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"WHITE SUPERCONTINUUM" AND "CONICAL EMISSION" OF FEMTOSECOND FILAMENTS IN BIREFRINGENT MEDIA

The observation of the polarization features of the "white supercontinuum" type and the "conical emission" of femtosecond laser filaments in quartz and sapphire is reported. The features are caused by the positive and negative birefringences of quartz and sapphire. The polarization directions of "white supercontinuum" and "conical emission" are mutually normal as a result of the difference between the group velocities of the ordinary and extraordinary rays. A physical mechanism of conical emission, which explains the features of its polarization, has been proposed.

Keywords: filamentation, femtosecond filament, conical emission, group velocity, birefringence.

1. Introduction

The propagation of powerful femtosecond laser pulses in transparent media is accompanied by a series of interesting, still little-studied phenomena such as filamentation, generation of conical waves, terahertz radiation emission, and femtosecond radiation emission with a "quasiwhite" spectrum [1]. Those phenomena are a matter of investigation for non-stationary nonlinear optics. For today, they form a forefront of researches in laser physics. Laser femtosecond filaments emerge as a result of the dynamical balance between the focusing of laser radiation due to the positive variation of the medium refractive index (the high-frequency Kerr effect) and its defocusing due to a negative variation of the refractive index (owing to the generation of an electron-hole plasma) [1]. The filamentation of laser light pulses favors the transformation of their frequency and angular spectra. This transformation manifests itself in the appearance of a "white supercontinuum" (SC) and the generation of "conical emission" (CE), respectively [1].

Earlier, the structure of filaments, conditions of their formation, and SC and CE properties were mainly considered for isotropic Kerr media [1–3]. The number of researches concerning femtosecond filaments in birefringent media is rather small [4–10]. However, the knowledge of the processes of formation and the properties of filaments at the propagation of ultrashort light pulses in such media – in particular, in sapphire – is of importance, since this material is used as an active medium in modern laser systems. Similar researches can provide information about such slightly studied phenomena that accompany the filamentation as, e.g., the CE generation. A periodic filamentation phenomenon, which was experimentally observed in sapphire and crystalline quartz [11], became another new effect revealed in the recent years. The physical origin of the observed periodicity consists in the cyclic transformation of the polarization state of a light pulse at its propagation in a birefringent medium owing to the incursion of the phase difference between the ordinary and extraordinary rays.

In this work, the unusual polarization properties of SC and CE generated by filaments in birefringent crystalline media – positive in quartz ($n_o < n_e$) and negative in sapphire ($n_o > n_e$) – are reported for the first time. A qualitative explanation of those properties is also proposed.

2. Experimental Part

The block diagram of the installation created for our researches is depicted in Fig. 1. In order to generate filaments in polished single-crystalline (Al_2O_3 or SiO_2) specimens 5 mm in thickness, we used hori-

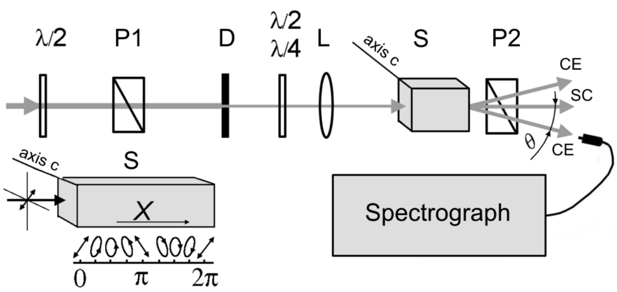

Fig. 1. Block diagram of the experimental installation

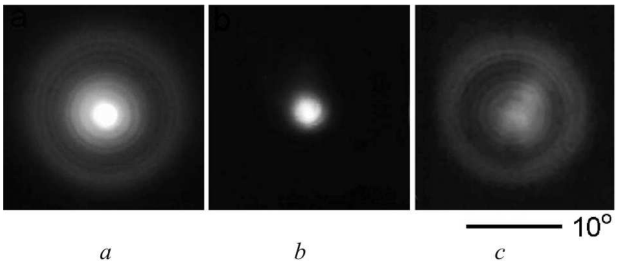

Fig. 2. SC and CE in a sapphire crystal in the far field: without polarizer P2 (*a*); polarizer P2 is oriented to transmit light with vertical (o-) polarization (*b*); polarizer P2 is oriented to transmit light with horizontal (e-) polarization (*c*)

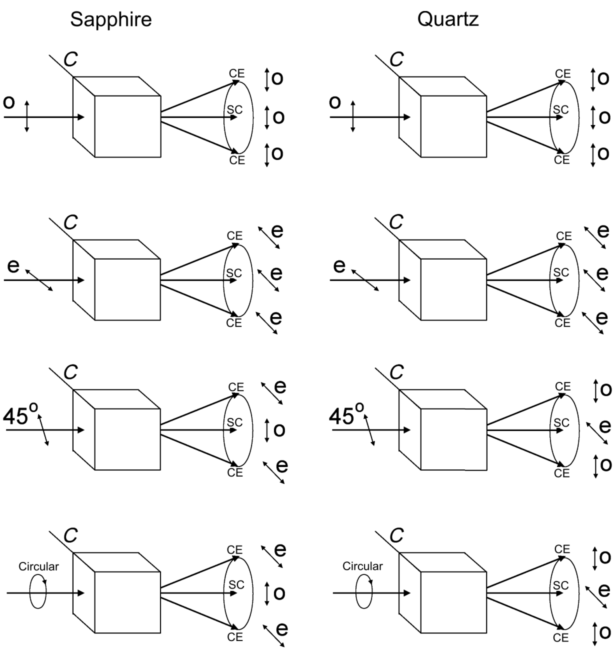

Fig. 3. CE and SC polarization states in sapphire and crystalline quartz depending on the input ray polarization

zontally polarized laser pulses with a wavelength of 800 nm, a duration of 150 fs, and a repetition frequency of 1 kHz. A half-wave phase plate ($\lambda/2$) and a Glan prism (P1) created a vertically polarized ray with controlled power. The ray passed through diaphragm D with a diameter of 3.0 mm. The orientation of the linear beam polarization was changed with the help of a $\lambda/2$-phase plate or transformed from linear into circular using a $\lambda/4$-phase plate. For the excitation of filaments, the ray was focused into specimen S with the help of lens L with a focal length of 8 cm. The crystallographic axis c was directed horizontally and complanarly to the input plane of specimen S, as is shown in Fig. 1. For probing the SC and the CE under various angles θ with respect to the axis of the input ray propagation, a flexible optical fiber 1 mm in diameter was used. A signal obtained at the optical fiber output was directed to the input slit of an SP-2500i spectrograph with a matrix silicon detector. The second Glan prism (P2) was used to analyze the polarization of SC and CE.

3. Experimental Results and their Discussion

In Fig. 2, the SC and CE registered at a distance of 20 cm from the output face of a sapphire specimen are shown. The laser beam was circularly polarized, and the pulse energy amounted to 1.5 μJ. The start point of the filament was located at a distance of 2.5 mm from the input face of the specimen. One can see that the ordinary (o-) polarization prevailed in the light of white SC (Fig. 2, *b*), whereas the visible CE and the weak red component of SC had the extraordinary (e-) polarization (Fig. 2, *c*).

The results of the analysis of the SC and CE polarizations in sapphire and quartz, and their dependence on the excitation ray polarization are exhibited in Fig. 3. In the case where the excitation ray has the o- or e-polarization, the SC and CE – both in sapphire and in crystalline quartz – have the same polarization as the excitation does, analogously to the case of isotropic media [12–15]. It is quite evident, because the polarization of a laser pulse remains invariable at its propagation in this case. On the other hand, if the excitation ray has a circular polarization or its polarization plane is directed at an angle of 45° with respect to the specimen crystallographic axis, the SC and CE polarizations are different. Actually, those two polarizations of the excitation ray are equivalent, because both in the case of circular polarization and when the polarization plane slope equals 45°, the polarization of the excitation pulse undergoes the same evolution cycle in the birefringent medium, as is illustrated in the inset in Fig. 1. The same cycle repeats with a period of 101 μm in sapphire and 90 μm in quartz: as soon as the phase incursion between the ordinary and extraordinary rays becomes equal to $2\pi n$, where n is an integer number [11]. Therefore, in what follows, we will discuss only the circular input polarization.

Attention is attracted by the fact that the CE (SC) polarization in the quartz crystal with the positive birefringence is orthogonal to the SC (CE) polarization in sapphire, where the birefringence is negative.

Besides the visible spectral interval, we have also analyzed the polarization of the infra-red (IR) component of SC in sapphire (see Fig. 4). The black curve exhibits the IR spectrum of SC for the e-polarized excitation, and the gray curve for the o-polarized one. The spectral interval of measurements was confined from the long-wave side by the sensitivity of a silicon detector, and from the short-wave one by the flare produced by the exciting radiation band. One can see from Fig. 4 that the IR component of SC is mainly e-polarized, whereas its visible component is o-polarized.

The experimental data presented above can be explained by a reduction in the spatial overlapping of the ordinary and extraordinary rays at their propagation in the birefringent medium. Figure 5 demonstrates the results of calculations that show the divergence of o- and e-pulses in sapphire due to the difference between their group velocities. The indication begins from the point of coincidence of those pulses in time and space, which corresponds to the input face of the specimen. The pulse duration equaled 150 fs, and the wavelength 800 nm. The value of refractive index was taken from the website http://refractiveindex.info.

From Fig. 5, one can see that, after passing a distance of 3 mm in sapphire, the pulses of ordinary and extraordinary rays so diverge that the periodic change of the polarization state occurs mainly in the central part of the resulting pulse, where the o- and e-components overlap. However, since $n_\mathrm{o} > n_\mathrm{e}$ in sapphire, the leading edge of the resulting pulse is mainly e-polarized, and the trailing one is o-polarized. This circumstance explains the polarization properties of

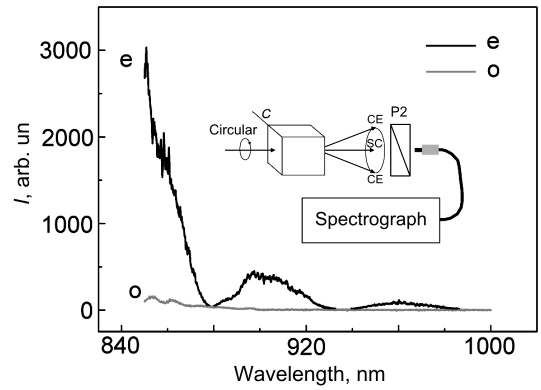

Fig. 4. IR spectrum of SC in sapphire for ordinary and extraordinary polarizations. The excitation ray is circularly polarized

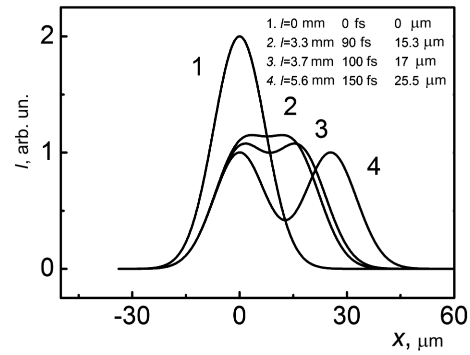

Fig. 5. Total intensity of Gaussian o- and e-pulses (a pulse duration of 150 fs) in a coordinate system that moves with the group velocity of an ordinary polarized pulse at various distances from the input face of the sapphire specimen, $l = 0$ (*1*), 3.3 (*2*), 3.7 (*3*), and 5.6 mm (*4*). The spatial and temporal differences between the o- and e-pulses are indicated for each l-value

SC, when excitation is performed by circularly polarized pulses. Really, as follows even from a simple consideration of the phase self-modulation, nonlinear changes of the refractive index at the trailing (leading) edge of the light pulse are responsible for the anti-Stokes (Stokes) broadening of its spectrum [16]. Therefore, in quartz, where $n_\mathrm{o} < n_\mathrm{e}$, the polarization of the trailing edge of the resulting pulse is determined by the slower e-pulse. As a consequence, the process of phase self-modulation at the pulse trailing edge gives rise to the e-polarized SC in quartz, and to the o-polarized SC in sapphire.

The presented speculations are supported by calculations [17], which testify that, in the case of linearly polarized excitation pulses, the SC intensity is higher

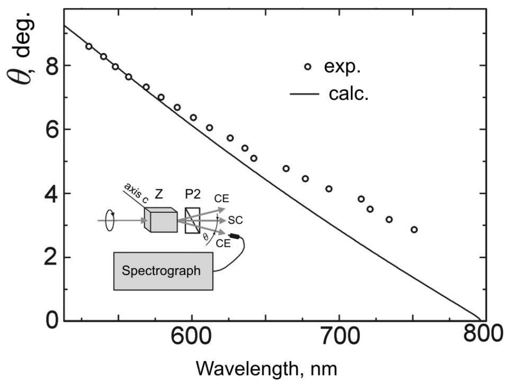

Fig. 6. Dependence of the angle θ (in air) for CE at the spectral maximum on the wavelength λ for the e-polarization in sapphire

than for the circularly polarized ones. Hence, it is the linearly polarized component of the pulse trailing edge that determines the SC polarization. Concerning the SC in the IR spectral interval, which is generated by the pulse leading edge, it has mainly the e-polarized component in sapphire. Note that, in the case of temporal splitting of a femtosecond laser pulse in the course of filamentation [1], the polarization of the leading and trailing edges of the cumulative pulse and, therefore, the SC polarization remain the same as in the case of unsplit pulse.

The issue concerning the CE polarization is more difficult. The polarizations of the SC and CE excitation rays are conventionally supposed to be identical in the isotropic medium [12–15]. The interrelation between the SC, CE, and dynamics of filamentation in isotropic media is explained differently. In work [18], the CE was associated with the Cherenkov radiation in the plasma channel of a filament surrounded by a Kerr shell and, in work [19], with the plasma defocusing of SC in the pulse tail section. In addition, a model for an enhancement of the modulation instability of the plane and monochromatic modes of a nonlinear Schrödinger equation in the angle–wavelength ($\theta - \lambda$) coordinates was proposed as a mechanism of CE generation in light filaments [20, 21]. In this model, the energy transfers from the main band of pulse excitation to the Stokes and anti-Stokes sidebands owing to a four-wave interaction. Recent numerical simulations of the spatiotemporal transformation of femtosecond laser pulses on the basis of a nonlinear Schrödinger equation [22, 23] predict rather well the appearance of new spectral components of radiation in filaments. However, simple physical interpretations still remain controversial.

Since the SC and IR CE polarizations are identical in sapphire, it is reasonable to assume that the CE generation occurs in the front section of the light pulse. In our opinion, the process of four-wave mixing (FWM), in which the IR CE and laser radiation with a central frequency of 800 nm participate, can also give a contribution to the mechanism of CE generation. The FWM can take place in the highly intensive core of a filament and at the leading edge of the pulse according to the following scheme:

$$\begin{aligned} &2\hbar\omega_0 = \hbar\omega_{\mathrm{IRSC}} + \hbar\omega_{\mathrm{CE}}, \\ &2k_0 = k_{\mathrm{IRSC}} + k_{\mathrm{CE}}\cos\theta. \end{aligned} \tag{1}$$

Here, the subscript 0 corresponds to laser radiation, and θ is the angle of CE with respect to the axis X. The first equation describes the energy conservation, and the second one is the equation of longitudinal phase synchronism, which is typical of nonlinear phenomena in the narrow filament channel. The preservation of only the longitudinal component of the wave vector is associated with the fact that the transverse size of the high-intensity section in the filament core, where the nonlinear interaction just takes place, amounts to a few wavelengths. As a result, requirements to the transverse phase synchronism become weaker.

Note that, unlike the FWM in which the Stokes, anti-Stokes, and excitation radiations participate [21], the proposed FWM scenario includes the excitation, CE, and IR SC. The result of calculations of the system of equations (1) is shown in Fig. 6 by a solid curve. Laser radiation and the IR SC are directed along the axis X, and the CE is generated at an angle θ. The value of the angle θ was corrected for air with regard for the refraction across the output face of the specimen. No fitting parameters were used in calculations.

Figure 6 also exhibits the experimental $\theta - \lambda$ spectrum of the CE excited in sapphire by a circularly polarized laser ray with a pulse energy of 1.5 μJ, as well as the results of calculations by formulas (1). The measurement setup is shown in the inset.

One should pay attention to a very good agreement between the experimental data and the results of calculations in the short-wave interval of 0.53–0.65 μm and to rather large deviations in the long-wave section. This fact may mean that the proposed mechanism is actual for the short-wave edge of CE. As follows from the equation of energy conservation (1), the short-wave limit of CE, which is observed at a wavelength of 530 nm, agrees well with a long-wave limit of 1600 nm for the IR SC in sapphire [6]. Note that, although the numerical models proposed in works [22, 23] adequately describe the $\theta - \lambda$ spectrum in a wide wavelength interval, the clear scenario of the nonlinear interaction becomes lost at that. Our interpretation is directed at resolving the major nonlinear processes that are responsible for the formation of the CE short-wave edge in the case of birefringent medium from a series of interconnected nonlinear interactions in the filament core, which govern the spatiotemporal and spectral transformations of a femtosecond laser pulse in the filamentation mode.

4. Conclusions

To summarize, the first observation of the polarization features of SC and CE in birefringent media with $n_o < n_e$ and $n_o > n_e$ has been reported. These features are shown to be a result of the temporal divergence between o- and e-polarized pulses. The CE polarization is found to be determined by the polarization of the trailing edge of the cumulative pulse, whereas its leading edge determines the SC polarization.

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“БІЛИЙ СУПЕРКОНТИНУУМ” І “КОНІЧНА ЕМІСІЯ” ВИПРОМІНЮВАННЯ ФЕМТОСЕКУНДНИХ ФІЛАМЕНТІВ У СЕРЕДОВИЩАХ З ПОДВІЙНИМ ПРОМЕНЕЗАЛОМЛЕННЯМ

Р е з ю м е

Повідомляється про спостереження поляризаційних особливостей “білого суперконтинууму” та “конічної емісії” фемтосекундних лазерних філаментів у кристалах кварцу та сапфіру, зумовлених їх додатним та від'ємним подвійним променезаломленням. “Білий суперконтинуум” і “конічна емісія” набувають взаємно ортогональних напрямків поляризації як результат різниці групових швидкостей звичайного і незвичайного променів. Запропоновано фізичний механізм генерації конічної емісії, який пояснює особливості її поляризації.