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Laser frequency standards at the P N Lebedev Physical Institute

V L Velichansky, M A Gubin

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Contents

Abstract. **A brief review is made of the investigations conducted at the Laboratory of Frequency Standards at the P N Lebedev Physical Institute (FIAN) Division of Quantum Radiophysics along an important line of inquiry into the development of quantum frequency standards — the elaboration of a subminiature clock based on the coherent population trapping effect in alkali metal atoms with semiconductor laser pumping (frequency stability of 10^{-11}–10^{-12} for an averaging time of $\sim 10^4$ s) and compact master oscillators/optical clocks with a frequency stability of 10^{-14}–10^{-15} (for an averaging time of 1 s) based on He–Ne/CH_4 and fiber femtosecond lasers.**

1. Introduction

One order of magnitude in the space of each ten years — such is the average rate of increase in the stability and precision of quantum frequency standards (QFSs) beginning from the 1950s. These figures are representative of the intrinsic potential of quantum radiophysics and demonstrate one of its most significant contributions to civilization.

The history of QFSs at FIAN goes back to the problem of narrowing the emission line of oscillators for radiospectroscopy. As soon as N G Basov and A M Prokhorov made the emission line a thousand-fold narrower by making an ammonia maser [1, 2], it became clear that the maser may be turned into a source of highly stable oscillations, i.e., into a frequency standard and permanently operating clock with an extremely uniform rate. The research performed at FIAN in the late 1950s–early 1960s (A N Oraevskii, G M Strakhovskii, V S Zuev, V V Nikitin, E P Markin, and some others) provided the basis for making hydrogen masers in the USSR (with a frequency of 1.4 GHz) by applied research institutes and production plants. Hydrogen masers, along with caesium beam standards, up to the present time serve as the basis for the State Time and Frequency Service, thereby securing national navigation.

The advent of lasers made it feasible to improve by 4–5 orders of magnitude the frequency stability and accuracy of frequency reproducing for a quantum transition selected as the reference, because optical frequency standards (OFSs) rely on transitions with a higher carrier frequency. The promise of optical frequency standards was substantiated even in the first review by N G Basov and V S Letokhov concerned with this research area [3]. It became possible to make use of several dozens new long-lived 'clock' quantum transitions and novel optical techniques for the highly sensitive recording of ultranarrow spectral lines arising from these transitions. Since that time, the OFS research area has been the 'engine' for quantum radiophysics itself, as well as for several branches of physics. Here are some examples: the elaboration of tunable lasers with a narrow (kilohertz) and ultranarrow (subhertz) emission spectra; the development of high-precision (in resolution and sensitivity) techniques of laser spectroscopy; ultradeep (to a temperature of 10^{-9} K) cooling and trapping of atoms and ions, and combining ultrafast (several femtoseconds long) processes with ultrastable cw oscillation regimes [4].

The last achievement of the physics of ultrafast phenomena in lasers solved one of the OFS problems and provided an efficient way of measuring optical frequencies. This enabled taking advantage of the precision potential of optical transitions and integrating different laser and microwave standards, which are spaced in frequency at several dozen or hundreds of terahertz, into a united system by means of compact laser devices [5–8].

An advantage of such integration consists in the transfer of the output parameters from one device to another, permitting the customer to approach the 'ideal' standard by selecting the needed combination of properties (relative frequency stability, spectral width, repetition, reproducibility, and precision).

Apart from the prime importance of raising the stability and accuracy in fundamental sciences (precision spectroscopy, basic physical experiments, radio astronomy relying on very-

VL Velichansky, M A Gubin P N Lebedev Physical Institute, Russian Academy of Sciences,
Leninskii prosp. 53, 119991 Moscow, Russian Federation
Tel. (7-496) 751 07 90; (7-496) 751 06 10; (7-496) 751 02 49
Fax (7-495) 938 22 51
E-mail: vlvlab@okb.lpi.troitsk.ru; gubin@sci.lebedev.ru

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long-base radio interferometers, geophysics), this research saw a significant broadening of its practical applications. The well-known satellite navigation systems GLONASS/GPS (GLObal NAvigation Satellite System/Global Positioning System) aim at forming a global coordinate–time field accessible at any point on Earth and in space for high-precision coordinate measurements, navigation, control, transmit-to-receive information timing between remote objects, etc. The key elements which govern the precision of timing of all onboard and ground-based devices are QFSs.

The diversity of 'clock' optical transitions, the methods for reference line extraction, and the requirements on the dimensions and other parameters of stable generators and clocks provide an objective basis for a broad cooperation of researchers. This paper briefly outlines a part of the research pursued in the Laboratory of Frequency Standards at the FIAN Division of Quantum Radiophysics along one of the promising avenues of research on quantum frequency standards: the development of compact and miniature optical and microwave frequency standards. It is planned to make a comprehensive review of the current status of research pursued at FIAN in this area in a separate paper.

2. Main lines of laboratory research activities

2.1 High-coherence diode lasers

Diode lasers with ordinary resonators (without external feedback) are characterized by a high level of phase noise arising from spontaneous emission. As a consequence, for relatively easy-to-meet requirements on the pump level stability the main contribution to the laser linewidth is made by quantum, rather than technical, noise. In the majority of other laser types the situation is the reverse: great pains are taken to reach the level of quantum noise. When embarking on a project aimed at the development of tunable external-cavity diode lasers (DLs), research workers from our laboratory realized that external feedback must strongly suppress the phase noise and transform the DLs to lasers of the ordinary class. This was borne out by Refs [9–11]. On establishing the critically important fact that there are no basic limitations on the external-cavity DL linewidth, the laboratory concentrated on improving the technical characteristics of tunable lasers, like the range of frequency tunability, regime and laser frequency stability, and output power [12, 13]. Special mention should be made of a series of papers concerned with the use of high-Q whispering gallery modes of quartz submillimeter spheres for capturing and narrowing the DL emission line [14, 15]. This ultimately offers possibilities of making subminiature lasers with extremely high monochromaticity of radiation. By taking advantage of all previous know-how, our laboratory elaborated a compact and reliable version of an external-cavity DL [16], which enjoys wide use in many laboratories in our country and abroad.

Simultaneously, we conducted work involving the use of elaborated DLs in high-resolution atomic spectroscopy. In studies of sub-Doppler spectra of alkali metals at low pressures, the emphasis was placed on the effect of atomic velocity-selective mechanisms for absorption saturation (pumping to an excited level, transfer of atoms to a different hyperfine sublevel, polarization and alignment at magnetic sublevels) [17, 18]. In the research on sub-Doppler atomic spectra of alkaline-earth metals (Ba, Sr, Ca), of prime concern were the metrological characteristics of resonances [19, 20]. A new sub-Doppler spectroscopic technique was developed — selective specular reflection — which enabled determining the cross section for collisional broadening at a high atomic concentration [21]. Further elaboration of this method at the University of Paris 13 allowed investigating van der Waals atom–wall interactions. Also noteworthy is the fact that Russia's first atomic trap was put into operation in the Laboratory of Frequency Standards (LFS) [22]. Among the numerous activities carried out with the use of external-cavity diode lasers in collaboration with foreign universities and the National Institute of Standards and Technology (USA), mention should be made of the work on an 'inversion-free laser' [23], which the American Physical Society put on the list of the five best contributions to physics in 1995.

2.2 Subminiature atomic clocks

High-technology near-infrared (IR) semiconductor lasers make it possible to realize the optical pumping of Cs, Rb, and K atoms, i.e., to produce nonequilibrium population distributions of the hyperfine or magnetic sublevels of the ground state. Under equilibrium conditions, these sublevels are evenly populated, because their splitting is much smaller than $k_B T$, and they can hardly absorb resonance microwave or radiofrequency (RF) fields. Optical pumping enables observing double radiooptical resonance (DROR) which is employed both in microwave standards and in magnetometers. The LFS pursued collaborative studies of the DROR in the cells with ^{87}Rb atoms [Gorky (now Nizhny Novgorod) Research Instrument-Making Institute [1]] [24] and in a rubidium atomic beam (Russian Institute of Radionavigation and Time) [25], which were aimed at the development of a microwave standard, as well as in a cell with ^{39}K (S I Vavilov State Optical Institute) [27] for applications in magnetometry.

Frequency standards and clocks based on ^{87}Rb atoms pumped with a resonance rubidium lamp with isotopic filtration have received wide acceptance. In traditional frequency standards, use is made of a double radiooptical resonance. The resonance lamp radiation transfers atoms from one hyperfine level to another (Fig. 1a). As a consequence, the absorption of light becomes weaker. The microwave field synthesized from a quartz oscillator excites a microwave cavity containing a cell with the atoms, and reverts a part of the atoms to the light-absorbing state. The dependence of resonance light absorption in the cell on the microwave frequency forms the reference resonance, which is employed to stabilize the frequency of the microwave oscillator. In order for the microwave field action on the atoms to be effective, the cell must be accommodated inside the microwave cavity. Its size is determined by the wavelength of the microwave field, thus constraining the possibility of miniaturization of the device. The wavelength of the metrological microwave transition is equal to about 3.26 cm for Cs, and to 4.4 cm for ^{87}Rb.

An alternative way of probing the microwave transition in the atomic ground state came under the scrutiny of researchers only in recent years. It is based on the effect of coherent population trapping (CPT) (Fig. 1b), which allows the volume and energy consumption to be reduced by one to

[1] The English version: Institute of Electronic Measurements KVARZ — IEM KVARZ. (*Translator's comment.*)

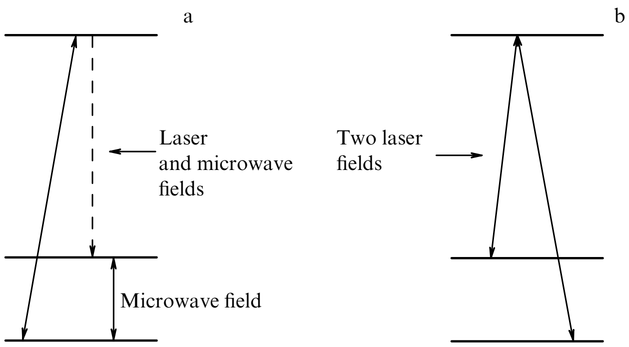

Figure 1. Traditional scheme (a) of a microwave standard and of the novel one, which makes use of coherent population trapping (CPT) resonance (b). In the case (a), atoms experience a microwave field and one optical field. The dashed line shows the spontaneous transition. In the case (b), the atoms are irradiated by two laser fields with a frequency difference determined by the microwave field.

two orders of magnitude. The CPT effect [26] is due to the formation of a coherent nonabsorbing superposition of two atomic states under bichromatic irradiation, each radiation component being tuned to resonance with one of the optical transitions and their frequency difference being equal to the splitting of the two levels involved. Stabilized in the traditional scheme is the frequency of the microwave field which interacts directly with the atomic ensemble accommodated in the microwave cavity. In the new scheme [28], the microwave field being stabilized does not act on the atoms directly but governs the frequency difference of the two optical fields which probe the atoms. It is precisely the application of CPT resonances to making frequency standards that permits the size of the standards to be radically reduced, because the necessity for a microwave cavity is obviated. Furthermore, it is significant that the possibility exists of employing compact semiconductor lasers as radiation sources. The microwave field modulates the LD current and produces the bichromatic spectrum required.

This research is being conducted in many countries (Japan, France, China, Russia, and some others) and especially vigorously in the USA — at the National Institute of Standards and Technology as well as by the companies Agilent, Kernco, Symmetricom, and Teledyne. By the order of and under the auspices of the Defense Advanced Research Projects Agency (DARPA), under development in the USA are new-generation atomic clocks with a unique combination of characteristics: a volume of the order of 1 cm^3, an energy consumption below 30 mW, and a stability of 10^{-11} over one hour. In June 2007, Honeywell Inc. reported the development of a production-ready prototype of atomic clock with the following parameters: a volume of 1.7 cm^3, an energy consumption of 57 mW, and a stability of 5×10^{-12} over one hour. Both by volume and by energy consumption this is at least an order of magnitude lower than for the highest-precision quartz oscillators, to say nothing of the higher stability.

The Laboratory of Frequency Standards, in collaboration with the Institute of Laser Physics of the Siberian Branch of the Russian Academy of Sciences (SB RAS), is engaged in work on the development of a small-sized atomic clock: the first investigation into the CPT effect employing two external-cavity DLs was carried out back in 1991 [29]. Recently, two new schemes have been proposed for the formation of high-contrast CPT resonances for atomic clocks [30, 31], the feasibility of making pure superposition atomic states with the aid of elliptically polarized bichromatic radiation was demonstrated [32], and shortened-cavity lasers were made, which lend themselves to microwave modulation in the lasing regime with single longitudinal mode [33]. The Laboratory of Frequency Standards continues the elaboration of such subminiature atomic clocks in cooperation with many institutes, because many of the applications will undoubtedly play an important role in the progress of scientific instrument making and technology. We list only some of them: noise-immune hot-start receivers of GPS and GLONASS signals with a high precision of navigation; mass applications in mobile communication systems, including broadband communications with ultrahigh-speed carrier frequency manipulation; the timing of networking computer equipment and data transmission in networks with many users; guidance, recognition, and tracking; ranging; improvement of the interference immunity of instrumentation and deployment of more stable wireless data transmission networks in field conditions, and the use of self-contained time standards when there is no way of receiving satellite signals, for instance, in an underwater environment.

2.3 He–Ne/CH_4 optical frequency standard

In 1968–1969, when studying the interaction of single-mode He–Ne laser radiation with methane molecules, J Hall, a 2005 Nobel Prize Laureate, demonstrated the extremely high resolution of nonlinear sub-Doppler spectroscopy and the potentialities of its application in time and frequency metrology [34]. This area of research began making rapid strides. In the USSR, work on these subjects commenced at the SB RAS Institute of Semiconductor Physics, FIAN, and the All-Union Research Institute of Physicotechnical and Radiotechnical Measurements [35–37]. Use was made of schemes for extracting a narrow methane spectral line, which based on lasers with single-frequency radiation. A different — 'two-mode' — method of sub-Doppler spectroscopy was elaborated at FIAN in collaboration with the Moscow Engineering Physics Institute. The high efficiency of this method was borne out in the development of high-stability compact optical frequency standards with a methane absorption cell (He–Ne/CH_4 OFS) [38–40].

There are two modifications of the two-mode method: the recording of saturated absorption resonances in mode intensities (the method of amplitude resonances) [41], and of saturated dispersion from the variation of intermode beat frequency (the method of 'frequency' resonances) [42]. In the case of amplitude resonances, the gain in signal-to-noise ratio arises from the resonance redistribution of competing mode intensities, when one of the modes experiences a resonance decrease in losses at the center of the methane line upon absorption saturation by the standing laser wave.

Recording frequency resonances offers several advantages in comparison with recording amplitude resonances in single- and two-mode lasers: the sensitivity is determined by fluctuations of spontaneous radiation, whose spectral density in the He–Ne laser is $W_e = 10^{-4} - 10^{-6}$ Hz^2/Hz, so that it is possible to record weak spectral lines with a linear absorption coefficient smaller than 10^{-11} cm^{-1} $Hz^{-1/2}$. For them, the requirements for the output laser power are relaxed and detecting does not necessitate the use of cooled photodetectors. In terms of sensitivity, this method is comparable to the method of frequency-modulation spectroscopy in an optical resonator [43] but is far simpler to implement.

Postponing until a more comprehensive paper the discussion of long-term frequency stability and frequency reproducibility of an He–Ne/CH_4 OFS, we indicate that the problem of making a master oscillator with a short-term frequency stability $(\delta\omega/\omega)_{\text{short}} = 1\times10^{-14} - 1\times10^{-15}$ (for an averaging time $\tau = 1$ s) is of great interest from the standpoint of today applications; the widely used hydrogen masers exhibit a high long-term stability $[(\delta\omega/\omega)_{\text{long}} \sim 1\times10^{-15}$ over one day], and yet do not meet present-day requirements for short-term stability: $(\delta\omega/\omega)_{\text{short}} \sim 3\times10^{-13}$ ($\tau = 1$ s).

The new-type high-stability master oscillators developed in foreign laboratories are microwave oscillators stabilized to sapphire cavities cooled to cryogenic temperatures [44] and lasers stabilized to superhigh-*Q* optical cavities [45]. Owing to the exceptionally high passive stability of the cavities and stabilization of ambient parameters, these devices exhibit a stability $(\delta\omega/\omega)_{\text{short}} = 1\times10^{-15}$ over $\tau = (0.1-10)$ s. For longer averaging time, the stability degrades due to a drift of the length of the optical cavity (structural changes in the cavity material, and residual temperature fluctuations). Cryogenic facilities are bulky, are expensive to operate, and are purely laboratory facilities.

The short-term stability of compact He–Ne/CH_4 OFSs elaborated at FIAN presently amounted to $(\delta\omega/\omega)_{\text{short}} \sim 1\times10^{-14}$ ($\tau = 1$ s), i.e., is 30–100 times higher than the short-term stability of the best radio frequency oscillators and shows promise of further improvement by a factor of 3–5 through raising the laser cavity *Q*-factor.

Oscillators with such characteristics are required in radioastronomy (as highly stable heterodyne oscillators), basic metrology (in the development of primary standards based on cold atomic 'fountains' and optical traps), experiments on elementary particle accelerators, the instrumentation of ground-based and space navigation systems, and radar stations [46].

2.4 Compact femtosecond methane optical clocks

The problem of transferring the high short-term stability of He–Ne/CH_4 OFSs to the radio frequency range and thereby making a master oscillator with a low level of phase noise became resolvable with the advent of the femtosecond technology of optical frequency division as discussed in the Introduction.

In collaboration with the Fiber Optics Research Center, RAS, and Avesta-project Ltd. set up on the basis of the Laboratory of Ultrashort Pulses at the FIAN Division of Quantum Radiophysics, the world's first highly stable compact master oscillator was realized at FIAN in 2008 — a femtosecond methane optical clock [47].

A scheme of the optical clock is presented in Fig. 2. The radiation of a two-mode He–Ne laser whose frequency is stabilized to a vibrational–rotational transition in methane ($\lambda = 3.39$ μm, $\nu = 88.4$ THz) serves as a 'pendulum' in the above clock, while the function of 'clockwork', which divides the optical frequency by a factor of 10^6, is fulfilled by a compact erbium fiber femtosecond laser ($\lambda = 1.55$ μm).

The fiber laser generates a continuous sequence of femtosecond (~ 100 fs) pulses with a repetition rate of 62 MHz, determined by the round-trip time of the laser resonator. In spectroscopic terms, this radiation is a supercontinuum — a comb of equidistant components spaced at 62 MHz, which cover the 1000–2000-nm range.

The challenge was to produce a supercontinuum spectrum with the specified properties and to transfer the stability of the

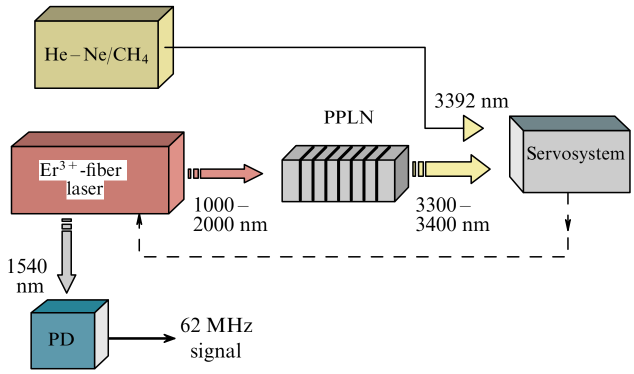

Figure 2. Schematic of a compact methane optical clock. PPLN is a crystal of periodically poled lithium niobate, and PD is a photodetector which records femtosecond laser pulses.

methane reference frequency to the repetition frequency of femtosecond pulses. To this end, the supercontinuum spectrum of the fiber laser was transferred from the 1.5-μm region to the three-micrometer spectral region by converting it in the nonlinear crystal of periodically poled lithium niobate (PPLN). Next, the frequency of one of the components of the converted spectrum was stabilized in phase to the methane reference frequency using a phase-lock loop. Since the feedback signal was fed to an element which controlled the length of the fiber laser resonator, the repetition frequency of femtosecond pulses acquired the stability of the optical reference, i.e., the 'division' of the optical frequency (88 THz) was effected by a factor of 1.4×10^6 .

In the development of the optical clock the problems arising from the extremely low power level (several picowatts) of the femtosecond comb components, the necessity of making broadband systems for the frequency control of the fiber laser, and some others were solved. Figure 3 displays the main elements of the optical clock created and the temporal instability of the output signal (for an averaging time of 1 s). One can see that the repetition-frequency fluctuations of stabilized femtosecond pulses lie in the range below 1 mHz.

A comparison [48] of the resultant signal with the signal of a commercial hydrogen maser revealed that the instability of

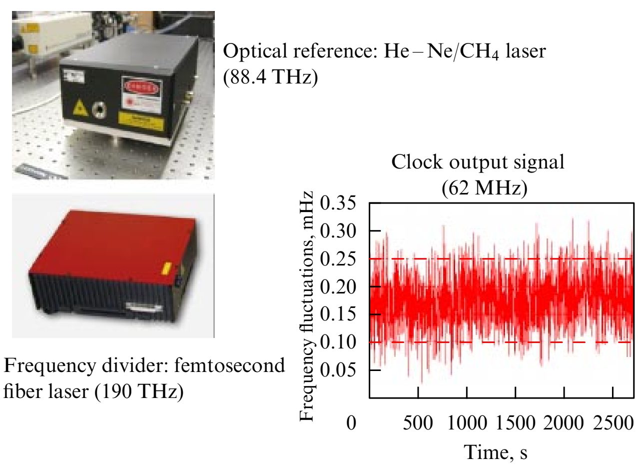

Figure 3. Main components of the compact optical clock and frequency-counter readings of the output signal (for an averaging time of 1 s). (The frequency counter is locked to a VCH-1006 hydrogen maser.)

their frequency difference amounts to 1×10^{-12} for an averaging time of 1 s and is limited by the frequency instability of the hydrogen maser. The highest frequency stability of the clock signal attained in the experiment was equal to 6×10^{-14} for an averaging time of 200 s. It is therefore valid to say that the optical and radio frequency spectral ranges were synchronized with the above precision in the experiment involved.

Elucidating the true short-term stability of the clock signal calls for another comparative system.

In the USA in 2004, FIAN, the Joint Institute for Laboratory Astrophysics (JILA) of the National Institute of Standards and Technology (USA), Colorado State University, and the Massachusetts Institute of Technology (MIT) carried out a joint experiment, in which they demonstrated for the first time the stationary version of a methane optical clock [49]. For a reference, advantage was taken of the FIAN He–Ne/CH_4 OFS (with a frequency stability of $\sim 1 \times 10^{-14}$, $\tau = 1$ s), which was delivered to JILA (Fig. 4). The repetition frequency ($f_{rep} = 78$ Hz) of femtosecond Ti:Sapphire crystal laser pulses (500–1000 nm supercontinuum spectrum) was stabilized, which was next compared with either the signal of a hydrogen maser or the repetition frequency of another Ti:Sapphire crystal laser stabilized to an iodine optical frequency standard (Nd:YAG/I_2 laser) [50]. In fact, this was the second (iodine) optical clock. The stability of the methane optical clock frequency ($f_{rep}^{CH_4}$) was found to exceed the hydrogen maser stability over averaging periods $\tau < 50$ s. Measuring the phase noise of the frequency difference of the two optical clocks ($f_{rep}^{CH_4}$ and $f_{rep}^{I_2}$) showed that their total noise spectral density scaled for a 1 GHz carrier frequency is, in the low-frequency (< 1.0 Hz) spectral domain, 30 dB lower than for the best quartz oscillators.

An experiment performed at FIAN in 2008 marked the passage from a purely stationary setup to a compact fiber laser and the realization of a new 'interface' for linking the spectra of femtosecond fiber and He–Ne/CH_4 lasers.

Although fiber lasers initially exhibit a higher noise level than Ti : Sapphire lasers, recently it was proved that they may be an aid in transferring the frequency to the radio frequency range with a precision higher than 2×10^{-15} ($\tau = 1$ s) [46]. On the other hand, they offer indisputable advantages like compactness, the ability to operate for days without human interaction, and a relatively low cost. Fiber lasers are seamlessly built into the existing optical telecommunication networks; furthermore, a start has been made on their use for transmitting reference optical frequency signals and comparing OFSs, which are spaced at several hundred kilometers, with a precision of 1×10^{-19} (!) [51].

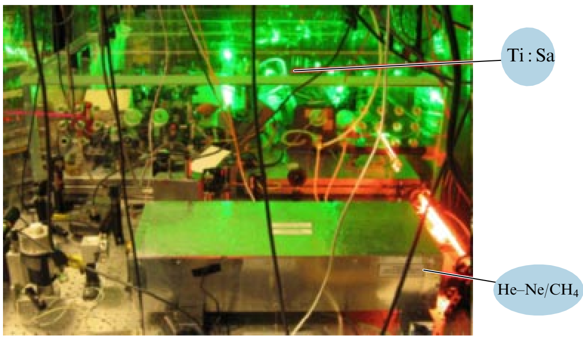

Figure 4. Stationary methane optical clock facility at JILA, Boulder, USA (2004). In the foreground is FIAN's He–Ne/CH_4 OFS. Ti:Sa is a Ti:Sapphire femtosecond laser.

3. Conclusion

Fundamental experimental and theoretical results have been obtained, which prove that in Russia in the immediate future it is possible to make semiconductor laser-based subminiature atomic clocks (with a frequency stability of $10^{-11} - 10^{-12}$ for an averaging time of $\sim 10^4$ s) and compact master oscillators/optical clocks (with a frequency stability of $10^{-14} - 10^{-15}$ for an averaging time of 1 s) on the basis of He–Ne/CH_4 and fiber lasers.

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