Raman–Ramsey pulsed excitation of coherent population trapping resonances in a ⁸⁷Rb cell with a buffer gas

V.N. Baryshev, G.V. Osipenko, M.S. Aleinikov, I.Yu. Blinov

Abstract. We report the results of experimental investigation of the Raman-Ramsey method of pulsed excitation of coherent population trapping (CPT) resonances in a rubidium vapour cell filled with a mixture of Ar and Ne buffer gases. For the development of compact quantum microwave frequency standards, this method is an alternative to the pulsed optical pumping method recently implemented using the same rubidium cell. Both methods have advantages over the traditional double radio-optical resonance technique, the main of which is the substantial suppression of the light shift of the clock transition frequency, since the evolution of the coherence of atomic states forming the clock transition occurs in the absence of laser radiation in the cell. The narrow (110-240 Hz) Raman-Ramsey resonances are obtained using the scheme of pulsed excitation of CPT resonances on the D₁ lines of the ⁸⁷Rb atom with the same linear (lin||lin) polarisations of the bichromatic laser radiation components. The process of optimising the central fringe linewidth, its contrast, and the linewidth-tocontrast ratio is described. The magnetic dependence of the Raman-Ramsey line central fringe linewidth is experimentally investigated.

Keywords: rubidium vapour quantum frequency standard, pulsed optical pumping, coherent population trapping, pulsed excitation of coherent population trapping resonances, diode laser, acousto-optic modulator, electro-optic modulator.

1. Introduction

In our recent paper [1], we presented the results of an experimental study of quantum frequency standards (QFS's) based on rubidium vapour with pulsed optical pumping (POP). In the process of optimising the duration and intensity of the pump and detection pulses, narrow (150–200 Hz) Ramsey lines with a contrast of the central fringe exceeding 40% were obtained. The signal-to-noise ratio (SNR) of the detected optical signal was ~30000, and the instability of the QFS in the shot noise limit, expressed as the Allan variation, can be estimated as $\sigma_y(\tau) \sim 2 \times 10^{-14}/\sqrt{\tau}$ (τ is the time of averaging). The aim of our work, as well as the work of other research groups [2–4], is to build a compact (volume ~10 L, mass

Received 24 October 2018; revision received 25 January 2019 *Kvantovaya Elektronika* **49** (3) 283–287 (2019) Translated by V.L. Derbov ~10 kg) microwave QFS based on a rubidium vapour cell with POP having a stability comparable to or even better than the stability of commercial passive hydrogen masers. The impressive results obtained in Refs [2–4] make it possible to consider the possibility of using a QFS with POP as a synchroniser of local time scales, including on-board scales of global navigation satellite systems.

However, at present, a frequency standard, based on a different physical effect - the QFS with pulsed excitation of coherent population trapping (CPT) resonances - is being widely developed as an alternative to QFS with POP. The contrast of the clock transition line in such QFS's (of the order of several tens of percent) is comparable to the contrast obtained using the pulsed optical pumping technique [5]. In the QFS based on the CPT effect in the traditional Λ -configuration of a three-level atomic system, when an atom is interrogated by bichromatic radiation with the same circular polarisation of components, only a small fraction of the atoms participate in the formation of a magnetically insensitive clock CPT resonance, thus reducing its contrast to only ~ 1 %. Most atoms are pumped to Zeeman magnetic sublevels with the maximum magnetic quantum number m. In order to avoid pumping atoms into trap states, several new schemes were proposed for the formation of high-contrast CPT resonances in both cw and pulsed regimes of interaction of bichromatic radiation with an atom: 1) a scheme with alternating direction of circular polarisation of the bichromatic radiation components (push-pull optical pumping) [5-7]; 2) a scheme with mutually perpendicular linear polarisations of the bichromatic radiation components ($lin \perp lin$) [8]; and 3) a scheme with parallel linear polarisations of the bichromatic radiation components (lin||lin) [9-11]. It was the third scheme, the simplest one, which we chose to study and develop the Raman-Ramsey technique of pulsed excitation of CPT resonances, since its implementation did not require significant changes in the experimental setup used before for the QFS with POP [1].

This paper presents the results of an experimental study of the Raman–Ramsey method of pulsed excitation of CPT resonances on the D₁ line of the ⁸⁷Rb atom in a cell with a mixture of Ar and Ne buffer gases with the same linear (lin||lin) polarisation of the bichromatic laser radiation components. The Raman–Ramsey line is presented. The process of optimisation of its central fringe linewidth, contrast value and linewidth-to-contrast ratio are described. The magnetic dependence of the central fringe linewidth, which is formed in the experimental lin||lin configuration by the CPT transitions between magnetic sublevels with $m = \pm 1$, is investigated [9].

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2. Experimental setup and results

The research and development of the Raman-Ramsey technique using pulsed excitation of CPT resonances on the D₁ lines of the 87Rb atom with the same lin||lin polarisation of the bichromatic laser radiation components were carried out on the same experimental setup that was used to study the operation principle of the QFS with POP. Described in Ref. [1], the prototype of the QFS with POP consisted of three main parts, which have undergone the following structural and functional changes. We did not introduce any constructive changes in the design of the prototype comprising a ⁸⁷Rb vapour cell filled with a mixture of Ar and Ne buffer gases at a total pressure of 14.7 Torr and a partial pressure ratio of 0.5, placed in a cylindrical microwave resonator; a solenoid producing a magnetic field B along the cavity axis; magnetic shields; and a thermostat. When conducting experiments on the development of the Raman-Ramsey technique of pulsed excitation of CPT resonances, the microwave cavity (6.834 Hz) was not used. An external-cavity diode laser (ECDL) based on a diffraction grating in the Littrow configuration provided radiation at $\lambda = 795$ nm, necessary for the formation of optical pumping and detection pulses. Developed at the VNIIFTRI, a single-pass acousto-optic modulator (AOM, -170 MHz) operating in Bragg anisotropic diffraction regime was used, like in Ref. [1], to generate optical pulses. To produce a bichromatic light field, the AOM output diffracted radiation was fed to an input of a polarisation-maintaining fibre coupled Photline NIR-MPX800-LN-10 electro-optical modulator (EOM). The EOM was controlled by a sinusoidal signal with a frequency of 3.417 GHz and a power of 100 mW. Almost all the power of the frequency-modulated radiation at the EOM output was almost equally divided between the two first-order sideband components. The spatially filtered radiation at the EOM output passed through a telescope, which increased the diameter of the light beam to 10 mm. As a result, the light beam had the same diameter and the same spatial position relative to the gas cell and the photodetector, as those of the prototype described in Ref. [1]. The system for generating the interrogation microwave signal and the digital control system that forms all the optical and electrical signals of the QFS prototype (now with pulsed excitation of the CPN resonances) have not been changed with the exception that the 3.417 GHz signal applied to the EOM served as the interrogation signal.

Thus, to test the technique of pulsed excitation of Raman-Ramsey CPT resonances on the D_1 line of the ⁸⁷Rb atom and produce the lin||lin polarisation of the bichromatic laser radiation components, we used a simple optical setup consisting of one laser source and an external optical phase modulator. Note that in our experiments the intensity of the frequencymodulated radiation directed into the cell with rubidium was limited by the maximum radiation power of the ECDL $(450\,\mu\text{W})$ and amounted to 570 μW cm⁻². The moderate value of the maximum contrast obtained in our experiments is explained only by this limitation. The linewidth of the clock CPT resonance in the cw regime with a maximum contrast of 4.0% at a cell temperature of 65 °C was ~1.5 kHz. Increasing the radiation intensity to the maximum value, we did not reach the regime, when the contrast of the CPT resonance becomes constant.

To observe high contrast CTP resonances in the lin||lin configuration of the light field, it is necessary to fulfill two conditions [9]. Excitation of CPT resonances in this scheme is possible only for alkali atoms with nuclear spin I = 3/2, in particular for the D₁ line of the ⁸⁷Rb atom, when two hyperfine levels of the ground state with $F_g = 1$ and $F_g = 2$ are coupled by a light field in Λ configuration with the excited level having $F_e = 1$ (Fig. 1). Another necessary condition is a good spectral resolution of the structure of hyperfine levels with F_e = 1 and $F_e = 2$ of the excited states. The hyperfine splitting of the excited level of the ⁸⁷Rb atom is 812 MHz.

In Fig. 1, the arrows show two CPT transitions between the levels $|F_g = 1, m = -1\rangle \leftrightarrow |F_g = 2, m = +1\rangle$ and $|F_g = 1, m =$ $+1\rangle \leftrightarrow |F_g = 2, m = -1\rangle$ of the ground state with the common level $|F_e = 1, m = 0\rangle$. Thus, the investigated magnetic field insensitive clock CPT resonance is formed by the superposition of these two transitions, whose frequencies are symmetrical with respect to the frequency Δ_{hfs} of the ground level hyperfine splitting. The linewidth of this clock CPT resonance depends on the magnitude of the magnetic field [9].

One can see from Fig. 1 that in addition to the central CPT resonance, there is a pair of magnetic field sensitive resonances, $|F_g = 1, m = -1\rangle \leftrightarrow |F_g = 2, m = -1\rangle$ and $|F_g = 1, m = +1\rangle \leftrightarrow |F_g = 2, m = +1\rangle$, between the levels with the same magnetic quantum number *m*. The frequencies of these CPT resonances are shifted from the frequency Δ_{hfs} of the hyperfine splitting by the Zeeman frequency $\pm f_Z$ [$f_Z = 700 \times 10^7 B$, where the magnetic field induction *B* is expressed in tesla].



Figure 1. Scheme of two-photon transitions between the levels $F_g = 1, 2$ and the level $F_e = 1$ in a bichromatic light field in lin || lin configuration.

Figure 2 shows the time sequence of pumping into the dark state t_p and detection t_{det} pulses. The optimal parameters are as follows: the pumping pulse duration $t_p = 3.5$ ms, the detection pulse duration $t_{det} = 0.02$ ms, and the time between the end of the pumping pulse and the start of the detection pulse T = 0.42 ms. The amplitude of the detection pulse is almost four times smaller than that of the pumping pulse. The cell temperature is 65 °C.

Figure 3 shows the Raman-Ramsey line obtained when the signal driving the EOM is swpt around the centre frequency of 3.4173413 GHz. The contrast of the central fringe is C = 3.6%. Its linewidth at half-maximum $\Delta v = 240$ Hz is significantly smaller than the linewidth expected for the Ramsey regime ($\Delta v < 1/(2T) = 1190$ Hz). Two field sensitive



Figure 2. Time sequence of pulses pumping the dark state t_p and detection pulses t_{det} .

Raman-Ramsey lines are clearly visible, corresponding to the CPT resonances $|F_g = 1, m = -1\rangle \leftrightarrow |F_g = 2, m = -1\rangle$ and $|F_g = 1, m = 1\rangle \leftrightarrow |F_g = 2, m = 1\rangle$ between the levels with the same magnetic quantum numbers. It is interesting to note that in Ref. [1], Ramsey lines on magnetic transitions with $\Delta F_g = 1$, $\Delta m = 0$ were not observed at all due to the inhomogeneity of the magnetic field. This fact means that in our experiment the pulsed regime of excitation of the CPT resonances is not the pure Ramsey regime. In our case, the Ramsey condition $T \gg t_p$ is not satisfied. The explanation why the central fringe linewidth, which also depends on the light intensity, is smaller than the width 1/(2T) determined by the Ramsey condition, is given in Ref. [5].



Figure 3. Raman-Ramsey line. The Zeeman frequency is $f_Z = 15.8$ kHz, the contrast of the central fringe is C = 3.6%, the central fringe linewidth at half-maximum level is $\Delta v = 240$ Hz, $t_p = 3.5$ ms, T = 0.42 ms, and $t_{det} = 0.02$ ms.

Since the light intensity did not change in our experiments, the optimisation procedure was to vary the duration T of the interval between pumping and detection pulses and to study the dependence of the Raman-Ramsey line central fringe linewidth and contrast on T. Figures 4 and 5 show the corresponding dependences. At a given light intensity, the Raman-Ramsey line with the linewidth of 110–240 Hz can be achieved. One can see in Figs 4 and 5 that the narrowing of the lines is accompanied by a decrease in their contrast. The contrast value decreases with increasing time T between pulses. Figure 6 shows the dependence of the ratio of Δv to the contrast *C* for the central fringe of the Raman–Ramsey line on the pumping pulse duration. It is interesting that at T =0.42 ms, $t_{det} = 0.02$ ms and the given light intensity, this ratio has an optimal minimum value. This fact is very important because the minimum value of the ratio $\Delta v/C$ provides the condition under which the photon shot noise associated with the process of optical detection of the clock signal makes the smallest contribution to the overall instability of frequency measurements in the pulsed regime, expressed as the Allan variation

$$\sigma_{y}(\tau) = \frac{1}{\pi} \frac{1}{Q_{\text{line}}} \frac{1}{\text{S/N}} \sqrt{\frac{T_{\text{c}}}{\tau}}, \qquad (1)$$

where T_c is the duration of the cycle; τ is the averaging time; and $Q_{\text{line}} = v/\Delta v$ is the Q factor of the atomic line. In the limit of photon shot noise, the SNR is proportional to the contrast C of the central fringe of the Raman-Ramsey line and the square root of the radiation power P_0 at the output of the cell [2,7]: SNR = $C\sqrt{\mu_q N_{\text{det}}} \sim C\sqrt{P_0}$. Here μ_q is the quantum efficiency of the photodetector and N_{det} is the number of detected photons. According to Ref. [1] and the present work, with the optimal parameters found ($t_p = 3.5 \text{ ms}$, T = 0.42 ms, $t_{\text{det}} = 0.02 \text{ ms}$), approximately equal values of the Q factors of



Figure 4. Dependence of the Raman–Ramsey central fringe linewidth Δv on the duration of the interval *T* at $t_p = 3.5$ ms, $t_{det} = 0.02$ ms.



Figure 5. Dependence of the Raman-Ramsey central fringe contrast on the duration of the interval T at $t_p = 3.5$ ms, $t_{det} = 0.02$ ms.



Figure 6. Dependence of the ratio $\Delta v/C$ for the Raman-Ramsey line central fringe on the pumping pulse duration t_p at T = 0.42 ms, $t_{det} = 0.02$ ms.

atomic lines and cycle durations, as well as with contrast of resonances differing by an order of magnitude, the SNR and instability of frequency measurements in the shot noise limit of the photodetection for resonant lines will be 30000 and 1000, $\sigma_y(\tau) \sim 2 \times 10^{-14}/\sqrt{\tau}$ and $\sigma_y(\tau) \sim 6 \times 10^{-13}/\sqrt{\tau}$, respectively.

The clock CPT resonance formed in the experimental lin||lin CPT configuration by magnetic field sensitive transitions is a superposition of two resonances. The dependence of their frequency on the magnetic field can be obtained by the Taylor expansion of the Breit–Rabi formula for small values of magnetic field induction:

$$v_{1,2} = \Delta_{\rm hfs} \pm 2g_I \mu_{\rm B} B + \frac{3}{8} \frac{(g_J - g_I)^2}{\Delta_{\rm hfs}} \mu_{\rm B}^2 B^2, \qquad (2)$$

where $\Delta_{\rm hfs}$ is the frequency of hyperfine splitting of the ground state in the absence of the magnetic field; g_I and g_J are the Landé g-factors; $\mu_{\rm B}$ is the Bohr magneton; B is the magnetic field induction; and subscripts 1 and 2 denote the transitions $|F_{\rm g} = 1, m = 1\rangle \leftrightarrow |F_{\rm g} = 2, m = -1\rangle$ and $|F_{\rm g} = 1, m = -1\rangle \leftrightarrow |F_{\rm g} = 2, m = 1\rangle$, respectively. The frequency shift of the clock transition due to the quadratic Zeeman effect is $(3/8)[(g_J - g_I)^2/\Delta_{\rm hfs}]\mu_{\rm B}^2B^2$. The coefficient at B^2 is 1.33 times smaller than the coefficient for the transition $m = 0 \leftrightarrow m = 0$ and is equal to 575.14×10^8 Hz T⁻².

In accordance with Eqn (2), the frequency difference between resonances $|F_g = 1, m = 1\rangle \leftrightarrow |F_g = 2, m = -1\rangle$ and $|F_g = 1, m = -1\rangle \leftrightarrow |F_g = 2, m = 1\rangle$ should linearly depend on the magnitude of the magnetic field. However, the dependence of the linewidth of the line formed by the superposition of these resonances on the magnetic field will be nonlinear, since we define the linewidth Δv as a total width (at half-maximum level) of superimposed lines of two magnetic field sensitive resonances, whose frequencies are symmetrically shifted by 2.8 kHz Gs⁻¹ from the hyperfine splitting frequency Δ_{hfs} [9]. Figure 7 shows the measured dependence of the linewidth Δv of the Raman–Ramsey line central fringe on f_Z at the optimal parameters $t_p = 3.5$ ms, T = 0.42 ms, $t_{det} = 0.02$ ms. It can be seen that only in a small range of Zeeman frequencies (up to slightly more than 20 kHz) the linewidth has a weak magnetic field sensitivity.

Figures 8 and 9 show, for clarity, the central fringes of the Raman–Ramsey lines at $f_Z = 13.31$ and 73.81 kHz. At $f_Z =$



Figure 7. Dependence of Δv on the Zeeman frequency f_Z at $t_p = 3.5$ ms, T = 0.42 ms, $t_{det} = 0.02$ ms.

73.81 kHz, the splitting of the central fringe is clearly observed – a phenomenon that was previously reported in the case of cw excitation of CPT resonances in the lin||lin configuration [9].



Figure 8. Raman–Ramsey line with the central fringe linewidth $\Delta v = 240$ Hz and the contrast C = 3.8% at $f_Z = 13.31$ kHz, $t_p = 3.5$ ms, T = 0.42 ms, $t_{det} = 0.02$ ms.



Figure 9. Raman–Ramsey line with the central fringe linewidth $\Delta v = 520$ Hz and contrast C = 1.7% at $f_Z = 73.81$ kHz, $t_p = 3.5$ ms, T = 0.42 ms, $t_{det} = 0.02$ ms.

3. Conclusions

The paper presents the results of the study of QFS's with pulsed excitation of coherent population trapping resonances on the D₁ lines of the ⁸⁷Rb atom using the Raman–Ramsey method with the same linear polarisations of the bichromatic laser radiation components. This method of obtaining narrow resonances is an alternative to the method of pulsed optical pumping, recently implemented in Ref. [1] in the same rubidium cell. The process of optimisation of the central resonance fringe linewidth, its contrast, and linewidth-to-contrast ratio is described. The optimal minimum value of this ratio is found, which determines the condition under which the photon shot noise makes the smallest contribution to the overall instability of frequency measurements in the pulsed regime. The Raman-Ramsey central fringe linewidth dependence on the magnetic field is investigated, and a range of magnetic field values is found, where this dependence is weak.

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