Proceedings of the International School and Conference on Photonics, PHOTONICA09

Population Loss in Closed Optical Transitions of Rb and Cs Atoms Confined in Micrometric Thin Cells

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We present the first experimental observation of narrow dips in the fluorescence profiles of completely closed hyperfine transitions in Rb vapor at high atomic density, which is attributed to the depolarization of the excited state. Moreover, at low atomic density, a narrow peak on the top of the fluorescence profile is demonstrated, centered at the completely closed transition within the D_2 line of Cs. Experiments are performed in thin (700 μ m) alkali cell by single light beam spectroscopy. The cell is filled with Cs containing a small portion of Rb.

PACS numbers: 32.30.-r, 32.70.Jz

1. Introduction

It has been shown that single light beam transmission through a $(10 \div 1000 \ \mu m)$ thin cell reveals weak sub--Doppler features centered at the resonance frequency of the hyperfine transitions of alkali atoms [1]. In later developed extremely thin cell (ETC, thickness $< 1 \ \mu m$) these resonances exhibit very good amplitudes [2]. Recently, it has been reported [3] that in ETC with thickness $L = \lambda$ (wavelength of the irradiating light), the fluorescence from the open transitions differs strongly from that of the completely closed transitions. More specifically, in the sub-Doppler width fluorescence profiles, tiny saturation dips appear only for the open hyperfine transitions, which suffer population loss due to hyperfine or Zeeman optical pumping. These fluorescence dips have been related to the velocity selective optical pumping, which is maximal for very slow atoms. In the case of closed transitions, the fluorescence profile does not present any feature.

Here we report an experimental observation of narrow dips in the fluorescence profiles for both open and closed transitions within the Rb D_2 line, for high atomic source temperature. Moreover, a narrow peak on the top of the fluorescence profile is demonstrated for the first time, centered at the closed transition within the Cs D_2 line. Experiments are performed with a thin cell (700 μ m in length) filled with Cs containing small portion of Rb.

2. Experimental results and discussion

The used cell thickness of $L = 700 \ \mu \text{m}$ is larger than the wavelength of the irradiating light. The cell is filled with Cs metal but, due to the presence of a small portion of Rb, it was possible to study Rb spectrum in the presence of significantly higher Cs density (serving as a "buffer gas" whose pressure can be precisely adjusted with temperature). At first, the thin cell was irradiated by mono-mode linearly polarized diode laser light, tuned around the D_2 line of Rb (Fig. 1). The light beam propagates orthogonally to cell windows. The transmission (PD1) and fluorescence (PD2) spectra of ⁸⁵Rb were measured in a large temperature interval — 120–300 °C. In a second experiment, the transmission and fluorescence spectra of Cs vapor on the D_2 line were measured at low temperature using DFB laser with linear polarization. The saturated absorption spectra, measured (PD3) in a conventional, cm sized cell are serving as a frequency reference.

During the experiment on Rb D_2 line (Fig. 2, left: only the energy levels of the ⁸⁵Rb transitions are presented), different behavior is demonstrated for sub-Doppler resonances centered at cross-over (CO) resonances involving $F_{\rm g} \rightarrow F_{\rm e} \leq F_{\rm g}$ transitions and at $F_{\rm g} \rightarrow F_{\rm e} > F_{\rm g}$ transitions. Here, $F_{\rm g}$ is the hyperfine structure quantum number of the ground state and $F_{\rm e}$ is the one of the excited state. Two sets of hyperfine transitions were studied (Fig. 3): (i) the first one involves transitions starting from $F_{\rm g} = 2$ level of ⁸⁷Rb and (ii) the second one involves $F_{\rm g} = 3$ level of ⁸⁵Rb. Each set forms a single fluorescence line, due to the strong overlapping of the

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Fig. 1. Experimental setup.

hyperfine transitions. At relatively low (130 °C) alkali source temperature, well pronounced dips in the fluorescence were observed (Fig. 3a) centered at CO resonance positions: namely CO: 22–23 and CO: 21–23 for ⁸⁷Rb and overlapping CO 32–34 and $F_{\rm g} = 3 \rightarrow F_{\rm e} = 3$ transition in case of ⁸⁵Rb. CO resonances involve open transitions with population loss due to hyperfine and Zeeman optical pumping. The resonances, (1) and (3), at $F_{\rm g} \rightarrow F_{\rm e} > F_{\rm g}$ transitions are much weaker.



Fig. 2. Energy-level diagrams: ⁸⁵Rb (left) and ¹³³Cs (right) D_2 lines. $F_{\rm g} \rightarrow F_{\rm e} \leq F_{\rm g}$ transitions (solid line) are distinguished from $F_{\rm g} \rightarrow F_{\rm e} > F_{\rm g}$ transitions (dashed line).

However, when increasing the temperature (Fig. 3b), the centered at the $F_{\rm g} = 2 \rightarrow F_{\rm e} = 3$ (⁸⁷Rb) and $F_{\rm g} = 3 \rightarrow F_{\rm e} = 4$ (⁸⁵Rb) transitions resonances are very well presented. Hence, in case of $F_{\rm g} \rightarrow F_{\rm e} > F_{\rm g}$ transitions, the dips in the fluorescence profiles, observed for the first time, increase their amplitude with temperature. The appearance and amplitude enhancement of these dips with temperature are attributed to the depolarization of the $F_{\rm e}$ state by collisions between Rb and Cs atoms. Let us note that without the depolarizing collisions the $F_{\rm g} \rightarrow F_{\rm e} > F_{\rm g}$ transition can be considered as completely closed, i.e. without population loss. Moreover, the light drives the transition in the most absorbing state. However, as it has been shown in Ref. [4]. the $F_{\rm e}$ level depolarization leads to atomic population accumulation to the ground level with the lowest probability of excitation.

Thus, due to the excited level depolarization, some effective "population loss" (only for very slow atoms) oc-



Fig. 3. Thin cell fluorescence spectra for $F_{\rm g} = 3 \, (^{85} {\rm Rb})$ and $F_{\rm g} = 2 \, (^{87} {\rm Rb})$ set of hyperfine transitions, for low (135 °C) and high (206 °C) temperature of alkali source. The laser detuning scale is relative to the $F_{\rm g} = 2 \rightarrow F_{\rm e} = 3$ transition of $^{87} {\rm Rb}$; (1) $^{85} {\rm Rb}$ 3–4 resonance, (2) overlapping $^{85} {\rm Rb}$ CO 32–34 and 3–3 transition, (3) $^{87} {\rm Rb}$ 2–3 resonance, (4) $^{87} {\rm Rb}$ CO: 22–23, (5) $^{87} {\rm Rb}$ CO: 21–23.

curs, because the atomic population is pumped in the least absorbing state. In result of this, the efficiency of atomic excitation is reduced. Hence, we can say that the excited state population mixing causes some "opening of the closed transition".

Let us note that unlike the resonances centered at the $F_{\rm g} \rightarrow F_{\rm e} > F_{\rm g}$ transitions, those at the CO resonance positions suffer strong amplitude (contrast) reduction due to the enhanced Cs atom density with cell temperature. In order to estimate Rb atom density we measured the absorption at $F_{\rm g} = 3$ set of transitions, which was of several percents.

To study the velocity selective optical pumping resonance on the $F_{\rm g} \rightarrow F_{\rm e} > F_{\rm g}$ transition under better defined conditions, transmission and fluorescence spectra were measured on the D_2 line of Cs at very low temperature (25 °C) (Fig. 4). Here the atomic density of Cs is lower and the depolarization of $F_{\rm e}$ is not expected to take place. Hence, no dip in the fluorescence of the $F_{\rm g} = 4 \rightarrow F_{\rm e} = 5$ transition is expected at such cell temperature. The performed experiment not only confirmed our expectation for the fluorescence dip vanishing but demonstrated markedly good amplitude peak in the fluorescence centered at the $F_{\rm g} = 4 \rightarrow F_{\rm e} = 5$ transition. To our best knowledge, this is the first observation of velocity selective optical pumping to state with enhanced absorption, obtained by single-light-beam spectroscopy. Hence, due to the Zeeman optical pumping in the most absorbing state of the $F_{\rm g} = 4 \rightarrow F_{\rm e} = 5$ transition, occurring predominantly for slow atoms (i.e. those flying orthogonally to the beam), peaks of almost natural width (10÷12 MHz) are measured in the fluorescence and absorption.



Fig. 4. Thin cell transmission and fluorescence spectra at very low atomic source temperature (25 °C), showing bright velocity selective optical pumping at the completely closed $F_{\rm g} = 4 \rightarrow F_{\rm e} = 5$ transition on the D_2 line of ¹³³Cs. Laser power $W = 20 \ \mu W$; (1) Cs 4–4 resonance, (2) Cs CO: 43–45, (3) Cs CO: 44–45.

In this way, it is shown that in thin cells the velocity selective optical pumping, which is usually considered as moving slow atoms (emitting in sub-Doppler spectral interval) out of the two-level system, under certain conditions can redistribute atoms within the system accumulating slow atoms in the most absorbing state. An important practical result is that the reduction of cell thickness does not compromise its performance in view of the development of a frequency reference, but brings new advantage: observation of good-contrast narrow sub-Doppler resonances by means of single-beam spectroscopy.

The presented results contribute to the further development of high resolution spectroscopy in thin vapor layers, which is promising not only for fundamental studies, but also for technological applications, for example in the construction of precise frequency references and photonic sensors.

Acknowledgments

The work was done under collaboration between the Italian National Research Council and the Bulgarian Academy of Sciences and is partially supported by the Bulgarian NCSR (grant No: DO 02--108/22.05.2009) and Indian (DST Govt. of India project no. DST/INT/BULGARIA/B-69/2006)-Bulgarian (BIn--2/07) bilateral contract.

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