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Diode-pumped ultra-short-pulse solid-state lasers

E. Sorokin, I.T. Sorokina, E. Wintner

Institut für Photonik, Technische Universität Wien, Gusshausstrasse 27/387, 1040 Wien, Austria
(Fax: +43-1/58801-38799, E-mail: ernst.wintner@tuwien.ac.at)

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Abstract. Many materials are good candidates for diode-pumped ultra-short-pulse lasers: several transition-metal-ion-doped crystals can or could support extremely short fs pulses. This goal, so far, has only been reached by Cr^{3+}:LiSAF, but there are good chances for other crystals like Cr^{4+}:YAG having its bandwidth within the third communication window, and the high-yield Cr^{2+}:ZnSe with its impressive bandwidth in the near IR. Rare-earth-ion-doped media deliver only sub-ps pulses but allow unprecedented and scalable high average powers, like a SESAM mode-locked Yb:YAG thin-disk laser described recently. In all ranges of pulse durations there are fascinating applications ready for widespread employment as soon as compact, reliable and moderately priced ultra-short-pulse systems will be available for the non-laser-skilled user. The highest impact in the near future is attributed to microstructuring of materials and processing of biological samples, including dental enamel, by ps and sub-ps pulses, and optical coherence tomography needing pulses in the 10-fs regime at very modest average powers.

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The generation of ultra-short pulses has been a challenging field of laser science since the very first results of mode locking of lasers and hence the demonstration of picosecond laser pulses [1–3]. It was a temptation to reduce pulse durations steadily within the more than 30 years which followed, reaching best values of just a few optical cycles generated directly in a laser resonator [4, 5]. The most impressive advances in this direction were essentially achieved by taking advantage of just two most versatile types of laser media, namely dye lasers and solid-state lasers, among which Rh6G [6] and Ti:sapphire [4] turned out to be the most successful ones, respectively. When employing external pulse amplification, nonlinear chirping and pulse compression, after many single steps of improvement the presently valid record value for pulse duration for ultra-short pulses, i.e. femtosecond pulses as often referred to, of 4.5 fs [7, 8] has been achieved. This comes very close to the ultimate limit of single cycles of approximately 2 fs for visible light.

Obviously, at any stage of ultra-short-pulse development there were many successful efforts to apply them, which are by far too numerous to even give a representative cross section of them in this paper. Basically, the focus of interest was either on time-resolved measurements of ultra-fast processes (see e.g. [9]) or on the measurement or employment of nonlinear processes (see e.g. proceedings of the series of conferences 'Ultrafast phenomena' [10]). The yield of the latter by nature is much higher if ultra-fast pulses associated with high peak powers are involved in such experiments, as compared to laser radiation of longer time duration. In many cases ultra-short pulses represent the only possible experimental condition because thermal side effects associated with longer pulses would destroy the samples before any measurement could be performed. Recently, another aspect of femtosecond pulses gained a lot of attention: the short coherence time together with the wide bandwidth of such laser radiation associated with the usual high brightness and excellent spatial coherence being by far superior to the light of superluminescent diodes. This led to impressive applications of optical coherence tomography (OCT) [11–13]. In general, all these applications have hardly reached a widespread distribution in a commercial sense because the lasers are still too bulky, too unreliable, too maintenance-demanding and, hence, simply too expensive.

A new era of ultra-short-pulse lasers was started when diode pumping gradually became more and more feasible. It took a very long time until semiconductor diode lasers became mature for technical applications and even longer until 'high-power diode lasers'[1] became available essentially in the form of array lasers [14–17], gradually replacing conventional pump sources like flashlamps or highly ineffective ion lasers. The first such diode lasers were GaAs/GaAlAs lasers emitting around 800 nm, luckily coinciding with absorption lines of Nd:YAG [18]. Later, more and more wavelengths were (at least potentially) covered by high-power diode lasers

[1] It may be considered practical to define 'high-power semiconductor diode lasers' to emit more than 0.5 W.

so that a large variety of mainly solid-state laser materials could be diode-pumped or became good candidates for direct diode pumping[2] [19, 20].

In a number of applications the generation of the shortest pulses is the focus of interest. In this case broad-band-emitting laser crystals as listed in Table 1 are potential active media of choice for laser oscillators which should be designed under the aspect of diode pumping: compactness and robustness are a high goal, and hence modern elements of ultra-fast resonator design are essential, i.e. mirror dispersion compensation [21–23] and passive mode locking via Kerr lenses (KLM) [24, 25] or semiconductor saturable absorber mirrors (SESAMs) [26–28], or a combination of both [29]. Fiber lasers, having been developed to a very high and useful standard in continuous wave (CW) and mode-locked operation modes and later having been commercialized, follow somewhat different rules and will not be covered in much detail within the scope of this paper. Pulse shortening within the resonator generally follows the solitary pulse formation concept [30–32] underlying limitations by high-order dispersion [33, 34] and generally suffering from smaller brilliance of diode lasers, yielding somewhat longer pulses than under conventional pumping [35]. If the emitted wavelength from a femtosecond resonator is important for the application, a systematic wavelength shift towards longer wavelengths has to be considered. This was observed in many solid-state lasers and can be understood on the basis of the Raman effect within the host material [36].

For a few years, applications of ultra-short pulses became more and more numerous where the pulse duration is not the important parameter in the focus of interest. On the contrary, subpicosecond or picosecond pulses might be more advantageous in this kind of application. High average power[3] and medium repetition rate[4] are considered to be essential for such new applications like materials processing. This is a very wide field covering ablation of solid inorganic matter as well as vaporization of biological tissue. High average power is needed for a competitive speed of material removal as compared to classical methods, while the repetition rate is limited by the time of dissipation of a plasma cloud in the vicinity of the location of impact. The pulse-duration time to be chosen is, on the one hand, governed by the energy-exchange time between electron gas and lattice and the diffusivity of hot electrons in the case of metals [37]. On the other hand, in the case of dielectrics with low concentration of mobile carriers multi-photon processes need to be excited, requiring high intensities generally associated with much shorter pulses [38]. Several laboratory measurements on metals and biological samples like dental enamel involving pulse durations around 1 ps yielded excellent results [39, 40]. Very promising results on high average power ultra-short-pulse lasers have been published recently [41–44], but much more work still has to be done until such lasers can become mature commercial products at affordable prices.

Section 1 briefly reviews the modern techniques of passive mode locking and dispersion compensation of diode-pumped solid-state lasers. Sections 2 and 3 consider the two main groups of solid-state laser media for ultra-short-pulse lasers: transition-metal-ion-doped crystals and rare-earth-doped media. Finally, in Sect. 4 we shall feature some of the most important applications of these laser sources, where we see the real motivation for the whole effort to develop such lasers.

1 Passive mode locking and dispersion compensation techniques

Passive mode locking of solid-state lasers made tremendous progress during the last decade, following the introduction of three novel techniques: additive-pulse mode locking (APM) [45], Kerr lens mode locking (KLM) [24] and semiconductor saturable absorbers (SESAMs) [46]. Of these, the first two techniques employ quasi-instantaneous third-order optical nonlinearity to create an artificial fast saturable absorber; SESAMs represent saturable absorbers in the classical sense based on fast relaxation times of carriers. The APM technique faces certain limitations for bulk solid-state lasers because its realization requires interferometric stabilization of the cavity arm lengths and fiber coupling to achieve enough nonlinearity. Both these requirements made APM systems overcomplicated and environmentally unstable. The most successful nonlinear optical scheme for bulk solid-state lasers in order to generate the shortest pulses is KLM, which was already commercialized as early as in 1992. The SESAM technique has matured very much since its invention and has been successfully implemented in a wide spectral region from 0.8 to 1.5 μm, in bulk and fiber lasers [47, 48], and is also commercially available. The KLM and SESAM techniques can be combined, so that a SESAM may serve as a starting element for a KLM-based laser [29].

At today's level of understanding, however, the mode-locking technique does not define the pulse-duration limits at a subpicosecond time scale; it rather exerts a stabilizing influence [31]. The critical mechanism for obtaining ultra-short pulses is the interaction of the distributed dispersion in the resonator with the optical third-order nonlinearity, often exclusively present in the active medium. This is a direct analog to soliton formation in optical fibers (*solitary* pulse formation [35, 49]). This mechanism requires at least one tight focus of an intracavity beam inside a nonlinear optical material and constant second-order dispersion over a broad wavelength region in the resonator. Consequently, resonator geometry and dispersion compensation are critical issues.

A standard most flexible cavity configuration for femtosecond lasers is the $X-$ or $Z-$shaped 4-mirror resonator, which makes dual use of the active element: as gain and as nonlinear medium at the same time. This is possible because the tight focus of the intracavity beam is located inside the gain medium, providing intensities in the $\sim 10^5$ W/cm^2 range over a length of 2–10 mm. In the case of diode-laser excitation, however, pumping through the dichroic folding mirrors is not always a convenient option. Better mode overlap is typically achieved with end-pumped systems often preferred for

[2] In this paper indirect diode pumping will be not considered: e.g. frequency-doubled a Nd:vanadate laser pumping Ti:sapphire, etc.

[3] Typical average powers of ultra-fast-pulse lasers are in the order of hundreds of milliwatts. We understand high average power as used in this context to be substantially higher and hence in the several watts regime.

[4] Typical ultra-fast resonator repetition rates are in the range of 100 MHz; repetition rates after amplification generally lie between tens of Hz and kHz. Our definition for medium repetition rate applies to the range of 100 kHz to 1 MHz.

diode-pumped oscillators. Improved pump overlap and better power-handling capability are possible with an elliptical mode shape inside the active medium [50–52]. For oscillators with tens of watts of output power, novel pumping concepts like the thin-disk laser [53] look very promising.

The beam waist in the active crystal often has to be made rather large in order to achieve good overlap with poorly focusable diodes or to prevent thermal damage in high-power systems. In this case, the nonlinearity in the gain medium may become insufficient for KLM or for solitary pulse formation. In this case an additional beam focus can be introduced into the cavity with a transparent nonlinear optical material placed there. This kind of design has been successfully used for mode locking of a diode-pumped Nd:YLF laser [54]. A suitable theory has been developed allowing us to characterize the KLM properties of an arbitrarily complex resonator [55].

Out of a number of different techniques for dispersion compensation only three are applicable within the resonator and have found successful use in femtosecond laser technology, namely prism pairs [56], chirped mirrors [57] and Gires–Tournois mirrors (GTMs) [22, 58, 59]. The main advantage of the prism-pair arrangement is the possibility to continuously and precisely adjust the amount of negative dispersion. At the same time, the prism pair introduces comparatively large additional third-order dispersion (TOD), becoming a main obstacle for pulse shortening at the sub-20-fs level. The prism pair also significantly increases dimensions and system complexity.

The chirped dielectric mirrors [57] exhibit the highest bandwidth available to date and allow true dispersion engineering, including also higher-order dispersion. However, they possess relatively low dispersion and high losses, about 50–80 fs^2 and up to 0.2% per reflection, respectively. These losses originate partly from transmission losses, but also from scattering losses in the thick coatings (up to 50 layers). The GTMs represent a certain trade-off in this respect, since their reduced bandwidth of ~ 10–30 nm is sufficient to support pulses in the subpicosecond regime, but at the same time they have larger dispersion and lower losses ($< 0.1\%$ per reflection, at least as good as high reflectors), even if made by conventional electron-beam technology.

Recently, a number of alternative techniques of dispersion compensating mirror design have been suggested, allowing us to expand the bandwidth in the Ti:sapphire wavelength range up to 300 nm and model virtually any kind of dispersion [23, 60, 61]. Especially interesting within the range of this paper is the technique of 'multiple Gires–Tournois mirrors' allowing us to reach bandwidths comparable to the chirped mirrors while keeping the low-loss properties of the GTMs [61]. The low-loss properties of GTM structures make them very suitable for application in diode-pumped systems.

Mode-locked glass-fiber lasers make a group of their own, and should be considered separately (for representative reviews, see [62, 63]). Some bulk laser techniques, such as KLM and mirror dispersion compensation, are not applicable here. A number of alternative techniques have been invented: the nonlinear active optical mirror and nonlinear polarization rotation for mode locking and the chirped grating for dispersion compensation. Around the 1.5-μm wavelength region (Er^{3+} in silica fibers), true soliton propagation can be employed due to the possibility of dispersion engineering of silica fibers.

2 Ultra-broad-band solid-state lasers

In this section we shall concentrate on transition-metal-ion-doped crystalline lasers. However, the class of ultra-broad-band solid-state laser materials includes a broad variety of crystals and glasses, also including color-center crystals. The lasers based on the latter type of media have been well studied for the purpose of tunable CW [64–67] and ultra-short-pulse generation [68, 69] and even for the purpose of diode pumping [70]. However, there are still two main obstacles on the way to their widespread use: the first is the frequent requirement for low-temperature operation, and the second consists in slow self-destruction of color centers at room temperature. Although the first limitation has been overcome in some lasers [67], the second, otherwise expressed as a short shelf life, still remains a problem.

An alternative option is to use transition-metal-ion-doped glasses as active media for broad-band solid-state lasers. Implementation of glass matrices for transition-metal ions may bring only marginal advantage in terms of line broadening. On the other hand, the obvious drawback is that the quantum yield of luminescence of active ions is usually low, the highest value measured so far in silicate glasses being 0.17 [71]. The practical use of laser glasses is also prevented by their inadequate thermo-optical and thermophysical characteristics. On the contrary, crystalline media are more robust and generally possess better thermo-optical properties.

Lasers employing transition-metal-ion-doped crystals have a long history. Indeed, the first solid-state laser was based on ruby, i.e. Cr^{3+}:Al_2O_3 [72], but the laser action took place on a narrow zero-phonon-line ($R-$line) transition. However, most radiative transitions in the crystals doped with Ti^{3+}, V^{2+}, Cr^{4+}, Cr^{3+}, Cr^{2+}, Ni^{2+} and Co^{2+} ions, and being interesting from the viewpoint of laser action, are vibrationally broadened up to a few hundreds of nanometers. The broadening takes place at the expense of the peak gain which is usually small in all these materials except Ti^{3+}:Al_2O_3 and Cr^{2+}:ZnSe, where there is practically no deteriorating influence on the gain by excited-state absorption (ESA). Such broad-band laser media allow the generation of extremely short pulses as long as a few optical cycles spanning a wide wavelength range from ~ 0.5 μm to 5 μm. Extremely short pulse durations have been achieved so far directly from the oscillator in Ti:sapphire [29, 60], Cr:LiSAF [73], Cr:LiSGaF [74], as well as Cr:forsterite [75]. Some of the mentioned lasers can be diode-pumped by the available high-power laser diodes. All this makes transition-metal-ion-doped crystals attractive active media for compact diode-pumped femtosecond laser sources.

The category of crystals suitable for CW room-temperature diode-pumped operation includes the colquiriites Cr^{3+}:LiSAF, Cr^{3+}:LiCaF, Cr^{3+}:LiSGaF, alexandrite (Cr^{3+}:$BeAl_2O_4$), Cr^{4+}:Ca_2GeO_4, Cr^{4+}:forsterite (Mg_2SiO_4) and Cr^{4+}:YAG. Table 1 gives a selection of the best results in these lasers in CW and mode-locked regimes achieved with III-V high-power diode-pump sources. Significant attention of the scientific community has been attracted towards the development of a compact ultra-short-pulse source based on Cr^{3+}:LiSAF representing a potential alternative to Ti:sapphire [83–86]. A few years ago, another crystal from the colquiriite family, Cr^{3+}:LiSGaF, was suggested for femtosecond pulse generation via Kerr lens mode locking [87, 88], having the ad-

Table 1. Diode-pumped continuous-wave (CW) and mode-locked (ML) broad-band crystalline lasers

Active medium	Pump-diode laser	Operation regime	Wavelength nm	Output power mW	Pulse duration fs	Ref.
(F_2^+)*:NaF(at 77 K)	AlGaAs	CW	1030–1120	7.8		[70]
Cr^{3+}:$BeAl_2O_4$	AlGaInP	CW	750	0.075		[76]
Cr^{3+}:LiSAF	AlGaInP	CW	860	1100		[77]
	AlGaInP	ML	900	6.2	12	[73]
Cr^{3+}:LiSGaF	AlGaInP	ML	840	61	78	[74]
Cr^{3+}:LiCaF	AlGaInP	ML	790	52	75	[79]
Cr^{4+}:Ca_2GeO_4	InGaAs MOPA	CW	1390–1475	20		[80]
Cr^{4+}:Mg_2SiO_4	InGaAs MOPA	CW	1236–1300	10		[80]
	AlGaInP	CW	1245	5		[81]
Cr^{4+}:YAG	InGaP/GaAs/InGaAs	CW	1415–1535	200		[82]

vantage of higher achievable output powers. In [88] up to 900 mW was demonstrated under Kr-ion laser pumping in the CW regime without any special cooling technique. This inspired a whole series of further works on diode pumping and passive mode locking of this crystal [78, 89–92]. It is remarkable that achieving the shortest pulses under diode-pumped operation was only feasible at the expense of output power (compare Table 1). This makes the requirement of optimal high-brilliance pumping (i.e. by a TEM_{00} laser-beam profile) indispensable. Most, if not all, of the novel concepts of mode locking and dispersion compensation of physical phenomena and limitations have been first studied under optimal pumping. Knowledge acquired in these studies could then be applied to the diode-pumped systems.

Along with diode pumping, the prism-less mirror dispersion control (MDC) approach [21], as described in Sect. 1, plays an important role in making resonator design really compact and stable. As an extension of this concept to the low-gain Cr^{3+}:LiSGaF and Cr^{3+}:LiSAF systems, Gires–Tournois mirrors for dispersion compensation were first suggested in [22]. Later, broad-band and sufficiently low-loss dispersive mirrors have been developed, permitting the generation of 14-fs pulses with considerable output power of ~ 100 mW [74, 93] in these lasers. It is interesting to note that in such systems the instantaneous intracavity power reaches tens of MW when operating on the sub-20-fs level. Correspondingly, 10 MW was also reported by Uemura and Torizuka [73].

At such pulse durations in the sub-20-fs regime we could observe a considerable pulse self-frequency shift (up to 60 nm) in our Cr:LiSAF and Cr:LiSGaF lasers, depending on the intracavity pulse peak power as well as on pulse duration (Fig. 1). Induced Raman scattering was identified as the physical mechanism causing this shift. Based on this observation a theory of Raman pulse shift in ultra-short-pulse crystalline lasers has been developed [94, 95]. Recently, induced Raman scattering in these materials has been also studied [96, 97].

Another phenomenon worth attention at very short pulse durations, when the net cavity group delay dispersion (GDD) approaches zero, is generation of side bands. In [98] a generalized condition for determining the resonant peaks in KLM lasers with an arbitrary dispersion compensation system has been derived analytically. As an example, Fig. 2 shows the measured pulse spectrum of a GTM-mirror-controlled Cr^{3+}:LiSGAF resonator [22] and the corresponding disper-

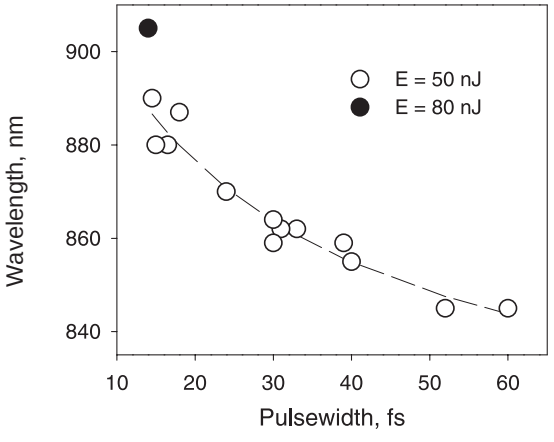


Fig. 1. Dependence of output wavelength vs. pulse width in a Cr:LiSGaF laser [95]. The data points (*empty circles*) on the graph correspond to approximately equal intracavity pulse energy of 50 ± 5 nJ but with different output couplers and net intracavity GDD. The *filled circle* corresponds to the pulse energy of 80 ± 5 nJ

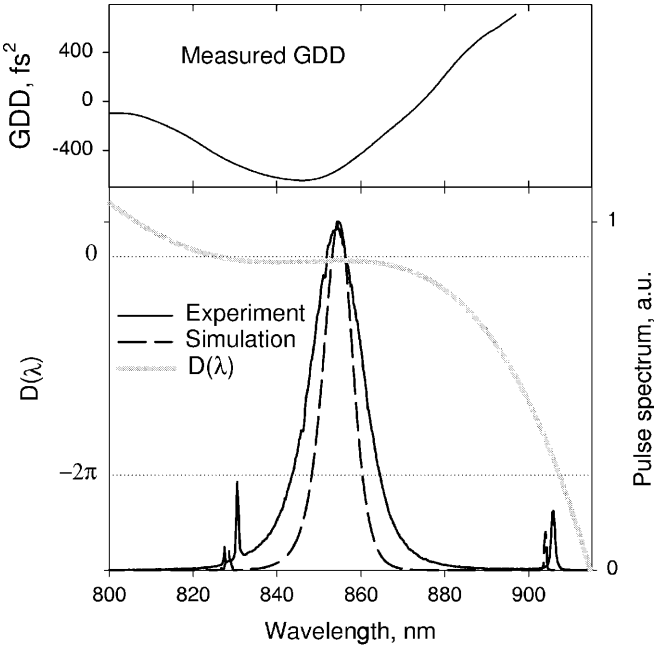


Fig. 2. *Upper part*: measured net GDD of the Cr:LiSGaF resonator, containing 3 GTMs and a 6-mm-long laser crystal. *Lower part*: Experimental spectrum of GTM-controlled Cr:LiSGaF laser with central wavelength at 855 nm and pulse duration (FWHM) of 40 fs; the corresponding dispersive function $D(\lambda)$ (*gray line*); results of a numerical simulation (*dashed line*) [98]

sive function $D(\lambda)$ for dispersions up to fourth order (FOD). A GTM-mirror-controlled laser has a dispersion curve which is well defined and rich in higher-order dispersion terms. Therefore, experimental data taken with such lasers is well suited for comparison with theoretical predictions. According to the theory, the points where $D(\lambda) = 0$ and -2π give the resonant peaks. It was shown that asymmetrically distributed multiple resonant peaks in the pulse spectrum can be explained not necessarily by the third-order dispersion, but also by higher-order dispersions (HOD). It was also shown that dispersive waves (DWs) are relatively long pulses whose durations and energies can be estimated through simple formulae. These long pulses propagate in company with the main short pulses forming wings to them. Furthermore, the energy transferred from solitary pulses to DWs increases rapidly with the decrease of net GDD. A practical condition on the relative amount of GDD and TOD provides minimum energy drain into DWs for the given intracavity pulse energy [98]. The theoretical predictions are in good agreement with experimental results, and may be useful for design of dispersion compensation systems and minimization of the adverse effects of DWs in diode-pumped mode-locked solitary lasers.

Besides the oscillators described above and Cr^{4+}:forsterite [99], Cr^{4+}:YAG [100–102] represents another well-investigated broad-band laser material emitting in the most interesting wavelength range around 1500 nm, corresponding to the third telecommunication window. This laser is also capable of producing pulses as short as 43 fs [103] and has a potential of supporting sub-20-fs pulses. During the last decade it has attracted a lot of attention as a compact source of ultra-short pulses [104–106]. However, for a wide acceptance of these lasers they will have to get less expensive and have higher wall-plug efficiency, which means direct diode pumping will have to be employed. The promising results on diode-pumped operation of Cr^{4+}:forsterite and the related Cr^{4+}:Ca_2GeO_4 reported in [80] required the ideal beam of a rather complex and very expensive master-oscillator power-amplifier (MOPA) diode-laser source or were obtained indirectly via a diode-pumped double-clad fiber laser [107]. Diode pumping of Cr^{4+}:forsterite by a 1.5-W AlGaInP diode at 670 nm has been demonstrated [81], yielding 5-mW of output power. The excitation route via 1-μm pump wavelength both in Cr^{4+}:YAG and in Cr^{4+}:forsterite seems to be more favorable due to lower excited-state absorption and higher efficiency, but is more difficult to realize due to lower absorption in this wavelength range relative to that in the visible region. We recently managed to accomplish this challenging task by combining highly concentrated excellent quality Cr^{4+}:YAG crystals with an optimized multi-diode pumping scheme [82] previously developed by us for a Cr^{3+}:LiSGAF oscillator. This allowed us for the first time to realize a compact directly diode-pumped room-temperature laser tunable over more than 120 nm around 1.5 μm, yielding more than 200-mW of output power at the highest electrical to optical efficiency reported for Cr^{4+}-doped laser systems, of at least $\sim 2\%$. Although this value might seem to be rather small, it is still by a factor of 4 higher than the corresponding values estimated for the Cr^{4+}:forsterite laser diode pumped by a double-clad fiber laser [107], being an alternative approach to the development of compact Cr^{4+}-doped systems. Hence, from the viewpoint of efficiency, cost, stability and compactness there is a clear motivation to develop directly diode-pumped femtosecond Cr^{4+}:YAG lasers.

Finally, there is another promising and rather new candidate for ultra-short-pulse diode-pumped operation associated with high tunability and high average power, i.e. the Cr^{2+}:ZnSe crystal [108, 109]. Diode-pumped operation of this crystal in a pulsed regime has been reported by Page et al. [110]. However, the available power and brightness of InGaAsP diode lasers were not sufficient to reach threshold in the CW regime. CW operation results under Co^{2+}:MgF_2 laser pumping demonstrate the achievable high output power of more than 500 mW tunable over 600 nm between 2200 and 2800 nm [111]. This laser also produced $\sim$ 5-ps pulses at considerable output power of 300 mW in an actively mode-locked regime [112]. Recently 1-W CW output power has been reported with Tm:YAG pumping [113]. All these works show that together with the progress in high-power InGaAsP diode lasers the development of a first compact ultra-short-pulse infrared laser oscillator based on Cr^{2+}:ZnSe is quite feasible.

3 Rare-earth-doped diode-pumped laser media

For a number of applications, pulse duration as short as possible is not the most important requirement. In such cases it can be traded off against other important laser parameters like particular wavelength region, efficiency, or average power, and hence other active media become the materials of choice, e.g. doped with rare-earth ions, such as Ho^{3+}, Er^{3+}, Nd^{3+}, Tm^{3+} and Yb^{3+}.

The optical spectra of these ions originate from the partially filled $4f$ shell shielded from the crystalline field by the filled $5s^2p^6$ shell. As a result of the lower electron–phonon coupling, the energy levels are significantly narrower in comparison to the transition-metal ions, typically of the order of 1 nm. This makes the rare-earth-ion doping not particularly suitable for generating ultra-short laser pulses. This is compensated by higher peak-emission cross section and long upper laser-level lifetime, resulting in lower threshold, high output power and efficiency. The energy-level structure of the rare-earth ions remains by nature essentially the same in every host, allowing a great variety of crystalline hosts or glasses to be used. It is possible to increase the bandwidth by selecting a host–ion combination with large homogeneous bandwidth or by making use of inhomogeneous broadening. Many of the rare-earth ions possess convenient pump bands in the near infrared, overlapping well with emission spectra of effective high-brightness AlGaAs and InGaAs laser diodes.

Rare-earth-doped glasses provide very smooth fluorescence spectra with typical bandwidth of 30–50 nm. Historically, Nd-doped glasses were the first rare-earth-doped materials to produce subpicosecond pulses in bulk [114] and in fiber lasers [115]. Compared to the crystals, glasses typically suffer from poor thermo-optical properties and higher nonradiative decay rates, making diode pumping more difficult. Nevertheless, diode-pumped femtosecond operation was first demonstrated in fibers doped with Nd^{3+} [116] and Er^{3+} [117]. Nowadays, diode-pumped femtosecond Er- and Yb/Er-doped fiber lasers in the 1.5-μm region are widely used in various configurations and are commercially available, with typical pulse duration of $\sim$ 300 fs.

Later, bulk-glass systems have bridged the gap and diode-pumped femtosecond Nd- and Yb-doped glass lasers have been reported. Nd-doped fluorophosphate glass delivered 100-mW of output power at 130-fs pulse duration [118], later improved to 60 fs [119] and, using the elliptical mode cavity [50, 51], to more than 1 W at 175 fs [120]. The latter result required 20-W of pump power, indicating that glasses are not a material of choice for efficient medium- and high-power operation [44].

A compromise between an increased bandwidth due to inhomogeneous broadening and good spectral and thermo-optical properties, typical for crystals, can be achieved by using rare-earth-doped disordered crystalline media. This approach for generating femtosecond pulses was first realized in Nd^{3+}-doped mixed garnets [121–124]. The disordered nature of such solid solutions results in strong inhomogeneous broadening of both the fluorescence line and the absorption bands. These characteristics, being typical for glasses, are combined with thermal and mechanical properties comparable with those of YAG and, additionally, allow two times higher Nd^{3+} doping levels. Most importantly, an isostructural solid solution permits continuous variation of composition in the whole substitution range, thereby making it possible to optimize the gain spectrum. Figure 3 illustrates the difference between the spectrum of an yttrium–scandium–gallium garnet (YSGG) and the corresponding spectrum of its 1 : 2 mixture with gadolinium–scandium–aluminum garnet (GSAG) (this composition was found to provide a symmetrical spectrum [123]). Using the additive-pulse mode-locking scheme, the bandwidth of the main spectral feature could be mode-locked, yielding 260-fs pulse duration in self-starting operation (Fig. 4). This result constituted a remarkable improvement over the values obtained in Nd^{3+}-doped crystalline lasers at that time and holds until now.

It is, however, not always necessary to exploit disordered host media to obtain a broad fluorescence bandwidth for rare-earth-doped crystals. Due to the stronger electron–phonon coupling [125], some rare-earth ions exhibit broader homogeneous linewidths, especially Yb^{3+} and Tm^{3+}. Compare, for example, the linewidth of a single Stark component at room temperature in Nd:YAG (~ 0.6 nm) and in Yb:YAG (~ 5 nm). Additionally, the positions and relative intensities of individual Stark components vary from crystal to crystal, allowing us to select such materials providing smooth continuous spectra as broad as ~ 150 nm for Tm^{3+} and Ho^{3+} in LaF_3 host [126] and up to ~ 30 nm for Yb^{3+} in $KY(WO_4)_2$ host [127]. Yb:$KGd(WO_4)_2$, being an analogous crystal, has recently produced subpicosecond pulses at 1.1-W of output power [128]. The most remarkable of the recent developments in rare-earth-doped high-power lasers is the achievement of 16.2-W of output power at 0.7-ps pulse duration in a SESAM-mode-locked and GTM-compensated thin-disk Yb:YAG laser by Aus der Au et al. [129], having the potential of even further output-power scaling.

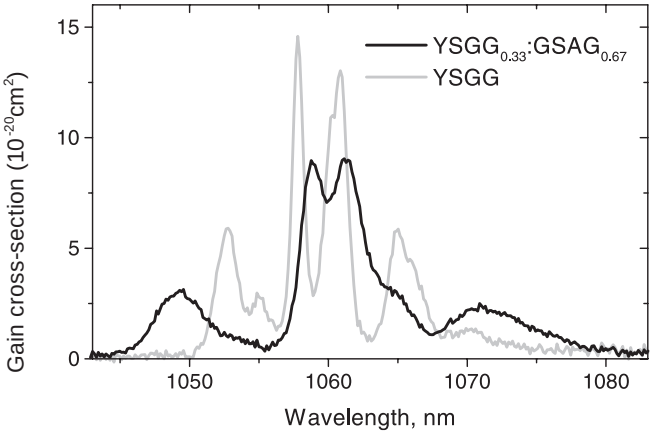


Fig. 3. Inhomogeneous gain broadening in mixed garnets. The *black line* shows the gain spectrum of a disordered mixed garnet. By choosing the right ratio of the components a smooth and symmetrical spectrum is obtained [123]. For comparison, the *gray line* shows the gain spectrum of one of the components, yttrium–scandium–gallium garnet

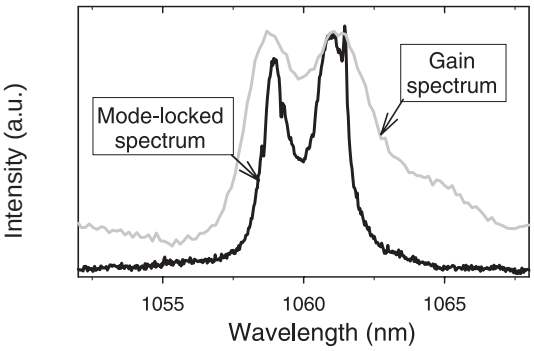


Fig. 4. Mode-locked spectrum of a mixed GSAG:YSGG garnet (*black line*). The *gray line* shows the gain spectrum for comparison

4 Applications

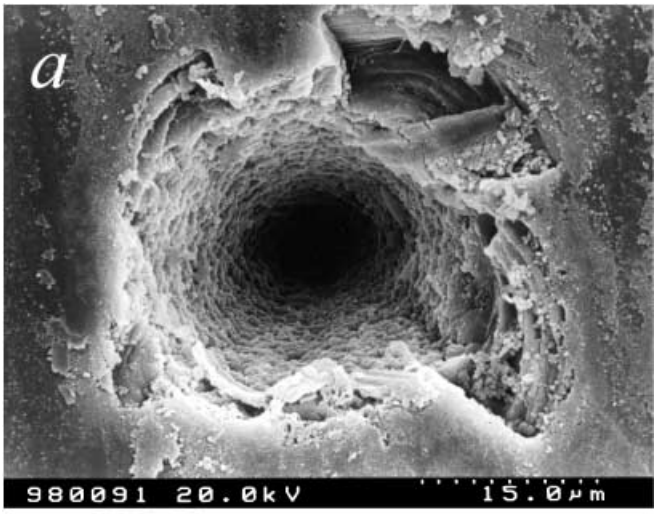
As mentioned in the introduction, there are a tremendous variety of applications which by far cannot and even should not be discussed within the context of this paper. Every kind of application performed so far by classical ultra-short-pulse laser systems (e.g. Ti:sapphire-based regenerative amplifier systems) can also be or will eventually be covered, maybe in a simpler and finally cheaper setup, by modern compact diode-pumped systems. This even applies to the largest ultra-short-pulse systems conceived so far, like for laser fusion including a fast ignitor concept (e.g. compare the Mercury Program at Lawrence Livermore National Laboratory [130]). Wavelengths so far being generated by lasers which are not realistic candidates for diode pumping might in the future be produced by diode-pumped systems involving parametric or other nonlinear wavelength-conversion processes being based on high-average-power ultra-short-pulse lasers and e.g. tailorable nonlinearities by quasi phase-matching of poled dielectrics [131]. Even terahertz radiation can be effectively generated by the use of femtosecond laser pulses [132].

The main emphasis in the following should be put on examples for successful applications of ultra-short pulses for which a wide dissemination will be important, crucially depending on the availability of compact, robust, maintainance-free and moderately priced laser systems. Generally, this does not apply in any strict sense to fundamental research in various fields like physics, metrology, chemistry, biology or medicine. However, applied sciences and the high-tech market derived from them will be the range where diode-pumped

ultra-short laser systems see their future. This will be in the first place materials processing covering a variety of substances like metals, dielectrics and even biological materials like organic tissue, but also biological science, medical diagnosis and therapy. Examples are given in the following:

Ablation of materials has been studied in a time-resolved way for a long time, thereby enabling the understanding of the physical processes more and more [133, 134]. Nowadays, it is proven that ablation by ultra-short laser pulses allows precise microstructuring of metals [135, 136], dielectrics [38, 137] or biological tissue [138–140]. The main advantage can be seen in the absence of thermal or acoustic shock leaving the remaining material in a basically unchanged condition. Figure 5 shows a microstructured sample of tungsten. As can be seen easily, there is no trace of melting and resolidification as is usually shown associated with nanosecond pulses [37, 141]. Nevertheless, there is still some controversy if ultra-short laser pulses are indispensable for such high quality of ablation during microstructuring. A recent paper at CLEO'2000 has shown comparable results achieved by nanosecond pulses indicating that the fluence with respect to threshold plays a major role [142]. Going to even finer dimensions, nanostructuring of metal films has been achieved when applying spatially localized femtosecond laser pulses [143], representing a combination of scanning optical near-field microscopy with ultra-short-pulse lasers.

For dielectrics like fused silica a clear dependence of damage-threshold fluence on pulse duration was measured in [38] as well as the dependence of ablated volume on fluence. Figures 6a and 6b give a comparison of the shape of the ablation crater and its depth for the same number of pulses ($N = 80$) at $\lambda = 780$ nm for two different pulse durations, 3 ps and 20 fs, respectively. The different precision of the ablated edges should be noticed, which may be understood by considering the different processes getting involved. When applying ps pulses at the given fluence level fewer multi-photon processes can occur than in the fs case so that impact ionization has to start mainly from the existing carriers having little density in such a high band gap dielectric like fused silica at room temperature ($\sim 10^8\,\mathrm{cm}^{-3}$). Therefore, some stochastic processes influence the nature of ps ablation shown here.



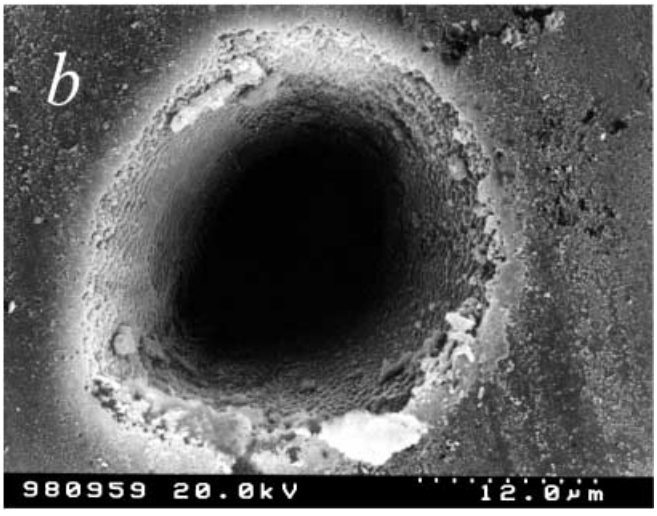


Fig. 6a,b. Ablation of fused silica by ultra-short pulses out of a Ti:sapphire oscillator–amplifier system at $\lambda = 780$ nm using $N = 80$ pulses. **a** $\tau_p = 3$ ps, fluence $19.9\,\mathrm{J/cm^2}$, depth $\approx 38\,\mu$m; **b** $\tau_p = 20$ fs, fluence $11.1\,\mathrm{J/cm^2}$, depth $\approx 20\,\mu$m. Photograph courtesy of M. Lenzner, Technische Universität Wien, Vienna, Austria

Fig. 5. Example for microstructuring of metals by femtosecond pulses: ablation of tungsten by amplified femtosecond Ti:sapphire pulses of pulse duration $\tau_p \approx 100$ fs, pulse energy $E_p \approx 1$ mJ at 1-kHz repetition rate at $\lambda = 780$ nm [135]. Photograph courtesy of S. Nolte and B.N. Chichkov, Laser Zentrum Hannover, Hannover, Germany

In the case of biological samples representing thermally very sensitive material in most cases, like dental tissue, the high efficiency of femtosecond ablation is crucial, allowing a reasonable speed of material removal without heating the tooth with its sensitive pulp beyond a critical level [144]. Figure 7 demonstrates the precision as well as the possible speed of ablation of dental enamel under the mentioned precautions [40]. Still, the speed of material removal is slower than by conventional mechanical drilling. It is the laser system, however, which represents a bottleneck by its 1-kHz repetition rate. There is a good chance that modern high-average-power laser systems will overcome this limitation. So far, no diode-pumped compact laser system has yielded the required results. But there are clear indications that such applications will be possible with one of the ultra-short-pulse laser systems described above, e.g. the femtosecond thin-disk laser.

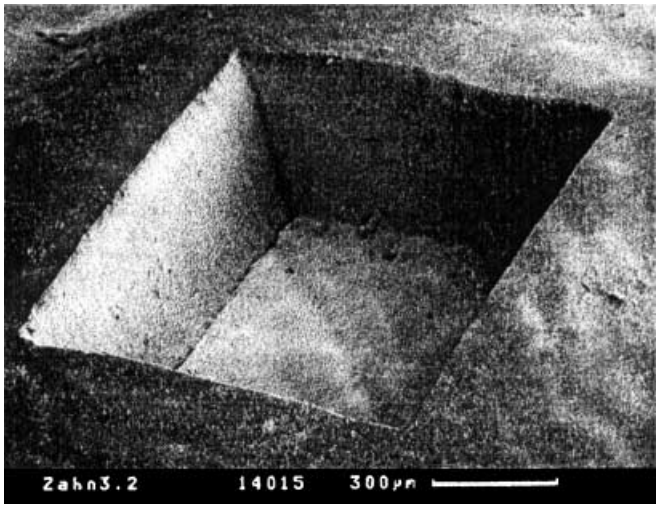


Fig. 7. Ablated cavity in human dental enamel: $\tau_p = 700$ fs, $E_p = 100\,\mu$J, repetition rate 1 kHz, depth $\sim 400\,\mu$m, scanning speed 10 mm/s, rate of material removal 0.04 mm^3/min. Photograph courtesy of A. Kasenbacher, Traunstein, Germany

The advantages of ultra-short-pulse micro- and nanostructuring can be seen in the possibility of efficient, fast and spatially selective coupling of optical energy into all samples which might also be transparent, i.e. linearly nonabsorbing. Very high precision can be yielded associated with minimal thermal and acoustic damage. It is also a contact-free method exerting no mechanical pressure and thus representing no risk of infection, which is essential in surgery. Hence, impressive results on cutting by nonthermal ablation of e.g. neural tissue were achieved, e.g. [140]. In the case of transparent biological samples like the cornea the nonlinear absorptive interaction allows a very good depth resolution of a few micrometers, opening the new possibility of precise internal cutting by lateral scanning of the focused ultra-short pulses, establishing a new technique for corneal shaping under conservation of epithelial layers [145]. The fast, direct and precise ablation by ultra-fast pulses also allows applications in the field of data storage, e.g. the fabrication of masters for DVDs without involving complicated multi-step photolithographic processes [146].

Ultra-short pulses are generally associated with nonlinear effects in matter, like nonlinear absorption or frequency conversion. As the yield in the confocal area is by far the highest, three-dimensional resolution can be achieved and consequently applied. This is well known in nonlinear microscopy, where the new contrast in comparison to linear microscopy might be based on second- or third-harmonic generation or fluorescence after two- or three-photon absorption, which originates from a well-defined tiny volume with dimensions of approximately the focal area and the focal depth [147]. Hence even thicker microscopic samples can be investigated in more detail. Based on similar considerations, rapid prototyping involving polymerization of monomers by irradiation with blue or UV light could be made much faster and hence more efficient if this hardening process would not be initiated by two-dimensionally structured illumination of surfaces generating patterns layer by layer. The three-dimensional figure could be written within the volume of the monomer via polymerization initiated by nonlinear absorption of ultra-short laser pulses [148]. This procedure resembles somewhat the way in which three-dimensional figures within glass cubes are engraved via optical breakdown spots (for their fabrication ns pulses are sufficient, however).

A very challenging new field of application of the shortest femtosecond pulses is optical coherence tomography. This method originally involved superluminescent diodes [10] having a rather short coherence length defining the resolution of measurement, originally of distances between optically different layers in the eye, like the thickness of the cornea or the depth of the eye down to the retina [149]. Soon, two-dimensional scanning of the background of the eye was achieved and, furthermore, this method was applied to a variety of scattering biological media like skin tissue or dental tissue [150]. A major step forward was made by taking advantage of the short coherence length of femtosecond pulses, which is in the µm regime and hence about one order of magnitude shorter than the one of superluminescent diodes but associated with the excellent brightness of a laser source. Figure 8 shows a very simplified scheme of a modern fiber-based optical coherence tomograph [13]. It seems to be worth mentioning that there is no problem or even contradiction when considering $\sim$ 10-fs pulses with a bandwidth of about 120 nm propagating through non-negligible lengths of optical fiber associated with a lot of dispersion. It is the coherence length and bandwidth which remain unchanged even if the originally bandwidth-limited pulses from the laser are torn apart by dispersion. A compensator within the reference arm should provide approximately equal dispersion conditions in both the measurement and the reference arms. In this way a resolution of $\sim 1\,\mu$m in the longitudinal direction is achieved, as can be seen in Fig. 9 showing sub-cellular details of African-frog tissue (Xenopus laevis [151]) with a lateral resolution of 3 µm. This most modern version of OCT, although so far only performed by Ti:sapphire pulses around 800 nm penetrating about 1 mm into normal tissue, seems to be ideally suited for the application of diode-pumped femtosecond lasers like Cr^{3+}:LiSAF or LiSGaF, as they can be directly diode-pumped and hence are substantially simpler and potentially cheaper. Their penetration depth is also somewhat deeper due to the center wavelength being about 100-nm longer. The average power needed for OCT lies in the sub-10mW regime; usually it is 3–5 mW, representing a rather modest requirement in general. For opthalmological appli-

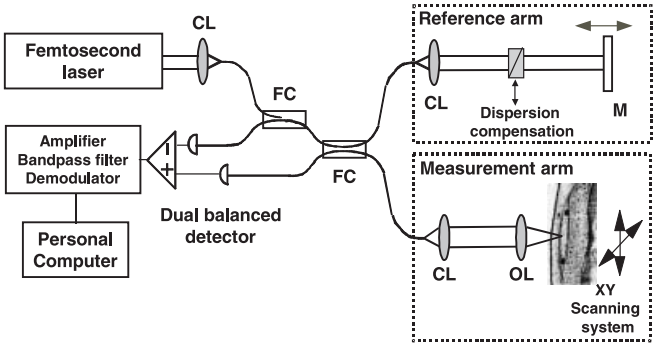


Fig. 8. Fiber-based setup for optical coherence tomography by femtosecond laser pulses comprising measurement and reference arms. CL, collimating lens; FC, fiber coupler; OL, objective lens; M, adjustable reference mirror. Courtesy of W. Drexler, Universität Wien, Vienna, Austria

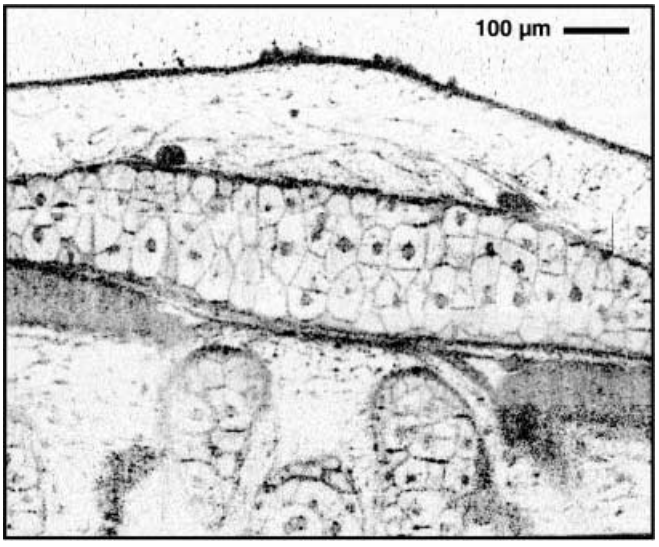


Fig. 9. In-vivo sub-cellular-resolution optical coherence tomography of Xenopus laevis (African frog). The scanned image area is $0.83 \times 1\ \mathrm{mm}^2$; the image consists of 1800×1000 pixels. Resolution: laterally 3 µm, longitudinally 1 µm. Measurement performed with a Ti:sapphire laser delivering 5.4-fs pulses having 250-nm bandwidth [4]. Courtesy of W. Drexler

cations only 300 µW are needed and only ∼1 µW would be permissible for safety reasons [152].

The implementation of OCT together with compact and moderately priced femtosecond laser sources in several wavelength regimes would be highly desirable: there are absorption windows around 950 nm, 1300 nm (yielding approximately 4-mm penetration depth) and 2200 nm. If also taking advantage of secondary information like spatially resolved dispersion and absorption this will revolutionize in vivo nondestructive medical diagnosis, being applicable to many areas of interest via endoscopic methods.

5 Conclusions and outlook

In the preceding sections it was shown that there are quite a variety of good material candidates for diode-pumped ultrashort-pulse laser operation. Most successful ultra-broad-band solid-state lasers emitting shortest pulses in the 10-fs regime and above have been realized by employing transition-metal-ion-doped crystals like Cr^{3+}:LiSAF or Cr^{3+}:LiSGaF. Much more work will have to be done using other crystals with prospective properties for fs-pulse generation, like Cr^{4+}:YAG or Cr^{2+}:ZnSe, thereby creating a very high demand towards high-brilliance, high-power laser diodes only gradually becoming available until now. When taking advantage of conventional high-brilliance pump lasers, first results showing the potential of a new laser material often could be demonstrated. Furthermore, important features within the resonator taking place in the sub-20-fs regime, like a systematic Raman shift towards longer wavelengths or the occurrence of side bands associated with higher-order dispersion, could be studied. If, like several applications suggest, moderately short pulses around 1 ps are within the focus of interest, rare-earth-doped crystals and glasses are the media of choice. In glasses the bandwidth is wider allowing shorter pulses, but there are limitations of heat conduction and hence output power. This partly can be circumvented by taking advantage of a typical weak point of existing diode lasers: the elliptical focus can be brought to a good overlap with a suitable resonator mode thereby yielding a rather linear heat flow out of the lasing area. Other choices are the design of crystalline laser media like mixed garnets, whose internal crystal fields sufficiently broaden the spectra allowing pulses in the 200-fs regime. Ultimately, specific wavelengths seem not to be in the focus of interest for high-average power ps or sub-ps diode-pumped lasers. Therefore the main emphasis can be put on high efficiency of the laser material and the high-power pump diodes. In this respect, the mode-locked Yb:YAG thin-disk laser is, for the time being, the best compromise of all the mentioned requirements. It can, consecutively, be wavelength-converted by standard nonlinear parametric or harmonic methods. Even terahertz generation is feasible. Therefore, this power-scalable ultra-short-pulse laser can become a workhorse for a wide range of applications.

Many applications have been mentioned in Sect. 4, which can also be grouped into two classes: the ones requiring ps and sub-ps pulses mainly involve ablation for micro- or even nanostructuring of materials. In this context the medical-application sector offers a tremendous potential. This might be either surgical cutting of tender tissue like neural, or heat-sensitive preparation of cavities in dental enamel or dentine. Taking advantage of the three-dimensional resolution of nonlinear processes, nonlinear microscopy or even sub-epithelium cutting within the cornea can become a routine application of the near future. Medical diagnosis might become revolutionized by advanced optical coherence tomography, allowing us to resolve sub-cellular details in a nondestructive in-vivo fashion. By varying the wavelength according to the several water windows even substantial penetration depths of a few millimeters will eventually be possible, given the development of advanced femtosecond diode-pumped laser systems. The enormous bandwidth of much more than 100 nm will allow us to analyze even further optical properties like absorption and index of refraction in a spatially resolved way and put a challenge to the medical specialists for interpretation.

Eventually, even large laser facilities involving short or ultra-short laser pulses will be fully reliant on diode pumping. Laser-driven inertial fusion programs like Mercury and its successors in the Lawrence Livermore National Laboratory will be totally based on generating optical energy via diode-pumped Yb crystals. Some specialists see the future of accelerators based on femtosecond high-power lasers offering unprecedented particle accelerations, which could substantially reduce the cost for the required size of building and apparatus.

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