

Spectroscopy of coherent population trapping with a light source based on a femtosecond laser

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Abstract. A new source of the bichromatic laser field with a stable mean value of the difference frequency is used to study coherent population trapping (CPT). A high stability of the difference phase of the fields of the radiation source is achieved by phase locking the radiation frequency of two semiconductor lasers to longitudinal modes of a femtosecond laser. The first experimental results on the spectroscopy of CPT resonances in rubidium are presented. The possibility of using the new method for spectroscopy of CPT resonances in rare-earth atom vapours is discussed.

Keywords: coherent population trapping, femtosecond laser, phase locking.

1. Introduction

Mixed states in atomic systems and related nonlinear effects have attracted great attention of researchers in the last years [1–6]. One of the interesting types of such states is the so-called dark coherent population trapping (CPT) states taking place in a three-level Λ -system (Fig. 1) [7]. The essence of this phenomenon is the preparation of a long-lived superposition state of the lower levels in a system irradiated by an external bichromatic field with the components E_1 and E_2 having a stable difference phase, this state no longer interacting with the bichromatic field. The presence of a long-lived dark state in the atomic system is manifested in a narrow hole in the absorption or fluorescence band – a CPT resonance, which is also called electromagnetically induced transparency [7].

Both the dark CPT state and the corresponding resonance have found numerous practical and fundamental applications. It was proposed recently to use a dark CPT state as a memory cell for storing information on the difference phase and amplitude characteristics of the two light fields. In this case, it is possible to reconstruct completely the recorded light pulses. The carrier frequency of the reconstructed pulse can be chosen not only equal to the initial frequency but also to any frequency which is

resonant in a given atom with one of the transitions from one of the lower levels of the initial Λ -system [5, 8].

In [9], the original method was proposed for laser cooling of atoms involving a specially prepared CPT state, which allows cooling of atoms below the limit imposed by the recoil momentum. The idea is that an atom finding itself in a dark state does not interact with cooling fields, which results in the accumulation of atoms in this state. Due to a long lifetime of the dark state, atoms are gathered during cooling into a narrow-velocity group corresponding to the CPT state, the half-width of the obtained velocity distribution being determined by the lifetime of the resonance rather than the recoil momentum. Note, however, that if the resonance is formed in a sufficiently broad velocity group, it can prevent the cooling of atoms [10].

Due to the possibility of detecting narrow spectral lines in cells with atomic vapours, the CPT resonances found applications in metrology for the development of magnetometric devices and frequency standards [6, 11]. The sensitivity of modern magnetoradiometers achieves a few pT cm⁻¹ [12].

Most of the studies of CPT resonances were performed with alkali atoms because the use of the fine-structure components of the ground state as the lower levels of the Λ -system allows one to anticipate that the lifetime of the CPT state will be quite long. In addition, the typical splitting of the ground-state components in these atoms lies in the radio-frequency range, which substantially simplifies the problem of producing a bichromatic field with a stable difference phase. One of the most efficient methods for producing a bichromatic field, which is used at present in the spectroscopy of CPT resonances in alkali atoms, is the method of direct modulation of the laser-diode current, which provides the required characteristics of the source in the setup of the minimal volume [11, 12]. This method, however, allows only the generation of fields with identically polarised components, which reduces the contrast of observed resonances [13].

The CPT resonances in systems with the splitting of lower levels corresponding to frequencies in the IR or even visible spectrum have the potential Q factor which considerably exceeds the Q factor of resonances in alkali atoms. An increase in the Q factor of the resonance in a properly selected atomic system improves the accuracy of ‘atomic clocks’ and magnetometers and also expands the scope of atomic systems used for data storage in quantum memory cells [8, 14, 15].

The theoretical and experimental study of CPT in the Λ -system of the ¹⁵⁴Sm atom with a large splitting of lower

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levels ω_{12} , which was equal to $2\pi \times 10$ THz, was performed in papers [14–16]. The Q factor of resonances demonstrated in experiments proved to be comparatively low and was mainly determined by the instability of the difference radiation frequency of two independent semiconductor lasers generating the components of a bichromatic field. For such a large splitting of the lower levels of the Λ -system, it is impossible to use modulation or other direct radio-frequency techniques.

Recently, we developed a source of bichromatic field based on the spectral properties of longitudinal modes of a passively mode-locked femtosecond laser (the so-called femtosecond frequency comb) [17]. Such a source produces a bichromatic field with the frequency difference lying within the femtosecond-comb spectrum, whose width is determined by the laser pulse duration. We used a femtosecond laser emitting ~ 800 -nm, 120-fs pulses with the spectral width of about 10 THz. Because the lines of the Λ -system of the samarium atom do not fall into this spectral interval, we used the $5s\ 5s\ S_{1/2}(F=2) \leftrightarrow 5s\ P_{3/2}(F=2) \leftrightarrow 5s\ S_{1/2}(F=3)$ Λ -system in the ^{85}Rb atom (components of the D_2 -line of rubidium) for testing and studying characteristics of the new source. In this paper, we present our first experimental results obtained with the help of the new bichromatic source.

The extensive development of fibre optics and its application for expanding the femtosecond-comb spectrum [18, 19] suggests the possibility of using this method for studying samarium atoms [14], barium ions [20] and other atomic systems with lower-level splittings lying in the optical spectral region.

2. Coherent population trapping resonance

The CPT effect was described theoretically in detail in a number of papers [7, 13, 14, 25]. Consider for simplicity a Λ -system consisting of the $|1\rangle$, $|2\rangle$ and $|3\rangle$ levels (Fig. 1). Transitions from the long-lived $|1\rangle$ and $|2\rangle$ levels to the upper $|3\rangle$ level are dipole-allowed transitions. The system is

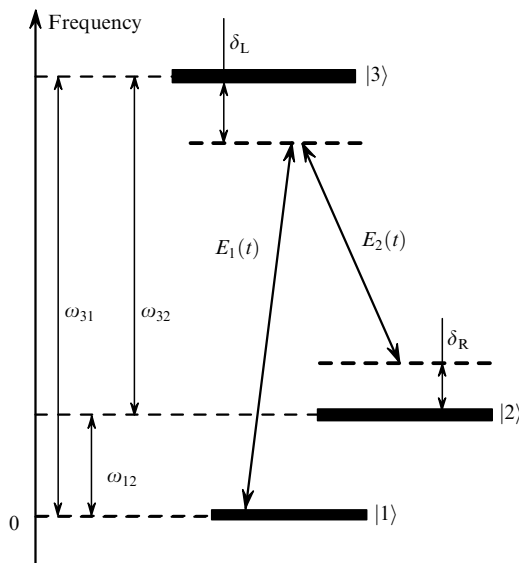


Figure 1. Three-level Λ -system. The $|1\rangle$ and $|2\rangle$ levels are long-lived levels of the same parity, from which the dipole transition to the $|3\rangle$ level is allowed; δ_R and δ_L are the Raman and laser detunings.

subjected to the action of an external bichromatic field, whose components $E_1(t)$ and $E_2(t)$ have the form

$$E_1(t) = E_1 \exp[-i\omega_1 t - i\varphi_1(t)], \quad (1)$$

$$E_2(t) = E_2 \exp[-i\omega_2 t - i\varphi_2(t)].$$

The system can be clearly represented in the orthogonal basis

$$|+\rangle = \frac{1}{\Omega} (\Omega_1^* |1\rangle + \exp[-i\omega_{12} t - i\Delta\varphi] \Omega_2^* |2\rangle), \quad (2)$$

$$|-\rangle = \frac{1}{\Omega} (\Omega_2 |1\rangle - \exp[-i\omega_{12} t - i\Delta\varphi] \Omega_1 |2\rangle).$$

Here, ω_{12} is the splitting between the $|1\rangle$ and $|2\rangle$ levels; Ω_1 and Ω_2 are the Rabi frequencies for the $|1\rangle \leftrightarrow |3\rangle$ and $|2\rangle \leftrightarrow |3\rangle$ transitions:

$$\Omega_1 = -\frac{d_{13} E_1}{\hbar}, \quad \Omega_2 = -\frac{d_{23} E_2}{\hbar}, \quad \Omega = (|\Omega_1|^2 + |\Omega_2|^2)^{1/2}. \quad (3)$$

By representing the electric dipole operator in the form

$$V = \frac{\hbar\Omega_1}{2} \exp[-i\omega_1 t - i\varphi_1] |3\rangle\langle 1| + \frac{\hbar\Omega_2}{2} \exp[-i\omega_2 t - i\varphi_2] |3\rangle\langle 2| + \text{H.c.}, \quad (4)$$

we can calculate the matrix elements $\langle 3|V|+\rangle$ and $\langle 3|V|-\rangle$ determining the interaction of the $|+\rangle$ and $|-\rangle$ states with the external bichromatic field. The most important is the case of a stable phase difference, for example $\Delta\varphi = \varphi_1(t) - \varphi_2(t) = 0$, and zero Raman detuning $\delta_R = \omega_1 - \omega_2 - \omega_{12} = 0$. In this case, as one can easily verify, the $|-\rangle$ state does not interact with the external field, i.e.,

$$\langle 3|V|-\rangle = 0, \quad \langle 3|V|+\rangle \sim \hbar\Omega. \quad (5)$$

Therefore, the $|-\rangle$ state is a dark state for the bichromatic field, and the entire population from the $|+\rangle$ state is accumulated in the $|-\rangle$ state due to optical pumping. Two substantial features should be emphasised: the high sensitivity of the resonance to the stability of the relative phase of light fields and the fact that the $|-\rangle$ state is a dark state only for the given bichromatic field (1) with indicated phase relations.

Indeed, when the relative phase $\Delta\varphi$ of the fields changes abruptly by π , the $|-\rangle$ state begins to interact with the field and is no longer dark. In the case of a sufficiently abrupt change in the relative phase $\Delta\varphi$ of components of the bichromatic field, the formation of the dark state becomes inefficient, resulting in the disappearance of the CPT resonance.

The main characteristics of the resonance determining the possibility of its various applications are its width, frequency, and contrast. The width Γ_{CPT} of the CPT resonance can be written within the framework of a simplified model as

$$\Gamma_{\text{CPT}} = \Gamma_{\text{coh}} + \Gamma_{\text{pump}}, \quad (6)$$

where Γ_{coh} is the contribution from dephasing of the lower levels $|1\rangle$ and $|2\rangle$ and Γ_{pump} is rate of pumping to the $|-\rangle$

dark state. The value of Γ_{coh} can be represented, in turn, as the sum of three contributions

$$\Gamma_{\text{coh}} = \Gamma_{12} + \Gamma_{\text{field}} + \Gamma_{\text{time}}. \quad (7)$$

Here, Γ_{12} is the width determined by the lifetime of the lower levels $|1\rangle$ and $|2\rangle$ of the Λ -system; Γ_{field} is the dephasing of the laser fields E_1 and E_2 ; and Γ_{time} is the time-of-flight broadening. Because CPT is usually studied in systems with long coherence lifetimes of lower levels, we can assume that $\Gamma_{12} \ll \Gamma_{\text{coh}}$.

As the pressure P of the buffer gas in a cell is increased, Γ_{time} and Γ_{pump} decrease as $1/P$ [21]. Therefore, in the case of completely phase-locked exciting fields ($\Gamma_{\text{field}} = 0$), the width of the CPT resonance proves to be inversely proportional to the buffer-gas pressure in a cell:

$$\Gamma_{\text{CPT}} \sim \frac{1}{P}. \quad (8)$$

This important result is often used for exciting CPT resonances in cells filled with metal vapours and buffer gas [6, 7].

At high buffer-gas pressures, transitions between the velocity groups of atoms induced by collisions accompanied by changes in the velocity begin to play a considerable role [21]. As a whole, the width of the resonance decreases down to pressures of a few thousand pascals, and then it begins slowly to increase due to collision broadening of lower levels and the corresponding increase in Γ_{12} .

The increase in the buffer-gas pressure causes not only the narrowing of the resonance but also eliminates parasitic effects produced by optical pumping [16]. At buffer-gas pressures of the order of 1 kPa, the spectral structure related to optical pumping of the upper and lower levels becomes completely blurred due to collisions, while the CPT resonance is not destroyed.

However, if the exciting laser fields are not phase-locked, the pressure dependence of the resonance width disappears due to a large contribution of Γ_{field} (of the order of the laser linewidth), and the limiting width is determined by the condition $\Gamma_{\text{coh}} \simeq \Gamma_{\text{field}}$. Therefore, it is convenient to study resonances by using phase-locked light fields E_1 and E_2 .

3. Phase locking

The main concepts of stabilisation of the relative phase (phase locking) of two initially independent lasers with a relatively broad radiation spectrum were proposed and realised for He–Ne lasers in 1987 [22]. However, this method gained wide acceptance in the optical range only after phase locking of two tunable semiconductor lasers with an external resonator was achieved in 1995 [23]. The main idea of phase locking of lasers with a broad emission spectrum consists in the use of digital feedback loops having the dynamic range of a few tens π for the difference phase. Compared to analogue schemes, such loops provide a considerably more stable phase locking because they allow one to work off phase jumps over several periods, although they work somewhat slower.

Note that the developed phase-locking methods provide the stabilisation of the relative phase only for radiation sources for which the difference of carrier frequencies lies in the radio-frequency range. The matter is that the phase-

locked loop control of the difference frequency of light fields is performed with respect to the frequency of a highly stable reference oscillator (Fig. 2). In this case, the input channels of the phase-locking circuit directly count the frequencies of the beat signal of the laser fields and reference oscillator and then compare them. The error signal obtained in this way is directed to the frequency control units of the controllable laser. Semiconductor lasers with an external resonator have the advantage of not only a prompt action on the lasing frequency by varying the pump current (fast phase-locking channel), but also of the ability of compensating for drifts by rotating the laser-resonator grating (slow phase-locking channel). In addition, they have a comparatively broad tuning range (~ 3 GHz).

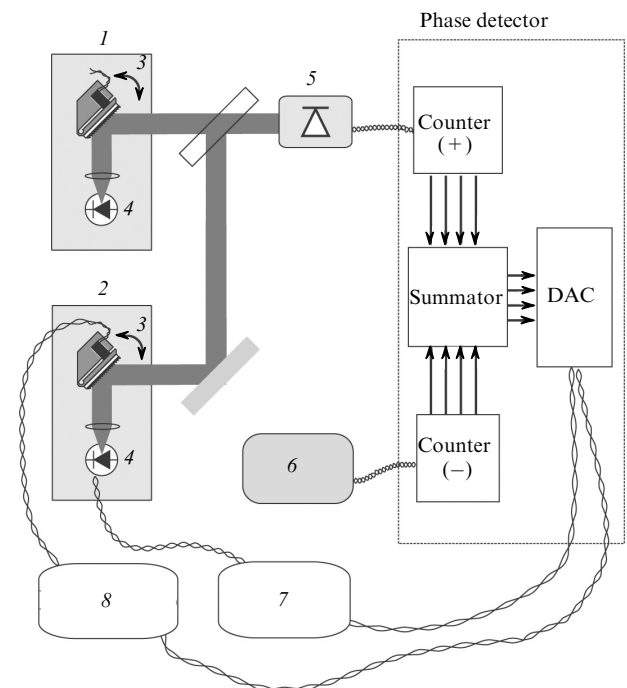


Figure 2. Principal phase-locking scheme for two semiconductor lasers: (1) master laser; (2) slave laser; (3) diffraction gratings on a piezoelectric ceramics; (4) laser diodes; (5) photodiode; (6) reference generator; (7) fast phase-locking scheme (to control the diode current); (8) slow phase-locking scheme (to control the rotation angle of the diffraction grating).

As mentioned above, the Λ -systems with a large splitting ω_{12} of the lower levels $|1\rangle$ and $|2\rangle$ (Fig. 1), which substantially exceeds the operation range of modern electronic devices, are promising for CPT studies. To lock these substantially different frequencies, we used a femtosecond frequency comb [17].

The method is based on the use of the spectral properties of radiation of a repetitively pulsed laser. The longitudinal radiation modes of such a laser with a stable pulse repetition rate prove to be strictly phase-locked. For example, it was shown in [24] that the modes of a femtosecond laser are equidistant with an accuracy of 3×10^{-17} and correspond to the pulse repetition rate with an accuracy of $\sim 6 \times 10^{-16}$. If two semiconductor lasers are phase-locked to different radiation modes of a femtosecond laser, the radiation of semiconductor lasers can be treated as a bichromatic field with a stable difference frequency and a stable phase

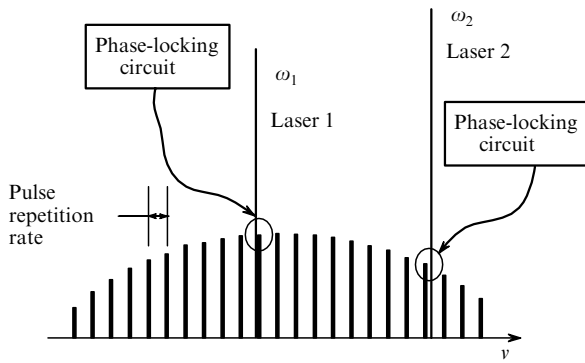


Figure 3. Principal scheme of a bichromatic source stabilised by a femtosecond laser. Feedback loops stabilise the phases of lasers 1 and 2 with respect to modes of the femtosecond comb, thereby stabilising them with respect to each other.

(Fig. 3). In this case, the stability of the difference frequency required for a reliable recording of narrow CPT resonances is determined only by the stability of the pulse repetition rate of the femtosecond laser and is independent of the absolute frequency of laser modes. This allows one to avoid the control or stabilisation of the absolute value of the carrier frequency of a femtosecond laser, which is commonly used for measuring optical frequencies with the help of a femtosecond comb [24].

4. Experiment

Figure 4 shows the scheme of the experimental setup. 120-fs pulses from a Verdi-V8 + MIRA-900F femtosecond system (Coherent Inc.) with a repetition rate of ~ 75 MHz propagated in two channels. In each of the channels, femtosecond radiation was combined with radiation from one of the diode lasers and was incident on a diffraction grating. A 1800-lines mm^{-1} diffraction grating was used for preliminary selection of modes of the femtosecond comb and also as a polariser of radiation required for obtaining a beat signal. In front of the grating, a $\lambda/2$ plate was mounted to adjust the relation between the intensities of light fields providing the optimal signal-to-noise ratio. A photodiode detected the beat signal of comb modes with radiation of the semiconductor laser. The beat signal filtered by a system of radio-frequency filters was directed to a phase-locking electronic circuit controlling the frequency of semiconductor lasers.

A fraction of radiation from each of the diode lasers was directed to the detection channel. Radiation from laser 1 passed through an acousto-optic modulator (AOM) with a central frequency of 200 MHz, which operated in a two-pass scheme allowing a continuous tuning of the difference frequency of phase-locked lasers. The difference frequency was roughly changed with the help of an oscillator generating pulses of a special shape, whose signal was fed directly to a piezoelectric ceramics of the laser resonator. The leading edge of the pulse is sufficiently steep ($\tau_1 > 1 \mu\text{s}$), and a slow phase-locking loop controlling the piezoelectric ceramics has no time to work off the leading edge. The pulse amplitude is adjusted so that a change produced in the laser frequency is equal to the mode interval in the femtosecond comb. The trailing edge of the pulse is quite flat ($\tau_2 > 30 \mu\text{s}$), and the phase-locking loop easily works it off, the laser frequency remaining invariable. The AOM tuning range

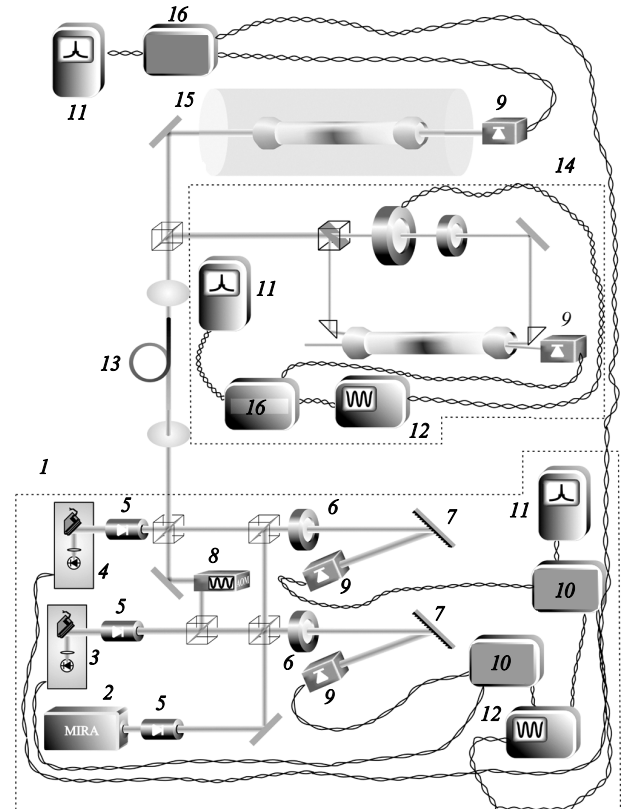


Figure 4. Scheme of the experimental setup: (1) bichromatic source with a stabilised difference frequency; (2) femtosecond laser; (3) diode laser 1 with an external resonator; (4) diode laser 2 with an external resonator; (5) optical isolators; (6) $\pi/2$ plates; (7) diffraction gratings; (8) AOM; (9) photodiodes; (10) phase-locking units; (11) digital oscilloscopes; (12) local oscillator; (13) single-mode fibre; (14) scheme for recording saturated absorption in counterpropagating beams; (15) magnetically screened cell with rubidium vapours; (16) lock-in amplifiers.

covers the mode interval (75 MHz) of the femtosecond laser, so that all the frequencies lying within the emission band of the femtosecond laser (~ 10 THz) become available.

The stability of the difference frequency of the bichromatic source was determined by studying the beat signal of two phase-locked semiconductor lasers. For this purpose, radiation of the lasers was combined on a photodiode and then the beat-signal frequency was reduced to 100 Hz by the heterodyne method. In this case, the difference frequency of the laser fields was equal to five mode intervals (375 MHz) of the femtosecond laser. The width of the beat-signal spectrum is mainly determined by the instability of the pulse repetition rate of the femtosecond laser and by the noise of phase-locking systems. In our case, the width of the signal spectrum recorded with an ASK-3106 spectrum analyser (ATAKOM) during 1 s was 1 Hz and was mainly determined by the drift of the pulse repetition rate of the femtosecond laser (0.2 Hz s^{-1}).

Radiation of semiconductor lasers in the detection channel was combined in a polarisation cube and transmitted through a polarisation-preserving single-mode fibre with the 0.16 numerical aperture and 4.2- μm fibre core diameter. An objective ($f = 20$ mm) behind the fibre formed one spatial mode for both components of bichromatic radiation. Bichromatic radiation obtained in this manner was directed either to the detection channel of saturated

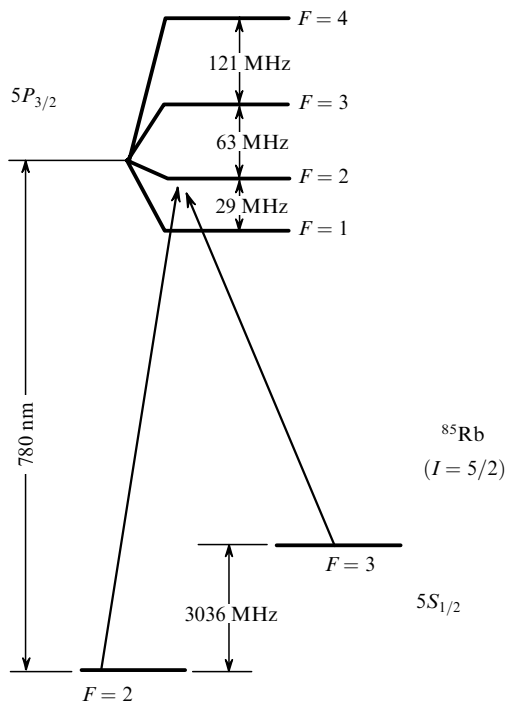


Figure 5. Energy level diagram of the rubidium atom and the Λ -system under study.

absorption or to the detection channel of CPT resonances. The intensities of field components were $\sim 50 \mu\text{W}$.

The maximum of emission of the femtosecond laser lies at the wavelength close to the D_2 line of rubidium (780 nm). Therefore, we first studied the Λ -system shown in Fig. 5. The diode lasers were preliminary tuned to the wavelength of the corresponding transition by the saturated absorption signal detected in a standard configuration with counter-propagating beams by modulating the saturating beam. The typical saturated absorption spectrum is shown in Fig. 6.

Because the contrast of CPT resonances for this Λ -system at high buffer-gas pressures proved to be very low (less than 1%), we detected resonances by the modulation technique. The frequency of a reference oscillator in the

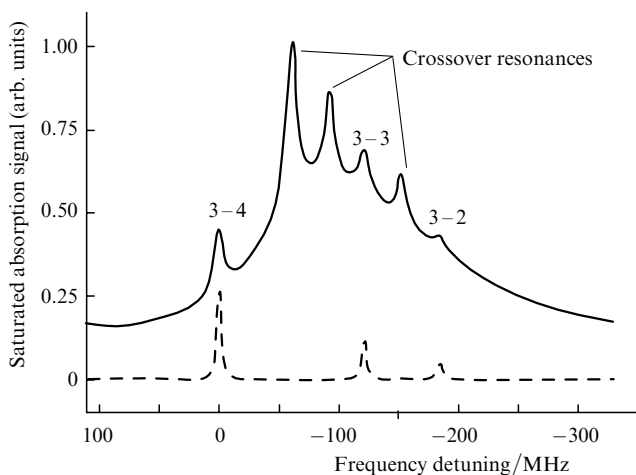


Figure 6. Typical saturated absorption spectrum in the region of the D_2 -line of the rubidium atom. The dashed curve shows the positions of spectral lines.

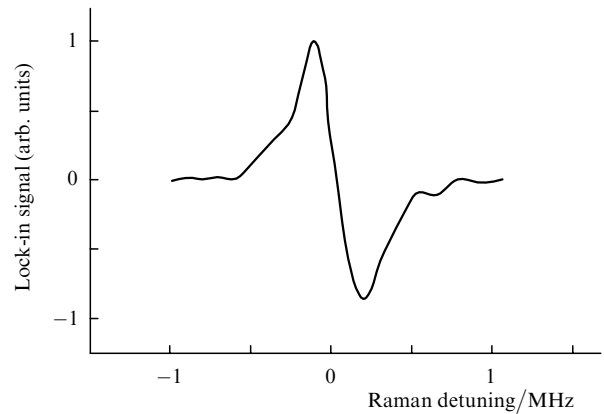


Figure 7. CPT resonance at a low buffer-gas pressure ($P < 1 \text{ Pa}$).

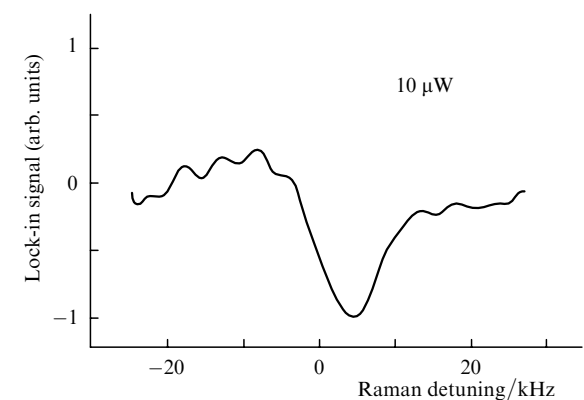
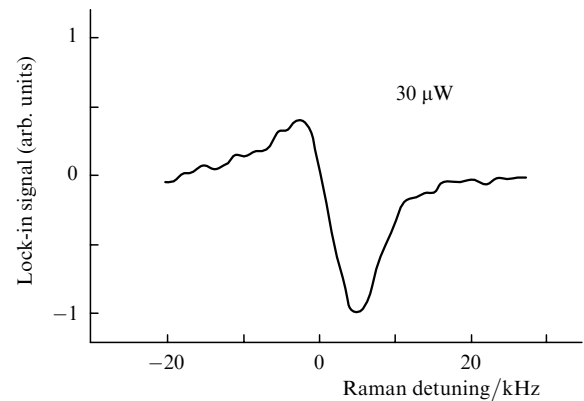
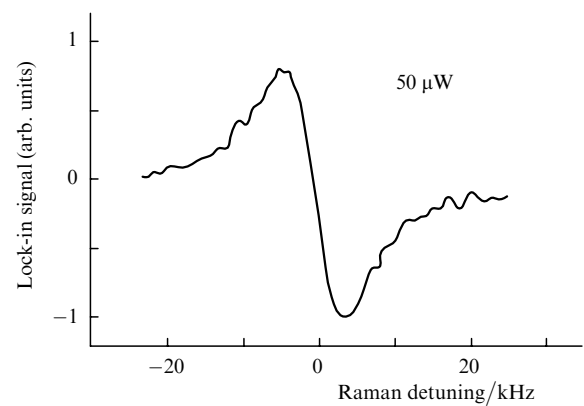


Figure 8. CPT resonances in a cell with a buffer gas for $P = 2.5 \text{ kPa}$ and different powers of laser 2.

phase-locking channel of laser 2 was modulated at the frequency 590 Hz, and the transmission of the bichromatic field in a cell with rubidium vapours was detected at the same frequency. In the absence of the buffer gas in the cell, the width of recorded resonances was ~ 500 kHz (Fig. 7). However, already at a pressure of 2.5 kPa, the characteristic width of the resonance decreased to ~ 10 kHz (Fig. 8). The modulation amplitude in the first case was 150 kHz, and in the second ~ 3 kHz.

When the power of laser 2 was reduced, the resonances became asymmetric. In the general case the shape of the CPT resonance for alkali atoms can be represented by a sum of Lorentzian and dispersion Lorentzian shapes [25, 26], the resonance asymmetry being determined by the ratio of light-field intensities and the laser detuning δ_L from zero, which was not controlled in experiments. For this reason, the resonances were approximated by a combination of the absorption and dispersion Lorentzians. The width of the resonance estimated at the zero power of laser 1 was 6.9 ± 0.5 kHz, and the width of the light broadening was 0.13 ± 0.1 kHz μW^{-1} .

5. Conclusions

We have demonstrated a new efficient method for recording CPT resonances by the example of the $5s\,S_{1/2}(F=2) \leftrightarrow 5s\,P_{3/2}(F=2) \leftrightarrow 5s\,S_{1/2}(F=3)$ three-level system of the rubidium atom. We recorded the CPT resonances in this system, studied the dependence of the width and shape of resonances on the relative intensity of components of a bichromatic field and buffer-gas pressure. The parameters of a developed bichromatic radiation source were analysed. A high stability of the difference frequency of the radiation source allows us to hope to record subkilohertz CPT resonances with the difference frequency up to 10 THz. The technique developed in our work will be used for the spectroscopy of CPT resonances in vapours of rare-earth elements, in particular, the samarium atom.

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