On the saturation of excitation-dependent dipole-dipole broadening in dense Rb vapour

Andrei Bobrov[1*], *Vladimir* Sautenkov[1], *Sergey* Saakyan[1], and *Boris* Zelener[1,2]

[1]Joint Institute for High Temperatures, Russian Academy of Sciences, 13, Izhorskaya St. 125412, Moscow, Russia
[2]National Research Nuclear University, Moscow Engineering Physics Institute, 31, Kashirskoye Chaussee, 115409, Moscow, Russia

Abstract. We study dependence of dipole-dipole interaction in dense Rb vapour on optical excitation. We measure the dipole-dipole contribution to the selective reflection spectrum of D2 line. Optical excitation is created by intense laser beam, which depletes the $5S_{1/2}$ population and decreases the dipole-dipole interaction, narrowing the observed spectrum. The limit of the dipole-dipole interaction change is determined by the optical saturation of the D2 transition and corresponds to the $5S_{1/2}$ population approximately equal to 1/3.

1 Introduction

In a gas of identical atoms the dipole-dipole interaction between an excited atom and an atom in the ground state can lead to the excitation of the ground state atom and, correspondingly, to de-excitation of the excited atom. The probability of this non-radiative excitation transfer process is proportional to the gas number density. For sufficiently high densities this process is the dominant process of spectral line broadening. For the first time the dipole-dipole broadening was studied in [1], where simple classical analogy for the process of excitation transfer was discussed as well as rigorous quantum mechanical study.

Recently, due to the development of quantum modelling and computing, there is growing interest to study dipole-dipole interactions in dense gases. In [2] many-body physics of atomic dipoles with collective dissipation and long-ranged interactions was studied. Tuning of the dipole-dipole interactions by rapid change of gas density was investigated in [3]. In [4] manipulating of dipolar interaction in confined geometries was discussed.

In this paper we continue our research [5-7] on the controlling the dipole-dipole interaction in dense gases, where the dipole-dipole broadening is much greater than the Doppler broadening. Unlike the other works [3] in our study we change the magnitude of the dipole-dipole interaction not by changing the gas density, but instead by change of ground state population of the atoms. The dependence of the dipole-dipole interaction on the media excitation was first studied experimentally in [8]. In our work in order to control the dipole-dipole interaction by changing the ground state population, we use optical excitation of the Rb vapour in the high-temperature cell by intense pump laser radiation.

* Corresponding author: abobrov@inbox.ru

As a diagnostic technique to measure the dipole-dipole interactions magnitude we used registration of the broadening of the selective reflection spectra of weak probe laser beam, directed to the pump beam spot at the window of the cell. This technique has an advantage over the absorption studies [2-4, 9] because it does not require using of nanocells. Control of the dipole-dipole interaction magnitude is achieved by varying the pump laser intensity. In the previous works we studied broadening of a selective reflection spectrum by a single laser beam [5,6]. Then we studied the broadening by using a probe beam [7].

In the current paper we experimentally investigate limits of the method of the optical excitation control of the dipole-dipole interactions and estimate the ground state population when the dipole-dipole broadening is saturated.

2 Experimental Setup and Methods

The core component of our experimental setup is the high temperature gas cell with natural abundance of the rubidium isotopes. The vapour cell is mounted inside the heater, and its temperature is maintained by a thermostat. The number density of the rubidium atoms vapour is determined by the temperature of the coldest spot of the cell (indicated by the arrow in fig. 1).

To create the optical excitation, the intense pump laser radiation resonant with Rb D2-line is directed to the cell window. The weak probe radiation is directed to the pump beam spot at the window to measure how the excitation change the selective reflection spectra. The pump and probe laser beams are combined in a polarizing beam splitter (PBS) cube and focused on the vapour cell windows by a lens with a focal length of 200 mm. Both beams are near normal in respect to the window surface plane. The focused beams have elliptical shape with $w_{(x,y)}^{pump} = (135, 57)\ \mu$m for the pump beam and $w_{(x,y)}^{probe} = (88, 56)\ \mu$m for the probe beam.

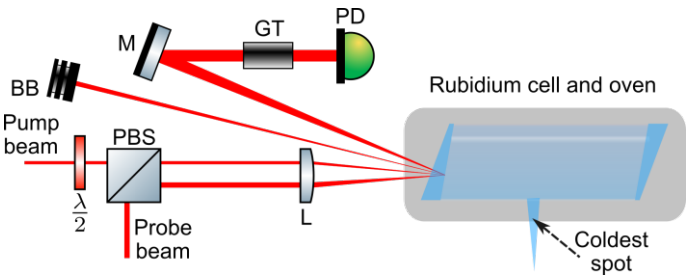

Fig. 1. Experimental setup for selective reflection pump and probe spectroscopy. The probe and the pump beam combined into polarizing beam splitter cube (PBS), focused into gas cell by using a lens (L). The reflected beam are filtered with narrow band mirror (M) and Glan-Thompson polarizer (GT) and recorded by photodiode (PD) and a scope. High-power pump beam, after reflection, guided into beam-block (BB).

The intensity of the pump beam I_{pump} was varied between 0 and 9 kW/cm^2 using a motorized half-wave plate and PBS. The probe beam intensity was kept below 4 W/cm^2 to avoid the saturation. The pump laser frequency was controlled with an accuracy of approximately 2 MHz by the Angstrom/High-Finesse WS-U wavemeter.

The pump beam was at resonance with Rb D2-line $5S_{1/2}$ (F=3) – $5P_{3/2}$ (F'=4) transition. To obtain the selective reflection spectrum, we measure frequency dependence of the probe beam reflection coefficient R by scanning the probe frequency detuning Δ_{pr} in vicinity of the resonance.

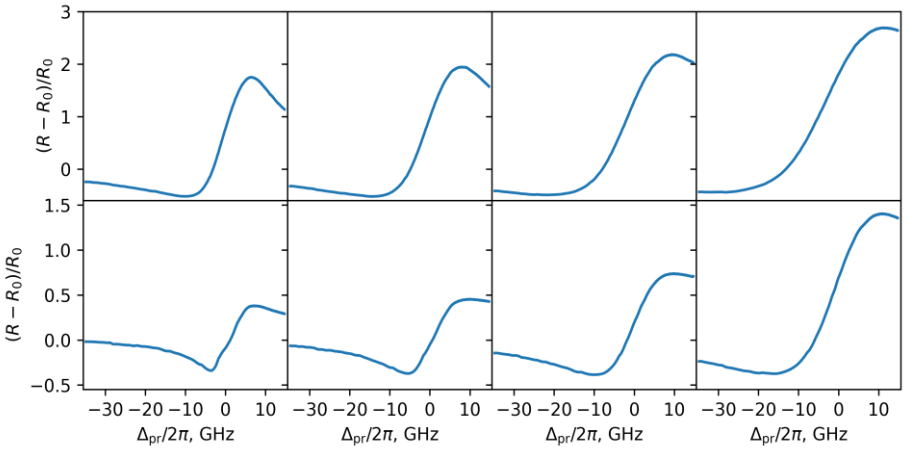

Fig. 2. Selective reflection coefficient vs probe beam detuning Δ_{pr} for different number densities $N = \{1.2, 1.7, 2.5, 3.6\} \times 10^{17}$ cm^{-3} from left to the right column. The upper row corresponds to the zero pump laser intensity, the lower row corresponds to the pump beam intensity $I_{pump} = 1.6$ kW/cm^2.

3 Results and Discussions

The dipole-dipole interaction is the dominant contribution to the broadening of the observed selected reflection spectra. The dipole-dipole broadening can be estimated as [5]:

$$\delta_{dd}(I_{pump}, N) = KN_g(I_{pump}, N) = K\eta(I_{pump})N,$$

where N_g is the number density of the atoms in the ground state ($5S_{1/2}$ in our case), N is the rubidium number density, η is the ground state population, K is constant. For the rubidium D2 line K can be estimated [10] as $K/2\pi = (1.1 \pm 0.17) \times 10^{-7}$ Hz/cm^{-3}.

Several samples of experimental selective reflection spectrum for several densities, with and without the pump radiation, are shown in fig. 2. For convenience we plot reduced reflection coefficient $(R - R_0)/R_0$, where R_0 is the reflection coefficient of the cell window. As can be seen, due to reduction of the ground state density N_g, the spectral width is reduced when the pump laser is on.

In fig. 3 the dependence of the selective reflection spectral width $\Delta\omega_{SR}$ on the atomic density is shown for several values of the pump radiation intensity I_{pump}. The dependencies are well fitted by linear functions.

We can approximately present the total selective reflection spectral width as a sum of the dipole-dipole broadening and broadening δ_0 corresponding to the other sources, such as Doppler broadening or unresolved hyperfine structure contribution:

$$\Delta\omega_{SR} = \delta_{dd}(I_{pump}, N) + \delta_0.$$

In the right side of the last equation only δ_{dd} depends on the gas density, so the derivative of the $\Delta\omega_{SR}$ on density is determined only by the derivative of the δ_{dd}:

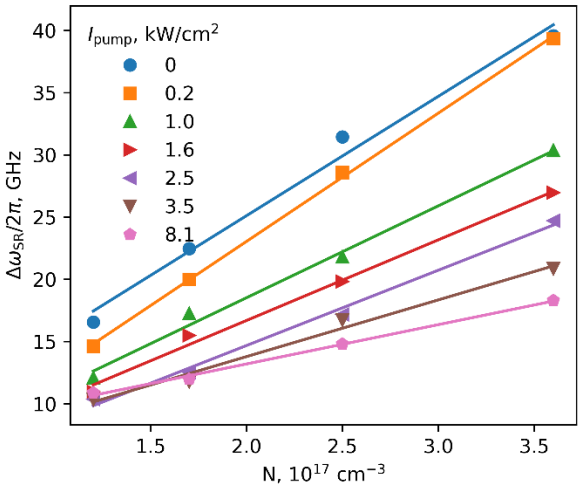

Fig. 3. Selective reflection width for different pump laser intensities I_{pump} versus Rb number density. Points are the experimental measurements; lines are linear fits.

$$\frac{\partial \Delta\omega_{SR}}{\partial N} = K\eta.$$

We see that, indeed, the derivative of $\Delta\omega_{SR}$ is constant in respect to N, so the dependence of $\Delta\omega_{SR}$ on the density should be linear. From the last equation we can easily obtain the ground state population η by dividing by K. In fig. 4 we plot η calculated from the slopes of the linear fits of the experimental results, shown in fig. 3.

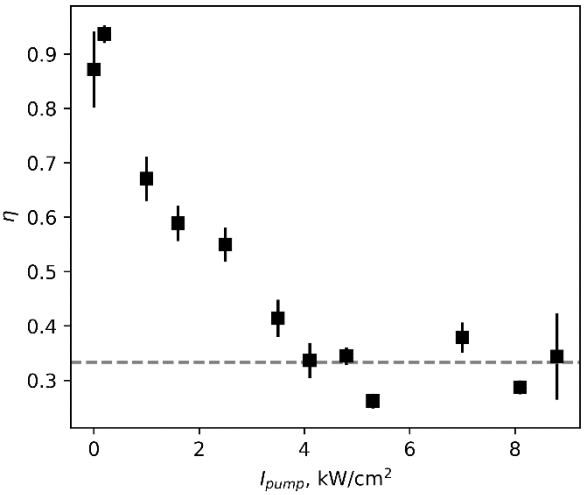

Fig. 4. Ground state $5S_{1/2}$ population η. Points are obtained from the slopes of the linear fits of the experimental results in fig. 3 (see text). Line corresponds to $\eta = 1/3$.

In our previous work [7] we made a suggestion that a dipole-dipole broadening stops to decrease with increasing intensity of a pump laser due to saturation of the optical transition. Now we have clear experimental evidence of this. From fig. 4 it can be seen that for high values of I_{pump} the calculated ground state density approaches value 1/3. This corresponds

to 2:1 ratio of population of the excited state to the population of the ground state, which is exactly the statistical weight ratio for the $5S_{1/2} - 5P_{3/2}$ transition.

4 Conclusion and Outlook

In conclusion, we studied possibility of control of the dipole-dipole interaction in a dense resonance gas using optical excitation. This method has advantage on the method of changing the density of the gas itself, since it is more precise. We changed the excitation level by intense pump radiation. It is all optical control, although, the limit of the dipole-dipole interaction change in this case is determined by the optical saturation. The limit can be extended by choosing a transition with greater ratio of statistical weights of the corresponding states.

As the experimental technique to obtain the dipole-dipole interaction magnitude we used measurement of the dipole-dipole contribution to the broadening of the selective reflection spectrum. This allowed us to conduct the experiments in a usual gas cell. Absorption measurements require nanocells since the absorption length for the densities considered here is about the wavelength of the laser radiation.

Acknowledgements

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