

## Formation of plasma channels in distilled water by femtosecond laser pulses in the mid-infrared range

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Received December 11, 2023

Revised January 09, 2024

Accepted January 16, 2024

Experimental studies of the parameters of plasma channels formed in distilled water under the action of high power laser pulses with wavelengths of 1050, 1105, 1200, 1300, 1500, 1700 nm with durations of 130, 310, 100, 150, 110 and 80 fs, respectively, were carried out. As a result, the nonlinear refractive index of water was experimentally determined and the quadratic dependence of the critical self-focusing power on the pump wavelength was confirmed. The values of the critical self-focusing power for the wavelengths considered in this work are in the range of 3.8–17.8 MW.

**Keywords:** Plasma channel, critical power for self-focusing, filamentation in water, ultrashort infrared laser pulses, nonlinear optics, distilled water.

DOI: 10.61011/0000000000

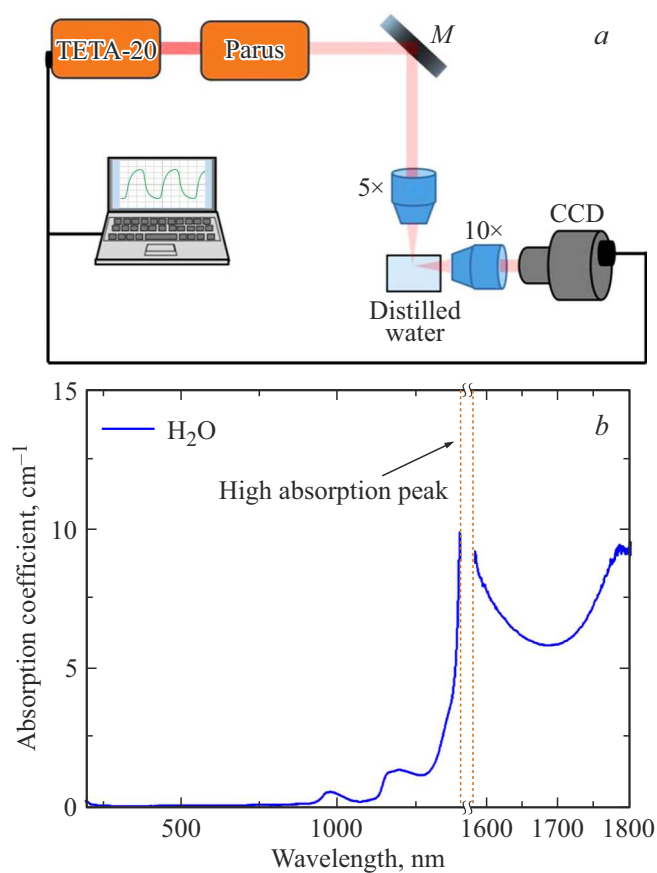
### Introduction

Physical phenomenon of powerful laser pulse self-focusing in various condensed media is followed by a series of non-linear processes such as formation of extended plasma channels [1,2], conical emission [3], supercontinuum generation [4]. When laser pulse propagates through a medium, non-linear polarization response of the material occurs. Refraction index in the area of maximum laser emission intensity becomes more significant and results in beam focusing and manifestation of self-focusing effect [5]. Laser sources with ultrashort pulses and point beam focusing are capable of achieving extreme emission intensities ( $10^{12}$ – $10^{13}$  TW/cm<sup>2</sup> and higher) by forming a plasma channel (filament) in the low focus waist area. Traces of this phenomenon are visually recorded by a luminous photoionized substance area [6]. Recent studies [7,8] has found a new nonlinear optical effect associated with multiple reduction of ultrashort (picosecond) laser pulse energy when optical breakdown was achieved in distilled water in the stimulated Raman back-scattering (SRS) and pumping field. Summation of nonlinear contribution of the refraction index and self-focusing provided the increase in intensity in the focal plane of the lens by more than an order of magnitude with a permanent laser pulse energy.

Investigations of filamentation in various media show that formation of a plasma channel is observed only when a particular critical laser emission power, that depends on the pumping wavelength, is achieved [9]. Beginning of

the filamentation area is defined on the assumption of the balance of self-focusing and diffraction induced, among other things, by defocusing effect of plasma formed in the focal plane. Critical power as well as nonlinear focus position during self-focusing and plasma channel formation are often described using the Marburger theory [10]. In case of point beam focusing of laser emission, the filament size may achieve several micrometers and this arouses the interest of researchers in precision laser machining of surfaces [11,12].

Many materials processing techniques, including generation of solid nanoparticles in solutions for biomedical applications are associated with utilization of distilled water. This has a direct effect on the process efficiency in particular when using powerful laser emission [13]. From this perspective, it is apparent that plasma channel parameter control conditions shall be studied. Filamentation in case of distilled water was investigated separately at various wavelengths mainly at 800 nm [14] and almost did not cover the infrared (IR) range above 1064 nm. The existing studies for direct measurement of nonlinear refraction index of water at wavelengths up to 1400 nm by the Z-scanning method [15] show wide data scatter [16,17]. Therefore, to update the existing description models of filamentation processes in dielectrics as a whole and water in particular, true data shall be obtained on nonlinear properties of a medium when powerful laser pulses are propagating in it in other wavelength ranges, especially, in near- and mid-IR ranges.



**Figure 1.** Experimental setup (a) and mid-IR absorption spectrum of water (b). Breakage at 1400–1580 nm is caused by the strong absorption band of water.

The study investigates the main parameters of plasma channels formed by powerful laser pulses in IR range with wavelengths of 1053, 1100, 1200, 1300, 1500, 1700 nm and durations of 130, 310, 100, 150, 110 and 80 fs, respectively, in distilled water. Nonlinear refraction index of water was assessed and quadratic dependence of critical self-focusing power on pumping wavelength in IR range was confirmed.

## Experimental

Parametric generation system (Parus, Avesta-Proekt) with solid-state pumping laser (TETA-20, Avesta-Proekt), maximum pulse energy  $E = 500 \mu\text{J}$ , pulse duration 250 fs, repetition rate 10 kHz and beam quality  $M^2 < 1.25$  were used to form plasma channels in a quartz cell with a wall thickness of 1.25 mm filled with distilled water. Focusing was conducted by a lens with numerical aperture  $NA = 0.1$  (LOMO) into a spot  $2.8\text{--}4.3 \mu\text{m}$  in radius depending on the pumping wavelength that corresponded to maximum peak intensities up to  $70\text{--}170 \text{ TW}/\text{cm}^2$  including the output idle wavelength power of the parametric oscillator.

Plasma channels were formed by laser pulses with varied energy at 1050, 1105, 1200, 1300, 1500 and 1700 nm

Experimental values of nonlinear refraction index of water for various pumping wavelengths

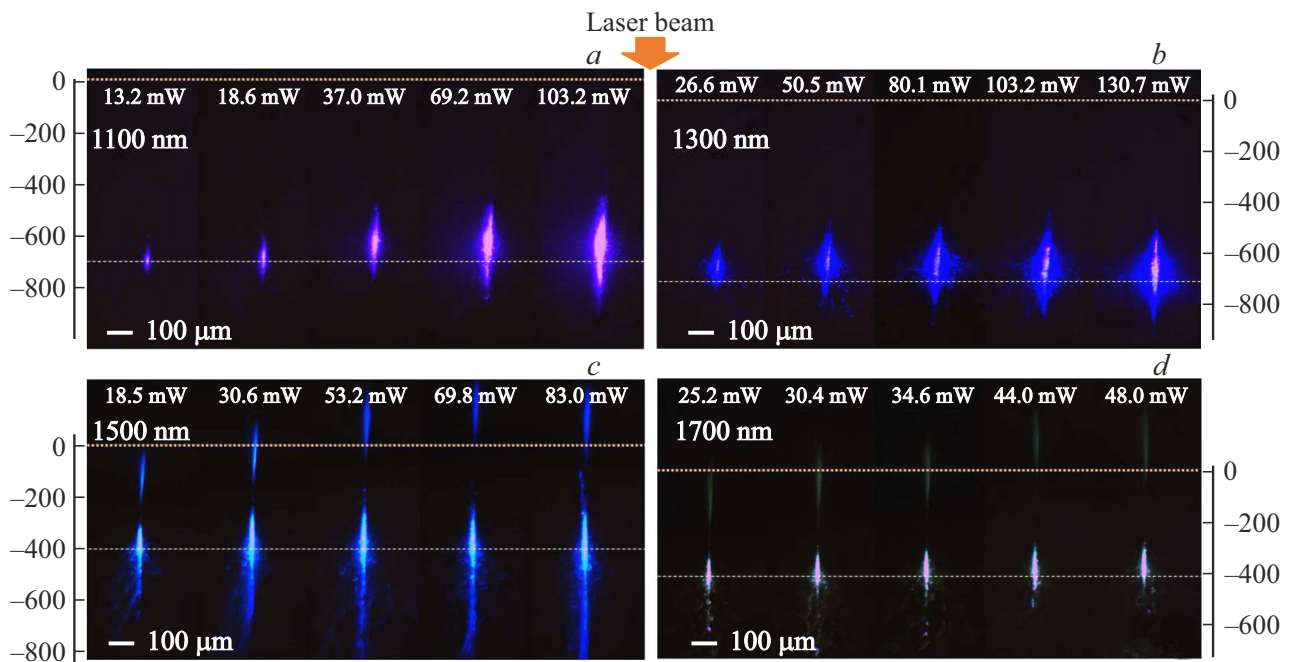
Pumping wavelength, nm	Pulse duration, fs	$P_{\text{cr}}$ , MW	$n_2$ , $10^{-16} \text{ cm}^2/\text{W}$
1053	134	$8 \pm 3$	$1.5 \pm 0.6$
1105	314	$4 \pm 1$	$4 \pm 1$
1200	102	$4 \pm 1$	$4 \pm 1$
1300	150	$8.4 \pm 2.5$	$2.3 \pm 0.7$
1500	112	$9 \pm 5$	$2.9 \pm 1.5$
1700	82	$18 \pm 9$	$1.8 \pm 0.9$

in a distilled water cell at a depth of 0.4–0.7 mm with vertical emission incidence (Figure 1, a). Water absorption in the specified wavelength range is  $\alpha = 1\text{--}10 \text{ cm}^{-1}$  with strong absorption bands within 1400–1600 nm and above 1800 nm as described in [18] and spectra recorded during the measurement of water flow through a quartz cell 10 mm in length using Vertex 70v IR Fourier spectrometer (Bruker). Plasma channel sizes were observed at right angle to the emission propagation direction using a lens with numerical aperture  $NA = 0.2$  (LOMO) and CCD camera equipped with 10x magnification eye piece (image pixel in the observation circuit is  $\sim 0.8 \mu\text{m}$ ). Pulse duration at the parametric oscillator outlet was recorded by ASF-30 autocorrelator (Avesta-Proekt) and was equal to 80 – 310 fs for all pumping wavelengths used in the study (Figure 1, b).

## Results and discussion

Figure 2 shows plasma channel images for 1100, 1300, 1500 and 1700 nm as most vivid examples. Bright luminous tracks  $100\text{--}200 \mu\text{m}$  in length occur at mean powers of  $10\text{--}25 \text{ mW}$  and increase in sizes up to  $500\text{--}800 \mu\text{m}$  with power growth. These images were used to determine critical power of self-focusing in water using a technique described in [19]. This technique involves identification of the difference between the linear and nonlinear lengths of the plasma channel where a track center obtained at the minimum pumping laser pulse energy is chosen to determine the linear (geometric) focus position. It should be noted that the luminous areas on the top of Figure 2, c, d are likely laser emission scattering near the water surface layer and do not act as the second plasma channel.

As a result, we get a dependence of the linear and nonlinear portions of the plasma channel on the mean pumping power whose intersection defines the critical self-focusing power (Figure 3, a, b for 1300 nm as an example). This approach used for other pumping wavelengths including the focusing optics and water absorption loss (absorption coefficient is  $0.3\text{--}5.9 \text{ cm}^{-1}$  for the employed wavelengths [18]) gives  $P_{\text{cr}} = 3.8\text{--}17.8 \text{ MW}$ . Saturation of



**Figure 2.** Plasma channel images in distilled water for 1100 nm (a), 1300 nm (b), 1500 nm (c) and 1700 nm (d) and various mean powers of laser pumping emission that are shown in the figure. Orange lines correspond to the cell water surface, white dashed line shows the focal plane position. Depth scales in  $\mu\text{m}$  are shown on the right and left of the pictures.

plasma channel length (Figure 3, b) may be caused by the effect of plasma defocusing and possible energy dissipation in favor of SRS stokes component generation in water [7], however, no detailed investigations of this effect have been carried out herein.

All experimental measurements of critical self-focusing power depending on the pumping wavelength are shown in Figure 3, c. Except 1053 nm and 1300 nm cases, experimental points are perfectly approximated by function quadratic in wavelength that is shown by the red dashed line on the diagram.

For the Gaussian profile beams, the expression of critical self-focusing power vs. wavelength becomes [1]

$$P_{\text{cr}} = \frac{3.77\lambda_0^2}{8\pi n_2 n_0}, \quad (1)$$

from which a value for  $n_2$  can be easily derived. Experimental dependence of the nonlinear refraction index for distilled water and 1050, 1105, 1200, 1300, 1500 and 1700 nm is shown in Figure 4 and has additionally marked points with corresponding errors. The points are the result of direct measurements of nonlinear characteristics by the Z-scanning methods in [15,16]. The resulting values are listed in the table.

In the wavelength range from 1053 nm to 1200 nm, our findings (Figure 4) match the previous values [16]. However, differences occur in case of pumping in the high water absorption area (1300–1700 nm) due to indirect measurement  $n_2$  in our study and finer focusing and, therefore, higher intensity in our experiment. In

addition, difficulties occur in correction of actual laser pulse power in the focusing and plasma channel formation area that require additional investigations, simulation of nonlinear response of the medium in a wide wavelength range, including the contribution of two-photon absorption and striction, and this is beyond the scope of the study.

## Conclusion

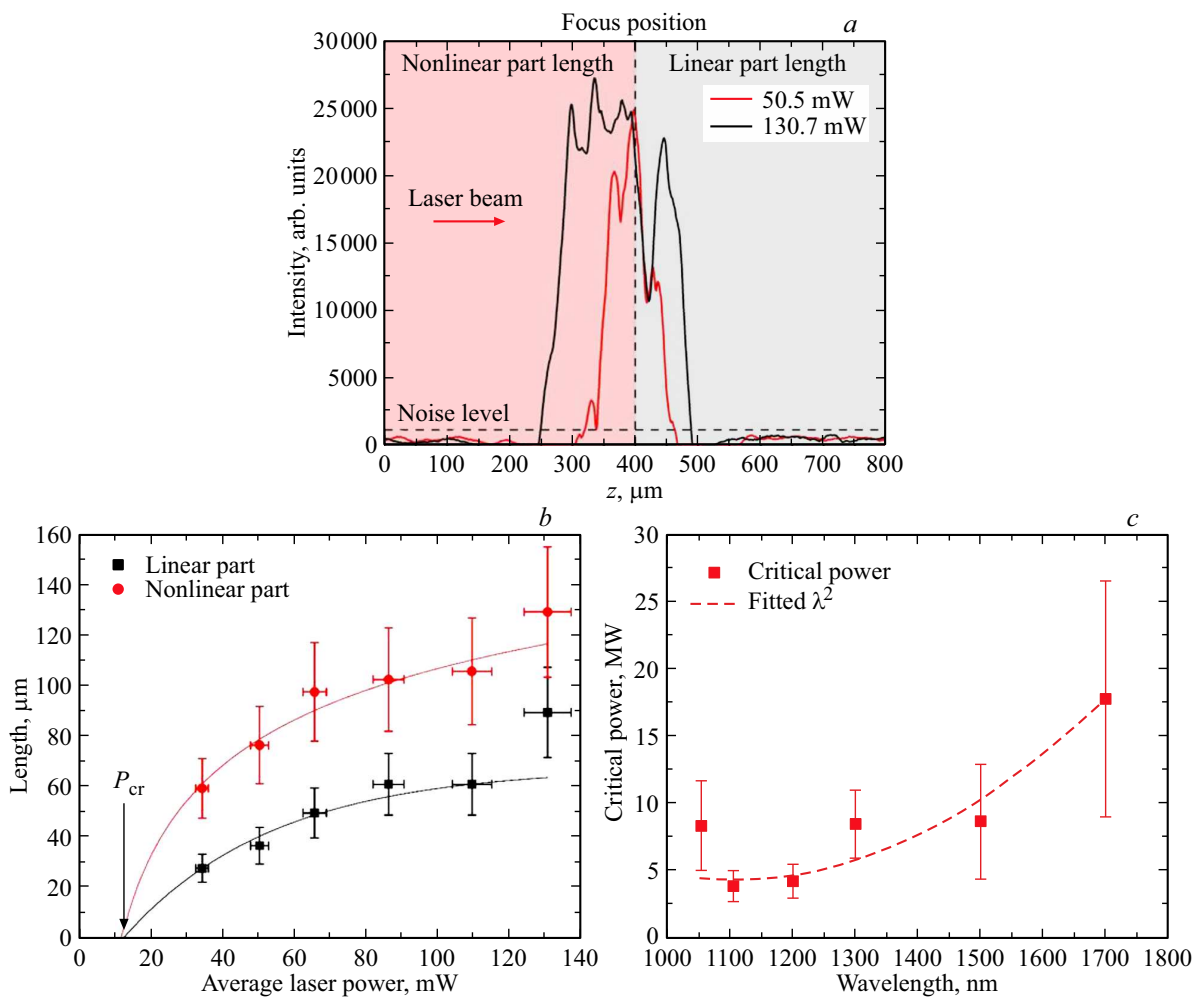
The study experimentally measures critical self-focusing powers and nonlinear refraction index of water for IR wavelengths (1050, 1105, 1200, 1300, 1500 and 1700 nm). The critical self-focusing power is proportional to the square wavelength in the given range. It should be noted that nonlinear refraction index  $n_2$  of distilled water at powers higher than the critical power is virtually unchanged with an increase in the wavelength of exciting femtosecond laser pulses within 1300–1500 nm.

## Funding

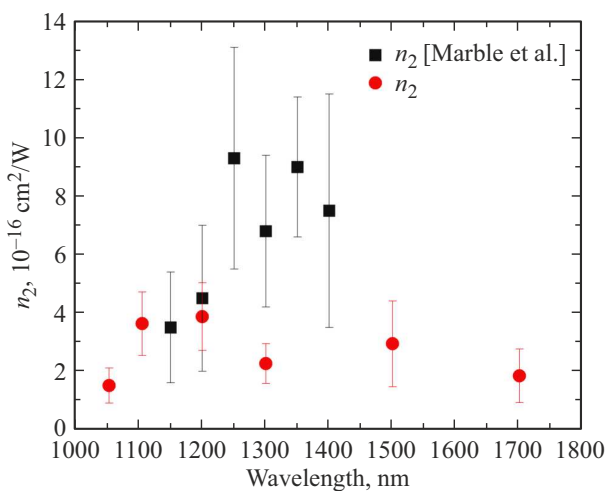
The study was supported by a grant provided by the Russian Science Foundation (project № 23-22-00453), <https://rscf.ru/en/project/23-22-00453/>.

## Conflict of interest

The authors declare that they have no conflict of interest.



**Figure 3.** (a) Example of identifying „linear“ and „nonlinear“ plasma channel length for 1300 nm and a mean power of 50.5 mW and 130.7 mW by the luminous intensity profiles of the channel on the  $z$  axis; (b) „linear“ and „nonlinear“ plasma channel lengths for 1300 nm pumping; (c) dependence of the critical self-focusing power  $P_{cr}$  on the pumping wavelength (red dots) and its approximation by function quadratic in wavelength (red dashed line).



**Figure 4.** Nonlinear refraction indices of water (red dots) calculated using equation (1). Comparison of the data with findings of [16,17] (black dots).

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*Translated by E.Ilinskaya*