

# Supplemental Material: Shifts of a Resonance Line in a Dense Atomic Sample

Juha Javanainen,<sup>1</sup> Janne Ruostekoski,<sup>2</sup> Yi Li,<sup>1</sup> and Sung-Mi Yoo<sup>1</sup>

<sup>1</sup>*Department of Physics, University of Connecticut, Storrs, Connecticut 06269-3046*

<sup>2</sup>*Mathematical Sciences, University of Southampton, Southampton, SO17 1BJ, UK*

In this Supplemental Material we demonstrate how the mean-field theory of the optical response of an atomic sample emerges from the complete theory of light-induced correlations. The full theoretical model is employed in the main section in the stochastic numerical simulations of the cooperative response.

## Hierarchy of correlation functions for resonant atomic sample

We will briefly review the coupled theory for light and atoms that was developed in Refs. [1, 2]. The total electric field  $\mathbf{E}(\mathbf{r})$  is expressed in terms of the atomic polarization density  $\mathbf{P}(\mathbf{r})$  by the monochromatic expression,

$$\mathbf{E}(\mathbf{r}) = \mathcal{E}_0(\mathbf{r}) + \int d^3r' \mathbf{G}(\mathbf{r} - \mathbf{r}') \mathbf{P}(\mathbf{r}'). \quad (1)$$

Here all the amplitudes  $\mathbf{E}(\mathbf{r})$ ,  $\mathcal{E}_0(\mathbf{r})$ , and  $\mathbf{P}(\mathbf{r}')$  correspond to the positive frequency components oscillating at the dominant frequency  $\omega$  of the incident light field. The incident field  $\epsilon_0 \mathcal{E}_0$  would be the electric displacement of the driving light if the matter were absent, and  $\mathbf{G}(\mathbf{r} - \mathbf{r}')$  represents the dipole field propagator, a  $3 \times 3$  matrix. The expression  $\mathbf{G}(\mathbf{r} - \mathbf{r}') \mathbf{d}$  is equal to the usual positive-frequency component of the electric field from a monochromatic dipole with the complex amplitude  $\mathbf{d}$ , given that the dipole resides at  $\mathbf{r}'$  and the field is observed at  $\mathbf{r}$  [3]. The explicit expression is

$$\begin{aligned} \mathbf{G}(\mathbf{r}) \mathbf{d} &= \frac{1}{4\pi\epsilon_0} \left\{ k^2 (\hat{\mathbf{n}} \times \mathbf{d}) \times \hat{\mathbf{n}} \frac{e^{ikr}}{r} \right. \\ &+ \left. [3\hat{\mathbf{n}}(\hat{\mathbf{n}} \cdot \mathbf{d}) - \mathbf{d}] \left( \frac{1}{r^3} - \frac{ik}{r^2} \right) e^{ikr} \right\} - \frac{\mathbf{d}}{3\epsilon_0} \delta(\mathbf{r}), \quad (2) \end{aligned}$$

with  $\hat{\mathbf{n}} = \mathbf{r}/r$  and  $k = \omega/c$ . The integral in Eq. (1) is not convergent at the origin. The expression (2) should be understood in such a way that the integral of the term inside the braces over an infinitesimal sphere enclosing the origin vanishes [1]. The delta function part then ensures that the Gauss law in a volume enclosing the origin is satisfied [3].

The expression (1) is the integral representation of Maxwell's equations for the electric field amplitude in an atomic medium. Although it provides the scattered field in terms of the atomic polarization, there is general no simple way of finding  $\mathbf{P}(\mathbf{r}')$ . In order to solve the coupled theory for light and matter we derived from quantum field theoretical formalism a hierarchy of equations of motion

for the correlation functions of atomic density and polarization. In the limit of low light intensity, these involve correlation functions  $\rho_p(\mathbf{r}_1, \dots, \mathbf{r}_p)$  for the ground state atomic positions at points  $\mathbf{r}_1, \dots, \mathbf{r}_p$  and correlation functions for ground state atoms at  $\mathbf{r}_1, \dots, \mathbf{r}_{p-1}$ , given that there is polarization at  $\mathbf{r}_p$ ,  $\mathbf{P}_p(\mathbf{r}_1, \dots, \mathbf{r}_{p-1}; \mathbf{r}_p)$ . For a  $J = 0 \rightarrow J' = 1$  atomic transition the response of the medium is isotropic, and we have a hierarchy of equations of motion for the positive frequency components of the correlation functions

$$\begin{aligned} \dot{\mathbf{P}}_p(\mathbf{r}_1, \dots, \mathbf{r}_{p-1}; \mathbf{r}_p) &= \\ (i\Delta - \gamma) \mathbf{P}_p(\mathbf{r}_1, \dots, \mathbf{r}_{p-1}; \mathbf{r}_p) &+ i\zeta \mathcal{E}_0(\mathbf{r}_p) \rho_k(\mathbf{r}_1, \dots, \mathbf{r}_p) \\ + i\zeta \sum_{q \neq p} \mathbf{G}(\mathbf{r}_p - \mathbf{r}_q) \mathbf{P}_p(\mathbf{r}_1, \dots, \mathbf{r}_{q-1}, \mathbf{r}_{q+1}, \dots, \mathbf{r}_p; \mathbf{r}_q) & \\ + i\zeta \int d^3r_{p+1} \mathbf{G}(\mathbf{r}_p - \mathbf{r}_{p+1}) \mathbf{P}_{p+1}(\mathbf{r}_1, \dots, \mathbf{r}_p; \mathbf{r}_{p+1}), & \quad (3) \end{aligned}$$

with  $p = 1, 2, \dots$ . Here  $\Delta = \omega - \omega_0$  is the detuning from the atomic resonance  $\omega_0$ ,  $\gamma$  is the HWHM linewidth of the transition,  $\zeta = \mathcal{D}^2/\hbar$ , and  $\mathcal{D}$  is the dipole moment matrix element.

The peculiar feature of Eq. (3) is the third line. It describes  $p$  atoms interacting with each other via the dipole-dipole interaction. The light mediates interactions in which photons are repeatedly exchanged by the atoms at positions  $\mathbf{r}_1, \dots, \mathbf{r}_p$ , corresponding to *recurrent* scattering events. Such processes are responsible for subradiant and superradiant resonances.

The full set of equations (3) can be solved by stochastic classical-electrodynamics simulations exactly, apart from statistical fluctuations [4]. In each stochastic realization we sample the positions of the atomic dipoles so that they have the proper correlation functions  $\rho_p(\mathbf{r}_1, \dots, \mathbf{r}_p)$ , and then solve the optical response for the given set of atomic positions from Eqs. (3) and (4) of the main text. Average over many realizations will give, say, the electric field that would be obtained by substituting the exact solution of Eqs. (3) to the right-hand side of Eq. (1). In the main text this is implemented for uncorrelated positions of the atoms in a disk-shaped container.

## Mean-field response

The lowest-order equation in the hierarchy (3) represents the equation of motion for the polarization and

reads

$$\begin{aligned} \dot{\mathbf{P}}(\mathbf{r}_1) &= (i\Delta - \gamma)\mathbf{P}(\mathbf{r}_1) + i\zeta\rho(\mathbf{r}_1)\boldsymbol{\mathcal{E}}_0(\mathbf{r}_1) \\ &+ i\zeta \int d^3r_2 \mathbf{G}(\mathbf{r}_1 - \mathbf{r}_2)\mathbf{P}_2(\mathbf{r}_1; \mathbf{r}_2). \end{aligned} \quad (4)$$

Suppose we factorize here the lowest nontrivial correlation function  $\mathbf{P}_2(\mathbf{r}_1; \mathbf{r}_2)$  as

$$\mathbf{P}_2(\mathbf{r}_1; \mathbf{r}_2) = \rho(\mathbf{r}_1)\mathbf{P}(\mathbf{r}_2), \quad (5)$$

ignoring any light-induced correlations between the ground state atom at  $\mathbf{r}_1$  and the polarization at  $\mathbf{r}_2$ . The polarization may be solved from Eqs. (4) and (5), and the scattered light is then obtained from Eq. (1). This represents the mean-field response of the system.

In this work we compare the full numerical solution of the stochastic simulations, which represents the solution of Eqs. (3), with the mean-field response. In order to derive the mean-field solution we assume an infinite slab of atoms with a uniform density and thickness  $h$ , such that the atoms fill the region  $z \in [0, h]$ . We illuminate the sample by an incident plane wave  $\boldsymbol{\mathcal{E}}_0(\mathbf{r}) = \mathcal{E}_0 \hat{\mathbf{e}} \exp(ikz)$ , where  $\hat{\mathbf{e}}$  denotes the polarization unit vector for light. Equation for the scattered light (1), together with the stationary mean-field equation for the polarization,

$$\mathbf{P}(\mathbf{r}) = \alpha\rho(\mathbf{r})\boldsymbol{\mathcal{E}}_0(\mathbf{r}) + \alpha\rho(\mathbf{r}) \int d^3r' \mathbf{G}(\mathbf{r} - \mathbf{r}')\mathbf{P}(\mathbf{r}'), \quad (6)$$

where  $\alpha = -\mathcal{D}^2/[\hbar(\Delta + i\gamma)]$  denotes the atomic polarizability, can then be solved exactly by substituting  $\mathbf{P}(\mathbf{r}) = P_+ \hat{\mathbf{e}} \exp(ik'z) + P_- \hat{\mathbf{e}} \exp(-ik'z)$ , for  $0 \leq z \leq h$ , where the complex  $k'$  [ $\text{Im}(k') > 0$ ] represents a damped plane wave. Specifically the transmission properties of light can be obtained by solving for the electric field amplitude  $\mathbf{E}(\mathbf{r}) = E_T \hat{\mathbf{e}} \exp(ikz)$ , for  $z > h$ . In this way, we find an analytic low-density expression for the resonance shift for the transmitted light [5]

$$\Delta_L = \Delta_{LL} - \frac{3}{4}\Delta_{LL} \left(1 - \frac{\sin 2hk}{2hk}\right); \quad \Delta_{LL} = -\frac{\rho\mathcal{D}^2}{3\epsilon_0 \hbar}. \quad (7)$$

There is an alternative approach to derive the mean-field result that further illustrates the role of recurrent

scattering. This argument is very similar to the one we used in the derivation of the Lorentz-Lorenz shift in Ref. [2]. We assume that the ground state atoms in the absence of light are initially uncorrelated, such that  $\rho_p(\mathbf{r}_1, \dots, \mathbf{r}_p) = \rho_1(\mathbf{r}_1) \dots \rho_1(\mathbf{r}_p)$ . Suppose we keep all the equations of the hierarchy (3), but neglect the recurrent scattering terms, i.e., the third line of Eq. (3) for each  $p = 1, 2, \dots$ . Then the steady-state solution is obtained by the following set of damped plane waves

$$\begin{aligned} &\mathbf{P}_p(\mathbf{r}_1, \dots, \mathbf{r}_p) \\ &= \begin{cases} \rho^{p-1} \hat{\mathbf{e}} (P_+ e^{ik'z_p} + P_- e^{-ik'z_p}), & 0 \leq z_1, \dots, z_p \leq h; \\ 0, & \text{otherwise,} \end{cases} \end{aligned} \quad (8)$$

With this substitution every steady-state equation in the hierarchy is effectively reduced to Eq. (6), and we find the same mean-field Lorentz-Lorenz shift and the collective Lamb shift as above.

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