LETTER

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Letter

Z-scan technique to study gain properties of optically pumped media

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Abstract

The opportunity to study the optical gain properties of active media using the open aperture z-scan technique without an additional probe beam is demonstrated. The method allows for the evaluation of pump-induced photodynamic process parameters in UV solid-state active media on pumping and lasing wavelengths: excited-state absorption and photoionization cross-sections of active ions, effective absorption and ionization cross-sections of color centers and recombination rates.

Keywords: z-scan, optical gain, amplified spontaneous emission, active media

(Some figures may appear in colour only in the online journal)

Introduction

The combination of the spectral features of the ground-state absorption (GSA) cross-section, gain cross-section, excited-state absorption cross-section (ESA) and other pump-induced losses determine the spectral and energy properties of active media (AM) [1–4]. That is why the measurement of these parameters is a very urgent issue. It is also important to assess the ability of the AM to amplify radiation to high-power levels, while maintaining stability of the characteristics for a long operation time [3–7]. To evaluate these parameters, the technique of pump-probe spectroscopy is usually used. Generally, pump-probe experiment requires using the high-power pumping radiation to create an inverted population in the medium and probe radiation to study its response. Although the pump-probe technique is a well-developed powerful tool, it has a number of limitations. First, two radiation sources with different wavelengths are needed. The first must efficiently pump a group of given excited states, while the second must provide radiation at the luminescence region of the sample. Second, to investigate the spectral and temporal dependencies of the gain, a wideband or tunable source of probe beam with high spectral radiance to exceed an intensive sample's luminescence is essential. This can be achieved if an appropriate laser is used. It should be noted that pulsed lasers can cause beam synchronization problems. Finally, the results of the experiments strongly depend on the accuracy of the spatial matching of the pumping and probing beams inside the sample. This severely complicates the studies and is the main source of experimental errors. However, there is another opportunity to study gain properties without any external probing radiation, using the amplified spontaneous emission (ASE) effect [8] that requires only the pump source. This technique proposes to measure the fluorescence yield from the face of the pencil-shaped AM as a function of its length and/or pumping rate. It has been successfully employed to study the gain properties of gas and metal vapor lasers [9, 10], organic dyes in liquid solution [11, 12], semiconductors [13] and solid-state AM [14, 15].

Here, we propose a new technique to study the gain properties of AM using the 'open aperture' z-scan method [16, 17]. Previously, the z-scan method was usually used for measuring non-linear features of the optically transparent sample, which were transferred through the focus of a laser beam (along the z-axis, to the sample depth direction). Because the pumping energy or the power density are changing, it can also be employed to study saturable absorption (SA) characteristics

and/or ESA and non-linear photodynamic processes in AM under pumping condition [18–20]. In the case of z-scanning, the last ones are often exhibited as reverse saturable absorption (RSA) [21–24]. Surprisingly, to the best of our knowledge, none of the researchers has taken into the account the stimulated emission (SE) as a mechanism that can affect the light transmission of the sample during z-scan measurements. In this work, we close this gap. The idea of the new technique is to use ASE that will cause a drop in the inversion of the population and will restore the population of the ground state in the AM. As a result, the absorption coefficient of the sample will depend on the cross-section of the SE and pump-induced transitions, the pumping rate, and the geometry of the experiment (the spatial distribution of pump radiation into the sample). Further analysis of the experimental data with various z-coordination, and pumping beam energy/power allows us to estimate the most important AM parameters.

Samples and experiment details

In order to demonstrate the ability of the z-scan technique to study the gain properties of the AM the Ce^{3+}: $LiCaAlF_6$ (Ce:LiCAF) wide band-gap dielectric crystal was selected as the sample. The crystal is a well-known high-gain degradation-free AM for lasers and amplifiers in the UV spectral range directly pumped by the fourth harmonic of a standard Nd:YAG laser [25–28]. It operates on a four-level scheme based on vibrational-broadened allowed electro-dipole 5d-4f transitions of Ce^{3+} ions. This is why the AM has a high GSA and SE cross-sections (about ~10^{-17} cm^2) and corresponding saturation fluence values (~100 mJ cm^{-2}) [25–28], comparable with ones for organic dyes. Together with the large active-ion density (~10^{17} cm^{-3} [19]), it essentially enforces the ASE effect. Also, Ce:LiCAF has a 5d-4f fluorescence quantum yield equal to 1, which leads to negligible losses of absorbed pumping energy to the heat and to the lack of noticeable pump-induced thermal distortion of the refractive index.

Generally, the problem of most solid-state UV AM based on the 5d-4f transitions of Ce^{3+} ions is the formation and accumulation of color centers (CC). The CC are formed because of the two-step $^2F_{5/2}$ (Ce^{3+}) → 5d (Ce^{3+}) → CB (6s-states of Ce^{3+} ions thermalized with the conduction band (CB) of the crystal matrix) Ce^{3+} ion photoionization by pumping radiation, which leads to degradation of the optical transparency and gain properties of the Ce-doped solid-state materials [4]. Unlike other solid-state UV AM, the Ce:LiCAF crystal demonstrates the unique high stability of laser properties for a long operation time even at a high pumping level and/or repetition rate [28–30] and in ultrashort pulse lasing regimes [31, 32]. In fact, the results of [15, 18, 19, 27, 33] demonstrate a low ESA cross-section at the pumping wavelength and ESA/CC formation has negligible impact on the laser properties of the Ce:LiCAF AM. This also means the lack of RSA [22].

LiCAF crystal doped with 0.5 at. % of Ce^{3+} ions in the melt had been grown by the Bridgman–Stockbarger technique in the Kazan Federal University. The optical axis of the crystal was perpendicular to the growth direction. The sample has a rod shape 6 mm in diameter and 9 mm in length, with uncoated polished ends.

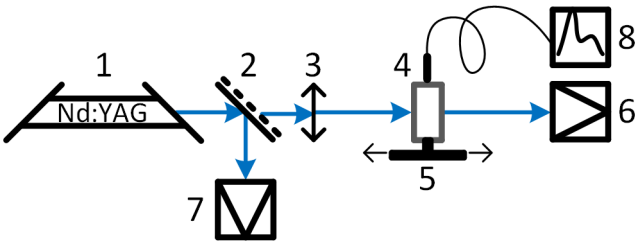

Figure 1. Open aperture z-scan experiment setup (1—Nd:YAG laser, 2—fused silica wedge, 3—30 cm focal length lens, 4—Ce:LiCAF crystal, 5—linear translation stage with micrometer screw, 6, 7—power meters, 8—Stellarnet CCD spectrometer.

The linearly-polarized fourth-harmonic ($\lambda = 266$ nm) beam from a Q-switched Nd:YAG laser LS2147A from JV LOTIS TII (1) with a 15 ns pulse width was used for the measurements (figure 1). The beam had a Gaussian energy distribution and it was focused on the sample (4) through the 30 cm focal length fused quartz lens (3). The lens provided the maximum pumping energy density up to 1.2–1.3 J cm^{-2} in the waist with the Rayleigh length at about 9 mm. The Ce:LiCAF sample (4) was mounted on a linear translation stage (5) and was transferred through the waist of the pump radiation focusing beam with a micrometer screw. The $z = 0$ coordinate corresponds to the focus position on the front of the crystal surface. Input and output pumping energy were controlled by a double-channel Ophir optical power meter (6, 7). No apertures were used in front of the power meters. Therefore, the experimental setup realized the typical open aperture z-scan arrangement for the thick sample, which is specifically sensitive only to non-linear absorption and free of any refractive index distortion [16, 17]. Fluorescence of the sample was recorded with a Stellarnet CCD spectrometer (8) perpendicular to the laser beam propagation.

Experiments and discussion

Z-scan experimental results of the non-linear absorption of Ce^{3+} ions in LiCAF crystal at 266 nm wavelength are shown in figure 2. It looks unpresumable from the point of view of known phenomena, which can become apparent in the experiments. As we have already pointed out above, this absorption coefficient behavior with z-coordination cannot be associated either with any pump-induced refractive index distortion, because the open-aperture z-scanning was applied, or with RSA, because the ESA cross-section is more than twice less than the GSA one and no CC are detected [15, 18, 19, 27, 33].

The classical trends of SA are observed at relatively low pumping flux (<0.4 J cm^{-2}) in areas $z < -20$ mm and $z > 15$ mm roughly. However, in the area where the pumping beam waist is located inside the sample, the trend of absorption coefficient dependence is significantly different from the classical one and demonstrates a sharp peak at $z = -2$ mm. This behavior is explained by the fact that instead the two-level medium usually considered in the theory of absorption

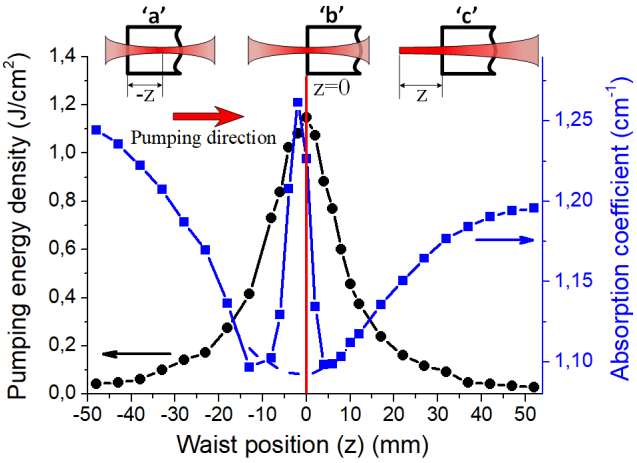

Figure 2. Pumping density (solid circles) and the Ce: LiCAF crystal absorption coefficient (solid squares) as a function of the front surface of the crystal position (z-coordinate) regarding the beam's focus. Dashed line roughly shows the view of 'classical' absorption saturation without considering the ASE. Waist center on the front sample surface corresponds to $z = 0$ coordinate. Negative values of z indicate that the beam waist is positioned inside the sample or behind it and, in contrast, positive ones mean that the focus position is situated before the sample.

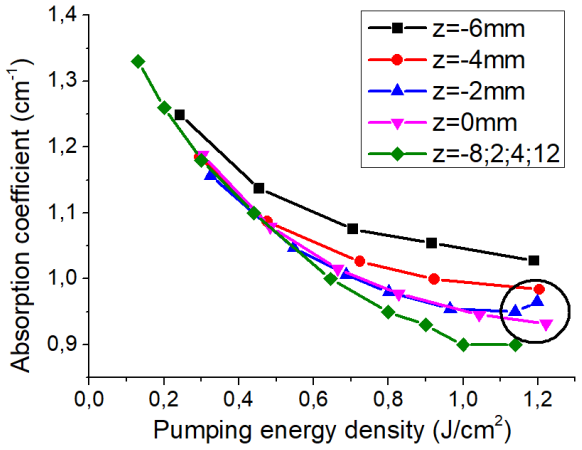

Figure 3. Absorption coefficient of a Ce: LiCAF sample as a function of pump energy density (P-scan), measured at various z-coordinates.

saturation, the four-level high-gain AM is studied here. In the case of the transference of the pumping beam waist into the Ce:LiCAF sample, the length of the area with population inversion and the total gain coefficient are increased. If the length and appropriated net gain exceeds the critical ones [34], the ASE intensity drastically grows, which effectively removes the inverse population between the 5d- and 4f-states of Ce^{3+} ions and, in turn, restores the ground-state population. Thus, the absorption coefficient of the sample again increases. Of course, it will take place when the rise time of the ASE process is less than the pumping pulse width, the sample has enough length and the Rayleigh length of the pumping waist is equal to or longer than the critical length of the of area with the given population inversion. For example, the ASE was revealed in Yb-doped crystals under continuous wave and long-pulse pumping [35].

A similar effect of ASE is observed in the dependence of the absorption coefficient versus pump radiation density (P-scan experiments) at fixed waist positions (z-coordinate) relative to the sample's front surface (figure 3). It can be seen from the figure that in the case of the waist position being outside the sample ($z > 2$ mm or $z < -8$ mm), the absorption saturation curves match each other. However, when the beam waist is positioned inside the sample ($-6\,\text{mm} < z < 1\,\text{mm}$) and the pump energy density is higher than 0.3–0.4 J cm^{-2}, the absorption coefficient dropping rate decreases with increasing radiation intensity. Moreover, the absorption coefficient demonstrates the rise trend at $z = -2$ mm and energy density higher than 1 J cm^{-2}. This area is marked in figure 3 by a circle.

The rough estimation of the critical AM length Z can be done using the formula for critical net gain G from [34]:

$$G = \frac{4\pi}{\eta \cdot \Omega} \cdot \sqrt{\ln(G)}, \tag{1}$$

where η—is fluorescence quantum yield, $\Omega = \pi D^2/4Z^2$—the solid angle subtended by one face of the medium with length Z, as can be seen from the central point of the opposite face, D—is the diameter of pumping beam waist (diameter of AM). Thus, the critical length Z is:

$$Z = \frac{D}{4} \cdot \left(\frac{\eta \cdot G}{\sqrt{\ln(G)}} \right)^{1/2}. \tag{2}$$

Taking into account roughly that:

$$G = \exp\left(\sigma_{SE} \cdot \Delta n \cdot Z\right) \tag{3}$$

and

$$\Delta n = \frac{\rho_0 \cdot \exp\left(-k_{abs}^{sat} \cdot Z\right)}{h\nu_{pump} \cdot Z}, \tag{4}$$

where σ_{SE}—is the stimulated emission cross-section at the pike of the net gain spectrum, Δn—is the population inversion, ρ_0—is the pump energy density on the front surface of the sample, $h\nu_{pump}$—is the energy of the pumping quantum and k_{abs}^{sat}—is the SA coefficient of the pumping radiation without an ASE effect, we have a transcendental equation, which can be resolved with respect to Z:

$$Z \cdot \left[\frac{\sigma_{SE}\rho_0}{h\nu_{pump}} \exp\left(-k_{abs}^{sat} \cdot Z\right) \right]^{1/4} = \frac{D}{4} \left\{ \eta \exp\left[\frac{\sigma_{SE}\rho_0}{h\nu_{pump}} \exp\left(-k_{abs}^{sat} \cdot Z\right) \right] \right\}^{1/2}. \tag{5}$$

In the case of averaged $\sigma_{SE} = 5 \times 10^{-18}$ cm^2 from [25–27, 33], $\rho_0 = 1.2$ J cm^{-2}, $\lambda_{pump} = 266$ nm, $k_{abs}^{sat} = 1\,cm^{-1}$, $D = 0.1$ mm and various $\eta = 0.3$–1 [36, 37], the critical length of AM $Z = 1.4$–1.9 mm that is almost the same as in the z-scan experiments.

The variation of the η value is stipulated by the Ce^{3+} ion multisite activation nature of the LiCAF crystal, which results in the redistribution of the pumping energy between three Ce^{3+} ion activator centers [36, 37]. Moreover, the lasing at about 290 nm is typically realized from only one type of Ce^{3+} center with the largest concentration corresponding to the substitution $Ce^{3+} \rightarrow Ca^{2+}$ and non-local positive charge compensation [38]. It distorts the total fluorescence spectrum of the Ce:LiCAF sample in the case of the laser action, because

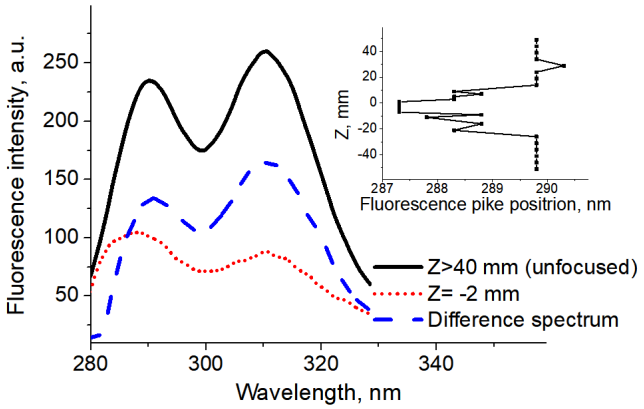

Figure 4. Typical 5d-4f luminescence spectra of Ce^{3+} ions in a LiCAF crystal for an unfocussed excitation laser beam at 266 nm ($z > 40$), for $z = -2$ and the difference of these spectra. Luminescence spectra pike's wavelength shift at around 290 nm versus the waist position (z-coordinate) is presented in the inset.

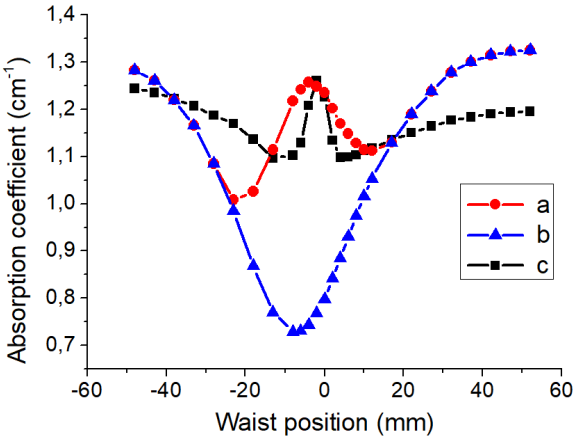

Figure 5. Approximation results of the non-linear absorption coefficient of the Ce:LiCAF sample versus the pumping waist position with (a) and without (b) considering the ASE. Data (c) is the experimental one.

the distribution of this center's spectrum to the total one is decreased [36]. The same is observed for the sample's spectra in the z-scan measurements due to the ASE effect (figure 4)—the intensity of the fluorescence of the Ce^{3+} activator center with pikes at about 290 and 310 nm decreases and the short wavelength band's maximum shift from $\lambda = 289.8$ nm to $\lambda = 287.3$ nm at around $z = -2$ mm is observed.

The observed non-linear absorption effects were interpreted using the model of photodynamic processes in cerium-activated materials [4] using the above-mentioned parameters determined from spectroscopic and pump-probe studies [25–27, 33, 36, 37]. The ASE was taken into account according to the approach from [39]. The results of approximating the Ce: LiCAF non-linear absorption coefficient as a function of the pumping waist position are shown in figure 5. Some discrepancy between the experimental and calculated curves is due to the uncertainty of the wavelength of ACE, its spectral width and divergence.

Conclusion

To the best of our knowledge, the impact of ASE on the results of z-scanning experiments of thick Ce:LiCAF active media is shown for the first time. Thus, the z-scan technique can be used not only to study non-linear features of transparent samples, non-linear absorption and/or saturation effects, but the gain performances. Comprehensive studies are now in progress and the fitting procedures that will allow us to determine the basic parameters of the processes in AM from z-scan experiments are being developed. Results of these studies will be submitted in the near future in the *J. of Laser Physics.*

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