Near-resonant light scattering by a sub-wavelength ensemble of identical atoms

N.J. Schilder,1 C. Sauvan,1 Y.R.P. Sortais,1 A. Browaeys,1 and J.-J. Greffet1

1Laboratoire Charles Fabry, Institut d’Optique Graduate School, CNRS, Université Paris-Saclay, F-91127 Palaiseau Cedex, France.
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We study theoretically the scattering of light by an ensemble of \(N\) resonant atoms in a sub-wavelength volume. We consider the low intensity regime so that each atom responds linearly to the field. While \(N\) non-interacting atoms would scatter \(N^2\) more than a single atom, we find that \(N\) interacting atoms scatter less than a single atom near resonance. In addition, the scattered power presents strong fluctuations, either from one realization to another or when varying the excitation frequency. We analyze this counter-intuitive behavior in terms of collective modes resulting from the light-induced dipole-dipole interactions. We find that for small samples, their properties are governed only by their volume when \(N \gtrsim 20\).

When deriving the optical properties of bulk materials, one usually starts from a microscopic description of the medium as a collection of atoms featuring, in the simplest case, one resonance with frequency \(\omega_0 = 2\pi c / \lambda\) and linewidth \(\Gamma_0\) [1, 2]. When the medium is dilute, such as in a vapor, the field scattered by the atomic ensemble is simply the interference of the fields scattered by the individual atoms considered as independent, i.e. non-interacting. For larger densities, the interactions between the light-induced atomic dipoles play an increasing role and modify the scattering. This raises the question of what happens if near-resonant light is scattered from an atomic sample with sub-wavelength size. By analogy with a resonant dipolar nano-particle [3], one would expect that the scattering cross section \(\sigma_{sc}\) is on the order of \(\lambda^2\) and independent of the exact number of atoms in the ensemble when it is dense enough. But how do we reconcile this intuition with the fact that for a sub-wavelength-sized sample the fields scattered by the \(N\) atoms should be in phase and hence the cross section should vary like \(N^2\)?

The answer to this question relies on a description of the atomic sample in terms of collective modes resulting from the interactions between the dipoles induced by the driving light field [4–10][11]. As analyzed by many authors, each of the collective modes has an eigen-frequency and a natural linewidth, which depend crucially on the exact spatial arrangement of the atoms [13–16]. We have shown in a previous work [17] that for atomic samples with a size \(L\) on the order of a few \(\lambda\), a few dominant modes delocalized over the entire system, called polaritonic modes, play an important role in the scattering of light. They correspond to the electromagnetic modes found when the ensemble of atoms is described by an effective dielectric constant. In a following work investigating the homogenization of these systems [18], we have also found that the total power scattered close to resonance saturates when increasing the atom number. However, for \(L\) around a few \(\lambda\), we could not find a simple expression for the saturation value.

Here we show that for a sub-wavelength volume \((L \ll \lambda)\), for which no polaritonic mode exists, the strong light-induced interactions between atoms lead to a scattering cross section averaged over the excitation spectrum that is actually smaller than the one of a single atom, i.e the sample scatters less than a single atom! Furthermore, the cross section presents large fluctuations as a function of the laser frequency. These two properties are independent of the atom number for \(N \gtrsim 20\). This behavior is at odds with the two naive pictures mentioned above: a particle with an effective refractive index and \(N\) atoms scattering coherently. The question of the optical properties of sub-wavelength-sized ensemble was raised by Dicke [19] for the case where all the atoms are initially excited. It was realized later that the interactions suppress the resulting super-radiant emission [20, 21]. Here instead, we concentrate on the scattering in the low light intensity limit where the atomic dipoles respond linearly to the field.

To investigate the scattering by a sub-wavelength ensemble, we apply the model developed in our previous works [17, 18] to the case of a cubic box with sides \(\sim \lambda/(2\pi) = 1/k\) containing \(N\) identical atoms at rest. We consider each atom as a resonant scatterer characterized by a classical polarizability \(\alpha\). For the sake of simplicity, we assume that the dipoles can only oscillate linearly along the \(z\)-axis [22]. The wavelength of the atomic transition is \(\lambda = 2\pi c / \omega_0 = 780\) nm, and the spectral width \(\Gamma_0 = 2\pi \times 6\) MHz (case of rubidium D2 line relevant for experiments [23–30]). We illuminate the system with a monochromatic plane wave propagating along the \(x\)-axis and linearly polarized along the \(z\)-axis. Each atom is driven by the incident light field (frequency \(\omega\)) and the sum of the \(z\)-components of the fields scattered by all other atomic dipoles. This leads to a set of coupled dipole equations from which we calculate, in steady state, the induced dipoles. We finally compute the total scattered field \(\mathbf{E}_{sc}\) and the total scattered power evaluated on a spherical surface \(\Sigma\) in the far field:

\[
P_{sc} = \frac{\varepsilon_0 c}{2} \oint_{\Sigma} |\mathbf{E}_{sc}|^2 dS. \tag{1}
\]
modes have a larger scattering cross section and small spectral width, which indicates that they are higher order modes resulting from the finite size of the ensemble. Indeed, the maximum scattering cross section of a $l$-order mode varies as $\sigma_l \sim (2l+1)\lambda^2/(2\pi)$ [31, 33], with the dipolar case corresponding to $l = 1$, the quadrupolar mode to $l = 2$, etc. We can now understand why the ensemble-averaged scattering cross section of the ensemble is smaller than $3\lambda^2/(2\pi)$: due to the spectral separation between the modes, a laser line centered on the atomic resonance $\langle \omega = \omega_0 \rangle$ excites mainly one mode with scattering cross section $\sim 3\lambda^2/(2\pi)$ or misses the modes. As their frequencies vary from one realization of the spatial distribution to another, the ensemble-averaged cross-section is smaller than $3\lambda^2/(2\pi)$.

We now turn to the discussion of the spectral widths $\Gamma_m$ of the modes. Figure 2(a) shows the distribution of frequencies $\omega_m$ and widths $\Gamma_m$ of the collective modes obtained from 1000 realizations of the cloud for $N = 10$ dipoles and $VK^3 = 1$. The widths vary significantly, indicating the presence of superradiant (broad) modes and subradiant (narrow) modes. However, their spectral widths are linked by a sum rule. To see that, we write the set of homogeneous linear equations describing the modes of the system: $[A][P] = (\omega_m - i\Gamma_m/2)[P]$ where $[P]$ is the vector containing the dipole moments of all the atoms and $[A]$ is the matrix connecting them:

$$[A] = \begin{pmatrix} (\omega_0 - i\Gamma_0/2) & -\frac{3\pi\Gamma_0}{\omega_0}G_{21} & \cdots \\ -\frac{3\pi\Gamma_0}{\omega_0}G_{12} & (\omega_0 - i\Gamma_0/2) & \cdots \\ \vdots & \ddots & \ddots \end{pmatrix}$$

Here, $G_{ij} = [G]_{xx}(r_j, r_i; \omega)$ is the $xx$ component of vacuum Green’s tensor describing the vectorial dipole-dipole interaction between atoms $i$ and $j$, including the $1/r$, $1/r^2$ and $1/r^3$ terms [3, 34]. As the trace of $[A]$ is basis-independent, its imaginary part is the same expressed in the atomic basis as above or in the collective mode basis, hence $\sum_{m=1}^N \Gamma_m = N\Gamma_0$. In Fig. 2(b), we plot the normalized probability distribution of the spectral widths extracted from Fig. 2(a). We observe two peaks around $\Gamma = 0$ and $\Gamma = 2\Gamma_0$. They correspond to pairs of closely-spaced atoms for which the dipole moments are anti-parallel (subradiant) or parallel (superradiant), respectively. The maximum spectral width is $\sim 5\Gamma_0$, showing that there is not a single mode dominating.

Having characterized the amplitude of the resonances and their width, we now investigate why the average scattered power does not depend on atom number for $N \gtrsim 20$. Let us define $\delta \omega$ as the typical frequency spacing between two eigenfrequencies. The probability that a monochromatic light with frequency $\omega$ excites a mode and scatters is thus $\sim \Gamma_0/\delta \omega$ so that the average scattering cross section of the cloud is approximately $\sigma \approx (\Gamma_0/\delta \omega) 3\lambda^2/(2\pi)$. To estimate this average spacing
\(\delta\omega\), we first estimate the spread \(\Delta\omega\) of the eigenfrequencies. As the atomic ensemble is dense, the dipoles are mostly in the near-field of each other, dominated by the \(1/r^3\) term. We thus assume that the frequency spread is given by the spectral shift \(\Gamma_0/(kr)^3\) due to the interaction between two atoms separated by a typical distance \(r\) given by \(N r^3 = V\) [35]. The spacing between modes is \(\delta\omega \sim \Delta\omega/N\), where the atom number \(N\) is also the total number of modes. It follows that

\[
\delta\omega \sim \frac{\Gamma_0}{VK^3} \tag{2}
\]

is independent of the number of atoms, and so is the average cross section. This scaling argument predicts a strong dependence of the frequency spacing with the volume. To check this prediction, we calculate the eigenmodes for a fixed number of atoms \((N = 20, 40, 80)\) while varying \(VK^3\) between 0.1 and 10. For each realization of a particular system we calculate all eigenfrequencies \(\omega_m\). We then compute the spacings between adjacent modes \(\omega_{m+1} - \omega_m\) and determine the median of this distribution as an indication of the typical spacing. We choose the median rather than the mean since it is insensitive to large spectral shifts originating from closely spaced pairs of atoms. The result is shown in Fig. 3: the mode spacing is indeed independent of the atom number and is proportional to \(1/V\), thus confirming the scaling of Eq. (2). It becomes larger than the average spectral width \(\Gamma_0\) when \(VK^3 \lesssim 10\). Furthermore, as the widths of the modes are bounded by \(\Delta\Gamma_0\), for small enough volumes the modes do not overlap, their spacing being larger than their width. In summary, the strong dipole-dipole coupling between atoms produces collective modes spectrally well separated with a frequency spacing that varies as \(1/(VK^3)\). This near-resonance scattering regime differs considerably from the one of a sequence of scattering events by independent atoms, valid for weak interactions. The calculations above indicate that the transition between these two regimes occurs for \(VK^3 \sim 10\). The factor controlling this transition is thus \(VK^3\), independent of \(N\) provided \(N \gtrsim 20\). Interestingly the atomic density at which the transition occurs is still orders of magnitude smaller than the densities encountered in condensed matter systems such as dielectric sphere or nano-particles.

In an actual experiment with a vapor, the atoms move so that the spectral positions of the modes fluctuate over time. For a monochromatic incident laser, we thus expect the scattered power to display giant fluctuations in time as the scattering cross section fluctuates between 0 and \(3\sqrt{2}/(2\pi)\) (see Fig. 1b). To characterize the amplitude of the fluctuations, we compute the standard deviation of the scattered power over an ensemble of random realizations of the system: \(\sigma_P = \sqrt{\langle P_{sc}^2 \rangle - \langle P_{sc} \rangle^2}\). Figure 4(a) shows the standard deviation normalized by the average scattered power as a function of \(N\): it becomes independent of the number of atoms as \(N\) increases. We can understand this behavior using the following argument. Assuming that the light is either scattered with a probability \(\Gamma_0/\delta\omega\) or transmitted (i.e. not scattered) with a probability \(1 - \Gamma_0/\delta\omega\), we find that \(\langle P_{sc} \rangle = \langle \Gamma_0/\delta\omega \rangle \sigma_{I} I\) and \(\langle P_{sc}^2 \rangle = \langle \Gamma_0/\delta\omega \rangle \sigma_{I}^2 I^2\), with \(\sigma_{I}\) the cross section of a mode and \(I\) the intensity of the incoming light field. We thus obtain \(\sigma_P/\langle P_{sc} \rangle = \sqrt{(\delta\omega - \Gamma_0)/\Gamma_0} \approx 1/(\sqrt{VK^3})\) where we used Eq. (2). We plot \(\sigma_P\) calculated numerically as a function of \(V\) in Fig. 4(b). We indeed observe that the fluctuations are very large for small volumes and follow the scaling derived above. In contrast, when the volume increases, \(\sigma_P\) tends to zero. The transition be-
between the fluctuating to non-fluctuating regime occurs when the frequency spacing $\delta \omega$ equals the average spectral width $\Gamma_0$. According to Fig. 3 and Eq. (2), this corresponds to $V k^3 \approx 10$. In summary, for interacting atoms, the condition $V k^3 \lesssim 10$ defines the large fluctuation regime with a small average scattering cross section. Remarkably for non-interacting atoms, the same condition defines the regime of coherent interferences leading to an enhanced scattering cross section $N^2 3 \lambda^2 / (2\pi)$.

So far, we have considered a nearly resonant illumination with a monochromatic source having a spectral width smaller than $\Gamma_0$. We now consider the case of a broad spectrum illumination with a width larger than the spectral width $\Delta \omega = N \delta \omega$. From the sum rule discussed previously, it follows that the spectrally integrated cross section is constant and does not fluctuate from one realization to another or when the atoms move. The integrated cross section is thus expected to be $N 3 \lambda^2 / (2\pi)$ and not to fluctuate around this value. This is an example of a self-averaging procedure [36]: the fluctuations of the scattered light are removed by integrating over the many eigen-frequencies. An analogous system consists of light scattered by a rough surface forming a speckle pattern, which is detected using a collection solid angle either smaller or larger than the angular aperture of a speckle grain. In the former case, the signal will display large fluctuations when changing the realization. In the latter case, it will be constant although it is a measurement performed on a single realization. In both cases, the self-averaging procedure stems from the increase of the number of channels (different frequencies or different angles) used to transmit electromagnetic power while using a single realization of the random system.

In conclusion, we have analyzed the near-resonance scattering of light by an ensemble of atoms in a volume $V$ smaller than $10 / k^3$. When this condition is fulfilled, all the light induced dipoles are strongly interacting. This interaction produces spectrally well separated collective modes. The system needs to be described in terms of these collective modes and the picture of a sequence of scattering events by each atom is no longer valid. The scattering properties of this type of systems are: (i) the number of atoms only influences the total spectral width of the cloud; (ii) for a laser on resonance, the average scattered power and the fluctuations do not depend on the number of atoms for $N \gtrsim 20$, but only on the volume of the system, and (iii) the smooth transition between the usual coherent behavior of non interacting atoms and the large fluctuation regime takes place for $k^3 V \approx 10$. This transition thus occurs in systems still very dilute with respect to condensed matter ones.

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