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New materials for short-pulse amplifiers

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Abstract: Laser amplifiers are seeking for high power, efficiency and short pulse durations. The research laboratories in this field have then focused their investigations towards new laser materials that can be efficiently diode-pumped, that can sustain the high-power pumping and that have broad emission bandwidth to achieved ultra-short pulse amplification. This is why, for more than ten years now, new Yb-doped materials are strongly investigated. In the actual state of the art, they represent the more promising and successful materials for this kind of applications. In the paper we will do a short review of the last and more impacting discoveries and demonstrations in this field for the last few years.

Directly diode-pumped femtosecond lasers delivering high powers is one of the hottest and most challenging topics of nowadays. From this point of view, numerous international groups of research are investigating the use of Yb-doped materials. In fact, it is admitted now that only ytterbium doped crystals can provide efficient femtosecond amplifiers at high repetition rate and high energy. In the past decade, laser development using Yb-doped materials and especially crystals has become one of the most active fields in laser research. It is now widely recognized that Yb-doped crystals have a significant potential in the development of directly-diode-pumped high power and ultra-short lasers [1-2]. This is possible thanks to the simple electronic-level structure based on only two manifolds of the Yb^{3+} laser active ions and the reduced "quantum defect" between pump and laser photons. This leads to high pumping efficiencies, very favourable thermal properties and their broad emission bands, due to a strong electron-phonon coupling, allowing then ultra-short-pulse generation. Since the laser performance of Yb-doped crystals is strongly correlated to the crystal-host properties, an important international research activity has been focused on the search for innovative Yb-doped crystals to improve efficiency, high average power and pulse duration.

For amplifiers, two main approaches are described in the literature. The first one considers classical and well-known Yb-doped materials such as glasses or YAG. In these works, the main development novelty is concentrated on the amplifiers architectures involving new crystal geometries with YAG and new large-mode-area fibre geometries with glass. The second approach concerns the materials themselves in order to find the perfect crystal gathering high thermal conductivity, high gain and broad emission bandwidth. In fact, since pulse duration is directly correlated to the spectroscopy of the Yb ions imbedded in their crystal, the crystal host choice is crucial to have good spectroscopic properties. The influence of the crystal -including Stark effect of the electric field, electro-phonon and vibronic interaction of the lattice- is very important on the spectra of this rare-earth dopant. It directly impacts on the intensity and the broadness of the emission spectrum lines. On the other hand, the potential to sustain high power is directly correlated to the capacity of the host matrix to well evacuate the heat (thermal conductivity, typically) brought by the high power pumping which is still an issue -even if the thermal loads with Yb^{3+} remains exceptionally low compared to other dopants. The high thermal conductivity is then a very important parameter to consider for high power amplifiers.

In the past decade, many novel crystals were proposed for a new generation of femtosecond diode-pumped solid-state lasers. On the one hand, for high average powers and very efficient lasers crystal such as tungstates (Yb:KYW[3-5,38] and Yb:KGW) or orthosilicates (Yb:YSO[6] and Yb:LuSO) have investigated, For amplifier systems (especially in the in the industry[5]), tungstates are exclusively used since they gather (which is atypical) reasonable emission spectral bandwidth and high cross sections. On the other hand, for ultra-short pulse generation crystals such as borates (Yb:GdCOB [12] or Yb:YCOB [13,14] Yb:BOYS [7]) or sillicate (Yb:SYS [8,9]) and vanadates (Yb:YVO_4 [16], Yb:$GdVO_4$ and Yb:$LuVO_4$ [17]) were investigated. But,in amplifiers, the gain of these crystals that is relatively low, imposes high number of passes with then a strong impact of the gain narrowing which was demonstrated at low (100 µJ) [10-11] and high (12 mJ) energy[15]. Moreover, the thermal conductivity is also an issue for these crystals that cannot deliver high output average powers in standard amplifier configurations. An alternative is to investigate crystals with high thermal conductivities (around 10 W/m/K for undoped crystals) with narrower spectral bandwidths: such as Yb-doped sesquioxide[18] or YAG. The narrow bandwidths can be overcome in oscillators with a strong amount of Kerr non-linearity to broaden the spectrum beyond the natural emission [19,20,21] but this cannot be extended to ultrashort amplification because of gain narrowing effect.

Another alternative way is to use exceptions to the basic rule according which in simple-matrix crystals phonons propagate well (thus with a high associated thermal conductivity) but do not have enough disorder to permit broad bandwidths, while on the other hand, disordered materials allow sufficiently different environments for Yb^{3+} to generate broad bandwidths but have a low thermal conductivity caused by their disorder. The strategy is to find highly structured crystal to allow high thermal conductivity but with *atypical* spectral properties (fig. 1).

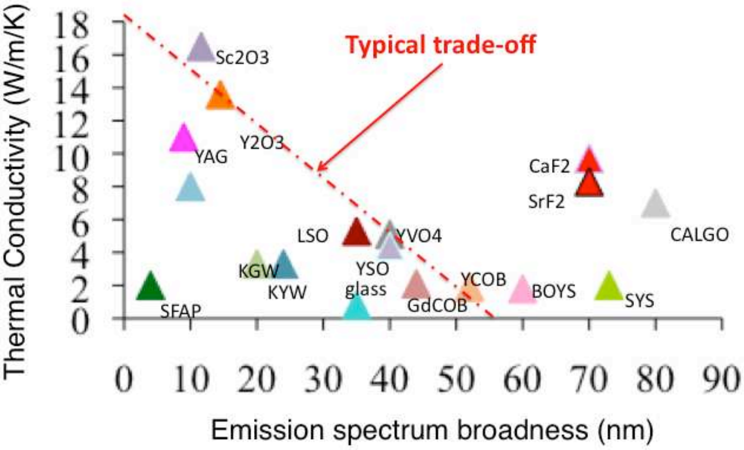

Figure 1: Thermal properties versus spectroscopic properties for different Yb-doped crystals. Yb:CALGO and Yb:CaF$_2$ clearly exhibit atypical properties.

Both CaGdAlO$_4$ (CALGO) and CaF$_2$ have relatively good thermal properties with a thermal conductivity (for undoped crystals) around 10 W/m/K. Measurements of the 2-at% Yb:CALGO thermal conductivity yielded 6.9 W/m/K and 6.3 W/m/K along the a and c axis respectively. This is similar to values obtained in Yb:YAG[23]. In the case of the CaF$_2$, the thermal conductivity values of undoped fluoride crystals are equal to 9.7 W/m/K [24] and decreases down to 6 W/m/K when doped at 2.6 % in Yb^{3+}. With this kind of values, one can expect narrow bandwidth for the spectra. Nevertheless, this is not the case while the reason for each crystal it is not the same.

In the CALGO crystal structure, Ca^{2+} and Gd^{3+} equally share the same crystallographic site, and can both be substituted by Yb^{3+} ions. This leads to the large inhomogeneous broadening in the emission spectrum. The presence of a plateau in the gain cross-section between 1000 nm and 1050nm can be explained by the exact complementarity of two different sites in the host. This peculiarity explains the unusually flat emission spectrum of Yb-doped CALGO crystal[25-27].

In the Yb:CaF$_2$ and its isotypes such as Yb:SrF$_2$, the broad bandwidth is due to substitution of Ca^{2+} or Sr^{2+} ions by Yb^{3+} ions that create hexameric clusters inside the matrix due to the unbalance valence of the substituted ions with Yb^{3+}. This clusters appear for “heavily” doped fluorites *id est* above 0.5 % at doping. The Yb^{3+} ion cannot then be considered as single isolated ion. This leads to a broad emission spectrum. Thanks to the cluster arrangement of the Yb^{3+}, the absorption and the emission spectra of the fluorites are relatively broad and represent an exception in the realm of Yb-doped crystals with good thermal properties. Moreover the Yb:CaF$_2$ and its isotypes such as YbSrF$_2$ have very long fluorescence lifetime above 2 ms (among the longest, with Yb:LnCOB, for Yb-doped laser materials). This makes them even more attractive for amplifiers since this allows a better storage of the energy[28-34].

This is why, we have intensively investigated and characterized these promising novel crystals within the strong collaboration between the laboratories expert in material science such as CIMAP Caen, LCMCP Paris and LCFIO Palaiseau; and why, additionally, a large number of ambitious scientific projects aim at using partly or entirely this technology to explore new fields of physics, such as attosecond physics in MPQ München, X-ray laser in MBI Berlin, proton generation for cancer treatment in FZD Dresden, and high field physics (electron and proton acceleration, nuclear physics) through the French ILE (Institut de la Lumière Extrême) and the corresponding European ELI project. We can also mention fusion projects (Genbu in Japan, HiPER in Europe) where efficiency at kilowatt average power level is a key parameter to achieve positive overall gain in next generation fusion power plants.

In the actual state of the art, the results for amplification chains using these new materials have demonstrated the shortest pulses ever produced with high-power, high-energy and efficient amplifiers. Siebold et al. have successfully developed a terawatt system within the Polaris project.[33-34] The production of 197 mJ, 192 fs pulses has been demonstrated at 1 Hz with the potentially of higher extractable energy with improvements on the crystal and coating quality.

On the other hand, at high repetition rate, seeded with pulses from an Yb:CALGO oscillator, a Yb:CaF$_2$ amplifier delivering short pulses (≈180 fs)[35-36] at up to 1 kHz repetition rate has been demonstrated. The shortest pulse duration generated is 178-fs which corresponds to our best knowledge, to the shortest pulses for a room-temperature Yb-doped-crystal amplifier. The corresponding energy is 1.4 mJ before compression (620 µJ after), at a repetition rate of 500 Hz for 16 W of pump power. The bandwidth is 10 nm centred at

1040 nm. At 10 kHz repetition rate, 1.4 W of average power before compression is obtained, corresponding to an optical-optical efficiency of 10%.

The future prospects, for these new materials for amplifiers will mainly consist in adapting to these crystals the novel amplifier architectures and crystal geometries already developed with Yb:YAG. In fact, another interesting point to note concerns the parallel progress done on geometries specifically developed for high power lasers based on crystals. These architectures involving crystals with a high surface/volume ration such as thin-disk[37-39], slab[40] and crystaline-fiber[41] have allowed strong improvements in the Yb:YAG amplifier performances especially in terms of power. For example the INOSLAB experiment has allows record of average power for femtosecond pulses with an average output power of 1.1 kW, a peak power of 80 MW, and a 615 fs[40] pulse width overwhelming then the fibre based amplifiers previous record [42]. On the other hand, very promising architectures closed to fibres but involving crystalline matrices (so called single crystal fibbers) are also very promising in terms of high power and pulse duration preservation. In fact, very recently fibre-crystal amplifier has demonstrated[41] 330 fs pulses with an average power of 12 W. This is the shortest pulse duration ever produced by an Yb:YAG amplifier. Moreover, the gain in the single crystal fibre can reach a value as high as x30 in a simple double pass configuration.

The other interesting way under exploration for Yb :CaF_2 concerns the cryogenic cooling. In fact, such as in Yb:YAG [43] and YLF [44] the laser amplification at low temperature will allow, with Yb:CaF_2, better performances in terms of average power and efficiency[45]. Firstly, the cryogenic cooling improved the thermal conductivity with a factor 7 from room temperature to LN_2 temperature for example. Secondly, the cross sections are also enhanced by a factor 3. On the other hand, the emission peaks tend to narrow which unfavoured ultrashort pulse amplification. Nevertheless, preliminary experiments[44] with Yb :CaF_2 have been realized and amplification of short pulses is possible.

In conclusion the main advances in the field of new materials for amplifiers really focus on the purpose of obtaining more average power in ultrashort-pulsed regime and mainly involved the research on new Yb-doped materials. Among them the actual breakthrough seems to come from exceptionally broad materials with high thermal conductivities such as Yb:CaF_2 or Yb:CALGO which is confirm by the excellent experimental performances: amplification up to the TW and record in pulse duration. In the same time, architectures optimal for very high power such as crystal-fibres, thin-disks and slab configuration have been developed for more standard crystal like Yb:YAG and really overcome the actual limitations in terms of high power amplification. The next generation of amplifiers for ultrashort pulses would certainly combine these to technological advances to produce ultra-high power, ultra-short pulse amplifiers with applications for precise ablation of a broad range of materials, from dielectrics to metals to be used in industrial applications, such as metal drilling or texturing in automotive field, or medical applications such as eye surgery, and is expected to find mass-production applications in semiconductor industry for its unique ability to achieve selective ablation, particularly required in photovoltaic industry. For academic applications, the high repetition rate (high-power) short pulses amplifiers are also an important issue such as, for example, in high harmonic generation.

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