POWER BROADENING OF SATURATION ABSORPTION RESONANCE ON THE D₂ LINE OF RUBIDIUM

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Doppler-free resonances in saturated absorption on the D₂ line ⁸⁵Rb are investigated by using a tunable external-cavity-diode laser. The linewidths of the most prominent crossover resonances are investigated as a function of the saturating beam intensity. In spite of the fact that many levels and different mechanisms of optical pumping are involved in the saturation, the dependences are found to fit well the simple formula, obtained for a two-level system with a phenomenologically defined saturation parameter.

1. Introduction

A Doppler-free saturated absorption resonance for "two-level" atoms and a weak probe beam has lorentzian shape with the width γ given by [1,2]

$$\gamma = \gamma_r (1 + \sqrt{1+G})/2$$
, (1)

provided that the Doppler width γ_D is much greater than the natural one (γ_r) and collisional broadening can be neglected. Here $G = 2I\sigma\tau/h\nu$ is the saturation parameter, I is the intensity of the saturating beam, σ is the absorption cross section. Eq. (1) holds if the relaxation constants of the two levels are far from being equal.

The nonlinearity of the absorption which is necessary for Doppler-free spectroscopy results from the decrease of the population difference of the lower and upper levels. With additional levels available for optical transitions a new possibility of the population redistribution, that is the optical pumping to another nonabsorbing level, occurs [2].

In this paper we consider an important case where the relaxation time of the lower level is much greater than the lifetime of the upper level $\tau \approx 10^{-8}$ s and the time of flight of an atom through the laser beam $T \approx d/v \approx 10^{-4} - 10^{-5}$ s. This is typical for alkali atoms.

As compared to the absorption saturation of "two-level" atoms the saturation by optical pumping in multilevel atoms has the following peculiarities:

- (i) The saturating intensity is two-three orders lower since the characteristic pumping time should be comparable with the time of flight of an atom through the laser beam rather than with the spontaneous lifetime of the excited level.
- (ii) The relationship between the saturation degree and the light intensity is nonlocal. The probability to find an optically pumped atom at a given point of space depends on the intensity distribution along the way by which the atom arrived to the point.

The first property has been taken into account [2-4]. For atoms with a "Λ"-level configuration a very small value of saturation intensity has been explained and crossover resonances have been shown to be a sum of two different lorentzians [3]. Rate equations have been used to evaluate the relative amplitudes of different saturation resonances for atoms with "N"-type configuration of levels [4]. The same type of equations has been applied to "A" configuration to analyse the shape, width and amplitude of Doppler-free resonances, and some polarization effects have been considered [2]. The power broadening has not been studied in the cited papers. It has been made in refs. [5,6], but the spectral resolution was not high enough, and the authors arrived to a wrong conclusion that optical pumping does not decrease the saturation intensity [6].

In a number of papers subnatural linewidth of Doppler-free resonances has been studied for sodium atoms. However, in this case the power broadening has been made deliberately very small [7].

The purpose of this work was to study the power broadening of saturated resonance induced by optical pumping and to find whether its specific features modify the dependence given by (1).

2. Experiment

The high spectral resolution of an external cavity diode laser (ECDL) has been demonstrated by displaying the natural linewidth in Doppler-free spectra of Cs resonance lines [8,9]. The ECDL is shown schematically in fig. 1, and discussed in detail in ref. [10]. An external dispersion cavity consists of a coupling objective lens (NA=0.65), PZT-driven mirror, and a holographic grating (50% efficiency, 3200 lines per mm). A DHS GaAlAs laser with 6% reflection coefficient of the mirror facing the external cavity was mounted on a copper heatsink with temperature stabilization provided by a Peltier element. The laser spectrum was controlled by a monochromator with 0.1 nm resolution and a confocal cavity having free spectral range of 320 MHz and finesse of 35.

The power of the beam collimated by the output objective was 2 mW. The single-mode laser linewidth in a free running regime did not exceed 200 kHz as measured by a heterodyne technique [11].

To observe nonlinear resonances of saturated absorption we used a standard optical scheme (fig. 1). The collimated laser radiation having a divergency of no more than 2×10^{-7} and 6×11 mm² cross section was divided into two counterpropagating, probe and pump, beams of the same linear polarization. The angle between them did not exceed 3×10^{-3} . To decrease the influence of the nonlocality of saturation the diameter of the probe beam was diminished to 2 mm, and it was sent through the central part of the saturating beam. A 35 mm long glass cell contained saturated vapors of 85Rb at room temperature. To simplify the measurements of the saturation resonance width we have used a standard method for the Doppler-background suppression (the modulation of saturating beam intensity and lock-in amplification). The frequency of the ECDL was stabilized

to a scanning confocal interferometer to ensure a constant rate of frequency tuning.

3. Results and discussion

Fig. 2 gives examples of Doppler-free spectra for the high-frequency component of the 85Rb D₂-line taken at different intensities of saturating beam and constant intensity of the probe one $(2 \mu W/cm^2)$. Three intrinsic resonances are observed at frequencies ν_{32} , ν_{33} , ν_{34} which correspond to transitions from F=3 sublevel of the ground state to the F'=2, 3, 4sublevels of the excited state, respectively. Three orders are the crossover resonances centered at frequencies $(\nu_{32} + \nu_{33})/2$, $(\nu_{32} + \nu_{34})/2$, $(\nu_{33} + \nu_{34})/2$. The resonance at ν_{34} frequency corresponds to the cycling transition. Optical pumping to another hyperfine level is not involved in its formation since the selection rules prohibit $F' = 4 \rightarrow F = 2$ transition. It results in smaller saturation parameter as compared with all the other components which involve optical pumping. This explains the small amplitude (in spite of the highest unsaturated absorption of this component) and width of the resonance.

Two crossover resonances at $(\nu_{32} + \beta_{34})/2$ and $(\nu_{33} + \nu_{34})/2$, combining cycling and pumping transitions, are prevailing [4]. The linewidth of the strongest resonance at $(\nu_{33} + \nu_{34})/2$ is plotted against the intensity in fig. 3. The experimental points fit well the dependence $\gamma = \gamma_r (1 + \sqrt{1 + I/I_s})/2 + \Delta \nu_{RD}$ at $I_s = 32 \pm 3 \, \mu \text{W/cm}^2$ (saturation parameter) and $\Delta \nu_{RD} = 0 \Delta \nu_D = 1.5 \, \text{MHz}$ (residual Doppler broadening) with $\theta \approx 3 \times 10^{-3}$ rad and $\Delta \nu_D \approx 500 \, \text{MHz}$ being the angle between the saturated and probe beams and the Doppler width (fwhm), respectively.

In order to estimate the saturation intensity we modify the results of ref. [2] according to the difference in the energy level configuration and the presence of a cycling transition. Assuming small absorption, $kL \ll 1$, and validity of the Doppler limit, $\gamma \ll \gamma_D$, (both conditions were fulfilled in the experiment) the shape of resonance is described by

$$\sigma_{34}\sigma_{33}(\gamma_{33}/2)^{2}/[(\nu-\nu_{0})^{2}+(\gamma_{33}/2)^{2}] +\sigma_{33}S_{34}(\gamma_{34}/2)^{2}/[(\nu-\nu_{0})^{2}+(\gamma_{34}/2)^{2}], \qquad (2)$$

where the background term is omitted and

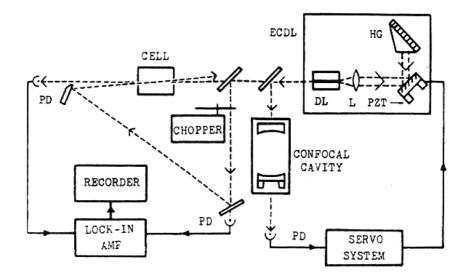


Fig. 1. Experimental set-up. ECDL, external cavity diode laser; DL, diode laser; HG, holographic grating; PD, photodiode; PZT, piezo-ceramic; L, microscopic objective lens.

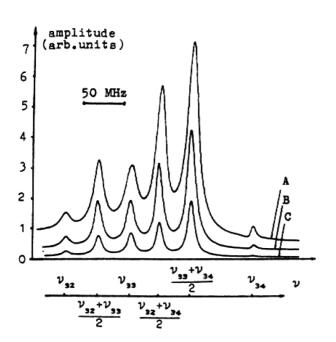


Fig. 2. Doppler-free resonances of saturated absorption for the high-frequency component of ⁸⁵Rb D₂ line recorded at different saturation intensities: (a) 0.34 mW/cm²; (b) 0.14 mW/cm²; (c) 0.03 mW/cm².

 $\sigma_{3F} = \sigma_0 A_{3F} / A g_3; \ \sigma_0 = 3\lambda^2 / 2\pi; \ g_3 = 7/12; \ A = \sum A_{FF} (A_{32} = 10; \ A_{33} = 35; \ A_{34} = 81; \ A_{21} = 27; \ A_{22} = 35; A_{23} = 28 \text{ are the relative amplitudes of the D}_2 \text{ line for }^{85}\text{Rb}); \ S_{3F} = 1 - (1 - G_{3F})^{-1/2}; \ \gamma_{3F} = (\gamma_r / 2) (1 + G_{3F})^{1/2}; \ G_{3F} = I / I_{3F}; \ \gamma_r = 1/2\pi\tau; \ I_{34} = h\nu (2\sigma_{34}\tau)^{-1}; I_{33} = h\nu (T\sigma_{33}\tau / \tau_{32})^{-1}.$

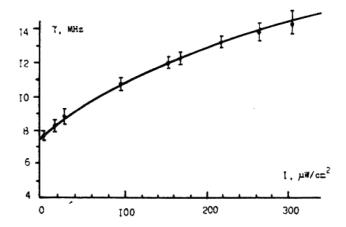


Fig. 3. The experimental dependence of the linewidth of the ⁸⁵Rb crossover resonance at $(\nu_{32} + \nu_{33})/2$ on the saturation beam intensity. The $\gamma = \gamma_r [1 + (1 + I/I_s)^{1/2}]/2 + \Delta \nu_{RD}$ dependence at $\gamma_r = 6$ MHz, $I_s = 32 \,\mu\text{W/cm}^2$, $\Delta \nu_{RD} = \theta \Delta \nu_D = 1.5$ MHz.

In the last two formulas determining the saturation intensity τ is neglected as compared with $T \approx d/v = 26 \, \mu s$; $\tau/\tau_{32} = 0.445$ gives the probability of the spontaneous-decay channel leading to the optical pumping. $1/\tau_{32}$ is the rate of spontaneous decay from F' = 3 to F = 2.

Two terms in (2) have simple physical meaning. As the laser frequency is passing the crossover resonance, $\nu_0 = (\nu_{34} + \nu_{33})/2$, the four independent groups of atoms distinguished by their longitudinal-velocity projection and interacting with the two counterpropagating laser beams at ν_{33} and ν_{34} merge

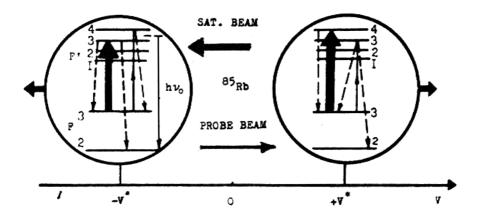


Fig. 4. Two groups of atoms in the velocity space contributing to the $(\nu_{33} + \nu_{34})/2$ crossover resonance and transitions involved in its formation. V is velocity projection on the laser beam, $V^* = (\nu_{34} - \nu_{33}) c/2\nu_0$, when $\nu_0 = (\nu_{34} + \nu_{33})/2$.

into two groups of atoms with $v = \pm (v_{34} - v_{33})/2v_0$ (fig. 4). The efficient optical pumping occurs in one of them via the $F=3\rightarrow F'=3\rightarrow F=2$ transitions induced by a strong wave so that S_{33} is large. The populations in the other group of atoms remain practically the same since the intensity of a strong wave is too small to saturate the $F=3\rightarrow F'=4$ cycling transition. Thus, for intensity used in the experiment $I < 300 \mu W/cm^2$ the ratio of the two terms contributing to (2) is $\sigma_{33}S_{34}/\sigma_{34}S_{33}$ < 0.01 which means that the first term can be neglected, and the experimental value of saturation intensity should be compared with $I_{33} = 86 \,\mu\text{W/cm}^2$. The distinction of this result from the experimental value of $I_s = 32 \pm 3 \,\mu\text{W/cm}^2$ seems to be quite small particularly if we bear in mind that the approach of homogeneous relaxation rate [2] does not include velocity distribution of atoms.

Thus the experimental $\gamma(I)$ dependence is well described by eq. (1) with phenomenologically determined saturation intensity. The last value is quite close to the one obtained for a monokinetic velocity distribution and without averaging over possible tracks.

The possibility to describe the power broadening by a single term is not so obvious since those Doppler-free resonances which are induced by repopulation of the ground state sublevels are actually the sums of independent lines for each magnetic sublevels [12].

The y(I) experimental dependences for other both intrinsic and crossover resonances in D_2 lines of Rb and Cs also fit well (1) when appropriate saturation

intensities are used. This result can be helpful when optimizing the conditions for a diode-laser frequency stabilization to saturation dips at resonant D lines of alkali atoms.

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