems where the energy levels and decay rates are different due to local environment effects such as Zeeman and hyperfine splittings induced by magnetic or electric
resulting spectra. To describe the dipole-dipole interaction we use a Green’s function method, developed by Grünér and Welsch [47], which has been successfully applied to describe superradiance in diverse systems such as atomic clouds [10] [11] [38] [39], one-dimensional waveguides [39] [40], and thin film x-ray reflection [46] [50]. A compact formula is found for the weak-excitation regime susceptibility, in terms of the coherent average of the emitters responses, with the collective interaction describable via a single complex constant. This allows for the effects of inhomogeneities and of the collective interaction to be analysed separately, allowing for a better understanding of their respective contributions to the collective spectrum.

We apply this formalism to the concrete example of x-ray quantum optics systems that comprise of ensembles of Mössbauer nuclei in thin-film x-ray cavities. The latter are a particularly suitable platform for exploring superradiance and collective interaction between emitters. In these systems, a thin layer of resonant nuclei is placed in the centre of a thin-film cavity, forming a waveguide like structure. Evanescent guided modes of the cavity radiance and collective interaction between emitters. In particular, these systems are a particularly suitable platform for exploring superradiant effects in various types of one-dimensional waveguide ensembles [39, 40], and thin film x-ray reflection [46, 50]. A brief overview are summarized in Sec. IV. Our examples for inhomogeneous nuclear hyperfine splitting are discussed in Sec. III B and III C. Conclusions and a brief outlook are summarized in Sec. IV.

II. MODEL

A. Hamiltonian and Lindblad operators

We begin with an ensemble of atoms in a one-dimensional waveguide, schematically illustrated in Fig. 1. The spectral parameters of the atoms are inhomogeneous, and the inhomogeneity is assumed to vary slowly over the inter-atomic length scale, such that the atoms can be divided into equally sized sub-ensembles that are approximately translationally and permutationally symmetric. The size of each sub-ensemble, and hence the number required, is determined by the gradient of the inhomogeneity over the spatial extent of the atoms. The variation of the inhomogeneity across each sub-ensemble should be taken to be small enough that it cannot be resolved within the linewidth of the transitions present, and can therefore be treated as a negligible perturbation. The atoms are driven by a probe field $E_p(t)e^{i(k_0x-\omega_0t)}$ of frequency $\omega_0$, wave vector $k_0$, and uniform illumination, with a possible time dependent envelope.

Following [10] [46], we work in the rotating frame of the driving field. The internal Hamiltonian for the atoms is given by

$$H_A = -\sum_n \sum_{i \in D_n} \sum_{\mu \in T_n} \hbar \Delta_\mu \left| e^{(i)}_\mu \right\rangle \left\langle e^{(i)}_\mu \right|,$$

where $D_n$ is the set of atoms in sub-ensemble $n$, $T_n$ is the set of excited states of sub-ensemble $n$, and $\Delta_\mu \ll \omega_0$ is the detuning of excited state $\mu$. Furthermore, $\hbar$ is the reduced Planck constant.

Incoherent decay is described by the Lindblad operator

$$L_A[\rho] = -\sum_n \sum_{i \in D_n} \sum_{\mu \in T_n} \hbar \gamma_\mu \mathcal{L} \left[ \rho, \left| e^{(i)}_\mu \right\rangle \left\langle e^{(i)}_\mu \right| \right].$$

where $\gamma_\mu$ is the natural decay rate of excited state $\mu$, and $\mathcal{L}[\rho, A, B] = AB\rho + \rho AB - 2B\rho A$.

At low saturations, the probe field only drives dipole transitions directly accessible from the ground state. It will be convenient to express the transition dipole vectors in the form $d_\mu \varphi$, with $d_\mu$ a dimensionless vector, and $\varphi$ the mean dipole magnitude.

The driving Hamiltonian is then given by

$$H_p = \varphi \sum_n \sum_{i \in D_n} \sum_{\mu \in T_n} d_\mu \cdot E_p(t)e^{i(k_0x-\omega_0t)} \left| e^{(i)}_\mu \right\rangle \left\langle g^{(i)} \right| + \text{h.c.}$$
The atoms couple collectively via a dipole-dipole interaction, described by the Green’s function formalism [40 46 47]. This gives the matrix elements for the dipole-dipole interaction via the classical dyadic Green’s function for the waveguide,

\[ H_{\text{dd}} = -\mu_0 \omega_0^2 \beta^2 \sum_{n,m} \sum_{\mu,\nu} d_\mu \cdot \text{Re}(\hat{G}(x_i,x_j)) \cdot d_\nu^* \left| e_\mu^{(i)} \right\rangle \langle g^{(i)} \right| \otimes \left| g^{(j)} \right\rangle \langle e_\nu^{(j)} \rangle + \text{h.c.} \]

\[ L_{\text{dd}}[\rho] = -\mu_0 \omega_0^2 \beta^2 \sum_{n,m} \sum_{\mu,\nu} d_\mu \cdot \text{Im}(\hat{G}(x_i,x_j)) \cdot d_\nu^* L \left[ \rho, \left| e_\mu^{(i)} \right\rangle \langle g^{(i)} \rangle \right|, \left| g^{(j)} \right\rangle \langle e_\nu^{(j)} \rangle \right] , \]

where \( \hat{G}(x_i,x_j) \) is the dyadic Green’s function for the waveguide, and \( \mu_0 \) the vacuum permeability, respectively.

Due to the one-dimensional propagation, and translational symmetry, the dyadic Green’s function \( \hat{G}(x_i,x_j,\omega) \) can be expressed in the form

\[ \hat{G}(x_i, x_j, \omega) = \hat{\Omega}^\perp \int \frac{dk}{2\pi L^{-1}} \hat{G}(k, \omega) e^{ik(x_i-x_j)}, \]

where \( \hat{G}(k) \) is a scalar function, \( \hat{\Omega}^\perp \) is a projection matrix for the 2D subspace perpendicular to the guided direction (Figure 1), and \( L \) is the quantisation length of the ensemble.

Following [46], we make the approximation, valid at low saturation, that the scattered radiation always has the same wave-vector as the driving field. Therefore, for the purposes of Eq. (5), we can substitute

\[ \hat{G}(x_i, x_j, \omega_0) \rightarrow \hat{\Omega}^\perp \hat{G}(k_0, \omega_0) e^{ik_0(x_i-x_j)}. \]

The plane wave phase factors can then be eliminated by the following unitary transformation:

\[ \left| e_\mu^{(j)} \right\rangle \rightarrow e^{-ik_0x_j} \left| e_\nu^{(j)} \right\rangle. \]

The model then becomes permutationally symmetric. Such systems can be analysed by a generalisation of the Holstein-Primakoff transformation [51]. This maps the collective transitions of the system to independent Bosonic modes, with the collective ground state mapping to the Bosonic vacuum. We introduce the bosonic creation (annihilation) operators \( b_\mu^\dagger \) (\( b_\mu \)) for each collective transition \( \mu \). For our system, the transformation reads

\[ \sum_{i \in D_n} \left| e_\mu^{(i)} \right\rangle \langle e_\nu^{(i)} \right| = b_\nu^\dagger b_\mu, \quad \mu, \nu \in T_n, \]

\[ \sum_{i \in D_n} e^{ik_0x_i} \left| e_\mu^{(i)} \right\rangle \langle g^{(i)} \rangle = b_\mu^\dagger \sqrt{Np_n - \sum_{\nu \in T_n} b_\nu^\dagger b_\nu} \approx \sqrt{Np_n} b_\mu^\dagger, \quad \mu \in T_n, \]

where \( p_n \) is the proportion of atoms in sub-ensemble \( n \), \( N \) is the total number of atoms in all ensembles, and

\[ [b_\nu, b_\mu^\dagger] = \delta_{\mu\nu} \delta_{mn}, \quad \mu \in T_n, \nu \in T_m. \]

As we are interested in the linear response of our system, we have \( \langle b_\mu^\dagger b_\nu \rangle \ll \sqrt{N} \), and we may linearise the collective transition operators,

\[ \sum_{i \in D_n} e^{ik_0x_i} \left| e_\mu^{(i)} \right\rangle \langle g^{(i)} \rangle \approx \sqrt{Np_n} b_\mu^\dagger, \quad \mu \in T_n. \]

In terms of these Bosonic operators, the Hamiltonian of
the linearised system reads

\[ H_A = - \sum_n \sum_{\mu \in T_n} h \Delta \mu b^\dagger_{\mu n} b_{\mu n}, \]
\[ H_{dd} = -h J \sum_{n,m} \sum_{\nu \in T_n} \sqrt{p_n p_m} d^\dagger_{\mu n} \cdot d^\dagger_{\nu m} b^\dagger_{\mu n} b_{\nu m}, \]
\[ H_p = \sqrt{N} \varphi \sum_{n} \sum_{\mu \in T_n} \sqrt{p_n} d^\dagger_{\mu n} \cdot E_p(t) b^\dagger_{\mu n} + \text{h.c.}, \]
\[ L_{A}[\rho] = - \sum_{n} \sum_{\mu \in T_n} h \gamma_{\mu n} \mathcal{L}[\rho, b^\dagger_{\mu n}, b_{\mu n}], \]
\[ L_{dd}[\rho] = -h \Gamma \sum_{n,m} \sum_{\nu \in T_n} \sqrt{p_n p_m} d^\dagger_{\mu n} \cdot d^\dagger_{\nu m} \mathcal{L}[\rho, b^\dagger_{\mu n}, b_{\nu m}], \]

where

\[ J + i \Gamma = N \mu_0 \omega_0^2 \nu^2 \mathcal{G}(k_0, \omega_0), \quad d^\dagger_{\mu n} = d_{\mu n} \cdot \mathcal{T} \cdot \mathcal{P}. \]  

(12)

Note that in transforming \( L_A \) to the Bosonic operators, we have assumed that the decay is dominated by the superradiance, \( \Gamma \gg \gamma_{\mu} \), such that we can approximate the single particle decay as collective. A more exact treatment of the single particle decay is given by Shammah et al. [52], and Gegg [23, 25], but is not significant in the low saturation regime we are considering (see Appendix [A]).

Finally, although we have considered only a single ground state, for systems with multiple ground states the resulting equations of motion are of the same form. The partitioning of the sub-ensembles can be extended to partition by the initial ground state configurations of the atoms. Terms that couple to a different ground state configuration are suppressed by a factor of \( \sqrt{N} \), and can be neglected (see Appendix [B]).

\[ (\omega + \Delta_{\mu} + i \gamma_{\mu}) b_{\mu n}(\omega) + (J + i \Gamma) \sqrt{p_n} d^\dagger_{\mu n} \cdot \sum_{m} \sqrt{p_m} \sum_{\nu \in T_n} d^\dagger_{\nu m} b_{\nu m}(\omega) = \frac{\sqrt{N} \varphi}{\hbar} \sqrt{p_n} d^\dagger_{\mu n} \cdot E_p(\omega). \]  

(17)

It will be useful at this stage to define a reference frequency scale \( \gamma_0 \). For example, this could be the natural linewidth of a single atom, which for an electric dipole excitation can be expressed in terms of the mean dipole magnitude as \( \omega_0^2 a_0^2 (3 \hbar \varepsilon_0 e_0 c^3)^{-1} \).

Multiplying both sides of (17) by

\[ \frac{\sqrt{N} \varphi p_n d^\dagger_{\mu n}}{\omega + \Delta_{\mu} + i \gamma_{\mu}}, \]  

(18)

and summing over \( n, \mu \in T_n \), we obtain

\[ \mathbf{P}(\omega) + G \mathcal{F}(\omega) \cdot \mathbf{P}(\omega) = \frac{N \varphi^2}{\hbar \gamma_0} \mathcal{F}(\omega) \cdot E_p(\omega), \]  

(19)

where \( G = \gamma_0^{-1}(J + i \Gamma) \), and

\[ \mathcal{F}(\omega) = \sum_n \sum_{\mu \in T_n} \gamma_0 d^\dagger_{\mu n} \otimes d^\dagger_{\mu n} \quad \omega + \Delta_{\mu} + i \gamma_{\mu} \]  

(20)

is the 'layer response matrix', the coherent average of the responses of each transition in each sub-ensemble. Note that in this context \( \otimes \) refers to the outer product. Such a quantity appears in the dynamical scattering approach to x-ray propagation [53, Eq. (4.13)], [54].

Solving for the polarisation we obtain

\[ \mathbf{P}(\omega) = \frac{N \varphi^2}{\hbar \gamma_0} \left( \mathcal{T} + G \mathcal{F}(\omega) \right)^{-1} \mathcal{F}(\omega) \cdot E_p(\omega). \]  

(21)
This directly gives the first order susceptibility,
\[
\chi'(\omega) = \chi_0 \left( \frac{1}{\Gamma + G \mathcal{F}(\omega)} \right)^{-1} \cdot \mathcal{F}(\omega), \tag{22}
\]
where \(\chi_0 = N\sigma^2 (h\gamma_0\omega)^{-1}\).

### C. Collective Lamb shift and cross-couplings

The coherent part of the collective coupling \(J\) has previously been referred to as a collective Lamb shift \([13\; 55\; 56]\). Indeed, in the limit of a single, uniform transition with natural decay width \(\gamma_0\), the susceptibility can be treated as a scalar, and is given by
\[
\chi(\omega) = \chi_0 \frac{\gamma_0}{\omega + \Delta + i\gamma + J + i\Gamma}, \tag{23}
\]
describing a single line, shifted by \(J\) and broadened by \(\Gamma\). However, with multilevel atoms, and inhomogeneous configurations, \(J\) does not act just as a Lamb shift, but also provides additional cross-couplings, analogous to an additional control field between transitions.

Specifically, a Lamb shift is a shift in an energy level due to emission and re-absorption of virtual photons from the same state. The analogue of this in Equation (12) is given by the diagonal matrix elements of \(H_{dd}\),
\[
-\hbar J d_\mu \cdot d^*_\mu. \tag{24}
\]

If the collective coupling acts purely as a ‘Lamb shift’, then each transition is simply shifted and broadened, giving a susceptibility of
\[
\chi'(\omega) = \chi_0 \sum_\mu \frac{\gamma_0 d^\lambda_\mu \otimes d^\lambda_\mu}{\omega + \Delta_\mu + i\gamma_\mu + (J + i\Gamma) d^\lambda_\mu \cdot d^\lambda_\mu}. \tag{25}
\]

However, we can see in Eq. (12) that due to additional cross-couplings from \(J\), and spontaneously generated coherences from \(\Gamma\) \([57]\), the resulting spectrum will not be so straightforward to interpret, and is more generally described by Eq. (22). Indeed, as we shall see in Sec. III B and III C, the spectrum for inhomogeneous two level atoms shows features that cannot be attributed to a simple Lamb shift. In the context of x-ray quantum optics, this was already hinted at in Ref. \([57]\) and later on discussed in more detail for the general case in Ref. \([26]\).

We note that in particular, Ref. \([26]\) has addressed the specific case of uniform magnetic hyperfine splitting, using the atomic cloud model of Svidzinsky et al. \([19]\) to describe the collective coupling. This method derives an interaction kernel for the effective inter-atomic interactions, which turns out to be identical to the free space Green’s function. As such, the Hamiltonian of our model reproduces the model of Ref. \([26]\) in the limiting case of atoms in free space, and uniform magnetic splitting.

### III. APPLICATION TO X-RAY QUANTUM OPTICS

We now discuss the application of the general model from Section III to x-ray quantum optics with Mössbauer nuclei, and the connection to existing formalisms. The relevant experimental setup comprises so-called x-ray thin-film cavities using grazing incidence reflection as illustrated in Fig. 2(a). In this setup, layers are stacked from alternating high and low atomic number \(Z\) materials (for instance, Pt or Pd alternating with C or B₄C) to form a waveguide structure for a pulse fired at grazing incidence to the layers. A thin resonant layer of Mössbauer nuclei, for instance \(^{57}\)Fe or \(^{57}\)Fe-enriched stainless steel \((^{57}\text{SS})\) is embedded in this stack, usually sandwiched between low-\(Z\) material layers. The scattering response of the system is recorded in the cavity reflectivity measured at the detector. As a function of incidence angle, the cavity reflectivity presents several minima, which correspond to the resonant driving of guided modes. An example is presented in Fig. 2(b). The minima are known as critical angles and they indicate the formation of a standing wave structure across the cavity layers. The resonant layer of Mössbauer nuclei is made sufficiently thin such that the guided mode field is approximately uniform across the depth of the layer.

The calculated probe field intensity profile for the cavity structure Pt 2.8 nm/C 22 nm/\(^{57}\text{SS}\) 0.6 nm/C 22.5 nm/Pt 15 nm and incidence angle \(\theta = 3.35\; \text{mrad}\) (corresponding to the third reflection minimum in Fig. 2(b)) is presented in Fig. 3. This example was chosen such that the thin layer of resonant nuclei is placed at the guided mode maximum. The low-\(Z\) layer is thereby used as an inert filler which allows the precise positioning of the resonant layer at the desired depth in the cavity. The guided mode ensures the uniform illumination of all Mössbauer nuclei in the layer.

In most cases, the driven Mössbauer transitions present hyperfine splittings. Expressed in a multipole expansion, the most significant are the isomer shift, corresponding to the monopole interaction with local electric field, the magnetic splitting due to a dipole interaction with the local magnetic field, and a quadrupole splitting resulting from the interaction with local electric field gradients. The splittings are determined by the respective magnetic fields or electric field gradients, which are in turn highly sensitive to the local electronic configuration. As most samples are polycrystalline, the hyperfine splittings will be inhomogeneous across the entire sample, but homogeneous within an individual crystal domain. For the case of the 14.413 keV Mössbauer transition in \(^{57}\)Fe, the natural width is approximately 4.6 neV and the transition has predominant magnetic dipole character. Magnetic splittings typically range in the interval \((1 - 50)\; \gamma_0\), while for example in iron carbides the isomer shift and quadrupole splitting range from \((2 - 10)\; \gamma_0\) and \((0 - 0.55)\; \gamma_0\) respectively, and vary for different crystal structures of the same chemical composition.
FIG. 2. (a) Schematic illustration of a thin-film cavity. x-rays in grazing incidence with angle \( \theta \) couple evanescently to the layered structure, exciting resonant transitions in stainless steel 95% enriched with \( ^{57}\text{Fe} \) (referred to as \(^{57}\text{SS} \)). The cavity reflectivity \( |R|^2 \) is measured at the detector.

(b) Example of theoretical reflectivity spectrum \(|R|^2(\theta)\) for a cavity with structure Pt 2.8 nm/C 22 nm/\(^{57}\text{SS}\) 0.6 nm/C 22.5 nm/Pt 15 nm, obtained using the Python library pyguss [55]. The reflectivity has various minima, that correspond to the resonant guided modes.

FIG. 3. Probe field intensity profile throughout the sample depth \( z \), for the third reflection minimum in Figure 2(b). The background shading illustrates the layer material, with the platinum capping layers forming the cavity, and the thin layer of \(^{57}\text{SS}\) placed at the guided mode maximum.

\[
|R|^2 \begin{cases} 
0 & \text{for } \theta \in [2, 6] \text{ mrad} \\
0.2 & \text{for } \theta \in [1, 7] \text{ mrad} \\
0.8 & \text{for } \theta \in [2, 6] \text{ mrad} \\
0.6 & \text{for } \theta \in [7, 10] \text{ mrad} \\
0.4 & \text{for } \theta \in [1, 10] \text{ mrad} \\
0.2 & \text{for } \theta \in [2, 10] \text{ mrad} \\
0 & \text{for } \theta \in [0, 2] \text{ mrad} 
\end{cases}
\]

\[
\theta \text{ (mrad)} \\
\begin{array}{c|c|c|c|c|c}
2 & 3 & 4 & 5 & 6 & 7 \\
0.0 & 0.2 & 0.4 & 0.6 & 0.8 & 1.0 \\
\end{array}
\]

Note that we have used electric dipole transitions in our general derivation, while the transitions in \(^{57}\text{Fe}\) are magnetic dipole. This can be dealt with trivially by making a duality transformation [60], replacing the incident electric field \( \mathbf{E} \) with the magnetic field \( \mathbf{B} \), electric dipole moment \( \mathbf{p} \) with magnetic moment \( \mu \), and so forth. In particular the electric dyadic Green’s function is replaced with its magnetic dual,

\[
\overleftrightarrow{G}(r, r') \rightarrow \overleftrightarrow{G}^*(r, r').
\]

In the effective refractive index model of x-ray scattering in matter, the propagation is described via a frequency dependent index of refraction. Outside of resonant interactions, magnetic scattering is orders of magnitude weaker than electronic scattering [61]. Thus, the magnetic permeability of the layers can be taken to be that of the vacuum, \( \mu_0 \).

Therefore, in the notation of Buhmann [60], we can obtain the dual Green’s tensor as

\[
\overleftrightarrow{G}^*(r, r') = \frac{1}{\mu_0} \nabla \times \overleftrightarrow{G}(r, r') \times \nabla' - \frac{1}{\mu_0} \delta (r - r').
\]

The dyadic delta term in the above transformation will modify each individual particle’s Lamb shift and linewidth equally, and as such can be absorbed into the definitions of \( \omega_0 \) and \( \gamma_0 \).
At grazing incidence, the partial Fourier transformed electric Green’s function was approximately given by
\[ \tilde{G}(k_0, \omega_0) \approx \sum_{\Delta} \tilde{G}(k_0, \omega_0), \quad (28) \]
\[ \tilde{G}^\dagger(k_0, \omega_0) \approx \sum_{\Delta} \tilde{G}^\dagger(k_0, \omega_0). \quad (29) \]
As seen in Eq. (13), the coherent and incoherent collective coupling strength \( J, \Gamma \) are obtainable directly from the Fourier transformed Green’s function. In addition, the quantum optical approach of Heeg and Evers [57] demonstrates that the these can also be obtained in terms of the cavity detuning \( \Delta_C \) and loss \( \kappa \) of the cavity mode excited by the probe field, such that
\[ J + i\Gamma \propto \frac{\Delta_C + i\kappa}{\Delta_C^2 + \kappa^2}. \quad (30) \]
In this model, the cavity detuning is minimized when the probe field is incident along one of the reflectivity minima of the cavity shown in Fig. [2b], and increases when the angle is shifted away from the minimum. As such, in grazing incidence cavities, the coherent coupling constant \( J \) is experimentally controllable by setting the angle of incidence of the probe field.

A. Semi-classical versus quantum models for x-ray quantum optics with Mössbauer nuclei

Before presenting some numerical examples, it is instructive to place the present formalism in the context of existing semi-classical and quantum models used in x-ray quantum optics. Previous approaches such as by Hannon and Trammel [59, 60], as well as Sturhahn [67, 68] have modelled grazing incidence x-ray reflection using a diagrammatic expansion for the photonic scattering. The nuclear interaction is treated semi-classically, with the nuclear transitions taken to be linear dipole oscillators. In this approach, the results of Eq. (22) are implicitly modelled, but not explicitly obtainable. Specifically, the response of the nuclei is modelled according to Eq. (20), and the re-scattering is implicitly included in the layer matrix formalism for the photonic propagation. However, this approach obscures the collective dynamics of the nuclei. In contrast, the present formalism makes the collective nuclear dynamics features explicit, and allows for the both the coherent and incoherent effects to be investigated separately.

A more recent model, developed by Heeg and Evers [45, 57] has focused on the quantum optical perspective, with the emphasis being on the resonant interaction of the nuclei with the cavity mode. A Green’s function approach for the scattering part of the Hamiltonian has been developed by Lentzrod et al. [46] and Kong et al. [30]. These works use analytic expressions for the Green’s functions in layered media, developed by Tomas [69] (and later on also by Johansson [70]), which can be expressed in terms of the layer matrix formalism for planar scattering. Refs. [46, 50] provide a connection between the linear response of the quantum optical model [15, 57] to the scattering model of Hannon, Trammel, Sturhahn et al. Our work extends this quantum optical Green’s function approach to include inhomogeneous hyperfine parameters, and provides an insight into how the inhomogeneity will enter into the non-linear dynamics of the system. In the following we provide a few illustrative examples.

B. Gaussian broadening for two level systems

As an illustrative example, we consider a layer of resonant Mössbauer nuclei with a Gaussian distribution of the isomer shift \( \delta \). Such Gaussian distributions of hyperfine parameters are typical of amorphous solids [41, 73]. For simplicity, we will consider the other hyperfine splittings to be negligible.

The isomer shift affects all excited states equally, and the distribution does not affect the dipole vectors or the natural linewidths. As we are taking the other hyperfine splittings to be negligible, all states with a given nuclear spin \( I \) are degenerate, and we can model the system as having a single transition. Thus, we can treat the problem as scalar.

In the continuum limit, the coherent average becomes an integral over the distribution of \( \delta \),
\[ \sum_n p_n \rightarrow \int_{-\infty}^{\infty} d\delta \ p(\delta), \quad (31) \]
where \( p(\delta) \) is the probability distribution finding a given value of \( \delta \) in the ensemble. The response function \( \mathcal{F}(\omega) \) is then given by
\[ \mathcal{F}(\omega) = \int d\delta \ p(\delta) \frac{\gamma_0}{\omega - \delta + i\gamma_0}. \quad (32) \]
With a Gaussian distribution,
\[ p(\delta; \sigma) = \frac{1}{\sqrt{2\pi} \sigma^2} \exp\left(-\frac{(\delta - \bar{\delta})^2}{2\sigma^2}\right), \quad (33) \]
this evaluates to a Voigt profile [74, Eq. 7.19],
\[ \mathcal{F}(\omega) = \frac{\gamma_0}{\sqrt{2\pi}} w\left(\omega - \bar{\delta} - i\gamma_0 \sqrt{2\pi}\right), \quad (34) \]
where
\[ w(z) = -i\sqrt{\pi} \exp\left(-z^2\right) \text{erf}(z) - i. \quad (35) \]
As the dipole vectors are all along a single direction, we need only consider the component of susceptibility in
The interaction of the coherent collective coupling $J$, and the broadening $\sigma$ is particularly interesting. Unlike the case of purely collective broadening $\Gamma$, for significant distribution widths $\sigma$ the coherent coupling factor $J$ no longer acts as a simple Lamb shift. Indeed, if $J$ were to act as a Lamb shift, from (25) one would expect instead a susceptibility of

$$\chi(\omega) = \chi_0 F(\omega + J + i\Gamma),$$

(37)

with a simple translation and broadening. Instead, the peak of the spectrum is shifted slightly further than would be in the absence of the Gaussian broadening, and is asymmetrically distorted. This is shown in Fig. 5 which presents the ratio $|\chi(\omega)/\chi_0|^2$ as a function of $\omega$ for three distribution widths $\sigma$.

For illustrative purposes, we have used values of $J, \Gamma$ in the range $(0-10)\gamma_0, (3-5)\gamma_0$, typical of x-ray cavities. For example, using pyms to simulate the single line spectrum, we find the cavity in Fig. 2 has $J = 8.5\gamma_0$ and $\Gamma = 3.36\gamma_0$ at an incident angle of 2.32 mrad, corresponding to just below the first reflection minimum. Going exactly to the first minimum gives $J = 5.5\gamma_0$, and increases $\Gamma$ significantly to $\Gamma = 18.6\gamma_0$. At an angle of 3.35 mrad, corresponding to the third reflection minimum, we have $J = 1.79\gamma_0$ and $\Gamma = 3.37\gamma_0$. Typical hyperfine distribution widths are between $(0-5)\gamma_0$. In optical contexts (for example Doppler broadening in atomic clouds, or size inhomogeneity in quantum dots), we would expect that the distribution widths could be substantially larger.

For significant distribution widths $\sigma \gg \gamma_0$, the line shape is that of a broad, almost Gaussian profile. However, as the collective coupling $J$ is increased, as well as being shifted and skewed, the effective linewidth tends to that of the incoherent coupling $\Gamma$. Figure 6 illustrates this behaviour for increasing coherent coupling $J$.

### C. Interference effects for magnetic splitting

Let us now consider the case of magnetic splitting in $^{57}$Fe with no isomer shift and an x-ray field which drives the two $m_e - m_g = 0$ transitions as shown in Fig. 4. Compared to the two-level system, the energies of these two transitions are now detuned by $\pm \phi = \pm \frac{1}{2}(\mu_e - \mu_g)B_{hi}$. Our model Hamiltonian reads

$$H = \hbar \phi (b_1^\dagger b_1 - b_2^\dagger b_2) - \hbar J (b_1^\dagger b_2^\dagger b_2 b_1 + h.c) + h.c.$$  

(38)

with Lindblad operator

$$L[\rho] = -\gamma \sum_{i=1,2} \mathcal{L}[\rho, b_i^\dagger, b_i] - \Gamma \sum_{i,j=1,2} \mathcal{L}[\rho, b_i^\dagger, b_j].$$  

(39)

The superradiant response of such a system was investigated by Kong and Pálffy [26] using an eigenvalue method. It was found that if the splitting $\phi$ is less than the incoherent part of the collective coupling $\Gamma$, the contributions from the two transitions interfere. The resulting spectrum has an interference dip in the peak, similar to electromagnetically induced transparency (EIT) [75], with the collective coupling $J$ playing the role of a control field. In addition, the coherent part of the collective coupling, $J$, was found not to act as a simple Lamb shift, but in fact non-trivially couple with the magnetic splitting, producing asymmetric, Fano-like spectra.

We now consider this system with the addition of a Gaussian distribution of magnetic field strengths across

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**FIG. 5.** Collective spectrum $|\chi(\omega)/\chi_0|^2$ as a function of the frequency $\omega$ for $J = 5\gamma_0$ and three different distribution widths $\sigma$. The peak is shifted further as the broadening is increased, and the shape is distorted.

**FIG. 6.** Collective spectrum $|\chi(\omega)/\chi_0|^2$ as a function of the frequency $\omega$ for $\sigma = 10\gamma_0$ and three different collective coupling values $J$. With increasing coherent collective coupling $J$ the shape of the spectrum is distorted asymmetrically, and the effective linewidth tends to $\Gamma$. 

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\[\chi(\omega) = \chi_0 F(\omega + J + i\Gamma), \quad (36)\]

\[\chi(\omega) = \chi_0 F(\omega + J + i\Gamma), \quad (37)\]
the sites. For a given site with splitting φ, the response matrix is given by [26]

$$F(\omega; \phi) = \frac{2\gamma_0(\omega + i\gamma_0)}{\omega^2 - \phi^2}. \quad (40)$$

In the case of a completely uniform magnetic field, the collective susceptibility is therefore given by

$$\chi(\omega) = \chi_0 \frac{2\gamma_0(\omega + i\gamma_0)}{(\omega + i\gamma_0)^2 + 2(J + i\Gamma)(\omega + i\gamma_0) - \phi^2}. \quad (41)$$

This has two poles in the denominator,

$$\omega_\pm = -i\gamma_0 - J - i\Gamma \pm \sqrt{(J + i\Gamma)^2 + \phi^2}. \quad (42)$$

When the collective coupling is completely incoherent, J = 0, the discriminant becomes \(\sqrt{\phi^2 - \Gamma^2}\). We can see that if \(\phi < \Gamma\), the argument of the square root becomes negative, and the poles become purely imaginary, describing overlapping Lorentzians with differing linewidths. This results in an EIT like dip. This behaviour is illustrated in Fig. 7 which presents the susceptibility ratio \(\chi(\omega)/\chi_0\^2\) as a function of \(\omega\) for four different values of the Gaussian distribution width \(\sigma\).

If we now consider the magnetic splitting to have a Gaussian distribution of width \(\sigma\), and mean \(\phi\), applying Eqs. (20) and (34) gives

$$F(\omega) = \int d\phi p(\phi; \bar{\phi}, \sigma) F(\omega; \phi)$$

$$= \frac{\gamma_0}{\sqrt{2\pi}2\sigma} \left( w \left( \frac{\omega - \bar{\phi} + i\gamma_0}{\sqrt{2}\sigma} \right) + w \left( \frac{\omega + \bar{\phi} + i\gamma_0}{\sqrt{2}\sigma} \right) \right), \quad (43)$$

with \(w(z)\) given by Equation (35). The susceptibility is as before given by

$$\chi(\omega) = \chi_0 \frac{F(\omega)}{1 + \gamma_0^{-1}(J + i\Gamma)F(\omega)}. \quad (44)$$

The overall envelope of the spectrum resembles that of the homogeneous case, and if the distribution width \(\sigma\) is narrow compared with \(\Gamma, \bar{\phi}\), we can see that the intensity minimum is still resolvable. However, increasing the distribution width gradually flattens the dip, and results in a flat, broad peak as shown in Fig. 7.

More interesting is the effect of different strengths of the coherent collective coupling \(J\). Rather than acting as a simple Lamb shift, the overall spectral shape is changed. One peak is flattened as the other increases, with large \(J\) resulting in a completely asymmetric picture with only a single one of the contributions being resolved. This can be seen in Fig. 7 which presents the same dependence as Fig. 6 but this time for different values of \(J\). While the peak locations are somewhat shifted, the shapes are distorted as well, and the location of the minimum is unchanged. This is in contrast to the single line case, where \(J\) acts as a pure Lamb shift.

![FIG. 7](image7.png)

**FIG. 7.** Comparison of collective spectrum \(|\chi(\omega)/\chi_0|^2\) as a function of the frequency \(\omega\) for \(\Gamma = 5\gamma_0\), \(J = 0\), with mean splitting \(\bar{\phi} = 17\gamma_0\) and various values of distribution width \(\sigma\).

As the distribution width increases, the dip is washed out to a very flat and broad peak.

![FIG. 8](image8.png)

**FIG. 8.** Comparison of collective spectrum \(|\chi(\omega)/\chi_0|^2\) as a function of the frequency \(\omega\) for small incoherent coupling \(\Gamma = 5\gamma_0\), a small distribution width value \(\sigma = 3.5\gamma_0\), mean splitting \(\bar{\phi} = 17\gamma_0\) and varying values of coherent collective coupling \(J\).

This holds even when the distribution width is large enough that the minimum is not resolved, as shown in Fig. 8. For vanishing collective coupling, \(J = 0\), the two peaks are merged, and the effective linewidth is very broad. Increasing \(J\) results in the left peak growing while the right peak diminishes, and for significant \(J\) only the left peak is individually resolved, with the linewidth approaching \(2\Gamma\). The result is an increase in the effective resolution of the spectrum, with an energy shift. As the coherent coupling strength is controlled via the angle of incidence of the driving field [46, 57], this provides a mechanism for mechanical control of the linewidth of such a sample.

To understand this, we consider the matrix form of the
corresponding equation of motion,

\[
M \begin{pmatrix} b_1(\omega) \\ b_2(\omega) \end{pmatrix} = \begin{pmatrix} \Omega(\omega) \\ \overline{\Omega}(\omega) \end{pmatrix}
\]

(45)

with

\[
M = (J + i\Gamma) \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix} + \begin{pmatrix} \phi + i\gamma_0 & 0 \\ 0 & -\phi + i\gamma_0 \end{pmatrix}.
\]

(46)

and \(\Omega(\omega) = h^{-1} \varphi E_p(\omega)\).

If \(J\) is large enough compared with \(\phi, \gamma_0\), we may treat the second term as a small perturbation of the first. The eigenvectors of \(M\) are then given by

\[
\hat{e}_\pm = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ \pm 1 \end{pmatrix} + O(\phi),
\]

(47)

with eigenvalues

\[
\lambda_+ = 2(J + i\Gamma) + O(\phi), \quad \lambda_- = O(\phi)
\]

(48)

The driving term couples to \(b_1, b_2\) equally, and is thus proportional to \(\hat{e}_+\). Therefore, only the symmetric state \(\hat{e}_+\) is strongly driven, and we will expect to see a single peak, with a Lamb shift of \(2J\) and a broadening of \(2\Gamma\). If the collective broadening \(\Gamma\) is significantly lower than the distribution width \(\sigma\), we will then see a reduction in the effective linewidth. This has potential applications in samples with significant magnetic texture, with the beam angle of incidence on the sample being used to control the collective coupling, and hence the effective linewidth.

**IV. CONCLUSION**

In this paper, we have examined an extension of the Dicke model for inhomogeneous atoms. We found a compact formula for the susceptibility, in terms of the coherently averaged nuclear/atomic responses, and the collective coupling constants \(J, \Gamma\). In addition to a collective Lamb shift and broadening, we found that the collective coupling also provides additional cross-couplings between transitions, as well as spontaneously generated coherences.

Previous work by Kong and Pálfy [29] has shown that for homogeneous multi-level atoms, the collective coupling does not act as an overall broadening and Lamb shift, and that in particular the coherent coupling \(J\) distorts the shape of the line asymmetrically. We have shown that this conclusion holds in the case of inhomogeneous ensembles, and that in addition, the coherent coupling \(J\) can counteract the inhomogeneous broadening.

Our work is applicable to one dimensional scattering geometries. For two and three dimensional geometries, if the approximation can be made that the scattering is elastic, with low recoil, then the system can be modelled as quasi one dimensional, and our approach holds. This is naturally the case for Mössbauer transitions. However, if this does not hold, the scattered radiation will be emitted in arbitrary directions, and will therefore couple differently to the various transitions depending on the direction of travel. The problem then becomes highly geometrically dependent [18, 49, 70].

In our model, we have only considered linear dynamics. The non-linear dynamics of permutationally invariant systems have been well studied [23, 25, 52], and can be applied numerically to the sub-ensembles. The Hilbert space of each sub-ensemble scales with \(N^{M+1}\) where \(N\) is the number of transitions, and \(M\) is the maximum excitation number being modeled [23,25]. If the distribution of hyperfine parameters can be modeled in a piecewise constant fashion with \(P\) pieces, then the Hilbert space can be modeled as the tensor product of \(P\) sub-ensembles, for a final dimension of \(N^{P(M+1)}\). The scaling remains polynomial in excitation number, but is exponential in sub-ensemble number, which poses a considerable challenge for numerical simulation.

Finally, we have made the assumption that there is no momentum redistribution during the scattering process. At larger saturations this will not be the case, and the permutational symmetry of the model will be broken. At this point it is not clear whether non-linearities due to the local emission, or momentum redistribution will be more significant. This is the subject of further work.

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Appendix A: Approximating single particle decay as collective

To justify approximating the single particle decay as collective, we note that we can write the exact form of the ground and excited bands, and dipole transitions are splittings compared with the energy difference between states, the ground states are considered to have small as well as multiple excited states. As with the excited coherent processes are dominated by collective interactions, and the internal decay can be approximated as a small correction to the collective decay rates.

\[ L_A[\rho] = -\sum_{\mu,i} \hbar \gamma_{\mu} \left( \langle \epsilon^{(i)}_{\mu} \rangle \rho + \rho \langle \epsilon^{(i)}_{\mu} \rangle - 2 |g^{(i)}_{\mu}| \rho |g^{(i)}_{\mu}\rangle \langle g^{(i)}_{\mu}\rangle \right) \]

(A1)

At low saturations \( \langle \epsilon^{(i)}_{\mu} \rangle \rho \epsilon^{(i)}_{\mu} \rangle \approx 0 \), therefore both the single particle and collective decays give the same result,

\[ L_A[\rho] \approx -\sum_{\mu} \hbar \gamma_{\mu} \left( b^{\dagger}_{\mu} b_{\mu} + \rho \delta_{\mu} \right) \tag{A2} \]

In addition, for x-ray quantum optics the cavities considered are in the so-called “bad cavity” regime, and the cavity mode lifetime is much shorter than the natural lifetime of the resonant transitions. Therefore, the incoherent processes are dominated by collective interactions, and the internal decay can be approximated as a small correction to the collective decay rates.

Appendix B: Multiple ground states

We now consider an atom with multiple ground states as well as multiple excited states. As with the excited states, the ground states are considered to have small splittings compared with the energy difference between the ground and excited bands, and dipole transitions are forbidden between ground states. We index here the excited states with Greek indices \( \mu, \nu \ldots \) and the ground states with Latin indices \( j \ldots \).

For nuclei with multiple ground states, we further partition the ensembles by the initial ground states of the atoms, creating permutationally invariant sub-ensembles. For a given sub-ensemble, we use the initial ground state as the ‘vacuum’ state for the generalised Holstein-Primakoff transformation. We then obtain

\[ \langle \mu | 0 \rangle \approx \sqrt{N} b^{\dagger}_{\mu} \]
\[ \langle j | 0 \rangle \approx \sqrt{N} e^{i} \]
\[ \langle j | k \rangle = e^{i} \]
\[ \langle \mu | j \rangle = b^{\dagger}_{\mu} e^{i} \]
\[ \langle \mu | \nu \rangle = b^{\dagger}_{\mu} b_{\nu} \]
\[ [b_{\mu}, b^{\dagger}_{\nu}] = \delta_{\mu\nu} \]
\[ [e_{j}, e^{i}_{k}] = \delta_{jk} \]
\[ [e_{j}, b^{\dagger}_{\mu}] = 0. \tag{B1} \]

The single particle Hamiltonian then reads

\[ H_A = \sum_{\mu} (\Delta_{\mu} - \delta_{0}) b^{\dagger}_{\mu} b_{\mu} + \sum_{j} (\delta_{j} - \delta_{0}) c^{\dagger}_{j} c_{j} \]

with the single particle decay reading

\[ L_A[\rho] = -\sum_{\mu} \gamma_{\mu j} \mathcal{L}[\rho, b^{\dagger}_{\mu}, b_{\mu}] - \sum_{\mu,j} \gamma_{\mu j} \mathcal{L}[\rho, b^{\dagger}_{\mu}, c_{j}, c^{\dagger}_{j} b_{\mu}] \]

(B3)

where \( \gamma_{\mu 0} \) is the decay rate to the initial ground state, and \( \gamma_{\mu j} \) is the decay rate to ground state \( j \). If we consider the action of this super-operator on \( b_{\mu} \), we obtain

\[ L_A[b_{\mu}] = -\gamma_{\mu 0} b_{\mu} - \sum_{j} \gamma_{\mu j} (1 + c^{\dagger}_{j} c_{j}) b_{\mu} \]

(B4)

The action on a ground state operator reads

\[ L_A[c_{j}] = \gamma_{\mu j} b^{\dagger}_{\mu} b_{\mu} c_{j} \]

(B5)

The rate of population transfer to the other ground state is thus proportional to the excited state populations, which are negligible in the linear response regime. Therefore, we can assume \( c_{j} c_{k} \approx 0 \) for all \( j, k \). However, the decay rate of the transition operator \( b_{\mu} \) is still affected by the left over terms,

\[ L_A[b_{\mu}] \approx - (\gamma_{\mu 0} + \sum_{j} \gamma_{\mu j}) b_{\mu} = - \gamma_{\mu} b_{\mu} \]

(B6)

i.e., the effective decay rate \( \gamma_{\mu} \) for a transition operator \( b_{\mu} \) is the sum of the decay rates of all decay channels for excited state \( \mu \). Thus, we may write

\[ L_A[\rho] \approx - \sum_{\mu} \gamma_{\mu} \mathcal{L}[\rho, b^{\dagger}_{\mu}, b_{\mu}] \]

(B7)

In addition to the internal decay, the cavity mediated coupling includes transitions to different ground states
than the initial. The interaction Hamiltonian reads

\[ H_{dd} = -J \sum_{\mu,\nu} d_{\mu0}^+ \cdot d_{\nu0}^+ b_\mu^+ b_\nu - \frac{J}{\sqrt{N}} \sum_{\mu,\nu} \sum_{m} d_{\mu0}^+ \cdot d_{\nu j}^+ b_\mu^+ c_\nu c_j^+ + \text{h.c.} \]

(B8)

We can see that terms involving population transfer between ground states are suppressed by a factor of \( \sqrt{N} \) or higher, and thus can be neglected in the linear response regime. An analogous argument holds for the dipole-dipole Lindblad term, and for the probe field driving.

Thus, we conclude that for the linear response in the presence of multiple ground states, only the initial ground state of a sub-ensemble needs to be considered, with the only contributions of the other ground states being to the decay rates of the transition operators.