Density-dependent modifications of the transition spectrum of an atom located inside cold atomic ensemble

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Abstract. On the basis of general theoretical results developed in our group previously [Sokolov I M *et al* 2011 *J. Exp. Theor. Phys.* **112** 246], the transition spectrum of a single atom inside cold atomic ensemble is analyzed under conditions when the averaged interatomic separation is less than or comparable with the wavelength of resonant radiation. Density-dependent shifts of resonance as well as distortion of the spectral shape of the atomic transition are investigated.

1. Introduction

Influence of the environment on atomic transition spectrum has attracted considerable interest both because of its fundamental importance and because of its significance in different applications such as quantum metrology, quantum information science, and lasing in disordered media [1] – [8]. These influences are of special interest for realization of highly stable and accurate optical atomic clocks [9] – [14], particularly neutral-atom-based optical frequency standards [15] – [20].

The question of density-dependent spectral line shifts for cold atomic gases recently gave rise to a wide discussion [21] – [31], and this question is especially important for optical frequency standards [17], [20]. In case when atomic density is quite big (so that average interatomic separation is comparable with wavelength of resonant light) spectral line shift caused by dipole-dipole interaction can be comparable with natural line width. In the vast majority of experiments the external quasi-resonant radiation excites whole atomic ensemble. Therefore, most of theoretical papers are focused on the analysis of this case.

In the present paper we will discuss local excitation inside cold atomic ensemble. It can be experimentally realized by illuminating of the ensemble with two narrow and off-resonant orthogonally propagated light beams. Each beam does not cause single-photon excitation, but their simultaneous interaction with atoms in the crossing region can cause two-photon excitation from the ground S to excited D state if conditions of two-photon resonance are satisfied. The subsequent spontaneous transition from D to P state leads to population of the studied P state. The thereby described method allows obtaining small cluster of excited atoms in the middle of the cloud. For simplicity thereafter in the paper we will consider the case of only one atom excitation. Note that possibilities of two-photon excitation $5s\ S-2(1/2) \rightarrow 5p\ P-2(j) \rightarrow 5d\ D-2\ (j)'$ of rubidium atoms have been already studied experimentally [32]. In our opinion the one-atom spectral response analyzed in this paper can be the foundation of a new practical method of studying of collective effects in dense media.

The main goal of the present work is to study theoretically the transition spectrum of an atom excited in a dense and cold atomic ensemble using quantum microscopic approach developed in our group [33]. A great advantage of this approach is that interatomic correlations are taken into account, including many-body correlations. Correct description of correlations is achieved by taking into account the discrete structure of the medium without the approximation of continuous medium.

2. Basic equations

The calculation of atomic transition spectrum in this paper will be made on the basis of a microscopic quantum approach developed in [33]. Here we will mention only the main approximations of this approach and will show the principal analytical expressions utilized for numerical calculations.

We consider an ensemble consisting of N motionless atoms. All atoms have the ground state with $J = 0$; $m = 0$ and the excited level with $J = 1$; $m = -1, 0, +1$. This model of atomic levels allows us to take into account the polarization properties of electromagnetic field correctly. For detailed comparison of this model and scalar model see [34], [35].

In the framework of quantum microscopic approach [33] we solve the non-stationary Schrodinger equation for the wave function of the joint system consisting of atoms and the field generated in the process of the evolution.

$$i\hbar \frac{\partial \psi}{\partial t} = H\psi \tag{1}$$

The Hamiltonian of the joint system can be presented as $H = H_0 + V$ where H_0 is and the sum of Hamiltonians of free atoms and free field, V is the operator of interaction between atoms and electromagnetic field. In the framework of dipole approximation used here, the operator of interaction V can be written as follows

$$V = -\sum_a \boldsymbol{d}^{(a)} \boldsymbol{E}(\boldsymbol{r}_a) \tag{2}$$

In this formula $\boldsymbol{d}^{(a)}$ is the dipole moment operator of the atom a, $\boldsymbol{E}(\boldsymbol{r})$ is the field operator.

We will seek the wave function ψ as an expansion in a set of eigenstates $\{|l\rangle\}$ of the operator H_0:

$$\psi = \sum_l b_l(t)|l\rangle \tag{3}$$

Here, the subscript l defines the state of all atoms and the field.

An important approximation used in the microscopic approach is in restriction of the total number of states $|l\rangle$ taken into account. We will calculate all radiative correction up to the second order of the fine structure constant. In this case we can consider only the following states (see [36]):

1) $\psi_g = |g, g, \dots, g\rangle|\boldsymbol{k}\alpha\rangle$ all atoms are in the ground state, and 1 photon is in the system
2) $\psi_{e_m} = |g, g, \dots, g, e_m, g, \dots, g\rangle|vac\rangle$ 1 atom is in the excited state, other atoms are in the ground state, and no photon in the system
3) $\psi_{e_m e'_{m'}} = |g, \dots, g, e_m, g, \dots, g, e'_{m'}, g, \dots, g\rangle|\boldsymbol{k}\alpha\rangle$ nonresonant states with two excited atoms and one photon

Here the index e indicates the number of excited atom, $e = 1,2, \dots, N$ and the index m indicates the angular momentum projection, $m = -1,0,+1$. The total number of one-fold excited states ψ_{e_m} is $3N$. The last states $\psi_{e_m e'_{m'}}$ are necessary for a correct description of the dipole-dipole interaction at short interatomic distances.

The restriction of the total number of states allows us to covert the infinite system of equations for quantum amplitudes of states of the joint system b_l to finite system $3N \times 3N$ for the amplitudes of the

one-fold atomic excited states b_{e_m}. The amplitudes of other quantum states b_g and $b_{e_m e'_{m'}}$ can be obtained via b_{e_m}. It allows us to obtain the wave function of the joint system.

In this paper we analyze the transition spectrum of one atom excited inside dense and cold atomic ensemble at the initial time. Let us consider the excited atom located in the center of the ensemble and we denote this atom number 1. For determinacy we will assume that at the initial time $t = 0$ only one substate $m = -1$ of the excited atom is populated. All the other atoms of the ensemble are in their ground state. Electromagnetic field at $t = 0$ is in the vacuum state. So we consider initial conditions as follows: $b_g(0) = 0;\ b_{e_m e'_{m'}}(0) = 0;\ b_{e_m}|_{e=1;m=-1} = 1;\ b_{e_m}|_{e=1;m=0,+1} = 0;\ b_{e_m}|_{e=1,2,\ldots,N;m} = 0$. Hereafter we will associate the Fourier-amplitude $b_0(\omega)$ with Zeeman sublevel $m = -1$ of the central atom $e = 1$.

The transition spectrum $b_0(\omega)$ contains fluctuations connected with random spatial configuration of atoms in the ensemble. In the real experiments the residual thermal motion of atoms makes all these configurations uniformly distributed. Therefore the experimental result is averaged over random spatial configurations of atoms in the ensemble. Accordingly, we perform multiple Monte-Carlo averaging of $b_0(\omega)$ over random spatial configurations of atoms.

3. Results and discussion

Interaction of a radiating atom with its surroundings causes a shift of the atomic transition as well as distortion of the spectral line. The specific type of transition spectrum depends both on the atomic density and on the size of the ensemble, i.e., on the number of atoms in it. We have analyzed how the function $b_0(\omega)$ changes with the number of atoms for the given density and found that for small ensembles when the mean-free path of photon less or comparable with linear size of atomic ensemble these changes are very essential. As the number of atom and linear size increase the changes in $b_0(\omega)$ become more and more weak. This dependence has an evident tendency to saturation. In this section we will discuss sufficiently big atomic ensembles when $b_0(\omega)$ for the given atomic density does not practically depend on the number of atoms. So results obtained in this paper can be used for description of single-atom excitation inside any macroscopic ensemble with reasonable accuracy. The saturation is a simplest scaling law; it is a feature of local excitation. For the excitation of whole atomic ensemble by probe radiation the scaling law is more complex. In [37] we have analyzed this law for cross section of scattered light.

Figure 1 shows the imaginary part of $b_0(\omega)$ (absorption curve) for different atomic densities. It demonstrates that the absorption curve is significantly modified as compared with the classical Lorenz curve typical for a free atom (dot-dashed curve). The maximum of the spectral line is shifted, moreover we observe both red and blue shift depending on the atomic density. The atomic densities $n = 0.05, 0.1, 0.2$ have been analyzed in [31] and we observed red shifts for all these values.

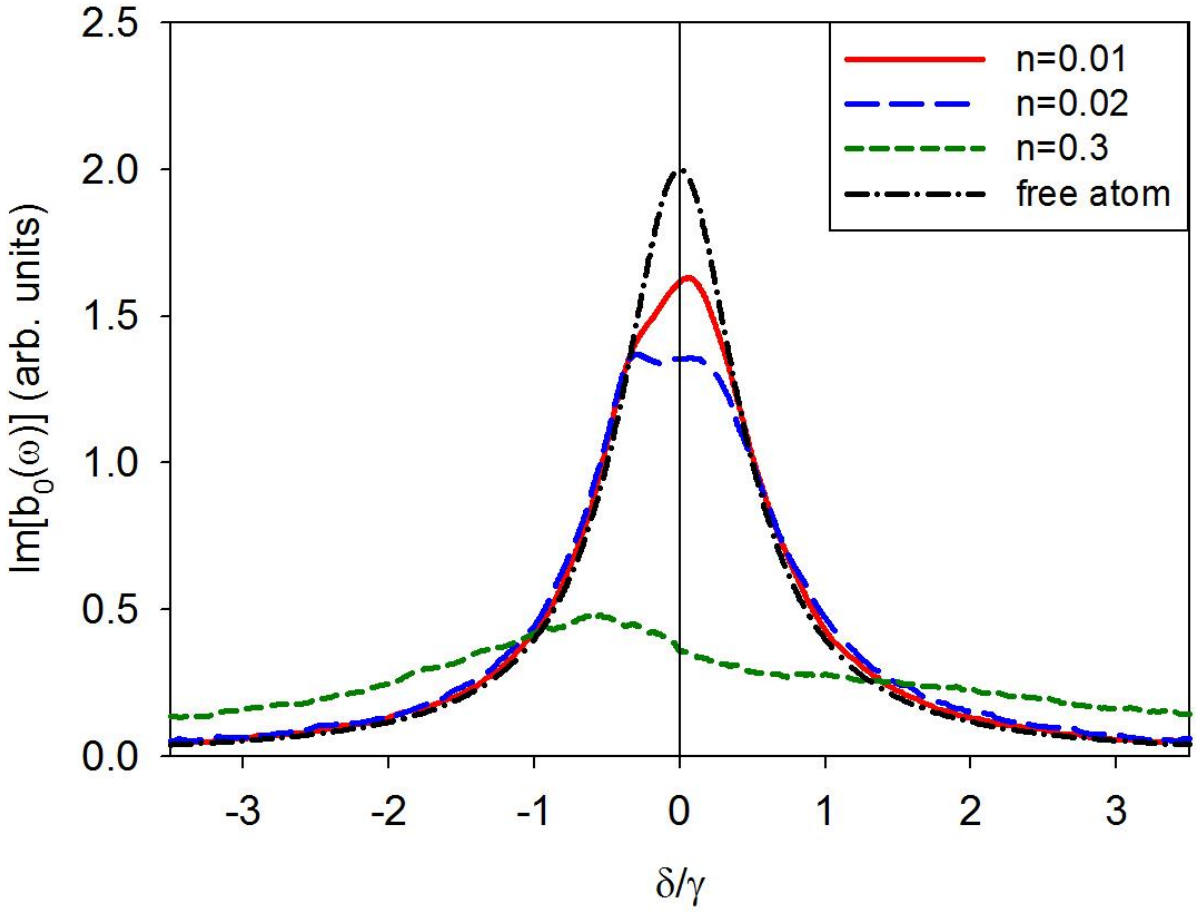

Figure 1. Absorption curve corresponding to an atom in ensemble that is excited at initial time; γ is the natural line width of free atom; $\delta = \omega - \omega_0$, where ω_0 is the resonant transition frequency of free atom; n is the atomic density in units k_0^3 where $k_0 = \omega_0/c$

Increasing the density of surrounding atoms causes enhancement of collective effects and consequently more considerable modification of atomic transition spectrum. It is clearly confirmed by the curve corresponding to the density $n = 0.3$ on the figure 1. The contour has relatively wide wings as compared with a free atom.

Figure 2 shows the real part of $b_0(\omega)$ (dispersion curve) for the same densities as figure 1. It also illustrates the enhancement of modifications of atomic transition spectrum caused by collective effects as density increases.

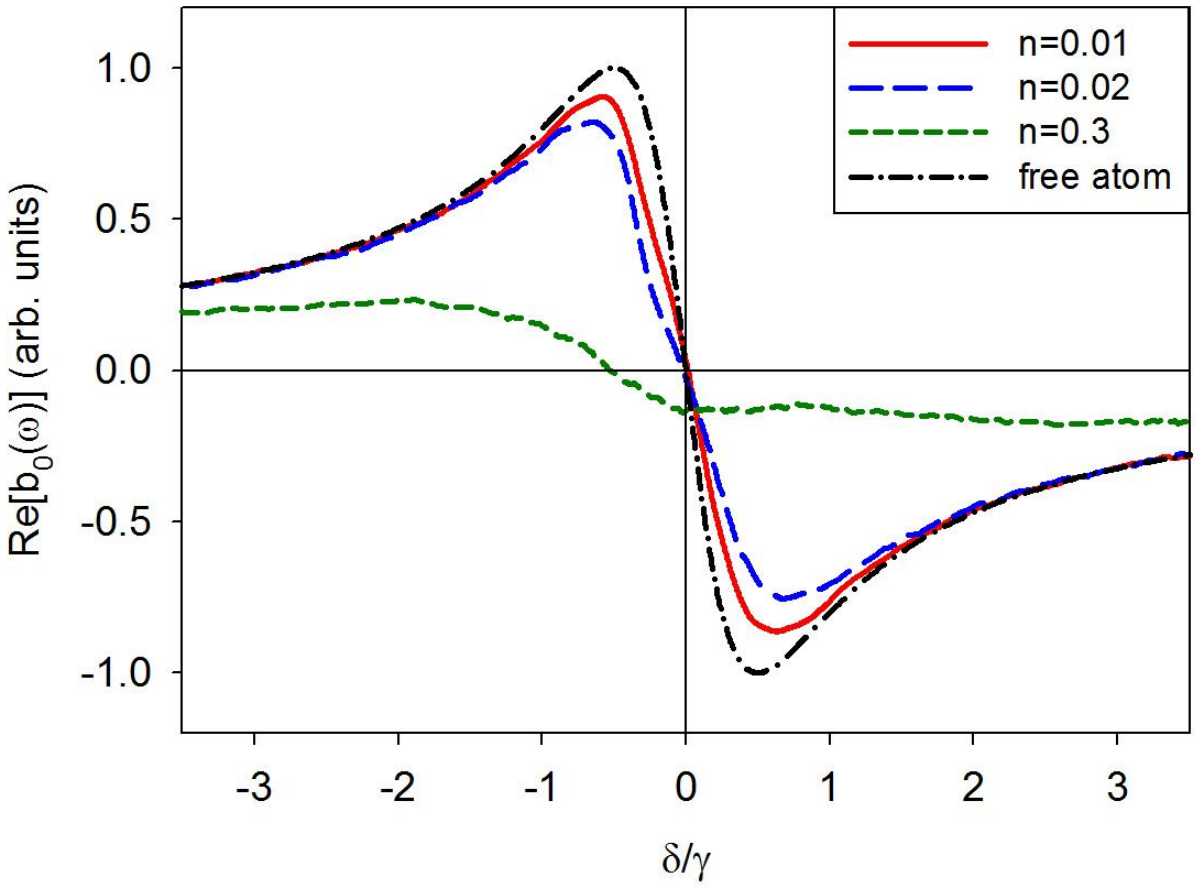

Figure 2. Dispersion curve corresponding to an atom in ensemble that is excited at initial time; all parameters are the same as in figure 1

The exciting feature of collective effects caused by resonant dipole-dipole interaction is that different observables have different spectral dependencies for dense atomic ensembles and these dependencies transform differently as density increases. Thus, the spectrum of the dielectric permittivity of cold and dense atomic clouds has a blue shift [28], [38], [39]. The maximal probability to excite the dense ensemble is observed for a negative detuning of exciting radiation [40]. The calculation of the total scattering cross section shows that the corresponding spectrum has several resonances [33]. Two of them are red shifted, and the third is in the blue region. Spectral dependence of fluorescence also has complicated behavior. It depends both on direction of fluorescence and on its polarization [33]. By now density dependence of spectral properties of cold clouds has been studied in several experiments [17], [41], [42], in which laser-induced fluorescence of atomic ensemble was studied. Experiments showed red shift and some distortion of the excitation spectrum.

4. Conclusion

In this paper we analyze the influence of the dipole-dipole interatomic interaction on the transition spectrum of an excited atom in cold atomic ensemble under conditions when the averaged interatomic

separation is less than, or comparable with, the wavelength of resonance radiation. Both red and blue shifts of resonance are obtained depending on the atomic density. The distortion of spectral shape of atomic transition is investigated.

The calculations were made on the basis of a quantum approach taking into account the vector nature of the electromagnetic field and Zeeman structure of atomic sublevels. A continuous medium approximation was not used. It allowed us to take into consideration random inhomogeneity of the atomic system and consequently the existing interatomic correlations.

Together with [31] the results of this paper give sufficiently complete information about the dependence of transition spectrum of an atom in cold atomic ensemble on the atomic density. In our opinion the results obtained in this paper are very important for future improvement in quantum frequency standards based on optical transitions in cold atomic ensembles. For optimization of these devices the density dependence of the main characteristics has to be taken into account.

Note that the model of motionless atoms used here can describe not only cold atoms but impurity centers in solids as well. Therefore the results can be relevant in the area of lasing from randomly disordered medium as well as in the investigation of lasers based on quantum dots. There are also a number of proposals about using cold atomic ensembles and ensembles of impurity centers in solids in quantum-information applications. Large coherence time makes possible to use such systems as elements of quantum memory.

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