Reversed Dark Resonance in Rb Atom Excited by a Diode Laser

Janis Alnis, Marcis Auzīnš*

Department of Physics, University of Latvia, 19 Rainis boulevard, Riga,

LV-1586, Latvia

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*Corresponding author, Fax +371-7820113, e-mail mauzins@latnet.lv
Abstract

Origin of recently discovered reversed (opposite sign) dark resonances was explained theoretically and verified experimentally. It is shown that the reason for these resonances is a specific optical pumping of ground state level in a transition when ground state angular momentum is smaller than the excited state momentum.

I. INTRODUCTION

Coherent population trapping was discovered in the interaction of sodium atoms with a laser field in 1976. [1]. Due to this effect a substantial part of population, because of destructive quantum interference between different excitation pathways, is trapped in a coherent superposition of ground state sublevels – dark states. With a coherent population trapping are associated dark resonances when due to this effect absorption and as a result fluorescence from atoms decreases, but intensity of the transmitted light increases when part of the atomic population is trapped in dark states. If in addition to the optical excitation an external magnetic field is applied, it can destroy coherence between ground state sublevels and return trapped population into absorbing states and, as a result, increase absorption and fluorescence, but decrease a transmitted light. A review of applications of dark resonances was published some years ago by Arimondo [2]. Coherence in an atomic ground state attracted substantial attention in connection with lasing without inversion [3], magnetometry [4] and laser cooling [5]. As a result dark resonances recently are studied in detail, including open systems [6] and systems with losses [7]. In course of these studies a new and unexpected phenomenon was observed by authors of [8]. In this study D2 line of 85Rb atoms was excited by a diode laser. Radiation was tuned to the absorption from optically resolved ground state hyperfine level \( F_g = 3 \) originating from atomic 5S\( _{1/2} \) state. The final state of the transition was Rb 5P\( _{3/2} \) excited state. Hyperfine components of this level was not resolved and all allowed in a dipole transition excited state hyperfine levels with quantum numbers
$F_e = 2, 3, 4$ were excited. In contrary to the usual dark resonance signal when in the absence of the magnetic field one can observe increased transmittance and decreased fluorescence intensity authors observed opposite effect — decreased transmittance which increases with magnetic field applied and an increased fluorescence intensity which decreased when field was applied. Authors of the paper [8] write that the physical reason of this effect remains still unclear. They suppose that one of the reasons for the peak in the fluorescence could be the non-coupled states on the Zeeman sublevels of the excited hyperfine levels. These non-coupled states, as it is supposed in [8], inhibit the stimulated emission induced by the laser field. The decrease of stimulated emission leads to an increase of the fluorescence.

In this letter we offer, in our opinion, very simple and straightforward explanation of the origin of these "reversed" dark resonances and perform experimental and numerical studies of them.

II. REVERSED RESONANCE

In a simple qualitative explanation traditional dark resonances can be connected with a well known optical pumping phenomenon. Let us assume that we excite atomic transition $F_g = 2 \rightarrow F_e = 1$ with a linearly polarized light. Direction of the $z$ axis is chosen along the light electric field vector $\mathbf{E}$. As a result $\pi$ absorption takes place and transitions occur between ground and excited state magnetic sublevels with $\Delta M = M_g - M_e = 0$, where $M_g, M_e$ are magnetic quantum numbers of the ground and excited states respectively, see Fig. If. According to this scheme absorption does not take place from ground state magnetic sublevels with quantum number $M_g = \pm 2$, because for these states there are no corresponding excited state magnetic sublevel with the same magnetic quantum number value.

In the spontaneous decay dipole transitions from optically populated excited state magnetic sublevels $M_e = \pm 1$ to the nonabsorptive ground state sublevels $M_g = \pm 2$ are allowed. As a result, if relaxation in the ground state is slow, in a steady state conditions substantial
part of the population will be optically pumped to the ground state sublevels with quantum numbers $M_g = \pm 2$ and will be trapped there. As a result traditional decrease of the absorption and fluorescence and increase of the transmittance will be observed, because the population of absorbing ground state magnetic sublevels will be reduced.

If we now apply an external magnetic field in a direction perpendicular to the $z$ axis, field will mix ground state sublevels effectively and will return trapped population into the states from which absorption takes place. As a result absorption and fluorescence will increase. This is a qualitative explanation of the usual dark resonance.

A similar reasoning can be exploited to explain the "reversed" resonance observed in \[8\] and in this paper. Let us assume that we excite with $\pi$ radiation atomic transition $F_g = 1 \rightarrow F_e = 2$. In this case there are no ground state sublevels not involved in the absorption that can trap atomic population. The actual relative transition rates proportional to the squared respective Clebsch–Gordan coefficients in this system of sublevels are shown in Fig. 2. As one can see ground state magnetic sublevel $M_g = 0$ is the most absorbing - with highest relative absorption rate. At the same time just to this sublevel intensively with high rates decay all three excited state magnetic sublevels populated by the light. One can expect that in a conditions of a steady state excitation, as a result of interplay of absorption and decay rates, population of the intensively absorbing ground state magnetic sublevel $M_g = 0$ will be increased and, as a result, one can expect increased absorption and fluorescence from this atom and decreased transmittance of the resonant laser light.

If an external magnetic field is applied perpendicularly to $z$ axis it will mix ground state magnetic sublevels and redistribute population between the ground state magnetic sublevels. As a result population of intensively absorbing magnetic sublevel $M_g = 0$ will be decreased. At the same time population of less absorbing magnetic sublevels $M_g = \pm 1$ will be increased. This means that the total absorption and fluorescence will be decreased and transmittance will be increased. This means that reversed dark resonance will be observed.

To prove this qualitative consideration let us solve balance equations for the magnetic sublevel stationary population $n_{M_g}$ in the scheme shown in Fig. 2. In a steady state
conditions for the ground state magnetic sublevels we will obtain

\[ n_{-1} = \frac{6(6\Gamma + 5\Gamma_p)}{51\Gamma + 100\Gamma_p} \overline{n}_g, \]
\[ n_0 = \frac{9(9\Gamma + 10\Gamma_p)}{51\Gamma + 100\Gamma_p} \overline{n}_g, \]
\[ n_{+1} = \frac{6(6\Gamma + 5\Gamma_p)}{51\Gamma + 100\Gamma_p} \overline{n}_g, \]

where \( \Gamma \) is excited state relaxation rate, \( \Gamma_p \) absorption rate, and \( \overline{n}_g \) ground state magnetic sublevel population in absence of the radiation. In a condition when absorption is slow \( \Gamma_p \ll \Gamma \) — weak absorption, we have

\[ n_{-1} \approx \frac{36}{51} \overline{n}_g \approx 0.706 \overline{n}_g, \]
\[ n_0 \approx \frac{81}{51} \overline{n}_g \approx 1.59 \overline{n}_g, \]
\[ n_{+1} \approx \frac{36}{51} \overline{n}_g \approx 0.706 \overline{n}_g. \]

If we now keep in mind absorption rates from different magnetic sublevels of the ground state, see Fig. 2 and calculate the overall absorption from such state and compare it with the absorption from the equally populated magnetic sublevels (when magnetic field is applied) than we see an increase in the absorption rate by a factor \( 18/17 \approx 1.059 \) or by approximately 5.9%.

The same calculation can be performed for the transitions \( F_g = 2 \rightarrow F_e = 3 \) and \( F_g = 3 \rightarrow F_e = 4 \). For these schemes in a similar way we will obtain even larger increase of the absorption due to this specific optical pumping. The increase will be by a factor \( 540/461 \approx 1.17 \) and \( 4004/3217 \approx 1.24 \) respectively. This means that the described effect increases with increase of the quantum numbers of involved levels.

Of course presented description is only qualitative, but in our opinion gives a good idea what is happening when reversed dark resonances are observed. To have a quantitative description of the phenomenon one must solve equations for the density matrix for an open system with losses. We will not do this in present paper. Instead a simple analysis will be carried out.
An analysis of the probabilities of optical transitions originating from \( F_g = 3 \) between excited hyperfine levels of the Rb atom show that levels \( F_e = 2, 3, 4 \) are populated in the ratio \((5/18 \approx 0.278) : (35/36 \approx 0.972) : (9/4 = 2.25)\). It means that a hyperfine transition leading to the reversed dark resonances discussed above is most strongly excited. For this scheme let us calculate a signal shape using a full density matrix approach. We solved a rate equations for a density matrix, see [9], Chapter 5, for \( F_g = 3 \leftarrow \rightarrow F_e = 4 \) transition. A broad line approximation was used. It means that we assume that in a magnetic field all magnetic sublevels are in equally good resonance with radiation. Secondly, we assumed that at a magnetic field strength used in the experiment (resonance width is less than 100 mG) hyperfine levels experience linear Zeeman effect. No substantial hyperfine level mixing at applied field strength takes place. Direct Rb atom magnetic sublevel splitting in a magnetic field calculations and measurements prove that these assumption are valid, see for example [10]. For signal simulation the following rate constants were used. Excited state relaxation rate \( \Gamma = 3.8 \times 10^7 \text{ s}^{-1} \) [11], absorption rate \( \Gamma_p = 3 \times 10^6 \text{ s}^{-1} \), ground state relaxation rate \( \gamma = 2 \times 10^5 \text{ s}^{-1} \) (mainly due to collisions with the walls of the cell and fly-through the excitation laser beam). Lande factors \( g_g = -0.3336, g_e = -0.5013 \) were calculated in a standard way from the atomic and nuclear data available in [12]. We suppose that a magnetic field is applied along \( z \) axis. Laser light excites \( F_g = 3 \rightarrow F_e = 4 \) transition and is linearly polarized along \( y \) axis. Intensity of the fluorescence with the same polarization is calculated and the intensity of the transmitted beam is also calculated as a function of the magnetic field. The results are presented in a Fig. [3]. They demonstrate well pronounced reverse resonances and are in a very good qualitative agreement with the measurements obtained in [8], see Fig. 5. there. Width of these resonances can be varied by changing ground state Lande factor value, ground state relaxation rate and absorption rate.

At week absorption \( \Gamma_p \ll \Gamma \) the dark resonance width is determined by a condition when ground state Larmor frequency is equal to the ground state relaxation rate. For \( F_g = 3 \) state of \(^{85}\text{Rb} \) atom at low concentration it can be as narrow as 20 – 30 mG. This is a width that was actually observed in [8].
Obtained signals in some sense are the same as the ground state Hanle effect measured in atoms as well as in molecules in great extent, see, for example [9,13]. In case of molecules also reversed structure in ground state Hanle effect was observed. In case of molecules, when optical pumping takes place in an open cycle and a total ground state population is substantially reduced, this structure can be attributed to a high order coherence created between ground state magnetic sublevels [14,15].

III. EXPERIMENTAL

We performed measurements of these reversed resonances also in our laboratory. In our experiment we use isotopically enriched rubidium (99 % of $^{85}$Rb) that is contained in a glass cell at room temperature to keep atomic vapor concentration low and avoid reabsorption. Transition $5s\ ^2S_{1/2}$ to $5p\ ^2P_{3/2}$ at 780.2 nm is excited using both temperature- and current-stabilized single-mode diode laser (Sony SLD114VS) with beam diameter 7 mm. Absorption signal (transmitted light) is monitored by a photodiode. As the laser is swept applying a ramp on a drive current, two absorption peaks with half-width of about 600 MHz separated by $\sim 3$ GHz appear due to the $^{85}$Rb ground state hyperfine structure. The excited state hyperfine structure is not resolved under Doppler profile.

During the resonance measurements, the laser wavelength is stabilized on absorption peak originating from ground state hyperfine level $F_g = 3$.

Helmholz coils are used to sweep the magnetic field over zero Gauss region, the Earth magnetic field components are compensated.

Signal detected in a transmitted light is averaged over 64 cycles and the result is presented in a Fig. 4, data points and Lorenz fitting, curve 1. On the same figure a calculated signal, curve 2, is presented. Model for this calculation is the same as in case on Fig. 3. Amplitude of the experimental and calculated signals are in arbitrary units and in figure are scaled in a way to make them easy to compare. For theoretical curve the parameters are chosen to have values that maximally reproduce our experimental conditions. The ground state
relaxation rate $\gamma = v_p/r_0 = 0.07 \mu s^{-1}$ was chosen as a reciprocal time of thermal motion of Rb atoms at room temperature with most probable velocity $v_p = 0.24 \text{ mm/}\mu s$ through the laser beam of radius $r_0 = 3.5 \text{ mm}$ \[^9\]. Absorption rate was chosen to be $\Gamma_p = 1.5 \mu s^{-1}$. Other parameters are as in Fig. 3. Agreement between calculated and measured signal is remarkable. As far as our model does not account for transitions to other hyperfine levels and so does not reproduce experiment in full, we do not attempt to fit experimental points with theoretical curve, nevertheless achieved agreement fully convinces us that the proposed explanation of the reversed dark resonances is correct. To obtain quantitative description of this signal in future one must take into account in the numerical model all other hyperfine levels involved in the process.

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REFERENCES

[1] G. Alzetta, A. Gozzini, L. Moi, and G. Orrioli, Novo Cimento B 36, (1976) 5

[2] E. Arimondo, Progr. Opt. 35 (1996) 257

[3] M. Scully, S-Y Zhu, A. Gavrielides, Phys. Rev. Lett. 62 (1989) 2813

[4] M. Scully, M. Fleischhauer, Phys. Rev. Lett. 69 (1992) 1360

[5] A. Aspect, E. Arimondo, R. Kaiser, N. Vansteenkiste, C. Kohen-Tannoudji, Phys. Rev. Lett. 61 (1988) 826

[6] Ferrucio Renzoni, Albrecht Lindner, and Ennio Arimondo, Phys. Rev. A 60, (1999) 450

[7] F. Renzoni, W. Maichen, L. Windholz, and E. Arimondo, Phys. Rev. A 55 (1997) 3710

[8] Y. Dancheva, G. Alzetta, S. Cartalava, M. Taslakov, Ch. Andreeva, Opt. Comm. 178 (2000) 103

[9] M. Auzinsh, R. Ferber, Optical Polarization of Molecules, Cambridge University Press, Cambridge UK, 1995, 305

[10] J. Alnis, M. Auzinsh, Phys. Rev. A, submitted

[11] G. Belin, S. Svanberg, Physica Scripta 4, 269 (1971)

[12] E. Arimondo, M. Inguscio, P. Violino, Rev. Mod. Phys, 49, 31 (1977)

[13] M.P. Auzinsh, R.S. Ferber, Phys. Rev. A, 43, 2374 (1991)

[14] M.P. Auzinsh, R.S. Ferber, Sov. Phys. Usp. 33, 833 (1990)

[15] M.P. Auzinsh, R.S. Ferber, Opt. Spectrosc. (USSR), 55, 674 (1983)
FIGURES

FIG. 1. Allowed dipole transition scheme for ground state optical pumping in case of π absorption for $F_g = 2 \rightarrow F_e = 1$.

FIG. 2. Transition scheme and rate constants for π absorption in case of $F_g = 1 \rightarrow F_e = 2$.

FIG. 3. Calculated intensity of fluorescence and transmitted light for reverse dark resonance for $F_g = 3 \rightarrow F_e = 4$.

FIG. 4. Measured (points and Lorenz fitting — curve 1) and calculated (curve 2) reversed dark resonance in $^{85}\text{Rb}$. 
Figure 1
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Figure 2
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Figure 3

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Figure 4
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