Selective reflection study of excitation-dependent dipole-dipole interaction in dense rubidium 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, 125412 Izhorskaya Street 13, Moscow, Russia
[2]National Research Nuclear University -- Moscow Engineering Physics Institute, 115409 Kashirskoye Chaussee 31, Moscow, Russia

Abstract. We conducted a comprehensive study of excitation dependent dipole-dipole interaction in dense rubidium gas using the resonant selective reflection of weak probe radiation from the dielectric-rubidium vapor interface. The excitation was created by the pump laser beam. The study involved exploring a diverse range of pumping radiation intensity and detuning for the number density of the rubidium vapour 2.8×10^{16} cm^{-3}. We have measured experimental selective reflection spectra in both linear and nonlinear pumping modes.

1 Introduction

The phenomenon of resonance behaviour in the reflection of monochromatic resonant light from the internal surface of a gas cell window was first demonstrated in experiments involving mercury vapour [1]. This discovery gave rise to what we now know as selective reflection (SR). This effect has significantly enhanced the study of dense atomic gases, which are non-transparent to resonant light. SR has emerged as a valuable tool of research, offering nuanced insights into the intricate atomic interactions within these dense environments.

In this paper we apply the SR technique to study a dipole-dipole (DD) interaction in a dense gas. The DD interaction occurs between the two identical atoms which are close enough to each other. If one atom is in an exited state and the other is in the ground state then nonzero probability of interaction exchange exists, i. e. the atom in the ground state becomes excited and the atom in the excited state falls to the ground state. The probability of this resonant process depends on the interatomic space separation and is proportional to the atomic number density. The classical analogy of this effect is the coupled Lorentz oscillating dipoles interaction [2]. The DD interaction is important in such phenomena as excitation blockade [3] or collective Lamb shift [4]. Knowing how to control the DD interaction can be of interest in developing new quantum devices [5].

* Corresponding author: abobrov@inbox.ru

The DD interaction leads to broadening of the SR spectra. If density is high enough, then the DD broadening becomes the dominant contribution to the spectral width and measuring the spectral width allows us to obtain information on the DD interaction strength. Since the dipole-dipole interaction involves one atom in the ground state and another atom in the excited state, the DD interaction is proportional to the ground state population. This was shown for the first time in [6]. Here we study experimentally how the DD interaction can be controlled by changing the ground state atomic population in the high-density rubidium cell. In order to change the population of the ground $5S_{1/2}$ state we use the pump laser driving the $5S_{1/2} - 5P_{3/2}$ transition. Unlike our previous work [7,8,9] where we registered the SR spectra of the single pump beam itself, in the present study we create excitation in the vapor by intense pump radiation and then register the SR spectra of the weak probe laser beam on the same transition, focused to the pump beam spot. In the next section we provide the details of our experimental setup.

2 Experimental Setup and Methods

The key component of our experimental setup is the high-temperature vapor cell containing a natural abundance of rubidium isotopes. We employ a sapphire cell with YAG windows, heated to a temperature of $T = 307°C$. The number density in units cm^{-3}, derived from the vapor-pressure model [10], is expressed as:

$$N(T) = 101325 \times 10^{-6} \frac{10^{4.312 - 4040/T}}{k_B T}, \qquad (1)$$

where T represents the temperature of the coldest part of the cell in Kelvins. The factor 101325 converts the vapour pressure from atm to Pa. Our investigation involves the selective reflection of a probe laser beam from the YAG-vapor interface within the glass cell. This reflection is measured across various values of pump beam intensity and detuning in proximity to the D2-line transitions of rubidium.

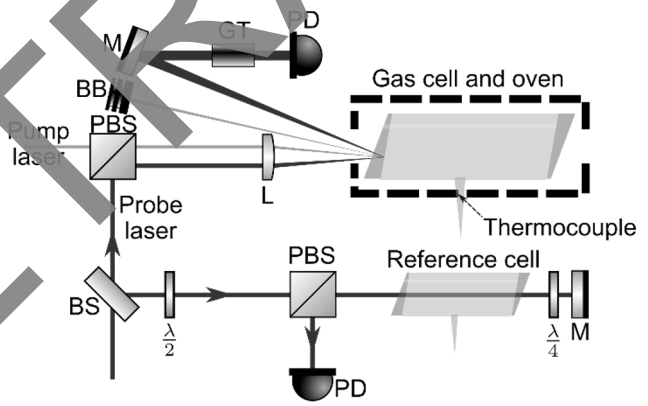

Fig. 1. The optical layout of the experimental setup for selective reflection measurements. The setup involves a combination of two orthogonally polarized laser beams, designated as the pump beam and the probe beam, through a polarizing beam splitter (PBS). These combined beams are then focused into a high temperature vapor cell using a lens (L) with a 200 mm focal length. The reflected probe beam is directed to the photodiode (PD) via a mirror (M) and a Glan-Thompson polarizer (GT). Meanwhile, the reflected high-power pump beam is obstructed by a beam block (BB). For precise control of the absolute frequency of the probe laser beam, we measure the saturated absorption spectrum of rubidium in a room temperature reference cell simultaneously with the SR spectrum.

For our pump and probe spectroscopy measurements, we employ two external cavity diode lasers. Radiations of the pump laser is further amplified using a semiconductor tapered amplifier to achieve an output power exceeding 1.5 W. The optical configuration of our experimental setup is shown in Fig. 1. Laser beams from the high-power pump laser and the probe laser are orthogonally polarized and combined using a polarizing beam splitter cube. Both beams are directed onto the YAG-vapor interface of the high-temperature cell through a lens having a 200 mm focal length. In the focal region, the pump beam and the probe beams overlap, forming ellipse-shaped beams with waists $w_{0x(y)}^{pump} = 135(56)\ \mu m$ and $w_{0x(y)}^{pr} = 88(56)\ \mu m$, respectively, at the e^{-2} level. Peak intensity of the beams is calculated as $I = 2P\left(\pi w_{0x} w_{0y}\right)^{-1}$. Intensity of the pump beam is varied using a motorized half-waveplate and a polarizing beam splitter, while the probe beam intensity is maintained below $I_{pr} = 4\text{W/cm}^2$.

Frequency of the pump laser beam is accurately determined by means of a calibrated wavemeter (Angstrom/High-Finesse WS-U) having absolute precision of 2 MHz [11]. Simultaneously, the saturated absorption spectrum (SAS) of the probe beam is registered together with the SR-spectrum, serving as a reference for absolute frequency calibration. The SR spectrum is recorded within a high-density vapor cell positioned inside an oven, while the SAS spectrum is obtained using an additional room temperature reference cell with natural abundance of rubidium isotopes. The probe laser's mode-hop-free tuning range is controlled by means of a Fabry—Perot cavity having a free spectral range of 1.2 GHz.

The reflected probe beam from the YAG-vapor interface of the glass cell is filtered using a narrow-band mirror and Glan-Thompson polarizer. The reflection coefficient was defined as a ratio of the probe beam intensity and the reflected signal intensity measured by a photodiode. The reflection coefficient was normalized relative to the non-resonant reflection coefficient from the YAG-vacuum interface, which is denoted as $R_0 = 8.5\%$ [12], resulting in $\delta R = (R - R_0)/R_0$. All the measurements are automated using a Python script, facilitating the acquisition of multiple SR-curves for different values of detuning and intensity.

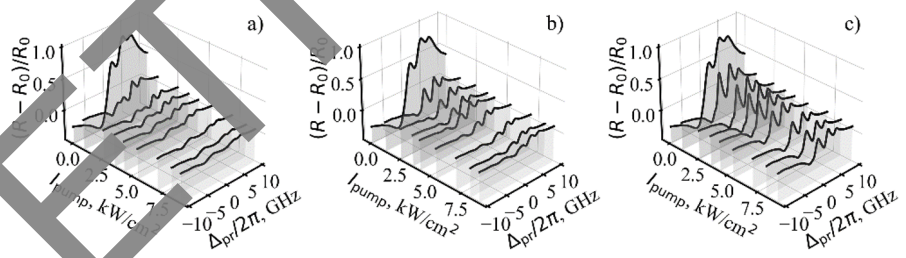

Fig. 2. Spectral dependence of selective reflection coefficient for the probe beam measured at various values of the pump beam detuning $\Delta_{pump}/2\,\pi = 0$ GHz (a), -2.2 GHz (b) and -8.2 GHz (c). Each panel features several selected curves depicting different pump beam intensities I_{pump}. In all the figures zero detuning of the probe beam and the pump beam $\Delta_{pr(pump)}/2\,\pi$, corresponds to the ^{85}Rb $5S_{1/2}$(F=3)-$5P_{3/2}$(F'=4) hyper-fine transition frequency.

3 Results and Discussions

The DD broadening of the SR spectral line is proportional to the ground state number density N_g and can be estimated as$\Gamma/2\,\pi = KN_g$. For the D_2-line of the Rb in the natural mixture studied here, the factor $K \approx 10^{-7}$ Hz cm^3. Without optical excitation by the pump beam, when all atoms are in the ground state, the DD broadening value for the atomic vapor

density considered is $\Gamma/2\pi \approx 3$ GHz, which is of order magnitude of ^{85}Rb hyper-fine splitting. The DD broadening in our experiments is much greater than natural linewidth of 6×10^{-3} GHz as well as the Doppler broadening, which is about 0.5 GHz.

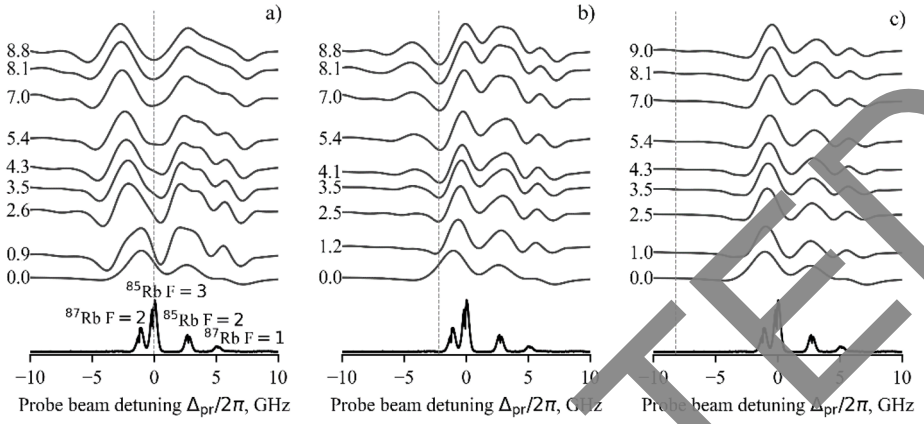

Fig. 3. Spectral dependence of the frequency derivative $dR/d\nu$ of the reflection coefficient for various values of pump beam detuning $\Delta_{pump}/2\pi =$ 0 GHz (a), -2.2 GHz (b) and for -8.2 GHz (c). Each figure shows several selected curves corresponding to different values of pump beam intensity I_{pump} indicated at the left side of each curve kW/cm^2. Dashed lines indicate the detuning values of the pump laser. The bottom panel presents the SAS in the reference cell.

In fig. 2 the SR spectra for the various values of the pump beam intensity and three values of detuning are presented as well as the reference spectrum with the pump beam turned off (i. e. with zero pump beam intensity). Zero detuning of the probe beam and the pump beam in all the figures, $\Delta_{pr(pump)}/2\pi$, corresponds to the ^{85}Rb $5S_{1/2}$(F=3)-$5P_{3/2}$(F'=4) hyper-fine transition frequency. As can be seen from the SR spectrum at the zero pump beam intensity, the hyper-fine components of D_2 line of Rb isotopes are overlapped and are practically unresolved. An increase of the pump intensity leads to the ground $5S_{1/2}$ state depopulation, that in turn leads to decrease of the DD broadening.

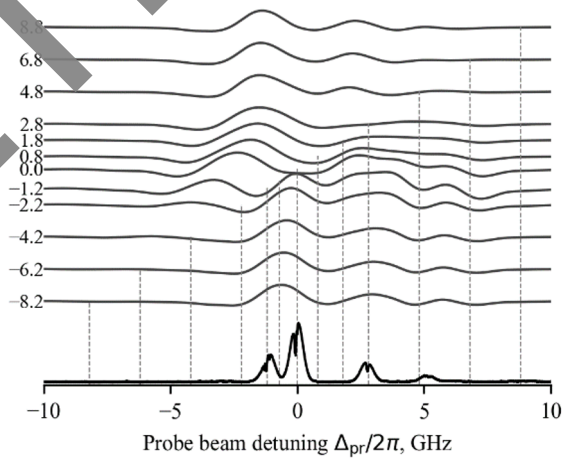

Fig. 4. Spectral dependence of frequency derivative $dR/d\nu$ of the reflection coefficient for the fixed pump intensity $I_{pump} = 5.4$ kW/cm^2 and different values of pump beam detuning $\Delta_{pump}/2\pi$ in GHz

For incoherent pumping and for large pump detuning in fig. 2(c) the decrease of the DD broadening leads to the hyper-fine components becoming more distinguishable. Of greater interest is the behaviour of the SR spectra for coherent pumping, i.e. in case when the pump detuning is low or zero, which is shown in Fig. 2(b) and 2(a). The strong pump field leads not only to lowering of the DD interaction, but also to power related nonlinear spectra changes. These changes in the spectra are more evident in Fig. 3 where the derivatives of the SR spectra are shown.

Fig. 4 shows the SR spectra for the fixed pump intensity. For high absolute values of the pump detuning, the spectra are identical regardless of the detuning sign. But for low absolute values there is asymmetry between the red and blue shifted pump.

4 Conclusion and Outlook

In conclusion, we present an experimental study of the possibility of control of the DD interactions in a dense gas. In order to tune the strength of the DD interaction we changed the ground state atomic population by means of optical excitation with the pump laser radiation. As a diagnostic technique we employed the SR spectroscopy by a weak probe laser beam. Increasing the pump intensity, we observed narrowing of SR spectra, which corresponds to a decrease of DD broadening, i. e. a decrease of the DD interaction strength. For coherent pumping with high intensity laser beam we observed intensity-dependent effects in the SR spectra.

The non-linear experimental spectra obtained cannot yet be described by the existing theoretical models. The theoretical description is complicated because of absence of any small parameter in the system since the high gas density and high laser intensity result in necessity to take into account both the optical saturation effect and field attenuation effect simultaneously. The existing models of SR spectroscopy were developed either in case of high laser intensity and low medium density [13,14] or in case of high density and negligible optical saturation [15]. The research presented here complements our previous experimental studies [7,8,9] where we investigated the non-linear SR spectra of the dense Rb vapour by means of single laser beam. We hope that our research will shed some light on the problem of interaction of intense resonance radiation with dense media and will help developing a new complex theoretical model of non-linear SR.

Acknowledgements

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