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High-precision laser spectroscopy of cold atoms and the search for the drift of the fine structure constant

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1. Introduction

This review presents the main scientific results obtained over the last several years at the Laboratory of Active Media Optics of the Optical department of the Lebedev Physical Institute. The work was aimed at the search for and the investigation of high-finesse optical resonances in atomic ensembles. This allows carrying out sensitive tests of fundamental physical theories and opens the possibility of creating prospective frequency references in the optical range. A new laboratory method has been presented to search for the drift of the fine structure constant by using a frequency comb of a femtosecond laser, and sensitive experiments are being performed on testing the quantum electrodynamic theory. Work has started on laser cooling of the thulium atom, which experiences a narrow transition near 1.14 μm. The possibility of cooling was analyzed experimentally and theoretically, and the transitions most promising for the cooling were determined. A new generation of ultrastable optical cavities was developed for stabilizing the frequency of laser systems, which allows detecting optical resonances with a sub-Hertz resolution. A compact magneto-optical trap for rubidium atoms was created, and the interaction of femtosecond-laser radiation with a laser-cooled ensemble of atoms was investigated.

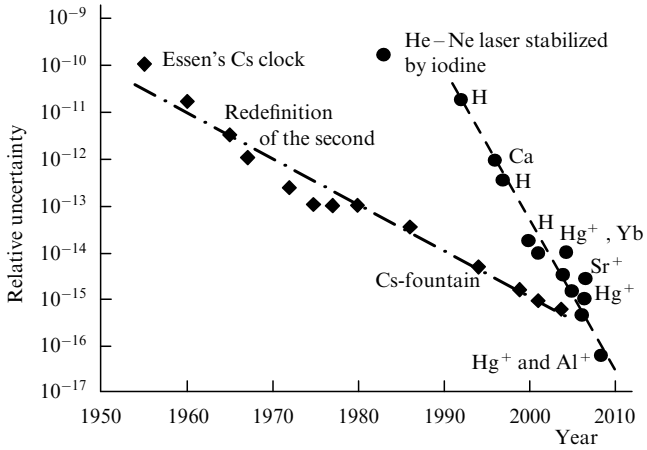

Figure 1. Uncertainty evolution for microwave (rhombs, dash-dotted line) and optical (circles, dashed line) frequency references. Noticeable progress in the development of optical standards is related to the evolution of new ultrastable laser systems, methods for measuring and comparing optical frequencies by means of an optical comb, and the development of new spectroscopic objects based on captured and cooled atoms and ions.

High-precision laser spectroscopy and laser cooling of atomic ensembles are rapidly developing fields of modern physics. In the last decade, the most impressive achievements have been awarded Nobel prizes for physics, specifically, the development of methods for cooling and trapping atoms by laser radiation (1997), the experimental discovery of Bose – Einstein condensation in dilute gases of alkali metals (2001), and the contribution to the development of methods for high-precision spectroscopy and the creation of a frequency comb on the basis of a femtosecond laser (2005) [1]. Intense investigations in the field of high-precision spectroscopy and metrology started about 30 years ago [2, 3]; however, it took a long time to approach the measurement uncertainty of $10^{-17} - 10^{-18}$, envisaged in pioneering works by A L Shavlov, V P Chebotaev, V S Letokhov, T W Hänsch, J L Hall, and other classics of nonlinear laser spectroscopy. As the result of long-term work, scientists from metrological and laser centers in the USA, Germany, Russia, France, England, and other countries succeeded in reducing the relative error of optical frequency references to 2×10^{-17} [4], which is by an order of magnitude better than the accuracy of the best primary standards, namely, cesium fountains [5]. In Fig. 1a, the relative errors of microwave and optical standards are compared.

Rapid progress in optical standards is mainly related to the development of simple-to-use laser systems with superior characteristics, which satisfy the most stringent requirements of experimenters. Laser systems are used for cooling and capturing atoms and ions in traps, for preparing their internal states, and in the spectroscopy of ultra-narrow 'clock'

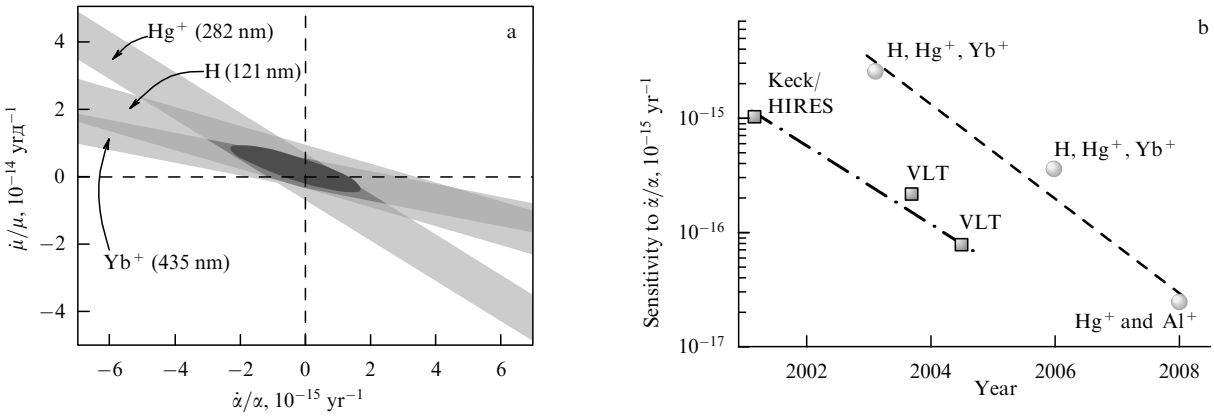

Figure 2. (a) Model-independent method for the search for a possible drift of fundamental constants based on a combined analysis of independent results on measuring the frequencies of absolute transitions in atoms and ions. The ellipse denotes the confidence interval (1σ) for the relative drifts of the fine structure constant $\dot{\alpha}/\alpha$ and the reduced magnetic moments $\dot{\mu}/\mu$ of the ^{133}Cs nucleus. (b) Comparison of the sensitivities of astrophysical (squares, dash-dotted line) and laboratory (circles, dashed line) methods to a possible linear drift of the fine structure constant.

transitions in single ions (see, e.g., [6, 7]), ensembles of laser-cooled neutral atoms [8, 9], and atomic beams [10, 11]. Principally new schemes are being developed for cooling, capturing, and spectroscopic measurements of atoms [4, 12, 13], which allow carrying out investigations at minimal influence of external perturbations on clock transitions. In turn, the use of a comb of optical frequencies based on the femtosecond laser with passive mode locking [14] opens the possibility of directly comparing reference frequencies in the RF and optical ranges, linking them by a simple and universal 'bridge.'

Such high-precision measurements and frequency comparison in the optical range not only are an important frontier in modern metrology but also open new possibilities for performing sensitive tests of fundamental theories. A relevant problem in modern physics is the question of the temporal stability of fundamental constants, in particular, the fine structure constant α. More than 70 years ago, Dirac [15] posed the question on the stability of α, which touches on the fundamentals of physical theories. The question has no answer yet, although the investigations in this field continue. A number of theories allow a possible drift of the coupling constant for electromagnetic interactions (see, e.g., reviews [16–18]), but the problem concerning the search for the drift resides in the experimental domain because no theory predicts the drift value.

In 2003, we measured the absolute frequency of the clock 1S–2S transition in the hydrogen atom (H) with the accuracy up to the 14th decimal place [19]. On this basis, a new laboratory model-independent method was developed for determining the possible drift of α, which is schematically shown in Fig. 2a [20]. The method suggests the experimental limitation on the drift of the frequencies of optical transitions measured relative to the frequency of a primary standard by means of a frequency comb. In addition, the different sensitivity of the transition frequencies to the parameter α due to relativistic effects [21] is taken into account. Data on the frequency of the 1S–2S transition in the hydrogen atom were used in a joint analysis with those from metrological centers in the USA (Hg^+ ion [22]) and Germany (Yb^+ ion [23]), which provided a bound on the drift of the fine structure constant at the level of $\dot{\alpha}/\alpha = (-0.9 \pm 2.9) \times 10^{-15}$ yr^{-1}. The sensitivity to the linear drift of α amounts to 3×10^{-15} yr^{-1} (the upper point in Fig. 2b), which is close to the accuracy of the analysis of quasar absorption spectra (under the assumption of a linear drift of α) performed by means of the complex Keck telescope/HIRES spectrometer (High Resolution Echelle Spectrometer) [24]. At the time, it was the strictest estimate obtained from the analysis of astrophysical data.

The results of the analysis in [24] indicated that about 10^{10} years ago, the parameter α differed from the modern value by $\Delta\alpha/\alpha = (-7.2 \pm 1.8) \times 10^{-6}$; this result stimulated further investigations in this area. In 2003–2004, the data obtained by the VLT (Very Large Telescope) were analyzed [25, 26], which, to the contrary, pointed to the invariability of α in the past relative to the present value, with the confidence level $|\Delta\alpha/\alpha| < 10^{-6}$. The collection of data detected from various astrophysical objects is inconsistent (for example, in Ref. [27], a nonzero drift is reported for the electron-to-proton mass ratio), which suggests that further investigations are necessary.

Nevertheless, the laboratory method developed in Ref. [20] based on the measurements of the absolute frequencies of clock transitions in various atomic systems by means of a femtosecond frequency comb allowed increasing the sensitivity to a linear drift by an order of magnitude already in 2005 (see Fig. 1b) [28, 29]. Presently, the drift of α is most strictly limited by a group from the National Institute of Standards and Technology (NIST) USA, who used a femtosecond comb to directly compare the frequencies of clock transitions in mercury and aluminum ions. It was found that the drift is bounded at the level $\dot{\alpha}/\alpha = (-1.6 \pm 2.3) \times 10^{-17}$ yr^{-1}, which is approximately an order of magnitude lower than the sensitivity of the best astrophysical tests [4]. A greater number of independent data, measurements of clock transitions in strongly relativistic systems, and an increased measuring duration have given the chance to enhance the sensitivity of laboratory methods to 10^{-18} yr^{-1} within the next few years and potentially to 10^{-20} yr^{-1}.

Some years ago, our group (Lebedev Physical Institute) suggested a new reference set of optical frequencies based on the dipole-forbidden transition at the wavelength 1.14 μm in the thulium atom. A potential finesse of this resonance reaches 2×10^{14}, and we therefore anticipate measuring the frequency in a cloud comprising 10^5–10^6 atoms with an

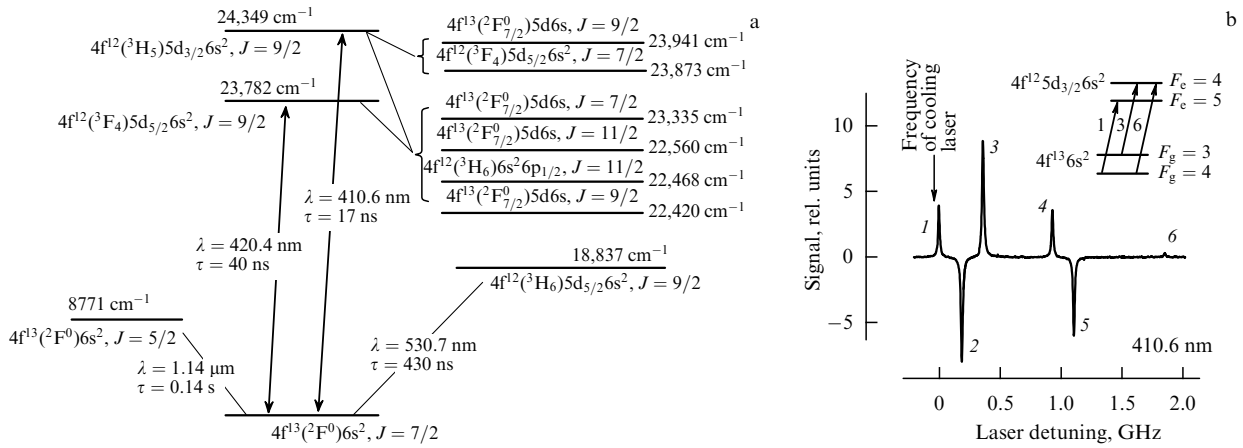

Figure 3. (a) Schematic diagram for thulium atom levels. (b) Experimentally recorded spectrum of saturated absorption at the wavelength $\lambda = 410.6$ nm. The required stabilized frequency for a cooling laser is shown.

accuracy comparable to that for standards on single ions and neutral atoms in optical lattices. Thus, we can obtain independent information on the bound on the drift of α by means of a high-relativistic thulium atom. Detection of resonances with a Hertz resolution requires a long-duration interaction of an atomic ensemble with laser radiation, which is only possible if atoms are cooled and localized. The spectroscopy of clock transition also requires the development of an ultrastable laser system. The thulium atom is widely used as a doping element in fiber and solid-state lasers, but the possibility of laser cooling and high-precision spectroscopy for this atom has not been considered so far.

In this review, we describe the strides towards solving the problem discussed. In Section 2, we investigate the possibility of laser cooling for the thulium atom using the results of sub-Doppler spectroscopy. In Section 3, the achievements of our group are presented on stabilizing laser systems by external reference cavities and some results are briefly described on using ultrastable laser systems in hydrogen atom spectroscopy. Section 4 is devoted to investigation results on the interaction of femtosecond radiation with an ensemble of laser-cooled rubidium-87 atoms in a magneto-optical trap.

2. Possibility of laser cooling for thulium

In the last decade, noticeable progress has been achieved in laser cooling of rare-earth elements. Compared to cooling by a buffer gas in a magneto-dipole trap demonstrated in Ref. [30], laser cooling exhibits lower temperatures, provides spectroscopy of narrow transitions almost free of external perturbations, and allows manipulation with separate atoms and groups of atoms. Despite the difficulty caused by the complicated structure of the spectra of lanthanides, scientific teams from the USA demonstrated laser cooling of ytterbium [31] and erbium [32] atoms in transitions residing in the blue spectral range (400 nm). The interest in rare-earth elements is explained by their prospective use in metrology and the study of ultra-narrow resonances [9], collision investigations at ultra-low temperatures and in Bose-condensates [33], and the possibility of implanting these atoms into solid substrates for solving problems in nanotechnology [34] and quantum informatics [35].

2.1 Study of cooling transitions

The thulium atom (Tm) has an electron vacancy in the 4f shell [the ground state configuration is $4f^{13}6s^2(^2F^0)$]. A natural isotopic mixture comprises only one isotope ^{169}Tm with the nuclear spin $I = 1/2$, which causes splitting of each electron level into two hyperfine-structure components. The ground state of the thulium atom comprises two fine-structure components (see Fig. 3a) with the quantum numbers $J_g = 7/2$ and $J'_g = 5/2$ of the total momentum of an electron shell with the splitting interval 2.6×10^{14} Hz ($\lambda = 1.14$ μm). Because the electron dipole transition between these sublevels is forbidden, it may be hoped that the $J'_g = 5/2$ state is long-lived (with the lifetime up to several tenths of a second). It is shown in [30, 36] that due to screening of the 4f shell by the external completed $6s^2$ shell, the influence of collisions on the frequency of the transition between the $J_g = 7/2$ and $J'_g = 5/2$ components of the ground state noticeably reduces.

The cooling of atoms is obligatory in creating a modern high-precision frequency reference because it eliminates the Doppler effect, provides a long period for their interaction with radiation, and allows the study of transitions virtually without external fields. For laser cooling of atoms, it is necessary to excite an intense cyclic transition that can be saturated by laser radiation. For the thulium atom, these may be dipole-allowed transitions from the sublevel ($J_g = 7/2$, $F = 4$) of the ground state $4f^{12}6s^2$ to the excited levels with ($J_e = 9/2$, $F = 5$), where F is the quantum number of the total momentum of the atom.

Among the excited levels of Tm, we can choose the following three candidates satisfying the criteria mentioned: $4f^{12}(^3H_6)5d_{5/2}6s^2$ (18,837 cm^{-1}), $4f^{12}(^3F_4)5d_{5/2}6s^2$ (23,782 cm^{-1}), and $4f^{12}(^3H_5)5d_{3/2}6s^2$ (24,349 cm^{-1}). The transition with the longest wavelength $\lambda = 530.7$ nm that can be excited by a second harmonic of the Nd:GSGG (Gadolinium Scandium Gallium Garnet) laser is totally cyclic; however, its relatively low probability $A = 2.3 \times 10^6$ s^{-1} prevents trap loading from atomic beams at room temperature. Nevertheless, this transition can be efficiently used with preliminarily cooled atoms. Two of the other transitions at the wavelengths 420.4 nm and 410.6 nm, which can be excited by a second harmonic of a Ti:sapphire laser, have noticeably greater probabilities (2.43×10^7 s^{-1} and 6.36×10^7 s^{-1} respectively [37]). This is sufficient for stopping atoms with velocities 200 m s^{-1} at a distance of

Table 1. Hyperfine splitting of the excited states in the thulium atom in our investigation [38]. Hyperfine splitting of the fundamental state is also presented.

Energy, cm^{-1}	Level	J	Splitting, MHz	References
0	$4f^{13}6s^2(^2F^0)$	7/2	−1496.550(1)	[39]
23,781.698	$4f^{12}(^3F_4)5d_{5/2}6s^2$	9/2	−1586.6(8)	[38]
23,873.207	$4f^{12}(^3F_4)5d_{5/2}6s^2$	7/2	+1411.0(7)	[38]
24,348.692	$4f^{12}(^3H_5)5d_{3/2}6s^2$	9/2	−1856.5(2,5)	[40]
			−1857.5(8)	[38]
24,418.018	$4f^{13}(^2F^0_{7/2})6s6p(^1P^0_1)$	5/2	−1969.4(1,3)	[38]

several tens of centimeters. The transitions are not fully cyclic, and there is a probability of the excited states decaying into the nearest levels of the opposite parity (see Fig. 3a).

We carried out a series of experiments and concluded that laser cooling of thulium atoms is possible.

The transitions with $\lambda = 420.4$ nm and $\lambda = 410.6$ nm were studied by means of saturation laser spectroscopy with counter-propagating beams of equal frequencies [38]. A sample spectrum is shown in Fig. 3b. The transitions were excited by the second harmonic of a Ti:sapphire laser. Thulium vapors were created by a vacuum oven heated to 700 °C.

The hyperfine splitting of excited states was determined for the transitions under study. For some transitions, this was made for the first time; for others, the error was reduced by several times (see Table 1). This investigation allows identifying the lines and adjusting the laser to the frequency necessary for laser cooling. The conclusion is made that in this scheme of levels, a swap laser is most likely unnecessary. In addition, the decay constants for the upper levels were specified, which determine the rate of atom cooling and the Doppler limit for a laser-cooled ensemble.

To determine the branching factor for the transitions under study (the ratio of the probability of unfavored decays for upper levels (see Fig. 3a) to the total decay probability), the individual probabilities were calculated for each pair of levels with opposite parities. The Cowan software package [41] was used for this purpose, which provides energy level calculations for multielectron neutral atoms and the corresponding transition probabilities. It was found that the branching factors for the transitions under study are

$$k_{410\,\mathrm{nm}} = 1^{+1}_{-0.5} \times 10^{-5}\,, \quad (1)$$

$$k_{420\,\mathrm{nm}} = 5^{+5}_{-2.5} \times 10^{-5}\,. \quad (2)$$

Because 35,000 photons should be scattered in order to cool a thulium atom with the initial velocity 200 m s^{-1}, the loss of atoms in the cooling cycle is about 97% (conservatively) for the $\lambda = 420.4$ nm transition and 50% for $\lambda = 410.6$ nm. Thus, the transition $\lambda = 410.6$ nm is preferable for laser cooling of thulium atoms and capturing them by a magneto-optical trap (MOT). The calculation for the Zeeman cooler was performed [42] and the results led us to the conclusion that about 5% of atoms in the atomic beam at the temperature 1100 K might be cooled by a laser to the temperature corresponding to the velocity 20 m s^{-1} and then captured by the MOT.

The probability of the magneto-dipole transition between fine sublevels of the ground state was calculated. This probability is $A = 7(2)$ s^{-1}, which corresponds to the natural linewidth 1.3 Hz and the transition Q-factor 2×10^{14}.

2.2 Conclusions

Laser-cooled thulium atoms are promising for solving problems in high-precision spectroscopy. The substantial screening of the 4f shell allows detecting narrow lines in dense atomic ensembles with discrete atomic-like spectra even if they are implanted into solid substrates. For example, the clock transition with $\lambda = 1.14$ μm, having the finesse 2×10^{14}, is strongly screened against collisions [36], which opens the possibility of detecting narrow unperturbed resonances in a dense cloud of atoms captured by a magneto-optical trap.

We have shown that efficient laser cooling of thulium can be realized by using the radiation tuned with the transition

$$4f^{13}6s^2(F=4) \rightarrow 4f^{12}(^3H_5)5d_{3/2}6s^2(F=5)$$

at the wavelength 410.6 nm. It is likely that a repumper laser would not be needed in this case. The calculation shows that at the temperature 1100 K, about 5% of atoms in the thermal beam can be decelerated to velocities 20 m s^{-1} and then captured by a magneto-optical trap. In this case, about 10^6 atoms can be captured in the trap at the temperature $T_D = 230$ μK (the Doppler limit). Further cooling of atoms is possible either by means of sub-Doppler cooling or by switching to the transition with the wavelength 530.7 nm, which corresponds to $T_D = 9$ μK.

Presently, we are working on creating a magneto-optical trap for thulium atoms.

3. Stabilizing lasers at a sub-Hertz level

For studying narrow optical transitions in atomic systems, laser sources are needed with characteristics not limiting the resolution of the system. To detect Hertz-width resonances (see, e.g., Refs [4, 9]), the laser frequency fluctuations must be within several tenths of a Hertz. This places exacting constraints on the design of reference cavities whose frequency is a reference for stabilizing the laser radiation frequency. Indeed, using the relation $\delta l/l = \delta\nu/\nu$ (where l is the cavity length and ν is the frequency of the laser field) for the cavity length $l = 0.1$ m, we obtain that the frequency changes by 1 Hz under the displacement of one of the cavity mirrors by less than 10^{-15} m, which equals the proton radius! Mirror vibrations, temperature variation, and fluctuations in the radiation power inside the cavity result in an instability of the cavity length and, hence, of the frequency of the laser stabilized. The creation of a compact and simple-to-use design of the reference cavity is a relevant problem, which determines the stabilization of the laser frequency at the sub-Hertz level.

3.1 Vertical cavities with temperature compensation

External active and passive systems designed for damping vibrations and temperature fluctuations in an ambient medium can only partially limit the influence of these factors, which is usually insufficient for satisfying the requirements considered above. Additional damping of fluctuations can be provided by a special construction of the cavity itself, which should be least sensitive to vibrations and temperature fluctuations. Several groups at the leading scientific centers are developing such systems (see, e.g., Refs [43, 49]). Noticeable progress in cavity stability was obtained through a special suspension whose plane passes through the center of mass of the system [43, 46].

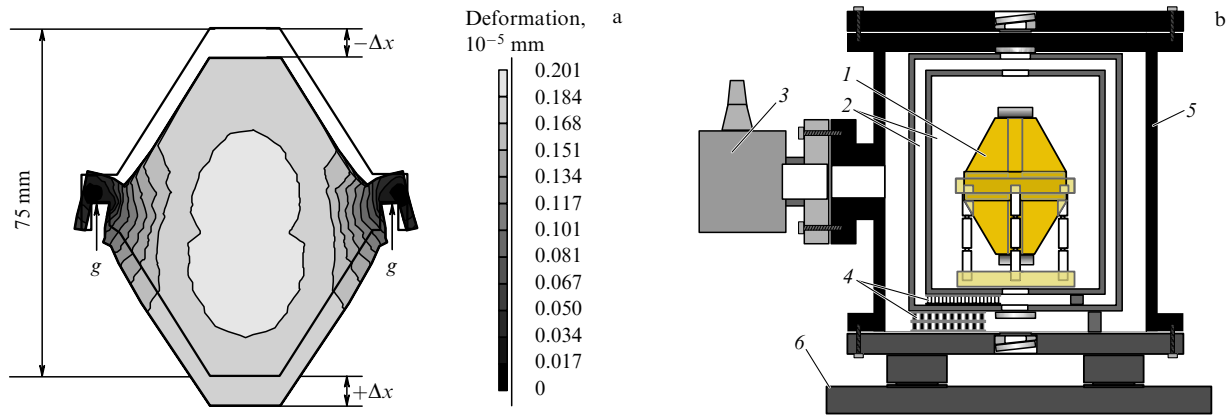

Figure 4. (a) Symmetric cavity with a vertical axis under the action of vertical acceleration. (b) Vertical cavity with the cooling and vacuum systems assembled: *1*, the cavity; *2*, the cooled duralumin screens; *3*, the getter-ion pump; *4*, Peltier elements; *5*, the vacuum chamber; *6*, the optical mount with an active system for suppressing vibrations.

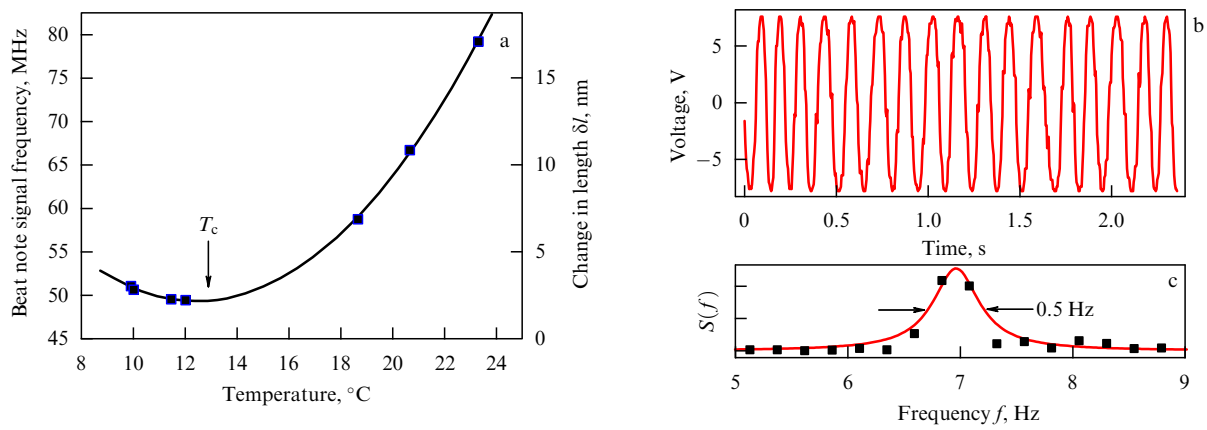

Figure 5. (a) Determination of the critical temperature T_c for a cavity made from ULE glass. (b) Oscillogram of the beat note signal for two lasers stabilized with respect to two independent cavities. (c) Spectral power density for the beat note signal presented in Fig. b.

Our group has combined the construction of a vertical cavity with the symmetric suspension shown in Fig. 4a. It can be seen that vertical acceleration reduces to a symmetric compression and tension of the top and bottom parts, respectively. The distance between cavity ends remains unchanged in the process. Such a construction of the cavity and suspension results in a weaker effect of vertical vibrations. Two mirrors with multilayer coatings, which provide the cavity finesse 4×10^5 at the wavelength 972 nm, are attached by an optical contact to the ends of the cavity made from special ULE glass (ultra low expansion glass [50]). This wavelength was chosen because it coincides with the fourth subharmonic of the radiation needed for a two-photon excitation of the 1S–2S transition in the hydrogen atom (243 nm). The cavity is placed in a vacuum chamber, which is continuously evacuated by a getter-ion pump (see Fig. 4b). The residual vacuum in the chamber is 10^{-8} mbar. For reducing horizontal vibrations, the cavity was placed on an optical table supplied with an active system for damping vibrations.

An important factor affecting the cavity stability is fluctuations in the temperature of the ambient medium. Usually, cavities are supplied with complicated multistage systems for temperature compensation and are made from a special material having a minimal thermal expansion coefficient. For example, ULE glass exhibits the specific dependence of length on temperature expressed as

$$\frac{\delta l}{l} \sim 10^{-9}(T - T_c)^2 , \qquad (3)$$

where T_c is the so-called critical temperature at which the length is minimal and the linear coefficient of thermal expansion vanishes. Usually, T_c is below room temperature and the system of external temperature stabilization cannot be used because of the moisture condensation. In fact, cooled cavities capable of providing laser stabilization for many days have not been used in practice yet.

We realized a method for cooling the cavity to T_c [49] by using Peltier elements placed in a vacuum chamber and multistage thermal screens (see Fig. 4b). The system for temperature stabilization is substantially simplified in this case and no condensation problem arises. A semiconductor laser in the Littrow configuration was stabilized with respect to the cavity transmission peak by means of an electronic feedback [51]. Two independent systems were made, which allowed carrying out experiments on determining T_c and studying the frequency stability of each of the systems.

In Fig. 5a, the frequency of the beat note signal is shown for two stabilized semiconductor lasers versus the tempera-

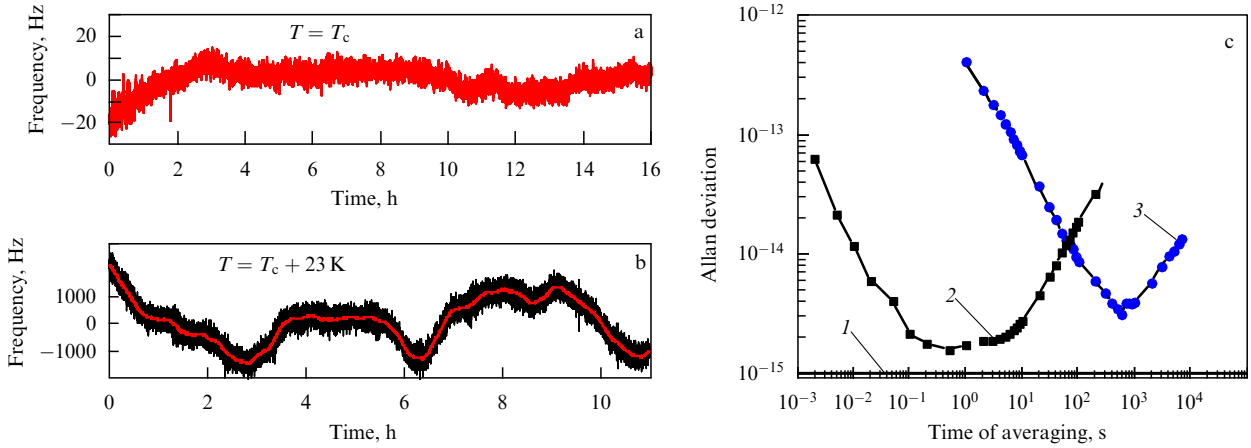

Figure 6. (a) The frequency of a cavity stabilized at the temperature T_c. The linear drift 63 mHz s^{-1} is subtracted. (b) Stability of the cavity frequency at the temperature $T_c + 23$ K. (c) Allan deviation graphs for the thermal noise of the cavity (curve *1*); for the beat note signal for two cavities, one of them stabilized at T_c and the other at $T_c + 23$ K (curve *2*); for the beat note signal for a cavity at T_c and the frequency comb stabilized by an active hydrogen maser (curve *3*).

ture of one of the cavities (see Fig. 4b). It can be seen that the cavity length is minimal at $T_c = 12.5\,^{\circ}$C; hence, the cavity exhibits the least sensitivity to variations of the external temperature in this case. The stabilizing system maintains the temperature near T_c with an accuracy about 1 mK, which corresponds to the sensitivity of the cavity frequency to temperature variations at the level of 50 Hz mK^{-1}.

For studying the short-time frequency characteristics of a stabilized laser, we recorded the oscillogram of frequency beatings with heterodyne conversion to low frequencies (see Fig. 5b). Fourier analysis of the signal shows that the radiation spectral width of each of the lasers is below 0.5 Hz at the averaging time 2 s (see Fig. 5c).

To characterize the long-time stability and determine the drift characteristics of the cavity, we measured the laser frequency by means of a frequency comb stabilized by the signal from an active hydrogen maser. A laser stabilized with respect to the cavity at the temperature T_c exhibits an almost linear drift of the frequency, of the order of 50 mHz s^{-1}, which arises due to material aging (recrystallization). As can be seen from Fig. 6a, deviations from the linear drift do not exceed 20 Hz for a time lapse of approximately 10 hours. For comparison, we measured the drift characteristics for a cavity stabilized at a temperature much higher than the critical, at $T_c + 23$ K. The sensitivity to temperature fluctuations increases to ~ 10 kHz mK^{-1}. This fact is confirmed by experimental observations (see Fig. 6b).

The results of the full analysis of characteristics of the laser system created are presented in Fig. 6c in the form of the Allan deviation [52]. Two independent laser systems were used, one of them stabilized with respect to the frequency of the cavity at the temperature T_c and the other with respect to the frequency of the other cavity at $T_c + 23$ K (curve *2*). The relative instability of the total system reaches 2×10^{-15} in the time interval 0.1 – 10 s, approaching the fundamental limit for cavity thermal noise (curve *1*) [53]. The cavity stability at T_c is also characterized by using a frequency comb (curve *3*). The measurements show that the instability of the laser system remains at the level of several units in the fifteenth digit for the time lapse of several hundred seconds.

3.2 Measurement of hyperfine splitting for the 2S level in a hydrogen atom

The laser system created was used for measuring the frequency of the hyperfine interval for the 2S level in the hydrogen atom. High-precision measurement of the frequency $f_{\rm HFS}(2{\rm S})$ of this interval allows carrying out a sensitive test for small quantum mechanical contributions [54, 55] based on the analysis of the specific difference $D_{21} = 8 f_{\rm HFS}(2{\rm S}) - f_{\rm HFS}(1{\rm S})$ [56].

Earlier, we suggested a new method for measuring $f_{\rm HFS}(2{\rm S})$ by two-photon spectroscopy on the 1S – 2S transition in the hydrogen atom [54]. As can be seen from Fig. 7a, the relation

$$f_{\rm HFS}(2{\rm S}) = f_{\rm HFS}(1{\rm S}) + f_{\rm triplet} - f_{\rm singlet} \qquad (4)$$

holds in vanishing magnetic fields, where $f_{\rm triplet}$ and $f_{\rm singlet}$ are the frequencies of triplet and singlet two-photon transitions. Hence, the problem of measuring the hyperfine splitting of the 2S level reduces to finding the difference between two optical frequencies because the value of $f_{\rm HFS}(1{\rm S})$ is known to the 13th decimal place [57].

The method of measurements of $f_{\rm HFS}(2{\rm S})$ used in 2008 is similar to that in [54], with the difference being that for a nonabsolute stable optical-frequency reference, we used the radiation of a diode laser with $\lambda = 972$ nm stabilized with respect to the cavity described in Section 3.1. The measurement results are presented as a histogram in Fig. 7b. The histogram is described by a Gaussian distribution with the width 85 Hz. We note that so small a statistical deviation is obtained in subtracting two optical frequencies of 2.26×10^{15} Hz each. The result of preliminary data processing gives $f_{\rm HFS}(2{\rm S}) = (177{,}556{,}840 \pm 5)$ Hz [58], which considerably refines the result $(177{,}556{,}860 \pm 16)$ Hz of the previous optical measurement carried out in 2003 [54]. We note that the new result, which is five times more accurate than that obtained by the direct radiofrequency method $(177{,}556{,}785 \pm 29)$ Hz [59] allows analyzing small (at the level of $< 10^{-7}$) corrections to the quantum electrodynamics of bound states in light atoms, and thus naturally complements tests based on an analysis of the Lamb shift.

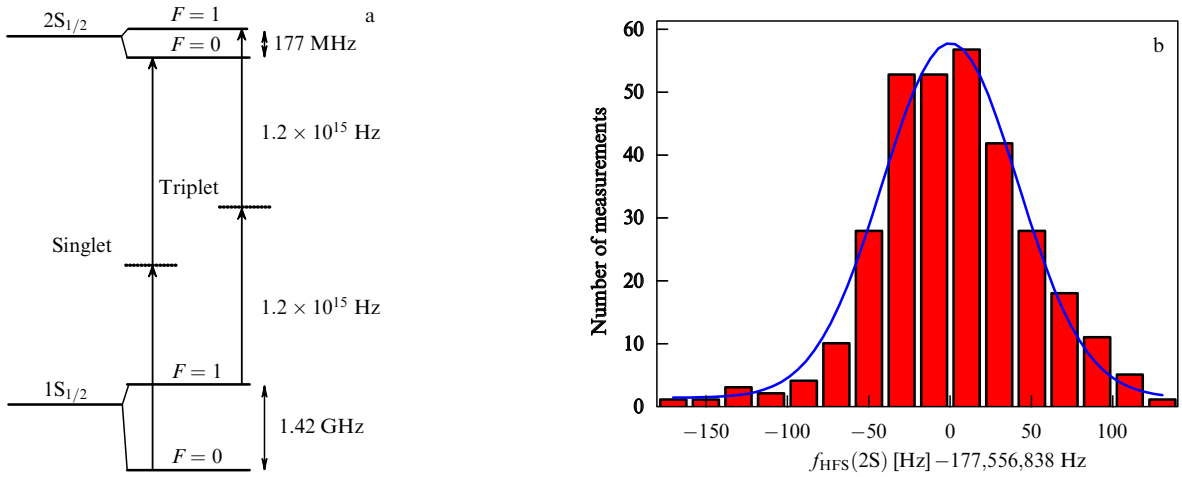

Figure 7. (a) Schematic diagram of measuring the hyperfine splitting for the 2S level by two-photon spectroscopy of the 1S – 2S transition. (b) Hystogram of the measuring results for $f_{\rm HFS}(2{\rm S})$ in the hydrogen atom and its approximation by a Gaussian profile.

3.3 Conclusions

New kinds of vibration- and temperature-compensated reference cavities capable of stabilizing laser systems with a sub-Hertz accuracy have been developed and realized. Such compact systems with typical dimensions $50 \times 50 \times 50$ cm^3 allow studying clock transitions in various atomic systems. We plan to use reference cavities of such a construction in the spectroscopy of clock transitions in thulium ($\lambda = 1.14$ μm) and hydrogen ($\lambda = 0.92$ μm) atoms. A laser system has been created with the wavelength $\lambda = 972$ nm, the spectral line width less than 0.5 Hz, the monotonic drift about 50 mHz s^{-1}, and the frequency instability below 5×10^{-15} over a time lapse in the range 0.1 – 100 s. At such durations, the frequency instability of the laser system is actually completely determined by the fundamental limit of the thermal noise of the reference cavity kept at a temperature about 300 K.

Using this laser system at the wavelength $\lambda = 972$ nm, we measured the hyperfine splitting of the 2S level in the hydrogen atom, which allows testing small corrections to the quantum electrodynamics of bound states. The result of preliminary processing of data measured in 2008 is $f_{\rm HFS}(2{\rm S}) = (177{,}556{,}840 \pm 5)$ Hz, which is five times better than the result obtained earlier by a direct radiofrequency measurement [59].

4. Interaction of laser-cooled ^{87}Rb with femtosecond radiation

Laser cooling of atoms opens unique possibilities for studying their intrinsic states. Most high-precision spectroscopic investigations are presently carried out with the help of laser cooling methods. For example, the use of such methods in cesium fountains has increased the accuracy of cesium standards reproducing the time unit in the SI system of units by more than an order [5]. Atom cooling in optical frequency references down to temperatures about 10 μK almost completely eliminates the influence of the Doppler effect and the limitation related to the finite time of the interaction with radiation. The development of special methods capable of scanning for atoms either in the free-falling mode [8] or in optical gratings at a 'magical' wavelength [12] ensures a minimal influence of external fields on the clock transition frequency.

The long lifetime of laser-cooled atoms in the trap domain allows reliably detecting processes occurring in a characteristic time about 1 s, which cannot be done, for example, with atomic beams. One such process is, in fact, the excitation of a clock transition. In the thulium atom, for example, the expected excitation rate for the $\lambda = 1.14$ μm transition is 0.1 – 1 s^{-1}. Detection of such processes is possible either by using the method of quantum hops for a small number of trapped atoms [60] or by measuring the population of the ground state in a large ensemble. In both cases, the act of excitation is detected via the two-photon process, namely, absorption at the clock transition wavelength and luminescence in another transition, which is usually the cooling transition.

For studying multi-photon processes in a cloud of laser-cooled atoms, we created a MOT for ^{87}Rb atoms and analyzed the interaction of the cloud with radiation from a femtosecond laser.

4.1 Magneto-optical trap for ^{87}Rb

We have created a MOT for ^{87}Rb atoms (see, e.g., Ref. [61]). A distinguishing feature of the configuration of this MOT is the compactness and reliability of the system. Atoms are trapped in a rectangular glass cell evacuated to a pressure below 10^{-9} mbar. As the source of atoms, we used the special dispensers produced by SAES Getters capable of precisely controlling the flux of rubidium atoms by varying the dispenser current. The cooling laser radiation at the wavelength 780 nm [the $5{\rm S}_{1/2}(F = 2) \to 5{\rm P}_{3/2}(F = 3)$ transition in Fig. 8a] is generated by a semiconductor laser stabilized by a resonance transition in rubidium with the laser frequency differing from the atomic resonance frequency by −12 MHz. To avoid populating the 'dark' sublevel $F = 1$ of the ground state, we used an additional repumping laser. After thorough intensity balancing, beams of the laser pass from six directions onto a glass cell. The crossing center of the six beams matches the three-dimensional minimum of the magnetic field produced by two coils in an anti-Helmholtz configuration with the field gradient about 10 G cm^{-1}. MOT luminescence at the wavelength 780 nm is detected by a CCD camera and a calibrated photodiode.

The cloud of cold atoms has the diameter 200 μm (at the level $1/e^2$) as shown in Fig. 8b. At the maximal current

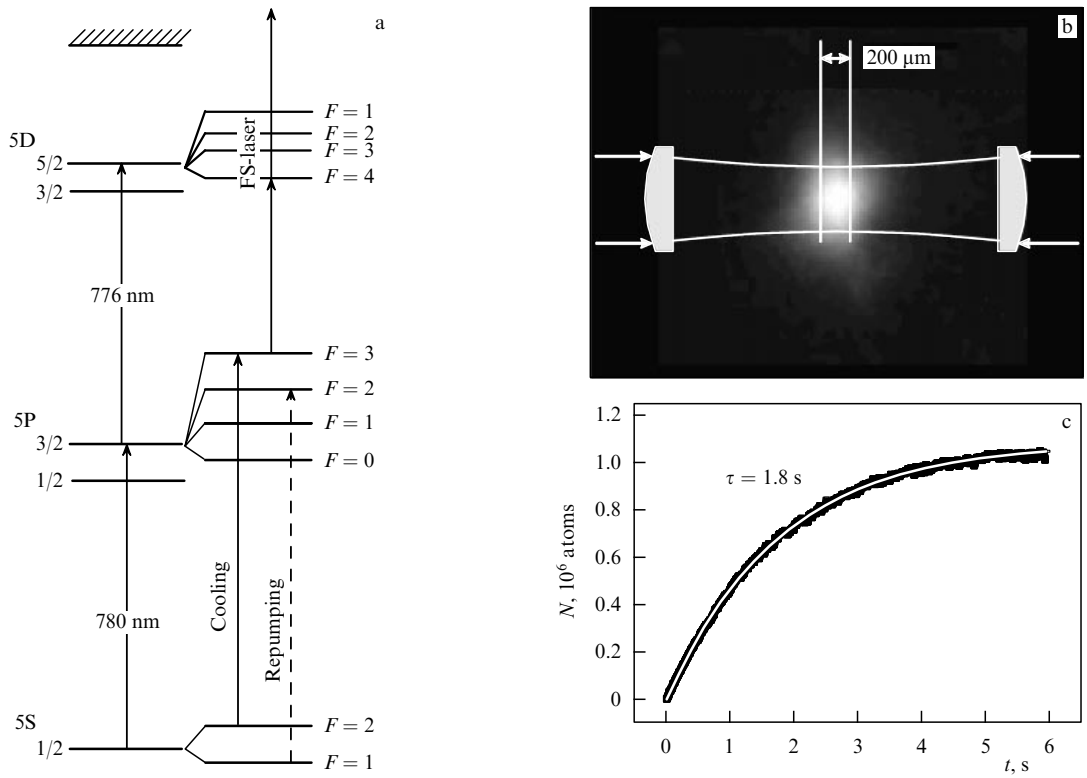

Figure 8. (a) Level structure of the ^{87}Rb atom used in the experiment. (b) MOT image at the wavelength of luminescence 780 nm. Focusing of femtosecond laser radiation is shown schematically. (c) Curve of trap loading. The approximation of experimental data by the exponential function of type (5) with the time constant $\tau = 1.8$ s is shown.

through the dispensers, the number of trapped atoms reaches 10^7, which corresponds to the cloud density 10^{12} cm^{-3}. The temperature of atoms measured by the method of recapture was 250 μK, which is close to the Doppler limit $h\gamma/2k_B = 140$ μK, where $\gamma = 6$ MHz is the cooling transition width.

An important MOT parameter is the atom lifetime in the trap domain. It can be determined from the characteristic time for trap load

$$N(t) = R\tau\left[1 - \exp\left(-\frac{t}{\tau}\right)\right], \tag{5}$$

where N is the number of atoms in the MOT and R and τ are the capture rate and lifetime. At a low atom concentration, we can neglect the losses caused by atom collisions inside the MOT. As can be seen from Fig. 8c, the characteristic trap load time is $\tau = 1.8$ s at the dispenser current 4.5 A. The lifetime of atoms in the MOT is mainly affected by collisions with rubidium atoms emitted from the dispensers. A reduction in the current results in longer τ; however, the capture rate and the total number of atoms $N(t \to \infty)$ simultaneously decrease. The regime in which τ is about 2 s is optimal for reaching both the maximal number of atoms in the trap (about several million) and their longest lifetime. It follows from Eqn (5) and Fig. 8c that every process with the frequency exceeding 0.5 s^{-1} that expels atoms from the cooling cycle (collisions, pumping into 'dark states', ionization) would substantially influence the shape of the load curve similar to the curve presented in Fig. 8c. This opens the possibility for a detailed analysis of such processes.

4.2 Interaction with a field from a femtosecond laser

The present section is devoted to studying the interaction of atoms captured by a MOT with radiation from a femtosecond (fs) laser operating in a cw mode. The specific features of this interaction were theoretically considered more than 30 years ago; however, intensive experimental investigation in this field started not long ago. In 1977, Baklanov and Chebotaev [62] predicted the possibility of detecting narrow two-photon resonances with the spectral width corresponding to the natural width despite a considerable spectral width of the laser source (up to several tens of nm for fs lasers).

This fact is explained by the specific spectrum of such lasers, which is given by a comb of optical modes whose frequencies are

$$f_n = f_0 + n f_{\text{rep}}, \tag{6}$$

where f_0 is the offset frequency caused by a difference in the group and phase velocities inside the laser cavity, f_{rep} is the laser pulse repetition frequency, and n is the mode number [14]. Successful use of fs lasers in spectroscopic studies of levels with the spectral width close to the natural width is demonstrated by the spectroscopy of anti-Stokes scattering [63], two-photon spectroscopy [64], and single-photon spectroscopy of atomic beams [65].

In our experiment, radiation from an fs laser ($f_{\text{rep}} = 76$ MHz, the pulse duration was 200 fs) was focused from opposite sides onto a cloud of atoms residing in a MOT (see Fig. 8b) with the beam waist $w_0 = 210$ μm overlapping the entire cloud. The central wavelength of the laser was 776 nm, which corresponds to the $5P_{3/2} \to 5D_{5/2}$ transition wavelength in ^{87}Rb (see Fig. 8a). Under variations in the

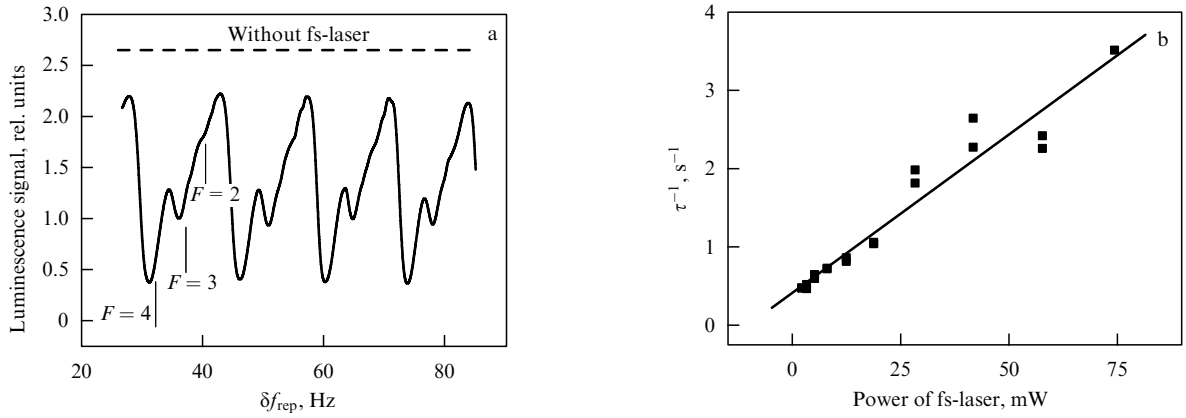

Figure 9. (a) Periodical dependence of MOT luminescence signal versus the detuning $\delta f_{\rm rep}$ of the repetition frequency of an fs laser. (b) Dependence of the atom inverse lifetime τ^{-1} on the power of fs laser radiation.

repetition frequency of the fs laser, periodic resonance variations of the luminescence signal for the $5P_{3/2}(F=3)$ level at the wavelength $\lambda = 780$ nm were detected as shown in Fig. 9a. The spectrum periodicity is explained by the periodicity of the fs-laser spectrum (6). Spectrum identification shows that the lines correspond to the $5P_{3/2}(F=3) \to 5D_{5/2}(F=2,3,4)$ transitions excited by separate modes of the fs laser from the upper populated level of the trap (see Fig. 8a). But so strong an effect resulting in the resonance depletion of the $5P_{3/2}(F=3)$ level and, correspondingly, in a tenfold suppression of luminescence cannot be explained in the framework of single-photon excitation because the light intensity power produced by the single mode of an fs-laser (6) is small in a cloud.

We analyzed this effect in a series of studies, where we separately investigated the effect of monochromatic radiation, mechanical action at the resonance transition, optical pumping, and ionization on the MOT excitation luminescence [66]. The conclusion was that a decrease in the number of atoms in a MOT is related to the two-photon process under which the atoms excited by a single mode of an fs laser and transiting to the $5D_{5/2}$ level are then ionized by the full power of the fs laser. The probability of this process is of the order of 1 to 10 s^{-1}, depending on the Fs laser power, and therefore the effect can be investigated based on the lifetime of atoms in the trap, Eqn (5).

To simplify interpretation of the phenomenon observed, the scheme of the experiment was modified. The $5P_{3/2}(F=3) \to 5D_{5/2}(F=4)$ transition was excited by a CW frequency-stabilized diode laser tuned to the transition ($\lambda = 776$ nm). The $5D_{5/2}$ level was ionized by the radiation from an fs laser whose central wavelength was tuned to 820 nm, free of Rb-atom resonances. Laser beams overlapped such that the spatial modes of the lasers coincided. In this case, the field of the fs laser, which does not itself affect the MOT, plays the role of a 'catalyst,' noticeably increasing the influence of the resonance field of the CW laser. In Fig. 9b, the inverse lifetime for an atom in the MOT is shown versus the radiation power of the fs laser. The power of the CW laser remained constant.

From experimental data on the known effective ionization cross section $\sigma_{\rm eff} = (1.2 \pm 0.2) \times 10^{-17}$ cm^2 for the $5D_{5/2}$-level [67], we succeeded in determining the efficiency of exciting the $5P_{3/2}(F=3) \to 5D_{5/2}(F=4)$ transition with the field of the CW laser with a power P_{776} in our experimental configuration. It was found that the efficiency is $\vartheta = (1.9 \pm 0.5) \times 10^{-4}\, P_{776}$ [μW^{-1}]. Despite the relatively high uncertainty, this method proves to be substantially more precise than direct determination of the level population by luminescence, which is especially difficult in the case of weak signals. The fraction of atoms passing to the $5D_{5/2}$ level as the result of excitation by a single mode of the fs laser with the average power 100 mW and the central wavelength 776 nm is 3×10^{-4}. This corresponds to the total number of excited atoms about 300.

4.3 Summary

The compact magneto-optical trap is created for rubidium-87 atoms, capable of capturing about $10^6 - 10^7$ atoms with the density $10^{11} - 10^{12}$ cm^{-3} at the temperature 250 μK. The lifetime of an atom in the trap is about 2 s, which gives the possibility to quantitatively study weak processes with the characteristic frequency above 0.5 s^{-1}.

It is shown that the interaction of atoms with the field of a passive mode-locked femtosecond laser at the central wavelength 776 nm is of a resonance character. The strong resonances observed in the experiments are explained by the excitation of the $5P_{3/2} \to 5D_{5/2}$ transition by a single mode of the fs laser and then by the ionization of the $5D_{5/2}$ level, which results in atoms being expelled from the cooling cycle. The width of resonances observed is close to the natural transition widths. By measuring the time constant for the trap load curve, we quantitatively interpreted this two-photon process and developed a sensitive method for determining the population of the $5D_{5/2}$ level in ^{87}Rb. The method allows a confident detection of the transition of several hundred excited atoms in a cloud to an upper electron level without collecting luminescence photons from this level, which is important in studying strongly forbidden clock transitions.

The methods developed are needed for solving problems at the next stage, such as studying the excitation of the two-photon metrological $5S_{1/2} \to 5D_{5/2}$ transition (778 nm) in ^{87}Rb with the field of a femtosecond laser and spectroscopy of laser-cooled thulium atoms (see Section 2). Substantial experience was acquired in working with laser-cooled atoms and the possibility was experimentally demonstrated to quantitatively analyze weak processes of interaction with radiation, which include the excitation of the clock transition.

5. Conclusion

A series of works is being performed with the aim of developing a new optical frequency reference for studying the time variation of the fine structure constant α. As a possible object for investigations, we suggested the metrological transition at the wavelength 1.14 μm in the strongly relativistic ^{169}Tm atom, whose frequency is weakly affected by a collisional shift. The possibility of laser cooling of a thulium atom was studied and was shown to be feasible for the transition at the wavelength 410 nm. A laser system was created whose frequency matches that of the cooling transition. Under construction are a magnetic system of a Zeeman cooler, a vacuum chamber, and an optical system for cooling and capturing atoms with a magneto-optical trap.

A new generation of optical cavities has been developed and created with low sensitivity to temperature fluctuations and vibrations. The spectral width of a semiconductor laser ($\lambda = 972$ nm) stabilized relative to such a cavity is below 0.5 Hz at the frequency drift about 50 mHz s^{-1}. The hyperfine splitting frequency $f_{\rm HFS}(2{\rm S}) = (177{,}556{,}840 \pm 5)$ Hz for the 2S-level in the hydrogen atom (a preliminary result) was measured by using such a laser system, which opens the possibility of performing sensitive tests in quantum electrodynamics. A similar laser system was developed for the spectroscopy of the clock transition in the ^{169}Tm atom.

A compact magneto-optical trap was created for ^{87}Rb atoms and the interaction of laser-cooled atoms with a femtosecond radiation was studied. It has been experimentally shown that weak processes with the characteristic rates down to 0.5 s^{-1} can be studied quantitatively by measuring the characteristic load time for the trap. The methods developed open the possibility of detecting the weak clock transition in thulium atoms.

The use of new objects in spectroscopic investigations, laser systems, and methods for measuring clock transitions is likely to favor achieving, within the next few years, sensitivity to the drift of the fine structure constant at the level of $\dot{\alpha}/\alpha \sim 10^{-18}$ yr^{-1}. This brings laboratory methods to a leading position in studying the drift of α in the modern age of the evolution of the Universe.

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