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300 YEARS OF PHYSICS IN RUSSIA AND 80 YEARS OF FIAN

Precision laser spectroscopy in fundamental studies

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Contents

Abstract. **The role of precision spectroscopic measurements in the development of fundamental theories is discussed, with particular emphasis on the hydrogen atom, the simplest stable atomic system amenable to the accurate calculation of energy levels from quantum electrodynamics. Research areas that greatly benefited from the participation of the Lebedev Physical Institute are reviewed, including the violation of fundamental symmetries, the stability of the fine-structure constant α, and sensitive tests of quantum electrodynamics.**

1. Introduction

In the middle of the 19th century, Robert W E Bunsen and Gustav R Kirchhoff started a new era in physics — the spectroscopy era. Even the simplest spectroscopy methods of that time were highly sensitive and selective, allowing not only the study of the composition of the burner flame, but also making conclusions about the chemical composition of the Sun. Thus, a new element — helium — was discovered in 1868 by indicating the 587.6 nm yellow line in the spectrum of the Sun's corona. J Janssen and N Lockyer, who made this discovery, succeeded in resolving a new yellow line, which was just 1.5 nm away from the resonant sodium line. The prism spectrometer had become a powerful tool in the hands of scientists studying surrounding world, a tool operating like an original dactyloscope detecting the 'fingerprints' of the chemical elements, which were invisible up to that time.

However, it took more than half a century for scientists to understand the nature of the spectra: only in 1913 did Niels Bohr introduce the hydrogen atom theory which explained the empiric spectral dependences that Johann Balmer had derived 18 years before. Scientists started to gradually unravel the mysteries of the simplest stable atom — hydrogen — establishing a new research area: quantum mechanics. All atomic spectroscopy textbooks start off with the Bohr model which correctly describes the stationary states of a single-electron atom and demonstratively uses wave–particle duality.

Later on, in 1926, Erwin Schrödinger will write down the famous wave equation. By solving this equation for an electron in the field of an infinitely heavy Coulomb center, the simple dependence of the energy E of the level on the principal quantum number n is obtained:

$$E_n = -\frac{\mathrm{Ry}}{n^2}\,, \qquad (1)$$

where Ry is the Rydberg constant. This result confirms the Bohr theory.

After increasing the accuracy of spectroscopic measurements, the fine structure of the Balmer series was discovered in the hydrogen atom, and this effect could not be explained in the framework of the Schrödinger equation. Indeed, in order to describe the fine structure, one needs to take into account the relativistic corrections for the electron, which moves in the first Bohr orbit with the speed $v = \alpha c$, where α is the fine-structure constant, and c is the speed of light. In 1928, P A M Dirac wrote down the relativistically invariant wave equation with the electron spin taken into account. The Dirac equation explained all features in the hydrogen spectrum known by that time and was successfully confirmed by many experiments in succeeding almost 20 years.

In 1947, Willis Lamb and Ernest Rutherford made a discovery which led to further change in the theoretical concepts. They performed a microwave spectroscopy of the hydrogen atom and discovered that the $2S_{1/2}$ energy level is placed higher than the $2P_{1/2}$ level, and the splitting between them is around 1 GHz. Such splitting could not be explained in the framework of the Dirac theory, and one needed to take

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into account the quantum mechanical corrections related to the existence of the physical vacuum. In the late 1950s, R Feynman, J Schwinger, and S-J Tomonaga developed the quantum electrodynamics theory (QED), which explained the origin of the Lamb shift and predicted its value with a high accuracy. To date, the QED is the most precise theory permitting us to predict different physical values (lepton g-factor, energy levels of simple systems, etc.) with an accuracy of up to the 12th decimal place.

We see that hydrogen-atom spectroscopy has played a significant role in the development of modern quantum mechanics. The improvement in measurement accuracy has led many times to the discovery of absolutely new effects calling for the revision of existing concepts on the nature of electromagnetic interactions. Precise spectroscopic methods continue now to provide a useful tool in the studies of many fundamental problems, such as confirmation of basic postulates in physics (principles of Einstein equivalence, basic symmetries, invariance of fundamental constants), tests of fundamental theories (for example, equations in quantum mechanics and electrodynamics, Standard Model or gravity theory), as well as high-precision measurements of fundamental constants.

Of course, the main goal for researchers is to discover new effects and develop new theoretical models in the field of fundamental interactions. Great success in this research area is achieved by using methods of high-energy physics: more powerful accelerators are built, astrophysical objects with matter in extreme states are studied, strong-field physics is being developed. However, investigations in the low-energy range are no less effective. Here, new physical phenomena can be observed by improving the sensitivity and accuracy of the measurements. The example of hydrogen-atom spectroscopy shows that such measurements often lead to unexpected results and open up new fields in physics.

The progress in the accuracy of spectroscopic measurements of the hydrogen atom is shown in Fig. 1. One can see that the tendency of this plot changes two times, indicating the classical spectroscopy era, the laser spectroscopy period, and the modern era of optical-frequency measurements. This evolution reflects the appearance of fundamentally new devices: in the late 1970s, lasers, being previously only

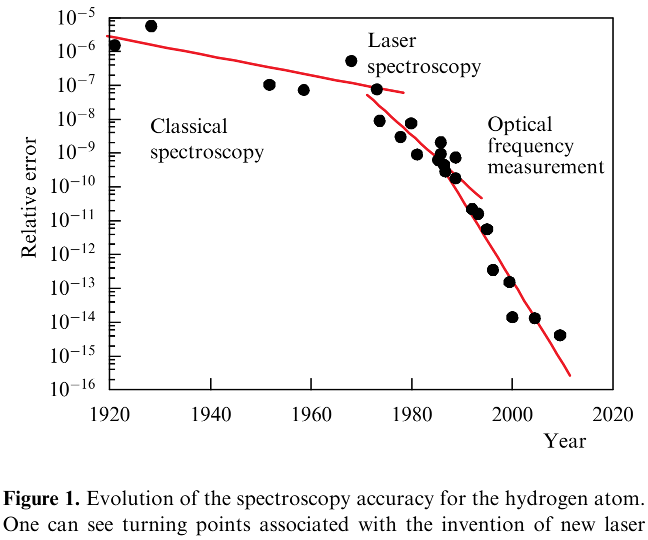

Figure 1. Evolution of the spectroscopy accuracy for the hydrogen atom. One can see turning points associated with the invention of new laser methods in the 1970s and new optical frequency measurement methods at the beginning of the 1990s.

subjects of investigation, started to become instruments which improved the resolution of the spectroscopic methods and led to fast progress in measurement accuracy. Precisely at that time, the work on laser spectroscopy and its applications to fundamental problems was started at the Lebedev Physical Institute (LPI).

2. First steps

2.1 Optical quantum generators for fundamental problems

One of the pioneering studies, where molecular frequency standards were suggested for the investigation of relativistic effects, is the article by N G Basov et al. [1], published in *Physics–Uspekhi* in 1961. The authors of this paper understood the huge opportunities offered by optical quantum generators and suggested that their unique properties be used to verify the postulates of general and special theories of relativity and to study some cosmological effects. In 1968, N G Basov, already a Nobel laureate, together with his postgraduate student V S Letokhov (now a famous scientist, the founder of some modern laser spectroscopy methods), published the famous paper "Optical frequency standards" [2]. In this study, along with a description of new methods for stabilization of the laser radiation frequency, the authors suggested using highly stable optical quantum generators to measure the speed of light and tests for the stability of fundamental physical constants.

The development of the suggested methods for the measurement of the speed of light led to a fast increase in the measurement accuracy and to a redefinition of the meter in 1983, when the XVII General Conference on Measures and Weights assigned the value of 299,792,458 m s^{-1} to the speed of light [3]. At the same time, one of the most sensitive methods for the investigation of fundamental constant stability is still the method where the frequencies of narrow atomic transitions of different natures are compared, as was suggested by Basov and Letokhov [2]. In 1970, Letokhov passed to the Institute of Spectroscopy of the USSR Academy of Sciences, and research at LPI was continued by several scientific groups in the quantum radiophysics and optics divisions.

Work on the precise laser spectroscopy of methane (wavelength $\lambda = 3.39$ μm) was performed in the LPI quantum radiophysics department. The hyperfine structure of the infrared spectrum of methane was studied in the He–Ne laser wavelength range, recoil doublets were registered, and the frequency stability of laser radiation was essentially increased [4]. As a result of many years of work, compact laser systems with a short-term relative frequency instability of up to 10^{-14} were assembled in the Frequency Standards Laboratory under the guidance of M A Gubin. These devices are assisted in performing studies in fundamental metrology and radio astronomy. High-stability lasers are also needed in many important practical applications.

2.2 Parity nonconservation studies

An important physical problem is the investigation of fundamental symmetries under parity transformation (P), charge conjugation (C), and time inversion (T). Until the middle of the 20th century, it was believed that the result of any physical measurement cannot depend on C, P, or T transformations. However, in 1956–1957, the anisotropy of ^{60}Co nuclei β-decay was discovered, and it was the first experimental examination of spatial-symmetry breaking: the laws of physics turned out to be different for 'right-hand' and 'left-hand' directions [5, 6]. Immediately after the experimental confirmation of the parity nonconservation, a number of outstanding scientists (M Gell-Mann, R Feynman, R Marshak, G Sudarshan) developed the universal theory of weak interaction (V – A-theory).

P-invariance violations, caused by weak interactions, could be clearly seen in nuclear physics, but for low energies (atomic physics) they turned out to be much weaker. The parity nonconservation effects in atoms are caused by a specific form of weak interactions — neutral currents. The theory, developed by S Weinberg, A Salam and others, was based on the prediction of weak neutral currents and succeeded in the unification of electromagnetic and weak interactions [7]. In order to verify the Weinberg–Salam theory, one needed a reliable observation of P-invariance violation in atomic systems.

The pioneering work published in 1959 by Ya B Zel'dovich [8] discusses the question of possible parity nonconservation in atomic systems. Zel'dovich considered an effect which led to the emission of circularly polarized photons from the hydrogen atom. However, this effect was too small for observation. The experimental search for parity nonconservation in atomic systems started much later — when sensitive experimental methods, based on laser spectroscopy, were developed.

Experimental work on P-invariance violation was initiated at LPI by I I Sobelman, who published in 1976, along with coauthors, the article "On parity nonconservation in atoms" in *Physics–Uspekhi* [9]. In the same issue, a group from Saint Petersburg (A N Moskalev, R M Ryndin, I B Khriplovich) published an article [10] which served as a basis for further experimental work [11] at the Budker Institute of Nuclear Physics in Novosibirsk. At LPI, it was decided to search for parity nonconservation effects in bismuth vapor by measuring the rotation of polarization plane of the laser radiation near the magnetic dipole resonance at the wavelength of 648 nm.

Experimental results on investigations of bismuth vapor optical activity were published by the LPI group in 1980 [12]. Figure 2 shows calculated spectra of the parity nonconservation effect and Faraday rotation in bismuth vapor [12]. One can see that for reliable observation of the effect the relative error (both statistical and systematic) of the optical rotation measurement needs to be on the order of 10^{-8}, which even now requires great effort to achieve. After overcoming several experimental difficulties, the LPI group managed to measure the value of the *R*-parameter, which characterizes the parity nonconservation effect [13]:

$$R_{\text{exp}}(\text{Bi}\,648) = -(7.8 \pm 1.8) \times 10^{-8}\,, \tag{2}$$

which is in good agreement with the theoretical prediction $R_{\text{theory}}(\text{Bi}\,648) = -(13 \pm 4) \times 10^{-8}$. As is shown in review [14], experiments on parity nonconservation in different atomic systems, performed by various scientific groups, perfectly confirm the Weinberg–Salam theory. This was the first step in building a more general Grand Unified Theory, which unifies strong, weak, and electromagnetic interactions.

The problem of investigating the basic symmetries is of great interest today as well. Despite the observations of C-, CP-, and T-symmetry violations, the baryon asymmetry of the Universe is still an open question, which cannot be

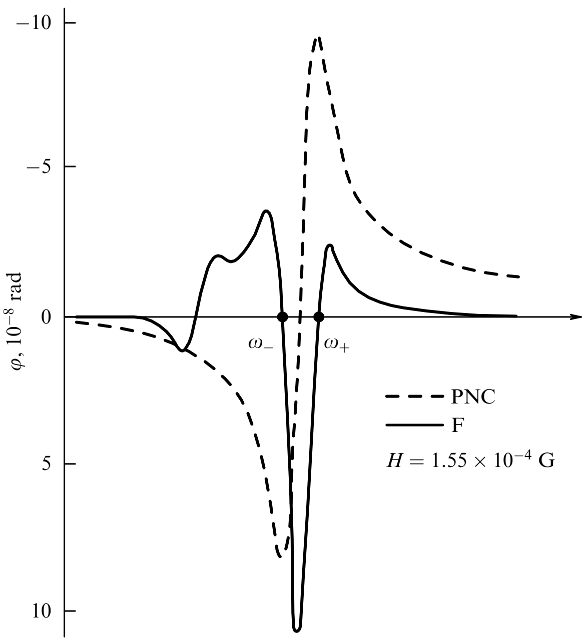

Figure 2. Calculated values of the parity nonconservation (PNC) effect and the Faraday rotation in optically dense bismuth vapor. (Taken from Ref. [12].)

explained in the framework of the Standard Model. The search for possible CPT-theorem violations continues, for example, through studying antimatter, which was successfully synthesized at CERN [15]. Modern technologies allow reliably confining antihydrogen ($\bar{\text{H}}$) in the ground state in magnetic traps. The first spectroscopic experiments are being prepared, and soon it will be possible to accurately compare the energy structures of atoms of matter and antimatter [16]. This will be one of the most sensitive tests for the CPT-theorem.

The antimatter investigations at CERN are performed with the participation of researchers from LPI, which is a member of the GBAR (gravitational behaviour of antihydrogen at rest) collaboration [17]. The GBAR collaboration was organized for experimental investigations into the gravitational interaction of matter and antimatter — one of the most topical fundamental problems in the field of low-energy physics. Antimatter gravitation is actually an unexplored field, and though there have been indirect observations based on Standard Model assumptions and the equivalence principle [18], this area has not been studied directly due to the fact that the gravitational interaction is very weak and the amount of antimatter synthesized in the laboratory is as yet small.

Within the GBAR project, single antihydrogen atoms are planned to be deeply cooled to temperatures which allow performing sensitive gravitational measurements by using the following method. As a result of low-energy antiproton-beam transmission through the positronium gas $\bar{\text{e}}\text{e}$, the emergence of positively charged $\bar{\text{H}}^+$ ions ($\bar{\text{e}}\bar{\text{e}}\bar{\text{p}}$ is the antimatter analog of a negative ion H^-) is expected. The probability of such a process is extremely small, but as soon as one single positive $\bar{\text{H}}^+$ antimatter-ion is produced, it can be separated and captured in an ion trap. The energy of the trapped ion would be around several electron-volts, which seems to mean that there can be no possible gravitational experiments. However, the positive charge of an ion allows cooling it through an ordinary ion, for example, the light beryllium ion, Be^+. The beryllium ion, which can be deeply laser-cooled, is widely used as a pair-ion [19]. Coulomb repulsion would prevent the annihilation, whereas collective oscillations in the trap potential would allow effectively cooling the system to temperatures of several μK. After terminating the cooling cycle, the $\bar{H}^+$ ion would be photoionized by a high-power laser pulse, and the produced neutral antihydrogen atom, in the absence of interaction with the trap, would begin free falling.

At present, the parts of the experimental setup are being planned and prepared; the GBAR project realization is scheduled to take place in 2018.

3. Research today

3.1 Search for drift of fundamental constants

The problem of the stability of dimensionless fundamental constants in time and space is one of the most intriguing in modern physics. What defines values of quantities like the fine-structure constant $\alpha = e^2/\hbar c$, the electron to proton mass ratio m_e/m_p, and many others? While using these quantities in our calculations, we seldom think about their origin and whether there is a reason for them to have exactly these values.

Fundamental constants are external parameters which are involved in one or another physical theories, making them incomplete and dependent on magnitudes that cannot be calculated within their own frameworks. An example of a complete theory in this sense would be the Bohr theory for the hydrogen atom spectrum (1), where the energy of the level is defined only by the integer n (Rydberg constant Ry can be defined as a unity by using the corresponding system of units). However, the Bohr theory inaccurately reproduces the observed spectrum, and for a full description, as we know, one needs several additional fundamental constants (see below).

The first attempt to explain the nature of fundamental constants, namely the fine-structure constant $\alpha \approx 1/137$, was made by P A M Dirac [20] in 1937. In this paper, Dirac questioned the stability of the α value, which laid the groundwork for a series of investigations that continue at the present time. There is hope that investigations of fundamental constants, the search for drifts and possible correlations can improve the understanding of the physics of the surrounding world, giving new knowledge about the origin and structure of the Universe.

Before the early 2000s, the main subjects of study were space object spectra, together with fossils and meteors (see review [21]). Laboratory investigations with masers and other microwave sources were performed as well. The method introduced by Basov and Letokhov [2] was applied. It was based on different sensitivities to the fundamental constants for transitions of different natures.

Achievements in the measurements of optical frequencies, namely creation of a femtosecond synthesizer [22] in 2001, formed the base for a diversity of high-precision laser experiments which exerted a pronounced influence on the progress of fundamental physics. Pioneering work on developing the optical frequency synthesizer was done at two large laser research centers: Max Planck Institute of Quantum Optics (MPQ) in Garching, Germany and Joint Institute for Laboratory Astrophysics (JILA) in Boulder, CO, USA. LPI maintains close cooperation with both these institutions. Using the data of these joint research has opened up fresh opportunities for making measurements in the optical range with a fantastically high precision up to the eighteenth decimal place [23].

At the beginning of the 2000s, LPI and MPQ performed a series of experiments on the high-precision spectroscopy of the hydrogen atom in order to specify the fundamental constants and search for their possible evolution. The LPI team suggested a new method for investigation of the α drift, which was discussed in detail in papers [24, 25]. The method is based on a comparison of absolute frequencies of the optical transitions in different atomic systems (H, Yb^+, Hg^+, and others), measured over several years with a femtosecond synthesizer. The sensitivity to α is determined by relativistic effects which strongly depend on the atomic system and transition type.

In 2004, this method resulted in a strong limitation imposed on the α drift in the modern era [26]:

$$\frac{\dot{\alpha}}{\alpha} = (-0.9 \pm 2.9) \times 10^{-15}\ \text{year}^{-1}\,. \tag{3}$$

It is important to note that the sensitivity of laboratory measurements to the possible linear drift of α at once reached values of the best astrophysical limitations of order 10^{-15} year^{-1}. Taking into account the fact that astrophysics investigates time intervals of up to 10 billion years, one can realize that the accuracy of experimental laboratory methods for laser spectroscopy that can accumulate data registered for only several years is extremely high.

This laboratory method was developed further and its sensitivity reached the value [19]

$$\frac{\dot{\alpha}}{\alpha} = (-1.6 \pm 2.3) \times 10^{-17}\ \text{year}^{-1}\,, \tag{4}$$

which is the strongest limitation on the α drift to date.

Figure 3 draws a comparison of the sensitivities of experiments performed using different methods. Astrophysical investigations were performed using a telescope/spectrograph [Keck Observatory and the Very Large Telescope (VLT)] system that registered absorption spectra of interstellar gas clouds. Laboratory measurements were performed using atoms and ions indicated in Fig. 3.

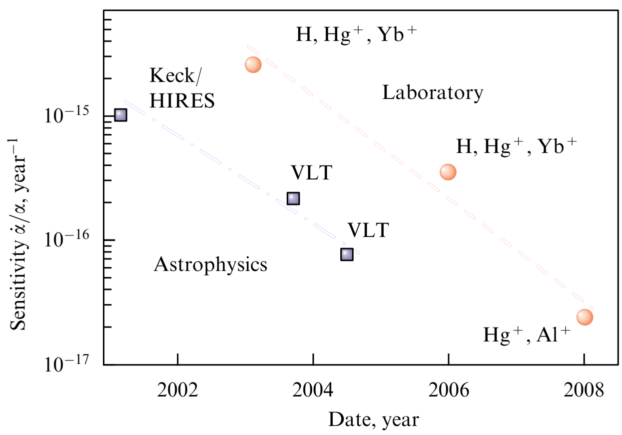

Figure 3. Comparison of sensitivity to the linear drift of α in modern astrophysical (squares) and laboratory (circles) methods. (HIRES — HIgh REsolution Spectrometer.)

These methods concern completely different eras of the Universe's evolution and are equally important for the problem under discussion. There is no doubt that, due to the constant increase in the precision of laboratory measurements in the optical range [23] and the large number of atomic systems available for research, the sensitivity to linear drift will reach in the nearest future the value of $\dot{\alpha}/\alpha \sim 10^{-20}$ year^{-1}. Possibly, a nonzero value of the drift will be registered at some moment, and we will witness many interesting discoveries.

3.2 Tm-based optical clocks

At the beginning of the 2000s, in addition to the joint research on the hydrogen atom, LPI started experimental work on laser cooling of thulium atoms and the search for the clock transition. Thulium is among rare-earth elements and its atom has features in the electron shell structure caused by the absence of one 4f-electron, whereas the outer (5s, 6s) shells are filled. Depending on the relative orientation of the atomic orbital momentum ($L = 3$) and the spin of the 'hole' in the f-shell ($S = 1/2$), the ground state of the atom splits into two sublevels: with $J = 7/2$, and $5/2$. Due to the same parity, the transition between them with the wavelength of 1.14 μm turns out to be forbidden in the electric dipole approximation, which makes it an interesting candidate for utilization in optical clocks. The magnetic dipole transition has a spectral line width of about 1 Hz [27] and is highly sensitive to α (as a transition probability between fine-structure components varies as $\sim \alpha^2$ Ry). These properties make it an interesting and promising object for studies of the possible α drift [28].

Experiments on laser cooling and the trapping of thulium atoms are held in the Optics Division of LPI. Highly stable laser sources for the spectroscopy of the 1.14-μm clock transition are being developed there as well. The thulium energy levels structure has been studied [27]; for the first time, laser cooling of thulium was carried out at the wavelength of 410 nm [29], its trapping was demonstrated using a magnetic trap [30], and secondary cooling was brought about using a weak transition at 530.7 nm [31]. The temperature of the cloud with 10^6 atoms reached the level of 25 μK, which allows reloading it into an optical lattice.

The optical lattice is a periodic potential formed by a light wave, where atoms are captured due to the dynamic Stark effect. The advantage of optical lattices lies in the realization of the Lamb–Dicke regime (atom localization in the volume with dimensions less than λ), which allows completely eliminating the influence of the first-order Doppler effect [32]. The disadvantages are the small depth of the optical potential (usually around several μK) and energy level shifts in the field of the light wave, which are, strictly speaking, responsible for the trapping in the lattice. In 2003, the H Katori group revealed that it is possible to choose a lattice wavelength for some systems in such a way that the shifts of the upper and lower metrological levels are the same [33]. If one forms an optical lattice with this 'magic' wavelength, the leading term in the differential polarizability vanishes. The most precise optical clocks based on neutral atoms, working on strontium [23], ytterbium [34], and others, all follow exactly this principle (see also review [35]).

The trapping of about 10^4 thulium atoms in the optical lattice was performed at LPI at the wavelength of 530 nm [31]. A photograph of the trapped atomic cloud (the luminescence is registered at the wavelength of the strong transition, 410 nm) is given in Fig. 4. When the optical lattice is formed, atoms are localized closer to the center of the waist,

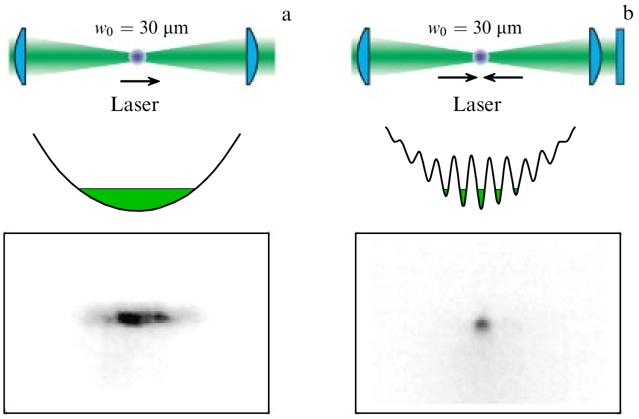

Figure 4. (a) Optical dipole trap. Thulium atoms are trapped in the intensity maximum of a propagating light wave at the wavelength of 530 nm. An optical schematics of the trap is shown, together with the potential profile and the photograph of the trapped cloud (figures are not to scale); w_0 is the radius of the beam waist. (b) Optical lattice which is formed in the waist of a standing wave, for which purpose a mirror is introduced in the experimental setup.

and the form of the cloud depends on the mutual polarization of the counterpropagating beams. The percentage of atoms trapped in the optical lattice from the magneto-optical trap is around 1% and is determined by the geometrical overlap of the laser waist with the cloud of cold atoms. In order to increase the efficiency of the retrapping, we are trying to decrease the temperature of the cloud of the secondary cooled atoms, which needs a narrowing of the cooling laser line width (530.7 nm). At the same time, calculations of the magic wavelength for the lattice are being carried out, as is the search for the clock optical transition at 1.14 μm, using the stabilized semiconductor laser system that we have assembled.

In concluding this section, it should be noted that, among the methods of laser cooling and trapping and manipulating atoms, thulium turned out to be a rather simple system (in comparison with widely used alkaline-earth atoms and neutral mercury), which additionally shows how promising and practically important the ongoing research is.

3.3 'Clock' lasers at LPI

The development of optical clocks, precise laser spectroscopy, and some of the laser cooling methods turn out to be impossible without exploiting so-called clock lasers — laser systems with an ultranarrow emission spectrum line. Clock lasers play an important role in many fundamental experiments (see, for example, paper [36]) and successfully find various applications due to their unsurpassed short-term stability. Thus, the relative stability of such lasers on time intervals from 1 to 10 s can reach 10^{-17} [37] (with the employment of cryogenic equipment), which is inaccessible for microwave range oscillators.

The heart of a stabilized laser system is a passive monolithic resonator with a large quality factor and a maximally high stability of the distance between the mirrors. The frequency of the laser source is stabilized around the transmission peak of the passive resonator through an active feedback loop. Various laser generators can be used for stabilization: semiconductor, fiber, solid state, and gas lasers, but the generated wavelength must be at resonance with the transition wavelength of the atomic system under study. The main difficulty lies in the construction of the passive resonator itself, because it needs to have minimal sensitivity to the temperature fluctuations and vibrations.

In 2006, we developed a temperature- and vibration-isolated resonator made from a special material — ultralow expansion (ULE) glass [38]. Attracting a set of novel approaches [27, 39], we decreased the laser frequency instability to the level of thermal noise, which corresponds to a spectral line width of less than 1 Hz. A number of systems were developed at LPI for different wavelengths: 698 nm (clock transition in strontium), 689 nm (secondary cooling transition in strontium), 1140 nm (clock transition in thulium), and 531 nm (secondary cooling transition in thulium), and the quality factors of the resonators reached values of up to 3×10^5. Unfortunately, the mirrors providing such good parameters are produced up till now only in the USA. However, all the other components of the system, including the resonator bodies of a special form, were made at LPI.

Figure 5 shows typical characteristics of the systems described above. For resonators with a moderately high quality factor (5×10^4), the instability does not exceed 10^{-13} over time intervals up to 3 hours, while for systems with improved characteristics (quality factor 3×10^5, additional intensity stabilization circuits, enhanced optical isolation for prevention of reference effects), the instability decreases by an order of magnitude, reaching the fundamental limit of thermal noise.

Stabilized laser systems provide a convenient tool for the problems of laser cooling and spectroscopy of atoms and ions. Using the ULE material near the critical point not only ensures short-term stability, but also minimizes the frequency drift. If the resonator body temperature is optimal, the dominating process responsible for the change in the resonator length is the recrystallization of the material. In reality, the observed frequency drift turns out linear, with a speed of about 0.1 Hz s^{-1}, which allows predicting the laser frequency with high precision on large time scales (up to several months). Applying these systems significantly simplifies the tuning to the resonant line and is irreplaceable in those cases when it is difficult to perform the spectroscopy in the additional cell (weak transition or ion transitions). For example, a laser system with a wavelength of 689 nm is successfully applied for the secondary cooling of strontium atoms in the weak intercombination transition $^1S_0 - {}^3P_1$ at the All-Russian Research Institute for Physical-Engineering and Radio-Engineering Measurements (VNIIFTRI) in Mendeleevo, Moscow region.

Clock lasers that we have made are integrated into setups for the laser cooling of the strontium atoms (VNIIFTRI) and thulium atoms (LPI), which are used for investigations of metrological transitions at 698 nm and 1140 nm, respectively. The field of optical clocks is developing rapidly in Russia in the last few years: various research groups are developing optical frequency standards with neutral atoms in gratings (VNIIFTRI, LPI), as well as with single ions (Institute of Laser Physics SB RAS, VNIIFTRI, LPI). The characteristics of produced laser systems allow decreasing the instability of optical clocks to $10^{-16} - 10^{-17}$, which is significantly lower than the instability of primary microwave frequency standards. These laser systems will help to solve the problem of improving time–frequency provision in this country and to perform a series of fundamental investigations.

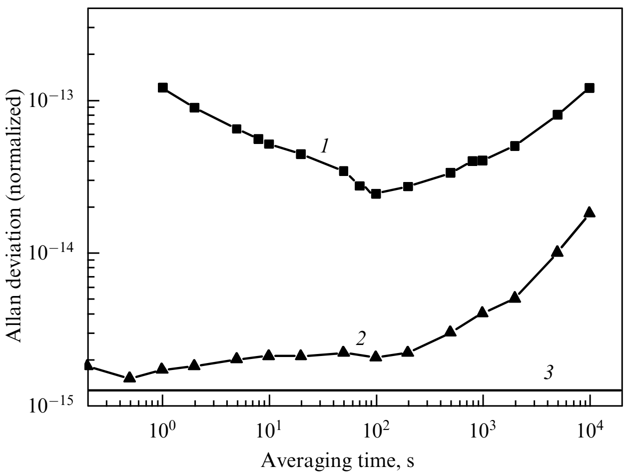

Figure 5. Typical relative instability of clock lasers (linear drift is not taken into account). Curve *1* represents the result of a comparison of two laser systems made at LPI with the standard stabilization system relative to ULE-resonators with a quality factor of 50,000 [39]. Curve *2* is the result of a comparison of two systems with additional feedback loops and a maximum ULE-resonator quality factor (300,000) [38]. Curve *3* is the theoretical estimation for the resonator thermal noise.

4. Undiscovered riddles. The proton radius problem

While performing fundamental research, a question always arises: are there any unsolved problems left in modern physics that are interesting and important, and which need joint efforts? On the cusp of the new millennium, Vitaly Lazarevich Ginzburg formulated the most important and interesting problems from his point of view [40]. Of course, the verification of the basic postulates and theories — the 'zero'-experiments — are an important part of research, which determines the application limits for theoretical assumptions. However, cases where the theory and experimental evidences do not agree are the most intriguing.

In this section, we will touch a problem from the field of laser spectroscopy of exotic atoms — the simplest atomic systems (mainly hydrogen-like), where the proton or electron are replaced by another unstable particle. Among exotic systems are positronium ($e\bar{e}$), muonium ($e\mu$), muonic hydrogen (μp), and, of course, antihydrogen. In modern particle physics, laser methods are a powerful tool for investigating simple exotic systems. These methods allow passing judgement upon the application limits for theoretical models at low energies. Although these simple systems seem to be predictable, they often puzzle scientists and reveal new unexpected properties.

The proton radius problem. With quantum-electrodynamic corrections taken into account, the energy level in a hydrogen-like system can be expanded in a power series of the small parameter α:

$$E_n = \mathrm{Ry}\left(-\frac{1}{n^2} + A_{20}\alpha^2 + A_{30}\alpha^3 + A_{31}\alpha^3 \ln\alpha + \ldots + \frac{16\pi^2 m_r^2 c^2 \alpha^2}{3n^3 h^2}\, \delta_{l0}\, r_p^2\right). \tag{5}$$

On the right-hand side of expression (5), the relativistic, radiative, polarization, and recoil corrections are grouped and represented as expansion coefficients A_{ij} with α raised to the corresponding power. Coefficients A_{ij} include the mass ratio of the particles in the system (for example, m_r/m_p in the case of hydrogen, where m_r is the reduced mass). The coefficients can be calculated with a high precision using QED methods.

Since a proton has a distributed charge, the Coulomb potential is distorted, which leads to the corresponding energy correction determined by the root-mean-square charge radius of the proton, r_p. This parameter cannot be calculated with a high precision and is determined either from spectroscopic measurements or from experiments on electron scattering by protons. Although the corresponding correction is small (1.25 MHz for the ground state), its error, being around 40 kHz, dominates when calculating the energy levels.

The measurement of r_p is one of the key aspects of determining the Rydberg constant Ry — the main scaling energy unit in atomic physics. The highest measurement accuracy for Ry was reached in spectroscopic experiments on hydrogen atoms [41]. As follows from expression (5), in order to determine Ry one needs, at least, the results of two energy measurements for different optical transitions. Indeed, the expression contains two unknown parameters Ry and r_p, while α and m_r/m_p, which are included as small corrections, are known accurately enough from other experiments. One of the transitions is traditionally chosen to be $1S_{1/2} \to 2S_{1/2}$, whose frequency has been measured with a relative error of 4.2×10^{-15} [42]. Other transitions (one- and two-photon) have been measured with an error of about 10^{-11}, and consequently they make the main contribution to the overall experimental error. The significantly lower accuracy is caused by the fact that all excited states in hydrogen, except for $2S_{1/2}$ and Rydberg states, have a short lifetime and broad width of the corresponding spectral lines. The Rydberg states, in turn, are very sensitive to the external disturbances, which makes it difficult to achieve high experimental accuracy.

In the upper part of Fig. 6, one can see the values of the proton charge radius, obtained as a result of combined frequency measurements for the $1S_{1/2} \to 2S_{1/2}$ transition and one of the transitions $2S_{1/2} \to n$P, nS, nD. Results of the $2S_{1/2} \to 2P_{1/2,\,3/2}$ Lamb shift measurements are also shown. By averaging these values, we obtain $H_{av} = 0.8758(77)$ fm. The experiments on electron–proton scattering give the value of $r_p = 0.879(8)$ fm [45], which confirms the hydrogen spectroscopy result. Based on this data, the CODATA group [43] suggested the following value of root-mean-square charge radius:

$$r_p^{\mathrm{CODATA}} = 0.8775(51)\ \mathrm{fm}\,, \tag{6}$$

and the corresponding Rydberg constant value

$$\mathrm{R}_\infty = \frac{\mathrm{Ry}}{hc} = 10{,}973{,}731.568\,539(55)\ \mathrm{m}^{-1}\,. \tag{7}$$

An experiment on the measurement of the Lamb shift in the exotic atom of muonic hydrogen was started at the beginning of the 2000s at the Paul Scherrer Institute (PSI) in Switzerland. The main motivation for the experiment was the refinement of the proton charge radius and, correspondingly, the further refinement of the Rydberg constant. Muonic hydrogen was chosen because the Bohr orbit of a heavy muon is 200 times less than that for the hydrogen atom, which results in a significantly larger overlap of the s-state wave function $\psi(r)$ with the nucleus than in an ordinary hydrogen. Correspondingly, the correction caused by the finite size of the nucleus and proportional to $|\psi(0)|^2$ will

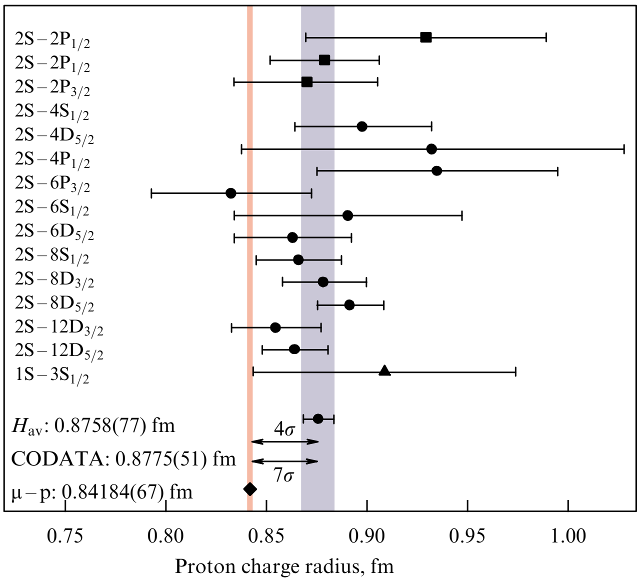

Figure 6. Mean-square values of the proton charge radius r_p, obtained in various spectroscopy experiments for the hydrogen atom. The combination of an accurately measured $1S_{1/2} \to 2S_{1/2}$ transition frequency and one of the frequencies $2S_{1/2} \to n$P, nS, nD allows one to simultaneously calculate the Rydberg constant and r_p. The figure shows the value of the averaged spectroscopic data on hydrogen, H_{av}, the value suggested by the CODATA (Committee on Data for Science and Technology) group [43] (where electron scattering data is additionally taken into account) and the value obtained from experiments on muonic hydrogen, μ–p [44]. The precision of the result obtained for muonic hydrogen is one order of magnitude higher, but its value is 4 and 7 combined standard deviations σ far from the H_{av} and CODATA values, respectively. This points to either problems of the theory that describes muonic and electronic systems or unknown experimental errors.

increase by a factor of 200^2 with respect to other contributions. At the same time, muonic hydrogen is a system which is completely analogous to ordinary hydrogen in the QED method.

The experiment at PSI has been performing for about 10 years, using the most advanced laser spectroscopy methods. It allowed detecting the transition $2S \to 2P$ ($\lambda \approx 6$ μm) in muonic hydrogen, and from QED calculations the proton charge radius value was obtained [44]:

$$r_p(\mu-p) = 0.84184(67)\ \mathrm{fm}\,. \tag{8}$$

The scientists managed to increase the accuracy of r_p by one order of magnitude, but as one can see from Fig. 6, it turned out to be seven combined standard deviations σ far from the CODATA value (6)! Even if we do not take into account the electron scattering data and limit ourselves to hydrogen spectroscopy, the difference will still be around 4σ, which points to a serious problem either in theoretical calculations or in experiment. During the last several years, many attempts have been made to explain this difference, which is now called the 'proton radius problem'. The spectrum of possible reasons extends from unaccounted systematical experimental errors to 'new' physics, which exceeds the limits of the Standard Model [46]. The problem is still unsolved and is, perhaps, one of the most interesting challenges in fundamental physics [47].

Continuing long-term scientific cooperation, researchers from LPI and the MPQ laboratory (Germany) started in 2011 an experiment on precise laser spectroscopy of hydrogen-atom excited states, in order to possibly explain the 'proton radius problem' [48]. We employ a beam of optically excited metastable hydrogen atoms at a temperature of 4 K, and time-of-flight measurements make the flying atomic groups with a velocity of up to 100 m s^{-1} to be accessible. Let us note that in all previous experiments on the spectroscopy of the $2S_{1/2} \to n\mathrm{P}, n\mathrm{S}, n\mathrm{D}$ transitions, the metastable state was excited by the electron impact, which led to hydrogen-atom characteristic velocities of about 3000 m s^{-1} and to a significant contribution from the Doppler effect. Moreover, the optical excitation allows aiming at specific magnetic and hyperfine structure sublevels, which makes the interpretation of the results easier. The aim of our experiment is to reach an error of about 1–2 kHz for the frequency measurement of the $2S_{1/2} \to 4P_{1/2,3/2}$ transitions. Such an accuracy corresponds to the accuracy of all previous combined measurements of the hydrogen atom, H_{av} (see Fig. 6). The new value will help either to confirm the CODATA value and sharpen the 'proton puzzle' or to solve it.

As is shown in Ref. [48], the assembled experimental setup allows reaching the desired statistical error, which corresponds to dividing the line into 10^4 parts (natural linewidth of the $2S_{1/2} \to 4P_{1/2}$ transition is 13 MHz). The significant suppression of the Doppler effect (by 10^5 times) and some other important systematical contributions were demonstrated. At present, the investigation of the interference effect influence on the $4P_{1/2,3/2}$-level decay is being performed. After the absolute measurement of the $2S_{1/2} \to 4P_{1/2,3/2}$ transition frequencies, the precise frequency measurements of the transitions to 6P, 8P, and 9P levels will be taken using a tried-and-true method. This will lead to a more precise determination of r_{p}.

It is important to note that some particular measurements, shown in Fig. 6, are not in strong contradiction with the results of the Lamb shift measurements for muonic hydrogen. A large difference appears only after averaging the results of over 10 experiments. Therefore, the general systematic effect in hydrogen measurements, masked by the statistical error, could have caused the observed contradiction. The significant decrease in the statistical error achieved in our experiments allows us to investigate the systematic shifts in more detail and increase the reliability of the result, which is necessary for explaining the roots of the proton charge radius problem.

5. Conclusion

In the present article, devoted to the 80th anniversary of LPI, we present the fundamental problems which are being solved by LPI researchers using precise laser spectroscopy methods. The invention of optical quantum generators — masers and lasers — opened absolutely new opportunities for highly sensitive investigations of fundamental symmetries, verification of quantum-mechanical and electrodynamical postulates and the Standard Model, and precise measurements of the fundamental constants. Laser spectroscopy is still one of the most rapidly developing fields of physics: new methods for synthesizing, preparing, and deep cooling the atomic–molecular samples are being developed, and new laser systems with advanced stabilization methods are being made. This leads to a fast increase in the measurement accuracy (by approximately one order of magnitude in three years), which opens new opportunities for performing more precise measurements. New research areas are arising, such as the laser spectroscopy of exotic atomic systems, antimatter spectroscopy, and the investigation of gravitational effects in quantum systems. The work of our Institute continues to play a key role in the development of this extremely interesting field of physics.

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References

1. Basov N G et al. *Sov. Phys. Usp.* **4** 641 (1962); *Usp. Fiz. Nauk* **75** 3 (1961)
2. Basov N G, Letokhov V S *Sov. Phys. Usp.* **11** 855 (1969); *Usp. Fiz. Nauk* **96** 585 (1968)
3. "Définition of the mètre. Résolution 1", in *17e Conf. Générale des Poids et Mesures (CGPM) 1983. Comptes Rendus* (Sèvres: Bureau Intern. des Poids et Mesures, 1984); Giacomo P *Metrologia* **20** 25 (1984)
4. Velichansky V L, Gubin M A *Phys. Usp.* **52** 1153 (2009); *Usp. Fiz. Nauk* **179** 1219 (2009)
5. Lee T D, Yang C N *Phys. Rev.* **104** 254 (1956)
6. Wu C S et al. *Phys. Rev.* **105** 1413 (1957)
7. Weinberg S *Phys. Rev. Lett.* **19** 1264 (1967)
8. Zel'dovich Ya B *Sov. Phys. JETP* **9** 682 (1959); *Zh. Eksp. Teor. Fiz.* **36** 964 (1959)
9. Alekseev V A, Zel'dovich B Ya, Sobel'man I I *Sov. Phys. Usp.* **19** 207 (1976); *Usp. Fiz. Nauk* **118** 385 (1976)
10. Moskalev A N, Ryndin R M, Khriplovich I B *Sov. Phys. Usp.* **19** 220 (1976); *Usp. Fiz. Nauk* **118** 409 (1976)
11. Barkov L M, Zolotorev M S, Khriplovich I B *Sov. Phys. Usp.* **23** 713 (1980); *Usp. Fiz. Nauk* **132** 409 (1980)
12. Bogdanov Yu V et al. *JETP Lett.* **31** 214 (1980); *Pis'ma Zh. Eksp. Teor. Fiz.* **31** 234 (1980)
13. Birich G N et al. "Optical rotation on 648 nm transition in atomic bismuth vapor", Preprint No. 136 (Moscow: Lebedev Physical Institute, 1984)
14. Fortson E N, Lewis L L *Phys. Rep.* **113** 289 (1984)
15. Amoretti M et al. (ATHENA Collab.) *Nature* **419** 456 (2002)
16. Kuroda N et al. (ASACUSA Collab.) *Nature Commun.* **5** 3089 (2014)
17. Gravitational Behaviour of Antihydrogen at Rest, http://gbar.web.cern.ch/GBAR/
18. Adelberger E G et al. *Prog. Part. Nucl. Phys.* **62** 102 (2009)
19. Rosenband T et al. *Science* **319** 1808 (2009)
20. Dirac P A M *Nature* **139** 323 (1937)
21. Uzan J-P *Rev. Mod. Phys.* **75** 403 (2003)
22. Udem Th, Holzwarth R, Hänsch T W *Nature* **416** 233 (2002)
23. Bloom B J et al. *Nature* **506** 71 (2014)
24. Kolachevsky N N *Phys. Usp.* **47** 1101 (2004); *Usp. Fiz. Nauk* **174** 1171 (2004)
25. Fischer M et al. *Lecture Notes Phys.* **648** 209 (2004)
26. Fischer M *Phys. Rev. Lett.* **92** 230802 (2004)
27. Kolachevsky N et al. *Appl. Phys. B* **89** 589 (2007)
28. Kolachevsky N N *Phys. Usp.* **54** 863 (2011); *Usp. Fiz. Nauk* **181** 896 (2011)
29. Sukachev D et al. *Phys. Rev. A* **82** 011405(R) (2010)
30. Sukachev D D et al. *Quantum Electron.* **41** 765 (2011); *Kvantovaya Elektron.* **41** 765 (2011)
31. Sukachev D D et al. *Quantum Electron.* **44** 515 (2014); *Kvantovaya Elektron.* **44** 515 (2014)
32. Riehle F *Frequency Standards: Basics and Applications* (Weinheim: Wiley-VCH, 2004); Translated into Russian: *Standarty Chastoty. Printsipy i Prilozheniya* (Moscow: Fizmatlit, 2009)
33. Katori H et al. *Phys. Rev. Lett.* **91** 173005 (2003)
34. Hinkley N et al. *Science* **341** 1215 (2013)
35. Ludlow A D et al., arXiv:1407.3493
36. Eisele Ch, Nevsky A Yu, Schiller S *Phys. Rev. Lett.* **103** 090401 (2009)
37. Kessler T et al. *Nature Photon.* **6** 687 (2012)

38. Alnis J et al. *Phys. Rev. A* **77** 053809 (2008)
39. Khabarova K Yu et al. *Quantum Electron.* **42** 1021 (2012); *Kvantovaya Elektron.* **42** 1021 (2012)
40. Ginzburg V L *Phys. Usp.* **42** 353 (1999); *Usp. Fiz. Nauk* **169** 419 (1999)
41. Biraben F *Eur. Phys. J. Special Topics* **172** 109 (2009)
42. Parthey C G et el. *Phys. Rev. Lett.* **107** 203001 (2011)
43. Mohr P J, Taylor B N, Newell D B *Rev. Mod. Phys.* **80** 633 (2008)
44. Pohl R et al. *Nature* **466** 213 (2010)
45. Bernauer J C et al. (A1 Collab.) *Phys. Rev. Lett.* **105** 242001 (2010)
46. Pohl R et al. *Annu. Rev. Nucl. Part. Sci.* **63** 175 (2013)
47. Bernauer J C, Pohl R *Sci. Am.* **310** (2) 32 (2014); *V Mire Nauki* (4) 4 (2014)
48. Beyer A et al. *Ann. Physik* **525** 671 (2013)