

Coherent population trapping resonances at lower atomic levels of Doppler broadened optical lines

E. Şahin, G. Özen, R. Hamid, M. Çelik, A.Ch. Izmailov

Abstract. We have detected and analysed narrow high-contrast coherent population trapping (CPT) resonances, which are induced in absorption of a weak monochromatic probe light beam by counterpropagating two-frequency pump radiation in a cell with rarefied caesium vapour. The experimental investigations have been performed by the example of nonclosed three level Λ -systems formed by spectral components of the D_2 line of caesium atoms. The applied method allows one to analyse features of the CPT phenomenon directly at a given low long-lived level of the selected Λ -system even in sufficiently complicated spectra of atomic gases with large Doppler broadening. We have established that CPT resonances in transmission of the probe beam exhibit not only a higher contrast but also a much lesser width in comparison with well-known CPT resonances in transmission of the corresponding two-frequency pump radiation. The results obtained can be used in selective photo-physics, photochemistry and ultra-high resolution atomic (molecular) spectroscopy.

Keywords: coherent population trapping, optical pumping, probe beam, sub-Doppler spectroscopy.

1. Introduction

The essence of the phenomenon of coherent population trapping (CPT) consists in the fact that a multilevel quantum system subjected to decay processes is coherently driven into a superposition state immune from the further excitation, in which the system population is trapped. CPT is the basis of a number of important applications: ultra-high resolution spectroscopy, atomic clocks, magnetometry, coherent population transfer among quantum states of atoms (or molecules) and also in some others described, for example, in reviews [1–3]. In particular, narrow CPT resonances are detected in the absorption spectrum of two-frequency laser radiation by three-level atomic Λ -systems and also in the corresponding induced-fluorescence spectrum of a gas medium. Such resonance excitation of atoms in the Λ -system scheme is realised by means of light induced optical transitions from long-lived

levels $|1\rangle$ and $|2\rangle$ to the excited state $|3\rangle$ (Fig. 1). Most Λ -systems are not closed because of a possible radiative decay of the upper state $|3\rangle$ to ‘exit’ quantum states, which do not interact with resonance laser radiation. In atoms such an ‘exit’ is formed by some hyperfine and Zeeman sublevels of the ground term or by metastable levels [4]. In molecules the considered three level Λ -systems are open because their excited states have channels of radiative decay also at numerous vibrational and rotational sublevels of the ground molecular term [5].

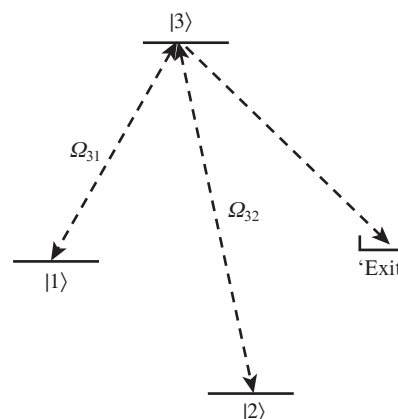


Figure 1. Scheme of the nonclosed Λ -system of optical transitions $|1\rangle \rightarrow |3\rangle$ and $|2\rangle \rightarrow |3\rangle$ (with central frequencies Ω_{31} and Ω_{32}) between the excited level $|3\rangle$ and long-lived states $|1\rangle$ and $|2\rangle$. The radiative decay also is shown from the level $|3\rangle$ to some quantum states (exit) which do not interact with the resonant laser radiation.

Some interesting features of CPT resonances in nonclosed atomic Λ -systems were detected and analysed in works [6–10]. An important experimental study of the CPT phenomenon in an open Λ -type molecular lithium system (Li_2) was presented in [11], where CPT resonances were detected in the fluorescence spectrum from the optically excited state. Lazoudis et al. [11] presented sufficiently large bibliography on corresponding optical coherence processes in molecules. Theoretical papers on possible CPT applications in considered nonclosed three level Λ -systems have also been published [12–14]. Thus, the possibility of selective photo-excitation of atoms (molecules) within the homogeneous width of their resonance optical lines was shown on the basis of the CPT phenomenon [12, 13], which is important for selective photo-physics and photochemistry. Moreover, a new method of ultra-high resolution spectroscopy was theoretically suggested, which allowed one to identify the overlapping optical

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lines in complex molecular (atomic) spectra even when frequency intervals between centres of given lines are less than their natural linewidths [14]. The possible applications of the CPT phenomenon [12–14] are directly based on the use of a trapped atomic (molecular) population at lower levels of a nonclosed Λ -system (Fig. 1). Therefore, further research of CPT features at these long-lived levels is important.

Let us consider interaction of a nonclosed Λ -system (Fig. 1) with two monochromatic laser fields. Frequencies ω_1 and ω_2 of these fields are close to centres Ω_{31} and Ω_{32} of electro-dipole transitions $|1\rangle \rightarrow |3\rangle$ and $|2\rangle \rightarrow |3\rangle$, respectively. Populations of lower levels $|1\rangle$ and $|2\rangle$ will diminish with increasing this resonance two-frequency radiation because of the light induced repumping of atoms from these levels to ‘exit’ states after the radiative decay of the optically excited level $|3\rangle$ (Fig. 1). However, sharp weakening of such a repumping process takes place due to a strong decrease in the population of the upper state $|3\rangle$ under the following CPT condition [1–3]:

$$|\delta_2 - \delta_1| \leq W, \quad (1)$$

where $\delta_1 = (\omega_1 - \Omega_{31})$ and $\delta_2 = (\omega_2 - \Omega_{32})$ are detunings of laser frequencies. The characteristic width W of the CPT resonance in Eqn (1) is determined by intensities of laser fields and by relaxation rates of populations and coherence of quantum states $|1\rangle$ and $|2\rangle$. Under definite conditions, the W value can be much less than homogeneous widths of spectral lines of optical transitions $|1\rangle \rightarrow |3\rangle$ and $|2\rangle \rightarrow |3\rangle$. The population of a nonclosed Λ -system (i.e., the total population of all the levels forming the Λ -system) almost completely vanishes at sufficiently intensive optical pumping when Eqn (1) is violated. However, according to the theoretical work [13], lower long-lived levels of an open Λ -system can keep about half the equilibrium population of an initial noncoherent molecular ensemble at the exact CPT condition $\delta_1 = \delta_2$ even in the case of such strong pumping. Thus, narrow high-contrast CPT peaks will arise in populations of long-lived levels $|1\rangle$ and $|2\rangle$ of a nonclosed Λ -system (Fig.1) versus the detuning difference $\delta_2 - \delta_1$. Corresponding CPT resonances will manifest themselves as dips in transmission of additional weak probe radiation resonant to a quantum transition from any lower level $|1\rangle$ or $|2\rangle$ at the scan of the frequency difference $\omega_2 - \omega_1$ of bichromatic pumping.

Recently we have studied experimentally such CPT resonances for nonclosed three-level Λ -systems formed by spectral components of the Doppler-broadened D_2 line of caesium atoms [15]. The CPT resonances were detected in absorption of probe monochromatic light beam under the action of counterpropagating two-frequency pump radiation. However, in [15], as monochromatic light source we used two independent diode lasers whose radiations were resonant to different optical transitions $|1\rangle \rightarrow |3\rangle$ and $|2\rangle \rightarrow |3\rangle$ (Fig. 1). Therefore, the detected CPT resonances had comparatively large characteristic widths (about 3–4 MHz) due to fluctuations of the frequency difference $\omega_2 - \omega_1$ of these lasers.

In this paper we applied the same method as in our previous work [15] but with the use of only one diode laser, which generates a monochromatic beam with a stabilised frequency ω_1 . The second coherent radiation component (with a frequency ω_2) was obtained from this initial beam by an electro-optical modulator. The frequency difference $\omega_2 - \omega_1$ was smoothly scanned around the microwave interval (9192.6 MHz) between hyperfine sublevels of the Cs ground term

(Fig. 2). In this experiment the use of the electro-optic modulator (EOM) has dramatically decreased the fluctuations of the frequency difference $\omega_2 - \omega_1$ of the bichromatic pump beam compared to the previous case of two independent laser beams described in [15]. Moreover, broadening effects due to nonideal parallelism have been eliminated in the configuration using a single laser and the EOM. Thus, the spectral resolution of our new setup was improved at least by an order of 1 as compared to the previous work [15]. As a result, new important features of nontrivial CPT resonances have been discovered. In particular, we have established that, at some conditions, CPT resonances in transmission of the weak probe beam exhibit not only a higher contrast but also are essentially narrower compared to well-known CPT resonances in transmission of the corresponding two-frequency pump radiation (or the light induced fluorescence of a gas medium).

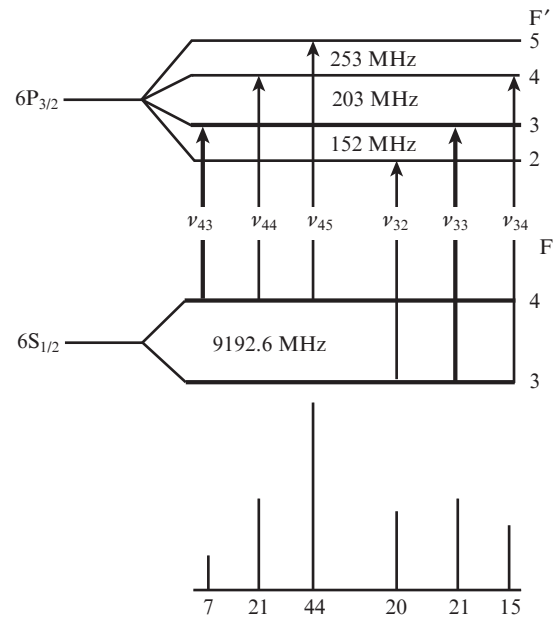


Figure 2. Energy level diagram of the ^{133}Cs D_2 line. The relative oscillator strengths of the lines, representing hyperfine transitions, are given at the bottom of the figure. CPT resonances are formed in the following two -systems of this scheme: $6S_{1/2}(F=3) - 6P_{3/2}(F'=3) - 6S_{1/2}(F=4)$ and $6S_{1/2}(F=3) - 6P_{3/2}(F'=4) - 6S_{1/2}(F=4)$.

2. Experimental method and setup

The scheme of the experimental setup is shown in Fig. 3. The laser frequency ω_1 was stabilised at the centre frequency ν_{33} of the caesium transition $6S_{1/2}(F=3) - 6P_{3/2}(F'=3)$ (Fig. 2) in a reference quartz Cs cell by using the saturation absorption spectroscopy technique. The output of the external cavity diode laser (ECDL) was phase modulated at the ^{133}Cs -atom hyperfine frequency 9192.6 MHz by the EOM, driven by the frequency synthesiser with harmonics less than 30 dB for obtaining the second component of the bichromatic pump beam with the frequency ω_2 close to the centre frequency ν_{43} of another caesium transition $6S_{1/2}(F=4) - 6P_{3/2}(F'=3)$ (Fig. 2). The sideband-to-carrier ratio of the resultant pump radiation was adjusted up to 50% when its component with the frequency ω_1 was two times stronger than another component with the frequency ω_2 . This adjustment was achieved by changing the radio frequency power of the frequency synthe-

various groups of atoms, whose velocity projections (along the wave vector \mathbf{k} of this beam) are approximately equal [16]:

$$V_{32} = \frac{\omega_1 - \nu_{32}}{|\mathbf{k}|}, V_{33} = \frac{\omega_1 - \nu_{33}}{|\mathbf{k}|}, V_{34} = \frac{\omega_1 - \nu_{34}}{|\mathbf{k}|}. \quad (2)$$

We note that narrow CPT resonances in absorption of the two-frequency pumping are formed in two different Λ -systems $6S_{1/2}(F=3) - 6P_{3/2}(F'=3) - 6S_{1/2}(F=4)$ and $6S_{1/2}(F=3) - 6P_{3/2}(F'=4) - 6S_{1/2}(F=4)$ (Fig. 2), which correspond to atomic velocity projections V_{33} and V_{34} in Eqn (2). Indeed, according to Fig. 2, the spectral interval (about 203 MHz) between these Λ -systems is essentially less than the Doppler width (460 MHz) of the corresponding optical transitions. At the same time, the counterpropagating monochromatic probe beam with the stabilised frequency $\omega_1 = \nu_{33}$ effectively interacts with pump radiation only via the single common group of atoms, whose velocity projections are close to the zero value. Thus, the sole Λ -system $6S_{1/2}(F=3) - 6P_{3/2}(F'=3) - 6S_{1/2}(F=4)$ mainly contributes to the CPT resonance in absorption of this probe wave.

Let us direct the quantisation axis along the same parallel linear polarisation of pump and probe laser beams. Under our experimental conditions (Fig. 3), when influence of an external magnetic field is negligible, according to known selection rules [16], only electro-dipole optical transitions between Zeeman degenerate Cs levels without a change in the magnetic quantum number m are induced (Fig. 4). Thus we have six nonclosed Λ -systems (corresponding to magnetic numbers $m = \pm 1, \pm 2$ and ± 3) for two adjacent resonant optical transitions $6S_{1/2}(F=3) - 6P_{3/2}(F'=3)$ and $6S_{1/2}(F=4) - 6P_{3/2}(F'=3)$ shown in Fig. 4. Optical repumping of the population of this resultant Zeeman degenerate Λ -system takes place at three magnetic sublevels of the ground Cs term $6S_{1/2}(F=4, m = \pm 4)$ and $6S_{1/2}(F=3, m = 0)$, which do not interact with incident pump and probe radiations. These three Zeeman sublevels are the analogue of the population 'exit' shown in Fig.1. The repumping of atoms to the 'exit' from other Zeeman states of lower levels $6S_{1/2}(F=3)$ and $6S_{1/2}(F=4)$ weakens the CPT condition (1). Taking into account the

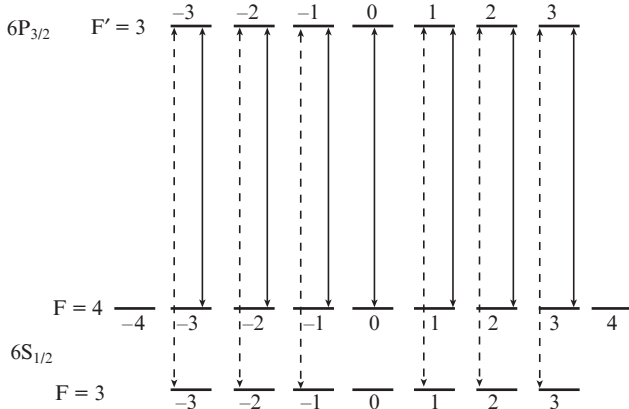


Figure 4. Diagram of Zeeman sublevels for the analysed Λ -system $6S_{1/2}(F=3) - 6P_{3/2}(F'=3) - 6S_{1/2}(F=4)$ of the Cs atom (Fig. 2). Arrows show allowed electro-dipole optical transitions, which are induced by pump and probe beams (with the same parallel linear polarisations) under our experimental conditions. One can see that three 'exit' magnetic sublevels of the ground term $6S_{1/2}(F=4, m = \pm 4)$, $6S_{1/2}(F=3, m = 0)$ do not interact with these pump and probe radiations.

given features, we will further analyse CPT resonances in absorption of the probe beam on the basis of the simple model of the nonclosed Λ -system (Fig. 1), where states $|1\rangle$, $|2\rangle$ and $|3\rangle$ correspond to levels $6S_{1/2}(F=3)$, $6S_{1/2}(F=4)$ and $6P_{3/2}(F'=3)$ (Fig. 2) with the exception of three indicated exit magnetic sublevels $6S_{1/2}(F=4, m = \pm 4)$ and $6S_{1/2}(F=3, m = 0)$.

Figure 5a presents the narrow CPT dip with the centre $\delta_1 = \delta_2 = 0$ in transmission of the weak probe light beam at the resonant transition $|1\rangle \rightarrow |3\rangle$. This transmission is defined by the difference between populations of levels $|1\rangle$ and $|3\rangle$. However, the detected CPT peak is determined mainly by the trapped atomic population at the lower level $|1\rangle$ in the non-closed Λ -system (Fig. 1) because of negligible population of its excited state $|3\rangle$ under the CPT condition (1). This statement was confirmed by our corresponding theoretical estimations, which were based on density matrix equations [1, 16] for the comparatively simple model of the nonclosed three level Λ -system. The well-known CPT resonance in transmission of the corresponding two-frequency laser pumping is shown in Fig. 5b. Unlike the CPT resonance in transmission of the probe beam (Fig. 5a), this resonance represents a peak because it is caused directly by a decrease in the population of the excited level $|3\rangle$ (Fig. 1) under the CPT condition (1). For both detected CPT resonances (Fig. 5), we have calculated contrasts C and widths W (at their half-heights) according to definitions given in [17]. In particular, the contrast of the CPT resonance was determined as the ratio of the amplitude of the resonance to the total transmission background level. The corresponding dependences of the values of C and W on the intensity of pump radiation are shown in Fig. 6. We clearly detected CPT resonances in absorption of the probe beam at the pump intensity $I > 2.5 \text{ mW cm}^{-2}$. Therefore, their parameters C and W in Fig. 6 are not indicated in the interval $0 < I < 2.5 \text{ mW cm}^{-2}$.

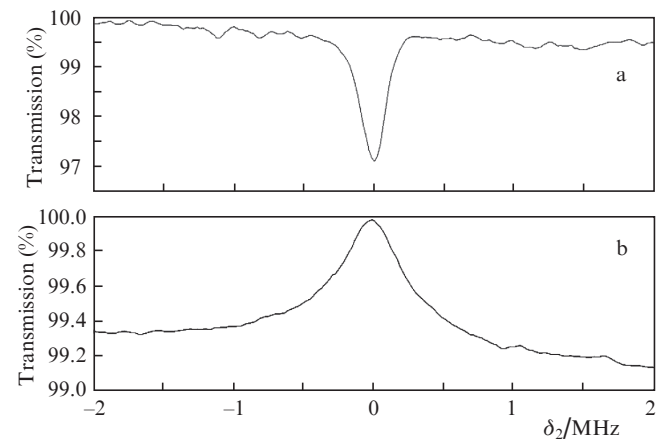


Figure 5. Detected CPT resonances in transmission of (a) the probe beam and (b) the corresponding two-frequency pump beam vs. the frequency detuning $\delta_2 = \omega_2 - \nu_{43}$ at the fixed detuning $\delta_1 = \omega_1 - \nu_{33}$ for the pump intensity $I = 7.64 \text{ mW cm}^{-2}$ and equal beam diameters $D_{\text{pump}} = D_{\text{probe}} = 5 \text{ mm}$.

Intensification of optical pumping causes more essential depletion of populations of lower levels $|1\rangle$ and $|2\rangle$ in the non-closed Λ -system (Fig. 1) when the CPT condition (1) is violated. Then, the amplitude of the CPT resonance (at point $\delta_1 = \delta_2$) increases. A similar situation takes place also with increasing duration of the optical pumping of atoms as they

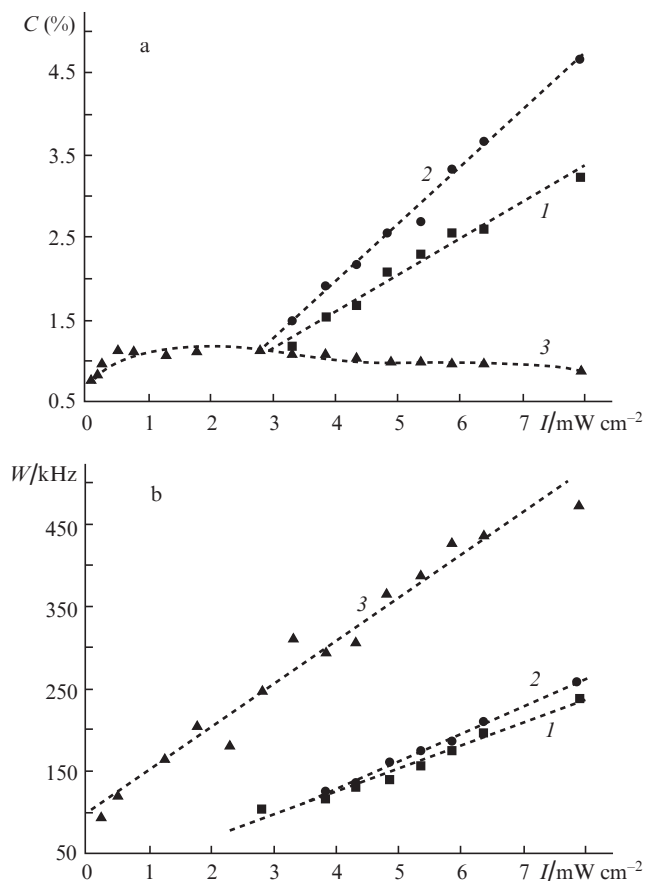


Figure 6. (a) Contrast C and (b) linewidth W of CPT resonances in transmission of the probe beam [curves (1, 2)] and the corresponding two-frequency pump beam [curves (3)] vs. the pump intensity I for the beam diameters $D_{\text{pump}} = D_{\text{probe}} = 5$ mm [curves (1)] and $D_{\text{pump}} = 5$ mm, $D_{\text{probe}} = 1.8$ mm [curves (2)].

transit to the central region of pump radiation in our rarefied gas medium. Therefore, the contrast C of the CPT resonance in transmission of the probe beam increases both with increasing pump intensity and with narrowing probe beam diameter to the central axis of the counterpropagating pumping with a fixed diameter $D_{\text{pump}} \geq D_{\text{probe}}$ [curves (1) and (2) in Fig. 6a]. At the same time, the nonmonotonic change takes place for the contrast C of corresponding known CPT resonances in transmission of two-frequency pump radiation (or fluorescence of the optically excited atomic state) [18, 19]. Indeed, the value of C decreases after some increase [curve (3) in Fig. 6a] at a growth of the pump intensity. Thus, the contrast of CPT resonances in absorption of the probe beam essentially exceeds the contrast of CPT resonances in absorption of the two-frequency pumping at its intensity $I > 3$ mW cm⁻² (Fig. 6a).

It is necessary to note that, even a small angle (\sim a few degrees) between linear polarisations of the pump and probe laser beams or a weak external magnetic field (~ 1 Gs) in the Cs cell (Fig. 3) lead to an essential decrease in contrasts of the recorded CPT resonances in transmission of the probe beam. Indeed, other schemes of allowed stimulated quantum transitions between Zeeman sublevels take place in comparison with those presented in Fig. 4. As a result, the analysed Λ -system $6S_{1/2}(F=3) - 6P_{3/2}(F'=3) - 6S_{1/2}(F=4)$ will not have any 'exit' states indicated in the simplified model in Fig. 1.

Under our experimental conditions with the irradiated rarefied Cs vapour (without a buffer gas), the widths of recorded CPT resonances (Fig. 5) are determined, mainly, by the power broadening and transit relaxation of atoms, connected with the atomic time of flight across laser beams. According to our theoretical estimations, characteristic widths of both considered CPT resonances should be close for a single nonclosed Λ -system (Fig. 1). However, experimental data (in Figs 5 and 6b) demonstrate essential difference between these widths. Indeed, we have established at the beginning of this section, that the CPT resonance in absorption of two-frequency pump radiation is a superposition of overlapping CPT resonances formed not only by the considered nonclosed Λ -system $6S_{1/2}(F=3) - 6P_{3/2}(F'=3) - 6S_{1/2}(F=4)$ (Fig. 4), but also by the additional Λ -system $6S_{1/2}(F=3) - 6P_{3/2}(F'=4) - 6S_{1/2}(F=4)$ (Fig. 2). Therefore, in our case of the Doppler broadened Cs D₂ line, the total width of the given CPT resonance is essentially larger than that of the corresponding resonance in absorption of the counterpropagating probe beam (Fig. 6b). An increase in the pump intensity leads to the power broadening of these CPT resonances (Fig. 5) according to quasi-linear dependences (Fig. 6b). However, the widths of the analysed CPT resonances in the indicated intensity interval are still much less than natural widths (about 5 MHz) of the corresponding optical transitions.

4. Conclusions

On the basis of the elaborated method and corresponding improved experimental setup (with a sufficiently high spectral resolution), we have detected and analysed narrow high-contrast CPT resonance characteristics for nonclosed atomic Λ -systems (Fig. 1). These CPT resonances are determined, mainly, by the trapped atomic population at the given lower level of the Λ -system. The resonant optical transition is induced exactly from this level by the weak probe monochromatic light beam counterpropagating with respect to the two-frequency laser pump beam. The applied method allows one to analyse the manifestation of the CPT phenomenon directly for a specific Λ -system between three selected levels (Fig. 1) even in sufficiently complicated spectra of atomic gases with large Doppler broadening. This is especially important when several Λ -systems contribute to the resultant CPT resonance in absorption of usual bichromatic radiation in given gases, which essentially complicates analysis of such resonances. Due to significant improvement of the spectral resolution of our experimental setup in comparison with the previous paper [15], we have established that CPT resonances in transmission of the probe beam exhibit not only a better contrast but also a much lesser width than the known CPT resonances in transmission of the corresponding two-frequency pump radiation (or fluorescence of the optically excited quantum state). Thus, our elaborated method can be used in the ultra-high resolution atomic spectroscopy for more detailed analysis of the CPT phenomenon. In our opinion, this method of sub-Doppler spectroscopy is promising also for corresponding research of open molecular Λ -systems, which can be qualitatively described by the simple scheme presented in Fig. 1. The results of this work are also important for the realisation of possible CPT applications in selective photophysics and photochemistry, which were suggested in papers [12, 13].

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