

Frequency stabilization of a diode laser on the $5P \rightarrow 5D$ transition of the Rb atom

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Received 16 November 2016, revised 25 February 2017

Accepted for publication 17 March 2017

Published 12 April 2017



Abstract

In this work, we investigated the frequency stabilization method of a diode laser that operates on the excited state transition $5P \rightarrow 5D$ of Rb atoms. The described technique allows simple control of the diode laser frequency stabilization to be performed. The critical parameters of stabilization, such as laser intensity and the temperature of a reference cell, have been investigated and long-time laser stability has been demonstrated with frequency stability of about 1 MHz.

Keywords: laser stabilization, rubidium atom, laser spectroscopy

(Some figures may appear in colour only in the online journal)

1. Introduction

Elements of alkali metals are some of the most used, both in basic research on laser spectroscopy [1] and atom optics [2] and in numerous practical applications [3, 4]. The development of the Doppler-free laser spectroscopy method [5] and, based on it, laser frequency stabilization, has made it possible to use alkali metal atoms in a vast number of experiments on the quantum control of atoms [6], nuclear magnetic resonance (NMR) experiments [7], coherent chemistry [8], quantum information processing [9] and quantum metrology [10], as well as in practical applications, such as quantum frequency standards [11], atomic rotation sensors [12], and local gravity sensors [13].

Generally, the application of alkali metals in modern experiments is based, as a rule, on the use of the first excited state (in the case of the rubidium atom, this is the $5S \rightarrow 5P$ transition) [14, 15]. But in a variety of experiments not only the ground and first excited atomic states are used but also higher-lying, primarily, D-states of atoms of alkali metals are involved (for example, in the case of rubidium, this is the $5P \rightarrow 5D$ transition). Thus, the second excited state of alkali atoms is used in experiments on studying Rydberg states [16] and building on quantum memory on their basis [17], single photon pair generation [18], in studies of the polarizability of

atomic states [19] aiming at the construction of exact models of the intra-atomic dynamics [20], as well as in experiments on photon transport by single atoms [21].

In order to operate with the excited atomic states, it is necessary to implement high frequency stability of the laser radiation at a corresponding transition. In the case of the use of the first excited state of alkali metal atoms, approaches to the frequency stabilization of lasers have been developed [22–25], which make it possible to stabilize the laser frequency in a wide range around the frequency of the atomic transition. There are also approaches to the frequency stabilization of lasers with the use of highly excited energy levels of atoms, but particular schemes of their realization either have not been described, or processes that affect the degree of stabilization of the laser have not been investigated.

This work is devoted to the experimental investigation of frequency stabilization of the diode laser operating at the wavelength of the $5P_{3/2} \rightarrow 5D_{5/2}$ transition of the rubidium atom. This transition makes it possible to excite rubidium atoms to the 5D state, and it is widely used in experimental investigations in recent times [16–18, 21]. The stabilization scheme of the laser considered in the paper is based on the absorption spectroscopy of the laser radiation in an atomic cell. We investigate the basic parameters that affect the frequency stabilization of the laser and present the main

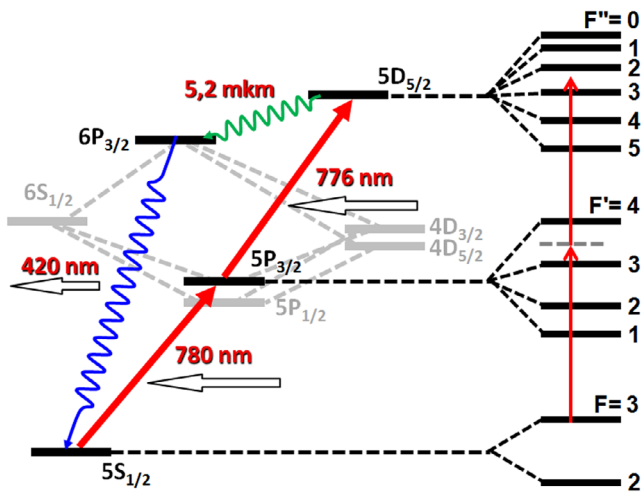


Figure 1. Scheme of the ^{85}Rb atom's energy levels with hyperfine splitting of the ground and the first two excited states. The red arrows show transitions for the excitation of the atom to the $5D_{5/2}$ state; the green arrow shows the decay of the $5D_{5/2}$ state to the $6P_{3/2}$ state, from which the atom returns to the ground state emitting a blue photon at a wavelength of 420 nm.

technological approaches that ensure stabilization of the laser without involving expensive equipment.

2. The structure of atomic levels

In order to describe the method of frequency stabilization of the diode laser at the $5P \rightarrow 5D$ transition of the rubidium atom, we will consider the energy level structure of this atom, which is presented on figure 1. The ground state of the rubidium atom is the $5S_{1/2}$ level. The transition to the first excited state, $5P_{3/2}$ (which corresponds to the D2 spectral line of the atom) is the strongest one. Precisely this transition is actively used in many experiments with alkali atoms. The wavelength of this transition is 780 nm. Its spectral width is about 6 MHz. Upon subsequent excitation of the atom by radiation at a wavelength of 776 nm, the atom passes to the second excited state, $5D_{5/2}$. The linewidth that corresponds to the $5P_{3/2} \rightarrow 5D_{5/2}$ transition is about 1 MHz [26]. This state can be used for further excitation of nP and nF Rydberg states of the rubidium atom.

Figure 1 also represented the hyperfine structure of the atomic states of ^{85}Rb . The hyperfine structure of the ground and the first excited states are well known. The crossover resonance $^{85}\text{Rb } F = 3 \rightarrow F' = 2, 4$ is used to stabilize the laser frequency for the first step excitation as described below. The hyperfine splitting of the $5D_{5/2}$ state is much smaller in comparison with $5P_{3/2}$ (from 9, 4 MHz to 2, 9 MHz for $5D_{5/2}$ state in comparison with several tens of MHz for the $5P_{3/2}$ state). Frequently only transitions to the states with $F'' = 5, 4, 3, 2$ can be resolved in experiment [26]. The strongest transition is $5P_{3/2}(F' = 4) \rightarrow 5D_{5/2}(F'' = 5)$.

Rb atoms can be excited to the $5D$ state by two methods: (i) via the two-photon excitation using the laser at a wavelength of 778 nm and (ii) by the two-step excitation via $5S \rightarrow 5P \rightarrow 5D$ transitions using two lasers. In the first method, there is no direct excitation of the intermediate $5P$ state. The

second method involves a considerable excitation of atoms to the $5P$ state. In this work, we used the second method, which makes it possible to create high concentrations of atoms in the $5D$ excited state. Theoretical calculations show that it is possible to excite up to 50% of atoms to the $5D$ state [27]. This technique is widely used for efficient frequency up-conversion in rubidium vapor [28, 29].

In the case of rubidium and two-step excitation with counterpropagating laser beams, the resonance wavelengths of transitions (780 and 776 nm) are close. It makes it possible to partially suppress the Doppler effect and to use almost all of the atoms of the velocity distribution upon their excitation by narrowband laser radiation [26, 30, 31]. This ensures obtaining resonances free from the Doppler broadening in the first order even if atomic vapors are used.

Let us consider in more detail the relaxation dynamics of the atom from the $5D_{5/2}$ state. There are two relaxation channels of the atom from the $5D_{5/2}$ state: $5D_{5/2} \rightarrow 5P_{3/2}$ (with a relative probability of 65%) and $5D_{5/2} \rightarrow 6P_{3/2}$ (with a relative probability of 35%) [32]. In the first case, the relaxation to the $5S$ ground state proceeds via the $5P$ state. In the second case, the relaxation process involves a larger number of levels and the $5D \rightarrow 6P \rightarrow 5S$ relaxation channel is the most interesting for practical applications. In this case, a photon at a wavelength of 420 nm is emitted (as a result of the $6P \rightarrow 5S$ transition). The emission of this photon can be used to detect the efficiency of the excitation process of atoms to the $5D$ state [30, 33], as well as to increase the detection sensitivity down to single atoms.

3. Stabilization schemes at the wavelength of the $5P \rightarrow 5D$ transition

There are several approaches to the stabilization of the laser at the wavelength of 776 nm, which corresponds to the $5P \rightarrow 5D$ transition. One of the simplest approaches is based on the use of a high-finesse and a highly stable Fabry–Perot interferometer in combination with a wavelength meter. In this scheme, the laser radiation is stabilized with respect to the transmission peak of the Fabry–Perot interferometer [34], and the stabilization frequency can be chosen arbitrarily. The wavelength meter is used to determine the wavelength of the laser radiation. The main problem of this scheme is the frequency drift of the Fabry–Perot interferometer, which restricts the stabilization time of the laser.

Another approach is based on the use of resonant absorption lines of the atom. Because the frequency of any atomic transition is stable in time under the constant external conditions, this approach does not have fundamental physical restrictions on the long-term stability of the laser. In this approach to the frequency stabilization of the laser, the characteristic stabilization time is determined only by the drift of electronic components of the laser and stabilization system.

The frequency stabilization of a laser on an atomic transition $5P \rightarrow 5D$ can be implemented by different methods. Three main approaches can be emphasized, which are based on the following physical processes: (1) the electromagnetically

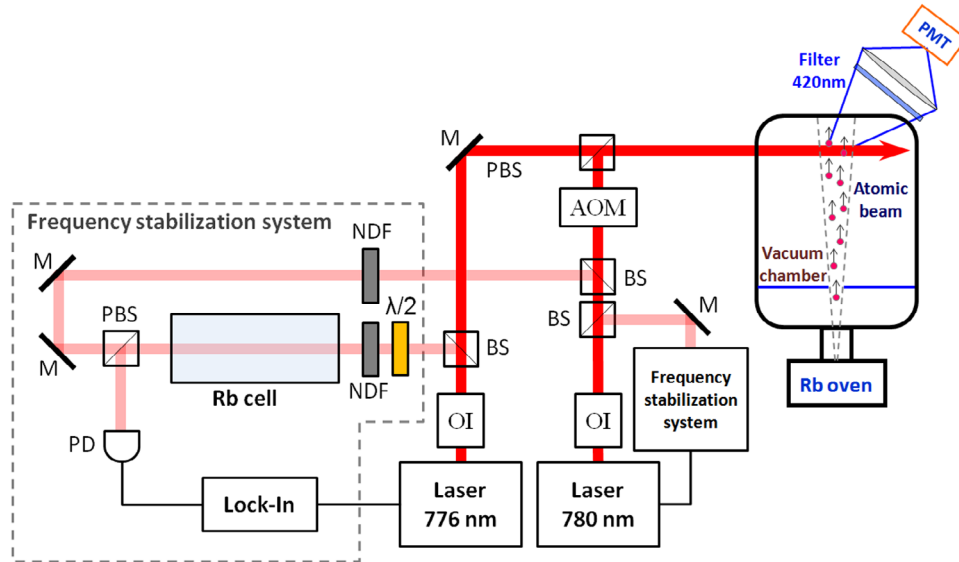


Figure 2. Schematic of the frequency stabilization system of a diode laser operating on the transition $5P \rightarrow 5D$ of Rb atom: (1) filter (NDF), (2) $\lambda/2$ plate, (3) beam-splitting cube (BS), (4) mirror (M), (5) photodiode (PD), (6) polarizing cube (PBS), (7) cell with Rb vapor, (8) optical isolator (OI), and (9) acousto-optic modulator (AOM). The experimental scheme of the investigation of laser frequency stability using co-propagating laser beam intersection with atomic beam is shown on the right.

induced transparency (EIT) [35] of the first atomic transition ($5S \rightarrow 5P$); (2) the fluorescence due to the decay of the second excited state ($5D$); (3) the absorption of radiation on the second transition ($5P \rightarrow 5D$). All three methods utilize multi step excitation scheme $5S \rightarrow 5P \rightarrow 5D$. Frequently, the laser for the first step of excitation is stabilized on the first transition ($5S \rightarrow 5P$) by using standard methods based on the saturated absorption spectroscopy with implementation of lock-in technique [36] or stabilisation on the slope of the nonlinear absorption resonances [37]. It should be mentioned that in the considered schemes the frequencies of both lasers are stabilized separately or the sum of the frequencies is stabilized. We do not consider here the techniques of the stabilization using polarization effects like in [38].

With the stabilization of the laser by the first method (using the effect EIT) [39] the absorption of laser radiation is detected at a wavelength of 780 nm, corresponding to an atomic transition $5S \rightarrow 5P$. The EIT effect is as follows. The absorption due to the first atomic transition ($5S \rightarrow 5P$, 780 nm) is cancelled if there is an intense radiation at the second transition frequency ($5P \rightarrow 5D$, 776 nm). That is the atomic cell, which was initially opaque at the wavelength of 780 nm, becomes almost transparent. The resonant character of the absorption process on the wavelength of 776 nm makes it possible to realize the frequency stabilization. The drawback of this approach is the need to use intense laser radiation up to 40 mW cm^{-2} at the wavelength of 776 nm in order to realize the EIT process. This requirement is not always convenient in real experiments because, as a rule, one tends to use an amount of the laser power as low as possible for the stabilization system and to use most of the power in the main experiment.

In the stabilization method of the laser at the frequency of the $5P \rightarrow 5D$ transition using the fluorescence at the wavelength of 420 nm [40], atoms are excited using the bichromatic laser field: (1) one of the lasers is stabilized on the

frequency of $5S \rightarrow 5P$ transition and (2) the second laser is stabilized near the resonance of the $5P \rightarrow 5D$ transition. In this case, the fluorescence signal is proportional to the population of the $5D$ state, which is maximal under the condition of exact frequency resonance of the laser to be stabilized.

The advantage of this scheme is the possibility of filtering the fluorescence at the wavelength of 420 nm from the scattered light of the exciting lasers. Apart from this, the realization of this scheme requires considerably lower intensities (about 6 mW cm^{-2} for the 780 nm laser and 1 mW cm^{-2} for the 776 nm laser) compared to the approach based on the use of the EIT effect. However, this scheme is also not free from drawbacks. The main drawback is the low quantum emission efficiency at the $5D \rightarrow 6P \rightarrow 5S$ decay channel: only 2% of atoms emit photons at the wavelength of 420 nm. This leads to the need to use photomultipliers for the atom detection at this wavelength, because highly sensitive solid-state radiation detectors in the wavelength range around 420 nm are unavailable. This complicates the functioning of the experimental setup. In this work, we experimentally demonstrate the use conventional photodiodes to detect absorption signals at the wavelength of the $5P \rightarrow 5D$ transition.

4. Experimental setup

The experimental scheme of frequency stabilization of a diode laser on the $5P \rightarrow 5D$ transition of the Rb atom is presented on figure 2. The scheme is based on the two-step excitation of Rb atoms by two counterpropagating laser beams. For its realization, we used two laser systems, a cell with vapor of Rb atoms, photodiodes, and accessory optical elements (figure 2). Rubidium vapor cell (6 cm long) was made of sapphire with YAG windows to prevent chemical reaction of Rb with them under the temperatures higher 100°C .

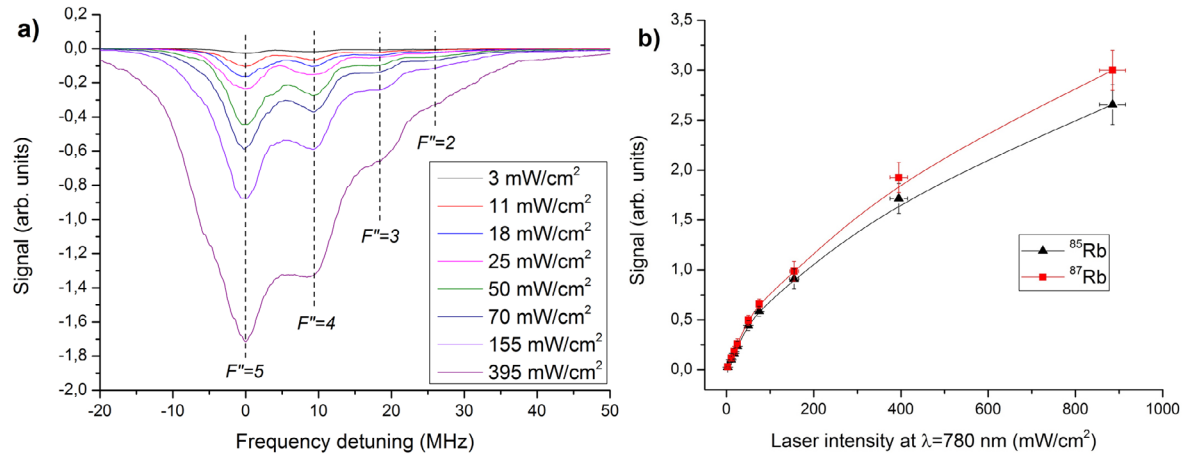


Figure 3. (a) Absorption spectra of ^{85}Rb atoms at the wavelength of 776 nm with resolved hyperfine structure of $5D_{5/2}$ state at the different intensities of the first-step exciting laser (780 nm); (b) dependence of the absorption signal on the laser intensity at 780 nm.

For the excitation of the first transition of the Rb atom, we used a laser system based on a DL 100 diode laser (Toptica Photonics), which was stabilized on the ^{85}Rb $5S_{1/2} \rightarrow 5P_{3/2}$ transition (wavelength 780 nm). The laser system included DCC-110 laser diode current control unit, DTC-110 diode temperature control unit, SC-110 frequency scanning electronic module, and LIR-110 lock-in amplifier. The frequency stabilization of this laser was realized based on the traditional saturated absorption spectroscopy on $F = 3 \rightarrow F' = 2, 4$ crossover resonance with the 92-MHz red frequency detuning from the $F = 3 \rightarrow F' = 4$ transition. The frequency detuning is necessary for the measurement of the laser frequency stability which was performed by using the fluorescence from atomic beam (see below). A small part of the laser radiation from the main laser beam was used in the stabilization system. The diameter of a beam was about 1.2 mm. The frequency stability of the laser for the first step of excitation was about 1 MHz.

A laser system to be stabilized is an external-cavity diode laser (VitaWave ECDL-7810R). The laser system includes a laser control unit and a lock-in amplifier. As a laser diode, DL-7140-201 diode (Sanyo Inc.) is used. The diameter of a laser beam was about 0.9 mm.

5. Experimental results

5.1. Frequency stabilization by the absorption in an atomic cell

In the laser frequency stabilization by an absorption on the $5P_{3/2} \rightarrow 5D_{5/2}$ transition, the following parameters are critical: (1) the laser intensities at the first and second atomic transitions, and (2) the temperature (correspondingly, the pressure) of atomic vapor. The laser intensity affects the value of the laser light absorption due to the second transition ($5P_{3/2} \rightarrow 5D_{5/2}$). Thus, it was shown theoretically in [27] that, at high intensities of the laser radiation, the population of the $5D_{5/2}$ state can reach 50%. However, at high laser intensity the atomic transition becomes saturated and its linewidth becomes broadened, which lowers the accuracy of the laser frequency stabilization.

The density of atomic vapor affects the value of the laser radiation absorption and should be maximal to create high optical densities. However, our experimental measurements showed that, upon an increase in the density of atomic vapor, the absorption signal caused by the $5P_{3/2} \rightarrow 5D_{5/2}$ transition increases only to a certain level, after which it begins to decrease. This effect is related to the contribution of collisional processes to the dynamics of population and relaxation of excited levels.

We determined optimal values of the temperature of the Rb vapor cell and the intensities of the lasers at the wavelength of the $5S_{1/2} \rightarrow 5P_{3/2}$ transition (780 nm) and the wavelength of the $5P_{3/2} \rightarrow 5D_{5/2}$ transition (776 nm). As was mentioned above, these parameters affect the value of the absorption of the stabilized laser radiation in the cell, and, consequently, the accuracy of the frequency stabilization system.

The laser frequency was stabilized using the peak of the atomic transition resonance. The feedback signal was formed by means of the frequency modulation of the laser to be stabilized in the lock-in amplifier mode. The long-term stability of the laser was measured via the fluorescence from the atomic beam of Rb atoms in the vacuum chamber.

Figure 3(a) presents the absorption spectra of Rb atoms at the wavelength of 776 nm for different intensities of the laser operating on the first transition (780 nm). The hyperfine structure of the $5D_{5/2}$ state of ^{85}Rb is resolved in agreement with other publications [26]. The laser intensity increase of the first excitation stage leads to the growth of the absorption signal of the 776 nm laser radiation. But under the high intensities of such radiation the hyperfine structure of the $5D_{5/2}$ state is unresolved because of power broadening. Figure 3(b) shows the dependence of the amplitude of the absorption signal at 776 nm in relation to the intensity of the laser operating at the wavelength of 780 nm. It is seen from this figure that, at intensities of up to 50 mW cm^{-2} , the absorption regime is linear.

The influence of the laser intensity to be stabilized was investigated similarly. The dependence of the absorption of the laser radiation in vapor of Rb atoms on the intensity at the wavelength of 776 nm is presented in figure 4 (laser intensity of the radiation at the wavelength of 780 nm was

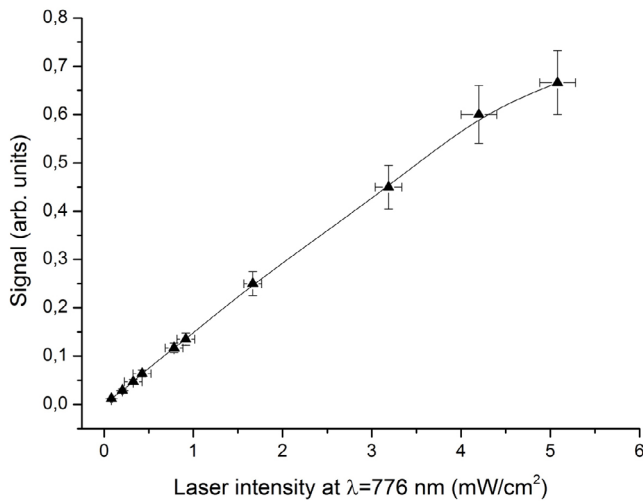


Figure 4. Dependence of the absorption of the laser radiation in vapor of Rb atoms on the laser intensity at the wavelength of 776 nm.

52 mW cm⁻²). It is seen that, in the whole intensity range, the absorption amplitude is proportional to the intensity of the laser, and the absorption coefficient remains constant.

The influence of the temperature of the Rb vapor cell on the absorption signal is presented in figure 5. With an increase in the temperature, the vapor pressure in the cell increases, and the absorption also increases, since it is proportional to the concentration of Rb atoms. This increase continues up to the cell temperature of 60 °C, which corresponds to the vapor pressure of 1×10^5 Torr and the concentration of rubidium atoms of 3×10^{11} cm³ [41]. The further increase in the temperature and, correspondingly, in the vapor pressure leads to an increase in the influence of atomic collisions processes [42]. The main contribution to the decrease in the absorption is made by the process of binary collisions of Rb atoms excited to the $5P_{3/2}$ state. In this process, which is referred to as the energy pooling collisions, the excitation energy is transferred from one atom to the other [42–46]. Due to this process, one of the two collided excited atoms passes to the $5S_{1/2}$ ground state, while the other atom passes to the $5D$ state. As a result, the population of the $5P_{3/2}$ level is depleted. A decrease in the population of the $5P_{3/2}$ level leads to a fall in the absorption at the $5P_{3/2} \rightarrow 5D_{5/2}$ transition of interest. It is seen from figure 5 that, for the stabilization of the laser on the $5P_{3/2} \rightarrow 5D_{5/2}$ transition, the optimal temperature of atoms in the cell is 60 °C.

5.2. The laser frequency stability

The laser frequency stability was investigated on the basis of the fluorescence measurement from Rb atomic beam at wavelength 420 nm (the $6P_{3/2} \rightarrow 5S_{1/2}$ transition). The atomic beam was formed in a vacuum chamber using several diaphragms. The co-propagating laser beams at the wavelength of 776 nm and at the wavelength of 780 nm crossed the atomic beam at an angle of 85°. The fluorescence signal at the wavelength of 420 nm was filtered and detected with a photomultiplier (figure 2).

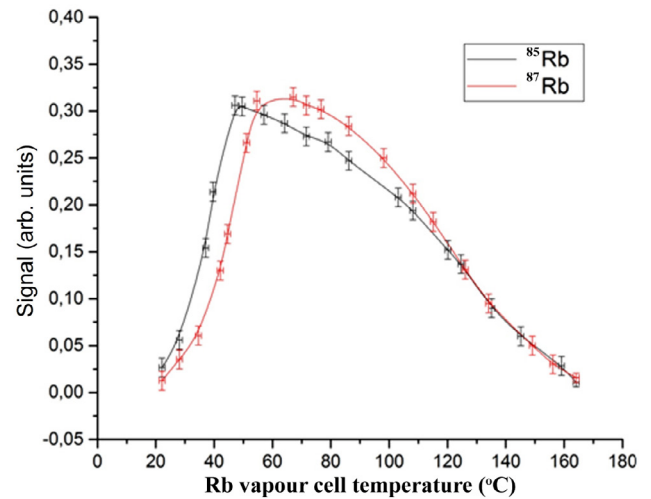


Figure 5. Dependence of the absorption amplitude on the temperature of a cell with rubidium vapor.

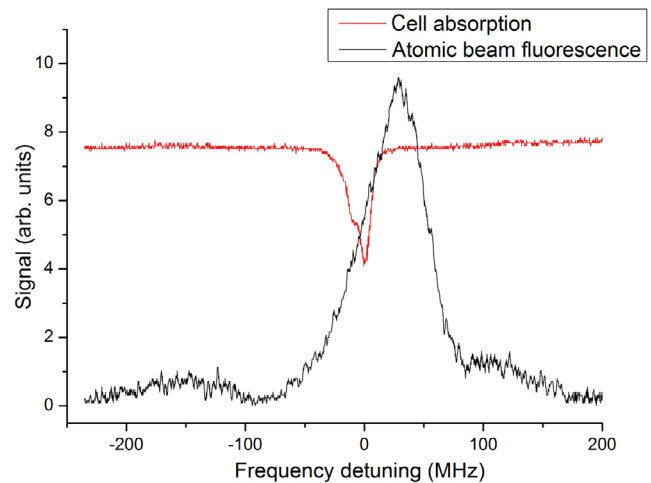


Figure 6. The absorption spectrum of Rb atoms in a cell ($5P_{3/2} \rightarrow 5D_{5/2}$, 776 nm) and the fluorescence spectrum of Rb atoms in a beam ($6P_{3/2} \rightarrow 5S_{1/2}$, 420 nm).

The laser at the wavelength of 780 nm was stabilized at the $F = 3 \rightarrow F' = 3, 4$ crossover resonance of ⁸⁵Rb atoms. Its laser beam was directed to the vacuum chamber via an acousto-optic modulator, which made it possible to perform the amplitude modulation of the laser radiation for the implementation of the lock-in amplifier of the fluorescence signal.

Figure 6 presents the absorption spectrum of Rb atoms in a cell at the $5P_{3/2} \rightarrow 5D_{5/2}$ transition (776 nm) and the fluorescence spectrum of Rb atomic beam due to $6P_{3/2} \rightarrow 5S_{1/2}$ transition (420 nm). The acousto-optic modulation of the laser radiation and the oblique excitation of the atomic beam cause a shift by ~ 20 MHz of the fluorescence peak with respect to the peak at which the stabilization of the radiation at the wavelength of 776 nm is performed. This makes it possible to measure the frequency drift of the laser to be stabilized via the fluorescence using the dependence presented in figure 6 as a reference.

The measurements of the laser frequency stability at the wavelength of 776 nm are presented in figure 7. The

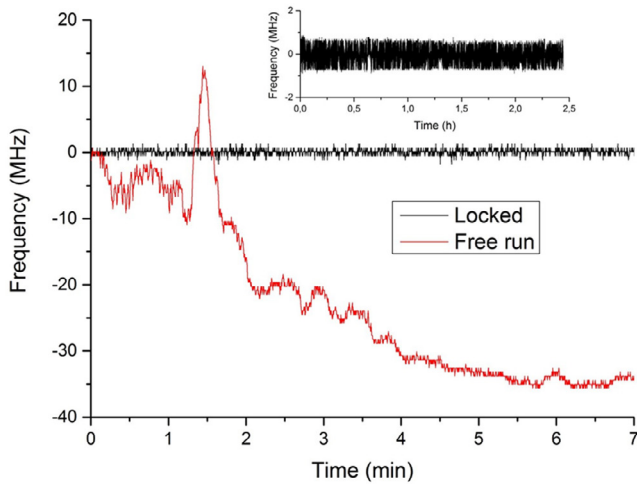


Figure 7. The measurements of the laser frequency stability at the wavelength of 776 nm. The fluorescence signals caused by the $6P_{3/2} \rightarrow 5P_{1/2}$ transition (420 nm) in a beam of Rb atoms for stabilized and unstabilized lasers are shown.

measurements were performed for the stabilized and unstabilized lasers. It is seen from the plots that, without stabilization, the laser goes out of resonance with the atomic beam in a time of about 5 min (frequency drift about 100 MHz). The stabilized laser retains its frequency for a few hours. The frequency stability of the laser is ~ 1 MHz, which is comparable with the stabilization accuracy of the laser on the first transition (780 nm).

6. Conclusion

In this work, we investigated the frequency stabilization method of a diode laser that operates on the excited state transition $5P \rightarrow 5D$ of Rb atoms. The $5D$ state is the excited state of the Rb atom, and it is frequently used in various experiments in atom physics. The described technique allows fast and simple control of the diode laser frequency stabilization to be performed.

Acknowledgments

This work was supported by Russian Science Foundation (Contract No. 14-12-00729).

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