Absorption and fluorescence in saturation regime of Cs-vapor layer with thickness close to the light wavelength

P. Todorov*[a], K. Vaseva[a], S. Cartaleva[a], D. Slavov[a], I. Maurin[b], S. M. Saltiel[c]
[a]Institute of Electronics, BAS, Tzarogradsko Shosse boulevard, 1784 Sofia, Bulgaria
[b]Laboratoire de Physique des Lasers UMR 7538 du CNRS, Université Paris-13, France
[c]Physics Department, Sofia University "St. Kl. Ohridski", 1164-Sofia, Bulgaria

ABSTRACT

Absorption and fluorescent spectra are obtained in saturation regime of a single beam laser spectroscopy of Cesium D_2 line, in a vapor layer with thickness close to the light wavelength. We compare experimentally and theoretically obtained spectra distinguishing between open and closed, in terms of optical pumping, hyperfine transitions. In absorption, we observe a persisting difference between open and closed transitions, in terms of Dicke narrowing, when increasing laser light intensity. In fluorescence, for open transitions we note saturation dips which do not change significantly when increasing intensity. In case of closed transition, a small feature at the fluorescence line center appears for relatively high light intensity.

Keywords: Thin cell, optical pumping, Dicke narrowing, saturation spectroscopy, coherent resonances

1. INTRODUCTION

Coherent absorption effects in a molecular gas 1D confined in a cavity have been studied a long time ago[1] revealing an effect of narrowing of the Doppler broadened molecular transition. This effect is due to the specific cavity length allowing the time of flight between collisions with the cavity walls to be comparable with the lifetime of the exited state. The effect is called Dicke coherent narrowing on the name of its finder R. H. Dicke. In the optical domain, observations of similar sub-Doppler features are reported for atomic gas of Cesium[2,3] confined in cells with thickness from 10 μm to 1 mm. After the creation of the Extremely Thin Cell (ETC)[4], whose thickness is comparable with the light wavelength, better pronounced sub-Doppler resonances in absorption and fluorescence are obtained for Cesium and Rubidium atoms. The Dicke coherent narrowing is also observed for ETC with thickness $\lambda/2$ and $3\lambda/2$, revealing a periodic dependence of the optical transition width on the cell thickness[5,6]. Here λ denotes the wavelength of the corresponding optical transition. Theoretical studies of a thin vapor layer have also been performed describing the atomic vapor spectra in transmission and selective reflection[7] and including optical properties of the ETC walls[8].

In addition to the previous observations of the ETC fluorescent spectra, recently it has been shown[9] that saturation dips appear in the fluorescence profile when irradiating the ETC with high intensity laser light. The experiment is performed in saturation regime on the D_2 line of Cesium, for the ETC thickness $\lambda/2$ and λ. It has been shown that the saturation dips appear for all open hyperfine transitions when increasing light intensity. For the closed ones, there is a saturation dip only in the $F_g=3\rightarrow F_e=2$ transition profile, which is attributed to Zeeman optical pumping. Here F_g and F_e denote the quantum numbers of Cesium ground and exited states, respectively. A simple theoretical model describing the saturation behavior of open and closed atomic transitions has also been developed.

In this communication we present an investigation, both theoretical and experimental, of the absorption and fluorescent spectra on the D_2 line of Cesium, for hyperfine transitions starting from $F_g=4$. Cesium atoms are confined in an ETC with thickness $5\lambda/4$ (where λ=852 nm). This work extends our previous research [9,10] to the intermediate situation of cell thickness where the maximum of Dicke revival in absorption is not yet reached but we are still able to see sub-Doppler features due to the coherent Dicke narrowing. In case of the fluorescence, our interest is focused on the different evolution of the saturation effects with the light intensity, for the open and the closed transitions.

*petkoatodorov@yahoo.com; phone +359 2 876 1423; fax +359 2 975 3201; http://www.nano-vapor-layers.net/

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In the absorption spectrum, we observe a well pronounced saturation behavior of the whole spectrum and Dicke coherent narrowing signal only for the closed transition. Surprisingly, the coherent Dicke signal survives even at very light intensities and its amplitude stays relatively independent of the increasing amplitude of the saturation deep. In the fluorescence of the open transitions, we register an increase of the transition profile width with the light intensity as well as an enhancement of the amplitude and the width of the saturation dips. For the closed transition, only a relatively small feature appears at intensities higher than 1000 mW/cm^2.

2. EXPERIMENTAL SETUP

In Fig. 1, schemes of the experimental set up as well as the Cs D_2 line energy levels are presented. As coherent light source an extended cavity diode laser is used that operates in single-frequency mode. The external cavity is realized in Littrow configuration. The laser frequency scan is accomplished by changing the length of the external resonator. The emission width of the laser is 3 MHz. The laser field is linearly polarized. The main part of laser power is directed by a beam splitter and a mirror onto the ETC at normal incidence. For frequency reference and scaling of the diode laser frequency tuning, we obtain saturated absorption signal in a volume (centimeter size) cell filled with Cesium vapor at room temperature. A scanning Fabry – Perot interferometer is used to control the single mode operation of the laser.

The ETC is positioned in a specially constructed oven which has three openings - two for transmission experiments and one for recording the fluorescence signal. The ETC construction is similar to that described in previous work[4].

The oven's construction keeps its upper part where the cell windows are positioned warmer than the lower part where the reservoir of Cesium is placed. This prevents Cesium atoms from condensing on the cell windows, which causes loss of transmission signal and a lot of scattered light.

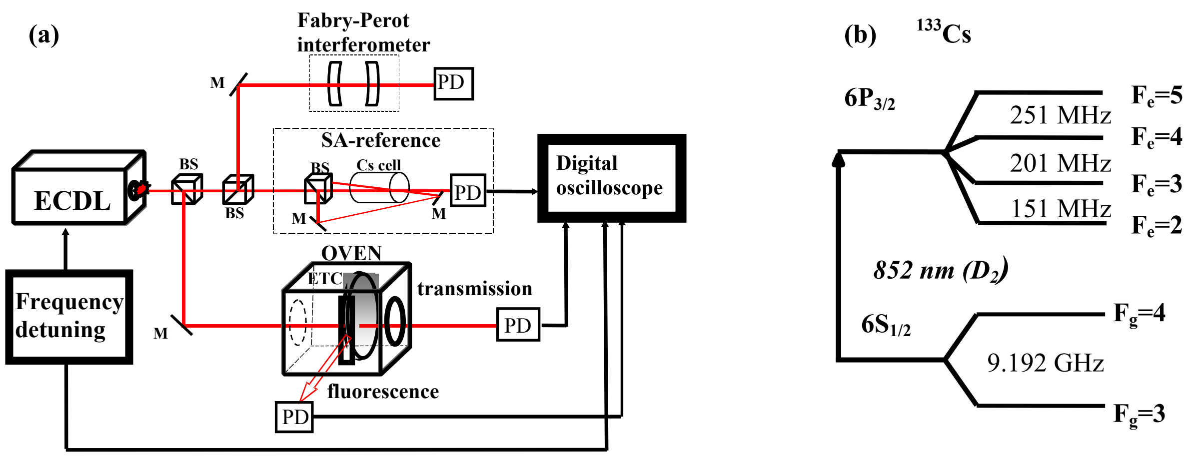

Fig. 1 (a) Experimental setup. ECDL – Extended cavity diode laser; M – mirror; BS – beam splitter; PD – photodiode; (b) The Cs D_2 line level scheme

The ETC transmission and fluorescence signals as well as this from the reference cell saturated absorption are registered by photodiodes simultaneously and stored in a digital oscilloscope. A frequency ramp generator and a high voltage amplifier are used to control the detuning of the laser frequency from the atomic resonance.

The atomic vapor density in the ETC is kept constant and low enough so that only atom-wall collisions have to be considered.

3. EXPERIMENTAL AND THEORETICAL RESULTS

3.1 Experimental and theoretical results in absorption

Experimental absorption spectra of Cesium D_2 line are presented in Fig. 2, for the three transitions starting from the $6S_{1/2}(F_g=4)$ ground level to the $6P_{3/2}(F_e=3, 4, 5)$ excited level. The thickness of the ETC is restricted to $5\lambda/4$ (where $\lambda=852$ nm). The different spectra are obtained for different intensities of the irradiating light.

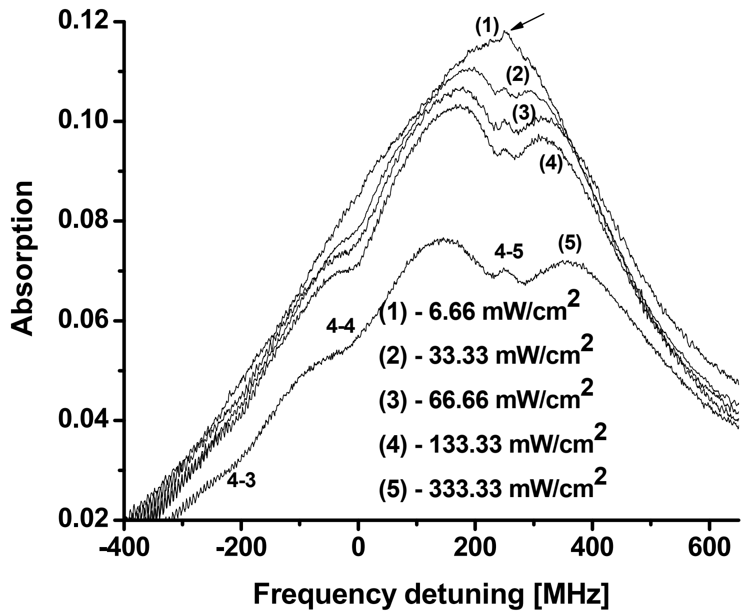

Fig. 2. Experimental absorption spectra of Cesium D_2 line observed in ETC, for different intensities of the irradiating light. The amplitudes of the corresponding spectra are inversely proportional to the light intensity.

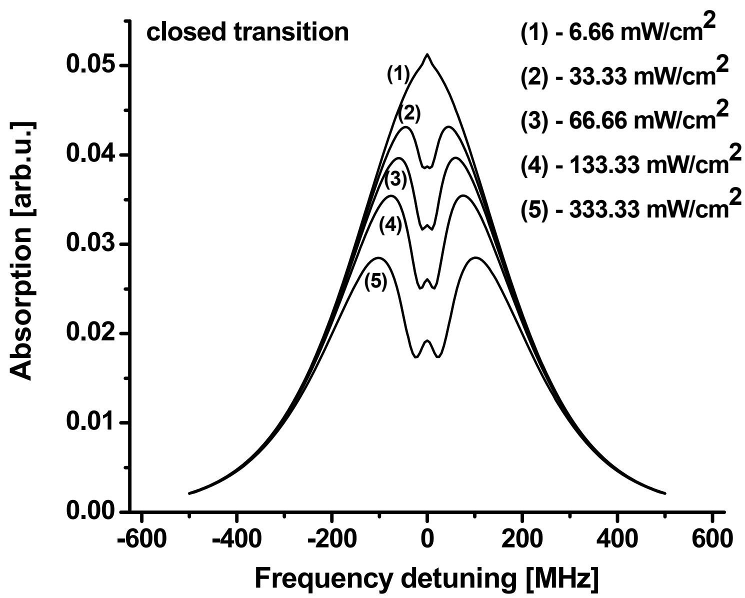

Fig. 3. Theoretical absorption profile for single closed transition and for different intensities of the irradiating light. The amplitudes of the corresponding spectra are inversely proportional to the light intensity. Here the natural width of the excited state - γ_{21}= 5 MHz, the Doppler width of the emitted light – ku = 250 MHz and the probability to decay from level 2 to level 1 - α = 0,99.

For an intensity of 6.66 mW/cm^2, one can distinguish only the F_g=4$\rightarrow$$F_e$=5 transition due to the small Dicke peak denoted by arrow in Fig. 2. When increasing the light intensity one can recognize the saturation behavior of the entire spectrum resulting in decreasing of its amplitude.

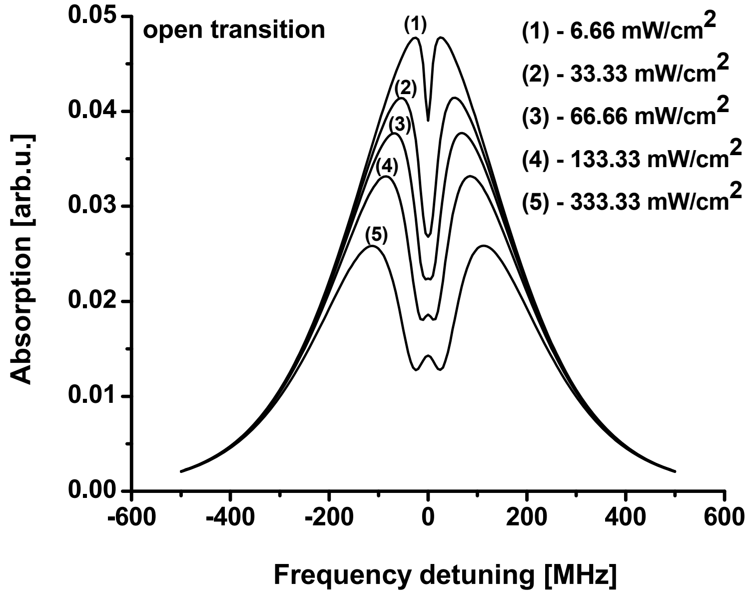

Fig. 4. Theoretical absorption profile for single open transition and for different intensities of the irradiated light. The amplitudes of the corresponding spectra are inversely proportional to the intensity. Here the natural width of the excited state - γ_{21}= 5 MHz, the Doppler width of the emitted light – ku = 250 MHz and the probability to decay from level 2 to level 1 - α = 0,6.

For the different hyperfine transitions, saturation dips around the transition center start to appear. These dips grow in amplitude and width with the light intensity.

The saturation dips are not the only sub-Doppler features that are recognizable in the absorption spectrum. In the position of the F_g=4 $\rightarrow F_e$=5 transition center, one can see an absorption peak superimposed on the saturation dip. This peak is observable at all intensities within the intensity range explored in the experiment. Moreover, for the light intensity 6.66 mW/cm^2, it is the only sub-Doppler feature in the spectrum. The narrow absorption peak originates from the coherent atom light interaction, and it is associated with the Dicke narrowing[5, 6]. For simplicity, further in the text we will call this absorption peak – Dicke signal. Increasing the light intensity, it can be seen that the Dicke signal does not change notably its amplitude even it falls in the saturation dip of the transition, which grows in amplitude. We could note only a broadening of the Dicke signal, which is better pronounced for the light intensity 333.33 mW/cm^2.

When comparing the open and closed (in terms of optical pumping) transitions, one could note that the saturation dips appear simultaneously for both types of transitions. The behavior of the saturation dip amplitude and broadening dependences on the light intensity is also similar.

However, one could immediately note that the Dicke signal is missing for the open transitions within the entire intensity range investigated, while the Dicke peak in absorption is always present, in case of the closed transition. The reason for this difference could be found in the transit time effects of atom light interaction, intrinsic to the nature of the ETC, combined with the optical pumping (in three-level system) or with the saturation effects in two-level system. It is well known[5, 6, 11] that Dicke narrowing is related to the appearance of a narrow Dicke signal over Doppler broaden pedestal, where Dicke signal originates from the transit times effects. The significant contribution to the narrow Dicke signal comes from the slow atoms. However, namely the slow atoms suffer the highest loss due to the optical pumping to the ground-state level non-interacting with the light, which occurs in case of the open transitions. Hence at the open transitions, the Dicke signal is missing.

Our theoretical results based on the model described in[9] are presented in Fig. 3 for the closed transition and in Fig. 4 for the open transition. In case of the closed transition, the theoretical model is in good agreement with the experimental results: for low light intensity only Dicke narrowing is observed, while for light intensity 33.33 mW/cm^2 the saturation dip is already well presented (together with the Dicke peak) in the absorption. The broadening and the increasing of the saturation dip amplitude take place with the light intensity.

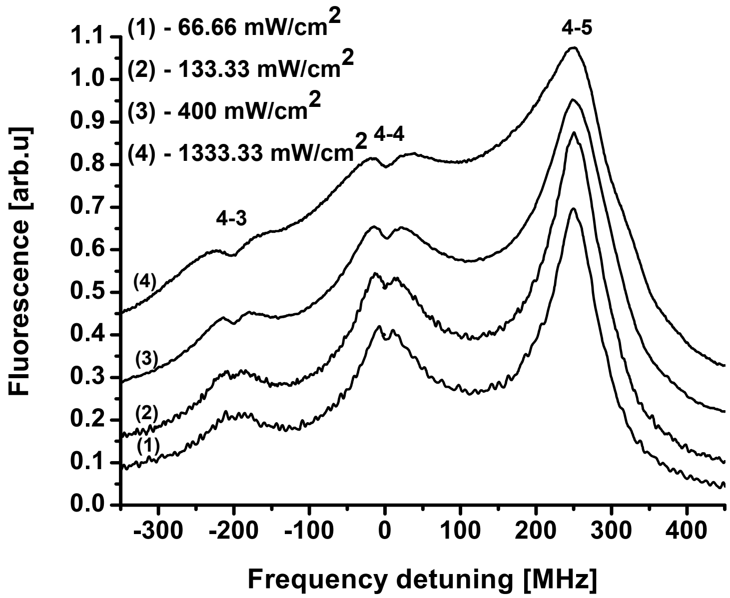

Fig. 5. Fluorescence spectra on the D_2 line of Cesium when exciting the F_g=4 level, for different intensities of the irradiating laser light. The amplitude of the corresponding spectrum is proportional to the light intensity.

In case of the open transition and low light intensity, the theoretical modeling shows (Fig.4) only dip in the absorption, which is in agreement with the experiment. Note however some difference between the experimentally and theoretically observed contrasts of the saturation dips. This difference is larger for the open transitions than for the closed ones. The dip contrast is larger in the theoretical spectrum than in the experimental one. The observed difference can be attributed

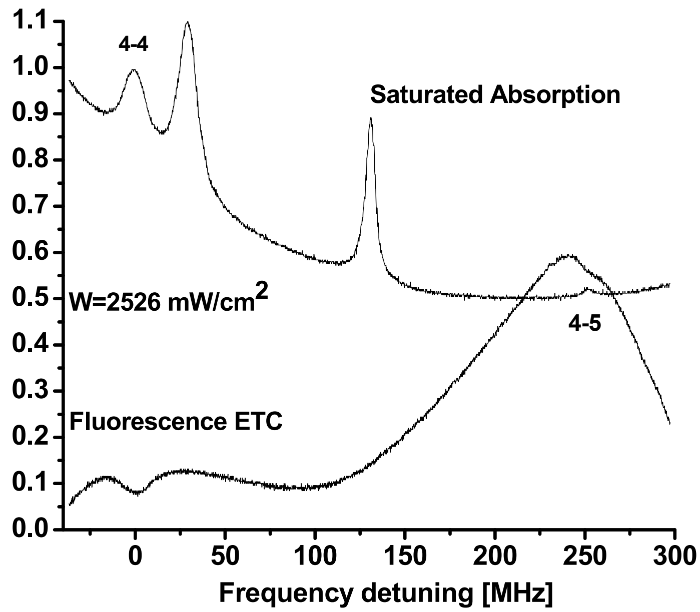

Fig. 6. Fluorescence and saturated absorption spectra on the D_2 line of Cesium illustrating the small saturation feature at the closed transition. Note the well pronounced dip at the open transition. The intensity of the irradiating laser light is equal to 2526 mW/cm^2.

to the intensity spatial distribution across the laser beam. In the experiment, the light intensity distribution is with close to Gaussian shape, while in the theoretical model we assume a plane wave. The Gaussian spatial intensity distribution makes the experimental spectrum to be a sum of spectra where each of them is obtained for different intensities. In this way, the experimental spectra exhibit lower contrast saturation dips than it is predicted by the simplified theory. In Fig. 4 we note that even for the smallest intensity there is a well pronounced saturation dip which is not observed in experimental spectra.

The theoretical amplitude of the transition profile reduces with the light intensity, confirming the result observed in the experiment. Dicke signal is missing for low intensity case (6.66 mW/cm^2 and 33.33 mW/cm^2), which is in agreement with the experimental results. When increasing the light intensity more than 66.66 mW/cm^2, a peak in the saturation dip appears. In the experimental spectrum, we do not observe such peak.

3.2 Experimental and theoretical results in fluorescence

The observed experimentally fluorescence spectra on the Cesium D_2 line are presented in Fig. 5. These spectra are obtained under the same experimental conditions with respect to the ETC thickness and the explored transitions as for the absorption spectra, discussed in the previous subsection. One notes that the three hyperfine transitions are very well resolved, for low light intensity. This is one of the advantages of the ETC, particularly in case of the fluorescence spectra registration.

In the ETC, the observed large difference between the fluorescence and absorption spectra is partly due to the strong anisotropy of the atom-light interaction time. In the fluorescence formation atoms with small velocity projections on the light beam direction are mainly involved. These atoms have longer interaction time with the laser light than those moving along the light beam. The later atoms could accomplish a single act of absorption. This is due to the fluorescence nature as a second order process[11]. The combination between the longer atom light interaction time needed for the fluorescence emission and the specific conditions in the ETC leads to much narrower fluorescence spectra than those in the absorption, for low light intensity (see Fig.2 and Fig.5).

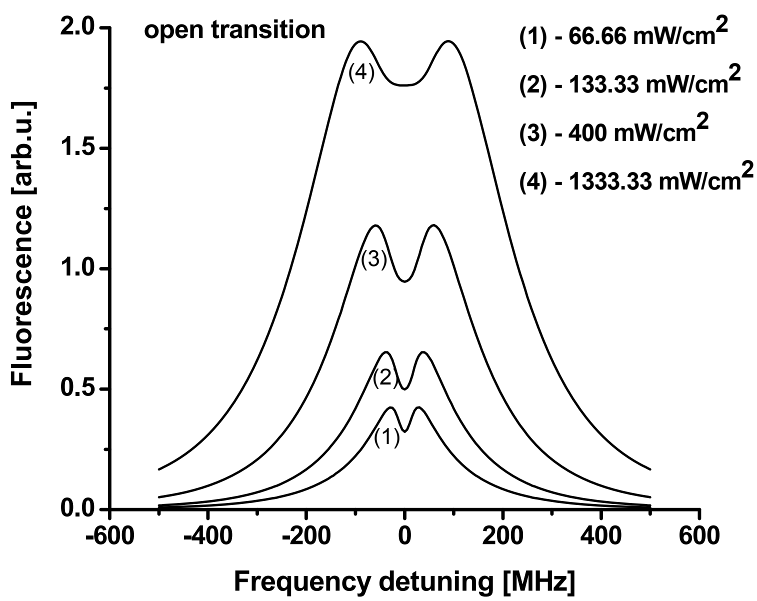

Fig. 7. Theoretical fluorescence profile for an open transition, at different intensities of the irradiating light. The amplitude of the corresponding spectrum is proportional to the light intensity. The calculations are made with the parameters used for Fig. 4.

Typically for the fluorescence behavior, the optical transition spectrum grows in amplitude and width with the light intensity. On the $F_g=4 \rightarrow F_e=3$ and $F_g=4 \rightarrow F_e=4$ open transitions (noted as 4-3 and 4-4 in Fig.5), saturation dips occur which have been reported for the cell thickness λ in[9] and for 2λ, 2.5λ, 3λ in[10].

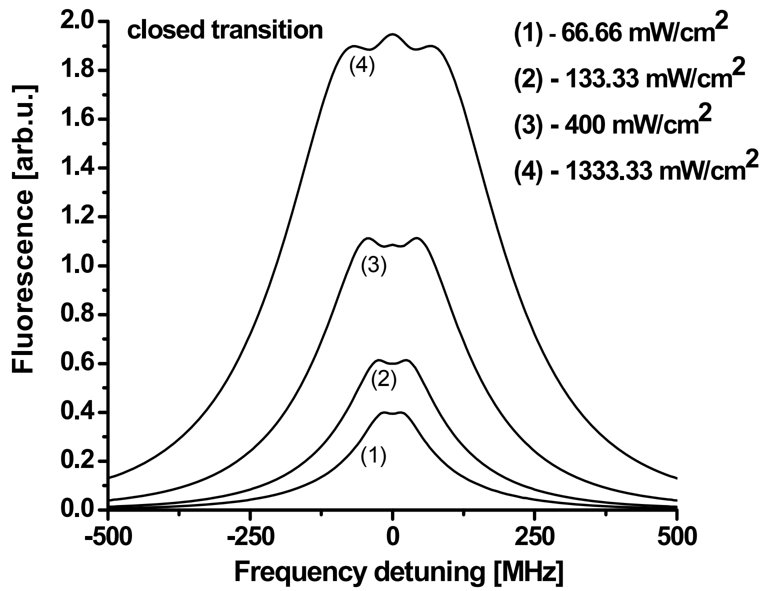

Fig. 8.Theoretical fluorescence profile for a closed transition, at different intensities of the irradiating light. The amplitude of the corresponding spectrum is proportional to the light intensity. The calculations are made with the parameters used for Fig. 3.

As it has been shown[9], these deeps are closely related to the velocity selective optical pumping process. For the $F_g=4 \rightarrow F_e=5$ closed transition (noted as 4-5 in Fig.5), no similar saturation dip in the fluorescence is observed. However, more precise measurement of the closed transition fluorescence profile shows that an extremely small feature is observed here at higher light intensity. This feature is illustrated in Fig.6. In order to be able to clearly recognize the small feature we present a zoom of the spectrum leaving in the frequency detuning only the 4-4 and 4-5 transitions. The feature is observed as a small slope change over the right side of the $F_g=4 \rightarrow F_e=5$ transition spectrum. This slope change could be a signature of a deep which is distorted because it appears on the slope of the resonance that is higher than the amplitude of the deep. The center of the feature coincides with the center of the 4-5 transition according to the saturated absorption reference also shown in Fig.6. The two additional saturated absorption resonances, which are situated between the 4-4 and 4-5 transitions, are the well known from the saturated absorption spectroscopy cross-over resonances. They are virtual resonances appearing due to the absorption at different real resonances which coincide in frequency scale thanks to different velocity groups in the Maxwell-Boltzmann velocity distribution.

Theoretically calculated by means of the model described in[9] fluorescence profiles are presented in Fig.7 and Fig.8, for different light intensities and for the ETC thickness $5\lambda/4$. Theoretical spectra obtained for an open transition are presented in Fig.7. One notes the saturation dip at the optical transition center. The width of the saturation dip and the width of the entire transition grow with the light intensity.

For a closed transition, the theoretical spectrum is presented in Fig. 8. One could easily note the significant difference between the open and closed transition saturation behavior. For the closed transition, one can recognize a peak in a dip (for the light intensity W=400 mW/cm^2 and W=1333.33 mW/cm^2), which reminiscent of the situation occurring in the absorption. One could note also that the ratio of the dip amplitude to the amplitude of the fluorescence transition profile observed in case of the closed transition is much smaller than that related to the open transition. The shape of the dip in the closed transition is different from that observed in the open transition. This could be a signature of different processes which govern the transition profile saturation in both cases.

For the closed transition, the small difference observed between the theoretical and experimental spectra can also be attributed to the fact that the model is for plane waves, while in the experiment the light beam with Gaussian intensity distribution is used.

4. CONCLUSION

We present the absorption and fluorescence spectra study for Cesium vapor layer confined in Extremely Thin Cell as a function of the laser light intensity. A different behavior of the absorption saturation has been found for the open and closed transitions, which is attributed to the coherent Dicke narrowing. We would like to point out that, at the chosen thickness of the ETC the Dicke narrowing is effective only in case of the closed transition.

In the fluorescence profiles, we observe simultaneously well pronounced saturation dip for the open transition and much smaller saturation feature for the closed transition. This feature could be associated with a deep of small amplitude and large width-to-amplitude ratio.

The presented theoretical results describe qualitatively the experimentally observed absorption and fluorescence profiles. For better analysis of the experimental results, a new theoretical model is under development, where the Gaussian beam shape is taken into account.

The presented results contribute to further advancement in the fundamental studies of the coherent effects, saturation and optical pumping processes in extremely thin vapor layers. They show that coherent and saturation processes strongly depends on the optical pumping in case of three level system.

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