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Continuous-wave femtosecond lasers

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Contents

Abstract. Studies in the field of laser science and technology have resulted in the creation of a unique passively mode-locked laser generating a continuous regular train of ultrashort pulses. The unique feature of this laser is that it combines the properties of a radiation source with the output spectrum consisting of numerous extremely narrow strictly equidistant spectral lines with the properties of a radiation source emitting ultrashort high-power laser pulses. This feature has permitted a wide range of remarkable results. The basic properties of continuous-wave passively mode-locked lasers are discussed and their main applications are considered.

1. Introduction

The creation of the laser signified the advent of a completely new light source with unique emission properties determined by the high degree of coherence. The world's first laser, created by Theodore Maiman, used a ruby crystal operating in a pulsed regime [1]. This laser demonstrated high spatial coherence, providing the concentration of laser radiation within a strictly directional beam with a diffraction-limited divergence angle. Because the radiation energy and power concentrated in such a beam can be rather high, it was reported at once in many United States papers that a science fiction death ray had been discovered.

The high spatial coherence provides a rather high radiation intensity in a focused laser beam. Soon, the boring of holes in a razor blade was demonstrated, and then the laser was modified by replacing one of the resonator mirrors with a rotating totally reflecting prism (Q-switching mode), which resulted in a great increase in the output peak power and therefore in the electric field strength of the laser wave. The focusing of laser radiation in air produced an electric breakdown in the lens focus.

The first continuous-wave laser, the He–Ne laser, was developed by Javan and colleagues [2] a few months after Maiman's success. This laser had high temporal coherence. The beams from two such lasers incident on a photodetector produced a beat signal, similar to beats observed upon interference of two sound waves from slightly detuned tuning forks. The beats of radiation from two lasers produced an audio frequency. The amplified signal was fed to a dynamic loudspeaker, and a whistle was audible. This was an outstanding result, proving that laser waves behave like classical radio waves. We note that the monochromaticity of laser waves was extremely high, deviations from the laser radiation frequency ($\sim 3 \times 10^{14}$ Hz) being no more than a few kilohertz.

Continuous-wave lasers with high temporal coherence have opened new avenues in high-resolution spectroscopy. On the other hand, the possibility of generating extremely high-intensity radiation by pulsed lasers has stimulated studies in a new scientific field, nonlinear optics. As a result, two directions in the investigation of lasers have appeared. The goal of one of them was to generate ultrashort laser pulses, because the peak power and the electric field strength of a light wave increase with decreasing the pulse duration. The aim of the other was to reduce the laser linewidth and to observe sharp optical resonances with the help of nonlinear

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laser spectroscopy, which can provide a spectral resolution better than that for usual interferometry.

Both these directions were developed, in fact, independently and have yielded outstanding results. The laser pulse duration has been reduced to one optical cycle, i.e., to the fundamental limit. The pulse duration can be further reduced by nonlinear optical conversion to ~ 100 as (1 as $= 10^{-18}$ s), but now in the frequency region at the boundary between vacuum ultraviolet and soft X-rays. On the other hand, the accuracy of frequency laser measurements has reached the 18th decimal place.

Researchers engaged in pulsed laser measurements have tried reducing the laser pulse duration using Q-switching and mode-locking techniques. At the same time, the efforts of researchers in the field of high-resolution laser spectroscopy were directed toward increasing the frequency stability of continuous-wave lasers. Both these scientific communities have been working independently of each other and have not felt a strong need to keep an eye on the achievements of each other. The difference has been in the investigation methods. The high-resolution and high-precision measurements of the emission spectrum of a continuous-wave laser are absolutely necessary. On the other hand, because ultrashort pulses have a broad spectrum, the high-resolution measurement of this spectrum was unnecessary. On the contrary, the measurement of the pulse duration was an extremely challenging problem, because even the most sophisticated photodetectors could not provide the required time resolution. Such measurements required fundamentally new methods based on nonlinear optics, so-called correlation methods for measuring ultrashort laser pulses.

The shortest pulses have been generated by mode-locked lasers, and the development of mode locking techniques, allowing, in principle, the generation of single-cycle pulses, has led to the necessity of using continuous-wave multimode lasers. The beats of many modes with certain phases produce a continuous train of pulses, with the duration determined by the width of the spectrum spanning all the modes. The emission spectrum here consists of a regular train of narrow spectral lines, the modes of a continuous-wave laser. Hence, a continuous-wave laser emitting ultrashort pulses is a source of an optical frequency comb. A continuous train of ultrashort pulses can be efficiently studied by correlation methods, which can be used for measuring both the pulse duration and its shape. Spectral measurements have also become important.

Thus, studies in the field of pulsed lasers have led to the necessity of using continuous-wave lasers in this field, with their specific features and investigation techniques taken into account. An amazing merging of two principally different approaches in laser physics and technology has occurred. Researchers in the field of femtosecond lasers have secured the possibility of reducing the laser pulse duration to one optical cycle along with techniques for measuring the femtosecond pulse duration and shape. At the same time, researchers in the field of ultrahigh-resolution spectroscopy have derived a unique radiation source emitting not a single ultranarrow laser line but a comb consisting of a huge number of equidistant ultranarrow lines at optical frequencies. Great achievements in the development of laser-based precision spectroscopy, including the optical frequency comb technique, were awarded the 2005 Nobel Prize in physics (John L Hall and Theodor W Hänsch). The history of these investigations and the results obtained are very well described in the Nobel lectures by Hall and Hänsch [3, 4].

2. Passively mode-locked lasers

The principles of operation of different types of lasers are described in many monographs (see, e.g., [5]). A number of reviews and monographs are devoted to ultrashort pulse lasers, including the types of lasers considered in this review [6–9].

2.1 Q-switching technique

The world's first laser based on a ruby crystal operated in the pulsed regime, emitting a random train of ≈ 0.3 ms pulses, which were somewhat shorter than ≈ 0.5 ms pump flashlamp pulses. To control the temporal characteristics of laser emission, the Q-switching technique was proposed [10] based on the insertion of a switch into the laser cavity, which could rapidly change the transmission of light. The operation principle of this technique is as follows. First the switch is in the closed state, providing a cavity Q-factor that is smaller than the Q-factor at the lasing threshold. In the absence of lasing, the pump radiation produces a high inverse population in the active medium. When the maximum inverse population is achieved, the switch is rapidly opened, thereby increasing the cavity Q-factor to a value exceeding the threshold value. The lasing conditions are then certainly satisfied and the energy stored in the active medium is emitted during a few round trips of light between cavity mirrors. Because this energy is concentrated in one comparatively short pulse (a few dozen nanoseconds), the peak output emission power exceeds the peak power emitted in the usual regime by a few orders of magnitude, reaching a few megawatts. For this reason, pulses emitted by Q-switched lasers were called giant pulses.

2.2 Mode-locking technique

Along with Q-switching, a technique for generating laser pulses with the help of continuous-wave lasers has been proposed. This technique is based on the fact that the gain bandwidth of active media typically exceeds the intermode interval of laser cavities. Therefore, a laser usually oscillates at several modes lying within the gain bandwidth of the active medium. As a result, the laser emits at several frequencies and its emission intensity is determined by interference (beats) between radiation at these frequencies. Emission in each of the modes begins with spontaneous emission. As a result, the phases of the modes are arbitrary and the output emission is a random distribution of intensity fluctuations in time [8, 9]. The maximum intensity can be achieved by properly selecting phases. In other words, a multimode laser is a synthesizer of optical frequencies.

The radiation intensity can be controlled in accordance with the Fourier transformation, by changing phases. This was experimentally demonstrated in a continuous-wave He–Ne laser with an intracavity modulator modulating radiation at the frequency equal to the intermode interval in the cavity (active mode locking). This leads to the parametric oscillation of neighboring modes with a certain phase, which means that emission in neighboring modes develops due to parametric lasing rather than spontaneously. As a result, phase matching occurs, and, due to constructive interference, radiation is emitted as a periodic train of pulses. The period T of this train is determined by the frequency interval between modes, $T = 2L/c$, where L is the distance between cavity mirrors and c is the speed of light, while the pulse duration τ is determined by the number of generated modes, i.e., by the total width $\Delta\nu$ of the emission spectrum, $\tau \sim 1/\Delta\nu$.

The narrow spectrum of a gas He–Ne laser consists of only 5–7 modes and its width is a few hundreds MHz. But even for this spectral width, the pulse duration does not exceed a few nanoseconds, being shorter than that in Q-switched lasers (a few dozen nanoseconds). However, the peak power is very low, because it was obtained by addition of continuous frequencies of the laser, whose total power does not exceed a few mW. For this reason, actively mode-locked lasers have not been considered as high-peak power radiation sources.

2.3 Passive Q-switching in combination with passive mode-locking

Studies of Q-switched lasers have shown that a nonlinear optical effect manifested in the light intensity dependence of the transmission coefficient of a substance can be used as a fast switch. Such switches can be based on dye solutions with absorption bands overlapped with laser wavelengths. If the absorption cross section greatly exceeds the emission cross section of the active medium of a laser, a small fraction of the laser radiation is sufficient for bleaching the absorber. Thus, we have an optical switch with transmission controlled by laser radiation itself (passive Q-switching). Bleachable dye solution absorbers are now widely used due to their simplicity in Q-switched lasers.

It has been found that when some dyes (with a short relaxation time of the bleached state) are used for Q-switching in a ruby laser [12] or a neodymium glass laser [13], a saw-tooth pulse is generated (Fig. 1), rather than a smooth pulse typical of Q-switched lasers.

A pulse envelope with a shape typical of a giant pulse contained a periodic train of rather short pulses. The period of this train was equal to the round-trip transit time for light in the cavity, i.e., the same as in the case of mode locking. It became clear that in this case, passive Q-switching was combined with passive mode locking. It is important that the duration of pulses contained within the envelope was rather short (a few dozen picoseconds), because solid-state lasers have a broad amplification band. A single pulse could be separated using an electro-optical gate, and its energy could be increased in an amplifier. Thus, a passively mode-locked solid-state laser has been created and used in high-power laser systems generating picosecond pulses.

The specific features of this laser operation are described by the so-called fluctuation mechanism of generation of ultrashort pulses [14]. The generation begins from amplified spontaneous emission in all modes. Because the radiation phases of the modes are arbitrary, interference (beats) leads to a random intensity distribution in the form of intensity fluctuations. The characteristic duration of such intensity spikes (pulses) determined by the total width of the spectrum of all modes is a few dozen picoseconds. The transmission of a bleachable absorber with a short relaxation time of the bleached state depends on the radiation intensity. Such an absorber is in fact an amplitude discriminator for fluctuation pulses circulating in the laser cavity. In combination with the active-medium saturation, this finally leads to only one ultrashort pulse left on the round-trip period of light in the cavity. A computer simulation of this process is shown in Fig. 2.

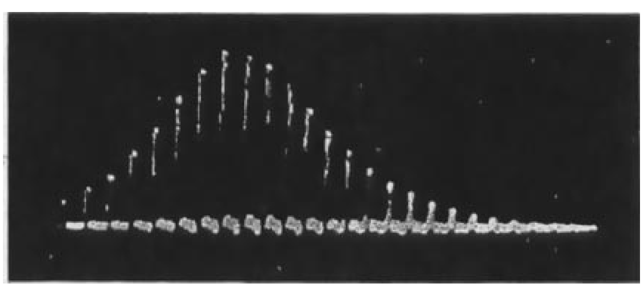

Figure 1. Shape of a pulse from a passively Q-switched and passively mode-locked laser with a saturable absorber [8].

The lifetime of the bleached state of the absorber is important. This time must be shorter than the characteristic duration of fluctuation pulses. However, the shorter this lifetime is, the higher the radiation intensity required to produce bleaching. Hence, a certain relation exists between the relaxation time of the nonlinear bleaching process and the radiation intensity inducing this process.

In practice, dyes were used with a bleached-state lifetime of about 10 ps. The output pulse duration obtained was of the same order of magnitude. The formation of an ultrashort pulse and therefore the reduction of its duration were also limited by the giant pulse duration. For these reasons, researchers have failed to obtain pulse durations shorter than 10–15 ps.

2.4 Passively mode-locked continuous-wave lasers

2.4.1 Dye lasers. The development of laser physics and technology resulted in the creation of continuous-wave gas lasers emitting a few watts in the visible range. The radiation beam from such a laser focused into a luminescent dye solution produced an inverse population in it. A continuous-wave dye laser emitting in a very broad spectral range was created. Such lasers can be tuned continuously in a broad wavelength range.

The large spectral width of continuous-wave radiation made it possible, in principle, to generate extremely short pulses in mode-locked lasers. Continuous mode-locking was achieved by using two dyes, one as an active medium and the other as a bleachable absorber [15].

The scheme of a laser setup is shown in Fig. 3. The 514.5 nm radiation from an argon laser is focused into a layer of a dye solution (Rhodamine 6G). The DODCI (3.3′-Diethyloxacarbocyanione iodide) solution is used as a bleachable absorber. These solutions are circulating in the form of plane jets freely flowing out and directed at Brewster's angle to the laser beam. The jets are located in the foci of confocal systems of concave mirrors. The radii of the mirrors are chosen so as to provide the relation between absorption and emission cross sections required for lasing. The passive mode-locking and the broad spectrum allows generating a continuous train of pulses shorter than 1 ps. Breaking through to the region of femtosecond pulses has thus been achieved. At the same time, a radiation source emitting a regular optical frequency comb has been created. However, the importance of this achievement was realized later.

2.4.2 Influence of dispersion. For very short pulses (shorter than 1 ps), the influence of the dispersion of the substance in the cavity becomes important. In considering mode locking, it was assumed that mode frequencies are equidistant. But this is the case only for an empty cavity. Mode frequencies in the cavity containing a substance are determined by their phase

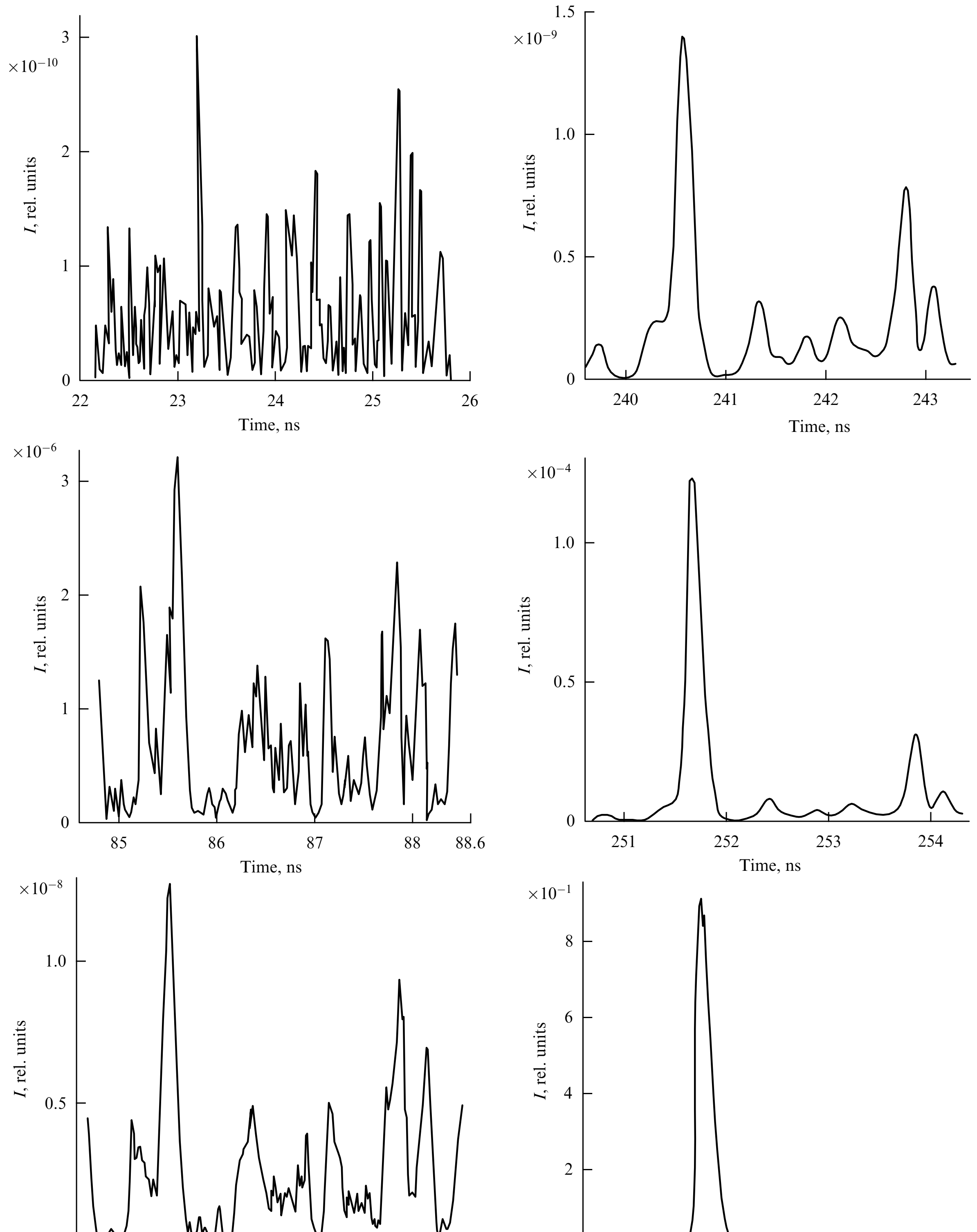

Figure 2. Computer-simulated time development of ultrashort-pulse generation in a laser with a fast saturable absorber. Shown are different stages of the time dependence of the pulse intensity [8].

velocities, i.e., the frequency (wavelength) dependence of the refractive index (dispersion). As a result, a pulse formed by such modes 'spreads' during propagation in the active medium and bleachable absorber layers due to the group velocity dispersion (GVD), and the carrier frequency changes linearly within a positive pulse. The group velocity dispersion can be compensated, in principle, by adding a layer of substance with the GVD of the same modulus but opposite sign. However, a substance with a certain anomalous dispersion required for this purpose can be absent in nature. A remarkable achievement of laser technology was the invention of optical systems providing controllable GVD of any sign.

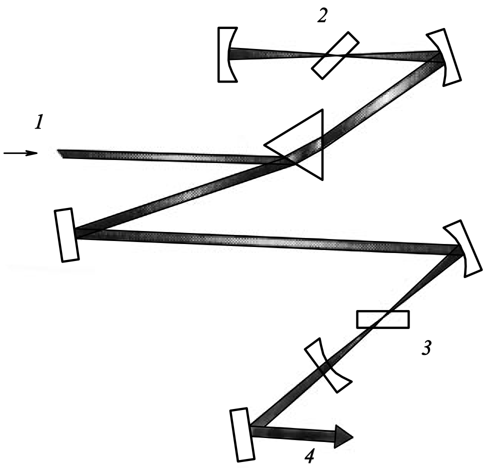

Figure 3. Continuous-wave passively mode-locked dye laser: (1) argon laser beam; (2) active medium (Rhodamine 6G) with confocal mirrors; (3) saturable absorber (DODCI) with confocal mirrors; (4) output beam [15].

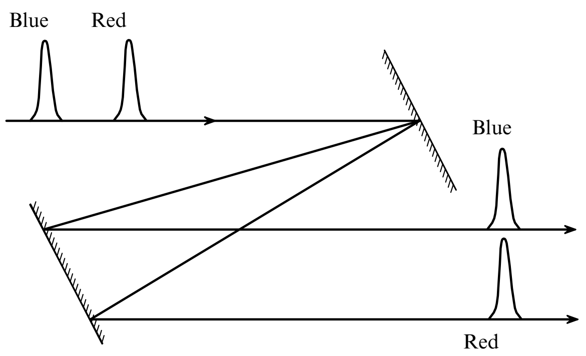

Figure 4. Pair of diffraction gratings forming an optical system with the anomalous GVD controlled by changing the distance between gratings [16].

These devices use diffraction gratings and prisms splitting laser beams into beams at different wavelengths propagating at different angles and combining them again into a parallel beam. Beams of different wavelengths then travel over different lengths, and the path difference can be controlled. The operation of these devices is similar to the action of dispersion. The principle of operation of a system of two reflecting diffraction gratings is explained in Fig. 4 [16]. This system allows controlling anomalous GVD in a broad range by changing the distance between mirrors. A disadvantage of the system is the high reflection losses ($\sim 50\%$), which strongly impedes its use in a cavity. The system with two prisms shown in Fig. 5 [17] is free of this disadvantage. The faces of the prisms are oriented at Brewster's angle, reducing losses in the beam to almost zero. Moving one of the prisms along the angle of deviation allows continuously controlling the GVD value, passing through its zero value. This system was immediately used in a continuous-wave dye laser to generate 27 fs ultrashort pulses [18].

Later, a system with multilayer dielectric mirrors was constructed [19]. The thickness and refractive index of the layers were selected to provide the reflection of radiation at different wavelengths from layers located at different depths (Fig. 6). This ensured different delays for the propagation of

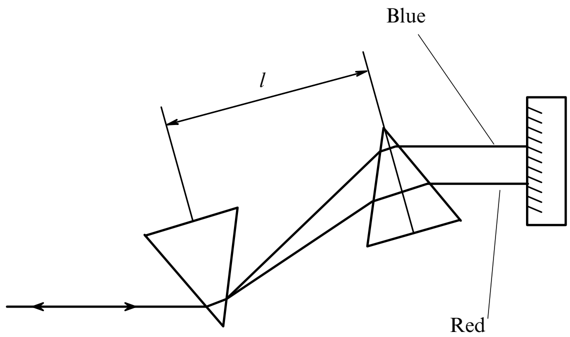

Figure 5. A prism GVD regulator for continuously changing GVD from positive to negative values by moving one of the prisms along the bisector of the deviation angle [17].

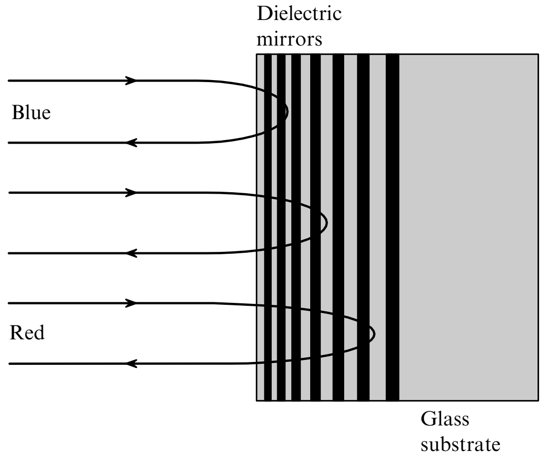

Figure 6. Scheme of a chirped mirror [19].

light at different wavelengths, resulting, as in the case of the dispersion of a substance, in an increase in the pulse duration and a change in the carrier frequency in time. This effect is called a chirp (or chirping).[1] The corresponding pulses are called chirped pulses, and mirrors having such properties are called chirped mirrors. Despite technological problems involved in manufacturing such mirrors, they are used in the cavities of the most advanced continuous-wave femtosecond lasers.

2.4.3 Solid-state lasers. Progress in the development of laser materials resulted in the discovery of a sapphire crystal doped with trivalent titanium ions ($Ti^{3+} : Al_2O_3$) [21]. This crystal has absorption and amplification bands close to those of Rhodamine 6G dye and can also be pumped by an argon laser to generate femtosecond pulses. However, the excited-state lifetime of approximately 3 s in this crystal greatly exceeds this lifetime of approximately 5 ns for Rhodamine 6G. Such a long lifetime of the excited state and the high heat conduction of sapphire can provide much higher pulse energies and

[1] The analogy between a frequency-modulated signal and the chirp of a sparrow was probably first pointed out by Rayleigh in his monograph *Theory of Sound* [20, p. 453]: "At Terling there is a flight of about 20 steps which returns an echo of a clap of the hands as a note resembling the chirp of a sparrow. In all such cases the action is exactly analogous to that of a grating in optics."

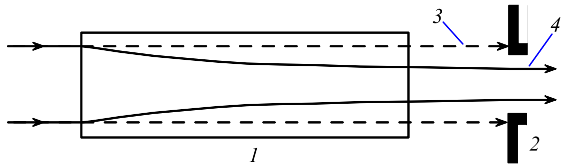

Figure 7. Kerr lens effect: (1) medium with a nonlinear refractive index; (2) aperture; (3) low-intensity radiation beam; (4) high-intensity radiation beam.

therefore higher peak powers for the same pumping. As a result, the generation of ultrashort pulses became strongly dependent on the nonlinear effects caused by the nonlinearity of the refractive index $n = n_0 + n_2 I$ (where n_0 is the usual refractive index, n_2 is the refractive coefficient due to the third-order electronic nonlinear susceptibility (nonlinear refractive index), and I is the light intensity), namely, radiation self-focusing and self-phase modulation (SPM).

Studies of the sapphire laser have led to a remarkable discovery. It was found that passive mode locking can also be achieved without an absorber [22]. Under certain conditions, a new type of a 'bleachable absorber' appears due to the self-focusing of laser radiation in the sapphire crystal itself.

Figure 7 shows how beams with different intensities propagate in a medium with a nonlinear refractive index. We can see that a lower-intensity beam propagating through an aperture is attenuated more strongly than a higher-intensity beam due to the self-focusing of the latter, i.e., the effect is the same as in the case of an absorbing dye. Because the nonlinear refractive index is caused by the optical Kerr effect, such an artificial bleachable 'absorber' was called a Kerr lens. It is important that although the self-focusing effect is rather weak, it is fast enough for generating femtosecond pulses.

Because dye jets were no longer used in this laser, its design became much simpler. For this reason, and also due to the higher output peak power compared to that of dye lasers, a Kerr-lens mode-locked (KLM) laser became a 'working horse' for numerous femtosecond studies. Figure 8 shows the typical scheme of a femtosecond solid-state KLM laser.

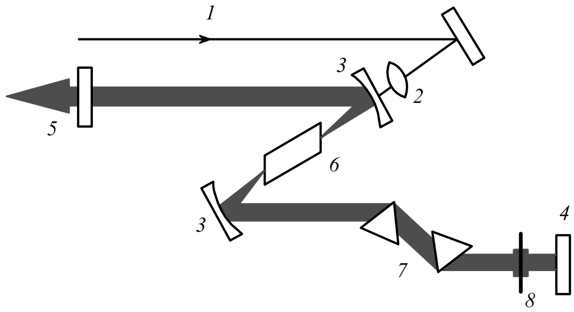

Figure 8. Typical scheme of a solid-state femtosecond laser. (*1*) pump laser beam (Ar laser or the second harmonic of a cw Nd laser); (*2*) lens focusing the pump beam into a sapphire crystal; (*3*) confocal mirrors; (*4*) highly reflecting mirror; (*5*) output mirror; (*6*) sapphire crystal; (*7*) prism GVD regulator; (*8*) aperture [9].

2.4.4 Efficient bleachable absorbers. We note that the optical Kerr effect is rather weak, and to provide the required operation of a Kerr lens, the laser cavity should be carefully adjusted, which requires high experimental skill. Without such an adjustment, the laser can oscillate, but without mode locking. But even when properly adjusted, the intensity of a fluctuation pulse should be high enough to provide the required operation of the Kerr lens. For this reason, in practice, the experimenter blocks the beam from the adjusted laser a few times until a fluctuation pulse with the required intensity accidentally appears. Only after the appearance of this pulse is the stable generation of femtosecond pulses achieved. Obviously, such a feature of laser operation is a disadvantage. The problem of laser self-firing appears, i.e., the development of a laser in which mode locking occurs simply above the pump threshold. This was achieved using bleachable absorbers operating at radiation intensities lower than in a Kerr lens.

As pointed out above, a relation exists between the relaxation time of the process and the radiation intensity inducing this process: the lower the radiation intensity is, the longer the relaxation time of the absorber. But to generate ultrashort pulses, the relaxation time of the absorber should be short. This contradiction was successfully surmounted after the invention of a special semiconducting absorber [23]. Semiconductors, like dye solutions, have broad saturable absorption bands at wavelengths determined by the band structure of the semiconductor. The relaxation of a bleached state is caused by two processes: interband relaxation with characteristic times of the order of a few nanoseconds and intraband relaxation for a few hundred femtoseconds. The radiation intensities corresponding to different absorption nonlinearities are determined by these relaxation times. Mode-locked lasing develops at comparatively low intensities corresponding to the interband relaxation time, while the final generation of femtosecond pulses is determined by the interband relaxation at high radiation intensities. Thus, unlike a dye, a semiconducting absorber can have two relaxation times of the bleached state.

Based on a semiconducting bleachable absorber, a remarkable device was built for continuous-wave ultrashort pulse lasers. A layer of a semiconducting material with required properties was incorporated into a highly reflecting multilayer mirror (Bragg reflector) consisting of layers of transparent semiconducting materials with certain refractive indices and thicknesses (Fig. 9). Lasers with such a device, which was called a SESAM (Semiconductor Saturable Absorber Mirror) [24], became an attractive alternative to KLM lasers.

A certain drawback of the SESAM design is the complexity of its manufacturing technology. Recently, remarkable results were obtained using bleachable absorbers of other types for femtosecond lasers. These absorbers, based on carbon structures, single-wall carbon nanotubes (SWCNTs), and graphene, having the properties of semiconductors used in SESAMs, were successfully used in femtosecond lasers. The manufacturing technology of laser devices based on carbon structures proved to be simpler, which stimulated the appearance of many successful works. Single-wall carbon nanotubes were used for the first time in femtosecond lasers in [25, 26]. Recently, graphene layers were used as a bleachable absorber [27, 28]. A remarkable property of graphene is the weak wavelength dependence of its absorption. This property and graphene's much greater accessibility than that of SESAMs and even SWCNTs makes graphene a quite promising material for the improvement of femtosecond lasers.

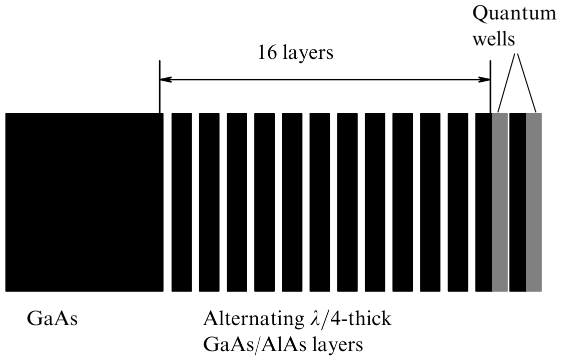

Figure 9. Semiconductor saturable absorber mirror (SESAM) [24].

We note that bleachable absorbers, such as SESAMs, SWCNTs, and graphene-based ones, are currently also used in lasers of a new type, femtosecond fiber lasers.

2.4.5 Fiber lasers. The development of new optical devices, single-mode optical fibers, is a remarkable achievement of science and technology. The advent of optical fibers not only initiated a real revolution in the development of communication links capable of transmitting huge amounts of information but also led to the development of lasers of a new type, in particular, femtosecond fiber lasers. Studies of nonlinear optical phenomena in single-mode optical fibers have made a great contribution to laser physics and technology [28].

An optical fiber (Fig. 10) consists of a core and a cladding made of transparent materials (glasses of different types). The refractive index of the cladding material is somewhat smaller than that of the fiber core, and therefore light propagating in the core undergoes total internal reflection at the core–cladding interface. This is the main property of an optical fiber. For certain relations between the refractive indices of the fiber core and the cladding and for certain core diameters, only a light wave in single transverse mode propagates in the fiber core. Such optical fibers are called single-mode.

Due to outstanding technological achievements in the manufacturing of optical fibers, radiation losses during the propagation of light in fibers can be extremely low. This achievement was awarded the Nobel Prize in physics in 2009 [30]. Fiber-optic communication lines literally wind around Earth and have become one of the most important parts of human civilization.

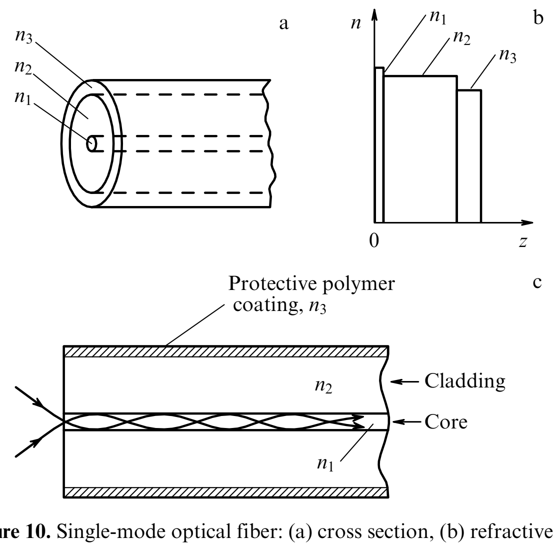

Figure 10. Single-mode optical fiber: (a) cross section, (b) refractive index profile, and (c) propagation of radiation in the optical fiber.

The core glass can be doped with rare-earth ions, which can be excited by efficient diode lasers. Thus, an optical fiber becomes an active laser medium.

We note that apart from fibers themselves, fiber lasers use other important outputs of fiber-optic technologies: collimators for coupling laser radiation to fibers, wavelength-division multiplexing (WDM) couplers for coupling radiation at one wavelength to a fiber and decoupling it from the fiber at another wavelength, fiber Bragg gratings used as cavity mirrors, and Faraday rotators providing unidirectional propagation of light in fibers. All this has resulted in the development of fiber lasers offering many advantages, such as:

(i) High efficiency due to pumping by efficient laser diodes;

(ii) low sensitivity to external mechanical perturbations, because radiation is concentrated in the fiber core;

(iii) no need for a special cooling system because the heat released upon pumping is scattered in the fiber cladding; and

(iv) compactness and a considerably lower cost compared to that of usual solid-state lasers.

For this reason, fiber lasers have begun to play a very important role in laser studies.

The laser cavity can have two configurations: a linear configuration of the Fabry–Perot type and a ring configuration. Both are usually realized by properly selecting mirrors. A remarkable specific feature of a fiber laser is that its ring configuration can be easily realized by simply connecting the fiber into a ring. A ring laser is preferable in a number of applications, because it is less subjected to the influence of the back reflected output laser radiation.

As noted in Section 2.4.3, solid-state lasers use a Kerr lens based on the nonlinear refractive index as a bleachable absorber. The nonlinearity of the refractive index can also be used in fiber lasers, but in a different way. The single-mode propagation of light excludes the self-focusing effect. For large propagation lengths of light in a fiber, birefringence appears in the fiber core, resulting in the elliptic polarization of light. Because the refractive index depends on the radiation intensity, the birefringence magnitude also depends on it. This leads to the intensity dependence of light transmission, i.e., to nonlinear absorption. Figure 11 shows the principal scheme of the self-amplitude modulation obtained due to a nonlinear rotation of the polarization ellipse [31].

As a birefringent plate, a fiber itself is used. It is well known that birefringence appears in isotropic media due to mechanical stresses. To obtain the required effect, a fiber is purposely bent (this device is called a polarization controller).

As a polarizer, a Faraday rotator is commonly used (a fiber Faraday isolator), which is introduced into a ring cavity to provide unidirectional lasing. Thus, a fast bleachable absorber and passive mode locking can be realized in a fiber.

Because light propagates over a long distance in the fiber glass, the influence of dispersion becomes very important. Fiber optics offers specific techniques for controlling the group velocity dispersion. The core of a single-mode fiber is a glass waveguide for an electromagnetic light wave. This means that the waveguide has both the material and waveguide dispersion, which depend on the wavelength differently in general. As a result, the GVD of the fiber depends not only on the glass dispersion but also on the waveguide configuration.

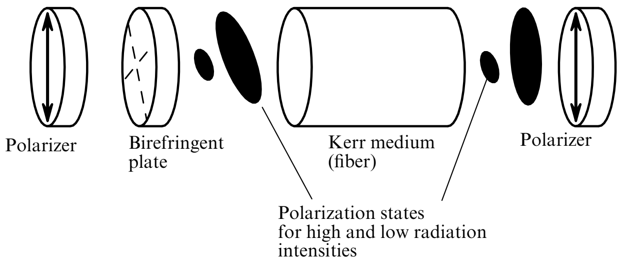

Figure 11. Principal scheme for obtaining the amplitude self-phase modulation (bleachable absorber) in an optical fiber due to the nonlinear rotation of the polarization ellipse [31].

Dispersion in fibers is usually described by expanding the mode propagation constant β into a Taylor series in the vicinity of the carrier frequency ω_0:

$$\beta(\omega)=n(\omega)\,\frac{\omega}{c}=\beta_0+\beta_1(\omega-\omega_0)+\frac{1}{2}\,\beta_2(\omega-\omega_0)^2+\ldots, \tag{1}$$

where β_1 determines the group velocity of the pulse envelope and β_2 determines the GVD and is responsible for a change in the pulse shape during its propagation. A remarkable feature of widespread single-mode silica fibers is that as the wavelength increases, the parameter β_2 and therefore GVD vanish (at a wavelength of 1.27 μm) and then acquire increasingly negative values (Fig. 12). Because the total GVD depends both on the material dispersion and on the dispersion determined by the waveguide properties of the fiber, it is possible to vary the GVD by changing the fiber structure affecting the waveguide propagation of light. A special technology allows manufacturing microstructured fibers with through holes arranged as shown in Fig. 13. These are so-called holey fibers or photonic-crystal fibers [32, 33]. A certain arrangement of the holes gives a certain GVD value, in particular, the zero of GVD can be displaced to the blue spectral range by changing the arrangement of the holes. Because fibers can have both positive and negative GVD, the GVD value can be controlled by selecting fiber pieces with different lengths and different CVD values.

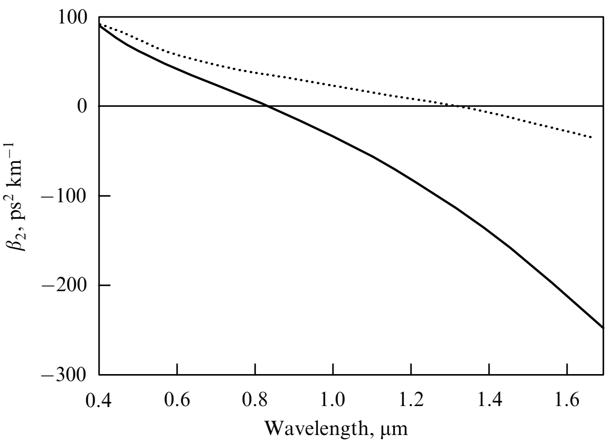

Figure 12. Wavelength dependence of the dispersion parameter β_2 for a glass rod in air (solid curve) and a standard single-mode fiber used in fiberoptic communication lines (dotted curve).

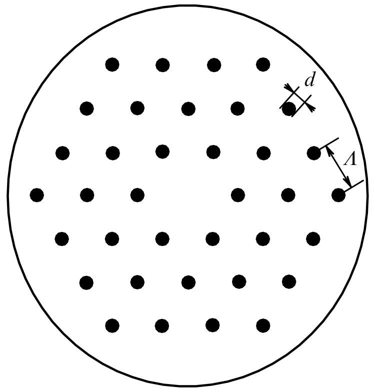

Figure 13. Cross section of a holey photonic-crystal fiber: d is the diameter of an air channel; Λ is the distance between channels [6].

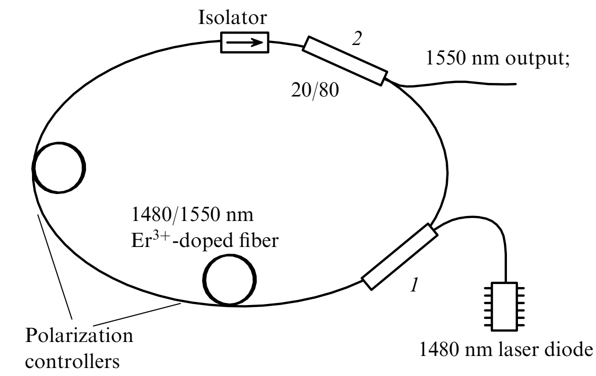

Figure 14. Typical scheme of a femtosecond fiber laser: (*1*) wavelength-division multiplexer (WDM) for pump and laser radiation; (*2*) optimal WDM radiation outcoupler [9].

Figure 14 shows the typical scheme of a ring femtosecond fiber laser. Along with ring lasers, a fiber femtosecond laser with a linear Fabry–Perot cavity containing a highly reflecting SESAM can also be used [34].

2.4.6 Theoretical description of a continuous-wave femtosecond laser. Progress in the development and advancement of continuous-wave femtosecond lasers is largely due to the detailed studies of physical processes underlying the operation of these lasers. The mechanism of generation of ultrashort pulses in passively mode-locked lasers was established in experimental and theoretical studies. It is not simply the separation of a single fluctuation pulse from multimode radiation that occurs with the help of a fast bleachable absorber, but an ultrashort pulse is also formed due to the nonlinearity of the refractive index of the medium inside the laser cavity. The continuous-wave regime facilitates the construction of an analytic model of the lasing process.

It was shown in [35] that under certain conditions, the propagation of a radiation pulse in a medium with dispersion and an inertialess nonlinearity of the refractive index can lead to the formation of an optical soliton. The propagation of a pulse in a medium with dispersion and nonlinearity is described by the nonlinear Schrödinger equation. The medium is assumed to be infinite. In the laser cavity, such a medium has a finite length. During the propagation of light in the medium and due to the transmission of cavity mirrors, losses occur. These losses are compensated by amplification in the active medium upon pumping. Therefore, the equation describing the laser operation in the continuous-wave regime

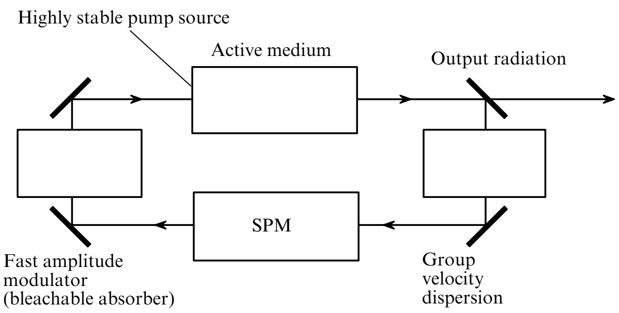

Figure 15. Scheme of a ring resonator with amplification, dispersion, a saturable absorber, SPM, and losses [38].

should be supplemented with the terms taking losses and amplification into account. A passively mode-locked laser has been theoretically studied in a number of papers (see, i.e., [36, 37]).

In [37], an equation describing the laser operation was obtained by considering a ring laser model (Fig. 15) containing successively arranged elements acting on a pulse: losses and the phase shift, amplification, GVD, SPM, and saturable absorption. A small change Δa in the complex amplitude $a = a(t)$ of the envelope of the laser-wave electric field with the carrier frequency ω_0 after the propagation of light through an element introducing amplitude losses l and the phase shift ik has the form

$$\Delta a = -(l + \mathrm{i}\kappa)\, a \tag{2}$$

(the intensity loss is $2l|a|^2$). For a small gain g in the corresponding element,

$$\Delta a = g\left(1 + \frac{1}{\Omega_g^2}\,\frac{\mathrm{d}^2}{\mathrm{d}t^2}\right) a\,, \tag{3}$$

where Ω_g^2 is the amplification bandwidth. The element responsible for the GVD effect produces the change

$$\Delta a = \mathrm{i}D\,\frac{\mathrm{d}^2}{\mathrm{d}t^2}\, a\,, \tag{4}$$

where D is the dispersion value, $D = (1/2)\,\beta_2 L$ for a fiber with length L.

The SPM element causes a phase shift proportional to $|a|^2$:

$$\Delta a = -\mathrm{i}\delta|a|^2 a \tag{5}$$

(for a fiber of length L with the nonlinear coefficient n_2, $\delta = (\omega_0/c)(n_2 L/s)$, where s is the core cross-sectional area). The action of the saturable absorber is expressed as

$$\Delta a = \gamma|a|^2 a\,, \tag{6}$$

where the coefficient γ is inversely proportional to the saturation intensity.

Because the laser operates in a continuous-wave stationary regime, a change in the pulse after a round trip in the cavity should be zero. This yields the equation describing the operation of a passively mode-locked continuous-wave laser:

$$\left[-(l + \mathrm{i}\kappa) + g\left(1 + \frac{1}{\Omega_g^2}\,\frac{\mathrm{d}^2}{\mathrm{d}t^2}\right) + \mathrm{i}D\,\frac{\mathrm{d}^2}{\mathrm{d}t^2} + (\gamma - \mathrm{i}\delta)\,|a|^2\right] a = 0\,. \tag{7}$$

In studies of ultrashort-pulse lasers [38, 39], Eqn (7) and its modifications are usually called the generalized Ginzburg–Landau equations. The solutions of Eqn (7) determine the characteristics of pulses for the specified initial parameters of a laser setup. For a negative GVD, typical of 1550 nm erbium-doped fiber lasers, the solution has the form

$$a = A_0\,\mathrm{sech}\left(\frac{t}{\tau}\right)^{1+\mathrm{i}\beta}, \tag{8}$$

where τ is the pulse FWHM and β is the chirp parameter in the pulse. For $\beta = 0$, a soliton with a minimum duration is formed. We note that a theoretical study is especially important in the case of fiber lasers, whose design does not allow controlling the GVD. But the optimal length of a fiber piece with the required dispersion can also be selected based on a theoretical analysis.

From the standpoint of the theoretical model, the generation of femtosecond pulses occurs as follows. A bleachable absorber causes the formation of a single pulse from fluctuation intensity spikes of amplified spontaneous emission. In other words, a fast bleachable absorber triggers passive mode locking. A stable pump source maintains stationary lasing. The pulse shape and duration are determined by a soliton that is formed due to the balance between SPM and GVD.

It is important to note that SPM causes spectral broadening. As a result, the pulse duration can become shorter than that corresponding to the amplification bandwidth of the active medium and, under certain conditions, can even reach the extremely small value close to the laser wave cycle.

3. Optical frequency combs

An important feature of continuous-wave ultrashort-pulse lasers is their specific emission spectrum. Because a continuous train of ultrashort pulses is obtained due to summation of many modes, the emission spectrum consists of narrow spectral lines, as noted above. The width of each of the spectral lines is determined by the lasing stability and can be extremely small. The interval between the lines is determined by the pulse repetition rate, i.e., it depends on the optical length of the laser cavity. The total number of lines depends, in turn, on the width of the emission spectrum, which cannot be smaller than $1/\tau$ (where τ is the ultrashort pulse duration). Thus, a continuous-wave passively mode-locked laser emits an optical frequency comb and can be used in two totally different contexts, as shown in Fig. 16. Using an optical gate, we can separate a single ultrashort pulse from a pulse train, thereby obtaining a source of ultrashort pulses with a continuous spectrum corresponding to an ultrashort pulse duration. These pulses can be amplified and subjected to various nonlinear transformations. On the other hand, we can separate a single line with a monochromator and use the laser as a source of highly monochromatic radiation.

Figure 17 shows a train of ultrashort pulses and the corresponding spectrum obtained by the Fourier transformation [40]. This spectrum exhibits equidistant spectral lines separated by the pulse repetition rate f_{repet}, forming an optical frequency comb. But the frequencies of the lines are not multiples of each other. We can see from the extrapolation of the frequency comb to zero in Fig. 17b that there is an offset frequency that is smaller than the mode separation f_{repet}. According to the Fourier transformation, this carrier offset

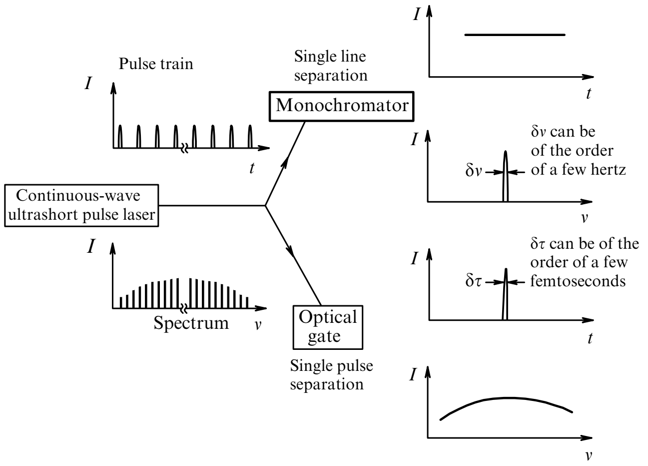

Figure 16. Fundamental specific features of a continuous-wave ultrashort pulse laser [8].

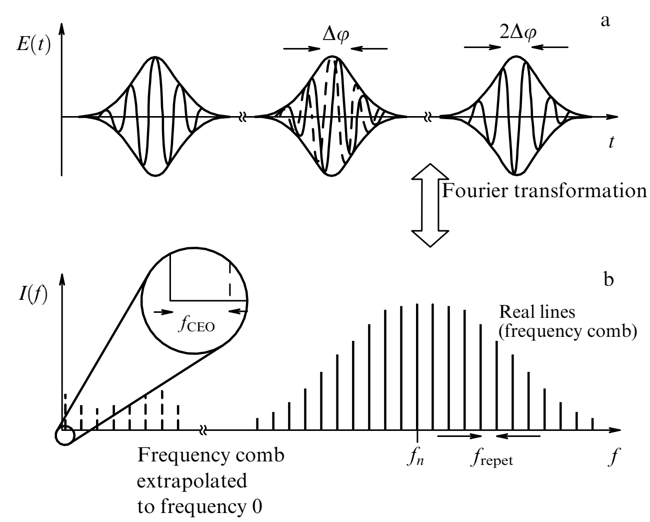

Figure 17. (a) Three consecutive femtosecond pulses in a pulse train; $\Delta\varphi$ is the phase shift of the carrier wave with respect to the envelope. (b) Shape of the corresponding emission spectrum (for clarity, is a small number of lines is shown, whereas their actual number can reach 1 million) [40].

envelope frequency f_{CEO} is determined by the phase shift $\Delta\varphi$ of the carrier wave with respect to the envelope: $f_{\text{CEO}} = (\Delta\varphi/2\pi)\, f_{\text{repet}}$. This shift appears because the envelope and the carrier wave propagate in the laser cavity with different velocities: the envelope moves with the group velocity, while the carrier wave moves with the phase velocity. These velocities are different because the active medium in the laser cavity has dispersion.

3.1 Principal possibility of direct optical frequency measurement

The frequency f_n of each of the lines can be expressed as $f_n = nf_{\text{repet}} + f_{\text{CEO}}$, where n is an integer. It is remarkable that frequencies f_{repet} and f_{CEO} lie in the radio frequency range. They can be directly measured with high accuracy by comparison with a microwave standard second using standard electronic methods. Knowing these frequencies allows finding the optical frequency of a line in the frequency comb.

To determine n, it is sufficient to measure the wavelength with an accuracy better than the mode separation in the frequency comb. This can be readily performed by standard spectroscopic methods. Knowing the speed of light, we obtain the frequency value close to the frequency f_n, which allows finding n. The frequency f_{repet} can be controlled using a fast photodiode and a piezoelectric transducer with a feedback circuit controlling the laser cavity length.

Finding f_{CEO} is more complicated. Because this frequency depends on the dispersion of the medium in the cavity, it can be controlled by changing the intracavity dispersion. This requires a rather small but quite definite change in the dispersion value. If a prism GVD controller is available, it is also used to control f_{CEO}. In the absence of a GVD controller, the intracavity dispersion can be controlled by slightly varying the pump power, because the dispersion value depends on the inverse population of the active medium.

Precise measurements of f_{CEO} are performed by the so-called f–$2f$ interferometer method (Fig. 18) based on comparing line frequencies in the long-wavelength (f^{red}) and short-wavelength (f^{blue}) regions of a frequency comb spanning more than an octave, i.e., $f^{\text{blue}} > 2f^{\text{red}}$. The frequencies f^{red} and f^{blue} are $f^{\text{red}} = nf_{\text{repet}} + f_{\text{CEO}}$ and $f^{\text{blue}} = 2nf_{\text{repet}} + f_{\text{CEO}}$. Radiation beams at the frequencies $2f^{\text{red}} = 2(nf_{\text{repet}} + f_{\text{CEO}})$ (second harmonic generation) and f^{blue} incident on a photodetector produce the difference signal

$$
\begin{aligned}
f_{\text{diff}} &= 2f^{\text{red}} - f^{\text{blue}} \\
&= 2nf_{\text{repet}} + 2f_{\text{CEO}} - (2nf_{\text{repet}} + f_{\text{CEO}}) = f_{\text{CEO}}\,,
\end{aligned}
$$

i.e., the required frequency is obtained. The f–$2f$ interferometer is in fact a Mach–Zehnder interferometer containing an SHG crystal in one of its arms. The output signal from the photodetector is fed to a radio-frequency spectrum analyzer recording frequencies f_{repet} and f_{CEO} (Fig. 19) [41]. These frequencies can be compared with the microwave frequency standard (standard second), which opens up the principal

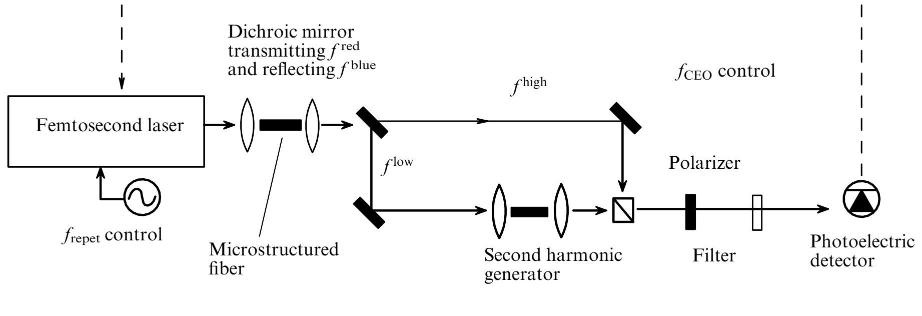

Figure 18. Scheme for measuring the f_{CEO} frequency [40].

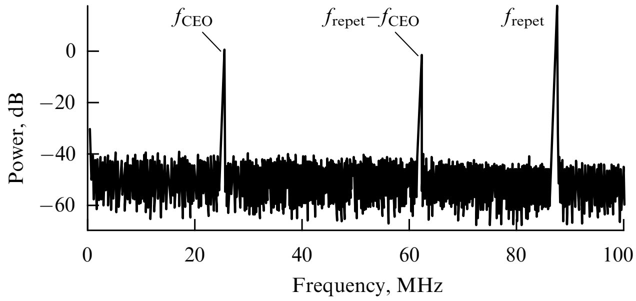

Figure 19. Signals in a radio frequency spectrum analyzer [41].

possibility of direct precise measurements of optical frequencies. However, this method can be used only for an octave-spanning comb: the doubled frequency of the low-frequency edge of the comb should not exceed the frequency of its high-frequency edge.

3.2 Spectral broadening of a frequency comb

The required width of a frequency comb, which should be no less than one octave, means that the duration of ultrashort pulses emitted by a continuous-wave laser should be close to one optical cycle. This is rather difficult to achieve. Usually, pulse durations are of the order of a few dozen optical cycles, corresponding to the spectral width of laser pulses a few times smaller than an octave. To use the method described in Section 3.1, the emission spectrum of a laser should be broadened, preserving the frequency comb. This was achieved using the nonlinear effect of supercontinuum generation.

The key factor of the supercontinuum generation effect is the dependence of the refractive index of a transparent medium on the laser radiation intensity. As noted in Section 2.4.3, $n = n_0 + n_2 I(t)$, where n_0 is the refractive index in the absence of radiation, $n_2 = (2\pi/n_0)^2 \chi^{(3)}(\omega)$ is the nonlinear refractive index at the frequency ω, $\chi^{(3)}(\omega)$ in the third-order nonlinear susceptibility of the medium, and $I(t)$ is the laser radiation intensity. The change in the laser radiation intensity in time causes SPM. The change in the phase after the propagation of radiation over a distance L in the medium is given by

$$\varphi(t) = \frac{\omega}{c}\, n_2 I(t)\, L\,. \tag{9}$$

The change in the frequency resulting in the broadening of the emission spectrum is

$$\Delta\omega(t) = \frac{\omega}{c}\, n_2 \frac{\mathrm{d}I}{\mathrm{d}t}\, L\,. \tag{10}$$

We can see that the maximum spectral broadening depends on the time dependence of the radiation intensity and increases with laser pulse shortening and upon increasing the propagation length of light in a nonlinear medium.

Supercontinuum generation was originally achieved by focusing a single ultrashort pulse on a nonlinear medium. As a result, emission with a very broad continuous spectrum was produced; this is why the effect was called the supercontinuum generation. To obtain a spectrally broadened frequency comb, a supercontinuum should be generated by a continuous train of ultrashort pulses. Emissions of each pulse are then added coherently and the resulting emission spectrum represents a frequency comb. However, the peak intensity

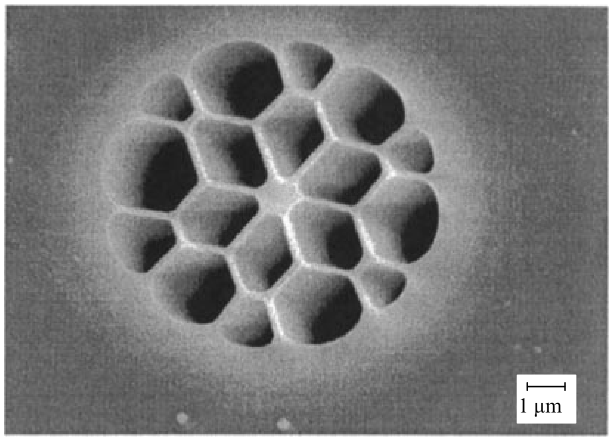

Figure 20. Microstructured fiber used for supercontinuum generation [42].

may not be high enough to produce the required spectral broadening, even upon focusing, and it is necessary to increase the length of interaction of intense radiation with a nonlinear medium. Radiation can be focused onto a single-mode fiber in which it can propagate over a long distance, interacting with the core glass. But because of dispersion, the increase in the pulse duration is accompanied by a decrease in its intensity, and the required result cannot be obtained. Therefore, microstructured fibers mentioned in Section 2.4.5 (Fig. 13), with the GVD shifted to zero at the laser wavelength, are used [32, 332]. These fibers provide, besides a reduction in dispersion effects, a concentration of radiation in a small-diameter core.

The example of a microstructured fiber used for broadening the frequency comb emitted by a cw femtosecond laser is shown in Fig. 20 [41]. To increase the radiation intensity, the core diameter is reduced, producing a so-called suspended-core fiber, whose core is suspended with very thin bands inside the fiber. This provides the greater widths of a frequency comb at the lower average power of femtosecond lasers. As a result, efficient supercontinuum generation can be achieved upon irradiation by cw femtosecond lasers. Figure 21 shows the spectrum of a femtosecond Ti:sapphire laser broadened in a microstructured photonic-crystal fiber.

We note that along with the SPM effect, supercontinuum generation also involves other nonlinear effects violating the regular structure of the frequency comb. This is manifested in the spectrum in Fig. 21. We can see that the supercontinuum spectrum is irregular. Studies have shown that the shorter the pulse duration is, the weaker the influence of side effects deteriorating the characteristics of frequency combs. For

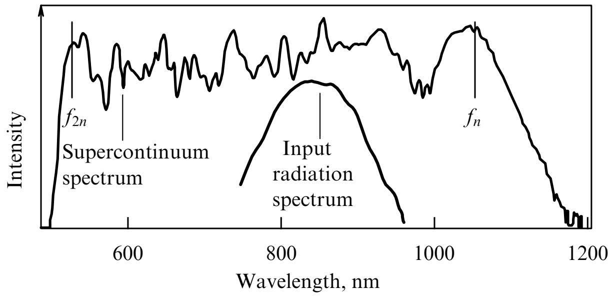

Figure 21. Spectral broadening during supercontinuum generation in a microstructured fiber [40].

precision measurements, femtosecond frequency combs are therefore preferable to picosecond ones, although the design of picosecond lasers is much simpler.

To summarize, a cw femtosecond laser, together with a supercontinuum generator and an $f-2f$ interferometer, is a generator or a synthesizer of frequency combs with well-known radio frequencies. Such a device operates like a few hundred thousand extremely stable, exactly aligned, simultaneously operating lasers emitting very narrow spectral lines. This remarkable feature of frequency combs underlies their numerous applications, especially in the field of precision metrology [3, 4].

4. Applications of optical frequency combs

Laser frequency combs synthesizing optical frequencies in the form of narrow spectral lines with direct measurement and extremely high accuracy have found important applications, especially in spectroscopy, where they have initiated a real revolution. Some applications of femtosecond laser frequency combs are considered in Sections 4.1–4.6.

4.1 Shaping of single-cycle pulses

As the laser pulse duration is reduced, the phase shift $\Delta\varphi$ between the carrier frequency and the envelope becomes important because it determines the pulse shape and duration. As the pulse duration approaches one optical cycle of laser radiation, the time dependence of the electromagnetic-wave field strength should be taken into account, because this field strength determines the action of laser radiation.

The electric field of a laser wave is represented in the form $E(t) = E_0(t)\cos(\omega_0 t + \Delta\varphi)$. The influence of the phase on the electric field strength is illustrated by the shape (envelope) of single-cycle pulses and the change in the electric field strength when changing the phase shift of the carrier frequency with respect to the envelope maximum. When the phase shift is $\pi/2$ (the envelope is superimposed on a sinusoid), the electric field does not attain its possible maximum (Fig. 22a). For a zero phase shift (the envelope is superimposed on a cosinusoid), the electric field reaches its maximum (Fig. 22b).

Hence, the electric field strength of a laser wave can have different maximum values for the same envelopes (pulse durations). This difference is manifested in the interaction of high-power femtosecond pulses with matter, in particular, upon generation of higher harmonics, which can lead to the emission of attosecond pulses.

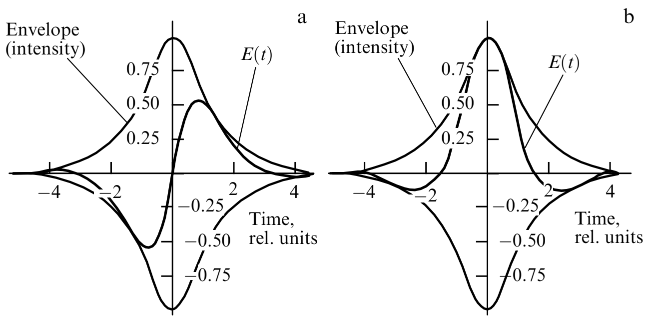

Figure 22. Computer-calculated envelopes and time dependences of the electric field (in rel. units) for single-cycle pulses. The envelope maximum coincides with (a) the zero amplitude of the carrier frequency and (b) the maximum amplitude of the carrier frequency [9].

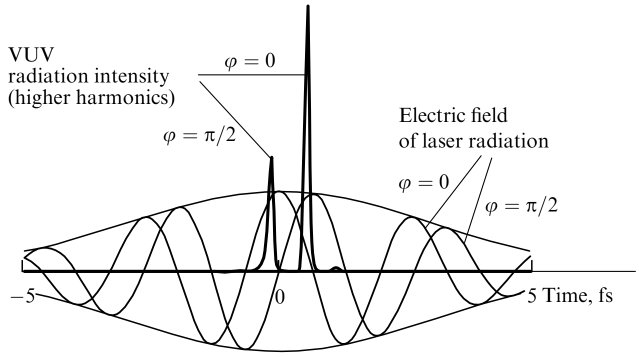

Figure 23. Higher harmonic radiation intensities calculated for the interaction of 5 fs pulses with different phases with a helium jet [41].

The value of $\Delta\varphi$ determining the pulse shape can be controlled with an $f-2f$ interferometer (see Section 3.1). The shape of high-power 6 fs pulses from a Ti : sapphire laser was controlled in [41]. The authors of [41] have demonstrated the influence of this control on higher harmonic generation upon interaction of laser radiation with a helium jet. Figure 23 shows the intensities of VUV higher harmonics calculated for different $\Delta\varphi$. We can see that this intensity in the VUV region reaches a maximum for the zero phase shift.

The generation of high-power nearly single-cycle laser pulses with a controllable shape has led to remarkable results in laser physics and nonlinear optics. In particular, attosecond pulses were generated in the VUV and soft X-ray regions [43], and the possibility of generating a regular VUV frequency comb was demonstrated [44].

4.2 High-power optical frequency comb generators

Many spectroscopic applications of optical frequency comb techniques require frequency combs spanning a broad spectral range, from the mid-IR to the VUV region. To generate such combs, high-average power cw femtosecond lasers are required. Femtosecond fiber lasers are currently the key component of frequency comb generators. As noted in Section 2.4.6, Er-doped fiber lasers are based on the generation of optical solitons that occurs due to a balance between phase contributions caused by the anomalous GVD ($\beta_2 < 0$) and SPM. By definition, a soliton has a particular, well-defined shape, which means that the femtosecond pulse energy is limited.

Experimental and theoretical studies resulted in the discovery of a new lasing regime in normal GVD [45–47]. In this regime, laser pulses are generated not owing to the balance effect; on the contrary, they are broadened and acquire a characteristic parabolic shape. But in this case, a regular linear frequency modulation (chirp) appears. Such broadened pulses can be compressed at the output using a system with GVD values of opposite signs (a pair of diffraction gratings). Due to the increase in the pulse duration, the constraints on the pulse energy caused by various nonlinear effects are reduced. Thus, this lasing regime provides a considerable increase in the pulse energy and peak power. Because this lasing regime is self-similar, the pulses were called similaritons.

This regime is especially important for Yb-doped fiber lasers emitting in the 1.1 μm range, where a Yb-doped fiber has normal dispersion. The similariton regime allows a drastic increase in the peak pulse power, thereby providing the development of high-average power optical frequency comb generators. The authors of [48] describe a Yb-doped fiber frequency comb generator emitting no less than 10 W of average power, with the width of the comb line being smaller than 1 mHz. In [49], the phase stabilization of pulses generated in this scheme was demonstrated.

4.3 Applications in precision spectroscopy

Perhaps the most important applications of cw femtosecond lasers are related to spectroscopy. During its development, spectroscopy played a key role in the discovery of the laws of quantum physics, the determination of fundamental physical constants, and the creation of the time, frequency, and length standards. Optical frequency combs exhibit extremely narrow lines at optical frequencies, which can be accurately compared with the frequency standard (the international definition of the second). This allows improving the resolution of spectroscopic techniques and developing perfect time, frequency, and length standards [50].

Because an optical frequency comb is in fact a ruler of evenly and closely spaced lines spanning a very broad spectral region, a spectral line under study can be compared by heterodyning with a certain 'tooth' of the comb to precisely determine its frequency.

The idea of using a cw ultrashort pulse laser for ultrahigh-resolution spectroscopy was proposed by Baklanov and Chebotaev [51] in 1977. Chebotaev had previously shown that the Doppler broadening of absorption lines in gases can be eliminated using two-photon absorption of two counter-propagating laser beams [52]. In this case, Doppler broadenings have the same magnitudes and opposite signs and are mutually compensated during two-photon absorption. Because the spectrum of cw ultrashort pulse lasers is a comb of evenly spaced lines with a readily controllable spacing, these lasers can be used in two-absorption experiments with counterpropagating laser beams. We note that different pairs of lines are involved in the required absorption, thereby increasing the efficiency of this process.

Baklanov and Chebotaev [51] drew a profound analogy between excitation by a coherent train of light pulses and the Ramsey excitation method in separated cavities. Applications of continuous-wave ultrashort pulse lasers were demonstrated in pioneering studies performed by Hänsch and collaborators [53–55]. A number of outstanding results were obtained. In particular, a femtosecond frequency comb was used to build highly accurate optical clocks.

4.4 Ultra-precise optical clocks

The international definition of the second is known to be based on the hyperfine-structure transition frequency in the ground state of the cesium isotope ^{133}Cs. This frequency (9.2 GHz) lies in the microwave region, its value being refined with time, in particular, using the atomic fountain technique based on the movement of atoms caused by resonance laser radiation. Cesium atoms are captured in a laser trap and cooled to temperatures below 1 μK. Then the cloud of cooled atoms is tossed upwards by laser radiation and subsequently falls downward in Earth's gravitational field (resembling the water in a fountain). During their motion, the atoms pass through the cavity twice. The Ramsey method with separated cavities is thus realized. As a result, a cesium atomic clock is built with a precision approaching the 10^{-18} level. This corresponds to an error of 1 s during the lifetime of our Universe.

The development of laser methods for obtaining ultra-narrow resonances in the optical range has led to the idea of using these methods to build optical atomic clocks. However, the problem of measuring an optical frequency based on the cesium microwave definition of the second has appeared.

An accurate frequency measurement involves the calculation of the number of oscillations per unit time. Obviously, the longer the measurement time is, the higher the frequency measurement accuracy (the error in one oscillation cycle). The use of optical frequencies $\sim 10^{10}$ Hz instead of microwave frequencies $\sim 10^{15}$ Hz requires, correspondingly, the time shorter by a factor of 10^5 for the accurate calculation of the number of oscillations. In other words, to achieve the maximum accuracy, only several seconds are required, rather than several days, as in the case of cesium clocks. These and other fundamental advantages of optical clocks initiated extensive studies aimed at creating them.

The optical frequency measurement involves its comparison with the cesium microwave frequency standard. For this, a chain of frequency generators is used with frequencies differing severalfold. The frequencies of these generators should be coherently coupled, and their number should be sufficient to span the gap between the optical frequency being measured and the cesium standard frequency. Such a chain is similar to the mechanism of a conventional mechanical clock consisting of several gears linking the periods of the hour, minute, and second hands.

Chain generators with successive frequency conversion (multiplication or division) should be coherently coupled with each other. The number of such generators (lasers, Gunn diodes, klystrons) exceeds ten, and the chain as a whole is a complex system. Only a few such systems have been built in the most advanced scientific centers in the world engaged in precision metrology.

Because a femtosecond frequency comb links an optical frequency to a microwave frequency, which can be easily compared with the cesium standard, the advent of such a comb became a giant revolutionary step toward the creation of optical clocks. As mentioned in the Introduction, this achievement was awarded the 2005 Nobel Prize in physics, to John L Hall and Theodor W Hänsch [3, 4]. Figure 24 conventionally shows a mechanical analogy of comparing an optical frequency with the cesium standard. Only two gears

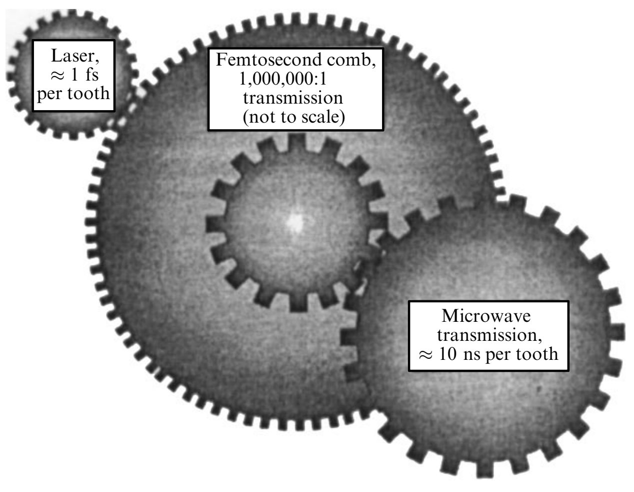

Figure 24. Mechanical analogy of optical frequency measurement using a femtosecond frequency comb [8].

are required on an axis instead of ten gears in the mechanical analogue of the frequency chain used previously. We note that complex and cumbersome setups in the frequency chain are now replaced by a compact and reliable cw femtosecond laser setup.

Considerable success in the development of precise optical clocks based on laser frequency combs was achieved by Bagayev and collaborators [56–59], who studied the properties of optical frequency combs in detail. They developed mode-locking techniques for narrowband diode lasers with certain frequency comb lines and described and studied the schemes of femtosecond optical clocks locked to the He–Ne (CH_4) frequency standard [57, 59]. Studies in this field are also being performed at the Lebedev Physics Institute, RAS (FIAN) [60].

Precision measurements of time and the speed of light of course allow measurements of length. The measurement of distances in space using a long-base heterodyne interferometer with an optical frequency comb is described in [61, 62].

To use precise optical clocks for experimental tests in fundamental theoretical physics, from cosmology to quantum physics, it is desirable to compare different unique systems that cannot be transported. This important problem was recently excellently solved in [63].

Researchers at the Institute of Physics and Technology (Braunschweig, Germany) studying a Cs fountain and researchers at the Max Planck Institute of Quantum Optics (Garching, Germany) involved in precision measurements of the 1S–2S transition frequency (a wavelength of 243 nm) have demonstrated the transfer of an optical frequency with high stability between these institutes with the help of a standard fiber optic communication link 929 km in length. For this, the frequencies being measured (the cesium standard microwave frequency and the optical frequency corresponding to a wavelength of 243 nm) were compared with the frequency corresponding to the fiberoptic communication link wavelength ≈ 1.3 μm. For this purpose, two femtosecond fiber lasers generating optical frequency combs were coupled to the ends of the communication link. As a result, exact correspondence between the optical frequency and the microwave frequency of the Cs fountain was achieved. The long-term frequency deviation did not exceed 4×10^{-19}. The results of this work demonstrate the possibility of the development of an all-European ultra-precise time network.

We note that the fiber-link technique was used for precision measurements of optical frequencies after overcoming some crucial limitations. The optical frequency can change during the propagation of light in a single-mode fiber due to variations in the optical propagation length with time. These variations can be caused by temperature variations or some mechanical perturbations. The change in the length with time causes a change in the phase with time, resulting, in turn, in the frequency change. Hence, the frequency coupled to the link input can change by a random quantity at the link output.

The frequency distortion can be compensated using the ingenious technique proposed and realized in [64]. The principle of this technique is as follows. We consider light at a frequency f_0 propagating through an optical fiber with a fixed length, where f_0 changes by a random quantity Δf. Then the frequency at the fiber output is $f_0 + \Delta f$. This frequency is immediately sent back. Assuming that the process producing the frequency variation does not change during the propagation of light in the fiber (several milliseconds), the frequency coming to the fiber input is $f_0 + 2\Delta f$, and it can be compared with the initial frequency f_0 to determine $2\Delta f$. The frequency f_0 is converted to $f_0 - \Delta f$ in a frequency modulator and is coupled to the link. As a result of distortions, the required frequency $f_0 - \Delta f + \Delta f = f_0$ is obtained at the fiber output.

4.5 Precision measurements in fundamental physics

Ultra-precise atomic clocks are successfully used for precision measurements of fundamental constants. For example, spectroscopic studies of a hydrogen atom are based on precision frequency measurements. Measurements of the 1S–2S transition frequency are being performed with constantly increasing accuracy [65]. A recent measurement was performed using fiber-link frequency standards. The 1S–2S transition frequency was measured to be 2,466,061,413,187,017 (11) Hz.

Measurements of the Rydberg constant (R_∞) with increasing accuracy are based on hydrogen spectrum studies. These measurements can be regarded as the method for testing quantum electrodynamic corrections or measuring the charge radius of the nucleus.

The fine structure constant α, one of the fundamental physical constants, is also being measured. This constant is related to the Rydberg constant as $\alpha^2 = (2R_\infty/c)(h/m_e)$, where c is the speed of light, h is Planck's constant, and m_e is the electron mass. The fine structure constant was measured with an accuracy of 6.6×10^{-10} [66].

An important part of such studies is the determination of the time drift of fundamental constants [67–69]. Progress in the development of ultra-precise atomic clocks and related precision measurements opened up the possibility of experimental studies of the time drift of fundamental constants. These data can be obtained by comparing the readings of clocks whose operation principle is based on different physical processes or different spectral transitions.

Precision measurements of the 1S–2S transition frequency in hydrogen were used to compare the readings of a $^{199}Hg^+$ optical clock and ^{87}Rb and ^{133}Cs microwave clocks. As a result, the limits of possible time drifts of some fundamental constants were found. For example, the limit of variation in the fine structure constant was estimated as $\Delta\alpha/\alpha = (-1.6 \pm 2.3) \times 10^{-17}$ year^{-1} [69].

4.6 Applications in astrophysics

Spectroscopic studies play a very important role in astrophysics. Appearing more than 200 years ago with the discoveries by Wollaston and Fraunhofer, spectroscopy has been one of the most powerful tools for learning about the nature of the Universe. Currently, similarly to how the highly stabilized laser has replaced the krypton lamp as the standard meter, laser frequency combs are replacing discharge lamps and absorption cells in calibrated spectrometers in telescopes.

The conceptual possibility of considerably increasing the resolution of spectral instruments used in astrophysical investigations initiated a number of studies aimed at realizing these possibilities. The Doppler effect gives a method for accurate measurements of radial velocities of stars, galaxies, and other cosmic objects by recording wavelengths (more accurately, frequencies) in their spectra.

A laser frequency comb with the mode spacing increased to many gigahertz and compared with the frequencies of atomic clocks has become an almost perfect tool for precision calibration of astronomical spectrographs. The high accuracy of measuring Doppler shifts opens up new fields of studies, such as the search for exoplanets and direct measurements of the expansion rate of the Universe.

4.6.1 Optical frequency comb with a gigahertz mode spacing. To use optical frequency combs for the accurate calibration of astronomical spectrographs, the spectrographs have to resolve the frequency comb lines. This requires a comb mode spacing exceeding 10 GHz. The optical length of a laser resonator with such a pulse repetition rate has to be equal to only 1.5 cm. The construction of such a laser is an extremely complicated problem. In [70], a femtosecond ring Ti:sapphire laser cavity 1.5 mm in length is described. The total size of the laser itself is only 8 mm. The pulse repetition rate of the laser is 10 GHz. The laser is so small that it can be placed on a 2-euro coin (with a diameter equal to that of a 50-kopeck coin) [71]. But this laser must be pumped by a power of 6.5 W, which requires the use of a Coherent Verdi pump laser, and therefore the system as a whole turns out to be expensive and not compact. In addition, Ti:sapphire lasers are very sensitive in alignment and are not suitable for long-term operation with telescopes.

More suitable are femtosecond fiber lasers, which are compact, are highly efficient, and have excellent operating characteristics. Another important advantage of femtosecond fiber lasers is the possibility of light amplification in fiber systems with cladding-pumped fibers. Such double-clad fiber amplifiers reliably provide average powers of a few watts. However, the large line spacing required in frequency combs generated by femtosecond fiber lasers is even more difficult to obtain because of the problems involved with decreasing the active fiber length. A maximum frequency of 3 GHz was obtained in a femtosecond Yb-doped fiber laser 1 cm in length [72].

To increase the spectral line spacing in an optical frequency comb, filtering the comb spectrum has been proposed [73, 74]. The transmission of a Fabry–Perot etalon with mirrors with a reflectance R separated by a distance L depends on the frequency v as

$$T(v) = \frac{(1-R)^2}{(1-R)^2 + 4R\sin^2(2\pi v L/c)} \,. \qquad (11)$$

The Fabry–Perot etalon thus has a regular set of transmission bands of width R spaced by the distance L. The length of the Fabry–Perot etalon can be selected to make the spacing between the etalon maxima a multiple of the spectral line spacing in the frequency comb. In this case, the line spacing in the comb can be increased by the corresponding factor. Despite the simplicity of this idea, its experimental realization involves considerable difficulties, which nevertheless are being successfully surmounted. We note that the bandwidth depending on the reflectance should be smaller than the spectral line spacing in the initial comb. This can require the use of mirrors with a very high reflectivity. To reduce the requirements imposed on the reflectance, a sequence of two or three etalons is used.

The attenuation of radiation after filtration can be compensated by placing laser amplifiers between Fabry–Perot etalons. In fiber lasers, such an approach is especially efficient because cw fiber amplifiers producing average powers up to a few watts can be used. As a result, the filtered radiation intensity can achieve the value required for efficient supercontinuum generation in a microstructured fiber. Thus, an octave spanning comb with a mode spacing of 10–25 Hz can be obtained.

In [75], an optical frequency comb setup for calibrating an astronomical spectrograph is described. The comb produced by a 1550 nm femtosecond Er-doped fiber laser with the pulse repetition rate $f_{\rm reper} = 250$ MHz was filtered by a system of two Fabry–Perot etalons to increase the spectral line spacing in the comb to 12.5 GHz. Then, radiation was amplified in diode lasers and in a high-power cladding-pumped Er-doped fiber amplifier. In this way, the required radiation intensity was achieved for generating a supercontinuum with the width exceeding an octave in a fiber with a high nonlinearity. This allowed the measurement of the frequency $f_{\rm CEO}$ required for stabilization. The frequencies $f_{\rm CEO}$ and $f_{\rm repet}$ were linked to the frequency standard. As a result, a frequency comb spanning the wavelength range from 1380 to 1820 nm with a mode spacing of 12.5 GHz was obtained.

4.6.2. Exoplanet searches. One of the urgent questions of modern astronomy is the search for new stars having exoplanets. Exoplanets cannot be discovered using even the most advanced telescopes. Exoplanet searches are therefore performed by indirect methods. We consider a method based on the measurement of the radial velocity of a star with respect to an observer on Earth. A planet rotates around the center of mass of the star and planet. As a result, the star experiences periodic perturbations with the radial velocity; the larger the planet mass is, the higher the maximum velocity. The radial velocity of the star can be measured by the Doppler shift of the wavelength (more accurately, the frequency shift) of the light emitted by the star.

The method for exoplanet searches by measuring the radial velocity from the Doppler frequency shift is quite successful. Currently, more than 600 planets have been discovered. Astronomical spectrographs are calibrated using thorium–argon lamps, which, however, cannot provide the measurement of radial velocities with an accuracy better than ~ 10 m s^{-1}. Because of this, the majority of the discovered exoplanets are Jupiter-like massive planets. One of the main goals of exoplanet searching is the discovery of Earth-like planets in the so-called living zone (where the star radiation provides conditions for the existence of water on the planet, which is required for the existence of life). But this requires a measurement accuracy of ~ 5 cm s^{-1}, which can, in principle, be provided by laser frequency comb calibrators of stellar spectra.

Figure 25 [76] shows the principal scheme with an optical frequency comb used for such measurements. The optical frequency comb spectrum controlled by the frequency standard is supplied to an astronomical spectrometer along with the stellar emission collected with a telescope. To provide the optimum detection, a line of the comb should cover 3–4 pixels of the CCD array of a detector. This means that the mode interval of the frequency comb should be 20–30 GHz.

The experimental implementation of this scheme is discussed in [75], where the Sun was observed with a telescope. The spectral lines of the solar photosphere were studied using a laser frequency comb for spectrometer calibration. These studies have shown that laser frequency combs are promising for these applications and promoted the development of work aimed at the creation of required frequency comb schemes and their investigations.

Some work aimed at discovering exoplanets using laser measurement techniques has already been performed. One

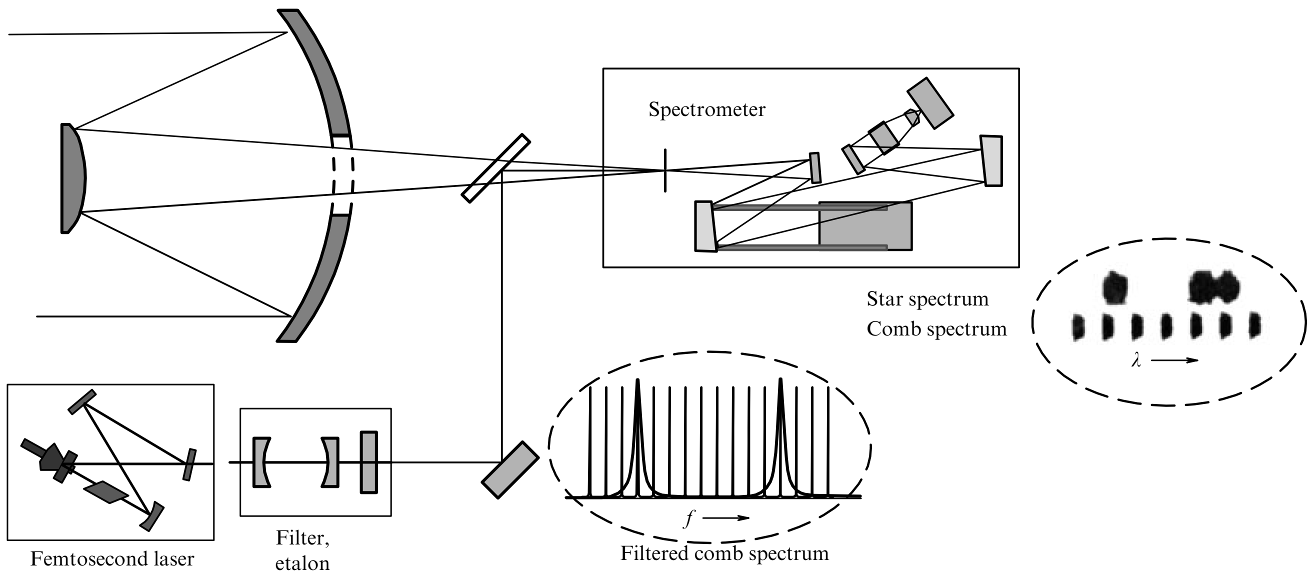

Figure 25. Scheme of using a laser frequency comb for calibrating an astronomical spectrograph [76].

study [77] was carried out using the Hobby–Eberly telescope with a 9.2 m mirror at the MacDonald Observatory (Texas, USA). The authors of [77] investigated the HD 168723 star having a planet that had previously been discovered with the help of a Pathfinder spectrometer calibrated by the conventional method using a thorium lamp. In this way, the conventional calibration method could be compared with frequency comb calibration. Spectral measurements were performed in the near-IR region.

This star is a dwarf star of the spectral class M, and its emission is weak in the visible spectral region because of its comparatively low temperature, but is quite bright in the near-IR region from 990 to 1800 nm. Such stars are very good candidates for exoplanet searches. Due to the relatively small mass of these stars, the radial velocity of an earthlike planet rotating in the so-called living zone (where liquid water can exist) causes a change in the radial velocity that is an order of magnitude greater than that caused by Earth rotating around the Sun.

A femtosecond Er-doped fiber laser with a pulse repetition rate of 250 MHz was used. Radiation was filtered with two identical Fabry–Perot etalons with mirrors with a reflectance of 99.8% and curvature radii 5 and 10 cm. A gap of approximately 0.6 cm between the Fabry–Perot etalons was adjusted with a piezoelectric ceramics to obtain a mode spacing of 25 GHz by filtering in the frequency comb. The comb spanned the wavelength range from 1450 to 1700 nm. The position of the comb was stabilized by comparing its frequencies with those of atomic clocks in the Global Positioning System. The star image and optical frequency comb radiation were delivered to a spectrometer through an optical fiber. Observations over four nights provided an accuracy of measuring the radial velocity about 10 m s^{-1}. This accuracy is higher than that obtained by standard methods. Laboratory studies have shown that the measurement accuracy can be improved to 1 m s^{-1}.

The calibration of an astronomical spectrograph with the help of an optical frequency comb was described in [78]. The frequency comb was generated by a commercial femtosecond Yb-doped fiber laser (Menlosystems Orange Comb 250) with a pulse repetition rate of 250 MHz. This frequency was converted to compare with the frequency of a rubidium atomic clock. In this way, the frequency stability at the level of $\Delta\nu/\nu = 10^{-11}$ was achieved, where $\Delta\nu$ is the frequency deviation per month. Such accuracy means that the Doppler shift of the velocity can be measured down to 0.3 cm s^{-1}. The mode spacing of the comb was increased using a set of Fabry–Perot etalons. Three or four successively mounted Fabry–Perot etalons were tested. The mode spacing in the comb was increased to 18 GHz. The attenuation of radiation caused by filtration was compensated using fiber amplifiers. The reflectance of the etalon mirrors was $R = 99.2\%$. To obtain a frequency comb required for measurements in the wavelength range from 380 to 690 nm, the laser radiation frequency was doubled in an LBO (lithium triborate) crystal. Measurements performed for calculating the orbit of a planet of the HD 75289 star showed that the accuracy of measuring the radial velocity can theoretically reach 2.5 cm s^{-1}.

4.6.3. Direct measurement of the Universe expansion acceleration. According to modern concepts, the Universe is expanding with an acceleration estimated as ~ 6 cm s^{-1} per year. In 1962 [79], it was proposed to determine the Universe expansion acceleration by measuring the radial velocity. The expansion of the Universe should produce a drift of the redshift of cosmological objects from which the acceleration can in principle be measured. However, it was pointed out that "with present optical techniques there is apparently no hope of detecting such small changes in redshifts for time intervals smaller than 10^7 years" [79]. Advances in laser science and technologies drastically changed the situation. The accuracy of measuring radial velocities by Doppler spectra can reach ~ 1 cm s^{-1}. This provided the basis for the International CODEX (Cosmical Dynamics Experiment) project of measuring redshift drifts with the largest telescopes [80]. It is assumed that with the measurement accuracy achieved at present, the Universe expansion acceleration will be directly determined during 10–15 years of observation.

4.7 High-resolution mid-infrared spectroscopy

Optical frequency comb techniques are successfully used in near- and mid-IR molecular spectroscopy. An efficient technique for molecular spectra studies is the so-called molecular fingerprinting. This technique provides the accurate recording of the intricate structure of near- and mid-IR spectra. Molecules of every type are here characterized by theirs own (individual) absorption spectra. This technique was considerably improved using optical frequency combs [81]. The high sensitivity and high resolution of this technique are efficiently used for detecting small amounts of impurities in gases, which is important for medical applications and applications in ecological and safety systems.

To generate an optical frequency comb in the required spectral range, various cw femtosecond lasers are used, emitting in the near- and mid-IR ranges, including crystal lasers such as $Cr^{2+}:ZnSe$ [82] and Tm- and Tm/Ho-doped fiber lasers [83] emitting in the 2 μm range. Emission at longer IR wavelengths is obtained using nonlinear frequency conversion processes such the as difference frequency generation [84] and optical parametric generation [85–87].

The detection of trace molecular gases in human breathing was demonstrated, which has an important medical application [88]: the highly sensitive detection of some molecules gives information on physiological processes in the human body and can be a useful diagnostic tool.

5. Trends in the development of optical frequency combs

The variety of applications of optical frequency combs imposes different requirements on their characteristics. For example, frequency comb matching of the newest schemes of atomic clocks should be performed without the impairment of accuracy (at the 10–18 level). In the case of astronomical applications based on Doppler shift measurements, radiation frequencies from cosmic objects with the minimal width of their spectra in the range of a few kilohertz are measured. This means that the measurement accuracy is at the 10^{-10}–10^{-11} level (the velocity 1 cm s^{-1} corresponds to the frequency shift of $\approx$ 25 kHz). However, the long-term (10–20 years) measurement stability is required.

Applications in the field of molecular spectroscopy require frequency combs in the mid-IR range. Frequency combs were also used in the VUV range between 40 and 200 nm [89]. A phase-coherent optical frequency comb is obtained by higher harmonic generation. The generation occurs in the amplifying cavity cell of a high-power cw femtosecond sapphire laser [90].

Femtosecond lasers are gradually becoming more accessible, their cost is decreasing, and the operational conditions are improving. The first systems used KLM sapphire lasers, which must be pumped by a stable laser emitting 5–10 W in the green region. Such a radiation source is based on a diode-pumped, intracavity-doubled neodymium laser. Millennia and Verdi commercial lasers of this type are produced by the companies Spectra-Physics and Coherent. However, they are quite expensive.

The use of a $Cr^{4+}:Mg_2SiO_4$ forsterite crystal instead of sapphire considerably reduced the laser cost. In this case, pumping was performed by an accessible and efficient fiber laser, without the need to double pump radiation. Although the output parameters of this laser are inferior to those of a sapphire laser (a wavelength of 1240 nm instead of 780 nm and lower peak powers), its advantage is the large wavelength of the pump laser ($\sim$ 1 μm instead of green light). Pumping can be performed by a Yb-doped fiber laser. The IRE-Polyus Scientific and Technical Association produces commercial PYL-10LP lasers. The cost of this laser is considerably lower than that of pump lasers for a sapphire laser. This made this femtosecond forsterite laser accessible for Russian laboratories, where excellent results were obtained in femtosecond studies.

The next stage in the development of cw femtosecond lasers is related to the use of diode-pumped active fibers. As a result, the most advanced, low-cost laser systems with excellent operational characteristics were created.

Semiconductor cw femtosecond lasers are currently being developed. A specific feature of such lasers is their compactness, which in principle allows achieving high pulse repetition rates and large mode spacings in frequency combs due to the small optical length of the cavity. In [91], a semiconductor disc laser emitting 110 fs pulses with a pulse repetition rate of 92 GHz was studied. In [92], the pulse repetition rate was 100 GHz. The proper choice of a semiconductor provides an emission wavelength coinciding with that of an efficient fiber laser. This ensures the radiation intensity required for supercontinuum generation.

Also, studies directed at the improvement of supercontinuum generators are being continued. To increase the intensity of radiation propagating in a fiber core, a microstructured fiber with a very small core diameter ($\approx$ 500 nm) was fabricated [93]. This fiber was used for supercontinuum generation upon irradiation by the second harmonic from a cw femtosecond Yb-doped fiber laser. By using radiation filtration in Fabry–Perot etalons, an optical frequency comb with a mode spacing of 14 GHz for calibrating astronomical spectrometers was created.

The supercontinuum generation efficiency can be increased by increasing the nonlinearity of the material irradiated by femtosecond pulses. It was proposed to use fibers with a core made of a material having a nonlinearity higher than that of glass. The manufacturing technology of special structured fibers allows the fabrication of single-mode, hollow-core optical fibers. Such fibers are peculiar capillaries that can be filled with a material with a nonlinearity higher than that of glass, thereby obtaining a supercontinuum generator with improved characteristics [94].

We note that the frequency comb of a cw femtosecond laser is not perfect. The comb radiation is accompanied by frequency and amplitude noises, and these noises should be minimized. Technical noises produced by temperature and acoustic fluctuations can be easily eliminated using good mechanical design with the necessary control feedback.

Besides these noises, amplitude and frequency noises caused by the pump laser noise appear, as does the fundamental noise of the laser itself [amplified spontaneous emission (ASE)]. The suppression of these noises is more complicated [95–97]. The ASE noise in the comb generated by high-power femtosecond lasers emitting shorter pulses is lower. In this respect, solid-state lasers (sapphire and forsterite lasers), which have comparatively short excited-state lifetimes of the active medium and high intracavity radiation intensities, offer certain advantages over fiber lasers.

During nonlinear supercontinuum generation, noises are so strongly amplified that they can destroy the comb itself. The study of noises in microstructured fibers is therefore quite important [98, 99]. Noises can be reduced by reducing the length of the nonlinear fiber and the pulse duration. For this reason, the use of femtosecond pulses is preferable, even though picosecond pulses can provide higher energies and intensities.

6. Conclusions

Passively mode-locked continuous-wave lasers can be used as sources of femtosecond frequency combs. Their advent is an outstanding achievement of laser science and technology. At present, optical frequency combs are generated by efficient and accessible femtosecond fiber lasers. As expressed by an American researcher, fiber lasers took femtoseconds to the masses.

The last 10–15 years saw many examples of remarkable applications of optical frequency combs, both in fundamental science and in technologies. The accuracy of optical and metrological measurements was drastically improved. A Michelson, who in 1907 received a Nobel Prize in physics "for his optical precision instruments and the spectroscopic and metrological investigations carried out with their aid," said in 1894 at the opening ceremony of the Ryerson Physical Laboratory at the University of Chicago: "...our future discoveries must be looked for in the sixth decimal place." Today, about 120 years later, the measurement accuracy has reached the 18th decimal place!

Laser measurement methods have been used for the creation of ultra-precise optical clocks, improvement of the Global Navigation Satellite System (GLONASS), the development of the latest spectroscopic techniques, and astrophysical applications. It is important that there is a clear tendency to make optical frequency combs more accessible by reducing their cost and improving their operational characteristics.

Studies in the field of continuous-wave femtosecond lasers and optical frequency combs based on them are of current interest and are opening up excellent opportunities for young people devoting their lives to serving science.

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