Saturated Absorption Spectroscopy: Elimination of Crossover Resonances with the Use of a Nanocell

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Abstract—It is demonstrated that the velocity-selective optical pumping/saturation resonances of the reduced absorption in a Rb vapor nanocell with thickness $L = \lambda$, $2\lambda$, and $3\lambda$ (resonant wavelength $\lambda = 780$ nm) allow for the complete elimination of crossover (CO) resonances. We observe well-pronounced resonances corresponding to the $F_g = 3 \rightarrow F_e = 2, 3,$ and 4 hyperfine transitions of the $^{85}\text{Rb}$ D$_2$ line with line widths close to the natural width. A small CO resonance located midway between $F_g = 3 \rightarrow F_e = 3$ and $F_g = 3 \rightarrow F_e = 4$ transitions appears only for $L \geq 4\lambda$. The D$_2$ line ($\lambda = 852$ nm) in a Cs nanocell exhibits a similar behavior. From the amplitude ratio of the CO and VSOP resonances, it is possible to determine the thickness of the column of alkali vapor in the range of 1–1000 $\mu$m. The absence of the CO resonances for nanocells with $L \sim \lambda$ is attractive for the frequency reference application and for studying the transitions between the Zeeman sublevels in external magnetic fields.

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INTRODUCTION

Saturated absorption (SA) spectroscopy is widely used in the realization of frequency references for atomic transitions [1–3]. In this technique, the laser beam is split into a weak probe field and a strong pump field, which are sent to the interaction cell as counterpropagating overlapping beams. Because of opposite Doppler shifts, only the atoms moving perpendicular to the radiation propagation direction resonantly interact with both laser beams. For these atoms, the pump beam saturates the transition, and the absorption spectrum of the probe shows a Doppler-free dip, the so-called velocity selective optical pumping/saturation (VSOP) resonance located at the line center. With properly chosen pump and probe beam intensities, careful adjustment of the geometry, and the elimination of stray magnetic fields, the line width of the resonance (to which we refer to as “VSOP resonance”) may be as narrow as the natural width of the transition.

The situation is more complex when two or more atomic (hyperfine) transitions overlap within the Doppler profile, which is the case for the majority of real atomic lines. The presence of multiple unresolved hyperfine transitions results in the formation of the so-called crossover (CO) resonances. These “spurious” resonances appear when the laser frequency is tuned midway between two transitions, so that, for a group of atoms moving with a certain longitudinal velocity, the pump beam drives one transition, while the probe drives the other one linked to the former by the same ground state. As a result of the partial depletion of the ground state by the pump beam, the probe “sees” a reduced absorption. Thus, the VSOP process is also responsible for the occurrence of CO resonances. The CO resonances may seriously complicate the applicability of the SA spectral reference technique, notably in the high-resolution spectroscopy of hyperfine levels with a small (10–30 MHz) frequency spacing. In that case, the CO resonances mask the real atomic VSOP resonances and, thus, hinder or even impede the identification of the relevant spectral features. We note that, as a rule, the amplitude of the CO resonances is larger than the one for the actual VSOP resonances.

There are several techniques that allow for the elimination of crossover resonances in atomic vapors. In [4], two copropagating laser beams (a pump laser with a fixed frequency and a probe laser with a tunable frequency) were used rather than two counterpropagating beams. In [5], it was shown that the use of a thin (20 $\mu$m-long) cell in a conventional SA setup allows for the reduction of the amplitude of the CO resonance. Note that the optical selective reflection (SR) spectroscopy technique [6, 7] also allows one to eliminate the CO resonances; but, for the correct determination of an atomic transition position using this technique, the spectra must undergo further nontrivial processing.
In this work, we aim at revealing the minimal thickness $L$ of the atomic vapor layer needed for the formation of the CO resonance, and studies of the further evolution of the CO development as $L$ increases. We also examine and demonstrate the advantages of using very small (wavelength-scale) thicknesses $L$ of the atomic vapor column (a “nanocell”) as a promising spectroscopic tool. We show that, by using nanocells, it is possible to completely eliminate the CO resonances. This advantage (along with several others) makes the use of a nanocell with the thickness $L = \lambda$, $2\lambda$, and $3\lambda$ as a frequency reference for an atomic transition attractive. It is shown that the absence of CO is of significant importance, particularly for the study of the Zeeman spectra of the $^{85}\text{Rb} \, D_2$ line in an external magnetic field.

**EXPERIMENTAL SETUP**

The scheme of the experimental setup is presented in Fig. 1. The beam from a single-frequency tunable diode laser (1-mm diameter, power $P_L = 30$ mW, central wavelength $\lambda = 780$ nm, and line width $\gamma_L \sim 5$ MHz) irradiated thin cell 1 under an incidence angle close to the normal. A Faraday isolator ($FI$) was inserted to prevent the optical feedback to the laser. In the experiment, we used different cells 1 with various thicknesses $L$ in the ranges $L = \lambda$–$6\lambda$ (nanometric, vertical wedged-shaped vapor column) [8–11]; 9, 60, and 700 $\mu$m and 2-, 4-, and 60-mm-long. The cell was inserted into an oven with two apertures for passing the radiation. The Rb density determining the temperature of the cell’s side arm was adjusted in the range of $40$–$115^\circ$C, depending on the cell thickness. To prevent the atomic vapor condensation on the cell windows, their temperature was kept $20^\circ$ to $30^\circ$ higher that the side-arm temperature. The laser intensity was controlled by neutral density filters $F$ and its frequency scanned over $\sim 10$ GHz, covering the hyperfine spectral components of the $^{85}\text{Rb} \, D_2$ lines $F_g = 3 \rightarrow F_e = 2, 3,$ and 4, where the subscripts $g$ and $e$ refer to the lower and upper levels, respectively. Mirror 3 and glass plate 4 (shown in Fig. 1 inside the dotted rectangle) were used to form the SA spectra. In order to vary thicknesses $L$ of the vapor column in the nanocell in the range $L = (1–6)\lambda$, the oven was translated along the vertical direction as indicated by the arrow in Fig. 1. A part of the laser beam was sent to a normal vapor cell with a thickness of 60 mm to obtain a reference SA spectrum. The upper set of filters $F$ was used to properly attenuate the laser beam in order to get the reference SA spectra with a line width close to the natural width. The spectra were registered by a four-channel digital storage oscilloscope (Tektronix, TDS 2014B).

**RESULTS AND DISCUSSION**

It was shown earlier [9–12] that the ratio $L/\lambda$, where $L$ is the thickness of the atomic vapor column and $\lambda$ is the laser wavelength resonant with the atomic transition, is an important parameter that determines the widths, shapes, and amplitudes of the absorption resonances in a nanocell. In particular, it was shown that the spectral width of the resonant absorption is minimal for $L = (2n + 1)\lambda/2$ (where $n$ is an integer), an effect which
was called the “Dicke-type coherent narrowing effect” (DCNE). It was also shown that, for \( L = n\lambda \), the spectral width of the resonant absorption reaches a maximal value close to the Doppler width (about several hundreds of MHz), an effect called collapse of DCNE [9, 10]. In [13], the DCNE and the collapse of DCNE were investigated up to thicknesses \( L = 6\lambda \) for the D\(_2\) line of Cs at \( \lambda = 894 \text{ nm} \). Here, we consider the case where \( L = n\lambda \), since as was shown in [10, 11], the VSOP resonances appear under this condition.

In Fig. 2, the transmission spectra are presented for \( L \) varying in the range from \( L = \lambda \) up to \( 6\lambda \) with steps of \( \lambda \) (for these recordings, mirror 3 and glass plate 4 were removed). The effective laser intensity (EI) is determined by multiplying the measured intensity by the coefficient \( \left[ \gamma_N / (\gamma_N + \gamma_L) \right] \), where \( \gamma_N \) is the spontaneous decay rate of the Rb D\(_2\) line, \( \gamma_L \approx 5 \text{ MHz} \), and EI is 10 mW/cm\(^2\). When \( L \) is varied from \( L = \lambda \) to \( 3\lambda \), it can be seen that only the VSOP resonances are detected. These peaks of a decreased absorption are located exactly at the atomic transition frequencies [5, 10–12], note that, when \( L = \lambda \), the effective laser intensity was \( ~10 \text{ mW/cm}^2 \). The temperature of the cell’s side arm is 115°C. The CO resonances are marked by arrows.

![Fig. 2. Transmission spectra of the nanocell for the transitions \( F_g = 3 \rightarrow F_e = 2, 3, \) and 4 of the \(^{85}\text{Rb D}_2\) line, for \( L \) varying in the range \( \lambda \)–6\( \lambda \). The effective laser intensity was \(~10 \text{ mW/cm}^2\). The temperature of the cell’s side arm is 115°C. The CO resonances are marked by arrows.](http://doc.rero.ch)

However, as can be seen from Fig. 2, a small CO resonance (marked by arrows) located midway between the \( F_g = 3 \rightarrow F_e = 3 \) and \( F_g = 3 \rightarrow F_e = 4 \) transitions appears in the transmission spectra when the thickness \( L \geq 4\lambda \). One can also see a different behavior for the VSOP peaks at different transitions. It is interesting to note that, when \( L = \lambda \), the amplitude ratios of the VSOP peaks is close to the ratio of the atomic probabilities of the corresponding transitions \( A(3–4')/A(3–3') \sim 2, A(3–3')/A(3–2') \sim 3 \) [16], while for the SA spectrum, this is not the case (compare with the lower curve of Fig. 4). As the thickness increases, the amplitude of the VSOP peak corresponding to the \( F_g = 3 \rightarrow F_e = 4 \) cycling transition decreases, while the amplitudes of the VSOP peaks for the noncycling transitions \( F_g = 3 \rightarrow F_e = 2, 3 \) increase. In particular, for \( L = 6\lambda \) the ratio \( A(3–4')/A(3–3') \sim 0.5 \).

Conversely, the ratio of the noncycling transitions is practically independent of the thickness, i.e., \( A(3–3')/A(3–2') \sim 3 \). Figure 3 presents the theoretical spectra for the same group of transitions calculated using the model presented in [13]. All of the important experimental parameters determining the VSOP resonance formation, such as the influence of the reflected beam by the second wall of the nanocell (which is important when the thickness \( L \sim \lambda \)), the line width and intensity of the laser, the type of atomic transition (cycling or noncycling), the Maxwellian velocity distribution of the atoms, etc. have been taken into account. The fol-
The following parameters were used in the calculations: natural line width $\gamma_N = 6$ MHz, Rabi frequency $\Omega = 0.4\gamma_N$, and laser line width $\gamma_L = 5$ MHz. One sees that the theoretical model [13] correctly describes the peculiarities of the VSOP of the cycling and noncycling transitions as well as the behavior of the CO resonance as a function of the thickness $L$. One can conclude from Figs. 2 and 3 that the optimum condition for the formation of an atomic reference spectrum (e.g., to determine the frequency position of a weak atomic transition as $F_g = 3 \rightarrow F_e = 2$) is obtained for $L = 3\lambda$, in which case, the narrow and large VSOP peaks are observed, along with the absence of the CO resonances.

The second set of measurements was done for the same group of transitions employing the SA configuration (mirror 3 and glass plate 4 are mounted as shown in Fig. 1). Thin cells with thicknesses $L = \lambda$ (780 nm), $5\lambda$ (3.9 $\mu$m), 9, 60, and 700 $\mu$m and cells with a length of 2, 4, and 60 mm have been used.

In Fig. 4, the SA spectra are presented for $L = \lambda$ (780 nm), $5\lambda$ (3.9 $\mu$m), and 9 $\mu$m. The lower curve is the SA spectrum recorded in a 60-mm-long cell serving as the reference. As before, the CO resonance is completely absent in the case of $L = \lambda$, while it appears when $L = 5\lambda$ (3.9 $\mu$m) and the amplitude of the CO resonance increases as $L$ increases from 3.9 to 9.0 $\mu$m.

Figure 5 shows the SA spectra for $L = 60$ $\mu$m, 0.7, 2, and 4 mm. The VSOP resonance of the $F_g = 3 \rightarrow F_e = 3$ transition is sensitive to the polarizations of the pump and probe beams [17]. For a quantitative description, it is, therefore, more convenient to use the ratio of the amplitude of the CO resonance appearing when the laser is tuned midway between the $F_g = 3 \rightarrow F_e = 3$ and $F_g = 3 \rightarrow F_e = 4$ atomic transitions (this is the largest CO resonance in the spectrum), and the amplitude of the VSOP resonance of the $F_g = 3 \rightarrow F_e = 3$ transition. Figure 6 shows the ratio of $A(\text{CO})/A(\text{VSOP} 3\rightarrow 3')$ as a function of the cell thickness. The monotonic smooth increase of this dependence permits us to infer the cell thickness from a measurement of the amplitude ratio. Note that, in the case of thinner cells ($L \sim \lambda$), the interferometric method presented in [9] allows for the determination of the thickness $L$ with a high accuracy ($\sim 15$ nm). It should be noted that the dependence shown in Fig. 6 is slightly affected by the pump and probe beam intensities, and is correct for the $^{85}$Rb, $F_g = 3 \rightarrow F_e = 2$, 3, and 4 atomic transitions.

![Fig. 3](http://doc.rero.ch)

**Fig. 3.** Calculated spectra for the conditions in Fig. 2. Rabi frequency $\Omega = 0.4\gamma_N$; $\gamma_L = 5$ MHz; and $\gamma_N = 6$ MHz.

![Fig. 4](http://doc.rero.ch)

**Fig. 4.** Saturated absorption spectra for Rb cells with $L = 780$ nm, 3.9 $\mu$m, and 9.0 $\mu$m. The lower curve is the SA spectrum of an ordinary 60-mm-long cell. The CO resonances are marked by the arrows.

![Fig. 5](http://doc.rero.ch)

**Fig. 5.** Saturated absorption spectra for Rb cells with $L = 60$ $\mu$m, 0.7, 2.0, and 4.0 mm.
There are several parameters which influence the CO resonance amplitude, in particular, the frequency separation between the upper levels (2e), the probabilities of the atomic transitions involved in the CO resonance formation, and the thermal atomic velocity of the alkali atom [2, 3]. A frequency-tunable distributed feedback diode laser with \( \lambda = 852 \text{ nm} \) (line width 5 MHz) and EI \(-10 \text{ mW/cm}^2\) has been used to obtain the transmission spectra (see Fig. 7) for a nanocell filled with a Cs vapor for \( L \) varying in the range of \( \lambda \) to \( 6\lambda \) in steps of \( \lambda \). \( F_g = 4 \rightarrow F_e = 3, 4, \) and 5 transitions of the D2 line. In this case, a small CO resonance appears at \( \lambda \approx 10 \) mW/cm², the temperature of the side arm is 110°C. The lower curve is the SA spectrum in an ordinary cell. The CO resonances are marked by the arrows.

The difference between the SA spectra for different atomic transitions is well seen if we compare spectra obtained in similar conditions (i.e., cell length, temperature, laser intensity, etc.), as is the case for the lower traces of Figs. 4 and 7 (\(^{85}\text{Rb} \) D2 line, \( F_g = 3 \rightarrow F_e = 2, 3, \) and 4 in Fig. 4, and the Cs D2 line, \( F_g = 4 \rightarrow F_e = 3, 4, \) and 5 in Fig. 7). For \(^{85}\text{Rb} \), the ratio \( A(\text{CO})/A(\text{VSOP} 3–3') \) is \(-10 \) (corresponding to the VSOP resonances for \( B = 0 \)). In the upper curve, the transmission spectrum is given from the nanocell for \( B = 30 \text{ G} \) (directed along the laser beam) with \( \sigma^+ \)-polarized excitation (numbers denote transitions marked in Fig. 8b). The splitting and shifts of the three VSOP resonances 6, 5, and 4 are seen clearly in Fig. 8a. For \( B = 0 \), the theoretical ratio of these transition probabilities is \( 10:3:1 \).
nances numbered 6, 5, and 4), while for $B = 30$ G, the ratio of probabilities (corresponding actually to the amplitudes of VSOP resonances) is 6 : 4 : 3. As to the VSOP peaks 2 and 3, they are spectrally partially overlapped, as shown in the left inset. The theoretical ratio of the probabilities for $B = 0$ is equal to 1, while for $B = 30$ G, the ratio is equal to 2. The transition $F_g = 1, m_F = -1 \rightarrow F_e = 0, m_F = 0$ (numbered 1) is almost invisible in Fig. 8a, because its probability (i.e., VSOP amplitude) for $B \approx 30$ G practically vanishes. The calculated probabilities are presented in [20], and a good agreement is observed between experiment and theory.

The SA spectrum is presented in Fig. 8a by the lower curve for $B = 0$ (for the SA in a magnetic field, see [19, Fig. 6]).

Note that Fig. 8a (the upper curve) enables one to determine the frequency shift of the atomic transitions from their initial positions in a zero magnetic field (the middle curve) and, thus, allows one to determine the external magnetic field with a submicron (780 nm) spatial resolution. This is important for the mapping of a strongly inhomogeneous magnetic field.

Since, for Na atoms, the resonant wavelength of the D line is the smallest (=590 nm) as compared with that...
of other alkali metals, one can expect the best spatial resolution when using a Na nanocell. For this purpose, we have recently prepared a nanocell filled with Na metal. The thickness \( L \) of the gap between the windows has a wedge in the vertical direction varying from 100 to 550 nm at room temperature. At the windows’ operating temperature of \(~200^\circ C\) the maximum thickness \( L \) slightly increases, reaching 800 nm. Thus, there is a region of thickness \( L = \lambda \) where the Zeeman spectra can be obtained. This experiment is in progress at the University of Siena.

CONCLUSIONS

We have demonstrated that the use of a nanocell as the frequency reference for atomic transitions in comparison with the use of the SA spectra provides several advantages as follows: (i) contrary to SA, in the case of a nanocell, the crossover lines are absent, and this is very important for the study of the Zeeman spectra; (ii) for the case of a nanocell with thickness \( L = \lambda \), the ratio of the amplitudes for the VSOP peaks is close to the ratio of the atomic probabilities of the corresponding transitions, while in the case of SA, this is not the case; moreover, as a rule, a strong cycling transition in the SA spectrum has a smaller amplitude in comparison with that of the weaker noncycling one; (iii) the SA geometry requires counterpropagating beams, while a single-beam transmission spectrum is needed for a nanocell; and (iv) the laser power needed for spectral reference applications in the case of a nanocell is more than an order of magnitude less than that needed for SA.

From the ratio of the amplitudes of the CO and VSOP resonances, it is possible to determine the thickness of the alkali vapor column in the range from 1 to 1000 \( \mu m \).

It is important to note that the absence of CO resonances in a nanocell with \( L \sim \lambda \) allows one to study the behavior of the atomic transitions between the Zeeman sublevels in an external magnetic field, since the short time of flight ensures that the CO resonances are absent, even when the VSOP resonance is split into several Zeeman components. Conversely, even if the CO resonances are absent in the SA spectrum of an ordinary cell (this happens when the frequency distance between the upper levels is larger than the Doppler width of the atomic transition), the CO resonance may appear again (due to the large time of flight) when the VSOP resonance is split into several components.

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