Light scattering and localization in an ultracold and dense atomic system

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The quantum optical response of high density ultracold atomic systems is critical to a wide range of fundamentally and technically important physical processes. These include quantum image storage, optically based quantum repeaters and ultracold molecule formation. We present here a microscopic analysis of the light scattering on such a system, and we compare it with a corresponding description based on macroscopic Maxwell theory. Results are discussed in the context of the spectral resonance structure, time-dependent response, and the light localization problem.

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Early studies of formation and dynamics of ultracold atomic samples were largely focused on obtaining the high density and low temperatures necessary to attain Bose-Einstein Condensation in atomic gas samples \cite{1}. At the same time, a large number of other research areas emerged from studies of ultracold atomic gas samples. Among these, there are several where physical processes are importantly modified at high atomic density. These include efforts in ultracold molecule formation \cite{2}, image storage in high-optical depth samples \cite{3}, and light storage and manipulation for possible atomic-physics based quantum repeaters \cite{4} and other quantum information applications. Another little explored area which may have significant impact in a range of scientific or technical areas is study of quantum optical processes at high density $n_0$, where $n_0 \lambda^3 \sim 1$ \cite{5}. Under these conditions, disorder-dependent phenomena such as an Anderson-type \cite{6} localization, random lasing \cite{7}, and other mesoscopic phenomena \cite{8,9,10} may appear. In these cases, the quantum optical response of the medium is of critical importance to dressing and probing the physical processes involved.

In this letter we are concerned with the collective dynamics of light and a sample of ultracold and high-density atoms. Under the conditions that $n_0 \lambda^3 \gtrsim 1$ the interaction of atomic dipoles via longitudinal and transverse electric fields strongly interferes with the scattering process. The atomic dipoles cannot be considered as independent secondary sources of the scattered waves freely propagating through the sample, as usually takes place for an optically dilute system. Here we present theoretical discussion of the scattering process based on two different and complementary approaches. In the first, we use a self-consistent description of the atomic sample in the spirit of the Debye-Mie model for a macroscopic spherical scatterer consisting of a homogeneous and dense configuration of atomic dipoles. We apply a self-consistent calculation of the macroscopic permittivity based on its relevant statistical expression given by the Kubo formula \cite{11}. In this model the scattered light is restored as secondary waves created by randomly distributed atomic dipoles, whose reradiation should be properly expanded in the spherical modes of the Debye-Mie problem. In the second approach we make exact numerical analysis of the completely quantum-posed description of the single photon scattering problem. The atoms, the incoming photon and the field modes participating in the process are considered in a second quantized formalism. This analysis gives us the exact value of the scattering amplitude and in particular shows the correct description of the cross section in those spectral domains where the Debye-Mie model fails or is not irreproachably applicable. We show the presence of collective microcavity structure, created by the atomic system, where a number of resonant modes have a sub-radiant nature, and discuss their physical substance in the context of light scattering and diffusion. The results have important implications for studies of light localization, and for manipulation of single and few photon states in ultracold atomic gases.

Light transport through a disordered system of atomic dipoles can be discussed in the Maxwell theory as a combination of the process of coherent propagation, described by configuration averaged Maxwell equations, and incoherent losses initiated by scattering from mesoscopically scaled permittivity fluctuations. In such a formalism one can visualize the light state as a local plane wave existing on a spatial scale $\sim \lambda$ and coherently interacting with the system of atomic dipoles located in a relevant mesoscopic volume. The dipoles interacting with the field are locally indistinguishable and the interaction process is assumed to be well approximated by the cooperative coherent response \cite{11} for the sample susceptibility $\chi(\omega)$ at frequency $\omega$. Then the permittivity $\varepsilon(\omega) = 1 + 4\pi\chi(\omega)$ can be extracted via a self-consistent approximation of the collective dipole dynamics driven by the transverse electric field and modified by the Lorentz-Lorenz effect coming from interference of
proximal atomic dipoles. If incoherent losses have only a radiative nature and atoms equally populate the ground state Zeeman sublevels we obtain a self-consistent expression for $\epsilon(\omega)$ for an infinite sample

$$
\epsilon(\omega) = \frac{1}{1 + \frac{4\pi n_0 |d_{F_0,F}|^2}{3\kappa}} \frac{1}{\frac{1}{1 + \frac{4\pi n_0 |d_{F_0,F}|^2}{3\kappa}}}
$$

This equation can be analytically solved and $\epsilon(\omega)$ expressed in terms of well defined external parameters, such as the natural decay rate $\gamma$ and $n_0$. The reduced matrix element $d_{F_0,F}$ for a given dipole transition can be also expressed by $\gamma$. We consider here a closed transition, such that for hyperfine levels only the selected lower and upper states with angular momenta $F_0$ and $F$ respectively can be radiatively. The above result can be applied to calculation of the scattering cross section of light by a spherical sample of large radius $a$ ($a \gg \lambda$) in the standard formalism of the Debye-Mie problem.

$$Q_S = \frac{\pi}{2k^2} \sum_{J=1}^{\infty} (2J+1) \left|1 - S_J^{(e)}\right|^2 + \left|1 - S_J^{(m)}\right|^2$$

$$Q_A = \frac{\pi}{2k^2} \sum_{J=1}^{\infty} (2J+1) \left|1 - S_J^{(e)}\right|^2 + \left|1 - S_J^{(m)}\right|^2$$

where the scattering matrix components for the TM and TE modes are respectively given by

$$S_J^{(e)} = -\frac{\epsilon(\omega)j_J(kr)[h_J^{(2)}(\bar{r})]^P_{r=a} - h_J^{(2)}(\bar{r})a]}{\epsilon(\omega)j_J(kr)[h_J^{(1)}(\bar{r})]^P_{r=a} - h_J^{(1)}(\bar{r})a]}$$

$$S_J^{(m)} = -\frac{j_J(kr)[h_J^{(2)}(\bar{r})]^P_{r=a} - h_J^{(2)}(\bar{r})a]}{j_J(kr)[h_J^{(1)}(\bar{r})]^P_{r=a} - h_J^{(1)}(\bar{r})a]}$$

Here $j_J(\ldots)$, $h_J^{(1)}(\ldots)$ and $h_J^{(2)}(\ldots)$ are spherical Bessel functions of $J$-th order, and $k = \sqrt{\epsilon(\omega)}\omega/c$. $Q_S$ is the elastic part of the cross section responsible for the coherent scattering of light from the sample boundary. The absorption part $Q_A$ is responsible for diffusely scattered light via the incoherent channel and the sum $Q_0 = Q_S + Q_A$ is the total cross section for the entire scattering process. Light emerging the sample via incoherent channels can be recovered in the Maxwell theory by considering secondary and multiply scattered waves generated by fluctuations of $\chi(\omega)$. This can be simulated by a Monte-Carlo scheme and crucially requires that the dipole sources generating these waves be indistinguishable on a mesoscopic distance $\sim \lambda$. Then secondary and multiply scattered waves can be simulated through the time dependence of the sample fluorescence when the excitation light is turned off. The analysis of the transient process and time dependence of the fluorescence is simplified by a diffuse approximation if the extinction length for the field penetration inside the sample is $\gg \lambda$.

We now turn to the quantum-posed description of the photon scattering problem. There photon scattering on an atomic system is expressed by the following relation between the total cross section and the T-matrix

$$Q_0 = \frac{\lambda^2}{\hbar^2c^4 (2\pi)^2} \int \sum_{\epsilon'} |T_{\epsilon'g \epsilon} e^{i\omega \epsilon} (E_i + i\epsilon)|^2 d\Omega$$

where $\mathcal{V}$ is a quantization volume. Here we keep only the Rayleigh channel and assumed that the atomic system is described by the same ground state $g$ before and after the scattering process, which includes the averaging over initial and sum over final Zeeman states. The initial energy of the entire system $E_i$ is given by $E_i = E_g + \hbar \omega$ and the incoming and outgoing photons have the same frequency $\omega' = \omega$. We consider below the simplest relevant example of a "two-level" atom, which has only one sublevel in its ground state and three Zeeman sublevels in its excited state, such that $F_0 = 0$ and $F = 1$ and in an isotropic situation the total cross section does not depend on the momentum direction and polarization of the incoming photon.

The T-matrix is expressed by the total Hamiltonian $H = H_0 + V$ and by its interaction part $V$ as

$$T(E) = V + V\frac{1}{E - H}V$$

In the rotating wave approximation the internal resolvent operator contributes to $\Pi$ only by being projected on the states consisting of single atom excitation, distributed over the ensemble, and the vacuum state for all the field modes. Defining such a projector as $P$ the projected resolvent

$$\tilde{R}(E) = P\frac{1}{E - H}P$$

performs a $3N \times 3N$ matrix, where $N$ is the number of atoms. For the dipole-type interaction between atoms and field this projected resolvent can be found as the reversed matrix of the following operator

$$\tilde{R}^{-1}(E) = P \left(E - H_0 - V Q \frac{1}{E - H_0} V Q \right) P$$

where the complementary projector $Q = 1 - P$, operating in the self-energy term, can generate only two types of intermediate states: a single photon + all the atoms in the ground state; and a single photon + two different atoms in the excited state and others are in the ground. For such particular projections there is the following important constraint on the dipole-type interaction $V$: $P VP = Q V Q = 0$. Due to this constraint the series for the reversed resolvent $\tilde{R}$ is expressed by a finite number of terms, which would be not the case in a general situation $\Pi$. The resolvent $\tilde{R}(E)$ can be numerically calculated and, for an atomic system consisting of a macroscopic number of atoms, when $N \gg 1$,
the microscopically calculated cross section \( \frac{\lambda}{n} \) can be compared with the Debye-Mie theory results \( \frac{\lambda}{n} \). Both the microscopic and self-consistent approaches have no fitting parameters. Compared with the mesoscopic approach, the essential difference is that in the microscopic approach any type of collective atomic excitations can contribute, which are not responsively cooperative. One can then expect significant difference in their predictions for the cross section and for the fluorescence behavior. The time dependence of the fluorescence signal can be built up via Fourier expansion of the outgoing photon wavepacket with the \( S \)-matrix formalism.

Below we consider the spectral dependence of the cross sections, calculated in both the self-consistent mesoscopic model and in an exact microscopic approach. Our central idea is to follow how this dependence is modified when the dimensionless density of atoms \( n_0 \lambda^3 \) is varied from smaller to greater values. In Fig. 1 we reproduce such a spectral dependence when the density of atoms is small, \( n_0 \lambda^3 = 0.02 \). There is excellent agreement between the Debye-Mie and microscopically calculated data and the microscopic result is insensitive to configuration averaging over random atomic distributions. This justifies that only cooperative modes, which allow the macroscopic Maxwell description, contribute to the scattering process. As a consequence the long term time dynamics of the fluorescence signal can be very well approximated by a Holstein mode. Such a spectral and temporal behavior is typical for dilute atomic systems as was verified by our numerical simulations done for the atomic ensembles of different sizes and consisting of different numbers of atoms. While varying the density to greater values \( n_0 \lambda^3 \sim n_c \sim 0.09 \) the solution of the self-consistent equation \( 1 \) turns the permittivity to negative values in a part of the spectrum where \( \epsilon' < 0 \) and \( \epsilon'' = 0 \). As is well known in, e.g. plasma physics \( 1,11 \) the negative permittivity can be associated with a forbidden spectral zone, where the radiation cannot penetrate inside the system and can exist only in the form of a surface light-matter wave. In the limit of even higher density samples, the light undergoes mainly surface scattering and the absorption (incoherent/diffusion scattering) channels suppressed. This is illustrated in Fig. 2 by those spectral dependencies of the cross section, which were calculated in the Debye-Mie model for \( n_0 \lambda^3 = 0.5 \). In contrast with similar results in Fig. 1 the contribution of the elastic channel now dominates such that a smaller portion of light can penetrate the sample. As a consequence, it is expected that fewer atomic excitations should contribute to the time dependent fluorescence decay and the self-consistent model predicts an even faster decay than would take place in a similar dilute system.

The situation can be differently posed if it is considered from a microscopic point of view. Then such a dense atomic vapor can be relevantly described as a microcavity system built up in an environment of randomly distributed atomic scatterers. Any distribution creates a specific quantization problem for the incoming field, whose mode structure can be properly defined in terms of standard scattering theory. The main difficulty for the quantization procedure is description of the complicated structure of the resonance states. The resonances are de-
The dilute system has a sample radius $a = 27\lambda$, and density $n_0\lambda^3 = 0.015$, and for the dense system these characteristics are given by $a = 5\lambda$ and $n_0\lambda^3 = 0.5$. Color online.

The analogy of sub-radiant states with a localization nature and manifest themselves via significantly slowed long time decay of the fluorescence in comparison with a classical Holstein mode. Fig. 3 illustrates the difference in the fluorescence decay for dilute and dense atomic systems initially excited by a short probe pulse. The external parameters for the systems are such that the optical depth for both the systems is nearly the same. For a dilute system the long term asymptote, extracted through exact microscopic calculations, is well described by a diffuse Holstein-type mode evaluated via the sample macroscopic characteristics. For the dense system there is evident deviation of the asymptotic behavior from such behavior. Such a deviation should be associated with the presence of sub-radiant resonance states in the resolvent poles.

The analogy of sub-radiant states with a localization process, usually discussed as a multiple wave scattering problem, suggests evolution with density of the qualitative scattering properties of the atomic gas. The discussed characteristics, such as the spectral dependence of the sample cross-section or the time-dependent fluorescence, have no direct relations with static thermodynamic properties of the system, which could be evaluated based on its partition function. However, they describe how the system in equilibrium responds to optical excitation near the atomic resonance transition. The atomic transition is dressed by cooperative dipole-field interactions and at higher densities, the disordered atomic system reveals a micro-cavity structure. Such a micro-cavity has unique properties for any particular configuration and its mode splitting is competitive with the mode decay rate. The modification of the dynamic response to an external field with density is an intrinsic property of the system, which may be visualized in terms of a cross over or a change of phase. In this sense our calculations show that the transformation of the system behavior from one of individual, independent atomic scatterers to the cooperatively organized micro-cavity structure mainly develops within a narrow and critical density zone. The configuration sensitive speckle structure of the spectral cross section manifests itself in those conditions when the self-consistent permittivity becomes negative in part of the spectrum. The description of the atomic system with the macroscopic Maxwell theory preferably yields surface scattering of light in this case. More precise microscopic description shows that part of the excitation can penetrate the system and be converted into a long-time decay of the fluorescence signal.

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