# HIGH RESOLUTION SPECTROSCOPY OF CS VAPOR CONFINED IN -MICRON THICKNESS OPTICAL CELLS

S Cartaleva[1], A Krasteva[1], A Sargsyan[2], D Sarkisyan[2], D Slavov[1] and T Vartanyan[3]

[1]Emil Djakov Institute of Electronics,
Bulgarian Academy of Sciences, 72 Tzarigradsko Chaussee, 1784 Sofia, Bulgaria;
[2]Institute for Physical Research, National Academy of Sciences of Armenia,
0203 Ashtarak, Armenia;
[3]St. Petersburg National Research University of Information Technologies, Mechanics and Optics, 49 Kronverkskiy Blvd., 197101 St. Petersburg, Russian Federation.

## 1. Introduction

Laser spectroscopy of alkali vapor contained in optical cells is widely used in various laboratory experiments and photonic devices. Reducing the cell thickness *L* is of importance not only for optical photonic sensor miniaturization, but it also results in the observation of new phenomena with *L* approaching the wavelength λ of the irradiating light. Recently, significant efforts have been devoted to the development of miniaturized atomic clocks and magnetometers based on electro-magnetically-induced transparency (EIT) resonances. The EIT resonances have been prepared in Cs atoms confined in cells of sub-millimeter thickness with a high-pressure buffer gas added to prevent frequent collisions of alkali atoms with the cell walls, which destroy the atomic coherence [1]. The EIT resonance contrast measured in such cells is similar in magnitude to that obtained in centimeter-size cells, but a substantially higher laser intensity is needed when sufficient buffer-gas pressure is used. The buffer gas broadens the optical transitions, which limits the pumping efficiency and compromises the performance of vapor cell clocks and magnetometers. A promising strategy for increasing the atom-light interaction time consists in the use of anti-relaxation-wall coated cells. However, the atomic-vapor density is limited owing to coating degradation with the temperature [2].

Our research concerns high-resolution laser spectroscopy of Cs vapor confined in unique optical cells with thickness of a few microns, further on called micrometric cells (MCs). The dimensions of such cells differ significantly. The distance between the high-quality cell windows *L* varies from 1 μm to 6 μm. At the same time, the window diameter is (1÷2) cm. If the irradiating laser light is steered normally to the cell window, a strong spatial anisotropy will be present for the time of interaction between the atoms and the laser radiation. Two kinds of atoms, “slow” and “fast”, can then be distinguished. The slow atoms are those flying in a direction nearly orthogonal to the laser beam (typically of a millimeter diameter) and reaching a steady state in the interaction with the laser light. The fast atoms have a significant velocity component along the laser beam propagation direction, which results in a time of 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) appearance of velocity-selective optical pumping (VSOP) resonances centered at hyperfine optical transitions [4]. A series of VSOP resonances have been observed and have been the object of intensive studies 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, we report here a new behavior of the EIT and VSOP resonances observed in Cs vapor confined in $L = 1.5\lambda$ and $L = 6\lambda$ cells, where $\lambda = 852$ nm is the wavelength of the laser light resonant with the Cs $D_2$ line. The $L = 1.5\lambda$ cell provides the possibility of studying the EIT jointly with the Dicke-type coherent narrowing of the optical transition, as the effect of Dicke coherent narrowing first revival has been observed in such an optical cell [4]. At the same time, for $L = 6\lambda$, the Dicke effect vanishes, thus allowing one to compare the two different cases.

In addition, we demonstrated that (i) in a MC a reasonably narrow EIT resonance can be observed without a buffer gas, and (ii) at the closed hyperfine transition, a high-contrast velocity-selective excitation (VSE) resonance occurs, which has not been observed in conventional optical cells under the same experimental conditions.

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

Both micrometric cells used in the experiment were filled with Cs vapor and irradiated by a bi-chromatic laser light emitted by two narrow-band (with spectral

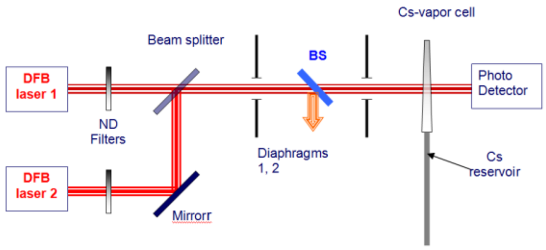



**Figure 1.** Experimental setup for VSOP, EIT and VSE resonance observation by pump-probe spectroscopy.

width of about 2 MHz) distributed feedback (DFB) diode lasers – a pumping one and a probing one (figure 1).

The frequency of the pump laser was fixed at a certain hyperfine optical transition, while that of the probe one was scanned within a set of hyperfine transitions starting from a single ground-state level. The laser beams were polarized orthogonally to one another. After careful overlapping, the two light beams propagated in a direction orthogonal to the MC windows. The transmission (absorption) and fluorescence (in a direction orthogonal to the laser beam) of the Cs vapor were measured on the $D_2$ resonance line.

## 3. Sub-Doppler-width resonances of Cs vapor confined in an optical cell with $L = 1.5\lambda$

Figure 2 illustrates the results for the MC spectrum on the set of transitions starting from the ground hyperfine level $F_g = 4$ obtained by scanning the probe laser.

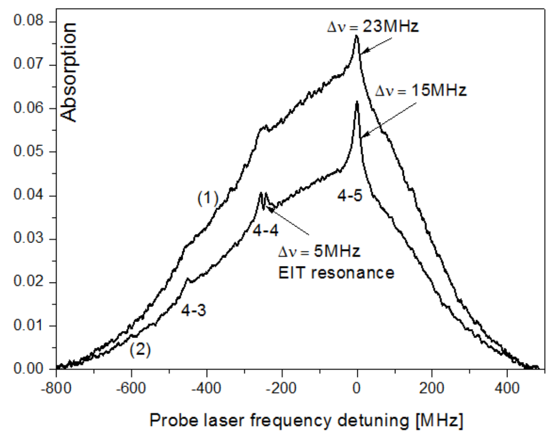



**Figure 2.** EIT and Dicke resonances at $L = 1.5\lambda$: (1) the probe laser is scanned within the $F_g = 4$ set of transitions, no pump; (2) the pump laser is fixed at the 3 - 4 transition and the probe is scanned as in (1); $T = 105$ °C, i.e. atomic density ~ $2\times10^{13}$ at/cm$^3$, $W_{probe} = 0.3$ mW/cm$^2$, $W_{pump} = 35.4$ mW/cm$^2$.

Initially, 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). The resonance at the closed $F_g = 4 \rightarrow F_e = 5$ transition is of the highest amplitude, since it does not suffer any atomic population loss due to hyperfine and Zeeman optical pumping. 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 ($\Delta\nu$ = 5 MHz), proving that it is based mainly on atoms flying parallel to cell windows. In fact, the dephasing rate of the ground levels for the atoms moving orthogonally to the windows is ~ 70 MHz, as in the $L$ = 1.5λ cell the atomic interaction time with the laser beam is highly anisotropic. The atoms moving nearly parallel to the cell windows have much longer interaction times compared to the atoms moving in the direction perpendicular to the windows. The advantage of this approach is that mainly the slow atoms contribute to the EIT signal, which exhibits a very good contrast.

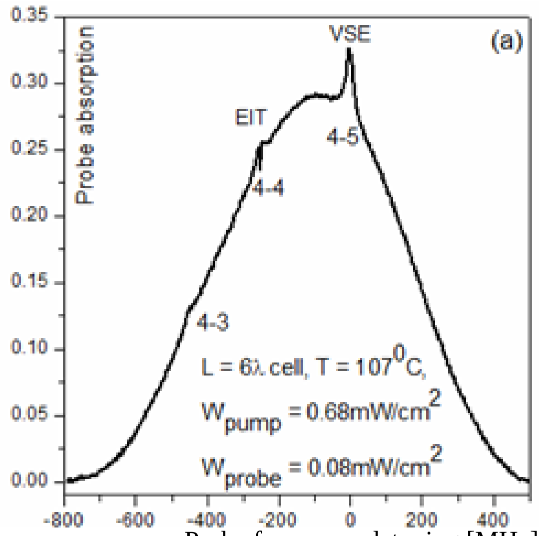
While it is clear that the laser light centered at the $F_g = 3 \rightarrow F_e = 4$ transition causes accumulation on the $F_g = 4$ level of Cs atoms with a small velocity component along the laser beam direction, it should be stressed that the Dicke resonance amplitude at the $F_g = 4 \rightarrow F_e = 5$ transition is strongly enhanced by the pump laser. Moreover, some narrowing of the Dicke resonance is observed. Hence, besides for optical cell miniaturization, the two-laser approach proposed could be advantageous for further Dicke effect studies in MCs, as the contributions of the atoms of different velocity classes to the Dicke narrowing are still to be clarified experimentally. Our experiment showed that if the pump laser frequency is shifted out of exact resonance with the transition, the amplitude of the narrow resonance decreases very quickly and broadens considerably, 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.

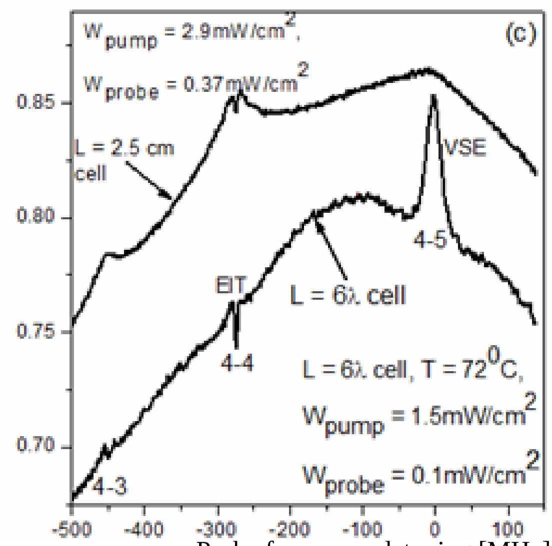
## 4. Sub-Doppler and sub-natural width resonances observed in cells with *L* = 6λ - difference between open and closed optical transitions

In order to check if the large amplitude of the sub-Doppler-width resonance at the $F_g = 4 \rightarrow F_e = 5$ transition (figure 2, curve (2)) is due to the Dicke phenomenon, the second cell with $L$ = 6λ was used, where no Dicke effect is expected.

Moreover, our aim was also to make an accurate comparison of the effects in the $L$ = 6λ cell with those in a conventional cell with $L$ = 2.5 cm. For this purpose, a beam splitter, BS, was inserted (denoted in blue in figure 1), directing the two precisely overlapped pump and probe beams to the centimeter cell as well. To make the analysis simpler, we used very low powers of both lasers, thus working in a linear regime. As can be seen in figure 3a, even without a Dicke effect, a significant accumulation of “slow” atoms occurs cycling at the $F_g = 4 \rightarrow F_e = 5$ transition. Since in the $L$ = 6λ cell this narrow resonance appears only with the pump laser switched on, it is determined as being a VSE resonance. An EIT resonance is observed in the absorption at the $F_g = 4 \rightarrow F_e = 4$ transition, as measured by the probe beam (figure 3a). While the EIT resonance width remains constant (less than 2 MHz) as the atomic source temperature is raised, the VSE resonance undergoes a noticeable broadening (figure 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 resonances for the $L$ = 6λ and $L$ = 2.5 cm cells shows two unexpected results (figure 3c): (i) despite the much higher rate of atomic collisions with the walls in the case of the $L$ = 6λ cell, the width of the EIT resonance is similar in both cells (about 2 MHz, mainly determined by the laser line widths); (ii) the thermalization of the atomic population at the $F_e = 4$ level selectively excited by the pump laser is dramatically lower for the $L$ = 6λ cell than that observed in the $L$ = 2.5 cm cell. Both results will be analyzed theoretically in a further study. In what concerns the different thermalization of the atoms excited by the pump laser, we assume that it is due to the radiation trapping effect [5]



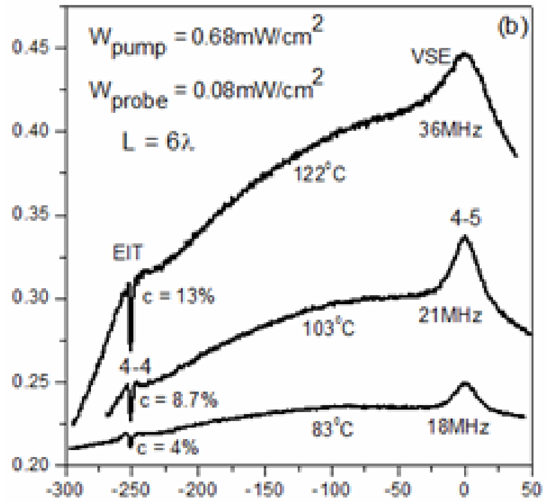




**Figure 3.** Probe absorption spectra in the $L$ = 6λ and the $L$ = 2.5 cm cells: (a) EIT resonance at the 4-4 transition and VSE resonance at the 4-5 transition; (b) EIT and VSE resonance behavior with the atomic source temperature; (c) illustration of the striking difference between the thermalization of the atomic population selectively excited by the narrow-band pump beam in the $L$ = 6λ and the $L$ = 2.5 cm cells. Atomic density: $(2.3\times10^{12} - 5.3\times10^{13})$ at/cm$^3$.

which is much more pronounced in the $L$ = 2.5 cm cell than in the MC. In fact, the extremely small thickness of the $L$ = 6λ cell reduces the probability for the Cs atoms re-absorbing the fluorescence, because of the extremely short time and distance of propagation of the fluorescence within the MC.

In general, our experiments showed that the MC is much less sensitive to the magnetic field gradients, the 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 cells (ETC) (with $L < 1$ μm) [4], it has been recently demonstrated [6] that even narrower dips appear in the already narrowed fluorescence profiles at an ETC thickness of $L$ = λ. These saturation dips have only been reported for the open hyperfine transitions of the 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 the completely closed 4-5 transition. For the latter transition, a tiny peak in the absorption has even been demonstrated [7] using a narrow-band (few MHz) diode laser. The results presented here concern dark (reduced absorption) and bright (enhanced absorption) VSOP resonances observed in a MC with thickness $L$ = 6λ. The two open $F_g = 4 \rightarrow F_e = 3, 4$ transitions show dark VSOP resonances in the transmission and fluorescence spectra at all Cs source temperatures used in the experiments. However, the profile of the closed $F_g = 4 \rightarrow F_e = 5$ transition exhibits a VSOP resonance of different sign at different atomic source temperatures. At $T = 60\,^{o}C$, a very well-pronounced narrow bright VSOP is observed in absorption (i.e. enhanced absorption), together with a sharp top of the fluorescence profile (figure 4a). This interesting absorption peak is observed only at low atomic concentration and low light intensity. However, raising the temperature to about $84\,^{o}C$ leads to a sign reversal of the VSOP resonance, i.e. a narrow dip occurs in the absorption (figure 4b).

Moreover, the fluorescence profile of the $F_g = 4 \rightarrow F_e = 5$ transition has a well-pronounced dip at $T = 84\,^{o}C$. At lower Cs-source temperatures, increasing the light intensity results in the same effect – a transformation of the bright resonance to a dark one. Note that no VSOP resonance broadening is observed during the resonance sign transformation.

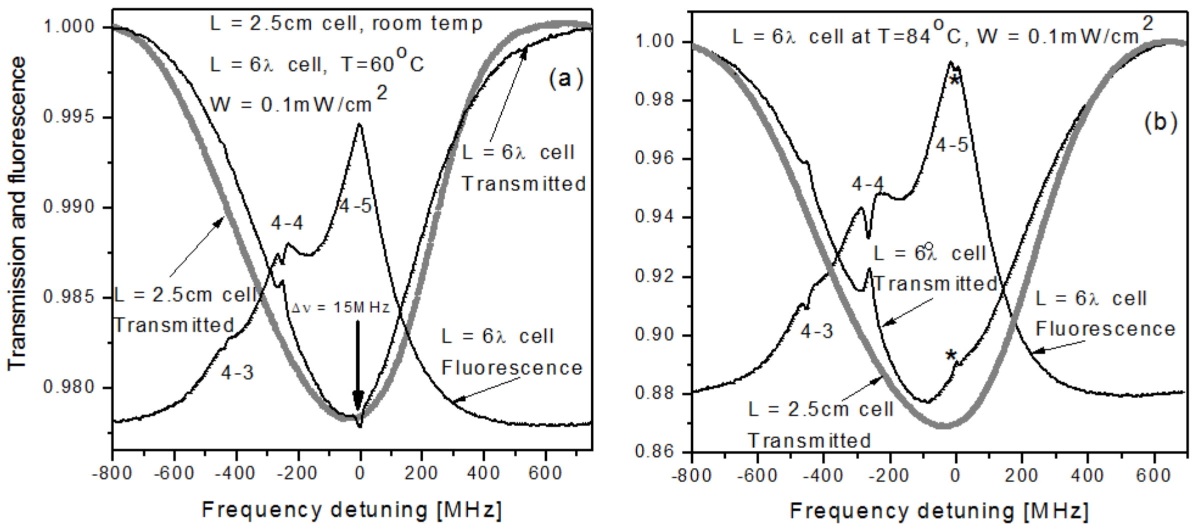



**Figure 4.** (a) Narrow VSOP resonances of different signs in the $L = 6\lambda$ cell fluorescence and transmission spectra for low atomic concentration ($9.2\times10^{11}$ at/cm$^3$) and very low light intensity – comparison with a conventional $L = 2.5$ cm cell profile, where no sub-Doppler-width feature occurs; (b) Illustration of the bright resonance (in the fluorescence and absorption of the $F_g = 4 \rightarrow F_e = 5$ transition) transformation into a dark one (denoted by asterisks) as the atomic concentration is raised ($5.2\times10^{12}$ at/cm$^3$).

The physical process behind the resonance sign reversal is assumed to be 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 at higher intensity and Cs vapor pressure. Two main processes can be responsible for the population mixing of magnetic sublevels of the excited state, namely: (i) elastic interaction between Cs atoms at higher atomic vapor pressures and (ii) elastic interaction between the cell windows and the Cs-atoms traveling parallel to the window surface [8].

## 6. Conclusions

VSOP, VSE and EIT resonances on the $D_2$ line of Cs were experimentally observed in a MC with a thickness of few microns confining Cs atoms. The resonances were detected by means of pump-probe spectroscopy using two narrow-band DFB lasers. In the $L = 6\lambda$ cell, a narrow VSOP resonance of very high amplitude occurred at the $F_g = 4 \rightarrow Fe = 5$ transition. In contrast, such a narrow VSOP resonance was not observed in the $L = 2.5$ cm cell. This fact was attributed to the higher rate of radiation trapping in the latter cell.

A very interesting result concerns the EIT resonance width, which for the $L = 6\lambda$ cell was about an order of magnitude narrower than that estimated from the atomic collision rate with the cell windows. The comparison with the EIT resonance in the $L$ = 2.5 cm cell under the same experimental conditions showed that EIT resonances of similar width are observed in both cells.

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

## Acknowledgements

This work was supported in part by the Bulgarian National Science Fund (grant No: DO 02-108), as well as by a Marie Curie International Research Staff Exchange Scheme Fellowship within the 7-th European Community Framework Program.

## References

[1] Knappe S, Hollberg L and Kitching J 2004 *Opt. Lett.* **29** 388
[2] Budker D and Romalis M 2007 *Nat. Phys.* **3** 227
[3] Sarkisyan D, Bloch D, Papoyan A and Ducloy M 2001 *Opt. Commun.* **200** 201
[4] Sarkisyan D, Varzhapetyan T, Sarkisyan A, Malakyan Yu, Papoyan A, Lezama A, Bloch D and Ducloy M 2004 *Phys. Rev.* A **69** 065802
[5] Allegrini M, Huennekens J, Namiotka R K, Sagle J and Jabbour Z J 1995 *Phys. Rev.* A 51 4472
[6] Andreeva C, Cartaleva S, Petrov L, Saltiel S, Sarkisyan D, Varzhapetyan T, Bloch D and Ducloy M 2007 *Phys. Rev.* A **76** 013837
[7] Cartaleva S, Saltiel S, Sargsyan A, Sarkisyan D, Slavov D, Todorov P and Vaseva K 2009 *J. Opt. Soc. Am.* B **26** 1999
[8] Krasteva A, Slavov D and Cartaleva S 2011 *Int. J. Opt*. 2011 ID 683415