Scattering properties of collective dipolar systems

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We present a theoretical treatment of light scattering by an ensemble of $N$ dipoles, taking into account recurrent multiple scattering. We study the intrinsic optical properties of collective dipolar systems without specifying a particular illumination condition. We apply this formalism to study the collective absorption modes for an ensemble of small nanoparticles and then to derive collective radiative corrections due to mutual interactions in dipolar ensembles, a topic of major importance in the development of collective nanophotonic systems or atomic networks used as optical clocks.

INTRODUCTION

Collective phenomena are involved in many innovative nanophotonic systems and they take benefit of the richness of interactions between emitters [1–7]. In atomic ensembles in particular, collective coupling can be a powerful mean to modulate the emission properties, by controlling superradiant emission [8–11] or generating a spectral shift [12, 13]. Yet it could also represent a genuine obstacle, as in the case of optical atomic clocks engineering. Atomic networks can indeed combine a large number of trapped atoms with ultra-narrow transitions to provide today’s most precise clocks [14–17] while in engineering, atomic networks can indeed combine a large number of trapped atoms with ultra-narrow transitions to provide today’s most precise clocks [14–17] while in particular, the dispersive interaction energy between two atoms or two nanoparticles can be directly derived from the collective coupling in a metrological context.

In this article we present a theoretical framework that enables to calculate the scattering matrix for an ensemble of dipoles. This framework is built up on the scattering formalism which turned out to be a very powerful tool to describe interaction of light with one or two scatterers. In particular, the dispersive interaction energy between two atoms or two nanoparticles can be directly derived from the scattering matrix [20, 21]. Meanwhile, we use theoretical and numerical methods which have been developed to tackle multiple scattering, in particular the Coupled Dipoles Model (CDM) which is very well suited to account for recurrent scattering taking place in a collective or dense system [22–24].

The new framework merges the two approaches and describes light scattering by an ensemble of dipoles. It calculates a collective scattering matrix including multiple scattering at all orders. Moreover it describes the intrinsic optical properties of the ensemble, without any specific choice of illumination or emission conditions. We will illustrate the advantages of this model, by studying first the collective absorption properties of an ensemble of dielectric nanospheres, and by computing then the collective radiative corrections to energy levels due to mutual interactions in an ensemble of dipoles.

THE BASIC APPROACH

A polarizable dipole located at $r_0$ and submitted to an incoming harmonic field $E_{\text{inc}}(r)$ of pulsation $\omega = ck$ acquires a dipolar momentum

$$p_0 = \varepsilon_0 \alpha_0(\omega) E_{\text{inc}}(r_0),$$

where its polarizability $\alpha_0$ has the dimension of a volume. It then generates a scattered field which can be expressed by using the Green tensor as

$$E_{\text{sca}}(r) = \mu_0 \omega^2 G_0(r, r_0, \omega) \cdot p_0.$$ 

In a collective system of $N$ interacting dipoles, each dipole additionally receives the field scattered by other dipoles. This is treated exactly by replacing for each dipole the incoming field by an exciting field accounting for the emission of other dipoles

$$E_{\text{exc},i}(r) = E_{\text{inc}}(r) + \mu_0 \omega^2 \sum_{i' \neq i} G_0(r, r_{i'}, \omega) \cdot p_{i'}.$$

Writing the exciting fields $E_i = E_{\text{exc},i}(r_i)$ as components of a column vector form, it is easy to express them from the incident fields by inverting a linear system

$$
\begin{pmatrix}
\vdots \\
\vdots \\
\vdots
\end{pmatrix} = [I - X]^{-1}
\begin{pmatrix}
\vdots \\
E_{\text{inc}}(r_i) \\
\vdots
\end{pmatrix},
$$

where $X$ is a structure matrix of size $3N$ by $3N$, whose blocks $(X_{ii'})_{i \neq i'}$ are 3-by-3 matrices containing the Green tensor that describes the propagation between the dipoles in vacuum (the diagonal blocks $X_{ii}$ are zero)

$$X_{ii'} = \left( \frac{k^3 \alpha_i G_0(r_i, r_{i'}, \omega)}{k} \right)_{i \neq i'}.$$ 

The field scattered by the ensemble of dipoles is then written by linearity as

$$E_{\text{sca}}(r) = \left[ \ldots, (k^3 \alpha_i G_0(r, r_i, \omega))_{i}, \ldots \right] \begin{pmatrix}
\vdots \\
E_i \\
\vdots
\end{pmatrix},$$

where
Riccati-Hankel functions which are naturally multiplied by the modes of the vacuum. For this subspace we also introduce a compact notation whose expressions are the same as those for collective systems where dipoles are closer to each other and in practice it must be increased until the result reaches convergence.

For those fields, we introduce the coefficients which have respective codomains \( E_{\text{exit}} \) and \( E_{\text{free}} \). These two matrices are related through the equality of the total field, using \( \phi_{\text{in}} = \phi_{\text{inc}} \) and \( \phi_{\text{out}} = 2\phi_{\text{sca}} + \phi_{\text{inc}} \),

\[
S = I + 2D .
\]

It must be kept in mind that this relation connects the coefficients \( A_{\ell,m}^{(j)} \) and \( B_{\ell,m}^{(j)} \) for \( S \) in the left-hand side to the coefficients \( A_{\ell,m}^{(1)} \) and \( B_{\ell,m}^{(1)} \) for \( D \) in the right-hand side.

The CDM, that uses incident and scattered fields \( \phi_{\text{inc}}, \phi_{\text{sca}} \) is well adapted to describe the multiple scattering events occurring among the dipoles. Hence we will first compute the \( D \) operator of the collective system by translating eqs. \( \text{[13]} \) into spherical modes. The obtained results will in the end be rewritten in terms of free fields in order to deduce the scattering operator \( S \) by using \( \text{[9]} \).

**THE COLLECTIVE SCATTERING MATRIX**

We start with eq. \( \text{[1]} \), for which one has to evaluate the incident field at the dipoles' positions \( (r_i) \) from its multipolar coefficients expressed in a common frame

\[
\phi_{\text{inc}} = (A_{\ell,m}^{(j)}, B_{\ell,m}^{(j)})^T .
\]

To do so, we proceed in two steps: first we translate the incident field by \( r_i \) to obtain the coefficients in spherical modes when it is expressed with respect to \( r_i \) with the operator \( T_{10} \) that transforms a free field to another free field \( \text{[20]} \); then we use these spherical coefficients to evaluate the vector field at the new origin \( r_i \). This second step can be realized through multiplication by an \( F \)
matrix of size 3 by $N_{\text{sph}}$, the number of spherical modes considered,

$$\frac{\mathbf{E}(0)}{P_0/\sqrt{\varepsilon_0}} = \frac{1}{\sqrt{12\pi}} \begin{pmatrix} i & 0 & -i \\ 1 & 0 & 1 \\ 0 & \sqrt{2} & 0 \end{pmatrix} \phi_{\text{inc}} .$$

$\mathbf{E}(0)$ is written in Cartesian coordinates and the only non-zero coefficients of the $F$ matrix correspond to $A^{(1)}_{\ell,m}$ with $\ell = 1$ and $m \in \{-1,0,1\}$. Finally, coming back to eq. (1) one obtains the exciting fields from the incident field as

$$\frac{1}{P_0/\sqrt{\varepsilon_0}} \begin{pmatrix} \mathbf{E}_i \\ \vdots \end{pmatrix} = [I - X]^{-1} \begin{pmatrix} \vdots \\ FT_{i0} \end{pmatrix} \phi_{\text{inc}} . \quad (7)$$

We then consider eq. (3), for which one has to express the coefficients $\phi_{\text{sca}} = (A^{(1)}_{\ell,m},B^{(1)}_{\ell,m})^T$ of the scattered field. Again, we do this in two steps, first by expressing in spherical modes the field scattered by a dipole at the origin and then by operating a translation by $-\mathbf{r}_i$ for the obtained field. If one considers a single dipole $\mathbf{p}_0 = \varepsilon_0 \alpha_0 \mathbf{E}_{\text{inc}}(0)$ located at the origin, the coefficients of $\phi_{\text{sca}}^0$ are obtained as a multiplication by a $N_{\text{sph}}$-by-3 matrix $Q$

$$\phi_{\text{sca}}^0 = k^3 \alpha_0 \frac{1}{\sqrt{12\pi}} \begin{pmatrix} 1 & i & 0 \\ 0 & 0 & \sqrt{2} \\ -1 & i & 0 \\ 0 & 0 & 0 \end{pmatrix} \mathbf{E}_{\text{inc}}(0) \frac{P_0/\sqrt{\varepsilon_0}}{Q}. \quad (8)$$

Non-zero components of $Q$ are relative to $A^{(1)}_{\ell,m}$ coefficients with $\ell = 1$ and $m \in \{-1,0,1\}$, and $\mathbf{E}_{\text{inc}}(0)$ is written in cartesian coordinates. We have separated the dimensionless factor $k^3 \alpha_0$ as it depends on the dipole polarizability, while $Q$ is a universal operator, related to $F$ by

$$Q = i F^\dagger .$$

Then the scattered field can be translated by $(-\mathbf{r}_i)$ such that it represents a field generated by a dipole at $\mathbf{r}_i$ in the basis associated with the global reference point $\mathbf{r}_0$. The translation is realized by the operator $T_{0i}$ that transforms an outgoing field to another outgoing field [20]. Finally doing this for each scatterer, we can express the total scattered field from the exciting fields as:

$$\phi_{\text{sca}} = (\ldots , (k^3 \alpha_i) T_{0i} Q , \ldots ) \frac{1}{P_0/\sqrt{\varepsilon_0}} \begin{pmatrix} \vdots \\ \mathbf{E}_i \end{pmatrix} , \quad (8)$$

where the constructed matrix on the left has dimensions $N_{\text{sph}}$ by $3N$.

Combining results (7) and (8), we obtain the matrix $\mathbf{D}$ of the total system as

$$\mathbf{D} = (\ldots , (k^3 \alpha_i) T_{0i} Q , \ldots ) [I - X]^{-1} \begin{pmatrix} \vdots \\ FT_{i0} \end{pmatrix} \quad (9)$$

and then the scattering matrix $\mathbf{S}$ by using (9), which is the main result of this article. The operator $\mathbf{S}$ fully describes the collective scattering by the ensemble of $N$ dipoles without specifying a specific choice of incident field, thus characterizing the intrinsic scattering properties of the collective dipolar system. As discussed in the following, this formalism is able to determine the proper scattering modes generated by collective interactions in such a system. Moreover, expanding the inverted operator into its Taylor series $(I + X + X^2 + \cdots)$ we get an expansion of the scattering matrix $\mathbf{S} = I + \mathbf{S}_1 + \mathbf{S}_2 + \cdots$ where $\mathbf{S}_k$ describes fields scattered $k$ times on various objects. This highlights the fact that this approach is non-perturbative as it includes multiple scattering at all orders.

### UNITARITY AND THE OPTICAL THEOREM

Before studying applications of this formalism, we note that the unitarity of the scattering matrix is linked to the optical theorem that ensures energy conservation or equivalently probability conservation in a situation where frequency conversion is disregarded.

For an isolated dipole located at the origin, formula (9) is simplified to

$$\mathbf{D} = (k^3 \alpha) \mathbf{Q} \mathbf{F} = (ik^3 \alpha/6\pi) I_{(A,\ell=1)} ,$$

where $I_{(A,\ell=1)}$ is the identity operator for coefficients $A_{\ell,m}$ with $\ell = 1$, $m \in \{-1,0,1\}$ and zero elsewhere. The condition of unitarity for the matrix $\mathbf{S}$ of a single dipole can then be written explicitly as

$$I - \mathbf{S}^\dagger \mathbf{S} = \frac{2k^3}{3\pi} \left[ \text{Im}[\alpha] - \frac{k^3 |\alpha|^2}{6\pi} \right] I_{(A,\ell=1)} = 0 , \quad (10)$$

where the term responsible for absorption by the dipole [27] appears within the brackets. We note that $\alpha \in \mathbb{R}$ is not the appropriate condition for $\mathbf{S}$ to be unitary, as could be expected. Instead, one has to include radiative reaction in the discussion, for instance by writing the polarizability as

$$\alpha = \frac{\alpha_0}{1 - \frac{\hbar c \omega}{6\pi \varepsilon_0}} .$$
With this notation, \( S \) is unitary if and only if \( a_0 \) is real. The scattering formalism thus naturally includes the optical theorem as it intrinsically incorporates radiative reaction in the treatment of energy conservation.

The fact that the operator \( D \) for a single dipolar object is directly proportional to the polarizability \( \alpha \) shows that the \( D \) operator can be interpreted as a generalization of the polarizability for a more complex system. As a consequence, the unitarity condition for the collective system is a generalization of eq. [10]

\[
I - S^\dagger S = -2 (D + D^\dagger) - 4D^\dagger D = 0 .
\] (11)

**COLLECTIVE ABSORPTION**

Using the collective scattering operator of the ensemble of dipoles, we are now able to study its intrinsic absorption properties. We can in particular define the absorption operator as the deviation of the scattering matrix from unitarity which is known to measure the quadratic norm of the noise [28]

\[
A = I - S^\dagger S .
\]

The scattering matrix can be rewritten as [25]

\[
S = U\Sigma V^\dagger = \sum_k \sigma_k u_k v_k^\dagger ,
\]

with \( \Sigma \) a diagonal matrix with real elements \( 0 \leq \sigma_k \leq 1 \), while \( U \) and \( V \) are the singular vectors that form a basis for \( \text{out} \) and \( \text{in} \) fields, respectively. For the absorption operator \( A \) the above decomposition is also valid as

\[
A = I - V\Sigma^\dagger \Sigma V^\dagger = \sum_k (1 - \sigma_k^2) v_k v_k^\dagger .
\] (12)

This means that for an incoming mode \( v_k \), the fraction \( \sigma_k^2 \) is re-radiated while a fraction \( 1 - \sigma_k^2 \) is absorbed by the system. This decomposition thus gives access to a complete knowledge on the intrinsic absorption properties of the system, by identifying the incoming modes responsible for small or large absorption by the system.

We illustrate this property by applying it to a simple system composed of \( N \) dielectric nanospheres, where one at the origin is absorbing and the remaining \( N - 1 \) are located at a fixed distance \( a \) around it and are non-absorbing. We place them on the vertices of a Platonic solid, which can be a tetrahedron \( (N = 5) \), an octahedron \( (N = 7) \), a cube \( (N = 9) \), an icosahedron \( (N = 13) \) or a dodecahedron \( (N = 21) \), and fix their dielectric function to \( \varepsilon = 10 \), while the central nanosphere features absorption with \( \varepsilon_C = (\sqrt{10} + 0.11)^2 \). The case \( N = 7 \) is sketched on fig. [1]. The nanospheres’ optical properties are described by a dipolar polarizability obtained from a Clausius-Mossotti model.

![Diagram of the system for \( N = 6 + 1 \) nanospheres with \( a = \lambda/2 \) (blue lines are guides to the eye).](image)

We numerically compute the scattering matrix of the collective system with a finite number of spherical modes, and extract the singular values and vectors for the absorption operator [12]. We then compare the results to the reference case with the central dipolar absorber only, in order to study the modifications in the absorption by the central dipole due to the non-dissipative outer shell that acts as a collective antenna. We note that the singular value decomposition enables the direct identification of the nature of the modes that yield maximal absorption for the collective system.

For an isolated dissipative dipole at the origin in the linear optics regime, the absorption is directly proportional to the incident electric field amplitude squared at the origin, and only three spherical modes are responsible for a non-zero absorption: electric modes with \( \ell = 1 \) and \(-1 \leq m \leq 1 \), which are associated with an identical singular value \( A_0 = 1 - \sigma_0^2 \). For the collective system described above, the absorption is now proportional to the amplitude squared of the excitation field at the origin. As a consequence, there are still only three singular incident modes responsible for non-zero absorption, but they can be associated with different singular vectors and singular values \( A = 1 - \sigma^2 \).

We present in fig. [2]a) the results of numerical evaluations for the relative change in the non-zero singular value for absorption \( (A - A_0)/A_0 \) due to the presence of the non-dissipative outer shell, as a function of the shell radius \( a \). We observe a strong increase in the system absorption for \( a \simeq \lambda/2 \), which can reach up to 60 % for the dodecahedron. This modification strongly depends on the size of the shell and can result in a decrease of absorption in some cases. In fig. [2]b) we present an histogram of the spherical mode coefficients for the singular mode responsible for maximum absorption for an octahedron with \( a = \lambda/2 \) which illustrates the appearance of
higher multipole components in their composition. These results show that the outer shell acts as an antenna for the central absorber as multiple scattering events enable to amplify the field at its position.

The scattering operator is also a very powerful tool to predict the absorption of the complex system for a series of incoming fields: for a new considered incident field $|\Psi\rangle$, one only needs to compute the module squared projection onto the three singular modes responsible for absorption $\sum_{i=1}^3 |\langle \phi_i |\Psi \rangle|^2$ and multiply it by the singular value $A$. For instance, for a linearly polarized planar wave incoming on a cube shell (N=9) with $a = \lambda/2$, the increase of the absorption singular value $A$ (+19.7%) observed in fig. 2(a) goes together with an increase of the projection coefficient due to the modification of the singular modes responsible for absorption (+15.0%) to almost double the increase of effective absorption (+37.6%) for this specific choice of incident field.

**COLLECTIVE RADIATIVE CORRECTIONS**

In collective dipolar systems, mutual interactions can lead to dispersive forces as well as energy shifts. In the latter case, the collective shifts can have crucial effect in atomic networks used in a metrological context by limiting the ultimate reachable accuracy. For instance, collective Lamb shifts may result from interactions that shift differently the ground and excited states energy levels of the system. Here we use the collective scattering formalism to derive the phase shift formula for a collective dipolar system. Applying these results to arrays of small nanoparticles, we show that that collective effect are already quite important when only considering the ground state energy of such an ensemble.

We start from the expression of the collective shift for an ensemble of dipoles as

$$\Delta(\omega) = \ln \det S(\omega) - \sum_{i=1}^N \ln \det S_i(\omega) \ , \quad (13)$$

where $S$ is the scattering matrix of the collective dipolar ensemble and $S_i$ is the scattering matrix for $i$th dipole.

Using the determinant lemma, one deduces from eq. (9)

$$\det [I - X + \left(\begin{array}{c} \vdots \\ FT_{0\ell} \end{array} \right) \left(\begin{array}{c} \vdots \\ 2(k^3\alpha_{\ell})T_{0\ell}Q, \ldots \right) \left(\begin{array}{c} \vdots \\ \vdots \end{array} \right)] = \frac{\det S}{\det [I - X]}$$

where the matrix appearing in the numerator has diagonal blocks $(1 + ik^3\alpha_{\ell}/(3\pi))I_3$, $I_3$ being the identity on $\mathbb{R}^3$, and non-diagonal blocks $(k^3\alpha_{\ell})[2FT_{0\ell}T_{0\ell}Q - G_0(r_i, r_{\ell'}, \omega)/k]$. Writing explicitly the translation operators for $\ell = 1$ one shows that

$$FT_{0\ell}T_{0\ell}Q = i\text{Im} \left[ G_0(r_i, r_{\ell'}, \omega)/k \right],$$

which is the quantity appearing in the cross density of states (CDOS) [29]. It follows that

$$G_0(r_i, r_{\ell'}, \omega)/k - 2FT_{0\ell}T_{0\ell}Q = G_0^*(r_i, r_{\ell'}, \omega)/k , \quad \Delta(\omega) = \ln \det[I - Y(\omega)] - \ln \det[I - X(\omega)] \ , \quad (14)$$

where $Y$ is a modified structure matrix, similar to $X$ but with non-diagonal blocks

$$\left[ \frac{k^3\alpha_{i\ell'}}{1 + ik^3\alpha_{i\ell'}/(3\pi)} \frac{G_0^*(r_{i\ell'}, r_{i\ell'}, \omega)}{k} \right]_{i\neq i'}$$

instead. In the particular case of non-dissipative dipolar objects, the left fraction in the latter expression is equal to $k^3\alpha_{i\ell'}^*$, which yields $Y = X^*$ and

$$\Delta(\omega) = -2i\text{Im} \left[ \ln \det(I - X(\omega)) \right] \ . \quad (15)$$

In such systems, the collective radiative corrections are hence entirely determined by the $3N$ by $3N$ structure.
matrix $X(\omega)$. Moreover, when looking at frequency-integrated quantity one can use imaginary frequencies $\omega = i\xi$ to express for instance dispersive energies as

$$E = \hbar \int_{0}^{\infty} \frac{d\omega}{2\pi} \frac{i\Delta(\omega)}{2} = \hbar \int_{0}^{\infty} d\xi \frac{\ln \det[I - X(i\xi)]}{2\pi}.$$  \hfill (16)

The latter result shows that the structure matrix $X$ emerges as the main ingredient to the collective energy shifts resulting from interactions in a dipolar ensemble.

We now illustrate these results by studying the collective radiative correction for the ground state of an array of $N$ nanoparticles, which in this case corresponds to the Casimir-Polder interaction energy, and by comparing it to the commonly derived pairwise quantity, obtained by summing up the radiative corrections obtained for all couples of atoms. For numerical application we consider separately the cases of 1D, 2D and 3D arrays, all being made of gold nanoparticles with radius $R = 20$ nm and optical properties modeled by a plasma model.

For collective systems with $N > 2$, we observe a deviation from the pairwise quantity, which increases when the array step size is reduced, as shown in fig. 3. This deviation is also increased by the number of atoms in the array, as expected, and the convergence between the two results at large spacings is slower for higher-dimensional arrays. In the case of a 3D atomic array, deviations of the order of the percent are obtained for array spacings up to 100 nm.

These results show that for simple dipolar systems, collective effects are already quite important when studying the radiative correction for ground-state energy. For collective dipolar systems, such as optical atomic clocks, where the collective shift of the transition energy is at play, the latter could be obtained by combining shifts for the ground-state as well as for the considered excited state. As the latter is expected to be of much longer range due to the slow decay of interactions between excited states [30–33], the presented results show that collective effects could play a major role in the energy shifts for atomic optical clocks, as already foreseen. The collective scattering formalism presented in this article, combined with a treatment of excited states, could be a way to investigate such collective energy shifts, a topic of major importance in the development of collective atomic clocks.

**Acknowledgments**

Thanks are due to T. Ebbesen, C. Genet and R. Guérout for stimulating discussions.

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