Cooperative decay rate and cooperative Lamb shift from a nanospherical configuration of two adjoining atomic species

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The eigenmodes of a nanosphere configuration where the core is an ensemble of two-level atoms with resonance frequency ω_C, and the shell is an ensemble of two-level atoms but with shifted resonant frequency ω_B are obtained for different values of the ratio of the core radius to the shell outer radius as function of $(\omega_C - \omega_B)$. I show that the eigenmodes belong to one of two branches. The dominant decay mode for superradiant emission may belong to either branch, depending on the system's parameters. In instances of configurations where the cooperative decay rate values for the two branches become equal, the cooperative exponential amplification (decay) is accompanied by temporal oscillations.

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I. INTRODUCTION

Since the seminal work of Dicke [1] on superradiance it has been widely assumed, with rare dissent [2], that the cooperative decay rate (CDR) of an ensemble of identical two-level atoms occupying any geometrical shape with dimensions much smaller than the wavelength of the transition radiation is simply the product of the number of particles in the ensemble multiplied by the isolated atom decay rate. As recently as 2010, Prasad and Glauber [3] critiqued the original Friedberg, Hartmann, and Manassah paper [2], which took exception to that prevailing view.

In Refs. [4,5], responding to the critique of Ref. [3] and the authors' concurrent desire to explore some interesting aspects of nanoscale electrodynamics, R. Friedberg and the author considered the same spherical geometry used in Ref. [3] but assumed, instead of the uniform distribution of the resonant atoms in the sphere, a varying radial density. They obtained approximate analytical results, confirmed by numerical calculations, which give non-Dicke rates for the CDR of the different cases considered, as they asserted that they would earlier in Ref. [2]. In addition to resolving the original issue of contention on the value of the CDR from a small sample with nonuniform density, the spherical eigenmode expansion used in Refs. [4,5] to compute the CDR and the cooperative Lamb shift (CLS) allowed these authors to obtain in a direct manner the plasmonics eigenmodes for different internal configurations of metallic nanospheres [6,7], without having to restrict derivation to the electrostatic approximation. This same method proved able, as well, to predict what was called the Dicke-Purcell effect, i.e., the enhancement of the CDR for an ensemble of atoms that are enclosed in a metallic nanoshell [8,9].

In this Brief Report, I use the same mathematical technique to analyze superradiance from a system consisting of two species of atoms with nonequal but close resonance frequencies. The configuration that I shall consider consists of a nanospherical structure, where one of the species fills the sphere core, while the other species occupies the surrounding outer shell. I compute the eigenmodes for this combined system and show that its solutions belong to one of two distinct branches. I show that, for a given geometry depending on the detuning between the two atomic resonant frequencies, the dominant mode may belong to one or the other branch. The possible crossing in the value of the real part of the system's two principal eigenmodes branches, for certain configurations, implies that a fast oscillatory time development will develop within the superradiant emission time temporal exponential envelope rise (decay) of an inverted (weakly excited) system.

This report is organized as follows. In Sec. II, I shall review the constitutive equations for each region of space and derive the expression relating the wave vector in the core and shell regions. In Sec. III, I review the form of the eigenmodes in spherical configuration, the boundary conditions, and the secular determinant relating the wave number of the different regions. In Sec. IV, the equations obtained by combining the constitutive equations and the secular determinant resulting from the boundary conditions are simultaneously solved, for some special values of the ratio of the core radius to the outer radius of the shell and the normalized detuning between the two species resonant frequencies to obtain the CDR and CLS for this system. I conclude in Sec. V.

II. CONSTITUTIVE EQUATIONS

The Gaussian units will be used throughout. The three regions of space shall be designated by the letters A, B, C from the outside in, thus,

$$\text{A passive dielectric } r \geqslant R, \tag{1}$$

$$\text{B species B } \beta R \leqslant r \leqslant R, \tag{2}$$

$$\text{C species C } r \leqslant \beta R, \tag{3}$$

where $0 \leqslant \beta \leqslant 1$. Thus R is the outer radius of the B species shell, and βR is its inner radius.

The eigenmodes of Maxwell's equations in spherical geometry [10] are designated by angular indices l,m (corresponding to the spherical harmonics Y_l^m) and a radial index s as well as a binary choice (E, M), where E and M refer respectively to the electric and magnetic modes. As the sphere is very small, we limit our attention to the cylindrically symmetric electric

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dipole modes, $E_{l,m,s} = E_{1,0,s}$. Accordingly we shall suppress the subscripts $l = 1$, $m = 0$ and keep only the subscript s. Hence the common (complex) eigenfrequency for a decay mode will be called ω_s and the associated wave numbers $k_s^{\rm A}, k_s^{\rm B}, k_s^{\rm C}$. The resonant frequency of the active atoms in the core (when isolated) is $\omega_{\rm C}$, while that in the shell is $\omega_{\rm B}$, and the corresponding wave number in the vacuum is $k_0 = \omega_{\rm C}/c$.

In each region there is a constitutive equation, specific to the material occupying that region, that links the wave number in that material to the complex eigenfrequency. In region A, (the passive dielectric) one has simply

$$\left(k_s^{\rm A}\right)^2 = \varepsilon^{\rm A}\,\omega_s^2/c^2 = n^2\omega_s^2/c^2, \tag{4}$$

where $\varepsilon^{\rm A}$ is a fixed (real) number, independent of the mode.

In region B we have

$$\left(k_s^{\rm B}\right)^2 = \varepsilon_s^{\rm B}(\omega_s)\omega_s^2/c^2, \tag{5}$$

where $\varepsilon_s^{\rm B}$ is given by the one-pole formula

$$\varepsilon_s^{\rm B}(\omega_s) = \varepsilon_\infty^{\rm B} - \frac{\mathbf{C}_{\rm B}}{\omega_s - \omega_{\rm B} + \omega_{L,{\rm B}} + i\gamma_{\rm B}}, \tag{6}$$

where $\varepsilon_\infty^{\rm B}$ is the background dielectric constant in region B, and the other quantities are defined following Eq. (8).

In region C we have

$$\left(k_s^{\rm C}\right)^2 = \varepsilon_s^{\rm C}(\omega_s)\,\omega_s^2/c^2, \tag{7}$$

where $\varepsilon_s^{\rm C}(\omega_s)$ is also given by a one pole formula

$$\varepsilon_s^{\rm C}(\omega_s) = 1 - \frac{\mathbf{C}_{\rm C}}{\omega_s - \omega_{\rm C} + \omega_{L,{\rm C}} + i\gamma_{\rm C}}. \tag{8}$$

Here $\mathbf{C}_{\rm C} = 4\pi\wp_{\rm C}^2 n_{\rm C}/\hbar$, $n_{\rm C}$ is the atomic number density in region C, $\wp_C$ is the reduced dipole matrix element for the two-level transition of species C, $\omega_{L,{\rm C}} = \frac{1}{3}\,\mathbf{C}_{\rm C}$ is the Lorentz shift due to the Clausius-Mossotti local field correction, and $\gamma_{\rm C}$ is the collisional half-width of the line ($\gamma_{\rm C} \cong 0.6\,\mathbf{C}_{\rm C}$) [11]. (Corresponding expressions for the region B hold, with C replaced by B.)

It is to be noted that the Kramers-Kronig relation that requires only causality for its validity allows the susceptibility and consequently the dielectric function to be analytically continued in the lower half of the complex-frequency plane. This allows us to use the same functional form of $\varepsilon^{\rm B}$ and $\varepsilon^{\rm C}$ for complex ω_s as the familiar one for real ω.

Before proceeding further, I shall replace the wave numbers by dimensionless equivalents: $u_0 = k_0 R$, $u_s = k_s^{\rm A} R$, $v_s = k_s^{\rm B} R$, $w_s = k_s^{\rm C} R$, and introduce the normalized quantities: $\Omega_s = \omega_s/\mathbf{C}_{\rm C}, \Gamma_{\rm C} = \gamma_{\rm C}/\mathbf{C}_{\rm C}$, $\Omega_{\rm C} = \omega_{\rm C}/\mathbf{C}_{\rm C}$. The normalized eigenfrequency can be written

$$\Omega_s = \Omega_{\rm C} - i\Lambda_s, \tag{9a}$$

where

$$-i\Lambda_s = \frac{u_0^2}{u_0^2 - w_s^2} - \frac{1}{3} - i\Gamma_{\rm C}. \tag{9b}$$

Introducing $\Omega_{\rm B} = \omega_{\rm B}/\mathbf{C}_{\rm C}$, $\Delta = \Omega_{\rm C} - \Omega_{\rm B}$, $\Re = \mathbf{C}_{\rm B}/\mathbf{C}_{\rm C}$, $\Gamma_{\rm B} = \gamma_{\rm B}/\mathbf{C}_{\rm C}$ and substituting in Eq. (6), one finds the first equation relating w_s and v_s,

$$v_s = u_0\left[\varepsilon_\infty^{\rm B} - \frac{\Re}{\Delta - i\,\Lambda_s + \frac{1}{3}\Re + i\Gamma_B}\right]^{1/2}. \tag{10}$$

In the next section, using the boundary conditions of the Maxwell fields, I shall find the other equation relating w_s and v_s. This equation will include the geometric parameters of the system's configuration.

III. EIGENMODES IN SPHERICAL CONFIGURATION

The coupling between species B and species C is that resulting from imposing Mawell fields boundary conditions at the different interfaces.

A. Fields and boundary conditions

For a cylindrically symmetric dipole mode $E_{1,0,s}$, the expressions for $\vec{B}$ and $\vec{E}$ in any one of the three regions depend on two constant coefficients specific to that region, but otherwise have the same form in each region. Letting F_i stand for A_i, B_i, or C_i in each respective region, where $i = 1$ or 2, we have [11]

$$\vec{\mathbf{B}}(r,\theta,\phi) = \left[F_1 j_1\left(k_s^{\rm F} r\right) + F_2 n_1\left(k_s^{\rm F} r\right)\right] P_l^1(\cos(\theta))\hat{e}_\phi \tag{11}$$

and

$$\begin{aligned}\vec{\mathbf{E}}(r,\theta,\phi) = -\frac{ik_0}{\left(k_s^{\rm F}\right)^2 r}\Big[&2\left[F_1 j_1\left(k_s^{\rm F} r\right) + F_2 n_1\left(k_s^{\rm F} r\right)\right]P_1(\cos(\theta))\hat{e}_r \\ &+ \left\{F_1\left[\left(k_s^{\rm F} r\right) j_0\left(k_s^{\rm F} r\right) - j_1\left(k_s^{\rm F} r\right)\right]\right. \\ &\left.+ F_2\left[\left(k_s^{\rm F} r\right) n_0\left(k_s^{\rm F} r\right) - n_1\left(k_s^{\rm F} r\right)\right]\right\} P_1^1(\cos(\theta))\hat{e}_\theta\Big],\end{aligned} \tag{12}$$

where $P_1(\cos(\theta)) = \cos(\theta)$ and $P_1^1(\cos(\theta)) = -\sin(\theta)$ are Legendre polynomial and associated Legendre function, and j_0, j_1 and n_0, n_1 are respectively zeroth- and first-order spherical Bessel and Neumann functions.

To avoid a singularity at $r = 0$, we must have

$$C_2 = 0 \tag{13}$$

and to have a pure outgoing wave in $r > R$, we must have

$$A_2 = iA_1. \tag{14}$$

In addition, Maxwell's equations require continuity of B_ϕ and E_θ at the boundaries $r = \beta R$ and $r = R$

$$C_1 j_1\left(k_s^{\rm C}\beta R\right) = B_1 j_1\left(k_s^{\rm B}\beta R\right) + B_2 n_1\left(k_s^{\rm B}\beta R\right), \tag{15}$$

$$\begin{aligned}&\left(k_s^{\rm C}\beta R\right)^{-2} C_1\left[k_s^{\rm C}\beta R j_0\left(k_s^{\rm C}\beta R\right) - j_1\left(k_s^{\rm C}\beta R\right)\right] \\ &\quad= \left(k_s^{\rm B}\beta R\right)^{-2}\left\{B_1\left[k_s^{\rm B}\beta R j_0\left(k_s^{\rm B}\beta R\right) - j_1\left(k_s^{\rm B}\beta R\right)\right]\right. \\ &\qquad\left.+ B_2\left[k_s^{\rm B}\beta R n_0\left(k_s^{\rm B}\beta R\right) - n_1\left(k_s^{\rm B}\beta R\right)\right]\right\}\end{aligned} \tag{16}$$

and

$$B_1 j_1\left(k_s^{\rm B} R\right) + B_2 n_1\left(k_s^{\rm B} R\right) = A_1 h_1^{(1)}\left(k_s^{\rm A} R\right), \tag{17}$$

$$\begin{aligned}&\left(k_s^{\rm B} R\right)^{-2}\left\{B_1\left[k_s^{\rm B} R j_0\left(k_s^{\rm B} R\right) - j_1\left(k_s^{\rm B} R\right)\right]\right. \\ &\quad\left.+ B_2\left[k_s^{\rm B} R n_0\left(k_s^{\rm B} R\right) - n_1\left(k_s^{\rm B} R\right)\right]\right\} \\ &\qquad= \left(k_s^{\rm A} R\right)^{-2} A_1\left[k_s^{\rm A} R h_0^{(1)}\left(k_s^{\rm A} R\right) - h_1^{(1)}\left(k_s^{\rm A} R\right)\right],\end{aligned} \tag{18}$$

where $h_l^{(1)}(q) = j_l(q) + i n_l(q)$.

B. Secular determinant

Equations (15)–(18) are a set of four linear homogeneous equations in the four unknowns A_1, B_1, B_2, C_1. For a solution to exist, the characteristic secular determinant for this system must vanish

$$\det\begin{pmatrix} j_1(w_s\beta) & -j_1(v_s\beta) & -n_1(v_s\beta) & 0 \\ v_s^2 j_V(w_s\beta) & -w_s^2 j_V(v_s\beta) & -w_s^2 n_V(v_s\beta) & 0 \\ 0 & j_1(v_s) & n_1(v_s) & -h_1^{(1)}(nu_0) \\ 0 & (nu_0)^2 j_V(v_s) & (nu_0)^2 n_V(v_s) & -v_s^2 h_V^{(1)}(nu_0) \end{pmatrix} = 0, \quad (19)$$

where $j_V(q) = q j_0(q) - j_1(q)$, and likewise for $n_V(q)$ and $h_V^{(1)}(q)$. Finding w_s and v_s reduces to simultaneously solving Eqs. (10) and (19).

Having computed $v_s = k_s^{\mathrm{B}} R$, $w_s = k_s^{\mathrm{C}} R$ as function of u_0 and the physical parameters, the quantities B_1/C_1, B_2/C_1, A_1/C_1 are uniquely determined by solving

$$\begin{pmatrix} j_1(v_s\beta) & n_1(v_s\beta) & 0 \\ j_1(v_s) & n_1(v_s) & -h_1^{(1)}(nu_0) \\ (nu_0)^2 j_V(v_s) & (nu_0)^2 n_V(v_s) & -(v_s)^2 h_V^{(1)}(nu_0) \end{pmatrix}\begin{pmatrix} B_1/C_1 \\ B_2/C_1 \\ A_1/C_1 \end{pmatrix} = \begin{pmatrix} j_1(w_s\beta) \\ 0 \\ 0 \end{pmatrix}. \quad (20)$$

IV. RESULTS

For a small sphere with single species uniform resonant atomic density, embedded in a passive dielectric with index of refraction n, the Dicke value of the CDR [12–14] is

$$\mathrm{Re}(\tilde{\Lambda}_{\mathrm{Dicke}}) \approx \frac{2n^5}{(1+2n^2)^2} u_0^3, \quad (21)$$

where $\tilde{\Lambda} = \Lambda - \Gamma$. In Dicke's theory, $\mathrm{Im}(\Lambda_{\mathrm{Dicke}}) = 0$.

I shall consider in the illustrations, the following values for the different parameters $u_0 = 0.2$, $n = 1$, $\Re = 1$, $\Gamma_{\mathrm{B}} = \Gamma_{\mathrm{C}}$, and plot the wave vector in region B, and Λ_s as function of the normalized detuning Δ for different values of β. Solving simultaneously Eqs. (10) and (19), I obtain two primary modes.

In Figs. 1 and 2, I plot the values of the wave vector in region B and Λ_s (where $\tilde{\Lambda}_s = \Lambda_s - \Gamma_{\mathrm{C}}$) as function of Δ, for $\beta = 0.8$. I denote by the subscript (1), the mode which at $\Delta = 0$ (i.e., only one species is present) has $\mathrm{Re}(\tilde{\Lambda}_{(1)}(\Delta = 0)) \approx \frac{2}{9} u_0^3$, and by the subscript (2), the mode which at $\Delta = \infty$ (i.e., the resonance frequency of the B species is far enough from the resonance frequency of the C species that no interaction between the species is present) has $\mathrm{Re}(\tilde{\Lambda}_{(2)}(\Delta = \infty)) \approx \frac{2}{9}\beta^3 u_0^3$, i.e., only the atoms in the core are cooperatively emitting.

In Fig. 3, I plot on the same panel the traces of both $\mathrm{Re}(\tilde{\Lambda}_{(1)})$ and $\mathrm{Re}(\tilde{\Lambda}_{(2)})$ as function of Δ, for different values of β. This permits us to determine the value of Δ_{int}, defined as $\mathrm{Re}(\tilde{\Lambda}_{(1)}(\Delta_{\mathrm{int}})) = \mathrm{Re}(\tilde{\Lambda}_{(2)}(\Delta_{\mathrm{int}}))$. As it can be observed that for small values of β, the two traces do not intersect.

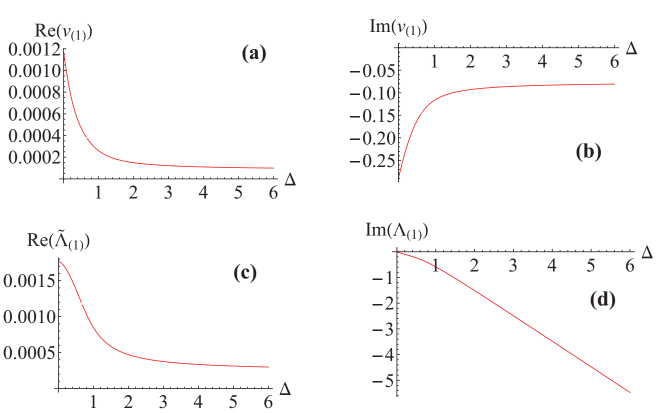

FIG. 1. (Color online) The variables associated with the first branch of the eigenmodes for $\beta = 0.8$: (a) The real and (b) the imaginary parts of the normalized wave vector in the shell; (c) the normalized cooperative decay rate, and (d) the normalized cooperative Lamb shift are plotted as functions of the detuning between the normalized resonant frequencies of the two species. $u_0 = 0.2, n = 1, \varepsilon_\infty^{\mathrm{B}} = 1, \Gamma_{\mathrm{B}} = \Gamma_{\mathrm{C}} = 0.6$.

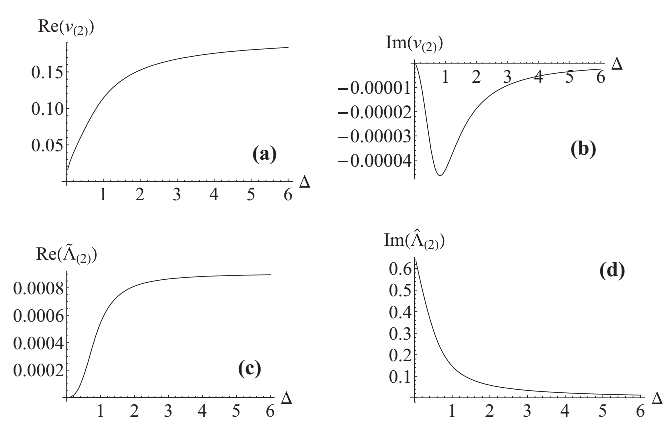

FIG. 2. The variables associated with the second branch of the eigenmodes for $\beta = 0.8$: (a) The real and (b) the imaginary parts of the normalized wave vector in the shell; (c) the normalized cooperative decay rate, and (d) the normalized cooperative Lamb shift are plotted as functions of the detuning between the normalized resonant frequencies of the two species. $u_0 = 0.2$, $n = 1$, $\varepsilon_\infty^{\mathrm{B}} = 1$, $\Gamma_{\mathrm{B}} = \Gamma_{\mathrm{C}} = 0.6$.

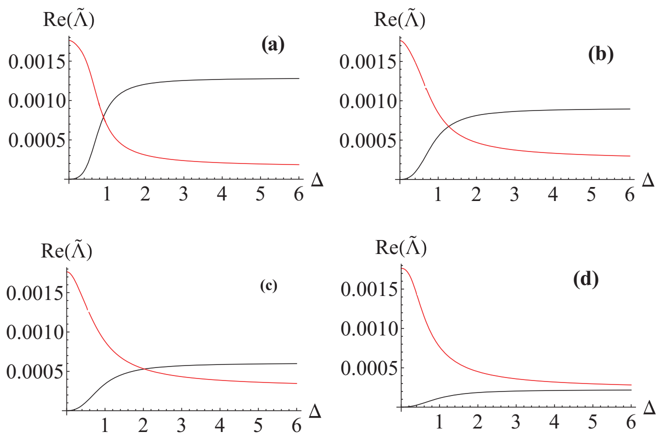

FIG. 3. (Color online) The two branches of the cooperative decay rate are plotted as function of the detuning between the normalized resonant frequencies of the two species for different values of β. $u_0 = 0.2$, $n = 1$, $\varepsilon_\infty^B = 1$, $\Gamma_B = \Gamma_C = 0.6$. (a) $\beta = 0.9$, (b) $\beta = 0.8$, (c) $\beta = 0.7$, (d) $\beta = 0.5$.

The values of Δ_{int}, $\text{Re}(\tilde{\Lambda}(\Delta_{\text{int}}))$, $\text{Im}(\tilde{\Lambda}_{(1)}(\Delta_{\text{int}})) - \text{Im}(\tilde{\Lambda}_{(2)}(\Delta_{\text{int}}))$ for $\beta = 0.9, 0.8, 0.7$ for the curves of Fig. 3 are given respectively:

(i) For $\beta = 0.9$, $\Delta_{\text{int}} = 0.902$, $\text{Re}(\tilde{\Lambda}(\Delta_{\text{int}})) = 0.0007903$, $\text{Im}(\tilde{\Lambda}_{(1)}(\Delta_{\text{int}})) - \text{Im}(\tilde{\Lambda}_{(2)}(\Delta_{\text{int}})) = -0.5129$

(ii) For $\beta = 0.8$, $\Delta_{\text{int}} = 1.275$, $\text{Re}(\tilde{\Lambda}(\Delta_{\text{int}})) = 0.0006738$, $\text{Im}(\tilde{\Lambda}_{(1)}(\Delta_{\text{int}})) - \text{Im}(\tilde{\Lambda}_{(2)}(\Delta_{\text{int}})) = -0.9354$

(iii) For $\beta = 0.7$, $\Delta_{\text{int}} = 2.027$, $\text{Re}(\tilde{\Lambda}(\Delta_{\text{int}})) = 0.0005296$, $\text{Im}(\tilde{\Lambda}_{(1)}(\Delta_{\text{int}})) - \text{Im}(\tilde{\Lambda}_{(2)}(\Delta_{\text{int}})) = -1.6877$.

For the above parameters: $\text{Re}(\tilde{\Lambda}_{\text{Dicke}}) = \text{Re}(\tilde{\Lambda}_{(1)}(\Delta = 0)) = 0.0017635$.

The value of $\text{Im}(\tilde{\Lambda}_{(1)}(\Delta_{\text{int}})) - \text{Im}(\tilde{\Lambda}_{(2)}(\Delta_{\text{int}}))$ determines the frequency of oscillations within the increasing (decreasing) exponential temporal envelope for emission for an initially inverted (weakly excited) system.

V. CONCLUSION

The mathematical steps previously used to compute the values of the CDR for a small sphere with a varying radial density of the resonant atoms; the Purcell-Dicke enhanced CDR of resonant atoms enclosed in a metallic shell; and the plasmonics eigenfrequencies for different metallodielectric configurations can be repeated to compute the CDR of a system consisting of two species with detuned resonant frequencies but spatially in contact.

The present approach provides a natural way to incorporate the interaction between the two species by requiring that the boundary conditions for the Maxwell fields at the interface between the two materials be satisfied.

The two branches of solutions obtained describe the mode of transition of the dominant mode from one branch of the solutions to the other. The computations summarized in this report give the value of the detuning at the intersection of the different eigenmodes CDR curves, the value of the CDR there, and the difference in the frequencies of the resonant modes frequencies at the same point.

Although the present Brief Report analyzed only the details of a spherical configuration, the methods used here can be generalized to different geometries. The observed eigenmode splitting and the strong correlation in the cooperative emission from atoms of two samples having different resonance frequencies are expected to be general features for values of the detuning not exceeding few cooperative Lamb shifts. The specific parameters leading to the crossover in the cooperative decay rates of the different modes is expected to be geometry dependent.

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