Hydrodynamic phenomena in optical discharges in liquids under self-focusing of periodic-pulse laser radiation

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Abstract. Quasi-stationary flows under the effect of focused periodic pulse femtosecond laser radiation were generated and observed in liquid solvents: water, heavy water, alcohols, ketones, chloromethanes. The mechanism inducing directional flows appears to be directional collapse of the gas bubbles produced by multiphoton dissociation in a focused laser beam. Laser pulses of 450 fs length, up to 220 μJ pulse energy at repetition rates up to 10 kHz have induced stationary flows of liquid originated from the laser beam waist directed along or transversely to the beam axis. The streams along the beam axis were observed under low pulse power (10-20 μJ), provided precise lens adjustment. Lens displacement transversely to the beam axis led to splitting beam waist in two astigmatic foci. Both foci generate the streams along the beam axis. Counter directed streams have collided in the gap between foci, forming the flow spreading transversely to the laser beam. The increase of the pulse energy was followed by formation of the filament of self-focusing. Repeating cycles of focusing and defocusing along the filament produced several beam energy dissipation zones, each one generating separate streams along the beam axis. Colliding of the counter directed streams gave rise to complex flow pattern transversely and upward with respect to the beam axis.

1. Introduction
The issues of optical hydrodynamic interactions of laser radiation with liquids are studied in connection with applications in nanotechnology and chemical technologies for the tasks of micro objects manipulation, nanoparticles contained solutions production, materials processing under layer of liquid technologies, and so on [1-4].

A number of papers (for example, [6-8]) are devoted to the study of similar dynamic mechanisms of directed jets formation in liquids. In liquids directional motion can occur as a result of asymmetric collapse of gas bubbles formed by laser induced evaporation [6], a phenomenon similar to the collapse of the region of reduced gas density that occurs due to gas expanding after optical breakdown [5]. When focusing femtosecond laser beam in a liquid, multiphoton processes occurring at a high intensity of laser radiation near the beam waist lead to dissociation of liquid molecules with the formation of gas bubbles similar to cavitation ones [8]. When gas bubbles collapse, directed jets form when hydrodynamic perturbation from a certain side affects the bubble during collapse. The perturbation may come reflected from a wall or from another bubble rapidly expanding or collapsing nearby [7]. In a pulse-periodic mode with a sufficiently high pulse repetition rate, collapsing bubbles will be under the constant influence of hydrodynamic perturbations distributed along the axis of the laser beam or the filaments in case of self-focusing (filamentation) [2, 9].
Therefore, one should expect the preferred direction of the resulting jets parallel to the laser beam axis. As shown below, along with the flows directed parallel to the laser beam, transverse flows are also formed as a result of a collision of counter directed longitudinal streams. In the literature one can find other flow generation mechanisms in liquids under the effect of laser radiation reported. Excluding thermocapillary and electro-optical mechanisms, which are not related to the subject of the current paper, we can note the ponderomotive mechanism [16] and the mechanism of generation of ultrasonic waves by nanoparticles contained in a liquid under the action of laser radiation [17].

This work is devoted to the production and observation of quasi-stationary flows in liquids under the action of repetitively pulsed laser radiation and to the study of the most general properties of the resulting flows.

2. Experimental setup
In current experiments, Amplitude Systemes s-Pulse femtosecond laser was used. The laser was adjusted to generate fixed energy levels per pulse of 13, 135, and 250 μJ at fixed pulse repetition rates of 1.43, 1.67, 3.33, 5, and 10 kHz with pulse duration less than 450 fs. The original radiation wavelength is \( \lambda \approx 1.023 \, \mu m \), the beam diameter is 2.6 mm, and the beam propagation factor is \( M^2 < 1.3 \), which corresponds to an angular divergence of less than 0.69 mrad (full angle). The laser radiation was focused by biconvex quartz lenses with a ratio of the optical surface curvatures 1:4, providing minimal spherical aberrations, with 13.2 and 33.3 mm focal distance, respectively. The beam was focused with focal numbers \( f/5n \) and \( f/12.8n \), where \( n \) is refractive index of the liquid medium. The minimum beam diameters in the beam waist were, respectively, less than 10 μm for the first case and about 20 μm for the second.

![Figure 1](image1.png)

**Figure 1.** Laser beam focusing schemes in rectangular quartz cuvettes with liquid. The radiation falls from left to right, the initial beam diameter is 2.6 mm. 1 - focusing lens, 2 – cuvette with a liquid, 3 - power meter beam stop. a) lens with a focal length \( f = 33.3 \, \text{mm} \) (\( f/12.8n \), \( n \) is the refractive index of the liquid); b), c) lens with a focal length \( f = 13.2 \, \text{mm} \) (\( f/5n \)); c) scheme with astigmatism induced - the lens is shifted 0.7 mm down and tilted 1° clockwise, the initial lens beam axis positions are shown in dashed lines. The beam paths were calculated by paraxial ray tracing method for \( n = 1.325 \) (water).
Table 1. Properties of nonlinear optical media under the effect of pulsed laser radiation of a pulse length $\tau = 450$ fs and pulse power up to 500 MW ($\lambda = 1.023$ μm; focusing lenses $f = 13.2, 33.3$ mm; beam propagation factor $M^2 = 1.3$, beam diameter $d = 2.6$ mm).

| Nonlinear medium   | Refraction index, $n_0$ ([14]) | Absorption coefficient, $k$ (cm$^{-1}$) | Nonlinear refraction index, $n_2$, $10^{-20}$ m$^2$/W | Critical power of self-focusing $P_{cr}$, MW [10] | Non-linear focus displacement at $P_n = 0.5 \times 10^9$ W, mm (in brackets ($P/P_{cr}$), $P = P_{cr} - losses$) | Nonlinear dispersion index, $\kappa_2$, fs$^2$/mm [15] |
|--------------------|--------------------------------|------------------------------------------|----------------------------------------------------------|-------------------------------------------------|---------------------------------------------------------------------------------|---------------------------------------------|
| Water              | 1.325                          | 0.264                                    | 3                                                        | 4.2                                             | 0.45 (79)                                                                        | 2.3 (55)                                    | 14.8                                       |
| Heavy water        | 1.322                          | 0.016                                    | 3.3                                                      | 3.8                                             | 0.54 (115)                                                                       | 3.1 (112)                                   | ---                                        |
| Ethanol            | 1.355                          | 0.153                                    | 4                                                        | 3                                               | 0.58 (124)                                                                       | 3.1 (99)                                    | ---                                        |
| Isopropanol        | 1.377                          | 0.116                                    | 4                                                        | 3                                               | 0.62 (126)                                                                       | 3.3 (109)                                   | ---                                        |
| Acetone            | 1.353                          | 0.033                                    | 6                                                        | 2.1                                             | 0.75 (210)                                                                       | 4.2 (200)                                   | ---                                        |
| Dichloromethane    | 1.420                          | 0.04                                     | 6.5                                                      | 1.8                                             | 0.88 (238)                                                                       | 4.8 (227)                                   | ---                                        |
| Chloroform         | 1.436                          | 0.0051                                   | 8                                                        | 1.5                                             | 1.0 (310)                                                                        | 5.6 (310)                                   | ---                                        |
| Tetrachloromethane | 1.451                          | 0.00055                                  | 6                                                        | 1.9                                             | 0.92 (240)                                                                       | 5.1 (240)                                   | 53.3                                       |
| Quartz glass       | 1.45                           | -                                        | 3                                                        | 3.8                                             | -                                                                              | -                                          | -                                          |
The laser beam focusing schemes are shown in fig. 1. The beam was fed into the cuvette with liquid through a flat optical quality quartz wall of 3 mm thick. At the exit from the cuvette, the laser beam was absorbed in a calorimeter laser power meter. As it was done in previous experiments [5], when the laser beam was focused with artificial astigmatism, the center of \( f/5n \) lens (13.2 mm focal distance) in some experiments was set off the beam axis by 1-2 mm. The induced astigmatism was characterized by focal parameter of 1-2 mm, significantly exceeding the length of a beam waist. This possibility was important for interpretation of the results. The astigmatism that occurred when the lens was set off and tilted related to the beam axis could also be calculated using paraxial ray tracing method.

Studies were carried out in water, acetone, alcohols, chloromethanes. In all the mentioned liquids, with an exception of minor features related to the difference in the refractive indices and absorption, similar patterns of laser-induced flows were observed. In interpreting the results of observation of laser-induced flows, it was important to take into account the absorption of laser radiation when passing through a liquid, as well as the nonlinear optical phenomenon of self-focusing, caused by the dependence of the refractive index on the intensity of the laser radiation. The necessary optical characteristics of liquids are given in Table 1. Formulas from Chapter 7 of [10] were used to estimate critical power of self-focusing and non-linear focus displacement due to self-focusing.

![Figure 2](image)

**Figure 2.** Self-focusing effect in non-linear medium in focused laser beam (shown by dashed red line). 1 – laser beam boundary without self-focusing (blue solid line); 2 – focusing lens, 3 – nonlinear medium \( n(I) = n_0 + n_2I \).

In order to use formulas 7.1.4 - 7.1.7 from Chapter 7 of [10] to estimate the non-linear focus displacement effect \( z_{sf} \) as shown in fig. 2, it was necessary to take into account the absorption of radiation when propagating from the cuvette wall to the focal point. Data on the absorption coefficients of radiation with a wavelength \( \lambda = 1.023 \mu \text{m} \) in the studied liquids can be found in the reference literature, for example [1], as well as in the original works [14]. In order to take into account the possible influence of the features of the liquids used in the experiments, as well as the specifics of the used laser radiation source – femtosecond periodic-pulse laser, the authors of this paper have measured the absorption coefficients of unfocused radiation in the studied solvents. The absolute accuracy of measuring, estimated at 0.001 cm\(^{-1}\), did not allow to measure reliably just the absorption in tetrachloromethane, which is 0.00055 cm\(^{-1}\), according to [14]. The obtained data on the measurement of absorption coefficients in the studied solvents, as well as estimates of the critical self-focusing power and the amount of focus shift due to self-focusing, calculated according to formulas of [10] for two focusing schemes (with focusing parameters \( f/5n \) and \( f/12.8n \), where \( n \) is the refractive index of the liquid) are presented in Table 1. It should be noted that the data on the absorption of radiation in liquids obtained in this work are consistent with the data of [14], as well as other reference data on the spectral absorption coefficients of near infrared radiation in liquids, for example [1]. This means, in particular, that in the range of intensities under consideration, the mechanisms of absorption of radiation differ little from the mechanisms of absorption at low levels of intensity.
Also it can be seen from Table 1 that, with the exception of water and alcohols, the studied liquids are characterized by small absorption coefficients (less than 0.05 cm$^{-1}$) having minor effect on self-focusing process. So in estimations of the focus shift during self-focusing, in most cases only a small decrease in the radiation power due to absorption as it passes through a nonlinear medium to the self-focusing point was taken into account.

As follows from the data presented in Table 1, in current experiments the critical self-focusing power in the studied liquids is from 1.5 to 4 MW, the laser power exceeds non-linear critical power from 55 to 310 times. According to the estimates based on formulas from [10], summarized in Table 1, the maximum focus shift ($z_m - z_{sf}$) in an optical system with a lens $f = 13.2$ mm ($f/5n$) depending on the type of liquid is from 0.45 to 1 mm, and in a system with a lens $f = 33.3$ mm ($f/12.8n$) - from 2.3 to 5.1 mm. As shown below, actually observed values of the focus shift turned out to be approximately twice as large as those calculated by the formula from [10].

The relations from [10] used for estimations of non-linear focus shift are valid for continuous radiation. In the case of pulsed radiation, it is necessary to solve the nonlinear differential equation for the propagation of radiation in a nonlinear medium. However, for estimates in this case as well, the expressions 7.1.4 - 7.1.7 from Chapter 7 of [10] can be used if one takes into account the dependence of the nonlinear addition of the refractive index $n_2$ on the pulse duration. Systematized data on the dependence of $n_2$ on the pulse duration for the studied liquids can be found, for example, in [12, 13]. The discrepancy between our estimates and experimental results mentioned in the above paragraph can be partially explained by this circumstance.

The last column in the Table 1 represents the coefficient of nonlinear dispersion $k_2$, which characterizes the increase in the pulse duration when passing through a nonlinear optical medium. The parameter $\tau^2/k_2$, having a dimension of length, gives an estimate for the characteristic dispersion length during the pass through a nonlinear medium, on which the pulse duration $\tau$ is doubled [10]. For the media under consideration, $k_2$ lies in the range 14-55 fs$^2$/mm, that is, for $\tau = 450$ fs and $d < 50$ mm, $\tau^2/k_2 >> d$ and the change in the pulse parameters due to dispersion can be neglected.

The resulting flows of the liquids were observed by schlieren methods. The cuvettes were illuminated by a beam collimated from point-like laser-plasma broadband radiation source LPS-50 (similar to that described in [5]). Another possibility of observing flows was due to the presence of gas bubbles in the liquid ejected from zones of increased laser beam intensity in the beam waist, where gas bubbles were formed due to multiphoton dissociation. The bubbles were illuminated by the beam of an auxiliary laser with a wavelength of 532 nm, deployed with a cylindrical lens in a line and focused by a spherical lens into a “light sheet”, with maximum intensity set in the region of interaction between the laser beam and a liquid. Schlieren images and patterns of moving bubble traces were recorded by video framing. The mixing of calibrated microparticles into the liquid to visualize the flow in this formulation was excluded, since the particles would affect the propagation, scattering, and absorption of the radiation of a femtosecond laser and the mechanisms of opto-hydrodynamic phenomena [17].

3. Results of the experiments

3.1. Axial and transverse flows under weak self-focusing

In experiments with a minimum pulse energy (taking into account losses in the optical path $E_p \approx 10$ мкДж, $P \leq 20P_{cr}$) at pulse repetition rates of 1.67 kHz and 5 kHz, a quasi-stationary flow of liquid was observed, flowing out of the focused beam waist region in both directions along the laser beam axis, as well as in the direction perpendicular to the laser beam (fig. 3). The flux along the laser beam was observed when the laser beam was focused exactly on the optical axis of the lens without astigmatism (fig. 3, a, c). To introduce artificial astigmatism, the center of the lens with a focal length $f = 13.2$ mm (focusing number $f/5n$ in a medium with a refractive index $n$) was shifted 0.7-1 mm transversely to the beam axis, while the lens was tilted by an angle of 1.5-2.2 degrees.

According to calculations by the ray tracing method, the focal beam waist was split into two astigmatic foci located at a distance of $a = 0.5$-1 mm from each other along the beam axis. With a
characteristic waist length < 0.25 mm, such distance meant that the foci were clearly separated. In this case, the flow along the optical axis weakened, and the appearance of a flow in the perpendicular direction was observed. When analyzing video frames, it was seen that weakened axial flows was generated in each of the astigmatic foci, and the transverse flow effect arises as a result of a collision of contra-directed axial flows from different foci. When the lens returned to the fine-tuning position, the transverse flow has ceased, and the flow along the axis has intensified. The observed velocities of the gas bubbles motion in the middle section of the jet in the fine-tuning position were more than ten centimeters per second, which indicated a possible velocity in the minimum beam cross section at the focus of a meter per second or more. The velocity was determined by the length of the tracks from the moving bubbles as in fig. 3, a) captured at the frames during the exposure time.

![Figure 3](image.png)

**Figure 3.** Flow pattern in the case of weak self-focusing with precise focusing a), c) and with artificial astigmatism introduced b), d). The flows are visualized using light sheet lit gas bubbles a), b) and by schlieren technique c), d). Focusing number $f/(5n)$, where $n$ – refraction index of the liquid; beam boundaries are shown with lines. Laser pulse energy $E \leq 20 \, \mu J \, (P \leq 10P_{cr})$, pulse repetition rate $v_r = 5 \, kHz$. Laser radiation falls from left to right. Liquids: a),b) water, $n = 1.325$, c), d) tetrachloromethane, $n = 1.45$. Frame size a), b) 3.8×2.1 mm$^2$, c), d) 19.6×11 mm$^2$. The position and size of focusing lens are shown schematically.

3.2. Axial and transverse flows with dominant effect of self-focusing

In the case when the pulse energy in the beam waist taking into account losses in the optical system was 100 $\mu J$ or more ($P \geq 100P_{cr}$), a more complicated flow pattern was observed. Self-focusing effect (Kerr lens) was manifested itself in a liquid at sufficiently high radiation intensity, thus before reaching the focal point the beam was collapsed in a filament. In the waist region, the amplified field of the light wave was interacted with the electrons of the liquid molecules, causing dissociation and at the same time defocusing the beam. This process was repeated several times until the beam stopped self-focusing by losing power due to absorption.

As a result, several constrictions are formed, in each of which the fluid moves along the axis of the beam, and between them the longitudinal flows collide and spread in the transverse direction. Fig. 4 shows the dynamics of the formation of the flow pattern after switching on the radiation with a pulse repetition rate of 1.67 kHz, as well as the steady-state picture of longitudinal and transverse flows at a
repetition rate of 5 kHz. The pulse energy in both cases is 200 μJ (taking into account losses in the radiation supply and focusing system). A feature of the regime with a high energy in the pulse and a high repetition rate is the heating of the liquid along the path of the beam due to the usual absorption, causing a local change in dielectric coefficients with heating. Therefore, the position of the filaments formed due to self-focusing changes: they are maximally stretched along the axis of the beam at the initial moment, when the heating is small, and approach as they heat up, as a result of which it becomes difficult to visually separate them when observing the flows. Accordingly, in the steady state, the flow pattern looks like two longitudinal streams moving along the beam channel towards each other and spreading to the sides at the collision site. The heat released due to the absorption of laser radiation, mainly in the region of constrictions, is transferred by these flows to the place of their collision, where, after the liquid spreads to the sides, thermo gravitational convection plume is formed, which carries the released heat into the upper layers of the liquid.

![Figure 4](image)

**Figure 4.** The formation of longitudinal and transverse flows in tetrachloromethane under the action of the focused beam of repetitively pulsed femtosecond laser in the presence of several constrictions along the filament resulting from self-focusing. Time marks - from moment of the laser beam turning on. Pulse energy $E_{pf} \approx 200$ μJ, focusing number $f/(12.8n)$, $n = 1.45$, the lines show the beam boundary in the absence of self-focusing. The radiation falls from left to right. Frame size $19.6 \times 11$ mm².
In the steady-state picture of flows with lower pulse repetition rate and lower energy release, it is still possible to separate several points from which the transverse streams originate. At a higher repetition rates separate transverse streams merge into a single convective plume, the flow in the transverse stream region is strongly turbulent and the individual spreading points become so close that they can no longer be distinguished.

4. Summary and conclusion
When interacting with liquids, focused radiation from a repetitively pulsed femtosecond laser at a radiation power of \( P \approx (10-20)P_{cr} \), where \( P_{cr} \) is the critical self-focusing power of laser radiation in the liquid as non-linear medium, generates quasistationary streams in the focused beam waist region directed along the laser beam axis in both directions from the waist.

In the case when there are several beam waist or several constrictions in the beam waist region, for example, when focusing with astigmatism (2 constrictions) or at \( P \geq 100P_{cr} \) when the filament is formed due to self-focusing (filamentation) of powerful pulsed radiation with the appearance of several focusing-defocusing regions, one can observe the streams along the beam axis together with the flows spreading around transversely to the beam. In this case, counter-propagating axial flows generated in the constrictions with a high intensity of laser radiation separated by gaps of a defocused beam collide in these gaps and spread to the sides, forming a flow transverse to the laser beam. At higher pulse repetition rate and significant heat release in the self-focusing region, axial flows transfer the released heat along the beam to the points of transverse flow spreading, from which a combined thermo gravitational convection stream is formed, which accumulates heat released in the self-focusing zone.

In the experiments, the possibility was realized of changing the shape and arrangement of energy release zones under laser irradiation in a liquid using an adjustable degree of astigmatism by changing the position of the focusing lens relative to the axis of the laser beam. It was shown that convective flows in the focal waist region, observed by schlieren technique or by the method of light sheet lit gas bubbles (deploying visible radiation from an auxiliary laser focused in a sheet), could be directed strictly along the laser beam axis and/or in the transverse direction, depending on the degree of the astigmatism introduced. With a significant excess of the laser radiation power over the critical one for self-focusing, when the length of the filament formed due to self-focusing exceeded 1 mm, the flows along the laser beam axis were observed, directed from the ends of the filament to the center, as well as in the direction transverse to the laser beam. Transverse flows were formed as a result of a collision of counter directed axial flows.

Since the general picture of the flows in all the studied liquids was similar, the authors of this work consider the dynamic mechanism of flow formation as a result of the collapse of photodissociation gas bubbles to be the most probable mechanism responsible for the formation of the observed flows. The mechanism of ponderomotive convection, described in detail in [16] for the case of the action of a continuous laser on a liquid, or convection as a result of the generation of ultrasonic waves by means of nanoparticles that are in plasmon resonance with incident laser radiation [17], does not explain all the phenomena observed in this study.

The results of the studies can serve as the basis for application in laser technologies for processing or synthesis of materials using a liquid medium. Further studies are required on specific mechanisms and applications of the phenomena observed.

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