Dipole–dipole interactions in a hot atomic vapor and in an ultracold gas of Rydberg atoms

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Abstract. In our paper ideal and non-ideal gas media of neutral atoms are analyzed. The first we discuss a dipole broadening of atomic transitions in excited dilute and dense metal vapors. Then the theoretical studies of the dipole–dipole interactions in dense ultracold gas of Rydberg atoms are considered. Possible future experiments on a base of our experimental arrangement are suggested.

1. Introduction
Studies of many–body interactions in a gas media with strong coupling between particles are very interesting and useful for fundamental science and for many different applications. To identify thermodynamic properties of a gas media by strength of inter-particle interactions it was introduced the useful dimensionless parameter [1]

$$\Lambda = \frac{V}{k_B T}. \quad (1)$$

Here $V$ is an average interaction potential, $T$ is a temperature of particles, $k_B$ is the Boltzmann factor. When parameter $\Lambda \ll 1$ it can be identified as an ideal gas medium, when $\Lambda \gg 1$ it is strong coupling non-ideal gas medium. Usually a gas media of charged particles with Coulomb type of interactions are investigated [1]. We would like to discuss gas media with dipole–dipole interactions between neutral particles. It is clear that such interactions are much weaker than Coulomb interactions.

The dipole–dipole interactions occur between atoms in different energy states with different parity. Between these atomic states the electro-dipole transitions are allowed. The inter-atomic interaction potential is described by the next relation [2]

$$V_d(r) = \frac{\vec{d}_1 \cdot \vec{d}_2 + 3(\vec{n} \cdot \vec{d}_1)(\vec{n} \cdot \vec{d}_2)}{r^3}, \quad (2)$$

where vectors $\vec{d}_1$ and $\vec{d}_2$ are dipole moments of the particles, $\vec{n}$ is unit vector in the direction joining the two particles, $r$ is distance between atoms. The interactions between the ground state atoms and excited atoms are quite well studied [3]. The average potential $V_d$ for a resonance gas with density $N$ can be estimated by using expression

$$V_d = d^2 N. \quad (3)$$
In hot gases \((T > 300 \text{ K})\) the potential \(V_d\) is much less than the energy of atomic thermal motion \(k_B T (\Lambda \ll 1)\). Therefore the dipole–dipole interactions induce only the spectral broadening of atomic transitions \(\Gamma = KN\). The factor \(K\) equals to \(4\pi\alpha d^2/\hbar\) \((\alpha \approx 1)\). The dipole broadening \(\Gamma\) at different atomic density \(N\) was measured for many chemical elements in gas phases [3]. The most suitable gases for experimental and theoretical investigations are atomic vapors of alkali metals. The alkali atoms have only one valence electron like hydrogen and as result they have simple energy spectra.

The dipole–dipole interactions can be characterized by ratio of dipole broadening \(\Gamma\) and Doppler broadening \(\Delta_D = 2\omega v_{th}/c\), where \(\omega\) is resonance frequency, \(c\) is speed of light, \(v_{th} = [k_B T/(2m)]^{1/2}\) is thermal velocity of atoms with mass \(m\). It is convenient for analysis to use dimensionless parameter

\[
h = KN/\Delta_D. \tag{4}
\]

The atomic vapor which characterized by small parameter \(h (h \ll 1)\) can be defined as a dilute gas and the atomic vapor with large parameter \(h (h \gg 1)\) can be defined as a dense gas [4]. In dilute gas the spectral profiles of atomic transitions are described by inhomogeneous broadening, in the dense gas the spectral profiles are homogeneously broadened. The dipole broadening in partly excited resonance gases is different in dilute and dense gases [4]. In the dilute atomic vapor \((h \ll 1)\) under standard experimental conditions the width \(\Gamma\) can only weakly depend on optical saturation. The possible induced variations are less than 20\% [4–7]. In the dense gas \((h \gg 1)\) the width can be strongly reduced in partly excited atomic vapor [4, 8–11].

The study of ultracold Rydberg atoms can reveal new opportunities and effects. Such atoms have unique properties as long radiative lifetime and large dipole moments [12]:

- In rarefied gas of ultracold Rydberg atoms \((h \ll 1)\), the interatomic interactions can induce energy shifts of Rydberg quantum states in neighboring Rydberg atoms [13–15]. This effect is used to create the Rydberg blockade which is perspective for applications in the field of quantum information.
- In dense gas of ultracold Rydberg atoms \((h \gg 1)\), the dipole–dipole interactions can produce variations of positions of the Rydberg atoms. Under these conditions chains and periodic structures can appear in the dense gas of ultracold Rydberg atoms due to attractive interactions [16, 17]. In presented paper our studies of dipole–dipole interactions in hot atomic vapor and in ultracold gas of Rydberg atoms are discussed.

2. Hot atomic vapor

In this part of the paper our results of experimental investigations of dipole broadening in optically excited atomic vapors are presented. Selective reflection spectroscopy was applied for measurements of optical properties of opaque metal vapors. The spectra of the selective reflection from vapor cells were recorded for different intensities of laser beam near the normal incidence (the incidence angle \(\Theta \ll 1\)). By using analytical expression for reflection resonances the spectral widths of atomic transitions were estimated. As result the dependence of the dipole broadening on the incident intensity was obtained.

2.1. Dilute vapor

In our experiment low density \(^{87}\text{Rb}\) vapor was served as a dilute gas \((h \ll 1)\). The experimental setup is described in paper [6]. The laser frequency was tuned at isolated atomic transition \(5S_{1/2} (F = 2)\) to \(5P_{1/2} (F' = 2)\) in \(D_1\) line of \(^{87}\text{Rb}\) \((\lambda = 780 \text{ nm})\). The atomic number density \(N = 2 \times 10^{14} \text{ cm}^{-3} (\Gamma/(2\pi) = 15 \text{ MHz} [18])\) was selected. At this density the parameter \(h\) is order of \(4 \times 10^{-2}\). The optical beam from external cavity diode laser was sent into the window of hot glass cell with rubidium vapor. The reflected beam was sent to photodetector. In spectra of the
selective reflection a sub-Doppler structure is appeared due to collisions atoms with a window surface [19]. The frequency modulation (FM) technique [6, 7, 19, 20] allows to obtain narrow Doppler-free resonances at atomic transitions with homogeneous width $W(0) = (\Gamma_{\text{nat}} + \Gamma)/(2\pi)$, where $\Gamma_{\text{nat}}$ is the natural width of the atomic transition ($\Gamma_{\text{nat}}/(2\pi) = 6$ MHz). This approach is called Doppler-free FM SR spectroscopy [7] or Doppler-free reflection spectroscopy [18, 20]. Under condition of the optical saturation in work [7] was obtained an expression for spectral broadening

$$[W(I)]^2 = [W(0)]^2(1 + I/I_{\text{sat}}).$$

Here $I$ is intensity of the optical beam, $I_{\text{sat}}$ is the saturation intensity. The experimental data and fit are presented in figure 1.

At the minimum intensity of 1.1 mW cm$^{-2}$ the width $W(I)$ is 27 MHz and at the maximum intensity of 74 mW cm$^{-2}$ it is 46.7 MHz. The data can be fitted by the linear function (5). The slope is linear with accuracy 15%. It is confirmed that dipole broadening $\Gamma$ can only weakly depend on optical excitation in dilute gases. Similar results were obtained in [7] with dilute cesium vapor. We shall note that due to the inhomogeneous broadening of the atomic transition only a small part of the rubidium and cesium atoms were excited to the atomic upper states. Obtained experimental results are in an agreement with results of calculations in the frame of theoretical model for the impact atomic collisions [5].

2.2. Dense vapor

For study a dense gas it was used hot potassium vapor ($h \gg 1$). The natural abundance consists two stable isotopes: $^{39}$K (73%) and $^{41}$K (7%). Here we will present experimental data for D$_1$ line ($\lambda = 770$ nm) with selected number density $N = 2.3 \times 10^{17}$ cm$^{-3}$. Description of experimental technique is presented in [9]. In the experiment two lasers were used: pump Ti–sapphire laser, which detuned for 50 GHz from D$_1$ line, and probe diode laser. The frequency of the probe laser was modulated. We recorded spectra of the FM selective reflection from interface of window–potassium. Then experimental spectra were compared with theoretical calculations. As result we can evaluate the main parameters of the spectral profile in optically excited dense vapor (the dipole broadening $\Gamma$ and excitation parameter $\eta$). The width is $W(I) = \Gamma/(2\pi)$. The dependence of the measured width on optical intensity is shown in figure 2.
Figure 2. Spectral width $W(I)$ vs optical intensity $I$ in dense potassium vapor.

The spectral width is decreased with optical intensity. One can say that dipole broadening can be reduced by more than two times. The relation between the width and intensity $I$ is nonlinear. In [9] it was demonstrated that dependence of $\Gamma$ on $\eta$ is expressed by linear function

$$\Gamma = \Gamma_0 (a + b\eta).$$

Here excitation parameter $\eta = (N_g - N_e)/N$, $N_g$ and $N_e$ are number densities of atoms in the ground state and the excited state. The maximum value of $\eta$ is equal to 1 for negligible optical saturation ($I = 0$). The analysis of experiments with potassium [9] and rubidium [10] confirmed the relation (6).

In dense vapors the dipole–dipole interactions cannot be described by model of impact collisions. The interaction time cannot be neglected. The dipole interaction between atoms is similar to quasi-molecular interaction. Interactions between excited atoms in dense gases are very weak (similar to van der Waals interactions) to compare with strong dipole–dipole interactions between excited and ground state atoms [8, 11]. As result the dipole broadening in the excited atomic vapor is proportional to the ground state population [8]

$$\Gamma = KN_g(\eta).$$

If only external degrees of freedom of atoms are considered, the dense atomic vapor will show the same thermodynamic properties as the ideal gas. If internal degrees, for example, quantum energy states of atoms, are considered, the dense atomic gas will demonstrate strong coupling between atoms. The differences in the properties of dilute and dense atomic vapors have to be taken into account especially when optical properties of such gases are important to know.

3. Ultracold gas of Rydberg atoms

The Rydberg atoms have unique properties as long radiative lifetime of the excitation and large dipole moments [12]. For ultracold atoms these properties allow to get such density $N_{Ry}$ of Rydberg atoms when criteria for non-ideal neutral gas will be satisfied ($\Lambda \geq 1$). According to [16] the parameter of non-ideality for gas of ultracold Rydberg atoms can be written as

$$\Lambda = d_{Ry}^2 N_{Ry}/(k_B T) \sim n^2 (e a_0)^2 N_{Ry}/(k_B T),$$

where $d_{Ry}$ is the Rydberg length, $a_0$ is the Bohr radius, $n$ is the density of atoms, $k_B$ is the Boltzmann constant, and $T$ is the temperature.
Figure 3. Spatial distribution of the Rydberg atoms for $\Lambda = 0.075$. Pink and dark blue colors of the spheres indicate the poles of the Rydberg dipoles.

Figure 4. Resonance on two-photon transition $2S-120S$ in $^7$Li atoms.

where $e$ is charge of electron, $a_0$ is the Bohr radius. It was calculated that for Rydberg gas with density $N_{Ry} = 10^{10}$ cm$^{-3}$, $T = 10^{-4}$ K, $n = 100$ the parameter $\Lambda = 1$. Figure 3 shows one of the possible spatial distribution of ultracold Rydberg atoms for $\Lambda = 0.085$. One can see that clusters and chains start to be formed. When parameter $\Lambda$ will be large enough then probably new structures will appear.

Our goal is experimental preparation and investigation of self-organized three-dimensional structures from ultracold dense gas of Rydberg atoms (Rydberg matter [21, 22]). In order to resolve this problem (to get $\Lambda \geq 1$) it is necessarily to prepare the required density of ultracold Rydberg atoms with principal quantum number $n \geq 100$. By using to two-photon excitation of $^7$Li atoms we observed resonances corresponding the Rydberg states $nS$ with $n$ from 38 to 165 [23] and the states $nP$ for $n$ with 42 to 114 [24]. The spectral resolution was limited by the spectral linewidth 30 MHz of uv laser. For effective population transfer from the atomic ground state to long-lived highly-excited state it is required a narrow band excitation. Recently the
linewidth was made less than 1 MHz. The narrow resonances on two-photon transition \(2S\rightarrow38D\) with width less than 6 MHz were observed [25]. In figure 4 resonance on \(2S\rightarrow120S\) transition is presented.

In figure 4 the solid curve shows the observed resonance. The dashed curve is a fit by Gaussian function. The spectral width of the resonance (FWHM) is 9 MHz. The temperature of Rydberg atoms \(T\approx0.5\) mK, Rydberg atoms number density \(N_{\text{Ry}}\sim10^5\) cm\(^{-3}\). The low density is due to low excitation efficiency in cw excitation regime.

4. Conclusion

In presented paper we discussed our studies of dipole–dipole interactions in hot atomic vapor and in ultracold gas of Rydberg atoms. As the next step in our project we would like to use a special sequence of intense optical pulses in order to realize the stimulated Raman adiabatic passage (STIRAP) for an efficient population transfer of the ground state atoms to Rydberg states [26]. Also a phase-space density of lithium atoms in a trap should be improved [27].

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References

[1] Fortov V, Iakubov I and Khrapak A 2006 *Physics of Strongly Coupled Plasma* (Oxford: Oxford University Press)
[2] Landau L D and Lifschitz E M 1959 *Quantum Mechanics, Non-relativistic Theory* (Oxford: Pergamon Press)
[3] Lewis E L 1980 *Phys. Rep.* 58 1–71
[4] Sautenkov V A 2016 *J. Phys.: Conf. Ser.* 774 012126
[5] Vdovin Yu A and Dobrodeev N A 1969 *Sov. Phys. JETP* 28 544–8
[6] Akulshin A M, Celikov A A, Sautenkov V A, Vartanian T A and Velichansky V L 1991 *Opt. Commun.* 85 21–5
[7] Vuletic V, Sautenkov V A, Zimmerman C and Hansch T W 1994 *Opt. Commun.* 108 77–83
[8] Sautenkov V A, van Kampen H, Eiel E R and Woerdman J P 1996 *Phys. Rev. Lett.* 77 3327
[9] van Kampen H, Sautenkov V A, Smeets C J C, Eiel E R and Woerdman J P 1999 *Phys. Rev. A* 59 271–4
[10] Li H, Sautenkov V A, Rostovtsev Y V and Scully M O 2009 *J. Phys. B: At., Mol. Opt. Phys.* 42 065203
[11] Sautenkov V A 2011 *Laser Phys. Lett.* 8 771–81
[12] Gallagher T F 1994 *Rydberg Atoms* (Cambridge: Cambridge University Press)
[13] Comparat D and Pillet P 2010 *J. Opt. Soc. Am. B* 27 A208–A232
[14] Sautenkov V A, Varzhpetyan T S, Li H, Sarkisyan D and Scully M O 2010 *J. Russ. Laser Res.* 31 270–5
[15] Manykin E A, Ozhovan M I and Poluektov P P 1981 *Phys. Dokl.* 26 974
[16] Manykin E A, Zelener B V and Zelener B V 2010 *JETP Lett.* 92 630–45
[17] Zelener B B, Saakyan S A, Sautenkov V A, Manykin E A, Zelener B V and Fortov V E 2015 *J. Exp. Theor. Phys.* 121 950–4
[18] Zelener B B, Saakyan S A, Sautenkov V A, Manykin E A, Zelener B V and Fortov V E 2016 *J. Exp. Theor. Phys.* 122 645–9
[19] Sautenkov V A, Saakyan S A, Vilshanskaya E V, Zelener B B and Zelener B V 2017 *J. Russ. Laser Res.* 38 11–5
[20] Deiglmayr J, Reetz-Lamour M, Amthor T, Westermann S, De Oliveira A L and Weidemüller M 2006 *Opt. Commun.* 264 293–8
[21] Gross N and Khaykovich L 2008 *Phys. Rev. A* 77 023604