CHERENKOV-TYPE CONDITIONS OF SYNCHRONISM FOR CONICAL EMISSION OF FEMTOSECOND BESSEL BEAMS

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It has been demonstrated that the angular distribution of wavelengths in the conical emission (CE) by a femtosecond Bessel beam in water is governed by the condition that the axial phase velocities of all CE spectral components are identical. The enhancement along the axial direction owing to the four-wave mixing results in the appearance of discrete rings of conical emission.

1. Introduction

Almost immediately after the introduction of pulse laser sources into the practice of scientific researches, the studies of the self-action of laser radiation in solid-state Kerr media were started and actively developed at the Institute of Physics of the NAS of Ukraine under the guidance of M.S. Brodyn. For semiconducting materials with various structures of their electron energy bands, the basic mechanisms have been established, which are responsible for the effects of self-focusing and defocusing of the laser radiation, a competition between focusing processes and multiphoton radiation absorption, the magnitude and the rate of nonlinear optical response of semiconductors, and other processes (see, e.g., works [1, 2]). These researches got a qualitatively new impetus in recent years. This fact was associated with the bringing into action of the "Laser femtosecond complex" at the Institute of Physics of the NAS of Ukraine, and M.S. Brodyn played a considerable role in its creation.

In this work, we discuss researches of the nature of new nonlinear optical phenomena, which accompany the propagation of powerful femtosecond laser pulses in Kerr media. One of such phenomena is the so-called conical emission (CE), which emerges, in particular, at the spontaneous filamentation of a Gaussian beam in transparent Kerr media. A characteristic attribute of such CE is an increase of the radiation frequency with the angle θ . As the filament length increases, the initially continuous frequency-angular distribution of CE becomes split into a series of discrete rings, every of them being characterized by a fixed angle θ and a specific radiation frequency.

2. Conical Emission Models

Since conical waves play a central role in the understanding of what follows, let us consider their main properties in brief. A linear monochromatic conical wave comprises a set of plane waves, the wave vectors \mathbf{k} 's of which are distributed over a cone surface at the angle θ with respect to the cone axis. In its transverse cross-section, the distribution of conical wave intensities, similarly to the filament case, has an intensive axial maximum, the diameter of which is constant along the axis. This maximum is surrounded by concentric rings with growing diameters, and the intensity distribution is described by the square of a Bessel function. Therefore, conical waves are also called the Bessel ones. In practice, a conical wave is created most often by focusing a Gaussian beam with the use of an axicon (Fig. 1). The axial phase velocity of its propagation in a dispersion medium, $V = c/(n \cos \theta)$, exceeds the light velocity at the given wavelength, c/n [3]. If the amplitude front of a conical wave is not inclined with respect to the phase one, the axial group velocity of the wave (the velocity, with which the axial maximum of a light pulse moves) equals $v = v_m / \cos \theta$, where v_m is the group velocity of light in the medium. However, the difference between the group and phase velocities in the



Fig. 1. Experimental set-up and the photo of CE on a screen at a distance of 500 cm from the axicone with a completely open iris diaphragm and a pulse excitation energy of 260 μ J: IR (1058 nm, 0°), Pump (802 nm, 5.7°), R (637 nm, 8.0°), G (531 nm, 10.0°), B (459 nm, 11.5°), and UV (390 nm, 12.9°). The lower part of the figure schematically illustrates the Bessel zone formed by the axicone, together with the corresponding intensity distribution

material of a focusing axicone results in an inclination α of the wave amplitude front. So, the expression for the group velocity v_0 of an excitation conical wave takes the form $v_0 = v_m \cos \alpha / \cos(\theta_0 + \alpha)$ [4].

The majority of modern models describing the CE formation (three-wave mixing [5], phase modulation [6–8], and formation of so-called X-waves [9–11]) are based on the idea of a point-like broadband source that moves together with the pulse in the region with a high gradient of field intensity, where, strictly speaking, the spectrum broadens out, first of all owing to the phase modulation. Under the superposition, this emission forms such a frequency-angular distribution for CE, in which the angle increases with the shift into the anti-Stokes region. The interference model [8] makes allowance for the interference of CE from a number of axial maxima along the filament, which leads to a split of the CE spectrum, continuous in the angle, into a set of discrete rings. Models of four-wave mixing (FWM) [12-14] and Cherenkov radiation (CR) at the dynamic border of an antiwaveguide created at the filamentation [15, 16] were also proposed. The authors of work [16] found that, for CE in air, the product $n \cos \theta$ is identical for all CE components. This means that the axial phase velocities are identical for all CE components. At the same time, a key role in the modeling of the nonlinear formation of X-waves is played by the assumption that the axial group velocities of all components in a polychromatic conical wave packet are identical, which gives rise to high intensities of nonlinear processes [17].

An experimental test that the angular spectrum of CE by filaments in air can be described using the FWM [14],

X-wave [11], and CR [16] models was carried out in work [18]. It was shown there that the expression describing the CE angular spectrum in the CR model can be considered as a special case of the more general expression obtained in the framework of the X-wave model, provided that the group velocity of a filament is equal to the phase velocity at the excitation frequency. The best agreement of theoretical calculations with experimental results was obtained just in this case.

However, in the experiments discussed above, the CE was studied in the regime of filamentation. As a result of the large nonlinear variation of the refractive index at the filament axis, we need to use the fitting parameters while comparing theoretical and experimental results. In this work, we use a regime of weak nonlinear excitation by a Bessel wave. In our opinion, namely the regime of weak nonlinearity before the transition into the strongly nonlinear regime of filamentation will allow one to attain a success in the quantitative description of the CE angular spectrum from the first principles and without any fitting parameters, because the complexity of such a description is associated to a great extent with the ambiguity of spatial and temporal parameters of the electromagnetic field (in particular, the axial group and phase velocities) in the strongly nonlinear regime of filamentation.

Another advantage of the Bessel excitation consists in a possibility to extend the measured angular dependence of CE onto the axial direction ($\theta = 0$) and into the infrared spectral range. On the other hand, the analytical expressions for CE angular spectra, which are presented in [11, 14, 16], cannot be applied directly to the Bessel

excitation case. Therefore, a straightforward verification of the conclusions made in those works is impossible. In this connection, we aimed at finding which principle of synchronism, the group or the phase one, is engaged at the CE generation. For this purpose, we used water as a Kerr medium, where the group and phase velocities are substantially different. We demonstrated unambiguously that, under the experimental conditions used in this work, the angular dispersion of CE is described by the Cherenkov-type dependence, which follows from the condition of equal axial phase velocities for all components in the polychromatic conical wave packet. The discrete behavior of rings is explained by the presence of a selected direction along the pumping beam axis, in which the FWM process is excited.

3. Experimental Results and Their Discussion

For the excitation of CE, we used horizontally polarized femtosecond laser pulses with a wavelength of 802 nm, the half-height duration $\tau = 150$ fs, a pulse-repetition frequency of 1 kHz, and a spectral width of 10 nm (Fig. 1). The beam was additionally confined using an adjustable iris diaphragm characterized by a maximum aperture diameter of 12 mm and focused with an axicon 12 mm in diameter, made of glass K8, and with a basis angle of 11.2° into a glass cuvette 30 mm in length and filled with water. It was so done that the axial intensity in the Bessel zone should be maximal at the cuvette center. The angle of the conical wave formed by the axicon with respect to the axis was $\theta_0 = 4.3^\circ$ in water. From the photo in Fig. 1, one can see that CE is characterized by a number of narrow discrete rings of red (R), green (G), and dark blue (B) colors, as well as by a weak ultraviolet (UV) ring. A narrow (a half-height width of 2°) beam of infra-red (IR) radiation was generated in the axial direction. In addition, the pumping conical radiation (Pump) was registered. We used a flexible quartz optical fiber 1 mm in diameter to probe CE at a distance of 50 cm from the axicon in the maxima of intensity rings at various angles β . The collected light was projected onto the input slit of a spectrometer.

The polarization of both CE and axial IR radiations coincides with that of the pumping radiation. The CE intensity has a threshold (185 μ J) and the superlinear dependence on the excitation pulse energy, which evidences its stimulated character. As the energy of excitation pulses grows, first, the red ring and the IR emission appear simultaneously; afterwards, the green, blue, and ultra-violet rings appear in turn. The rings are most pronounced and monochromatic, if the excitation level CHERENKOV-TYPE CONDITIONS OF SYNCHRONISM



Fig. 2. Angular dependences of the CE spectrum for the Cherenkov-type synchronism. Hollow circles correspond to experimental points. Results of calculations: phase synchronism (solid curve 1), group synchronism (dashed curve 2)

is minimal. If the energy of excitation pulses grows further, the structure of rings becomes more complicated, and, in the axial direction, there emerges radiation belonging to a quasiwhite supercontinuum, which may be a result of the transition to a strongly nonlinear regime of filamentation in the Bessel beam. Therefore, in what follows, we confine the discussion of CE to the minimum excitation level.

In Fig. 2, circles demonstrate the energies of quanta and the localization angles for rings of various orders obtained at an excitation pulse energy of 260 μ J. Points "Pump" and "IR" are also shown. The spectral width of CE bands within a ring are close to 30 meV, somewhat exceeding the pumping band width (20 meV).

Those experimental data were compared with the calculated angular spectra of CE, provided that either of two types of synchronism – phase (solid curve) or group (dashed curve) one – was realized. In the former case, the phases of all components in a polychromatic conical wave packet were assumed to be identical at the beam axis, i.e. the axial phase velocities in water, $V = c/(n\cos\theta)$, are identical for all wavelengths and equal to the phase velocity of the excitation conical wave, $V_0 = c/(n_0 \cos \theta_0) = 2.2627 \times 10^{10}$ cm/s. In the latter case, the axial group velocities in water, $v = v_m / \cos \theta$, were assumed to be identical for all wavelengths and equal to the group velocity of the excitation conical wave, $v_0 = v_m \cos \alpha / \cos(\theta_0 + \alpha) = 2.2391 \times 10^{10} \text{ cm/s.}$ According to the results of our calculations for the axicon material (glass K8), the slope of the amplitude wave front

with respect to the phase one was $\alpha = 0.15^{\circ}$ in water; for all CE frequencies, but the exciting one, it was considered to equal zero. The angles θ 's in water were recalculated into the angles β 's in air. In contrast to work [18], we used no analytical expressions for the dependence of the CE angle on the wavelength. Instead, the problem was solved numerically, making allowance for the dispersion of the refractive index in water according to the results of work [19]. Note that, for a charge, whose velocity V_c exceeds that of light, the angular dependence of the Cherenkov radiation wavelength in a dispersion medium arises as a consequence of the similar condition for the Cherenkov synchronism, $V_c = c/(n \cos \theta)$.

The presented plot makes it evident that the experimental points, including the data for the axial IR radiation, are well described by Cherenkov angular dependence 1 for phase synchronism. This result confirms the conclusion made in works [16, 18] about the equality of axial phase velocities of all components of a CE filament in air and extends it onto the cases of condensed media (water) and the excitation with a Bessel wave. For the axial radiation, the condition of phase synchronism is especially simple: $V_0 = c/n_{\rm IR}$.

Note that the Cherenkov synchronism condition is not rigidly connected with the Cherenkov mechanism of radiation, i.e. the polarization of a medium by a moving charge. Nevertheless, the longitudinal separation of charges in the plasma trace is really probable in our experiment, so that it can be a source of CE. In particular, this mechanism was used as a basis for the explanation of the terahertz radiation in femtosecond filaments [20]. However, first, a dipole cannot emit in its axial direction and, second, for symmetry reasons, the polarization of the CE generated by a dipole must be radial. Since the radiation is plane-polarized, and its IR component is directed along the axis, the Cherenkov mechanism of CE has to be excluded from consideration.

At the same time, the condition of phase synchronism of the Cherenkov type describes a continuous angular spectrum of CE and gives no answer concerning the nature of the observable ring discreteness. In this connection, let us consider the phenomenon of FWM, which is promoted by both the configuration of Bessel beams – owing to a high intensity in the axial maximum (of about 50 TW/cm² at a pulse energy of 300 μ J) – and the large length of the central peak. Therefore, it is possible to suppose that IR and R radiations can be generated by two pumping quanta in the course of FWM. Really, in Fig. 2, vertical lines mark the wavelengths that correspond to the combination wavelengths obtained in the FWM scenario, $1/\lambda_{\rm R} = 2/\lambda_0 - 1/\lambda_{\rm IR}$; for

higher orders, the conditions are $1/\lambda_{\rm G} = 2/\lambda_{\rm R} - 1/\lambda_0$, $1/\lambda_{\rm B} = 2/\lambda_{\rm G} - 1/\lambda_{\rm R}$, and $1/\lambda_{\rm UV} = 2/\lambda_{\rm B} - 1/\lambda_{\rm G}$. One can see that the measured wavelengths of lines R, G, B, and UV agree well with the expected ones obtained at FWM. In addition, the specificity of FWM in a conical beam also brings about a modification of the usual phase synchronism conditions: $2\mathbf{k}_0 = \mathbf{k}_{IR} + \mathbf{k}_R$. Since the density of Bessel beam intensity in the axial maximum is 6.25 times as high as that in the first ring, the generation of new components, which is proportional to the squared pumping intensity, near the threshold will take place exclusively locally on the beam axis. In this case, the ordinary conditions of phase synchronism for FWM lose their meaning. It is important only that the sum of the axial phase velocities of generated conical waves should be equal to the doubled phase velocity of the conical pump wave, $V_{\rm IR} + V_{\rm R} = 2V_0$ or $c/(n_{\rm IR}\cos\theta_{\rm IR}) + c/(n_{\rm R}\cos\theta_{\rm R}) = 2c/(n_0\cos\theta_0)$. This condition is equivalent to the condition of longitudinal phase synchronism [21], being undoubtedly obeyed on curve 1, where all phase velocities are identical.

From the aforesaid, the origin of the discrete behavior of CE rings becomes clear. In the described geometry of the experiment, there exists a unique selected direction, the axial one, in which the radiation in the course of FWM is most stimulated. That is why, among a continuum of possible CE angles, only a single variant, $\theta = 0$, is realized, in which the infra-red radiation that satisfies the condition $V = c/n_{\rm IR}$ is generated. The spectral composition of other rings consists of the combination wavelengths obtained at FWM: $1/\lambda_{\rm R} = 2/\lambda_0 - 1/\lambda_{\rm IR}$ and so forth. One can see that the points, where curve 1 intersects vertical lines corresponding to the combination wavelengths at FWM, are close to the experimentally observed ones. Note that, in the present consideration, FWM does not define the condition of CE synchronism, being only a secondary phenomenon that creates the selective conditions for discrete frequencies to be generated.

Concerning the group synchronism condition, which is necessary for the maximal efficiency of CE generation, its realization is also possible, provided the proper selection of a correcting parameter α ; i.e. new generated CE waves will possess a necessary inclination of their amplitude wave front. However, we reserve this issue for further researches.

4. Conclusions

To summarize, we showed in this work that, under the described experimental conditions, the angular spectrum

of CE is determined by a condition that the axial phase velocities of all its components are identical. The discreteness of CE rings is explained by the presence of a selected axial direction of the pump beam, along which the generation and the amplification of IR radiation in the course of FWM take place, together with the simultaneous generation of red radiation, as well as radiation with other frequencies in the course of FWM processes of higher orders.

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УМОВИ СИНХРОНІЗМУ ЧЕРЕНКОВСЬКОГО ТИПУ ДЛЯ КОНІЧНОЇ ЕМІСІЇ ФЕМТОСЕКУНДНИХ БЕССЕЛІВСЬКИХ ПУЧКІВ

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Резюме

Показано, що кутовий розподіл довжини хвилі конічної емісії фемтосекундного бесселівського пучка у воді визначається умовою рівності осьових фазових швидкостей всіх її спектральних компонент. Підсилення в осьовому напрямку внаслідок чотирихвильового змішування приводить до появи дискретних кілець конічної емісії.