High resolution spectroscopy of Cs vapor confined in optical cells of few-micron thicknesses

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Abstract. We present here the new behavior of Electromagnetically Induced Transparency (EIT), Velocity Selective Optical Pumping (V SOP) and Velocity Selective Excitation (VSE) resonances observed in Cs vapor confined in unique cells with thicknesses $L = 1.5\lambda$ and $L = 6\lambda$. It is shown experimentally that in both cells, the EIT resonance is significantly narrower than would be expected from the ground state dephasing rate due to atomic collisions with the cell windows. The enhanced absorption (fluorescence) narrow VSOP resonance at the closed transition transforms into reduced absorption (fluorescence) one with small increase of atomic concentration or light intensity. A striking difference appears between the VSE resonance broadening due to excited atom thermalization, in $L = 6\lambda$ and conventional $L = 2.5$ cm cells.

1. Introduction

Laser spectroscopy of alkali vapor contained in optical cells is widely used in various laboratory experiments and photonic devices. The reduction of cell thickness $L$ is of importance not only for optical photonic sensor miniaturization but it also results in observation of new phenomena with $L$ approaching the wavelength $\lambda$ of the irradiating light. Recently, significant effort has been devoted to the development of miniaturized atomic clocks and magnetometers based on Electromagnetically Induced Transparency (EIT) resonances. The EIT resonances have been prepared in Cs atoms confined in sub-millimetre-thickness cell with high pressure buffer gas added to prevent frequent collisions of alkali atoms with the cell walls, which destroy atomic coherence [1]. The EIT resonance contrast measured in such cells is similar in magnitude to that obtained in centimetre-sized cells, but substantially more laser intensity is needed when sufficient buffer-gas pressure is used. Buffer gas broadens optical transitions, which limits the pumping efficiency and compromises the performance of vapor cell clock and magnetometer. A promising strategy for increasing atom-light interaction time consists in the use of anti-relaxation-wall coated cells; however, high atomic-vapor densities are limited owing to coating degradation with temperature [2].

Our communication concerns the high resolution laser spectroscopy of Cs vapor confined in unique optical cells with thickness of few microns, further on called Micrometric Cell (MC). The dimensions of such cell differ significantly. The distance between the high-quality cell windows $L$ varies from 1 $\mu$m to 6 $\mu$m. At the same time, the window diameter is (1±2) cm. If the irradiating laser light is adjusted in orthogonal to the cell window direction, a strong spatial anisotropy will present for the
time of interaction between the atoms and the laser radiation. Two kinds of atoms can be distinguished – “slow” and “fast”. Slow atoms involve those flying predominantly in direction orthogonal to the laser beam (typically of a millimeter diameter) and reach the steady state in the interaction with the laser light. The fast atoms have significant velocity projection on the laser beam propagation direction, which results in a time of atomic flight between the MC windows shorter than the lifetime of the excited state of the atom. Due to transient effects, this anisotropy leads to: (i) observation of a significant difference between the fluorescence and transmission (absorption) spectra [3] and (ii) occurring of Velocity Selective Optical Pumping (VSOP) resonances centered at hyperfine optical transitions [4]. A series of VSOP resonances are observed, which are subject of intensive study, motivated by the possible applications for development of wavelength references as well as for investigation of atom-atom and atom-cell window collisions.

More specifically, here we report about the new behavior of the EIT and VSOP resonances observed in Cs vapor confined in L = 1.5λ and L = 6λ cells, where λ = 852 nm is the wavelength of the laser light resonant with Cs D2 line. The L = 1.5λ cell provides the possibility to study the EIT jointly with the Dicke-type coherent narrowing of optical transition, namely at such thickness of optical cell the effect of Dicke coherent narrowing first revival has been observed [4]. At the same time, for L = 6λ, the Dicke effect vanishes, thus allowing the two different cases to be compared.

In addition, we demonstrate that (i) in MC a reasonably narrow EIT resonance can be observed without buffer gas using and (ii) at the closed hyperfine transition, a high-contrast Velocity Selective Excitation (VSE) resonance occurs, which is not observed for conventional optical cells, under the same experimental conditions.

2. Experimental set up for sub-Doppler and sub-natural width resonance study

Both used in the experiment micrometric cells are filled with Cs vapor and irradiated by bi-chromatic laser light from two narrow-band (with spectral width of about 2 MHz) distributed feedback (DFB) diode lasers – pump and probe one (Fig.1). The frequency of the pump laser is fixed at certain hyperfine optical transition while that of the probe one is scanned along a set of hyperfine transitions starting from a single ground-state level. The beams of the lasers are with orthogonal to each other polarizations. After careful overlapping, the two light beams propagate in direction orthogonal to MC.
windows. Transmission (absorption) and fluorescence (in an orthogonal to the laser beam direction) of Cs vapor were measured on the D₂ resonance line.

3. Sub-Doppler-width resonances of Cs vapor confined in optical cell with L = 1.5λ
We illustrate (Fig.2) the results related to the MC spectrum on the set of transition starting from the ground hyperfine level \( F_g = 4 \), obtained by scanning of the probe laser. In this case, the pump laser is switched off. If the probe laser is of low intensity, narrow Dicke resonances occur, centered at the \( F_g = 4 \rightarrow F_e = 3,4,5 \) transitions - curve (1). Of highest amplitude is the resonance at the closed \( F_g = 4 \rightarrow F_e = 5 \) transition, which does not suffer any atomic population loss due to hyperfine and Zeeman optical pumpings. To obtain an EIT resonance, a pump laser is involved and fixed at the \( F_g = 3 \rightarrow F_e = 4 \) transition. The EIT resonance, centered at \( F_g = 4 \rightarrow F_e = 4 \) transition - curve (2), is of natural width (Δν = 5 MHz), proving that it is based mainly on atoms flying parallel to cell windows. In fact, the dephasing rate of ground levels for the atoms moving orthogonal to the windows is ~ 70 MHz, as in the L = 1.5λ cell the atomic interaction time with a laser beam is highly anisotropic. Atoms moving nearly parallel to the cell windows have much longer interaction times compared to atoms moving in the direction perpendicular to the windows. The advantage of such approach is that mainly the slow atoms contribute to the EIT signal, which exhibits very good contrast.

While it is clear that the centered at the \( F_g = 3 \rightarrow F_e = 4 \) transition laser causes accumulation on the \( F_g = 4 \) level of Cs atoms with small velocity projection on the laser beam direction, it should be stressed that the Dicke resonance amplitude at the \( F_g = 4 \rightarrow F_e = 5 \) transition is strongly enhanced by the pump laser. Moreover, some narrowing of the Dicke resonance is observed. Hence, beside for optical cell miniaturization study, the proposed two-laser approach could be advantageous for further Dicke effect study in MC, because still it is not experimentally clarified what are the contributions of different-velocity-class atoms to the Dicke narrowing. Our experiment has shown that if the pump laser frequency goes out of exact resonance with the transition, the amplitude of the narrow resonance decreases very fast and it is getting much broader, i.e. the narrow Dicke resonance at the \( F_g = 4 \rightarrow F_e = 5 \) transition is mainly due to the “slow” atoms flying along the cell window surface.

4. Sub-Doppler and sub-natural width resonances observed in cell with L = 6λ - difference between open and closed optical transitions
In order to check if the reason for the large amplitude of the sub-Doppler-width resonance at the \( F_g = 4 \rightarrow F_e = 5 \) transition (Fig.2, curve (2)) is namely the Dicke phenomenon, the second cell with
L = 6\lambda is used, where no Dicke effect is expected. Moreover, our aim is also to make an accurate comparison of the L = 6\lambda cell behavior with that of a conventional cell with L = 2.5cm. For this, a beam splitter, BS, is inserted (denoted in blue in Fig.1), directing both precisely overlapped pump and probe beams to the centimeter cell as well.

To make the analysis simpler, we use very low powers of both lasers, thus working in a linear regime. From Fig.3a, it can be seen that even without Dicke effect a significant accumulation of “slow” atoms occurs, that are cycling at the F_g = 4 \rightarrow F_e = 5 transition. As for L = 6\lambda cell this narrow resonance appears only with the pump laser switched on, it is determined as Velocity Selective Excitation, VSE, resonance. An EIT resonance is observed in the F_g = 4 \rightarrow F_e = 4 transition absorption, measured by the probe beam (Fig.3a). While the EIT resonance width stays constant (less than 2 MHz) with the atomic source temperature enhancement, the VSE resonance suffers a measurable broadening (Fig.3b). Further theoretical studies are in progress for clarifying the physical processes behind the VSE resonance broadening under different experimental conditions.

The comparison of the EIT/VSE resonance for L = 6\lambda and L = 2.5 cm cells shows two unexpected results (Fig.3c): (i) despite the much higher rate of atomic collisions with the walls in the case of L = 6\lambda cell, the width of the EIT resonance is similar for both cells (about 2 MHz, mainly determined by the laser widths); (ii) the thermalization of selectively excited atomic population at the F_e = 4 level by the pump laser is dramatically lower for the L = 6\lambda cell than that observed in the L = 2.5 cm cell. Both results will be analyzed theoretically in further study. Here, in relation with the different thermalizations of the excited by the pump laser atoms, we assume that it could be due to the radiation trapping effect [5] which is much more pronounced in the L = 2.5 cm cell than in the MC. In fact, the extremely small thickness of L = 6\lambda cell reduces the probability of the fluorescence re-absorption by Cs atoms, because of the extremely short time and distance of the fluorescence propagation within the MC.

In general, our experiments have shown that the MC is much less sensitive to the magnetic field gradients, laser beam overlapping and the mutual coherence of the two independent lasers.
5. VSOP resonance sign reversal

In addition to the first observation of narrow and well resolved fluorescent spectra in Extremely Thin Cell (ETC) (of $L < 1 \mu m$) [4], recently it has been demonstrated [6] that even narrower dips appear in the already narrowed fluorescence profiles, at ETC thickness $L = \lambda$. Those saturation dips have been reported only for the open hyperfine transitions of Cs $D_2$ line, which suffer atomic population loss due to hyperfine or/and Zeeman optical pumping. No dip has been observed in the fluorescence of completely closed $4\rightarrow5$ transition. For later transition and using a narrow band (few MHz) diode laser, even a tiny peak in the absorption has been demonstrated [7].

Here, results are presented related to dark (reduced absorption) and bright (enhanced absorption) VSOP resonances observed in MC with thickness $L = 6\lambda$. The two open $F_g = 4 \rightarrow F_e = 3, 4$ transitions show dark VSOP resonances in the transmission and fluorescence spectra, at all experimentally examined Cs source temperatures. However, the profile of the closed $F_g = 4 \rightarrow F_e = 5$ transition shows different sign VSOP resonance at different atomic source temperatures $T$. At $T=60^\circ C$, very well pronounced narrow bright VSOP in the absorption (i.e. enhanced absorption), as well as a sharp top of the fluorescence profile is observed (Fig.4a). This interesting absorption peak is observed only at low atomic concentration and low light intensity. However, rising the temperature to about $84^\circ C$ leads to VSOP resonance sign reversal, i.e. narrow dip occurs in the absorption (Fig.4b). Moreover, the fluorescence profile of the $F_g = 4 \rightarrow F_e = 5$ transition shows a well pronounced dip at $T = 84^\circ C$. For lower temperatures of Cs source, the increase of light intensity results in the same effect – transformation of the bright resonance to a dark one. Note that no VSOP resonance broadening is observed during the resonance sign transformation.

![Figure 4](image-url)

Fig.4 (a) Narrow VSOP resonances of different signs in the $L = 6\lambda$ cell fluorescence and transmission spectra, for low atomic concentration ($9.2.10^{11}$ at/cm$^3$) and very low light intensity – comparison with a conventional $L = 2.5$ cm cell profile, where no sub-Doppler-width feature occurs; (b) Illustration of the bright resonance (in the fluorescence and absorption of $F_g = 4 \rightarrow F_e = 5$ transition) transformation into a dark one (denoted by asterisks) with atomic concentration enhancement ($5.2.10^{12}$ at/cm$^3$).

Physical processes behind the resonance sign reversal are attributed to the depolarization of the excited level, which transforms the completely closed system at low light intensity and atomic...
concentration to one with effective loss in the excitation process, for higher intensity and Cs vapor pressure. Two main processes can be responsible for population mixing of magnetic sublevels on the excited state, namely: (i) the elastic interaction between Cs atoms at enhanced atomic vapor pressures and (ii) the elastic interaction between the cell windows and the Cs-atoms travelling parallel to the windows surface [8].

6. Conclusions
VSOP, VSE and EIT resonances are experimentally observed in MC with few micron thickness confining Cs atoms, on the D\textsubscript{2} line of Cs. The resonance registration is performed by means of the pump-probe spectroscopy, using two narrow-band DFB lasers. In the L = 6\lambda cell, very high amplitude and narrow VSOP resonance occurs at the F\textsubscript{g} = 4 \rightarrow F\textsubscript{g} = 5 transition. In opposite, such narrow VSOP resonance is not observed for the L = 2.5 cm cell. This observation is attributed to the higher rate of radiation trapping in the last cell.

Very interesting result is that related to the EIT resonance width, which for the L = 6\lambda cell is about an order of magnitude less than estimated from the atomic collision rate with the cell windows. The comparison with the EIT resonance observed in the L = 2.5 cm cell under the same experimental conditions shows that in both cells EIT resonances of similar width are observed.

The obtained experimental results are of significant importance for the development of spectroscopy of alkali atoms confined in optical cells with micrometric thickness, for the purpose of photonics sensor miniaturization, as well as for new scientific effects study.

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