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Recent advances in crystal optics/Avancées récentes en optique cristalline

New laser crystals for the generation of ultrashort pulses

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Abstract

Since the beginning of the 1990s, titanium sapphire has become the crystal of choice for the development of ultrashort laser systems producing very short and powerful pulses using the Chirped Pulse Amplification technique. In parallel to these developments leading to commercial products, new laser crystals have been studied in order to reach directly other wavelength ranges and to overcome the need to develop cw or pulsed green laser to pump the titanium sapphire crystal. In order to be able to directly pump the crystals with a very efficient and high-power semiconductor laser, new crystals doped with chromium or ytterbium ions have been developed. This article will review the latest development in this research field with the best performances obtained in terms of pulse duration. ***To cite this article: F. Druon et al., C. R. Physique 8 (2007).***

Résumé

Nouveaux cristaux laser pour la génération d'impulsions ultra-brèves. Le saphir dopé au titane est devenu le cristal de référence pour le développement de systèmes laser produisant des impulsions ultra-brèves et de forte puissance crête, grâce notamment à la technique d'amplification à dérive de fréquence. Ce cristal est donc à la base des lasers femtoseconde commerciaux actuels. Cependant, de nouveaux cristaux de laser ont été étudiés, d'une part, pour avoir accès directement à d'autres gammes de longueurs d'onde et, d'autre part, pour permettre le pompage direct par diodes laser de puissance et ainsi être beaucoup plus efficace qu'avec les lasers verts utilisés pour pomper le saphir dopé au titane. Pour cela, de nouveaux cristaux dopés au chrome ou à l'ytterbium ont été développés. Cet article se propose de faire un état de l'art des derniers développements dans ces domaines de recherche en mettant en avant les meilleures performances obtenues en termes de durée d'impulsion. ***Pour citer cet article : F. Druon et al., C. R. Physique 8 (2007).***

Keywords: Laser; Femtosecond pulses; Laser crystals; Diode-pumping

Mots-clés : Laser ; Impulsions femtoseconde ; Cristaux laser ; Pompage par diode laser

1. On the progress of femtosecond laser technology

Ultrafast laser technology involving pulses of light with durations of typically a few tens of femtoseconds (10^{-15} s) has become one of the most active fields in physics. This technology dealing with very short pulse durations and a very high temporal confinement of the energy (implying giant electromagnetic fields) has actually many applications. These

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applications cover numerous fields of physics, engineering, chemistry, biology and medicine, with applications such as time-resolved spectroscopy, multiphoton imaging, micromachining, refractive surgery, acceleration of particles, X-rays generation, fusion, etc. These applications have in common the use of one or a few specific and very interesting properties of the femtosecond laser pulses: femtosecond pulses allow a very precise time resolution; their strong electric field induces important and unusual non-linear effects; the localization of these non-linear-effects is very confined (in the 3D) which allows a high spatial resolution. Thanks to the confinement of the energy it is possible to obtain a high peak power without necessarily having a high average power.

The production of ultrashort pulses is based on locking in phase many longitudinal modes in a laser cavity. When these modes are locked together, there is constructive interference which leads to short pulse generation. These lasers are the so-called mode-locked laser oscillators. Since the first generation of mode-locked lasers [1–4], in 1964, numerous major evolutions occurred. The first main evolution took place in the 1970–80s with the use of femtosecond dye lasers [5,6]: this is the second generation of mode-locked lasers. However, perhaps the most important evolution concerns the breakthrough that occurred in the early 1990s with the emergence of mode-locked lasers using a new solid state laser medium: titanium-doped sapphire (Ti:sapphire) [7,8]. This event really marks the beginning of the third generation [9] of femtosecond lasers: the all-solid-state self-mode-locked femtosecond lasers. Such tunable femtosecond lasers have opened the area of non-linear microscopy for biological imaging. In combination with the chirp pulse amplification technique (CPA) [10,11], these lasers actually revolutionized the physics of ultrafast phenomena thanks to a high improvement in terms of reliability, efficiency and compactness. They have allowed the production of ultrashort ultrahigh-power (up to the petawatt: 10^{15} W) pulses which correspond to the beginning of ultrahigh-intensity physics and to a large expansion of femtosecond laser chain applications.

The aim of this article is to make a review of the major crystals involved in this latest generation of mode-locked lasers. In a first part, we will present the different criteria that define a crystal 'well-suitable' for the development of femtosecond mode-locked lasers; then in a second part, the main properties of these crystals will be described. The last part will give a state of the art of the main performance obtained with these crystals in mode-locked oscillators.

2. Optimization strategy of crystals for developing ultrashort pulsed lasers

2.1. Spectroscopic properties of the emission bands

The most important point when dealing with ultrashort pulses is related to the conservation of the time-bandwidth product. In fact, the time-bandwidth product has to satisfy

$$\Delta t \Delta v \geqslant 0.32 \tag{1}$$

where and are the full width half maximum (FWHM) duration and spectrum of the pulse, respectively. The equality in Eq. (1) corresponds to transform-limited pulses (and the strict inequality corresponds to chirped pulses). Eq. (1) means that, if one wants to generate ultrashort pulses, the laser crystal has to be able to allow amplification of very broad spectra. The emission bandwidth of the crystal has to be in the order of the spectral bandwidth of the pulse. The influence of the crystal host on the broadening of the spectral lines for a given doping ion is fundamental. On the one hand, the crystal host matrix will induce on the doping ion an electric field that tends to spread its energy level distribution. This broadening effect may be emphasized if the doping ions are inside different types of sites in the host matrix. On the other hand, a strong electron–phonon coupling (or coupling by vibronic transitions) in the crystal will homogeneously broaden the electronic lines. The addition of these two phenomena (splitting of the energy levels and electron–phonon coupling, effects that strongly depend on the crystal) are the main effects that lead to very broad and smooth emission spectra required for developing femtosecond lasers. These phenomena are thus crucial in the choice of a crystal in the development of an ultrashort pulsed laser.

For laser efficiency, it is also crucial to consider the emission cross-section (σ_{em}) and the lifetime of the upper state (τ) of the laser transition. The higher the product $\sigma_{\mathrm{em}}\tau$ is, the lower the laser threshold will be. In conclusion, a crystal well adapted for the development of an efficient ultrashort pulsed laser, will require both high peak and very broad emission band. However, these two criteria are often in contradiction and a compromise has to be made between short duration and efficiency.

2.2. Other important criteria

For the development of femtosecond lasers, the evaluation of the spectral emission properties of a crystal is not sufficient. Many other criteria have to be taken into account as well. Other criteria, to which it is important to pay attention, are, for example, thermal properties, excited-state absorption levels or absorption properties.

For developing efficient, reliable and high-power oscillators, the thermal properties play a key role. In fact, if the crystal tends to store too much heat during the pumping process, some problems of efficiency, stability or even fracture of the crystal may occur. A good thermal conductivity is thus an essential condition in the strategy of choosing a laser crystal. Moreover, it is interesting to notice that this good conductivity condition tends to frustrate, in the case of rare-earth crystals, the high electron–phonon coupling that is needed to obtain a broad emission band. Here again a compromise has to be made between short duration and efficiency. This is the major reason why doped glasses exhibit broader emission bands than doped crystals (for a given doping ion). This is due to the high structural disorder of the glass host. However, glasses are less attractive than crystals; their efficiency is relatively weak because of their poor thermal conductivity and low emission cross sections.

Another spectroscopic parameter which plays an important role in laser efficiency is the influence of possible excited states at energy levels higher than the laser transition. In fact, this can lead to excited state absorption, concentration quenching and up conversion, which are very deleterious phenomena for laser efficiency.

The last important parameter which is interesting to focus on, is the absorption spectrum of laser crystals. Actually, the technological breakthrough brought by the use of high-power laser diodes for the optical pumping of laser crystals is considerable in terms of simplicity, reliability, efficiency and compactness. The development of directly diode-pumped femtosecond lasers is then one of the most significant improvements of the last decade in the laser technology. This latest generation of femtosecond lasers (which could be called the fourth generation of mode-locked lasers) is in full expansion and is very promising for many applications. From this point of view, the absorption bands of the laser crystal must fit the emission spectra of high power diodes. The high power diodes available generally emit around 800 nm for GaAlAs/GaAs type semiconductor lasers and around 900 nm–1 μm for InGaAsP/InGaAs type semiconductor lasers; some power diodes (but less efficient) are also available in the red around 670 nm with InGaAlP/GaAs diodes and in the far infrared around 1600 nm with InGaAsP/InP diodes. Laser crystals exhibiting an absorption band in one of these regions are thus very interesting.

3. Major crystals in the ultrafast technology

The classification of the crystals presented is based on the development maturity of the femtosecond technology using these crystals: starting with crystals used for the development of the all-solid-state self-mode-locked femtosecond lasers (Sections 3.1 and 3.2), followed by crystals involved in the development of the diode-pumped all-solid-state self-mode-locked femtosecond lasers (Sections 3.3 to 3.4), and concluding by new crystals with a high potential (Sections 3.5 and 3.6).

3.1. Titanium doped sapphire

The Ti:sapphire (Ti^{3+}:Al_2O_3) was discovered in 1986 at the Lawrence Livermore Laboratory by Peter Moulton [7] and quickly became the reference crystal in the realm of femtosecond laser technology. At that time, the femtosecond laser technology was mainly based on femtosecond dye lasers. Thanks to its remarkable properties—very broad emission band from 650 to 1080 nm (shown in Fig. 1) and a very high thermal conductivity (34 $W\,K^{-1}\,m^{-1}$)—Ti:sapphire was a good candidate to replace the femtosecond dye lasers. Up to now, in the actual state of the art, Ti:sapphire has allowed the production of the shortest pulses ever produced from a laser oscillator. Moreover, Ti:sapphire technology allows the production of very intense pulse generation up to the petawatt level in CPA-based amplifiers. This is why it has become the standard crystal of modern commercial femtosecond systems.

The laser transition in Ti:sapphire is between the ^2E and $^2T_2$ levels of the Ti^{3+} ion. The Al_2O_3 matrix (whose crystal structure is hexagonal rhombohedric) allows a broad emission band by a splitting of Ti^{3+} levels due to the combination of effects such as cubic-field, trigonal-field, spin-orbit interaction, Jahn–Teller effects leading to a strong vibronic-transition coupling.

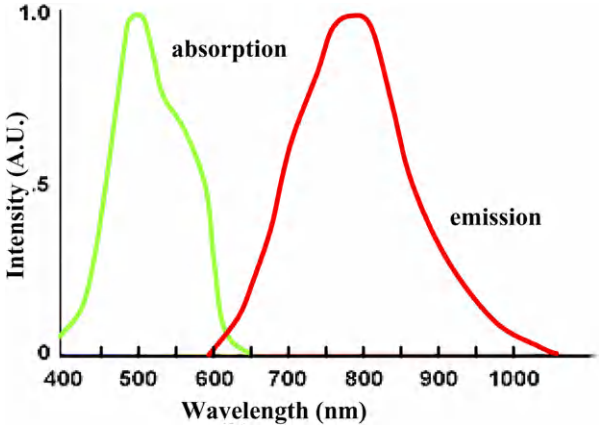

Fig. 1. Emission and absorption spectra of Ti:sapphire.

The main disadvantage of the Ti:sapphire is related to its absorption spectrum (presented in Fig. 1) which is located around 500 nm. It is thus impossible to directly diode pump the Ti:sapphire crystal. In the actual state of the art, the most efficient way to pump it is to use a diode-pumped Nd:YAG (or Nd:YVO_4) laser emitting at 1064 nm and to convert its beam in the green by second harmonic generation. The total electrical-to-optical efficiency of Ti:sapphire systems is then quite low, which leads to important problems for the developing of high average power Ti:sapphire lasers.

3.2. *Cr^{4+}-doped crystals*

For some applications, it is very interesting to have femtosecond lasers emitting at longer wavelengths than the Ti:sapphire. Materials doped with Cr^{4+} ions are thus very attractive because they emit in the 1.3–1.5 μm region which corresponds to an eye-safe region. Another advantage of Cr^{4+}-doped crystals consists in their relatively broad emission spectra (Fig. 2). Moreover, Cr^{4+} ions have an absorption line around 1064 nm (Fig. 2), which is very interesting because this line corresponds to the emission peak of Nd-doped-crystal lasers (or Yb-doped fiber lasers) that are very efficient diode-pumped lasers. The other characteristics of the Cr^{4+}-typical crystal are summarized in Table 1.

The typical Cr^{4+}-doped crystals used for developing femtosecond oscillators are the Cr:YAG [12] (Cr^{4+}:$Y_3Al_5O_{12}$) and the Cr:forsterite [13] (Cr^{4+}:Mg_2SiO_4 whose emission and absorption spectra are plotted in Fig. 2 as an example of spectroscopic properties of a Cr^{4+}-doped crystal). In these crystals, the laser transition is between the $^3T_2$ and $^3A_2$ levels of the Cr^{4+} ion. This corresponds to a peak laser emission at 1250 nm for the Cr:forsterite and 1450 nm for the Cr:YAG. As for the Ti:sapphire, the vibronic-transition coupling plays an important role in the spectral broadening.

However, Cr^{4+}-doped crystals reveal some important problems. First, as shown in Fig. 2, the absorption and emission spectra strongly overlap. This leads to a quasi-4-level type laser which presents re-absorption at the laser

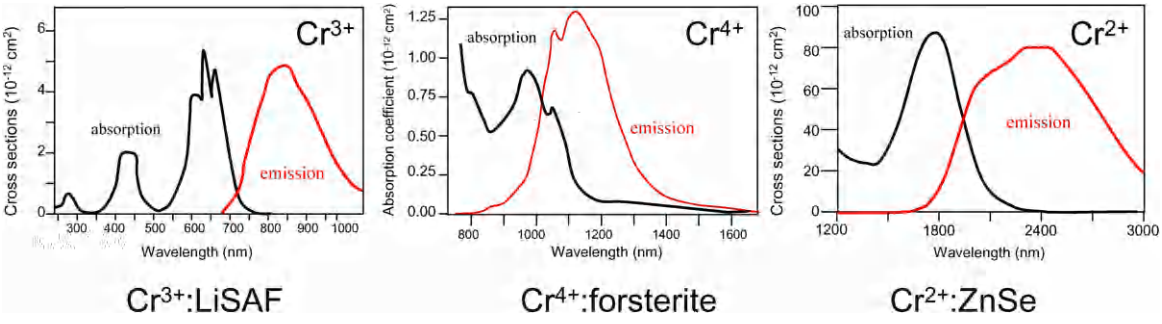

Fig. 2. Emission and absorption spectra (from left to right) of three Cr^{3+}-, Cr^{4+}-, Cr^{2+}-doped crystals.

Table 1
Main properties of laser materials involved in the development of femtosecond lasers

Material	Emission band width (FWHM)	Minimum theoretical duration	Central emission peak	Absorption (usual pumping)	Diode pumping	Emission cross-section (10^{-20} cm^2)	Fluorescence lifetime	$\sigma_{em}\tau$ (μs cm^2)	Thermal conductivity (undoped) (W K^{-1} m^{-1})
Ti:sapphire $Ti^{3+}:Al_2O_3$	180 nm	3.6 fs	790 nm	∼500 nm	No	41	3.2 μs	131	34
Cr:forsterite $Cr^{4+}:Mg_2SiO_4$	150 nm	11 fs	1250 nm	1064 nm	No	20	3 μs	60	5
Cr:YAG $Cr^{4+}:Y_3Al_5O_{12}$	200 nm	11 fs	1450 nm	1064 nm	No	20	4 μs	80	11
Nd:glass	22 nm	53 fs	1053 nm	808 nm	Yes	4	360 μs	1440	0.8
Cr:LiSAF $Cr^{3+}:LiSrAlF_6$	100 nm	7.6 fs	850 nm	670 nm	Yes	4.8	67 μs	322	3.1
Cr:LiCAF $Cr^{3+}:LiCaAlF_6$	100 nm	6 fs	760 nm	670 nm	Yes	1.3	175 μs	228	5
Cr:LiSGaF $Cr^{3+}:LiSrGaF_6$	100 nm	7.2 fs	830 nm	670 nm	Yes	3.3	80 μs	264	4
Yb:YAG $Yb^{3+}:Y_3Al_5O_{12}$	9 nm	124 fs	1031 nm	942 nm	Yes	2.1	951 μs	1997	11
Yb:glass	35 nm	31 fs	1020 nm	975 nm	Yes	0.05	1300 μs	65	0.8
Yb:GdCOB $Yb^{3+}:Ca_4GdO(BO_3)_3$	44 nm	26 fs	1044 nm	976 nm	Yes	0.35	2600 μs	910	2.1
Yb:BOYS $Yb^{3+}:Sr_3Y(BO_3)_3$	60 nm	18 fs	1025 nm	975 nm	Yes	0.3	1100 μs	330	1.8
Yb:KGW $Yb^{3+}:KGd(WO_4)_2$	25 nm	44 fs	1023 nm	981 nm	Yes	2.8	600 μs	1680	3.3
Yb:KYW $Yb^{3+}:KY(WO_4)_2$	24 nm	46 fs	1025 nm	981 nm	Yes	3	600 μs	1800	3.3
Yb:SYS $Yb^{3+}:SrY_4(SiO_4)_3O$	73 nm	16 fs	1040 nm	979 nm	Yes	0.44	820 μs	361	2
Yb:YVO_4	30 nm	36 fs	1008 nm	984 nm	Yes	1.25	250 μs	312	5.1
Yb:CaF_2	30 nm	36 fs	1047 nm	980 nm	Yes	0.25	2400 μs	600	9.7
Yb:CALGO $Yb^{3+}:CaGdALO_4$	80 nm	14 fs	1050 nm	980 nm	Yes	0.8	420 μs	336	6.5
Cr:ZnSe $Cr^{2+}:ZnSe$	600 nm	11 fs	2500 nm	1600 nm	Yes	90	7 μs	630	16
Cr:ZnS $Cr^{2+}:ZnS$	500 nm	12 fs	2350 nm	1600 nm	Yes	140	4.5 μs	630	27.2

wavelengths. This re-absorption effect tends to limit the broadness of the effective emission band. Another important limiting effect in Cr^{4+}-doped crystals is the existence of (higher) excited state levels ($^3T_1$ band). This leads to an excited state absorption (from $^3T_2$ to $^3T_1$) at the pump and laser wavelengths which is a strong deleterious effect for the laser efficiency. Moreover, efficiency of Cr^{4+}-doped crystal lasers is very sensitive to temperature because of an important thermal quenching of fluorescence (which corresponds to the decrease of the lifetime by non-radiative transitions versus temperature) [14,15]. Finally, the Cr^{4+} doping in YAG and in forsterite often leads to severe problems because of a strong competition between Cr^{4+} and Cr^{3+}. In fact due to the existence of two different site geometries: octahedral (favoring Cr^{3+}) and tetrahedral (favoring Cr^{4+}), chromium ions can be found in both 4+ and 3+ oxidation states, which frustrates the Cr^{4+} doping and modifies the spectral properties of the crystal [16].

3.3. Nd^{3+}-doped materials

To improve further laser efficiency, the use of directly diode-pumped laser seems the paramount method. Nd^{3+}-doped crystals (in particular Nd:YVO_4 and Nd:$Y_3Al_5O_{12}$ or YAG) are well known to have the possibility to be directly diode pumped at 808 nm and to be very efficient laser materials. However, the spectral bandwidth of Nd-doped

materials is too narrow to allow the production of ultrashort pulses. For example, the Nd:YAG that has a 0.8-nm-broad emission bandwidth, cannot produce pulses shorter than 1.5 ps. The only way to broaden the emission spectrum with a Nd-doped materials is to use glasses (Table 1) but in these cases, the emission cross-section and the thermal conductivity are very low and it is thus very difficult to develop a high-power femtosecond laser. However, there is some commercial femtosecond laser based on Nd:glass with limited output power [17,18].

3.4. Cr^{3+}-doped colquiriites crystals

An interesting family of crystals that can be directly diode pumped and that has broad emission bands are Cr^{3+}-doped colquiriites [19,20]: Cr:LiSAF (Cr^{3+}:$LiSrAlF_6$), Cr:LiSGaF (Cr^{3+}:$LiSrGaF_6$) and Cr:LiCAF (Cr^{3+}:$LiCaAlF_6$). In fact, as shown in Fig. 2, with the example of the Cr:LiSAF case, these crystals combine a broad absorption peak around 650 nm (including a line at 670 nm where InGaAlP/GaAs laser diode emits) and a wide emission spectrum from 750 to 1000 nm. This broadness of the laser transition is mainly due, as for the Cr^{4+}-doped crystal, to a strong vibronic transition coupling. The laser transition is between the $^4A_2$ and $^4T_2$ of the ion Cr^{3+}. This corresponds to a peak laser emission around 850 nm. The other characteristics of the Cr:LiSAF, Cr:LiCAF and Cr:LiSGaF are summarized in Table 1.

Despite all these promising advantages, the results obtained in laser operation by Cr^{3+}-doped colquiriites are very poor in terms of average power. In fact, Cr^{3+}-doped colquiriites have many drawbacks that strongly counteract the diode-pumping efficiency [21,22]. First, like Cr^{4+}, Cr^{3+} exhibits higher excited states levels ($^4T_{1a}$ band) [23]. This band leads to various deleterious effects such as excited state absorption and up-conversion. These effects not only affect the laser efficiency by decreasing the population inversion, but also generate heating by non-radiative transitions. This heating (which is emphasized by the relatively poor thermal conductivities of colquiriites) is disastrous in the case of Cr^{3+}-doped colquiriites. In fact, Cr^{3+}-doped colquiriites show an important thermal quenching of fluorescence. When the temperature in the crystal increases, the laser performance then rapidly drops thus preventing an efficient laser emission.

3.5. Yb^{3+}-doped crystals

Another family of interesting crystals that allows diode-pumping is that of the Yb^{3+}-doped crystals [24,25]. Owing to its simple electronic structure based on two electronic manifolds ($^2F_{7/2}$–$^2F_{5/2}$), the ytterbium ion exhibits advantageous laser properties. First, the low quantum defect of this ion reduces the thermal load and thus the thermal problems. Second, the absence of additional parasitic levels in Yb^{3+} prevents undesired effects such as up-conversion, excited-state absorption and concentration quenching. Consequently, ytterbium provides the possibility of a high doping configuration. Moreover, the absorption band in ytterbium-doped media (ranging from 900 to 980 nm as shown, for example, in Fig. 3) is covered by high-power InGaAs laser diodes. This enables direct diode-pumping and the

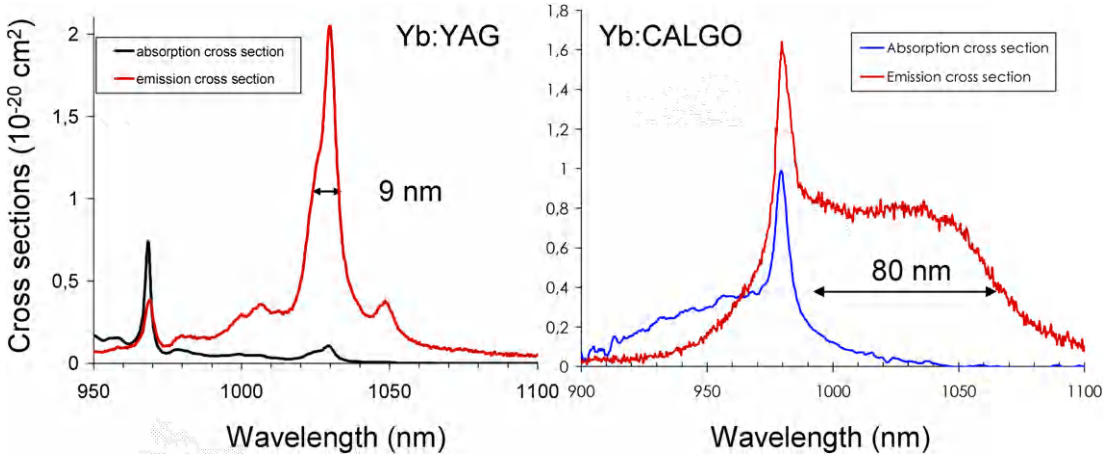

Fig. 3. Emission and absorption spectra of Yb:YAG and Yb:CALGO.

development of efficient and compact all-solid-state lasers. Moreover, Yb-doped materials exhibit—with respect to their Nd-doped counterparts (which are also rare-earth-ions-doped materials used for efficient diode-pumped lasers)—relatively broad emission spectra, which makes them very attractive for the development of ultrafast lasers.

All these advantages of the Yb^{3+}-doped crystals have led to a strong interest for many host matrixes favoring either short pulses or laser efficiency. During the past few years many interesting studies and results have been reported in literature; and Table 1 summarizes the properties of the most interesting Yb-doped crystals for the femtosecond laser development [26–33]: Yb:YAG (Yb^{3+}:$Y_3Al_5O_{12}$), Yb^{3+}:CaF_2, Yb^{3+}:YVO_4, Yb:BOYS (Yb^{3+}:$Sr_3Y(BO_3)_3$), Yb:SYS (Yb^{3+}:$SrY_4(SiO_4)_3O$), Yb:CALGO (Yb^{3+}:$CaGdAlO_4$), Yb:KGW (Yb^{3+}:$KGd(WO_4)_2$), Yb:KYW (Yb^{3+}:$KY(WO_4)_2$) and Yb:GdCOB (Yb^{3+}:$Ca_4GdO(BO_3)_3$). Fig. 3 shows the absorption and emission spectra of two different kinds: on the first hand, the highly peaked Yb:YAG spectra and the other hand, the very broad and smooth Yb:CALGO spectra (particularly well-adapted for the production of ultrashort pulses.) This last crystal is interesting because of its very broad and smooth emission band, in comparison to the Yb:YAG for example.

However, the Yb-doped crystals exhibit some disadvantages. First, there emission cross sections are in general relatively low and only the narrowest-emission-band materials (such as Yb:YAG or Yb:KYW, Yb:KGW) have a relatively high gain. Choosing the ideal Yb-doped material for femtosecond laser development strongly depends on the application: either favoring a crystal matrix host with high disorder (broad emission band but poor thermal conductivity and low emission cross-section) or favoring a crystal matrix host with low disorder (good thermal conductivity, high emission cross-section but narrow emission band) [34,35]. Another drawback of the Yb-doped crystals is the quasi-3-level structure of these lasers [36,37]; in fact, as shown in Fig. 3, there is a strong overlap between the emission and the absorption bands which leads to strong re-absorption effects and to a reduction of the effective emission band broadness. The last disadvantage of Yb-doped crystals is related to the very long fluorescence lifetime which implies some difficulties in the mode-locking process since the laser gain tends to store too much energy and to favor the Q-switched regime instead of the mode-locked regime [38].

3.6. Cr^{2+}-doped chalcogenide crystals: promising crystals for the far IR

For some applications, femtosecond sources delivering some atypical wavelengths are very interesting. For the 2.5 μm region, Cr^{2+}-doped chalcogenide crystals [39–42], such as Cr^{2+}-doped zinc selenide (Cr:ZnSe) or Cr^{2+}-doped zinc sulfide (Cr:ZnS), seem very suitable and are very promising. In fact—as shown in Fig. 2 with the example of Cr:ZnSe—these crystals have very smooth and broad emission bands (at least 500 nm wide—Table 1) centered at 2350 nm for ZnS and centered at 2500 nm for ZnSe. Moreover, they can be pumped at 1600 nm by InGaAsP/InP-type laser diodes. Furthermore, Cr^{2+} (contrary to Cr^{3+} and Cr^{4+}) does not suffer from deleterious effects due to excited state absorption and up-conversion. The other properties of these Cr^{2+}-doped crystals are summarized in Table 1.

4. Performance of different femtosecond oscillators

We will now make a tour of the performance obtained with femtosecond laser oscillators for the different laser crystals presented in Section 3. Due to the wide spectrum of ultrashort pulses, we know that managing the dispersion is probably the major concern for obtaining the shortest pulses from a given crystals. This is why the development of femtosecond mode-locked laser with a new crystal generally includes two steps. The first step involves the use of a versatile mode-locked oscillator with prisms compensator. This oscillator (Fig. 4(a)) allows one to demonstrate the mode-locked femtosecond regime with a pulse duration in the 50–100 fs range (except, of course, in the case where the emission spectrum of the crystal could not sustain so short pulses). However, this system does not give the shortest pulses obtainable with the crystal since the limitation is due, in this case, to higher-order uncompensated dispersion effects. In the second step—when the dispersion of the cavity including the crystal is fully characterized—a second generation of oscillators will allow one to obtain femtosecond pulses with a duration close to the limit imposed by the crystal emission bandwidth. This second generation of oscillators generally uses chirped mirrors (Fig. 4(b)) to compensate higher orders of the dispersion (one can notice the exception of the Cr:LiSAF for which, in very particular conditions, it is possible to compensate perfectly the dispersion only with prisms).

The experimental results for different crystals are listed in Table 2. One can notice that these results corroborate the advantages and drawbacks of the different crystals presented in Section 3 thanks to the analysis of the laser properties. Actually, the shortest pulses, up to now, have been obtained with a Ti:sapphire laser, with a duration around 5 fs. The

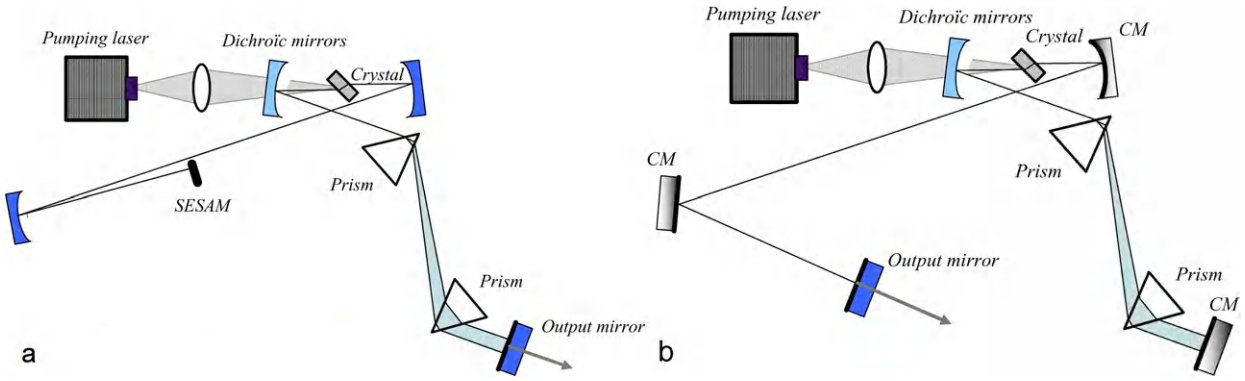

Fig. 4. Examples of typical mode-locked femtosecond laser cavities: using prisms to compensate the GVD (Group Velocity Dispersion) and a SESAM (SEmiconductor Saturable Absorber Mirror) to initiate the mode-locked (a), using both prisms and chirped mirror (CM) to compensate the GVD and KLM (Kerr Lens Mode Locking) to initiate the mode-locked (b).

shortest pulses from a directly diode-pumped laser were obtained with the Cr:LiSAF with duration of 11.5 fs but with an extremely low efficiency. The highest average powers and the best efficiencies have been obtained with Yb-doped materials; this explains the important growth of interest concerning these crystals whose potential has not been fully exploited hitherto in terms of pulse duration. Recently pulses as short as 47 fs have been obtained in a diode-pumped ytterbium-doped CALGO crystal, indicating the large potential of this kind of crystals. In the infrared, pulses as short as 106 fs have been very recently obtained with Cr:ZnSe indicating the huge potential of this crystal [43].

The interest for these femtosecond oscillators is very important and the most advanced developments have already led to commercial products. Thus Ti:sapphire lasers using prism sequence for GVD compensation are now very standard products [44,45], such as fully integrated and computer controlled femtosecond oscillators devoted to specific applications such as multiphoton microscopy [46,47]. The new technology involving Ti:sapphire with chirped-mirror dispersion compensator has also led to commercial products with even shorter pulses [48,49]. Moreover, directly diode-pumped femtosecond lasers also exhibit a rapid expansion on the femtosecond-source market. The main commercially available technologies until now is based on Nd-doped glasses [50,51], Yb-doped glasses [52] and Yb-doped tungstate crystals [69], but very promising systems using new Yb-doped crystal begin to emerge [53].

5. Conclusion

The breakthrough brought by new crystals very suitable for the production of ultrashort pulses has revolutionized the technology of ultrafast lasers and their applications. Actually, a first revolution occurred in the 1990s with the development of the first compact, efficient and reliable all-solid-state mode-locked lasers using Ti:sapphire; a second, which is now in full expansion—in particular thanks to new Yb-doped crystals—concerns the development of very efficient directly diode-pumped femtosecond lasers. These new generations of mode-locked lasers have allowed a fast expansion of applications and thus have raised a great interest.

In this article, we have tried to make a tour of the major crystals involves is these revolutions. These crystals demonstrated a very large range of laser properties. Their more remarkable advantages and drawbacks have been reported for each of them trying to highlight there specificities. A state of the art of performances has been then reported for the most relevant crystals. Some of these crystals such as Ti:sapphire, Cr^{3+}- and Cr^{4+}-doped crystals where work is most advanced, have already led to excellent and significant results with the production of pulses very close to the limit imposed by the crystal emission bandwidth. Others, such as Yb-doped crystals, have shown very innovating and significant results with a non-fully exploited potential. The last class of crystals (Cr^{2+}-doped crystals) includes crystals whose properties have exhibited very promising results in the infrared.

Future improvements need then to be made. First, a better knowledge and characterization of the most recent crystals is necessary in order to improve the laser performance of these crystals whose potential has not been fully exploited. Second, the development of new crystal hosts is still very important in order to extend to new application domains by finding crystals with atypical or better laser properties. Finally, to reach shorter durations, a method consisting in putting together several different crystals with slightly-shifted spectra—in order to artificially

Table 2
Main experimental results of laser materials obtained in femtosecond mode-locked laser oscillators

Material	Experimental duration	Central laser emission	Minimum theoretical duration	Average power	Diode pumped	Type of saturable absorber	Dispersion compensation system	Ref. (year)
Ti:sapphire	60 fs	880 nm	3.6 fs	450 mW	No	KLM	Prisms	[60] (1991)
Ti^{3+}:Al_2O_3	4.8 fs	790 nm		300 mW	No	SESAM	Chirped mirrors and prisms	[61] (1999)
Cr:forsterite	50 fs	1250 nm	11 fs	45 mW	No	KLM	Prisms	[62] (1993)
Cr^{4+}:Mg_2SiO_4	14 fs	1300 nm		80 mW	No	KLM	Chirped mirrors and prisms	[63] (2001)
Cr:YAG	20 fs	1420 nm	11 fs	400 mW	No	KLM	Chirped mirrors	[64] (2002)
Cr^{4+}:$Y_3Al_5O_{12}$	70 fs	1510 nm		50 mW	No	KLM	Anomalous Dispersion	[65] (1994)
Nd:glass	60 fs	1053 nm	53 fs	80 mW	Yes	SESAM	Prisms	[66] (1997)
Cr:LiSAF	90 fs	870 nm	7.6 fs	20 mW	No	KLM	Prisms	[67] (1994)
Cr^{3+}:$LiSrAlF_6$	11.5 fs	900 nm		6.2 mW	Yes	KLM	Prisms	[68] (1999)
Cr:LiCAF Cr^{3+}:$LiCaAlF_6$	9 fs	825 nm	6 fs	220 mW	No	KLM	Chirped mirrors and prisms	[69] (2002)
Cr:LiSGaF Cr^{3+}:$LiSrGaF_6$	14.8 fs	880 nm	7.2 fs	100 mW	No	KLM	Chirped mirrors	[70] (1997)
Yb:YAG	340 fs	1031 nm	124 fs	110 mW	Yes	SESAM	Prisms	[71] (1999)
Yb^{3+}:$Y_3Al_5O_{12}$	810 fs	1030 nm		60 W	Yes	SESAM	Chirped mirrors	[72] (2003)
Yb:glass	58 fs	1020 nm	31 fs	65 mW	Yes	SESAM	Prisms	[73] (1998)
Yb:GdCOB Yb^{3+}:$Ca_4GdO(BO_3)_3$	89 fs	1044 nm	26 fs	40 mW	Yes	SESAM	Prisms	[74] (2000)
Yb:BOYS Yb^{3+}:$Sr_3Y(BO_3)_3$	69 fs	1062 nm	18 fs	80 mW	Yes	SESAM	Prisms	[75] (2002)
Yb:KGW	112 fs	1045 nm	44 fs	200 mW	Yes	SESAM	Prisms	[76] (2000)
Yb^{3+}:$KGd(WO_4)_2$	240 fs	1028 nm		22 W	Yes	SESAM	Chirped mirrors	[77] (2002)
Yb:KYW	71 fs	1025 nm	46 fs	120 mW	Yes	KLM	Prisms	[78] (2001)
Yb^{3+}: $KY(WO_4)_2$	101 fs	1045 nm		100 mW	Yes	SESAM	Prisms	[79] (2002)
Yb:SYS	70 fs	1066 nm	16 fs	156 mW	Yes	SESAM	Prisms	[80]
Yb^{3+}:$SrY_4(SiO_4)_3O$	110 fs	1070 nm		420 mW	Yes	SESAM	Prisms	[81] (2004)
Yb:SYS//YAG	130 fs	1070 nm		1 W	Yes	SESAM	Chirped mirrors	[82] (2005)
Yb:YVO_4	61 fs	1050 nm	36 fs	54 mW	Yes	KLM	Prisms	[83]
	120 fs	1045 nm		300 mW	Yes	SESAM	Prisms	[84] (2005)
Yb:CaF_2	150 fs	1043 nm	36 fs	880 mW	Yes	SESAM	Prisms	[85]
	230 fs	1047 nm		1740 mW	Yes	SESAM	Chirped mirrors	(2004)
Yb:CALGO Yb^{3+}:$CaGdAlO_4$	47 fs	1050 nm	14 fs	38 mW	Yes	SESAM	Prisms	[86] (2006)
Cr:ZnSe Cr^{2+}:ZnSe	106 fs	2460 nm	11 fs	140 mW	No	SESAM	No	[87] (2006)
Cr:ZnS Cr^{2+}:ZnS	1100 fs	2460 nm	12 fs	125 mW	No	SESAM	No	[88] (2006)

get broader spectra—seems to be also a very promising solution which starts to be investigated [54]. The development of ultrashort-pulsed lasers using new laser crystals is then still one of the most daring challenges for the future laser technology.

Another very promising way to develop very efficient and high-power solid-state ultrafast lasers consists in using Yb-doped fibers as laser gain media [55–57]. The main advantages of these lasers are based on the guiding properties of this gain medium. The gain medium can be extremely long which neutralizes the problems of low emission cross section of thermal loading and can then exhibit very high gain; moreover the laser can be compact and monolithic. On the opposite, these long and very confined gain media has also the disadvantage of a strong influence of the GVD and non-linear effects (non-linear effects such as stimulated Raman scattering, four wave mixing that can be very deleterious effects). Anyway, the results obtained with femtosecond fiber systems are already very interesting. Moreover, since very few years, new promising fibers, photonic crystal fibers, are emerging; they have very attractive properties in terms of the dispersion control [58] and large mode propagation [59] (that reduces the non-linear effects). The femtosecond lasers based on rare-earth doped fibers are thus a very challenging domain in full expansion, but this is another story.

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