Narrow structure in the coherent population trapping resonances in rubidium and Rayleigh scattering

S Gateva, L Gurdev, E Alipieva, E Taskova and G Todorov

Institute of Electronics, Bulgarian Academy of Sciences, 72 Tzarigradsko Chaussee, 1784 Sofia, Bulgaria
E-mail: sgateva@ie.bas.bg

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Abstract
The measurement of the coherent population trapping (CPT) resonances in uncoated Rb vacuum cells has shown that the shape of the resonances is different in different cells. In some cells the resonance has a complex shape—a narrow Lorentzian structure, which is not power broadened, superimposed on the power-broadened CPT resonance. The results of the performed investigations on the fluorescence angular distribution are in agreement with the assumption that the narrow structure is a result of atom interaction with Rayleigh-scattering light. The results are interesting for indication of the vacuum cleanliness of the cells and building of magnetooptical sensors.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The development of the applications of the coherent population trapping (CPT) resonances in high-resolution spectroscopy, quantum information storage and processing, metrology (atomic clocks), magnetometry, lasing without inversion, laser cooling, ultrashort group velocity propagation of light, etc [1–6] increases the interest in the processes which determine the shape of the resonances. For many applications narrow resonance signals and high signal-to-noise ratios are important. Narrow structures in the CPT resonance shape were measured at different experimental configurations, in vacuum, buffer gas and coated cells. The effect was studied for spatially and temporally separated laser fields.

Optical Ramsey fringes induced by Zeeman coherence were investigated in Rb by Zibrov et al [7, 8]. Experimental data were presented showing Ramsey fringes in frequency and time domain for a vacuum cell, as well as for a buffer gas cell. In a series of later works [9–12], the observed sharp central peak on a broad pedestal in the electromagnetically induced transparency (EIT) resonance shapes (the diffusion-induced Ramsey narrowing [10, 11]) was studied. The broad pedestal is associated with the single pass interaction time and is power broadened. The sharp central peak is the central Ramsey fringe, which adds coherently for all Ramsey sequences. Its width changes with the laser beam diameter. At low laser power, small beam diameter and low buffer gas pressure, the sharp central peak is not Lorentzian in shape and is insensitive to power broadening. At high laser intensity, the central peak loses its contrast and is Lorentzian in shape and power broadened.

Another narrow structure in the CPT resonance was registered in Rb by Alipieva et al [13]. In an uncoated room temperature vacuum cell, the CPT resonance was prepared in the so-called Hanlé effect configuration. The narrow resonance observed in fluorescence is of Lorentzian shape, is not radiation broadened and its amplitude increases with the laser power. Its width (FWHM) $\Delta L$ does not change with the laser beam diameter and corresponds to a relaxation time, equal to that due to atomic collisions with the cell walls. Details about the shape of the CPT resonances and the narrow structure are given in [13–18]. The resonance shapes, measured at different geometries of excitation and registration, show that the narrow structure at the centre of the resonance can be considered as a result of a weak field–atom interaction [17].

Narrow structures in the CPT resonance have also been measured in Na [19, 20]. In [19], the non-Lorentzian structure is explained, supposing the existence of scattered light in the volume of the cell. In [20], the narrow structure in Na was...
attributed to multiple interactions of the atoms with the main beam. In the latter case, the comparison of the experimental and theoretical shapes shows that in the range of ±1 mG there is good coincidence between them, but in the range of ±10 mG the experimental shape is narrower than the theoretical one.

One of the possible scattering processes influencing the CPT resonance shape is Rayleigh scattering [21] because in this case the scattered light maintains coherence with the incident beam and in this way a diffusion of the coherent light is created. For example, Rayleigh scattering has been used for studying the properties of cold atoms [22] and optical lattices [23] and references therein.

The purpose of the present work is to investigate the hypothesis of the influence of the Rayleigh-scattering light on the formation of the narrow structure in the CPT resonance described in [13].

2. Experimental setup

All the investigations were performed under the experimental conditions of [13] in the so-called Hanlé effect configuration. The experimental geometry is shown schematically in figure 1. The resonances were examined in uncoated vacuum cells containing a natural mixture of Rb isotopes at room temperature (295 K). A single-frequency linearly polarized diode laser beam (2 mm in diameter, 22 mW in power) was propagated along the cell’s x axis. A scanning magnetic field \( B_{\text{scan}} \) created by a solenoid, was applied collinearly to the laser beam. The gas cell and the solenoid were placed in a three-layer \( \mu \)-metal magnetic shield. The fluorescence was detected at 90° to the laser beam direction by a photodiode. The signals from the photodiode were amplified and stored in a PC, which also controlled the magnetic field scan. All measurements were performed on the \( F_e = 1 \rightarrow F_x = 2 \) transition of the \(^{87}\text{Rb} \) D\(_1\) line. The spectra in figures 2, 4 and 6 are plotted as a function of the magnetic field. The conversion of the magnetic field to frequency is straightforward on the basis of the Zeeman splitting between adjacent magnetic sublevels of the ground level \( \sim 0.7 \text{ MHz G}^{-1} \) [24].

The resonances were examined in three Rb vacuum cells with internal dimensions: cell a (length \( l_a = 4.3 \text{ cm}, \) diameter \( d_a = 2.1 \text{ cm} \)), cell b (length \( l_b = 4.5 \text{ cm}, \) diameter \( d_b = 2.9 \text{ cm} \)) and cell c (length \( l_c = 2.0 \text{ cm}, \) diameter \( d_c = 2.0 \text{ cm} \)). Cell a is a new one, which was sealed off some months before the experiments. It was manufactured using an oil-free vacuum installation—a turbomolecular pump and ion pump. All coating and filling steps are performed in a vacuum below 10\(^{-5} \) Pa. Cells b and c (as all cells from [13]) are very old (more than 20 yr), manufactured with an oil diffusion pump connected by a liquid nitrogen cold trap to the cell. Checking the vacuum with a Tesla coil (high-voltage transformer) shows that there is no light emission (glowing) in cell a, while the cell glass is glowing in cells b and c (as in all cells from [13]).

The experimental geometry is shown schematically in figure 1.

3. Shape of the CPT resonances

3.1. Experimental shapes

The investigations of the CPT resonance in different uncoated Rb vacuum cells show that the shape of the resonances is different in different cells. In figure 2(a), the CPT signals measured in cells a and b are given. In cell a, the resonance has triangular shape. In cell b, the resonance contains a narrow structure superimposed on a broad pedestal.

The difference in the dimensions of the two cells cannot be the reason for the existence of the narrow resonance. Following [13], we can calculate the narrow resonance width (FWHM) \( \Delta L = 1/(2\pi \tau) \), where the coherence time \( \tau \) is defined by two successive atom collisions with the cell walls \( \tau = L/v \). The estimated mean path between two collisions with the wall L is 1.6 cm in cell a and 2.1 cm in cell b. As the measurements are made at the same temperature, the mean velocity of the atoms is the same. Then, the evaluated difference between the widths of the eventual narrow resonances is of the order of 25–30% and is detectable.

3.2. Theoretical shapes

For explanation of the difference in the shape of the CPT resonances in cells a and b, a numerical solution of the full system of the density matrix equations for the investigated transition and geometry of excitation was performed following the procedure described in [15–18]. The quantization axis is chosen parallel to the polarization vector of the laser field (see figure 1) and for this geometry the coefficients in the system of equations described in the irreducible tensor operator presentation were calculated. The velocity distribution of the atoms and the Gaussian distribution of the laser beam intensity were taken into account. The photon reabsorption was not included in this model, because in our experiments the medium is optically thin on the length scale of the laser light–atom interaction zone [25].

The intensity of the registered fluorescence \( I_{F_eF_x}(\tilde{n}(\theta, \varphi)) \) in our experiments from the upper to the lower level in the direction \( \tilde{n}(\theta, \varphi) \) is described with

\[
I_{F_eF_x}(\tilde{n}) = C_0 (-1)^{F_e+\kappa} (2F_e+1)^{-1/2} |d_{\kappa}\rangle \sum (2\kappa + 1) \times \left\{ \frac{1}{F_e} - 1 \right\} \frac{1}{F_e} \frac{1}{F_e} \sum_q (-1)^q f_q^* \Phi^\kappa_q (\tilde{n}(\theta, \varphi)) \right) \right) (1)
\]
Figure 2. CPT resonance normalized shapes: (a) experimental in cell a (open circle curve) and cell b (dashed curve); (b) experimental in cell a (open circle curve) and theoretical (solid curve); (c) experimental in cell b (dashed curve), theoretical (solid curve) and the difference between them (dash-dotted curve).

where $|d_{eg}|$ is the reduced matrix element of the dipole transition $F_e \rightarrow F_g$, the curly brackets denote the 6–j Wigner symbol, $f_{jk}^l$ are the tensor components describing the upper level, $\Phi_{q}^n(\theta, \varphi)$ is the observation tensor, $\theta$ is the angle between the laser light polarization and the direction of registration (the inclination angle) and $\varphi$ is the azimuth angle.

In a dipole approximation, the fluorescence is defined only by the tensor components $f_{jk}^l$ with rank $k \leq 2$ and when the observation direction is in the plane orthogonal to the laser beam

$$I_{F,F}(\vec{n}) = C_0 |d_{eg}|^2 \left[ f_{00}^0 - \frac{1}{\sqrt{3}} \begin{pmatrix} 1 & 1 & 2 \\ F_p & F_e & F_g \end{pmatrix} \right] \times \left( f_{02}^0 \Phi_{22}^2(\vec{n}) + 2 \text{Re}(f_{22}^2 \Phi_{22}^{2-2}(\vec{n})) \right).$$

Although the components of the observation tensor $\Phi_{q}^n(\vec{n})$ depend on the direction of observation, the performed evaluations for the investigated transition have shown that the main contribution to the intensity of the unpolarized fluorescence has the population $f_{00}^0$ and $I_{F,F}$ practically does not depend on the observation direction.

The theoretical shape obtained for the laser beam–atom interaction coincides with the shape of the fluorescence resonance in cell a (figure 2(b)) and the pedestal of the resonance in cell b (figure 2(c)). The difference between the experimental and theoretical shapes in cell b (figure 2(c)) is a Lorentzian whose width is of the order of $\Delta_L = 1/(2\pi \tau)$, defined by the mean time $\tau$ between two atom collisions with the cell’s walls. This is the narrow structure from [13].

4. Rayleigh-scattering light angular distribution

To check the supposition that this narrow structure is a result of Rayleigh scattered light, we have performed a series of experiments and evaluations related to the light field angular distribution. For the linear polarization of the incident light, the power of the Rayleigh-scattering light $P_R$ registered at 90° to the laser beam direction is [21, 26]

$$P_R(\lambda, \theta) = P_0 N \sigma(\lambda) \sin^2 \theta \quad (3)$$

where $P_0$ is the power of the incident light, $\alpha$ is the registration efficiency coefficient, $N$ is the density of the scattering particles, $\sigma(\lambda, \theta) = \sigma(\lambda) \sin^2 \theta$ is the differential cross-section of the Rayleigh scattering of polarized light with wavelength $\lambda$ in the plane perpendicular to the beam direction and $\theta$ is the angle between the polarization of laser light and the direction of registration. According to equation (1), the Rayleigh-scattering light has a maximum at $\theta = 90^\circ$ and is zero at $\theta = 0^\circ$.

In a Rb vacuum cell at room temperature, the Rb pressure is of the order of $4 \times 10^{-5}$ Pa and at this pressure the Rayleigh scattered light from Rb atoms cannot be registered in our experiment [21, 26]—this is the case of cell a. However, if the vacuum cell is not pumped very well, there will be some residual air, water and oil vapour, as well as rare but relatively strongly scattering submicron particles, which will scatter the light—this is the case of cell b. In this case we are able to register the Rayleigh scattered light. The measurement of the power of the scattered light when the laser is tuned out of line has the typical $\sin^2 \theta$ dependence from equation (3) with maximum at $\theta = 90^\circ$ and minimum at $\theta = 0^\circ$ (figure 3).
5. Angular dependence of the narrow structure amplitude

In figure 4 are given the shapes of the CPT resonances measured at angles $\theta = 0^\circ$ and $90^\circ$ when the laser is tuned on the $F_e = 2 \rightarrow F_e = 1$ transition of the $^{87}$Rb D$_1$ line. The results are presented on two different scales. The comparison of the resonances on the broader scale shows that at the wings they have the same shape and that this shape is in good coincidence with the calculated one using the model from section 3.2. The comparison of the shapes in figure 4 on the narrower scale shows that at $\theta = 90^\circ$ there is a narrow structure. This narrow structure is Lorentzian in shape with width of the order of that defined by the mean time between two collisions with the cell walls $\Delta L$. The amplitude of this narrow structure (figure 5) has the same $\sin^2 \theta$ dependence on $\theta$ as in figure 3; it is maximum at $\theta = 90^\circ$ and minimum at $\theta = 0^\circ$.

Qualitatively, the observed narrow resonance can be described by including a weak, resonant, polarized light field into the numerical model from section 3.2. Such an approach corresponds to the supposition that the signal in the cell is formed by two subensembles of atoms interacting with different light fields. The general solution for the fluorescence intensity is the sum of the numerical integrations of the density matrix equations for each of the subensembles.

For the subensemble interacting with the laser beam the evaluations in section 3.2 have shown that this part of the signal practically does not depend on the observation direction. For the subensemble interacting with the weak Rayleigh scattered light, the angular distribution of the Rayleigh scattered light has to be included in the excitation tensor. This angular dependence in the excitation reflects on all tensor components $f_{kq}$ and in this way on the fluorescence signal $I_{F_eF_g}(\vec{n})$.

As the scattered light is very low in power, the resonances are with Lorentzian shape and not power broadened [1]. When the Rabi frequency of the light field $\Omega$ is small enough, so that $\Omega^2/\gamma_e < \gamma_g$ ($\gamma_e$ is the population decay rate from the
Figure 6. Narrow structure of the CPT resonance at different geometries of registration in cell c (length $l_c = 2.0$ cm, diameter $d_c = 2.0$ cm) the photodiode is on the cell wall (solid curve) and the light from the laser beam is projected by a lens on the photodiode (open circle curve).

excited state into the ground state, and $\gamma_g$ is the ground state coherence relaxation rate), there is no power broadening. So long as the mean free path of the atom is longer than the cell dimensions, $\gamma_g$ is defined by the mean time between two successive collisions with the cell walls. Since the weak field is due to Rayleigh scattering, the amplitude of the Lorentzian will depend on the density of the scattering particles and can be used as an indicator of the level of the vacuum cleanliness of the cell.

6. CPT resonance dependence on the geometry of registration

The shape of the resonances was measured at two different geometries of observation—when the photodiode is on the cell wall and the observation field of view is considerably larger than the laser beam diameter (figure 6, solid curve), and when a lens is used in front of the photodiode in order to restrict the observation field of view just to the laser beam volume (figure 6, open circle curve). On the other hand the angle of view in the former case is sufficiently small to ensure satisfactory angular resolution of observation. Because of the limited dimensions of the $\mu$-metal shield, the measurements were performed in a smaller cell, cell c. In cell c the mean path between two collisions with the cell walls is 1.33 cm and the expected (evaluated) width of the narrow resonance result of collisions with the cell walls is 3.2 kHz (4.6 mG) [13]. The comparison of the shape of the resonances in these two cases shows that the broad pedestal does not change, while the shape and the width of the narrow structure change (figure 6). In the first case (figure 6, solid curve), when the fluorescence mostly of atoms out of the laser beam is registered, the narrow structure is Lorentzian in shape and its width $\Delta_L$ is defined by the relaxation on the cell walls. In the second case (figure 6, open circle curve), when fluorescence mostly from the laser beam volume is registered, the measured narrow structure is narrower than $\Delta_L$. For the explanation of such narrowing, the influence of Ramsey narrowing has to be taken into account.

This explanation is in accordance with the results in [10, 13]. In [13], the registered fluorescence is mainly from atoms out of the laser beam volume, which interact with the Rayleigh scattered light. In this case, the narrow component is Lorentzian and its width is defined by the relaxation time due to atomic collisions with the cell walls. In [10], the resonance is registered in EIT and only atoms from the laser beam volume contribute to the signal. In this case the diffusion-induced Ramsey narrowing is responsible for the narrow structure.

7. Conclusion

The measurement of the CPT resonances in the fluorescence of uncoated Rb vacuum cells by means of the Hanlé effect configuration has shown that the shape of the resonances is different in different cells. In some not very well pumped cells, where residual gases exist, the resonance has a complex shape—a narrow structure which is not power broadened, superimposed on the power broadened CPT resonance [13]. The results of the performed investigations on the fluorescence angular distribution are in agreement with the assumption that this narrow structure is due to atom interaction with the Rayleigh-scattering light. The shape of the narrow structure is Lorentzian and so far as the mean free path of the atom is longer than the cell dimensions, its width is defined by the mean time between two successive collisions with the cell walls. The investigation is interesting for indication of the vacuum cleanliness of the cells and building of magnetooptical sensors.

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