Observation of suppression of light scattering induced by dipole-dipole interactions in a cold atomic ensemble

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Dated: February 19, 2014

We study the emergence of collective scattering in the presence of dipole-dipole interactions when we illuminate a cold cloud of rubidium atoms with a near-resonant and weak intensity laser. The size of the atomic sample is comparable to the wavelength of light. When we gradually increase the atom number from 1 to 450, we observe a broadening of the line, a small red shift and, consistently with these, a strong suppression of the scattered light with respect to the noninteracting atom case. Numerical simulations, which include the internal atomic level structure, agree with the data.

PACS numbers: 42.50.Ct,42.50.Nn,42.25.Fx,32.80.Qk,03.65.Nk

When resonant emitters, such as atoms, molecules, or quantum dots, with a transition at a wavelength $\lambda$ are confined inside a volume smaller than $\lambda^3$, they are coupled via strong dipole-dipole interactions. In this regime, the response of the ensemble to near-resonant light is collective and originates from the excitation of collective eigenstates of the system, such as super- and sub-radiant modes \[.\] Dipole-dipole interactions profoundly affect the response of the system, as they modify the decay rate and shift the energy of each state in a different way, leading, e.g., to the collective Lamb shift \[.\]. The collective scattering of light can strongly differ from the case of an assembly of noninteracting emitters \[ and has been predicted to be suppressed for a dense gas of cold two-level atoms \].

The collective response of interacting emitters is different between ensembles exhibiting inhomogeneous broadening, such as solid state systems \[ or thermal vapors \], and those that are homogeneously broadened, such as cold atom clouds \[.\] Ensembles of cold atoms allow in principle the study of collective scattering beyond the mean-field regime \[ when the atoms respond cooperatively. Several experiments in this direction have been reported recently that employ large and optically thick ensembles of cold atoms \[ and arrays of trapped ions \[.\] In particular, the experiments of Refs. \[ infer a modification of the collective scattering rate from the measurement of the radiation pressure force acting on a cloud. However, the important case of an atomic ensemble with a size comparable to the optical wavelength has not been studied experimentally, nor has the transition between the well-understood case of scattering by an individual atom \[ to collective scattering. In particular, the suppression of light scattering when the number of atoms increases in a regime of collective scattering has, to our knowledge, never been directly observed.

Here, we study--both experimentally and theoretically--the emergence of collective effects in the optical response of a cold atomic sample due to dipole-dipole interactions, as we gradually increase the number of atoms. To do so, we send low-intensity near-resonant laser light onto a cloud containing from 1 to about 450 cold $^{87}$Rb atoms and with a size comparable to the wavelength of the optical transition at $\lambda = 780$ nm. Starting from one atom, we observe a broadening of the line as the number of atoms increases, as well as a small red shift and a strong suppression of the amount of scattered light with respect to the case of noninteracting atoms. We show that this suppression is consistent with the measured broadening and shift. We find that our measurements are compatible with numerical simulations of the response of the system in the low excitation limit, accounting for all the scattering processes between atomic dipoles and the internal level structure of the atoms.

The suppression of light scattering by resonant dipole-dipole interactions can be understood qualitatively as follows. Consider an ensemble of classical radiating dipoles with resonance frequency $\omega_0 = 2\pi c/\lambda$ and radiative decay rate $\Gamma$ in the absence of interactions. When the ensemble of dipoles interact through the dipole-dipole potential, given by

$$V_{ji}^{\alpha\beta} = -V_{dd} \left[ p_{\alpha\beta}(1 - ikr) + q_{\alpha\beta}(kr)^2 \right] e^{ikr}, \quad (1)$$

one can describe the system as an ensemble of collective modes with various eigen-frequencies and decay rates. Here, $j$ and $l$ denote two dipoles separated by a distance $r$, $V_{dd} = 3\Gamma/4(kr)^3$, $k = 2\pi/\lambda$, and the angular functions $p_{\alpha\beta}$ and $q_{\alpha\beta}$ depend on the polarizations $\alpha$ and $\beta$ and the relative orientations of the dipoles $j$ and $l$. \[32, 23, 25\]. An incident laser with weak intensity couples better to modes with decay rates larger than $\Gamma$ (super-radiant modes). Combined to the fact that the eigen-frequencies then span a large range, this results in a
reduced excitation rate, and therefore a reduced scattering. This effect is all the more pronounced when the average distance between dipoles \(\langle r \rangle\) is smaller than \(\lambda/2\pi\). The argument also implies a broadening of the excitation line with respect to the case of non-interacting dipoles.

To study the collective scattering by an ensemble of atoms coupled via resonant dipole-dipole interactions we use the setup depicted in Fig. 1(a). We prepare small clouds containing up to 450 atoms at a temperature \(\sim 100 \mu\text{K}\), confined in a microscopic dipole trap \(\Gamma_0=0.3\text{ mK}\), and illuminate them with laser light nearly resonant with the atomic transition at \(\lambda = 780 \text{ nm}\). The Doppler width of the sample (150 kHz) is much smaller than the atomic linewidth \(\Gamma/2\pi = 6 \text{ MHz}\), making inhomogeneous broadening negligible. The anisotropy of the trap results into an elongated cloud with calculated root-mean-square thermal sizes \(\sigma_x = 0.3\lambda\) and \(\sigma_z = 2.4\lambda\). The maximal density is \(\rho = 2.5 \times 10^{14} \text{ at/cm}^3\) and the minimal average inter-atomic distance \(\langle r \rangle = \rho^{-1/3} = 0.2\lambda\). Figure 1(b) represents the distribution of nearest neighbors in a single stochastic realization of a cloud of 450 atoms. In this regime, \(k(r) \sim 1\), leading to \(V_{dd} \sim \Gamma_0\), and the resonant dipole-dipole interaction will therefore have an effect on the scattering.

Experimentally, we prepare the trapped atoms in the \(F = 2\) hyperfine manifold with an efficiency better than 95\%, release them in free space by switching off the trapping light, and illuminate them with \(\sigma_+\) polarized light at a frequency \(\omega = \omega_0 + \Delta\) tuned near the \((5S_1/2, F = 2)\) to \((5P_{3/2}, F' = 3)\) transition (see Fig. 1). In this way we avoid extra light-shifts induced by the trapping beam that would obscure the measurement of small collective shifts and broadening. Also, we choose the intensity saturation \(I/I_{\text{sat}}\) to be in the low excitation limit \((I_{\text{sat}} = 1.6 \text{ mW/cm}^2)\). We interleave excitation pulses with duration 125 ns and recapture periods in the dipole trap with duration 1 \(\mu\text{s}\). This sequence is repeated 200 times using the same cloud of atoms, in order to improve the duty cycle of the experiment. Finally, we prepare a new atomic sample and repeat the set of excitation pulses a few hundred times. The scattered light that we collect in the \(z\) direction is therefore the result of an average over many spatial configurations of the atoms. The choice in the number of pulses (200) is a trade-off between getting a good signal-to-noise ratio and avoiding light-assisted losses or heating of the cloud, both of which would lower the density. We ensured that both effects do not exceed 5\% over the entire set of pulses and that less than 5\% of the atoms are depumped in the \((5S_1/2, F = 1)\) hyperfine level during the excitation.

Figure 2 shows the number of photons \(n_x(N, \Delta)\) detected by the I-CCD as a function of the detuning \(\Delta\) of the excitation laser, for various atom numbers \(N\). A Lorentzian fit agrees well with the data for \(N \lesssim 300\). As expected from the qualitative argument described above, we observe that the full-width-at-half maximum (FWHM) increases with the number of atoms (see Fig. 3), since the interatomic distance then decreases, leading to stronger dipole-dipole interactions. We also measure a small red shift \(\delta \omega\) of the center frequency (Fig. 4). For \(N = 1\) atom, the FWHM is \(1.35 \pm 0.15\Gamma\), in agreement with the short duration of

![FIG. 1: (a) Experimental setup. The atoms are initially confined in a microscopic single-beam dipole trap (not shown) (wavelength 957 nm, depth 1 mK, and a waist 1.6 \(\mu\text{m}\), oscillation frequencies \(\omega_x = \omega_y = 2\pi \times 62 \text{ kHz}\) and \(\omega_z = 2\pi \times 8 \text{ kHz}\). The excitation laser propagates along the quantization axis \(z\), set by a \(B \sim 1 \text{ G}\) magnetic field. We collect the scattered light along \(z\), after a polarizer \(P\) oriented at an angle of 55\(^\circ\) with respect to \(x\), using a lens \(L\) with a large numerical aperture (NA\(\sim 0.5\)) and an image intensifier followed by a CCD camera (I-CCD). (b) Histogram of the distances to the nearest neighbor for \(N = 450\) atoms. (c) Structure of \(^{87}\text{Rb}\) atoms relevant to this work. The excitation light at frequency \(\omega\) is near-resonant with the transition at \(\lambda = 2\pi c/\omega_0 = 780 \text{ nm}\).]

![FIG. 2: Amount of scattered light detected \(n_x(N, \Delta)\), versus the detuning \(\Delta\) of the excitation light for numbers of atoms \(N = 1, 5, 20, 50, 200, 325, 450\) (from bottom to top). The amplitudes of the curves have been normalized to the amount of light detected at resonance for \(N = 1, n_x(1, \Delta = 0)\). The lines are Lorentzian fits to the data. Typical uncertainties: 10\% (vertically) and 20\% (horizontally).]
the excitation pulses (125 ns), which broadens slightly the resonance. Figure 2 also shows that the amount of light scattered in the z direction at resonance does not increase linearly with the number of atoms as one would expect for noninteracting atoms, but actually increases more slowly. Fig. 3(a) indicates that this is also the case off resonance, where we plot $n_z(N, \Delta)/n_z(1, \Delta)$ for different atom numbers and detunings. For noninteracting atoms this ratio is equal to the number of atoms $N$ (and is thus independent of the detuning $\Delta$), as we verified by collecting the scattered light after letting the atomic cloud expand in free space for a sufficiently long time [32]. By contrast, here we observe that the amount of scattered light is strongly suppressed on resonance as the number of atoms increases, and that we gradually recover the behavior of noninteracting atoms as we detune the laser away from resonance.

All the observations reported above can be reproduced by a unique functional form:

$$n_z(N, \Delta) = C \frac{N}{\Gamma_c(N)^2 + (\Delta - \delta \omega_c(N))^2},$$

for $N \lesssim 300$. This is illustrated in Fig. 3(b): we find that the quantity $R(N, \Delta)/R(1, \Delta)$, where $R(N, \Delta) = n_z(N, \Delta) \times |T_z^2 + (\Delta - \delta \omega_c)^2|$ and $\Gamma_c$ and $\delta \omega_c$ are respectively the phenomenological fits of FWHM and the shift (see Fig. 2) collapses on a single curve whatever the detuning. For $N \lesssim 300$, this curve is linear with $N$ with a slope of 1, in agreement with Eq. 2. It emphasizes that in this regime, the scattered intensity is suppressed by a factor $(\Gamma/\Gamma_c)^2$ at resonance. We note that this fact cannot be explained by a model where the suppression would come from an incoherent superposition of the intensities scattered by each atom with resonant frequencies inhomogeneously distributed over a distribution of width FWHM: that would lead to a suppression that would scale as $\Gamma/\Gamma_c$ near resonance, instead of the $(\Gamma/\Gamma_c)^2$ scaling observed here.

We now compare the experimental data with a theoretical model. We have performed numerical simulations of the collective many-atom response to near-resonant light that solve the radiative interactions in the low excitation limit essentially exactly. In this model, each atom, located at position $\mathbf{r}_j$ ($j = 1, \ldots, N$) and with dipole $\mathbf{d}_j$, is driven by the incident laser field and by the fields scattered by all the $N - 1$ other atoms, i.e., each dipole is coupled to the $N - 1$ other dipoles via the resonant interaction of Eq. (1). Each atom emits a dipole radiation with the characteristic short-range $e^{ikr}/r^3$ and the long-range $e^{ikr}/r$ contribution. This classical electrodynamics simulation approach incorporates all the radiative interactions between an ensemble of non saturated discrete dipoles. This approach has been used to study dielectric media comprising two-level or spatially averaged isotropic electric dipoles [10, 13, 16, 18, 19, 21, 33] as well as magneto-dielectric circuit resonator systems.

Here, we also incorporate into the scattering processes the Zeeman level structure of the atoms [12] and the shifts associated to the presence of a magnetic field. To calculate the dipoles $\mathbf{d}_j$ in our experimental configuration, we stochastically sample the positions of the atoms according to a Gaussian density distribution; each atomic position is treated as an independent and identically distributed random variable. At each realization we then have the $N$ atoms fixed at positions $\mathbf{r}_j$ ($j = 1, \ldots, N$). For each such realization, we also stochastically sample the magnetic quantum number of the Zeeman states $m_j$ of each atom $j$. The probability of atom $j$ being in state $|g, m\rangle$ ($m = \pm 2, \pm 1, 0$) is the initial population of that Zeeman state $p_m$ ($0 < p_m < 1$; $\sum_m p_m = 1$). The optical pumping, used in the preparation step before the excitation sequence, skews the initial populations; here we use the values $p_0 = p_1 = p_2 = 1/3$ and $p_{-1} = p_{-2} = 0$. Working in the low excitation limit, we write the positive frequency component of the dipole produced by each atom $j$ that oscillates at the dominant laser frequency as $\mathbf{d}_j = D \sum_{\sigma} \mathbf{\hat{e}}_\sigma C_{m_j}^{(\sigma)} |P_{j, \sigma}\rangle$, where the sum runs over the unit spherical polarization vectors $\sigma = \pm 1, 0$. The amplitude of the atomic dipole $j$ associated to the optical transition $|g, m_j\rangle \rightarrow |e, m_j + \sigma\rangle$ is proportional to the reduced dipole matrix element $D$, the atomic coherence $|P_{j, \sigma}\rangle$, and the corresponding Clebsch-Gordan coefficient $C_{m_j}^{(\sigma)}$. The steady-state coherences are given by the coupled set of equations

$$(\Delta_{ja} + i \Gamma/2) P_{ja} = \Omega_{ja} + \sum_{l \neq j} \sum_{\beta} C_{m_j}^{(\beta)} C_{m_l}^{(\alpha)} P_{j, \beta} C_{l, \alpha},$$

where $\Omega_{ja}$ is the Rabi frequency of the driving laser field on atom $j$, $\Delta_{ja} = \omega - \omega_{ja}$ is the detuning of the laser.
with respect to the Zeeman shifted transition of the \( \alpha \)-polarized atom \( j \) (with frequency \( \omega_{j\alpha} \)), and \( \beta = \pm 1, 0 \). The last term resonantly couples the \( \alpha \)-polarized dipole \( j \) to the \( \beta \)-polarized dipole \( l \) separated by \( r = r_j - r_l \) according to Eq. (1). We have solved Eqs. (2) numerically in the presence of a \( B = 1 \) G magnetic field to calculate the intensity that is coherently scattered into the solid angle encompassed by the aspherical lens in the far field. Finally, accounting for the polarizer in the detection, we calculated the measured light intensity.

The simulation predicts that the spectra \( n_z(N, \Delta) \) present an increasing broadening and asymmetry (Fig. 5), a negligible shift (Fig. 3b), as well as a suppression of the scattered light (Fig. 4) when the number of atoms increases. These features are in good agreement with our data for \( N \lesssim 50 \). In this range, the simulated spectra are well fitted by a Lorentzian for \( N \lesssim 50 \), thus justifying our fitting of the data by Eq. (2) and the collapse of the data shown in Fig. (1b). For \( N \gtrsim 50 \), the agreement is only qualitative, as the effects are found to be less pronounced experimentally. We attribute these discrepancies to two possible reasons. Firstly, the volume occupied by the atoms may increase with \( N \) during the sequence of pulsed excitations. This effect is hard to check experimentally since the sample is smaller than the diffraction limit of our imaging system. We found numerically, however, that an increase by a factor 2 in the widths \( \sigma_p \) and \( \sigma_z \) already restores a nearly Lorentzian profile close to the measured spectra (see Fig. 5), and yields the observed suppression of light scattering (see Fig. 4). Secondly, the simulation predicts that the number of detected photons increases by a factor 2 when the initial distribution of Zeeman state populations varies from \( p_0 = p_1 = p_2 = 1/3 \) to \( p_2 = 1 \). For large atom numbers, optical pumping during the set of excitation pulses may change the distribution of populations, an effect not accounted for in our model.

In conclusion, we have directly measured the suppression of light scattering induced by dipole-dipole interactions in an ensemble of cold atoms driven by a near-resonant weak laser field. We have found a good agreement with a model of coupled dipoles. In the future, we plan to investigate to what extent the observed collective scattering involves beyond-mean-field scattering processes, i.e. is cooperative in nature. Experimental investigations of the temporal response of the system, and comparisons to the case of a single atom [26], should also provide insight into the interplay between dipole-dipole interactions and collective scattering.

We acknowledge support from the E.U. through the ERC Starting Grant ARENA, from the Triangle de la Physique, EPSRC, and Leverhulme Trust. We thank P. Pillet, J.-J. Greffet, and J. Javanainen for fruitful discussions.

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