Spatio-Temporal Characterization of Pump-Induced Wavefront Aberrations in Yb$^{3+}$-Doped Materials

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A comprehensive spatio-temporal characterization is presented describing the pump-induced wavefront aberrations in Yb$^{3+}$-doped YAG, CaF$_2$, and fluorophosphate glass. Time-resolved interferometric measurements were performed to reveal the profiles of the total optical path differences (OPDs), which are described by the spatio-temporal superposition of thermal as well as electronic contributions, across the free aperture of the considered diode-pumped active materials. These contributions were individually determined by a COMSOL-based thermal profile model along with a detailed characterization of the electronic changes by measuring the single-pass gain and the spatial fluorescence profile. Due to the low quantum defect, the amplitude of the electronic component becomes comparable for all three materials and, in the case of Yb:CaF$_2$, almost completely compensates the thermal component resulting from a pump pulse during the time frame of laser pulse amplification. Finally, all relevant material constants such as the photoelastic constant $C'$ and the polarizability difference $\Delta \alpha$ could be determined during this investigation, allowing the accurate modeling of the total pump-induced wavefront aberrations and subsequent optimization for laser systems worldwide employing these Yb$^{3+}$-doped materials.

1. Introduction

The ongoing development of high power laser systems towards higher peak and average powers enables various potential industrial and scientific applications, such as advanced materials processing and laser shock treatment,[1] pulsed electron[2] or ion sources,[3] the generation of ultrashort XUV and X-ray radiation,[4] and medical applications based on intense ion beams.[5] For these purposes, in addition to the repetition rate and pulse duration, the energy contained within the laser pulse is of crucial importance. Here, diode-pumped systems based on Yb$^{3+}$-doped laser materials are promising candidates that are capable of delivering high energy pulses at a high repetition rate and efficiency. Currently developed systems have demonstrated pulse energies of more than 50 J with pulse durations below 100 fs[6] and more than 100 J for pulses in the nanosecond regime.[7]

However, because of the fixed saturation parameters and the limited laser induced damage threshold (LIDT) of the laser materials, an energy scaling of laser systems is possible only with respect to the cross-sectional area of the laser beam. For high pulse energies, a large pumped area in the laser material – and thus, a large beam diameter – is necessary. In this case, a spatially homogeneous flat-top-like gain profile and therefore a homogeneous profile of the pump radiation is desirable in order to ensure a high beam quality, even with large beam diameters. Due to the LIDT, the spatial beam quality is often the limiting parameter for the output energy of a laser system. However, in addition to the spatial gain profile, the beam quality is also influenced by the wavefront of the amplified laser pulse. Here, the pumping of the active materials induces aberrations that can impair the quality of the near field profile and the final focusability. These aberrations are a consequence of spatially inhomogeneous optical path differences (OPDs) $\phi(t)$ of the active material, which occur due to the thermal load[8] and pump-induced changes of the electronic configuration,[9] that cause different parts of the laser beam profile to traverse different optical path lengths through the material. The OPD profiles directly imprint themselves onto the wavefront of the laser pulses to be amplified, and thus, can cause significant modulations of the spatial profile during the subsequent laser pulse propagation. These spatial beam distortions limit the total output energy as well as the focusing of the final laser pulses, eventually resulting in a significant reduction of the available peak power and intensity. Therefore, pump-induced aberrations must be characterized and, if possible, corrected in order to further increase the peak power of state-of-the-art laser systems. This is of utmost importance for

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the development of laser systems with a peak power in excess of 10 PW, which are currently in development under the Apollon\textsuperscript{[10]} and the Extreme Light Infrastructure (ELI\textsuperscript{[11]}) projects.

For Yb\textsuperscript{3+}-doped materials, the small energy difference between the pump level and the laser level, along with pumping using narrowband high-power laser diodes, results in a quantum defect of approximately 9\%, which is significantly lower compared to other dopants, e.g., Ti\textsuperscript{3+} (33\%) or Nd\textsuperscript{3+} (24\%). This leads to a reduced heat load and lower thermally-induced OPDs of the pumped material. Consequently, electronically-induced OPDs become relatively more important. For our investigations, we have studied two different categories of Yb-doped materials. We have characterized Yb:YAG, which is well suited for the generation of high energy pulses with nanosecond durations\textsuperscript{[7]} due to the high small signal gain. Furthermore, we have also selected Yb:CaF\textsubscript{2} and Yb-doped fluorophosphate glass (Yb:FP\textsubscript{15}\textsuperscript{[8]})\textsuperscript{[9]}, which are characterized by the amplification of a broad spectral bandwidth, thus enabling the generation of high energy pulses with durations of less than 100 fs.\textsuperscript{[6,13]} The large amplification bandwidth, however, results in a lower small signal gain, and therefore, a large number of amplification passes are required for a high output energy.

With these low gain materials and a high number of passes, the beam quality is more predominantly influenced by the pump-induced aberrations than with high gain materials (e.g. Yb:YAG), which only require a lower number of passes to reach the same final energy. Furthermore, the three characterized materials differ in their thermal properties. Since Yb:YAG and Yb:CaF\textsubscript{2} provide a cubic crystal structure, both materials have significantly higher thermal conductivities than Yb:FP\textsubscript{15} glass. Due to the different material structures, there are also considerable differences in the thermally-induced stress. In addition, Yb:CaF\textsubscript{2} and Yb:FP\textsubscript{15} also exhibit a negative thermo-optic coefficient \(dn/dT\), resulting in a partial compensation of the thermally and electronically generated OPDs. Until now, detailed investigations have been performed for Yb:YAG, in which the two contributions have been characterized within a small area of only 150 \(\mu m^2\), which was pumped by a Gaussian shaped profile.\textsuperscript{[9]} The spatial dependencies have been described by assuming a “lens-like” profile. For larger beam diameters, however, it is necessary to consider the complete, spatial OPD profile at the time of the laser pulse amplification. By means of this characterization, the aberrations with higher spatial frequencies can be distinguished from a pure lens effect. Characterizations of the electronically-induced phase change have not yet been published for Yb:CaF\textsubscript{2} or Yb:FP-glass. Previous investigations of Yb:CaF\textsubscript{2} neglected the electronic aberrations, which led to large deviations of the measured and theoretically determined OPDs.\textsuperscript{[14,15]} With the complete understanding of these aberrations, present-day laser systems employing Yb\textsuperscript{3+}-doped active materials, e.g., at the Helmholtz-Zentrum Dresden-Rossendorf (PEnELOPE\textsuperscript{[16]}) and the Helmholtz-Institute Jena (POLARIS\textsuperscript{[6]}), which are designed for 1 PW peak power, can be optimized towards higher energies and an improved beam quality.

In this paper, we report on the results of a comprehensive spatio-temporal characterization of the total pump-induced OPD profiles of the materials Yb:YAG, Yb:CaF\textsubscript{2} and Yb:FP\textsubscript{15}. The profiles across the free aperture of the considered materials were extracted from time-resolved interferometric measurements. Since

2. Experimental Setup

The experimental setup utilized for the characterization is depicted in Figure 1a. Each active material (AM) was placed in a water-cooled mount and end-pumped from one side using a 940 nm, 6.8 kW spatially homogenized laser diode source. The pump source (PS) exhibits a square-shaped flat-top profile as shown in Figure 1c, necessary for a homogeneous pump distribution inside the active material, which was reduced to a beam full width of 4.5 mm (FWHM) after a zoom lens as shown in Figure 1b. A frequency-doubled Nd:YVO\textsubscript{4} continuous wave laser (CW1) operating at 532 nm was split into two and recombined in a Mach-Zehnder configuration around the active material. The wavelength of the CW1 laser lies outside the emission and absorption spectra of the Yb\textsuperscript{3+}-doped active materials and could be isolated from the fluorescence light using a spectral filter. This prevents a disturbing influence of the fluorescent light on the fringe patterns collected by the camera C1. The CW1 laser was...
magnified to measure the spatially resolved OPDs within the entire clear aperture of the considered active materials.

The relevant parameters of the characterized active materials including the dimensions and the doping concentration, along with the utilized pump pulse duration \( \tau_p \) and repetition rate, which were adjusted with respect to the materials’ lifetime and thermal conductivity, are displayed in Table 1. The length of the material was chosen such that 90% of the irradiated pump power is absorbed in each case. Each material was pumped by the pulse train for several minutes before measurements were recorded, in order to approach a steady-state ("thermalized") regime in which the energy absorbed by the pump throughout the material is balanced by the removal of heat via the water-cooled mount and the surface-air interfaces. Thus, the average temperature no longer increases with the number of cycles. This results in the observation of the behavior of the pump-induced aberrations in an equilibrium state during a periodic pump process.

The pump-induced OPD profiles were determined from the interferometric measurements via the Fourier-transform method. Here, the OPD profile imprinted onto the phase of the CW laser was extracted and unwrapped from the recorded fringe patterns. To obtain the full OPD profiles throughout the pumped active medium, the rear surface of the medium was imaged by an objective mounted on the camera C1 as depicted in Figure 1 (C1: Allied Vision Manta G-032B CCD with F/1.8 objective and 2x focal length extender). In addition, a reference measurement of the unpumped material was subtracted from each measurement in order to avoid influences of potential material inhomogeneities and the measurement setup itself. The camera was programmatically triggered and delayed (Stanford Research Systems, Model DG645) with respect to the pump pulse. Recording multiple images with various delays allows for the retrieval of the OPD profiles throughout a complete pump cycle, which spans the period during the pump pulse and the cooling of the material up until the subsequent pump pulse. The delay times during the pump pulse were chosen with a 50 \( \mu s \) resolution, which is nearly twice the minimum exposure time of the CCD of 26 \( \mu s \). After the pump pulse, we chose a gradually decreasing resolution until the end of the pump cycle.

### 3. Thermal Contributions

The OPD profiles (\( \phi(t) \) [nm]) at the end of the pump pulse are displayed in Figures 2a–c, along with the corresponding vertical lineouts through the center of the profiles at various timestamps within the pump cycle (Figures 2d–f). The OPD profiles at the initial state directly before the pump pulse at time \( t = 0 \) ms, which are shown by the purple lines in Figures 2d–f, are non-zero due to the thermal gradient from the thermalized state of the materials. Here, Yb:YAG and Yb:FP15 show a positive residual profile for our employed pump power, while the profile for Yb:CaF\(_2\) is reversed. During the pump pulse, the amplitude of the OPD profiles of Yb:YAG and Yb:FP15 increases as depicted by the blue line, which corresponds to the OPD profiles at the end of the pump pulse at \( t = \tau_p \). After the pump pulse, the OPD profiles of both materials relax and finally return to the initial state at the end of the pump cycle (red lines). The relaxation of the OPD profile of Yb:CaF\(_2\), however, does not show such a clear behaviour.

For a complete understanding of the thermally-induced components of the OPD profile, the temperature of the pumped active material was tracked with a FLIR P620 thermal infrared imaging camera (IR in Figure 1a). In addition, the spatio-temporal temperature profile of the pumped active materials was simulated in three dimensions using the finite element analysis software COMSOL (Version 3.5, Sweden) and knowledge of the thermal properties of Yb:YAG, Yb:CaF\(_2\), and Yb:FP15. The comparison between the simulated and measured front and back temperature profiles for the case of the pumped and thermalized Yb:FP15 sample is displayed in Figure 3a, along with a thermal image of the pumped Yb:FP15 sample in Figure 3b and a slice through the pumped Yb:FP15 model in Figure 3c. The maximum relative discrepancy between the simulated and measured results was less than \( \pm 3\% \) for each material.

Due to the good agreement between the temperature simulation and measurement, the thermal contributions to the OPD profile can be determined. By computing the mean of the simulated temperature shift \( (T(r, t) - T_0) \) along the material thickness \( L \), the OPD profile \( \phi(t, r) \) can be calculated using

\[
\phi(t, r) = \int \frac{d\phi}{dT} + 2n_0\alpha_T C + (n_0 - 1)(1 + \nu)\alpha_T \left( T(r, t) - T_0 \right) L.
\]

and compared to the results of the interferometric measurements. In Eq. (1), the three terms within the squared brackets form the total thermo-optic coefficient \( C \), which describes the refractive index changes due to the thermo-optic coefficient \( d_n/dT \), the stress via the photoelastic effect, and the thermal expansion of the material considering Poisson’s ratio \( \nu \), the coefficient of thermal expansion \( \alpha_T \), and the initial refractive index \( n_0 \) of the active material. The temperature of the unpumped active material \( T_0 = 20.5 \) °C corresponds to the cooling temperature at the lateral surfaces.

In published literature, the thermo-optic coefficient \( d_n/dT \) as well as the thermal expansion \( (n_0 - 1)(1 + \nu)\alpha_T \) are well-characterized for all three materials. To determine the stress-induced OPD, the photoelastic constant \( C \), along the radial direction was calculated using a reduced form of the elasto-optic tensor and the elasto-optic coefficients \( p_{11} \) and \( p_{12} \) [22]. However, both coefficients are available in literature only for undoped YAG and CaF\(_2\) crystals, while none are available for FP15 glass. For this reason, we experimentally determined the stress component for all three Yb(\( L \)) doped materials by fitting the calculated OPD profile (see Eq. 1) to the measured profile at the thermalized state – before the pump pulse arrives (\( t = 0 \) ms) – using \( C \) as the free parameter. The \( C \) values were determined to be \( 3.2 \times 10^{-11} \) for

| Yb:YAG | Yb:CaF\(_2\) | Yb:FP15 |
|--------|------------|---------|
| Yb-Doping \( [\times 10^{20} \text{ ions} / \text{cm}^3] \) | 4.2 | 3.8 | 6 |
| Diameter [mm] | 23 | 28 | 12.7 |
| Length [mm] | 4.5 | 30 | 13 |
| Pump Duration \( \tau_p \) [ms] | 0.9 | 2 | 1.4 |
| Repetition Rate [Hz] | 2 | 4 | 0.2 |
Figure 2. Pump-induced OPD profiles for a) Yb:YAG, b) Yb:CaF$_2$, and c) Yb:FP15 at the end of the pump pulse (denoted by the timestamps). Lineouts of the OPD profiles at various times during the pumping and cooling periods are displayed for d) Yb:YAG, e) Yb:CaF$_2$, and f) Yb:FP15.

Figure 3. a) Comparison of measured (solid) and simulated (dashed) Yb:FP15 front and back thermal profiles, displayed for times 0, 0.1, 1, 2.5, and 5 seconds within the pump cycle (rep. rate 0.2 Hz; pump cycle = 5 s). b) Thermal image of the pumped Yb:FP15 (front). c) Slice through the pumped Yb:FP15 model in COMSOL.
Yb:YAG, $-3.7 \times 10^{-2}$ for Yb:CaF$_2$, and $1.9 \times 10^{-3}$ for Yb:FP15. The resulting profiles as well as the residual errors are depicted in Figure 4. Furthermore, all values regarding the total thermo-optic coefficient $\chi$ are summarized in Table 2 for the three characterized materials. We found a good agreement of the determined stress component with published values for Yb:YAG. The $C_r$ value for Yb:FP15 is also comparable to that of Yb-doped silicate and phosphate glasses ($C_r = 1 \times 10^{-3}$)\cite{23}. However, the stress-component for Yb:CaF$_2$ is an order of magnitude larger than for the case of Yb:YAG and Yb:FP15. We have further found a discrepancy between the theoretically calculated value for undoped and the experimentally determined value for Yb$^{3+}$-doped CaF$_2$.

Due to the replacement ratio of 2 Yb-ions for 3 Ca-ions, the face-centered-cubic lattice structure is considerably disturbed by the doping\cite{24}. This results in, e.g., a strongly doping-dependent thermal conductivity\cite{21} and thus, can also influence the internal stress distribution of the pumped material. An additional independent measurement of the thermal-stress-induced birefringence was conducted to confirm this value, by recording the laser profile through the pumped Yb:CaF$_2$ sample placed between two crossed polarizers. The measured relative transmitted amplitude and spatial profile, which are shown in Figure 5, confirm our $C_r$ value. The measurement was also carried out with 50 $\mu$s resolution at the time $t = 0$ ms, directly before the arrival of the pump pulse. The Yb:CaF$_2$ crystal was cut in the (111) direction, which was experimentally verified using the Laue X-ray diffraction method\cite{25}. Possible causes involving the dependence of the stress-optic coefficients on the doping of the material are currently being investigated.

After the pump pulse is over, the thermal gradient in the active material – and thus, the thermal OPD profile – relaxes due to the outward transfer of heat towards the cool surface of the mount. The thermal diffusivity $D$ – formed from the thermal conductivity $K_{th}$, specific heat capacity $C_p$, and density $\rho$ – along with the pump beam radius $w_p$ can be used to give an approximate value for the characteristic timescale of the relaxation of the temperature increase per pump pulse\cite{23} which will be referred to as the thermal diffusion time:

$$\tau_D = \frac{w_p^2}{4 \cdot D} = \frac{w_p^2 \cdot C_p \cdot \rho}{4 \cdot K_{th}}. \quad \text{(2)}$$

4. Electronic Contributions

Pumping an active material generates not only a temperature gradient within the material, but a localized region of excited laser-active ions as well. Since the excitation directly leads to a
change in the charge distribution within the laser-active ions, the dynamic response of the material to external fields changes. For Yb-doped active materials, this response manifests itself as the formation of excited-state near-UV absorption bands, caused by ligand-to-metal charge transfer transitions \((O^2 \rightarrow Yb^{3+}\) for Yb:YAG\)\(^{[20]}\) along with interconfigurational \((4f^n \rightarrow 4f^{n-1}d\) for Yb:YAG) UV transitions of the Yb\(^{3+}\)-ions \(^{[27]}\). As a consequence, the active material provides a polarizability difference between different excited states, and will, according to the Lorentz-Lorenz formula \(^{[28]}\) exhibit a change in the refractive index that scales with the population inversion. This results in an additional OPD profile, also referred to as a population lens, that the material imbues onto the incident seed laser. The amplitude and shape of this electronic aberration can be calculated by \(^{[9]}\)

\[
\Delta \phi_e(r, t) = \frac{2\pi}{n_0} \left( \frac{n_0^2 + 2}{3} \right)^2 \Delta \alpha \Delta N(r, t) L, \tag{3}
\]

which requires the knowledge of the spatio-temporal profile of the inverted population \(\Delta N(r, t) \) \([\text{ions/cm}^2]\) within the pumped active material, the polarizability difference \(\Delta \alpha \) \([\text{cm}^2]\), and the unperturbed refractive index of the material \(n_0\). However, the polarizability difference is strongly dependent on the host material of the Yb\(^{3+}\)-ions and, as seen in Table 3, can vary nearly an order of magnitude between materials. References for Yb:CaF\(_2\) and Yb:FP15 are also not available here. Much like the thermal OPD profile, the electronic OPD profile is expected to relax after the pump has ended, yet with a characteristic time according to the fluorescence lifetime. Therefore, both effects can be distinguished temporally.

**Table 3.** Previously reported magnitudes of the polarizability difference \(\Delta \alpha\) for various Yb\(^{3+}\)-doped active materials.

| Material    | \(\Delta \alpha \times 10^{-26} \text{ cm}^2\) |
|-------------|---------------------------------------------|
| Yb:YAG\(^{[6]}\) | 1.95 ± 12.8% |
| Yb:KGW\(^{[21]}\) | 10.3 ± 4.9% |
| Yb:YVO\(_4\)\(^{[21]}\) | 8.1 ± 4.9% |
| Yb:Lu\(_2\)O\(_3\)\(^{[24]}\) | 2.6 ± 23.1% |
| Yb:Sc\(_2\)O\(_3\)\(^{[24]}\) | 1.9 ± 26.3% |

**Figure 6** shows the temporal progression of the mean center values (between ±1 mm) of the total OPD profiles throughout the pumping and cooling periods, obtained by the time-resolved interferometer measurements. The black points in each plot represent an average over 10 measurements and display the temporal progression of the total OPD profiles, with the grey points corresponding to the standard deviation. In order to distinguish between the different contributions, the measured temporal OPD profiles were fitted to a superposition of two exponential decay functions with two different lifetimes. **Table 4** compares the amplitudes and time constants of the two types of pump-induced OPD profiles via their relaxation profiles \(\phi_{relax}(r, t)\), which spans the complete cooling period beginning at the end of the pump pulse – marked by the dashed red line in Figures 6a–c. The amplitudes of the relaxation profiles \(\Delta \phi_0\) and \(\Delta \phi_e\) correspond to the magnitudes of the thermal and electronic OPD profiles generated from each pump pulse. The time constant from the electronic OPD component \(\tau_e\) closely resembles the fluorescence lifetime \(\tau_0\), yet is slightly longer due to reabsorption over the material length. The time constant from the thermal OPD component \(\tau_th\) is on the order of the thermal diffusion time \(\tau_D\), which is approximated \(^{[28]}\) by neglecting the heat transfer rate through the unpumped volume of the material. The comparability of the approximation \(\tau_D\) and the determined thermal relaxation time \(\tau_{th}\) is thus visibly reduced with an increase in the ratio between the volume of the full material and the pumped region \(V_{total}/V_{pump}\) (FP15 ≈ 6, YAG ≈ 20, CaF\(_2\) ≈ 30).

The amplitudes and temporal profiles of the population inversion, knowledge of which is necessary to determine the electronic aberrations, can be extracted from measurements of the unsaturated small signal gain \(G\) and information regarding the emission and absorption spectra of the active material:\(^{[31]}\)

\[
G = \exp \left[ L \cdot N_{tot} \cdot \left( \frac{\Delta N}{N_{tot}} \cdot \sigma_e - \left( 1 - \frac{\Delta N}{N_{tot}} \cdot \sigma_e \right) \right) \right], \tag{4}
\]

The number density of the Yb\(^{3+}\) ions is denoted as \(N_{tot} \) \([\text{ions/cm}^3]\), while the emission and absorption cross-sections are given as \(\sigma_e \) \([\text{cm}^2]\) and \(\sigma_a \) \([\text{cm}^2]\) respectively. Reabsorption, due to the overlap of emission and absorption spectra in Yb\(^{3+}\)-doped active materials, has been included in Eq. 4. To measure the temporally-resolved small signal gain, a 1030 nm wavelength cw-laser (CW2 in Figure 1) passes through the center of the pumped active material and onto a Thorlabs DET36A/M photodetector (PD) connected to an oscilloscope (OSC). The power of the cw-laser was sufficiently low, such that the gain within the pumped active material was not saturated. In addition, the 2.2 mm \((1/e)^2\) beam diameter was small enough to guarantee a perfect overlap between the pumping area and the cw-laser.

From the temporally-resolved thermal simulations, which are depicted in orange in Figure 6, and the temporal gain measurements (blue) for the three Yb\(^{3+}\)-doped active materials, a superposition of the thermal and electronic OPD profile was constructed. This superposition was fitted to the measured temporal OPD profiles with the polarizability difference as the free parameter. The resulting simulated temporal OPD profiles for the three materials are depicted by the purple curves in Figures 6a–c. The determined polarizability difference for Yb:YAG of 1.93 \(\times 10^{-26} \) \([\text{cm}^3]\) ± 3.7 % closely matches the value given by Antipov et al. \((1.95 \times 10^{-26} \) \([\text{cm}^3]\) ± 12.8 %\)\(^{[9]}\), but with a higher precision. The polarizability difference values for Yb:CaF\(_2\) and Yb:FP15, which we determined as 0.79 \(\times 10^{-26} \) \([\text{cm}^3]\) ± 2.9 % and 1.15 \(\times 10^{-26} \) \([\text{cm}^3]\) ± 6.8 %, respectively, have not yet, to the best of our knowledge, been reported in literature.

For Yb:YAG, the electronically-induced OPDs (Figure 6a, blue) compared to the thermally-induced OPDs (Figure 6a, orange) are similar in both sign and magnitude for each pump shot. For the Yb:CaF\(_2\) case, both OPDs are similar in magnitude but are competing (opposite in sign), and cancel each other out (Figure 6b, purple) during a time interval near the end of the pump pulse. The signs of both forms of aberrations are positive for Yb:FP15, and the electronic OPDs (Figure 6c, blue) are larger than the thermal OPDs (Figure 6c, orange) by nearly a factor of three. The slight undershooting of the measured OPDs for Yb:FP15 within
Figure 6. Temporal comparison of the measured (black) and superposition (purple) of the thermal (orange) and electronic (blue) OPDs for a) Yb:YAG, b) Yb:CaF₂, and c) Yb:FP15. The temporal profiles display the magnitude of the OPD profiles generated per pump pulse, shown along a logarithmic time axis. The dashed red lines in a - c) denote the end of the pump pulse.

Table 4. OPD relaxation \( \phi_{\text{relax}}(t) = A_{\text{e}} e^{-t/\tau_{\text{e}}} + A_{\text{th}} e^{-t/\tau_{\text{th}}} \).

| Material   | \( A_{\text{th}} / A_{\text{e}} \) [nm] | \( \tau_{\text{th}} / \tau_{\text{e}} \) [ms] | Fit R²-value | \( \tau_{\text{D}} \) [s] | \( \tau_{\text{fl}} \) [ms] |
|------------|----------------------------------------|------------------------------------------|--------------|----------------|----------------|
| Yb:YAG     | 46.6 / 46.0                            | 0.49 / 1.0                               | 0.98         | 0.43 \( ^{[19]} \) | 0.95 \( ^{[31]} \) |
| Yb:CaF₂    | 50.6 / 65.8                            | 0.25 / 2.7                               | 0.96         | 0.46 \( ^{[21]} \) | 1.9 \( ^{[31]} \) |
| Yb:FP15    | 19.5 / 68.2                            | 4.9 / 2.3                                | 0.92         | 4.86 \( ^{[12]} \) | 1.4 \( ^{[12]} \) |

the 10∼100 ms range is most likely a result of acoustic vibrations due to the rapid expansion of the glass and is currently under investigation. The impact of the electronic aberrations on the total pump-induced OPD profiles for Yb:CaF₂ and Yb:FP15 is particularly strong – in comparison to Yb:YAG – due to the low value of the total thermo-optic coefficient. Here, the negative \( \mathrm{dn}/\mathrm{dT} \) compensates the positive value of the material expansion, thereby reducing the total thermo-optic coefficient and leading to a larger role for the electronic aberrations. The ratio between the magnitudes of the electronic and thermal OPDs remains valid with increasing pump power until pump saturation, after which the increase in population inversion \( \Delta N/dt \) is no longer linearly proportional to the pump power.

5. Pump-Induced Wavefront Aberrations

The superposition of the two types of aberrations can also be verified spatially within the thermalized regime. Figure 7 reveals the pump-induced OPD profiles at the end of the pump pulse for each material. Subtracting the measured OPD profile (black) from the simulated thermal OPD profile (orange) results in the spatial OPD profile corresponding to the electronic aberrations (blue), which matches the calculated OPD profile for the electronic aberrations (red) using Eq. 3. Here, the amplitude of the population inversion at the end of the pump pulse was determined by the temporally-resolved small signal gain measurements and Eq. 4, with the spatial profile of the population inversion given by the emitted fluorescence light of the material. The fluorescence light was recorded as the integrated profile along the thickness of the pumped materials (Figure 1b for Yb:CaF₂) at the end of the pump pulse using the camera C2 in Figure 1a operating with a CCD exposure time of 26 \( \mu \)s. The baseline of the measured OPD profiles (black) were adjusted to match the offset of the thermal simulations (orange) to account for the temperature change on the edges between the unpumped and thermalized states, which produces a constant phase offset that is removed during the unwrapping process of the fringe analysis.

The impact of these aberrations can be further explored by separating the total pump-induced OPD profiles into a defocus component and the residual high frequency spatial phase distortions. The defocus term represents a pure parabolic curvature of the wavefront, which can be compensated in an amplifier layout by implementing additional curved surfaces – e.g., lenses or mirrors – or by altering the distance between imaging optics. The high frequency phase distortions, however, cannot be corrected using these simple methods. In order to distinguish between these two consequences, a parabolic defocus term was fitted to the pump-induced OPD profiles (black profile, Figure 7a–c).
within the pumped region at the end of the pump pulse, using an effective focal length (EFL) under the thin lens approximation as the free parameter. Removing this defocus component from the pump-induced OPD profiles reveals the residual phase distortions imprinted onto the wavefront of an incident seed laser. Figure 8 depicts the phase distortions (red) resulting from a single pass through the pumped region of the considered thermalized materials after a single subsequent pump pulse ($t = \tau_p$), with the amplitude given in units of wavelengths ($\lambda_0 = 1030$ nm). The fitted EFLs for the defocus (blue) terms were 31 m ± 1.4% for Yb:YAG, −35 m ± 0.5% for Yb:CaF$_2$, and 53 m ± 1.5% for Yb:FP15. The wavefront of an incident seed laser passing through the pumped region of Yb:YAG or Yb:FP15 experiences an oscillatory structure of phase distortions with opposite signs in the center and near the edges of the pumped region. Despite the low peak-to-valley magnitudes of ~0.03 $\lambda$, the spatial phase distortions within an amplifier providing, e.g., up to 20 material passes---which is necessary due to the low gain of Yb$^{3+}$-doped broadband materials---rapidly accumulate, leading to an enormous influence on the spatial intensity profile upon propagation. In the case of Yb:CaF$_2$, however, such an influence is significantly lower due the opposite signs of the electronic and thermal aberrations, which produces a parabolic-like OPD profile within the pumped region that almost completely vanishes with the correction of the defocus term, such that an optimal amplification of the laser pulses can be guaranteed.

6. Conclusion

In conclusion, a comprehensive spatio-temporal characterization of pump-induced optical path differences and the resulting wavefront aberrations has been completed for Yb$^{3+}$-doped YAG, CaF$_2$, and FP-glass. Interferometric measurements of the total OPD profiles could be matched to the superposition of the spatial and temporal profiles of the electronic and thermal contributions. The thermally-induced OPD profiles were simulated using a COMSOL-based thermal profile model, and verified by surface temperature measurements with a thermal infrared imaging camera. The electronically-induced OPD profiles were determined by performing single-pass gain measurements and recording the spatial fluorescence profile. The photoelastic constant $C$, and polarizability difference $\Delta \alpha$, extracted for each material, agree with previously published values for Yb:YAG, verifying the presented characterization method. We have found a discrepancy of the stress-induced OPD component regarding Yb$^{3+}$-doped and undoped CaF$_2$, which was validated by an additional birefringence measurement. Additionally, we precisely characterized the polarizability difference of Yb:CaF$_2$ and Yb:FP15, which enables the simulation of electronic OPDs for both materials in the future. The electronic contributions per pump pulse were found to be significantly large: 3 times larger than the thermal OPDs for Yb:FP15 and large enough to cancel out the thermal OPDs in Yb:CaF$_2$ near the extraction time. Furthermore, the magnitude of the pump-induced OPD profiles---and thus, the wavefront aberrations---are inhomogeneous and scale with each material pass, rapidly resulting in a significant influence on the propagation and focusing of high energy laser pulses amplified in Yb$^{3+}$-doped materials. With the understanding of these results, the impact of the pump-induced wavefront aberrations on the performance of the laser can now be modeled in order to optimize the pump laser parameters and the amplifier design, leading to an improvement in the spatial profile of the laser pulse and a subsequent increase in the maximum pulse energy for high power laser systems worldwide employing Yb-doped active materials.

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Conflict of Interest

The authors declare no conflict of interest.
