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FILAMENTATION IN THE INTERSECTION REGION OF TWO FEMTOSECOND LASER BEAMS IN SAPPHIRE

The filamentation phenomenon arising at the intersection of two femtosecond laser beams in a sapphire single crystal has been studied. Conditions for a regular multifilament structure (MFS) to emerge with the parameters depending on the pulse energy, intersection angle, and phase difference between two exciting beams are determined. For the first time, the formation of a single filament under the action of two different excitation beams is analyzed as an MFS implementation. The number of filaments in the MFS is demonstrated to depend on the number of interference maxima in the intersection region of beams, the power of which exceeds the critical power of self-focusing. Attention is paid to the possibility to control the multifilament structure by varying the phase difference between the interacting beams. Optical manifestations of the interaction between the filaments with the "attractive" or "repulsive" character are observed. The spectrum of the axial emission by a single filament, as well as its dependence on the filament length, is studied, and the process of four-wave mixing is shown to play a key role in its formation.

Keywords: femtosecond laser pulses, Kerr effect, filaments, sapphire, focusing, axial emission.

1. Introduction

A significant progress has been reached recently in the technology allowing various optoelectronic materials to be modified with the use of ultrashort laser pulses. It was demonstrated that strongly focused laser pulses – in particular, in the filamentation mode – are especially useful while creating the wave guides in the material volume, optical splitters and gratings, and optical memory cells [1, 2]. Therefore, the research of peculiarities in the propagation of femtosecond laser pulses in such materials, including the filamentation effects, is important not only from the viewpoint of fundamental science, but also for probable practical applications.

If the laser pulse power exceeds a threshold value, the self-focusing of the beam dominates over its diffraction-driven divergence. As a result, the beam gets filamented [2], and the regime of pulse propagation substantially changes. If the threshold power is exceeded by several times, the modulation instability gives rise to the emergence of plenty of filaments, with the interaction and the energy exchange between them being possible. Therefore, the study of the interaction between filaments is important to understand and to precisely control the multifilamentation phenomenon, in particular, in order to modify optoelectronic materials with the help of femtosecond laser pulses.

Some experimental and theoretical works were devoted to the consideration of interaction between filaments in air [3–10] and transparent solids [11]. The authors of work [9] studied the formation of periodic modulations of the density of a plasma, whose dimensions are comparable with the wavelength and which are capable of transporting the input pulses, similarly to a photonic crystal, at intersection angles from 2 to 16°. The authors of work [10] simulated the interaction between femtosecond filaments at intersection angles within the interval $0.01 \div 0.1°$. They showed that the interaction brings about the attraction between co-phased filaments; on the contrary, if the filament phases are opposite, the repulsion dominates. It was predicted that the merging of two co-phased filaments should result in the formation of a single stable filament. As far as we know, there is no experimental confirmation of this theoretical conclusion till now, in particular, in the case of solid transparent insulators. In work [11], a regular MFS was formed in a quartz glass pumped with two intersecting beams, and every filament generated its own supercontinuum at that. The authors of work [11] confined the scope of their research to the mutual coherence of multiple supercontinuum sources, but the influence of the phase difference between excitation beams on the MFS was not considered.

This work is aimed at studying the interaction between two filaments intersecting in a single-crystalline sapphire at angles ranging from 1.2 to 5° and with the excitation-beam phase difference varying from 0 to 180°. The spatial period of the interference pattern at such intersection angles is smaller than the diameter of a pump beam in the intersection region. Therefore, depending on the energy and the phase in this region, either a single filament or an MFS can be formed.

2. Experiment

The processes of filamentation and interaction between filaments were visualized by registering the time-integrated sapphire fluorescence emitted from the plasma channel in the direction perpendicular to the pulse propagation axis. The spectra of axial emission by filaments were also registered. The experimental installation is schematically shown in Fig. 1.

A regenerative amplifier 1 generated a train of pulses (2.5 mJ, 150 fs, 818.5 nm) with a repetition frequency of 1 kHz. The set of neutral filters 2 provided a required pulse energy. Diaphragm 4 cuts out two narrower parallel beams, 1 or 1.5 mm in diameter, the axes of which are separated by a distance of 2.4 or 3 mm. They are directed into objective lens 6 with a focal distance of 43 or 65 mm. Two identical plates of fused quartz 1 mm in thickness are inserted in the beam optical path before diaphragm 4. The slope of one plate is varied to provide a required phase shift between the two beams. Lens 6 is mounted on a motorized one-coordinate stage that could move along the axis Z. The distance between diaphragm 4 and lens 6 equals 10 cm, when the coordinate stage is

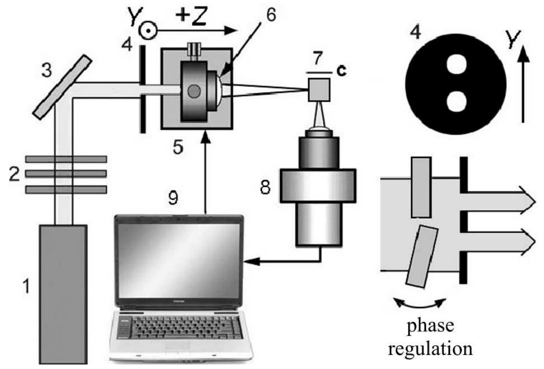

Fig. 1. Schematic diagram of the experimental installation: regenerative amplifier Legend F-1K-HE (*1*), neutral filters (*2*), rotary mirror (*3*), diaphragm (*4*), coordinate stage (*5*), objective lens (*6*), specimen (*7*), microscope objective and CCD-chamber (*8*), and personal computer (*9*)

located at $Z = 0$. Polished specimen 7 of a single-crystalline sapphire $3 \times 3 \times 20$ mm^3 in dimensions is mounted behind the lens in such a way that, at $Z = 0$, the lens focus is in the specimen, and the crystal axis of the specimen is oriented in parallel to the axis Z. Objective lens 8 (×3.7 or ×10) exposes the filament interaction region on a 1200×1600 monochrome CCD-matrix in chamber 8 with a pixel size of 4.4×4.4 μm^2 on a scale of 1.08 or 0.4 μm/pixel with a spatial resolution of 3 or 2 μm, respectively.

In another experiment, the spectra of axial filament emission are measured. For this purpose, the light of axial emission, with the help of an optical fiber waveguide fixed at a distance of 15 cm from the back specimen face, is directed into the entrance slit of an SP-2500i spectrograph with a focal length of 500 mm and equipped with a SPEC-10 CCD-detector.

As an experimental material, we select a single-crystalline sapphire (the energy gap width was 9.9 eV, the refractive index $n_0 = 1.76$ at a wavelength of 0.8 μm, and the nonlinear refractive index $n_2 = 2.5\times$ $\times10^{-16}$ cm^2/W). Sapphire specimens $3\times3\times20$ mm^3 in dimensions were fabricated, in which the crystal axis is oriented along the shorter side.

3. Results and Discussion

Figure 2 illustrates the interaction between two coherent femtosecond beams that intersect each other at the angle $2\alpha = 3.2°$. With the help of CCD-chamber 8, we registered the luminescence light of sapphire emitted owing to the relaxation of free

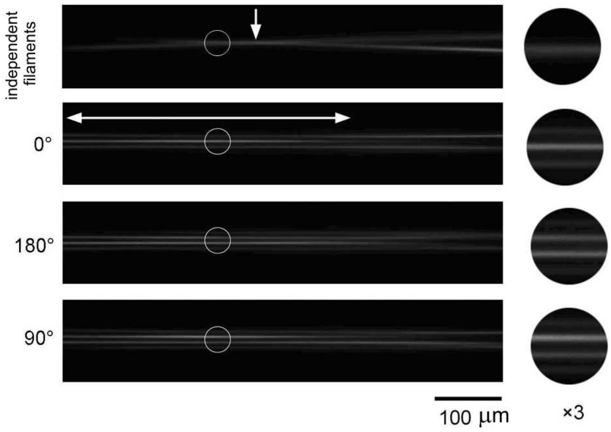

Fig. 2. MFSs formed by two coherent femtosecond beams intersecting at the angle $2\alpha = 3.2°$ in sapphire for various phase differences between them. The corresponding phase difference is indicated to the left of each fragment. Enlarged views of multifilament structure fragments are shown to the right

charge carriers in the filament plasma channel. The excitation beams were directed from left to right. The energy of every excitation pulse equaled 2.0 μJ. The upper fragment in Fig. 2 demonstrates the intersection between two non-coherent and non-time-synchronized filaments. This fragment was obtained when one of the phase compensating plates was removed. A time difference of about 2.5 ps between two pulses that emerged in this case was much longer than the pulse duration, so that the interaction of pulses according to the Kerr mechanism was excluded. On the other hand, a small energy disbalance because of the plate absence explains the difference between the intensities of two filaments. The intersection point of two independent filaments (it is marked by a white arrow in Fig. 2) lays in the focal plane of lens 5. In this case, the filamentation began before the geometrical focus.

The next fragment (marked as 0°) demonstrates a scenario of the interaction between two time-synchronized pulses at the zero phase difference between them. In the intersection region (it is indicated by a white arrow), an MFS emerged in the form of three equidistant parallel filaments. Attention should be paid to that, if the pump pulses were synchronized in time, the MFS began much closer to the specimen entrance face in comparison with the case of independent filaments. This fact testifies that both pump pulses were engaged in the MFS formation process. Proceeding also from the fact that the MFS length (about 400 μm) considerably exceeded the length of the region where the filament kernels intersected, it becomes clear that a wider energy reservoir of each filament, where the energy intensity was low, participated in the MFS formation. The filaments in the MFS were located at the distance $d = 8$ μm, with the most intensive central filament being symmetrically arranged along the bisectrix of two non-interacting filaments. The positions of the filaments and the distance between them in the MFS corresponded to the calculated positions of maxima in the interference pattern formed by two co-phased pulses with a wavelength of 820 nm and an intersection angle of 3.2° in sapphire. A phase difference of 180° between two filaments (see the next fragment below) gave rise to a vertical shift of MFS by $d/2$, in accordance with a shift of the interference pattern maximum. In this case, there emerged a symmetric MFS consisting of two identical central filaments with a high intensity and two weak peripheral filaments; the distance between filaments equaled 8 μm. If the phase shift was 90°, an asymmetric structure of four filaments was formed (see the lower fragment). The filament positions and the intervals between them in the interference patterns for two beams with a phase shift of 90°, as well as 0 and 180°, corresponded to the calculated positions of maxima.

The results presented above allow us to draw conclusion that two coherent filaments, first, disappeared in the intersection region and transferred their energy to the energy reservoirs. Instead, there emerged an MFS in the intersection region. The number, intensity, and position of MFS were determined by the interference pattern of two reservoirs. A total MFS width of 24 μm qualitatively estimates the diameter D of the filament energy reservoir.

In the transient region between the filaments and the MFS, the effects of filament attraction and repulsion manifested themselves. In essence, this transient region was an area where the interference pattern contrast was reduced because the energy reservoirs intersected each other there only partially. A superposition of low-contrast interference strips and own gradients of the intensity of the light field created by excitation pulses shifted the local intensity maxima with respect to perfect strip positions and, as a consequence, the filament positions themselves. To make this interaction more evident, we diminished the pulse energy and the intersection angle values.

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Figure 3 demonstrates the MFS zone of two filaments and the surrounding regions located along the axis Z. The energy of each pulse was 1.5 μJ and the intersection angle equaled $2\alpha = 1.2°$. Pump pulses propagated from left to right. At this pulse energy, the filamentation of the pulses shifted in time began already behind the geometrical focus. In the MFS region, only a single filament was formed if the phase difference was 0°, and two parallel filaments located at a distance of 20 μm if the phase difference was 180°. The right edge of the photo coincides with the specimen exit face. As was in the previous case, the position and the distance between MFS filaments corresponded to the calculated positions of maxima in the interference pattern. The lower fragment, which was obtained at a phase difference of 180°, exhibits a continuous transformation from an MFS (two filaments separated by $d = 20$ μm) to two independent diverging filaments. Attention should be paid to that the distances between the filaments at the exit face (their values are indicated to the right of each fragment) are different in all three cases. Co-phased filaments attract, whereas counter-phased ones, on the contrary, repulse each other. However, the situation can differ at different positions along the axis Z.

In Fig. 4, the dependences of the distance between filaments on the coordinate Z are shown for a phase difference of 180° and, for comparison, for independent filaments shifted in time (according to the data of Fig. 3). One can see that the attraction dominating in the interval $Z = 800 \div 1500$ μm transforms into the repulsion in the interval $Z = 1400 \div 1700$ μm. Note that the authors of work [10] predicted only the repulsion of two filaments with opposite phases that intersect each other in air at the angle $\alpha = 0.01°$. We think that the physical reason for this discrepancy consists in the ratio $r = D/d$ between the energy reservoir diameter and the distance between the fringes in the interference pattern. In air and provided $\alpha = 0.01°$, this ratio equals $r \approx 0.5$ if $D = 1$ mm. However, in sapphire and for $\alpha = 0.6°$ and $D = 24$ μm, we obtain $r \approx 1$. This means that a destructive interference between two pulses dominates in air, even if they intersect weakly, so that only a filament repulsion is observed. Unlike that, the interval between the strips in sapphire equals D. Hence, besides the central minimum, there emerge constructive maxima located at a distance of 20 μm from each other. As the coordinate Z increases, the own pulse maxima diverge, which gives rise to a shift of the resulting maxima owing to the superposition of low-contrast interference strips and the own distribution of the light field intensity of pump pulses; firstly toward the center and afterward away from it. Just this circumstance results in that both the filament attraction and repulsion are observed.

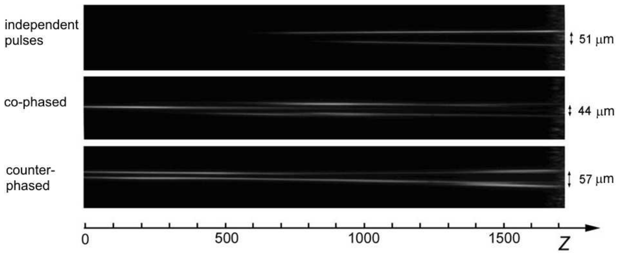

Fig. 3. Interaction of two filaments intersecting at the angle $2\alpha = 3.2°$ for various phase differences

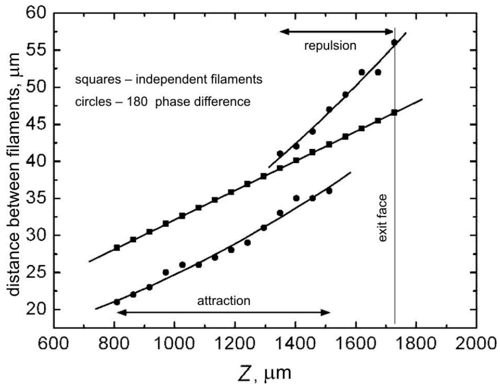

Fig. 4. Dependences of the distance between filaments on the coordinate Z

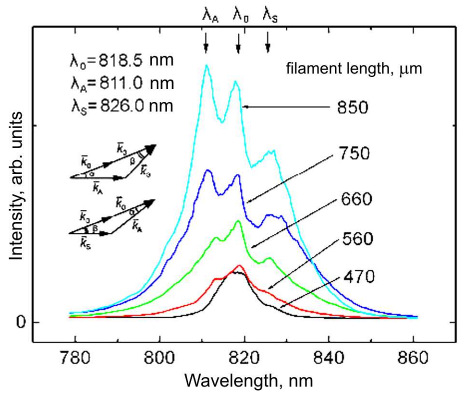

Fig. 5. Spectra of axial emission by a single filament with various lengths. $E_{\text{pulse}} = 2.4$ μJ

In Fig. 5, the spectra of axial emission are shown for various lengths of a single filament excited by two beams with a total energy of 2.4 μJ that intersect at an angle of 1.5°. One can see that, starting from a single filament 560 μm in length, new Stokes and anti-Stokes peaks appear at the wavelengths λ_S = 826.0 nm and λ_A = 811.0 nm, respectively, which are equidistant from the excitation wavelength λ_0 = 818.5 nm. We suppose that those peaks are generated in the course of the four-wave mixing in the high-intensity filament kernel, in which the recombination of two laser photons creates a Stokes photon and an anti-Stokes one (see the inset in Fig. 5). The dispersion of the refractive index in sapphire allows the momentum and energy conservation laws in this process to be obeyed,

$$2\mathbf{k}_0 = \mathbf{k}_S + \mathbf{k}_A, \quad (1)$$

$$2h\omega_0 = h\omega_S + h\omega_A. \quad (2)$$

Really, using the tabulated data for the refractive index of sapphire and the measured wavelengths λ_S, λ_A, and λ_0, the solution of the system of equations (1), (2) gives $\alpha = 0.72°$ and $\beta = 0.7°$. The values obtained are in good agreement with an experimental value of 0.75° for the angle between the pump beam and the axis Z.

4. Conclusions

In this work, the interaction of two femtosecond filaments in single-crystalline sapphire is studied experimentally at various pulse energies, intersection angles, and phase differences. It is shown that both a regular multifilament structure (MFS) and a single filament can be formed in the intersection region of energy reservoirs. A possibility to control the MFS by varying the phase shift between the interacting beams is demonstrated. Owing to the superposition of low-contrast interference strips in the own distribution of the light field intensity of exciting beams, both the attraction and the repulsion of filaments are observed in the near-MFS region. The axial emission of a single filament formed in the zone, where the energy reservoirs overlap, is a result of the nondegenerate four-wave mixing.

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ФІЛАМЕНТАЦІЯ В ОБЛАСТІ
ПЕРЕТИНУ ДВОХ ФЕМТОСЕКУНДНИХ
ЛАЗЕРНИХ ПРОМЕНІВ У САПФІРІ

Р е з ю м е

Досліджено явище філаментації при перетині двох фемтосекундних лазерних променів у монокристалічному сапфірі. Визначено умови утворення регулярної мультифіламентної структури (МФС), параметри якої залежать від енергії імпульсу, кута перетину і різниці фаз між двома збуджуючими променями. Вперше, як окрему реалізацію МФС, досліджено процес формування одинарних філаментів при дії двох різних променів збудження. Продемонстровано, що число філаментів в МФС залежить від кількості інтерференційних максимумів в області перетину променів, потужність яких перевищує критичну потужність самофокусування. Звернуто увагу на можливість керування мультифіламентною структурою шляхом зміни різниці фаз між взаємодіючими променями. Спостережено оптичні прояви "притягувального" і "відштовхувального" характеру взаємодії філаментів. Вивчено спектр осьового випромінювання одинарного філамента залежно від його довжини. Показано, що у формуванні осьового спектра його випромінювання визначальну роль відіграє процес чотирихвильового змішування.