Collective dipole-dipole interactions in planar nanocavities

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The collective response of an atomic ensemble is shaped by its macroscopic environment. We demonstrate this effect in the near-resonant transmission of light through a thermal rubidium vapor confined in a planar nanocavity. Our model reveals density-dependent line shifts and broadenings beyond continuous electrodynamics models that oscillate with cavity width and have been observed in recent experiments. We predict that the amplitudes of these oscillations can be controlled by coatings that modify the cavity’s finesse.

When a dense ensemble of quantum emitters is interrogated by near-resonant light, strong dipole-dipole interactions cause a collective response of the system and alter its spectroscopic properties [1–3]. Collective behavior has been studied intensively to build enhanced light-matter interfaces such as a perfect mirror out of a single layer of atoms [4, 5] or to control unwanted effects such as collective line shifts that can bias optical clocks [6–8]. In the past two decades, novel nanophotonic light-matter interfaces were developed [9] that offer new handles to study and tune collective interactions — especially with respect to two parameters: emitter geometry [10, 11] and modification of the local density of states by cavities, waveguides and photonic crystals [12–14].

A prominent example for the geometry-dependence of collective effects is the collective Lamb shift (CLS). Friedberg et al. [10] predicted that a continuous slab of atoms of thickness $d$ in free space exhibits a line shift

$$\Delta_{\text{CLS}} = \Delta_{\text{LL}} - \frac{3\Delta_{\text{LL}}}{4} \left(1 - \frac{\sin(2kd)}{2kd}\right). \tag{1}$$

Here, $\Delta_{\text{LL}} = -\frac{N\Delta_{\text{d}}^2}{3\epsilon_0\hbar(2J_\ell+1)}$ is the Lorentz–Lorenz shift that occurs in bulk media, where $N$ is the atomic density, $\Delta_{\text{d}}$ is the transition dipole moment and $(2J_\ell+1)$ is the number of (degenerate) ground states. Despite its name, the collective Lamb shift originates from classical single scattering events (see Refs. [15, 16] and below). The CLS has been measured in x-ray scattering from iron layers in a planar cavity [17], whereas cold atomic vapor experiments either found a shift compatible with a CLS prediction [18], or an altogether negligible shift [19–21].

Recently, experiments studied the CLS in wedged nano-cells [11, 16] containing a thermal atomic vapor layer of varying thickness $d$ between two planar walls. This confinement intertwines the sample geometry with the cavity properties and complicates the evaluation of experimental results. Although an earlier study [11] seemed to conform with Eq. (1), an improved analysis revealed that it does not extend to vapors in cavities and concluded the agreement to be fortuitous [16]. Instead, the latter study found that the transmission through the vapor-filled nano-cavity can be described by a continuous medium model with an additional broadening and shift that themselves depend on density and cavity width.

In this paper, we present a generalized microscopic interaction model that consistently accounts for the modified non-collisional atom-atom and atom-wall interactions in a planar nanocavity environment. When treating the atomic vapor as a continuous medium, we retrieve the well-known transmission profiles of a Fabry–Perot etalon. When solving our model for discrete ensembles of thermal atoms, the resulting transmission spectra can be fitted to a continuous medium model with an additional collective broadening and shift that compare well to the experimental observations in Ref. [16]. We find an oscillatory dependence of the collective broadening and shift on the cavity width. The amplitudes of these oscillations can be tuned by cavity coatings that increase or decrease the cavity’s finesse. This prediction offers a feasible approach for experimental tests. Further, such a test also has implications on collective interactions in other systems of different shape, dimension, or local density of states as the Green’s formalism employed here can be adapted to any macroscopic environment.

The scattering of light in atomic samples is most conveniently described in a Green’s function formalism, where

FIG. 1. (a) Free-space (solid blue line) and cavity-mediated (solid red line) atom-atom and atom-wall interactions (dashed lines). (b) Sketch of the setup: Incident light is scattered by atoms in a cavity of width $d$ and focused onto a detector by a lens of radius $R$. 

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the classical Green’s tensor $G(r_1, r_2, \omega)$ is simply the electric field at $r_1$ radiated by an electric dipole $d$ at $r_2$, viz.
\[ E(r_1, \omega) = \frac{k}{i\omega} G(r_1, r_2, \omega) d(r_2, \omega) \]. The Green’s tensor consists of a free-space and a cavity contribution,
\[ G(r_1, r_2, \omega) = G_{\text{free}}(r_1, r_2, \omega) + G_{\text{cav}}(r_1, r_2, \omega). \] (2)

Explicit expressions are provided in the Supplemental Material. If a low-intensity field $E_{\text{inc}}$ impinges onto an atomic ensemble, the steady-state response follows the classical coupled dipole model (see Supplemental Material)
\[ d_i = \alpha_i E_{\text{inc}}(r_i) + \alpha_i \sum_{j \neq i} \frac{k^2}{\epsilon_0} G(r_i, r_j, \omega_0) d_j, \] (3)

where $\omega_0$ denotes the single-atom resonance frequency in vacuum. The dipole moment of atom $i$ is created by the sum of the incident field in an empty cavity, and the field scattered from all other dipoles either directly or via the cavity (see Fig. 1a).

Without loss of generality, we take a plane wave polarized along $x$ as input field. For near-resonant light, the atomic polarizability is well described as a system with two energy levels $g, e$ with degenerate substates $\mu, \nu$
\[ \alpha_i = -\frac{1}{\hbar} \sum_{\mu, \nu} \frac{f_{g\mu} d_{\mu e} \otimes d_{\nu g}}{\delta - \Delta_{\mu \nu}(r_i) + \frac{1}{2} \Gamma_{\nu}^e(r_i)}, \] (4)

with ground-state population number $f_{g\mu}$, detuning $\delta = \omega - \omega_0$, total line shift $\Delta_{\mu \nu}(r) = \omega_D + \omega_{\text{CP}}(r)$ including the Doppler shift $\omega_D = kv$, and the Purcell-enhanced emission rate $\Gamma_{\nu}^e(r)$ that depends on the dipole’s position and orientation. The modified emission rate is accompanied by a line shift $\omega_{\text{CP}}(r)$ deriving from the Casimir–Polder interaction between atoms and cavity walls [22, 23]. We only include the fine structure of the atoms. However, Eq. (4) can be straightforwardly extended to include hyperfine states as well [20].

From the dipole moment in Eq. (3), we compute the transmission profile of the sample. Adopting a method established in free space [24], we consider a lens of radius $R$ (see Fig. 1b), operating in the Fraunhofer limit, that focuses the forward scattered light onto a detector. The resulting transmission coefficient is
\[ t = t_0 + \int \frac{k^2}{\epsilon_0} f_{\text{inc}} dA j G_{\text{cav,out}, xx}(z, r_j) d_{x, j} \]
\[ = t_0 + \frac{2i k e^{i \Delta_k} t_{21}}{4 \pi \epsilon_0 R^2} \sum_j \frac{e^{-ikz_j} + r_{21}e^{ikz_j}}{1 - r_{21}^2 e^{2ikd}} \frac{d_{x, j}}{E_0}, \] (5)

where $t_0$ is the transmission through an empty cavity, $t_{12}, r_{21}$ are the Fresnel coefficients at the interfaces of an empty cavity, $k_1$ is the wavenumber in the cavity material, $\Delta k = k - k_1$, $E_0$ is the incident field amplitude in free space, and $G_{\text{cav},\text{out}}$ is the Green’s tensor of the light propagating out of the cavity (details of the integration can be found in the Supplemental Material).

When treating the atomic vapor as a continuous medium with a spatially homogeneous polarizability, our model results in the familiar transmission coefficient of a Fabry–Perot etalon. In a high-density vapor, strong line shifts and broadenings exceed the single-body Casimir–Polder and Purcell effects that do not scale with density. In this regime, it is justified to neglect the spatial dependence of the polarizability in Eq. (4). Then, the coupled dipole model (3) results in a field governed by new Fresnel coefficients $t_{12}, r_{21}$ between cavity walls and a gas with refractive index $n_G = \sqrt{1 + N \alpha_{xx}/\epsilon_0}$. Inserting this field into Eq. (5) yields the well-known transmission coefficient of a Fabry–Perot etalon (see Supplemental Material)
\[ t = \frac{t_{12} r_{21} e^{i \Delta_k} + r_{21} t_{21} e^{ikd}}{1 - t_{21} r_{21} e^{2ikd}} \] (6)

The collective Lamb shift Eq. (1) originates from the classical physics contained in Eq. (6). In the low-density limit in free space, Eq. (3) can be solved by the single scattering (first Born) approximation resulting in a Lorentzian transmission profile that features the shift $\Delta_{\text{CLS}}$ (see also Refs. [15, 16] and Supplemental Material). As noted in Ref. [15], deviations from the low-density limit already occur for densities as low as $N = 0.01 k^3$. Therefore, the CSL cannot be used to study the properties of atomic vapors in the intermediate ($N \sim 1 k^3$) or dense ($N \gg 1 k^3$) regimes, even in free space. On the other hand, the Fabry–Perot relation (6) applies in the dense regime but does not account for the discontinuous distribution of atoms and their mutual correlations [15].

For a full description, we conducted numerical simulations with discrete particles. In each simulation run, we assigned random positions, ground-state populations and Doppler shifts sampled from the Maxwell–Boltzmann distribution to the otherwise static atoms. Then, we found the self-consistent solution to the scattering model (3) for different detunings $\delta$ and computed the transmission spectra according to Eq. (5). Finally, we computed the mean over many random atomic realizations until the transmission profile converged. The transmission profiles can be very well fitted to continuum versions of Eqs. (3) and (5) that allow for an additional broadening $\Gamma_p$ and a line shift $\Delta_p$ that are added to the atomic polarizability (4). The fit function also includes non-collisional atom-wall interactions and is detailed in the Supplemental Material. Analogously to the treatment in Ref. [16], the fitting parameters describe all effects that reach beyond the continuous medium model. Repeating the simulations for different densities, we found that $\Delta_p, \Gamma_p$ scale linearly with density. Their slopes $\partial N \Delta_p, \partial N \Gamma_p$ describe the non-trivial collective effects in the system.

In the simulations, the slab of atoms was truncated to a cylinder of radius $R$, with $R = \sqrt{256 \pi} k$ already yield-
ing reasonably well converged fit parameters. We con-
ducted the simulations for the D2 line of $^{85}$Rb at room
temperature with a Doppler width of $\Delta \omega_{\text{FWHM}} \approx 85 \Gamma_0$.
Although the polarizability (4) explicitly involves only
two energy levels, the Casimir–Polder shift $\omega_{\text{CP}}(r) =
\Delta \omega_{5S_{1/2},3P_1}(r) - \Delta \omega_{5S_{1/2},3P_0}(r)$ is computed using the full
atomic multi-level structure. The Casimir–Polder shift
scales asymptotically as $1/R^3$ close to the cavity walls,
shifting nearby atoms far away from resonance. This
results in an inert layer of gas close to the cavity walls
which gradually changes into a normally responding vapor.
Under our conditions this normal state is reached
about 30 nm away from the surface where the Casimir–
Polder shift is comparable to the natural linewidth.
This spatially inhomogeneous response alters the transmission
profile through the cavity and is included in our fitting
function. We performed simulations in a sapphire cavity
with and without non-collisional atom-wall interactions
and found that the atom-atom interaction parameters $\partial N \Delta \mu_p$, $\partial N \Gamma_p$ remain unchanged within the statistical un-
certainty. Hence, non-collisional atom-wall interactions
do not introduce effects beyond mean-field theory.

As a result, we can introduce dimensionless units based
on the two-level structure of the atoms that describe the
generic behavior of any alkali vapor by rescaling the cav-
ity size $d$ to the vacuum transition wavelength $\lambda$ and the
frequency shifts and broadenings to the Lorentz–Lorenz
shift $\Delta \LL$. Using these dimensionless units, we show in
Fig. 2 the results of the $^{85}$Rb simulation, together with a
recent experiment [16] on $^{39}$K vapor where, in both cases,
the gas is confined in a sapphire cavity. The error bars
of the simulation show the random error of the fits and
originate from finite statistics. Whereas the experiment
reaches up to atomic densities as high as $100 k^3$, our sim-
ulations were limited to densities around $1 k^3$. This was
due to the large numerical effort in solving the system of
linear equations (3) that scales as $n^3$ with the number of
atoms $n$. Further, in the experiment the cell temperature
is varied to change the density, and reaches higher values
($T = 600 \text{K}$) using lighter $^{39}$K atoms at the same time.
Given these differences, the simulation results compare
remarkably well with the observed collective shift and
broadening. In particular, the simulation reproduces a
$\lambda/2$-oscillation in the collective shift that is, however, dis-
placed between simulation and experiment.

On the other hand, the experiment shows no oscilla-
tory behavior in the broadening, and the pronounced dip
observed in the experiment does not show up in the simu-
lation. In sample computations, we found no signifi-
cant changes to our results when further increasing the
temperature. Therefore, these deviations should be con-
sidered in the context of four effects that are not cov-
ered by the theoretical model. First, atom-wall collisions
change the velocity distribution of the atoms away from
a Maxwell–Boltzmann shape as investigated in recent ex-
periments [25]. Once known, the real velocity distribu-
tion should be incorporated in the model. Second, the
model neglects atom-atom collisions. One could discrim-
nate this effect from the collective broadening by inves-
tigating different atomic lines. Whereas the collisional
broadening differs by a factor of $\sqrt{2}$ for the D1 and D2
lines [26, 27], the Lorentz–Lorenz shift, and hence the col-
lective broadening, differs by a factor of 2. Third, a mov-
ing atom can absorb a photon in one place and radiate at
another leading to a nonlocal susceptibility. This effect
has recently been experimentally observed in the meso-
scopic regime but can be neglected for dense vapors with
large homogeneous broadening [28]. Therefore, it is con-
sistent to compare the results in Ref. [16] to a simulation
that does not account for nonlocality. Fourth, we do not
account for the finite spectrum of the scattered light, and
only incorporate the Doppler shift to 'zeroth' order [15]
in the polarizability. However, modeling moving atoms
emitting a finite spectrum of light greatly increases the
numerical complexity, and no successful implementation
of such an approach is known to us.

We attribute the deviations between simulation and
experiment mainly to collisions and propose an exper-
imental design to test this assumption, consisting of
coated cavities that are equipped with distributed Bragg
refectors (DBR) made out of $\lambda/4$ stacks (see Fig. 3c,d).
We used a combination of silica ($n = 1.45$) and sapphire
($n = 1.77$) which provides adequate reflectance. With in-
creasing cavity finesse we find a prominent enhancement

FIG. 2. Slope of collective shift (top) and broadening (bottom) over width $d$ of a vapor-filled sapphire cavity in simulation ($^{85}$Rb, room temperature, density $\approx 1 k^3$) and experi-
ment [16] ($^{39}$K, temperature varied up to 600 K, density up
to $100 k^3$). In textbook electrodynamics, a constant Lorentz-
Lorenz shift (dotted line) and no additional broadening is
expected. Simulation error bars correspond to $1 \sigma$, solid lines
are spline interpolations.
of the amplitudes of the oscillatory features in the collective shift and broadening as shown in Fig. 4. The $\lambda/2$ period suggests the oscillations to be caused by correlated atom-atom interactions that are mediated by photonic round trips in the cavity. With increasing finesse, the oscillation amplitudes increase, too. As all cavity designs feature the same sapphire surface, the collisional atom-wall interactions and the resulting changes to the velocity distribution should be the same. Therefore, if future experiments investigate systematic changes in shift and broadening for different cavity designs, the results can be compared against our prediction even without knowledge about the atom-wall collisions.

Complementary to cavities with large finesse, we considered a cavity with an anti-reflection (AR) coating (see Fig. 3b). We chose a $\lambda/4$ magnesium fluoride ($n = 1.38$) layer, which provides decent reflection suppression ($R \approx 0.1\%$). In practice, one would add a thin sapphire layer ($\sim 10\,\text{nm}$) on top of the AR coating to protect it against the chemically aggressive alkali vapor [29]. The collective shift and broadening in the AR-coated cavity shown in Fig. 4 no longer feature pronounced oscillations which can be attributed to the fact that the AR coating suppresses photonic round trips in the cavity. However, the results for the AR-coated cavity still deviate from results obtained in free space. Although the AR coating suppresses reflection at normal incidence, the light fields exchanged between atoms also involve finite oblique reflection that modify atom-atom interactions.

The presented model is not exclusive to thermal vapor but also applies to ultracold atoms. However, one finds that at low temperatures ($T < 1\,\text{K}$) the resulting transmission profiles can no longer be described by continuum models, even when introducing more fit parameters such as an effective density. As a result, one cannot easily predict or discriminate to which amount a line shift is caused by mean-field effects as contained in Eq. (6), or by additional collective effects. This may be one reason for the seemingly conflicting outcomes of cold vapor experiments mentioned above.

In conclusion, we constructed a microscopic model that accounts for the effects of a macroscopic cavity environment on the dipole-dipole interaction between atoms. We showed that these modified interactions can explain the oscillatory dependence of the collective shift of thermal atoms on the cavity width observed in Ref. [16]. Furthermore, we predicted that the amplitudes of these oscillations can be tuned in coated cavities which can be tested in future experiments. The existing literature has explored the atom-atom interactions in one- and two-dimensional photonic crystals [12–14], and is now complemented by our three-dimensional description. Our model can be used to evaluate the role of dipole-dipole interactions, Casimir–Polder and Purcell effects in many macroscopic environments that have been interfaced with atoms such as microresonators [30], nanofibers [31], hollow-core fibers [32], superconducting chips [33], and atomic cladding waveguides [29, 34]. The necessary changes have to be incorporated in the respective Green’s tensor. In a next step, our model should be extended to account for nonlocal susceptibilities and saturation effects that are more likely to occur in cavity environments. The difficulties to reproduce the bulk Lorentz–Lorenz shift in thermal vapors in free space raised questions as to whether the microscopic modeling of atom-atom interactions can be considered complete [15]. We hope that future tests to our model can help address this fundamental question.

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Supplemental Material: Collective dipole-dipole interactions in planar nanocavities

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1. DERIVATION OF THE COUPLED DIPOLE MODEL

In this section, we derive the coupled dipole model from quantum theory at zero temperature. The zero-temperature limit is justified because room temperature provides no significant population at the optical transition frequencies. Furthermore, in our setting the separation between atoms and surface is never larger than the transition wavelength, and thus small compared to the wavelength of thermal photons. We start with the atom-field coupling Hamiltonian in dipole approximation [1, 2]

\[ H = \int d^3r \int_0^\infty d\omega \hat{\mathbf{r}} \cdot \hat{\mathbf{f}}(r, \omega) + \sum_{A} \sum_{n} E_{A,n} \sigma_{A,n} - \sum_{A} \int_0^\infty d\omega [\hat{d}_A \cdot \hat{E}(r, \omega) + \hat{E}^\dagger(r, \omega) \cdot \hat{d}_A], \]  

(S1)

where \( \hat{d}_A = \sum_{m,n} \hat{d}_{A,nn} \hat{\sigma}_{A,nn} \) and \( \hat{\sigma}_{A,nn} = |m_A\rangle \langle n_A| \) are the atomic transition operators. The equation of motion for the atomic operators becomes

\[ \dot{\hat{\sigma}}_{A,nn} = \frac{1}{\hbar} \{ \hat{\sigma}_{A,nn}, H \} = -i\omega_{A,nn} \hat{\sigma}_{A,nn} + i \int_0^\infty d\omega \left[ \sum_k (\hat{\sigma}_{A,nn} \hat{d}_{A,nn} - \hat{\sigma}_{A,kn} \hat{d}_{A,kn}) \cdot \hat{E}(r, \omega) \right. \\
\left. + \hat{E}^\dagger(r, \omega) \cdot \sum_k (\hat{\sigma}_{A,nn} \hat{d}_{A,nn} - \hat{\sigma}_{A,kn} \hat{d}_{A,kn}) \right], \]  

(S2)

where \( \omega_{A,nn} = (E_{A,n} - E_{A,m})/\hbar \). We expand the electric field operator in terms of the Green’s function \( G(r, r', \omega) \) and a set of bosonic vector fields \( \hat{\mathbf{f}}(r, \omega) \) describing collective excitations of the electromagnetic field and the linearly absorbing matter [1, 2]

\[ \hat{E}(r, \omega) = i \sqrt{\frac{\hbar}{\pi \epsilon_0 \omega^2}} \int d^3r' \sqrt{\text{Im} \epsilon(r', \omega)} G(r, r', \omega) \hat{\mathbf{f}}(r', \omega), \]  

(S3)

and solve its equation of motion in Markov approximation leading to [1, 2]

\[ \hat{E}(r, \omega, t) = e^{-i\omega t} \hat{E}(r, \omega, 0) + i \mu_0 \sum_{A} \sum_{m,n} \left( \delta(\omega - \omega_{A,nn}) - \frac{i}{\pi} \frac{\mathcal{P}}{\omega - \omega_{A,nn}} \right) \omega^2 \text{Im} G(r, r_A(t), \omega) \hat{d}_{A,nn} \hat{\sigma}_{A,nn}(t). \]  

(S4)

Here, we introduced the effective frequencies \( \omega_{A,nn} \) that govern the time evolution of the atomic operators due to the atom-field interactions, and that can be determined self-consistently later. We insert the field back into Eq. (S2) and explicitly denote the coherence between degenerate substates \( \nu, \mu \) of the energy levels \( n, m \) by \( \hat{\sigma}_{A,\nu\mu} \). Its expectation value becomes

\[ \langle \hat{\sigma}_{A,\nu\mu} \rangle = i \omega_{A,ge} \langle \hat{\sigma}_{A,ge} \rangle + i \sum_k \left\{ \langle \hat{\sigma}_{A,k\nu} \rangle \hat{d}_{A,\nu k} - \langle \hat{\sigma}_{A,k\mu} \rangle \hat{d}_{A,\mu k} \right\} \cdot \hat{E}_{\text{inc}}(r_A, t) \\
+ \sum_{B} \sum_{kpq} \left\{ \left( \langle \hat{\sigma}_{A,kp} \rangle \hat{d}_{A,pk} - \langle \hat{\sigma}_{A,\mu k} \rangle \hat{d}_{A,\nu k} \right) \cdot \hat{d}_{B,pp} \right\} \left( \frac{1}{2} A_{B,qp}(r_A, r_B) - i B_{B,qp}(r_A, r_B) \right) \hat{d}_{B,pp} \\
+ \hat{d}_{B,pp} \left\{ \langle \hat{\sigma}_{A,\nu\mu} \rangle \right\} \left\{ \langle \hat{\sigma}_{A,\nu\mu} \rangle \hat{d}_{A,\nu k} - \langle \hat{\sigma}_{A,\nu\mu} \rangle \hat{d}_{A,\mu k} \right\}. \]  

(S5)

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where we introduced the abbreviations
\[ \mathcal{A}_{A,nm}(r, r') = \frac{2\mu_0}{\hbar} \Theta(\tilde{\omega}_{A,nm}) \tilde{\omega}_{A,nm}^2 \text{Im} G(r, r', \tilde{\omega}_{A,nm}), \quad (S6) \]
\[ \mathcal{B}_{A,nm}(r, r') = -\frac{\mu_0}{\hbar \pi} \mathcal{P} \int_0^\infty d\omega \frac{\omega^2 \text{Im} G(r, r', \omega)}{\omega - \tilde{\omega}_{A,nm}} \]
\[ = -\frac{\mu_0}{\hbar} \Theta(\tilde{\omega}_{A,nm}) \tilde{\omega}_{A,nm}^2 \text{Re} G(r, r', \tilde{\omega}_{A,nm}) - \frac{\mu_0}{\hbar \pi} \int_0^\infty d\xi \frac{\tilde{\omega}_{A,nm}^2}{\tilde{\omega}_{A,nm}} + \xi^2 \xi^2 \text{Re} G(r, r', i\xi). \quad (S7) \]
We define the Casimir–Polder line shift and the corresponding Purcell decay rate as
\[ \Delta \omega_{A_{\nu\delta}}^n = \sum_k \sum_{\kappa} d_{A_{\nu\delta}} B_{A,nk}(r, r') d_{A_{\kappa\delta}}, \quad \Gamma_{A_{\nu\delta}}^n = \sum_k \sum_{\kappa} d_{A_{\nu\delta}} \mathcal{A}_{A,nk}(r, r') d_{A_{\kappa\delta}}, \quad (S8) \]
Assuming that the modification of the resonance frequency due to interactions is small compared to the bare resonance frequency, we can replace \( \tilde{\omega}_{A,nm} \) \( \rightarrow \) \( \omega_{A,nm} \) in the definition of \( \mathcal{A}_{A,nm}(r, r') \) and \( \mathcal{B}_{A,nm}(r, r') \) and compute the line shifts and decay rates perturbatively.
In order to retrieve the classical coupled dipole model, three classical assumptions are necessary [3]. First, we assume that we start from and remain in an incoherent mixture of ground states \( \langle \tilde{\sigma}_{A,\mu}^g \rangle = \delta_{\mu.\nu} f_{A_{\nu}^g} \) with occupation numbers \( f_{A_{\nu}^g} \). Second, we assume that the ground-state population of atom \( A \) is not correlated to the coherence of atom \( B \), i.e. \( \langle \tilde{\sigma}_{A_{\mu}^g} \tilde{\sigma}_{B_{\nu}^g} \rangle = \langle \tilde{\sigma}_{A_{\mu}^g} \rangle \langle \tilde{\sigma}_{B_{\nu}^g} \rangle \) when \( A \neq B \). Third, we assume that the atoms are unsaturated, and hence all excited states are not significantly populated. Furthermore, we assume a monochromatic incident field with frequency \( \omega_L \), i.e. \( E_{\text{inc}}(r, t) = E_{\text{inc}}(r)e^{-i\omega_L t} \). Then, we transform the equation of motion into the rotating frame of the incident field leading to the slowly varying coherence amplitude \( \langle \tilde{\sigma}_{A_{\mu}^g} \rangle = e^{i\omega_L t} \langle \tilde{\sigma}_{A_{\mu}^g} \rangle \). Assuming that the incident light is near-resonant to the \( e \leftrightarrow g \) transition but far detuned from all other atomic transitions, we discard other \( \langle \tilde{\sigma}_{B_{\nu}^g} \rangle \) terms in rotating-wave approximation. The effective equation of motion becomes
\[ \dot{\langle \tilde{\sigma}_{A_{\mu}^g} \rangle} = i(\omega_L - \omega_{A,eg}) \langle \tilde{\sigma}_{A_{\mu}^g} \rangle + \frac{i}{\hbar} f_{A_{\mu}^g} \hat{d}_{A_{\mu}^g}^g \cdot E_{\text{inc}}(r_A) + \frac{i}{\hbar} \sum_{\nu \neq \mu} (\Delta \omega_{A_{\nu}}^e + \frac{i}{2} \Gamma_{A_{\nu}^g}) \langle \tilde{\sigma}_{A_{\nu\delta}}^g \rangle + i \sum_{\delta} (\Delta \omega_{A_{\delta}^e} + \frac{i}{2} \Gamma_{A_{\delta}^g}) \langle \tilde{\sigma}_{A_{\nu\delta}}^g \rangle + \frac{i\mu_0}{\hbar} \tilde{\omega}_{A,eg} f_{A_{\mu}^g} \sum_{\nu \neq \mu} \sum_{\delta,\nu} \hat{d}_{A_{\mu}^g}^g \cdot G(r_A, r_B, \tilde{\omega}_{A,eg}) \hat{d}_{B_{\delta}^g}^g \langle \tilde{\sigma}_{B_{\nu\delta}}^g \rangle \quad (S9) \]

Different coherences of a single atom \( \langle \tilde{\sigma}_{A_{\mu}^g} \rangle, \langle \tilde{\sigma}_{A_{\nu}^g} \rangle \) can mutually couple when an atom emits and reabsorbs a photon that in between has changed its polarization after being reflected from a macroscopic body. In our planar cavity system, the Green’s tensor \( G(r, r, \omega) = \text{diag}(G_{\|}(r, r, \omega), G_{\|}(r, r, \omega), G^\perp(r, r, \omega)) \) is diagonal and the z-axis is chosen perpendicular to the surface. We take the quantization axis of the atoms along the z-direction and encounter no coupling, i.e. \( \Delta \omega_{A_{\nu}^e} = \Delta \omega_{A_{\nu}^e} = 0 \), \( \forall \nu \neq \delta \). When the quantization axis is chosen along the x-direction parallel to the surface, coupling occurs, e.g. \( \Delta \omega_{A_{\nu}^e} \neq 0 \) for \( \nu = -3/2, \delta = +1/2 \) because \( G^\perp(r, r, \omega) = G^\perp(r, r, \omega) \neq 0 \). Then, a linear equation system has to be solved. Depending on the choice of basis, the individual sublevels may experience different shifts and broadenings. However, the polarizability, which sums over all sublevels, is the same in all bases. Introducing the abbreviation \( \Gamma_{A_{\nu}} = \Gamma_{A_{\nu}} \), the operator equation of motion with a quantization axis in z-direction becomes
\[ \dot{\langle \tilde{\sigma}_{A_{\mu}^g} \rangle} = i(\omega_L - \omega_{A,eg} - \Delta \omega_{A_{\mu}}^e + \Delta \omega_{A_{\mu}}^g + i \frac{1}{2} \Gamma_{A_{\mu}^g} + \Gamma_{A_{\mu}^g}) \langle \tilde{\sigma}_{A_{\mu}^g} \rangle + \frac{i\mu_0}{\hbar} \tilde{\omega}_{A,eg} f_{A_{\mu}^g} \sum_{\nu \neq \mu} \sum_{\delta,\nu} \hat{d}_{A_{\mu}^g}^g \cdot G(r_A, r_B, \tilde{\omega}_{A,eg}) \hat{d}_{B_{\delta}^g}^g \langle \tilde{\sigma}_{B_{\nu\delta}}^g \rangle \quad (S10) \]
In the steady state, \( \langle \tilde{\sigma}_{A_{\mu}^g} \rangle = 0 \), one obtains a linear set of equations for the \( \langle \tilde{\sigma}_{A_{\mu}^g} \rangle \). Identifying the classical dipole moment with the expectation value \( \langle \hat{d}_{A_{\mu}^g} \rangle = \sum_{\mu,\nu} \langle \tilde{\sigma}_{A_{\mu}^g} \rangle \hat{d}_{A_{\mu}^g} \), we obtain Eq. (3) of the main text.
II. EXPLICIT FORM OF THE GREEN’S TENSOR FOR A PLANAR CAVITY

The Green’s tensor is the unique solution of the Helmholtz equation

$$[\nabla \times \nabla \times -k^2 \varepsilon(\mathbf{r}_1, \omega)] \mathbf{G}(\mathbf{r}_1, \mathbf{r}_2, \omega) = \delta(\mathbf{r}_1 - \mathbf{r}_2),$$

(S11)

with the Sommerfeld radiation boundary condition \( \mathbf{G}(\mathbf{r}_1, \mathbf{r}_2, \omega) \to 0 \) when \( |\mathbf{r}| \to \infty \) where \( \mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2 \). The free-space (bulk) part of the Green’s tensor can be expressed as [4]

$$G_{\text{free}}(\mathbf{r}_1, \mathbf{r}_2, \omega) = -\frac{\delta(\mathbf{r})}{3k^2} + \frac{e^{ikr}}{4\pi k r^3} \left[(k^2 r^2 + i kr - 1) I + (3 - 3ikr - k^2 r^2) \mathbf{e}_r \otimes \mathbf{e}_r\right].$$

(S12)

Next, we derive the cavity (scattering) contribution \( \mathbf{G}_{\text{cav}}(\mathbf{r}_1, \mathbf{r}_2, \omega) \) in Eq. (2). Due to cylindrical symmetry, we can utilize a coordinate system in which the source is located at \( \mathbf{r}_2 = (0, 0, z_2) \), the receiver at \( \mathbf{r}_1 = (x, 0, z_1) \), with the \( z \)-axis perpendicular to the surfaces. Any other pair of points can be transformed to this set of coordinates. By defining the distance \( x = \sqrt{(x_1 - x_2)^2 + (y_1 - y_2)^2} \) and the angle \( \cos \phi = (x_1 - x_2)/x \) to the \( x \)-axis, we can write the Green’s tensor as

$$\mathbf{G}_{\text{cav}}(\mathbf{r}_1, \mathbf{r}_2, \omega) = \mathbf{G}_{\text{cav}}(x, \phi, z_1, z_2, \omega) = \mathbf{R}^T(\phi) \mathbf{G}_{\text{cav}}(x, z_1, z_2, \omega) \mathbf{R}(\phi),$$

(S13)

$$\mathbf{R}(\phi) = \begin{pmatrix} \cos \phi & -\sin \phi & 0 \\ \sin \phi & \cos \phi & 0 \\ 0 & 0 & 1 \end{pmatrix}.$$

(S14)

Next, \( \mathbf{G}_{\text{cav}}(x, z_1, z_2, \omega) \) is constructed in the basis of \( s \)- and \( p \)-polarised plane waves [4]. We decompose the wavevector in layer \( j (j = 1 \text{ outside and } j = 2 \text{ inside the cavity, see Fig. 1) into components parallel and orthogonal to the surface, } \mathbf{k}_j = \mathbf{k}^\parallel + \mathbf{k}^\perp_j. \) Using cylindrical coordinates \( \mathbf{k}^\parallel = \mathbf{k}^\parallel(\cos \phi_k, \sin \phi_k, 0)^T \), we write the basis as

$$\mathbf{e}^j_\pm = \mathbf{e}^j_\parallel \otimes \mathbf{e}^j_\pm = (\sin \phi_k, -\cos \phi_k, 0)^T,$$

(S15)

$$\mathbf{e}^j_\pm = \frac{1}{k_j} (\mathbf{k}^\parallel \mp k^\perp_j \mathbf{e}_z) = \frac{1}{k_j} (\mp k^\perp_j \cos \phi_k, \mp k^\perp_j \sin \phi_k, \mathbf{k}^\parallel)^T.$$

(S16)

Considering the different pathways connecting source and receiver sketched in Fig. 1, we can write down Green’s tensors analogously to the treatment in Ref. [4],

$$\mathbf{G}_{\text{cav}}(x, z_1, z_2, \omega) = \frac{i}{8\pi^2} \int_0^\infty dk^\parallel \frac{k^\parallel}{k^2} \int_0^{2\pi} d\phi_k e^{ik^\parallel x \cos \phi_k} \sum_{\sigma=s,p} \left( \frac{(r_{\sigma}^{21})^2 e^{i k^\perp_j (2d-z_1-z_2)}}{D_{\sigma}} \mathbf{e}_\sigma^2 \otimes \mathbf{e}_\sigma^2 \right.$$

$$\left. - (r_{\sigma}^{21})^2 e^{i k^\perp_j (2d-z_1+z_2)} D_{\sigma} \mathbf{e}_{\sigma}^2 \otimes \mathbf{e}_{\sigma}^2 \right),$$

(S17)

with reflection and transmission coefficients

$$r_{\sigma}^{21} = \frac{k_2^\perp - k_1^\perp}{k_2^\perp + k_1^\perp}, \quad r_{\sigma}^{21} = \frac{\epsilon_1 k_2^\perp - \epsilon_2 k_1^\perp}{\epsilon_1 k_2^\perp + \epsilon_2 k_1^\perp},$$

(S18)

$$t_{\sigma}^{21} = \frac{2k_2^\perp}{k_2^\perp + k_1^\perp}, \quad t_{\sigma}^{21} = \frac{k_1 + 2\epsilon_2 k_2^\perp}{k_2^\perp + k_1^\perp},$$

(S19)

$$D_{\sigma} = 1 - (r_{\sigma}^{21})^2 e^{2idk^\perp_j}.$$

(S20)
For walls consisting of \( n-1 \) layers, one computes \( r_{21}^{j+1,j} \) as above and infers the effective reflection coefficient of the stack layer by layer using the recursion relation

\[
r_{\sigma}^{j+1,j} = \frac{\tilde{r}_{\sigma}^{j+1,j} + r_{\sigma}^{j-1,j} e^{2i k \hat{r}_{\sigma}^{j-1,j} d_{j}}}{1 + r_{\sigma}^{j+1,j} r_{\sigma}^{j-1,j} e^{2i k \hat{r}_{\sigma}^{j-1,j} d_{j}}}.
\]

Here, \( \tilde{r}_{\sigma}^{j+1,j} \) denotes two-layer coefficients analogously to Eq. (S18). The integral over \( \phi_k \) in Eq. (S17) leads to cylindrical Bessel functions \( J_n(x) \),

\[
J_n(x) = \frac{1}{2\pi i n} \int_0^{2\pi} d\phi e^{i x \cos \phi} \cos(nx) \quad \text{(S21)}
\]

The remaining integral over \( k^\parallel \) can be solved numerically by using an appropriate integration contour [5].

Furthermore, we encounter integrals of the form

\[
\int dA' \mathbf{G}(\mathbf{r}, \mathbf{r}') = \text{diag}(\tilde{G}^\parallel(z, z'), \tilde{G}^\parallel(z, z'), 0),
\]

\[
[\partial^2_z + k^2 \varepsilon(z, \omega)]\tilde{G}^\parallel(z, z') = -\delta(z - z').
\]

This is also valid for the Green’s tensor \( \mathbf{G}_{\text{cav, out}}(\mathbf{r}, \mathbf{r}') \) that propagates light from a point inside of the cavity to a point outside of the cavity that is needed in the derivation of Eq. (5). The one-dimensional Helmholtz equation has the intuitive planar wave solutions (see Fig. 1)

\[
\int dA' \mathbf{G}_{\text{cav, out}}(z, \mathbf{r}') = \text{diag}(1, 1, 0) \int \frac{e^{i k (d-|z'| - z)}}{2k} + r_{21} e^{i k (d+|z'| + i k |z'|)} + \frac{r_{21}^2 e^{i k (2d-|z'|)}}{1 - r_{21}^2 e^{2 i k d}}
\]

\[
\int dA' [\mathbf{G}_{\text{free}}(z, \mathbf{r}') + \mathbf{G}_{\text{cav}}(z, \mathbf{r}')] = \text{diag}(1, 1, 0) \int \frac{e^{i k |z|}}{2k} + r_{21} e^{i k |z| + i k |z'|} + \frac{r_{21}^2 e^{i k (2d-|z'|)}}{1 - r_{21}^2 e^{2 i k d}}.
\]

Note that these results are not limited to the far field.

### III. Continuous Medium Model

In this section, we derive the transmission coefficient of the Fabry–Perot etalon, Eq. (6), from our microscopic interaction model. The transition from discrete atoms to a continuous gas is accomplished by the replacement \( \sum_{j \neq i} N \rightarrow N \int dV \). Averaging over the ground state populations, the polarizability tensor becomes diagonal \( \alpha(z) = \text{diag}(\alpha^\parallel(z), \alpha^\parallel(z), \alpha^\perp(z)) \). We introduce the susceptibility \( \chi(z) = N \alpha^\parallel(z)/\varepsilon_0 \) and the normalized field \( \tilde{E}_x(z) = d_x(z)/(\alpha^\parallel(z)E_0) \). The coupled dipole model Eq. (3) becomes

\[
\tilde{E}_x(z) = \tilde{E}_{x, \text{inc}}(z) + k^2 \int_0^d dz' \int dA' G_{xx}(z, \mathbf{r}') \chi(z') \tilde{E}_x(z') \quad \text{(S26)}
\]

\[
\approx \tilde{E}_{x, \text{inc}}(z) + k^2 \chi \int_0^d dz' \int dA' G_{xx}(z, \mathbf{r}') \tilde{E}_x(z')
\]

\[
= t_{12} \frac{e^{i k z} + r_{21} e^{i k (2d-z)}}{1 - r_{21}^2 e^{2 i k d}} + \frac{i k \chi}{2} \int_0^d dz' \tilde{E}_x(z') \frac{e^{i k |z|}}{1 - r_{21}^2 e^{2 i k d}}.
\]

In the second step, we neglected the non-collisional atom-wall interactions, replacing \( \alpha^\parallel(z) \) with a constant \( \alpha^\parallel \). This approximation is valid in a regime where density-dependent interactions dominate as argued in the main text. In Eq. (S26), we do not explicitly account for the singular contribution of the free space Green’s tensor (S12). Its effect can be incorporated by inserting the Lorentz–Lorenz shift into \( \chi \) and redefining \( \tilde{E}_x(z) \) as the locally corrected field. The solution of Eq. (S27) then assumes the intuitive form

\[
\tilde{E}_x(z) = \frac{t_{12} (e^{i k n \alpha z} + r_{21} e^{i k n \alpha (2d-z)})}{1 - r_{21}^2 e^{2 i k n \alpha d}} ,
\]

\[
\text{(S28)}
\]
with new Fresnel coefficients $\tilde{t}_{12}, \tilde{r}_{21}$ between cavity walls and the refractive index $n_G = \sqrt{1 + \frac{N_\alpha}{\epsilon_0}}$ of the gas. Inserting this result into the continuous version of Eq. (5), we obtain the transmission profile of the Fabry–Perot etalon

$$t = \frac{t_{12}}{1 - r_{21}^2 e^{2ikd}} + \frac{ik\chi}{2} e^{i(k-k_1)} \int_0^d dz \frac{e^{-ikz} + r_{21} e^{ikz}}{1 - r_{21}^2 e^{2ikd}} t_{21} \tilde{E}_x(z) = \frac{t_{12} \tilde{t}_{21} e^{i(kN_G-k_1)}}{1 - r_{21}^2 e^{2im_0kd}}. \quad (S29)$$

### IV. COLLECTIVE LAMB SHIFT

In this section, we derive the collective Lamb shift of a continuous atomic slab in free space. In the low density limit, one can approximate the solution of Eq. (S26) by considering only a single scattering event (first Born approximation). In free space, the result reads

$$\tilde{E}_x \approx \tilde{E}_x^{(0)} + \tilde{E}_x^{(1)} = e^{ikz} + k^2 \int_0^d dz' \int dA' G_{xx}(z, z') e^{ik(k_1-z')}. \quad (S30)$$

The transmission coefficient according to Eq. (S29) then assumes the form

$$t = 1 + \frac{ik\chi d}{2} (1 + \xi), \quad \xi = \frac{k^2}{d} \int_0^d dz \int_0^d dz' \int dA' G_{xx}(z, z') e^{ik(k_1-z')} \quad (S31)$$

We can express the susceptibility $\chi = \frac{3\Delta_{LL}}{\pi(i+2k)}$ in terms of the Lorenz-Lorentz shift $\Delta_{LL} = -\frac{Nd_g^2}{3\alpha_{\text{eff}}(2k+1)}$, where we assumed equally populated ground states with $\sum_{\mu,\nu} f_{\mu} g_{\nu} d_{\mu} \otimes d_{\nu} = I_{(2J_g+1)}$. Employing the Taylor expansion $\frac{1}{1-x} \approx 1 + x$ for $x \ll 1$, we can approximate the transmission profile by a Lorentz curve

$$t \approx 1 - \frac{k\chi d}{2i} \left(1 - \frac{3\Delta_{LL} k^2}{2i(\delta - 3\Delta_{LL} \Re \xi) + 6\Delta_{LL} \Im \xi - \Gamma_0} \right), \quad (S32)$$

from which we can read off the collective Lamb shift. The resulting expression is identical to Eq. (4.1) in the original derivation of the collective Lamb shift by Friedberg et al. [6], which reads

$$\Delta = 3\Delta_{LL} \Re \xi - \frac{3\Delta_{LL} k^2 \Re}{d} \int_0^d dz \int_0^d dz' \frac{i e^{ik|z'-z|}}{2k} e^{ik(k_1-z')} \quad (S34)$$

$$= -\frac{3\Delta_{LL}}{4} \left(1 - \frac{\sin(2kd)}{2kd} \right). \quad (S35)$$

Subsequently, Friedberg et al. added a term $\Delta_{LL}$ accounting for the local-field correction that stems form the singularity of the free-space Green’s tensor, Eq. (S12). We obtain the collective Lamb shift from an entirely classical theory.

### V. FITTING FUNCTION

Here, we discuss the fitting function used to extract the parameters $\Delta_p, \Gamma_p$. Both parameters modify the susceptibility of the vapor $\chi(z)$ for which we have to solve a continuous version of the coupled dipole model that accounts for non-collisional atom-wall interactions. Abbreviating $P(x) = \chi(z) \tilde{E}_x(z)$, Eq. (S26) becomes

$$\tilde{E}_x,\text{inc}(z) = \int_0^d dz' \int dA' G_{xx}(z, z') P(x). \quad (S36)$$

The $z'$ integral can be approximated by a Gauss–Laguerre quadrature, and the resulting linear set of equations is solved for $P(x)$. Eventually, one obtains the transmission profile

$$t = \frac{t_{12} \tilde{t}_{21} e^{i(k-k_1)}}{1 - r_{21}^2 e^{2ikd}} + \frac{ik}{2} e^{i(k-k_1)} \int_0^d dz \frac{e^{-ikz} + r_{21} e^{ikz}}{1 - r_{21}^2 e^{2ikd}} t_{21} P(z). \quad (S37)$$
The susceptibility $\chi(z) = N\alpha^\parallel(z)/\epsilon_0$ is computed from the ground-state averaged polarizability

$$\alpha(z) = \frac{1}{(2J_g + 1)} \sum_{\mu,\nu} \alpha_{\mu\nu}^{ge}(z) = \text{diag}[\alpha^\parallel(z), \alpha^\parallel(z), \alpha^\perp(z)].$$  \hfill (S38)

It contains an average over the Maxwell–Boltzmann velocity distribution

$$\alpha_{\mu\nu}^{ge}(z) = -\frac{1}{\hbar} d_{\mu\nu}^{ge} \otimes d_{\nu\mu}^{eg} \sqrt{\frac{\alpha}{\pi}} \int_{-\infty}^{\infty} dv \frac{1}{B_{\mu\nu} - kv} e^{-av^2}$$

$$= -\frac{1}{\hbar} d_{\mu\nu}^{ge} \otimes d_{\nu\mu}^{eg} \frac{\sqrt{\pi a}}{k} \left( D \left( \frac{\sqrt{a}B_{\mu\nu}}{k} \right) - i e^{-\frac{aR_{\mu\nu}^2}{k^2}} \text{sign}(\text{Im} B_{\mu\nu}) \right),$$  \hfill (S39)

where $D(x) = e^x \int_0^x dt e^{-t^2}$ is the Dawson function, $a = \frac{m}{2\pi k_B T}$ and

$$B_{\mu\nu} = \delta - \left[ \omega_{CP}^{ge}(z) + \Delta_p \right] + \frac{i}{2} \left[ \Gamma_{\nu}(z) + \Gamma_p \right].$$  \hfill (S40)

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