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Transverse amplified spontaneous emission: The limiting factor for output energy of ultra-high power lasers

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ABSTRACT

For the new generation of the ultra-high power lasers with tens of PW of output power, kJ-level energies have to be reached. Our modeling, applied to Ti:sapphire amplifiers, demonstrates for the first time, according our knowledge, that Transverse Amplified Spontaneous Emission (TASE) places an additional restriction on storing and extracting energy in larger gain apertures, even stronger than transverse parasitic generation (TPG). Nevertheless, we demonstrate that extracting during pumping (EDP) can significantly reduce parasitic losses due to both TASE and TPG.

1. Introduction

The main goal of Chirped Pulse Amplification (CPA) is to reduce the size of optical elements and amplifier crystals. It gives an alternative way to increase pulse energy while maintaining a compact geometry. Instead of enlarging the transverse beam sizes, the pulse duration is increased for further amplification, then, compensated by the pulse compression [1,2]. This method has allowed an increase in the output power of laser systems from the GW to the PW level with commensurate efficiency increase for modest energies up to a few tens of Joules [3]. However, for the new generation of ultra-high power lasers with tens-of PW output powers [4] these unpretentious energies will no longer suffice, and the kJ-level has to be reached. The modern ability to stretch pulse duration is restricted to a few orders by existing grating technology, thus, further energy increase while avoiding harmful nonlinear effects requires the enlargement of amplifier apertures. There are two solid-state candidates for the final amplifier in such systems: Optical Parametric Amplification (OPA) [5] and laser amplification. Both possess advantages and shortcomings. OPA has such attractive properties as: gain bandwidths large enough to support sub-10 fs pulses; “optical cooling,” to enable high repetition rates; availability of large high-quality nonlinear amplifier crystals; and high pulse contrast due to the absence of Amplified Spontaneous Emission (ASE) before and after the pump pulse. But, OPA demonstrates low efficiency and places severe requirements on pump-beam spatial amplitude fluctuations. It requires control of pump pulse duration (few hundreds of ps – 1 ns) along with accurate spatial and temporal matching between pump and the signal pulses and so on.

Large aperture laser amplifiers, being free from the most of these restrictions, are subjected to severe losses due to Transverse ASE (TASE), and Transverse Parasitic Generation (TPG). These effects cause significant depletion of the inverted population, leech stored energy, and limit extracted energy. Consequently, enlarging the amplification aperture fails to be an unlimited means of obtaining higher output energy under the constraints imposed by nonlinear effects. In laser amplifiers the conventional procedure used to prevent parasitic generation is to reduce the reflectivity of the sidewall of the gain crystals by grinding, sandblasting and/or coating with an index-matched absorptive polymer or liquid layer [6,7]. However, the difficulty found in introducing exact index matching with existing absorbers still restricts the diameter of the pump area to ~6 cm, corresponding to an extracted energy to around 30 J from Ti:sapphire [8]. Attempts to enlarge amplifier apertures, or to further increase pump fluence have lead to severe parasitic generation and have failed to increase extracted energy.

Previously it was shown that TPG is the limitation on energy extraction and we provided a new solution to this critical amplifier problem, namely Extraction During Pumping (EDP), that allows laser amplification to remain viable through the next generation of the ultrahigh power laser systems [9,10]. In this paper we demonstrate that TASE places an additional restriction on storing and extracting energy in larger gain apertures. This restriction is

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even stronger than parasitic lasing because the threshold for the latter can be increased with the development of the new index matching materials for absorbers, while TASE necessarily increases with the aperture size, limiting the maximum stored energy. We also demonstrate that EDP is able to successfully resolve this problem too.

2. TASE calculation

In Ref. [11] the losses connected with ASE following pump absorption are calculated. Here, we estimate the total TASE radiated from a disc-shaped gain material *during* pump excitation. This calculation is simplified by assuming transverse uniformity in the density of the population inversion, which is realistic for typical super-Gaussian pump beam profiles. Further, we assume a spectral and polarization average value for the emission cross-section [12] rather than integrating over the spectral line shape and angle. We have checked the validity of this assumption by substituting in the calculation the values of the cross sections for each polarization and observing that the mean of these results matches the result based on the mean cross section. In contrast to Refs. [13,14] we also neglected emissions that pass through the working parallel surfaces. All photons reflected from these surfaces due to Total Internal Reflection (TIR) pass through the side surface of the crystal and are being taken into account in our simulation. The other emissions experience significantly lower gain due to low amplification path length. For example, under typical pumping conditions (3 J/cm^2) in a 15×4 cm Ti:sapphire amplifier, maximal gain for emission through the face is ~ 15, whereas for TASE it is $\sim 2 \times 10^6$. The good match between these numerical calculations and the experimental results reported below support the validity of this assumption.

With these assumptions, the differential equation governing the temporal dependence of the population inversion in an element of the gain volume is a first order nonhomogeneous equation

$$\frac{dn}{dt} + an + W_s(n,t) = W_p \tag{1}$$

where n is the population inversion density, $a = 1/\tau$ is the inverse lifetime of the laser transition, $W_s(n,t)$ is the loss rate due to stimulated emission, and W_p is the pump rate.

Splitting the pump duration into i-time steps, each being shorter than time required for significant change in population inversion, W_s is essentially constant during each interval, and Eq. (1) becomes linear (with initial conditions: $t_0 = 0$, $n_0 = 0$, $W_s^0 = 0$ and $\delta t_i = t_i - t_{i-1}$) and has the well known iterative solution

$$n_i = n_{i-1} e^{-\delta t_i/\tau} + \tau (W_p - W_s^{i-1})(1 - e^{-(\delta t_i/\tau)}) \tag{2}$$

Now, we have to find the value of W_s^i for each step of integration. For that, we integrate the ASE radiated during each time-step. In the wide gain volumes of interest the majority of the ASE passes through the sidewall of the medium due to the high transverse gain, so, for simplicity, we concentrate on this portion of the emission.

For the integration, we take an element of the gain volume *dldrds* (with respective lengths of arc, radius, and elevation as indicated in Fig. 1a), find the fluence of ASE from this element through element of the side surface *ds*'*dl*', and integrate over the whole volume, using a cylindrical geometry

$$dF_i = dl' ds' \int\int\int \frac{h\nu_e n' e^{\sigma n r} \cos\theta \cos\alpha}{4\pi r^2} dl dr ds \tag{3}$$

where dF_i is the fluence through an element of the sidewall area *dl*'*ds*', $h\nu_e$ is the emitted photon energy, and n' is the total population inversion decay rate for the volume element *dldrds*.

Splitting the crystal into thin discs as shown on Fig. 1a, we integrate fluence from an arbitrary disc, and then integrate over the angle α, in the range $\pm\alpha_{c'}$ ($\alpha_{c'}$ is the complement of the critical angle for TIR, γ. This takes into account ASE experiencing total internal reflection from the flat surfaces of the crystal, as illustrated in Fig. 1b. The remainder of the crystal volume adds negligible flux.

The distribution of the population inversion in a longitudinal direction can be written as follows: when the gain volume is pumped from both sides: $n(s) = -aP_0\,(e^{as} + e^{a(S-s)})\,/h\nu$, where α is the coefficient of pump absorption, P_0 is the pump energy incident on one face, S is the crystal thickness, $s = D\sin\alpha$, and the crystal, for purposes of the calculation, can be considered as the virtual multilayer crystal of Fig. 1b.

For integration through a disc we draw a circular arc, L, with radius r from the element *ds*′*dl*′ and take the layer *dr* of Fig. 1c. The gain for ASE flux from each element of this layer through the element *ds*′*dl*′ is approximately constant. Therefore, by replacing in (3), $dl = r \bullet d\beta$, $r = D\cos\theta \cos\alpha / \cos^2\theta \cos^2\alpha + \sin^2\theta$; $dr = (-D\sin\theta \cos\alpha\,(\cos^2\theta \sin^2\alpha + 1)\,/(\cos^2\theta \cos^2\alpha + \sin^2\theta)^2) d\theta$; $ds = r \bullet d\alpha$, and taking into account that $\int_0^L \cos\beta dl = \int_{-\theta}^{\theta} r\cos\beta d\beta = 2r\sin\theta$, we have the following integral for total ASE through the side surface, which can be evaluated numerically

$$F_i = \frac{h\nu_e S D^2}{2} \int_{-\alpha_t}^{\alpha_t} \int_0^{\pi/2} \frac{n' G \cos^2\alpha \sin^2\theta (\cos^2\theta \sin^2\alpha + 1)}{(\cos^2\theta \cos^2\alpha + \sin^2\theta)^2} d\theta d\alpha,$$

where $G = \exp(\sigma n_{av} D \cos\theta \cos\alpha / \cos^2\theta \cos^2\alpha + \sin^2\theta)$; and n_{av} is the average population inversion along r. Thus $W_s^i = 2F_i / h\gamma_e S\pi D^2$. Making this integration iteratively for each time-step throughout the pump duration we obtain the time dependence of $F(t)$, the TASE losses during the pumping process.

For verification, this method of calculation was compared with the commonly accepted Franz–Nodvik equation for 1-D geometry [15]. With a 15-cm crystal diameter and a pump fluence of 2.6 J/cm^2 incident on a Ti:sapphire gain crystal. The results of the calculation shown in Fig. 2a demonstrate good agreement.

The deviation seen in Fig. 2a, for high input flux, results from the development of a spatially non-uniform population inversion during the amplification of the long pulse. This is small for the 3-D geometry due to the absence of directional selectivity in the gain distribution, and is experimentally confirmed.

A second verification was made, comparing calculated results with experimental data measured in an existing 8-cm-diameter Ti:sapphire amplifier with a 5-cm-diameter pump area (see Fig.2b). Although the absolute volume of ASE varies with direction, the form of the fluorescence' time-dependence should be the same because it is determined by the time-dependence of total population inversion decay. Thus, the normalized curves coincide.

3. Results of calculations

Fig. 3a shows the evolution of the gain volume's normalized fluorescence to its maximal value as a function of pumping time for a 100 ns pump pulse given different gain apertures. Here E_{max} is the theoretical maximum of the extracted energy, and E_{loss} the lost energy due TASE. As seen in the plot, ASE grows dramatically after a certain time of pumping (even for a 10 cm gain aperture) and soon approaches the pump energy (flat part of the curves). This means that with further pumping all additional energy will be radiated out of the crystal as ASE. These critical points of “Anomalous” ASE (APs) advance to the beginning of the pumping process with increasing gain diameter. Under these conditions, no more than 50% (30%) of the pump energy can be stored in a 15 cm (20 cm) crystal.

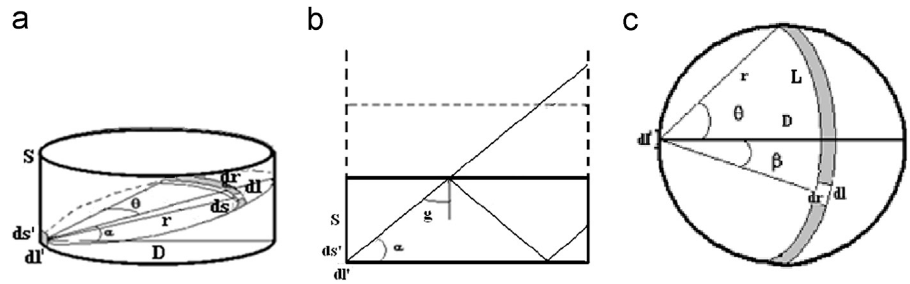

Fig. 1. Amplifier crystal (picture for calculation of transverse ASE through side wall). (a) perspective view; (b) side view; and (c) view from the top.

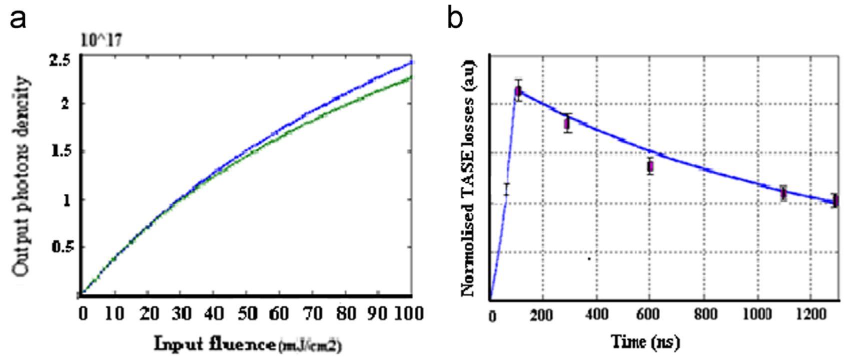

Fig. 2. (a) verification of the above calculation vs. the Franz–Nodvik equation ((1)-D geometry), $S=15$ cm, pump fluence – 2.6 J/cm^2, blue curve – F–N, green – used; (b) demonstrates good agreement between the calculated temporal dependence from the 5-cm-diameter pump area blue curve and the experimental purple points. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

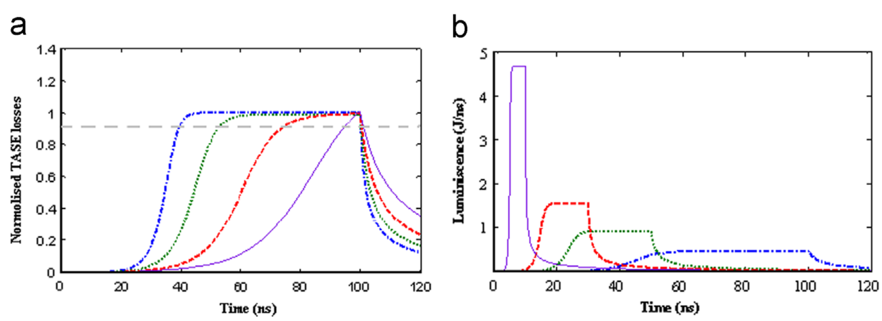

Fig. 3. Fluorescence evolution of gain volumes vs. pumping time. (a) Normalized ASE to its maximal value for different crystal apertures, D: blue dash-doted curve $D=20$ cm ($E_{max}=824$ J, $E_{loss}=590$ J), green dots $D=15$ cm ($E_{max}=460$ J, $E_{loss}=290$ J), red dashes $D=10$ cm ($E_{max}=210$ J, $E_{loss}=100$ J), and violet solid $D=6$ cm ($E_{max}=74$ J, $E_{loss}=20$ J); (b) TASE for different pump pulse durations, τ_{pump} (15 cm crystal diameter): violet solid curve $\tau_{pump}=10$ ns ($E_{loss}=325$J), red dashes $\tau_{pump}=30$ ns ($E_{loss}=321$ J), green dots $\tau_{pump}=50$ ns ($E_{loss}=315$ J), blue dash-dots $\tau_{pump}=100$ ns ($E_{loss}=290$ J). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Shortening the pump pulse duration does not help to reduce the losses; at least, not until pulse durations become shorter than the transit-time of the TASE across the gain aperture, and this is in the range of several hundred ps, where the damage threshold of the gain material is below the fluence required for efficient operation.

Fig. 3b shows crystal fluorescence during pumping for different pump-pulse durations for a 15 cm crystal diameter. As seen in Fig. 3b, the losses grow slightly with the reduction of the pump pulse duration, because the higher pump rate enhances TASE more than the total ASE in each time step.

In order to reduce the transverse gain in the volume near the parallel surfaces, the method of double-pass pumping of a lightly doped crystal was suggested in Ref. [16]. We calculate the losses due to ASE for differently doped crystals with various diameters. Results presented in Fig. 4a show that this method reduces losses for smaller gain apertures. When the aperture is increased, the losses increase in spite of the low doping and eventually supersede the losses experienced by the same aperture with higher doping. There are, in fact, two transverse modes: generation along the two working parallel surfaces due to the maximal population inversion [6] and the z-pass between these surfaces due to total internal reflection [17] as illustrated in Fig. 4b.

The z-pass TASE surpasses parallel TASE in large-aperture-gain geometries with high aspect ratios and relatively low crystal doping because they exhibit higher gain for the first mode. Moreover, with the deeper pump light penetration in the gain volume, the increased ASE reaches the AP later but with a higher rate as seen in the leading edges of the solid curves.

As shown in Fig. 3a, the onset of TASE imposes a practical upper bound for the pump fluence based on the gain geometry's diameter, doping, and thickness. Therefore, it is important to

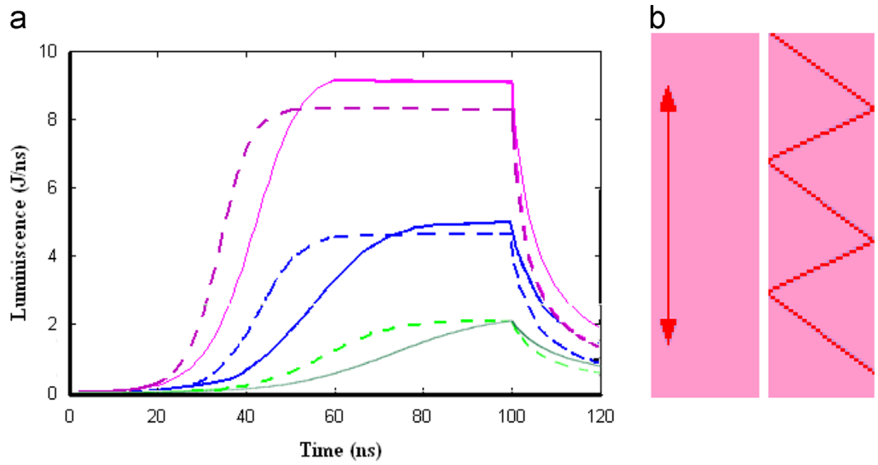

Fig. 4. (a) ASE for differently doped gain volumes with various diameters, D: green solid curve – $D=10$cm (pump transmission coefficient $B=0.1$), green dashes – $D=10$ cm ($B=0.01$), blue solid – $D=15$ cm ($B=0.1$), blue dashes – $D=15$ cm ($B=0.01$), purple solid – $D=20$ cm ($B=0.1$), purple dashes – $D=20$ cm ($B=0.01$); and (b) two transverse optical paths with highest gain for TASE within a crystal. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

understand where this boundary resides in comparison with the limitation given by TPG.

The key parameter for calculating the output fluence is the initial pump fluence corresponding to the parasitic generation threshold [10]. As mentioned above, there are two transverse parasitic modes. The product of the pump fluence and the pump area can be explicitly written as

$$D \times F_p = K \text{ where } K = \frac{\nu_p}{\nu_{em}} \times \ln G_{pt} \times \frac{zF_s}{\ln B} \text{ for near – surface,}$$

$$\text{and } K = \frac{\nu_p}{\nu_{em}} \times \ln G_{pt} \times \frac{zF_s}{\ln B} \text{ for } Z\text{–pass gain,} \quad (4)$$

where F_p is the initial pump fluence of one side of the crystal, F_s is the saturation fluence, D is the pump area diameter, G_{pt} is the highest transverse gain that can be compensated by an index-matched absorptive coating, z is the crystal thickness, n is the index of refraction of the crystal, and B is the absorption coefficient for the pump frequency.

The product of the crystal diameter and the pump fluence is a constant for each set of the values of the critical transverse gain (absorber index matching); crystal thickness; and/or doping. So, ultimately, the use of a crystal with a given aperture fixes the maximum pump fluence. Further, enlargement of the crystal aperture requires reduction of the pump fluence. In Fig. 5, curves showing the dependence of the optimal pump fluence through both crystal faces on the crystal diameter are presented. There are three TGP-curves with the varied K-factors.

From another perspective, we can build the same dependences for the TASE limitation based on the APs (Fig. 3a, crosses with gray dashed line indicating 0.9 of the maximum volume).

As seen in the Fig. 5, the TASE curve for crystals with $B=0.01$ nearly match the TGP curve with K values of 32. These values correspond to existing absorbers [6,8,18]. The main conclusion to be drawn from this correspondence is that there remains no motivation to develop better absorber materials, owing to restrictions of TASE.

According to this discussion we have to conclude that the situation for regular laser amplifiers with large apertures appears bleak, especially with respect to their application as final amplifiers in very high power laser systems. Nevertheless, in this paper we will demonstrate that the parasitic losses due to both TASE and TPG can be significantly reduced using EDP [9,10].

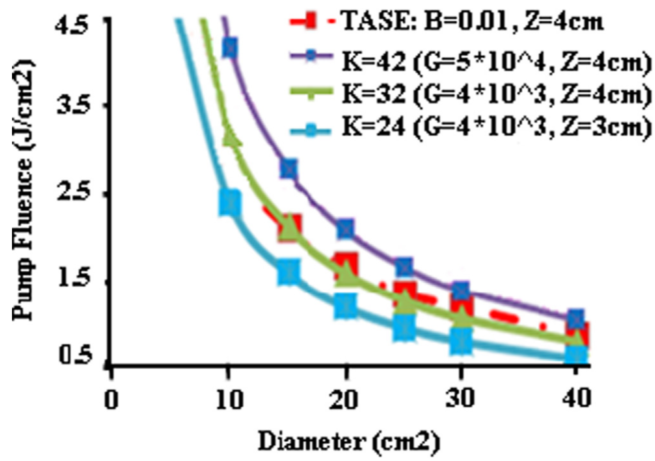

Fig. 5. Dependence of pump fluence on crystal diameter: solid curves – calculated for TPG ($K=24$ – light blue squares, $K=32$ – green triangles, $K=42$ – blue squares); dashed curves – calculated for TASE (red squares – $B=0.01$). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

4. Extraction during pumping (EDP)

To overcome restrictions discussed above we suggested deviating from the conventional method of pumping and amplifying in multi-pass amplifiers, which is based on the energy stored in the upper laser level prior to the arrival of the first pass of the input pulse [19]. By continuing to pump after the arrival of the amplified pulse, we are able to forestall TASE/TPG and increase the extracted energy. In other words, the energy extracted during one pass of the amplified pulse through the crystal can be restored by pumping up to the AP or the parasitic lasing threshold before the next pass. The EDP process requires an extended pump-pulse duration ranging from tens to hundreds of nanoseconds, or several delayed pulses. This allows sufficient time for proper pumping between passes, avoiding problems with temporal jitter and allowing increased total pump fluence due to the longer pump duration. This approach was shown to double the output flux above the parasitic lasing limit in our experiments [10]. The method has also been applied on several Ti:sapphire booster amplifiers of petawatt scale, and has allowed output energies in excess of 60 J with the current record power of 1.5 PW from a single channel [8,20].

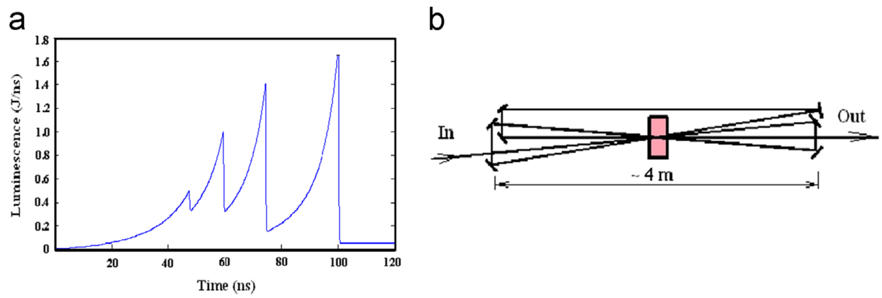

Fig. 6. Fluorescence temporal dependence for 15 cm-diameter crystal with EDP amplification. (a) 400 J pump energy, 230 J theoretical maximum output, and 3.8 J losses, (b) schematic diagram of the 4-pass EDP amplifier.

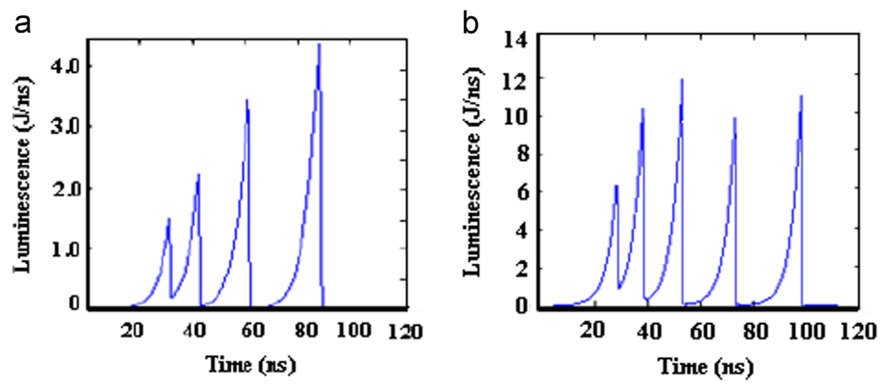

Fig. 7. Fluorescence temporal dependence (a) for 20 cm-diameter crystal with EDP amplification (1.25 kJ pump energy, 824 J theoretical maximum output, and 61 J losses); (b) for 40 cm-diameter crystal (~5 kJ pump energy, 3.3 kJ theoretical maximum output, and 207 J losses)

The optimal conditions for EDP amplifiers were discussed in [10], and the capability of the EDP method to significantly increase the output energy to the kilojoule level with existing technology was demonstrated vis-a-vis TPG. Here we will demonstrate the ability of the EDP-amplifiers to eliminate losses connected with TASE.

In cases where ASE losses reach the AP before the development of parasitic generation (see Fig. 5), EDP amplification is also able to significantly suppress the ASE component. Estimations indicate that a four-pass EDP amplifier with a 15 cm diameter pump area and 100 ns pulse duration can approach its theoretical energy extraction limit. Modeling of the pumping of this crystal is presented in Fig. 6a, With 400 J of pump energy (in green), the EDP amplifier supplies 230 J with very low losses, using a steady pump rate. As seen in the plot the delays between the 1st and 2nd passes and the 2nd and 3rd are 12 ns and 15 ns and between the 3rd and 4th passes, 25 ns. This corresponds to the specific geometry of Fig. 6b where a bypass is placed between the 3rd and 4th passes.

A four-pass EDP amplifier with a 20-cm-diameter pump area can approach 730-J energy with a 62-J input, when pumped by 1.25 kJ. Fig. 7a shows the temporal dependence of fluorescence for this scenario. Consequently, losses are limited 61 J or ~7% of the theoretical maximum, as opposed to the 590 J or more than 70% lost in conventional amplification (compare with Fig. 3a). The timing between passes is close to the optimum with respect to the maximal point of energy extraction.

Now, we consider the possible boundaries of this EDPCPA technology. As demonstrated above, the APs advance toward the zero pump fluence when the diameter of amplifier crystal is increased. The same happens when the thickness of the crystal is reduced, a choice which might be made to improve cooling for increased repetition rate. Generally speaking, an increase in the aspect ratio of the crystal leads to reducing the initial part of the stored pump energy for EDP-amplifiers. Reduction of that part to 20% of the total pump is about the minimum acceptable level, because a four to five-pass configuration is reasonable for large aperture crystals. Under this consideration, a 40-cm Ti:sapphire crystal with a 6-cm thickness is nearly maximal for EDP amplification. The dependence of luminescence from a crystal with such parameters is presented in Fig. 7b. The amplifier is able to supply 3 kJ of energy with 250 J of seed energy while suffering TASE losses of about 207 J.

Placing EDP into a broader perspective: if one were to extrapolate the idea to continuous pumping, it would move first toward quasi-*cw* and then toward *cw* pumping regimes, where instantaneous and longer-term thermal effects would certainly play a significant role.

5. Summary

1) We have suggested a method of calculating TASE in laser amplifiers during pumping, and our estimations reveal that TASE is a stronger restriction than TPG for larger Ti:sapphire apertures and that it imposes more severe losses (up to 70% for crystals having diameters more than 15 cm).
2) We have demonstrated that the limit for amplifier pump fluence connected with TASE fits within the parasitic generation limit for existing absorbers, so further improvement of the absorber material is not expected to lead to much improvement.
3) We have concluded that the only existing method capable of overcoming this limitation with existing technology is EDP amplification.
4) We have demonstrated the capability of these amplifiers to reduce the TASE-losses to 5%; using an exemplary design.
5) Finally, we have demonstrated the ability of EDPCPA technology to achieve kJ-level output energy.

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