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SCATTERING OF GINZBURG–FRANK AND CHERENKOV TYPES UNDER SELF-FOCUSING OF NANOSECOND LASER PULSES IN LIQUIDS [1]

We study the dynamics of nonlinear optical processes such as self-focusing, self-phase modulation, and stimulated Raman scattering in Kerr-liquids under the nanosecond laser pulse excitation. The results prove the existence of the transition Ginzburg–Frank-type effect, which promotes the appearance of new spectral components of the laser radiation at the medium boundary. The generation of extended anti-Stokes frequency-angular bands of stimulated Raman scattering is explained. When the velocity of a self-focusing focal spot matches the phase velocity of the non-linear polarization at the anti-Stokes Raman frequency and the phase velocity of the scattered axial radiation, the most intense frequency-angular bands appear. They are described by the equations typical of the Cherenkov radiation.

K e y w o r d s: self-focusing, self-phase modulation, stimulated Raman scattering.

1. Introduction

The self-focusing (SF) of laser pulses in the nanosecond range in a Kerr medium leads to the movement of a focal spot [1]. The focal spot speed $v_{\rm fp}$ is defined by the laser pulse envelope. At the front and back of a pulse, $v_{\rm fp}$ takes positive and negative values and is not limited by the speed of light in vacuum [2].

In the practical aspect, SF creates a new situation – the dynamics of nonlinear optical processes such as the self-phase modulation (SPM) and the stimulated Raman scattering (SRS), which cannot be achieved within other technical methods.

Previously, we identified the stop point location of a focal spot [3], possibility of Cherenkov-type radiation of SRS under SF [4], effect of SF on angular spectra of SRS [5], angle-selective inverted SRS [6], frequency dependence of anti-Stokes SRS on the focal spot speed in the approach of "ideal thin lens"

[7], transition effect of SPM [8], and physical mechanism of anti-Stokes SRS of the Cherenkov type under SPM [9].

Now, it is possible to state the principles of the Ginzburg–Frank transition and the Cherenkov (or Vavilov–Cherenkov) superluminal scattering [10] under SF of nanosecond laser pulses in Kerr liquids.

2. Consideration and Analysis

A simplified scheme for describing the processes is presented in Fig. 1. The focal spot has velocity [2] $v_{\rm fp} = v_{\rm fd} v_{gL}/(v_{\rm fd} + v_{gL})$, which can exceed the light speed c in vacuum ($v_{\rm fd}$ – velocity of focal $z_{\rm f}$ distance change, v_{gL} – group speed of laser radiation).

The laser spot has velocity $v_{\rm fp} = v_{\rm fd} = 0$ for the top of a laser pulse. At this stop-point of a laser spot, classical SRS can be observed, which has asymmetric

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[1] The paper was presented at the XXIII Galyna Puchkovska International School-Seminar "Spectroscopy of Molecules and Crystals".
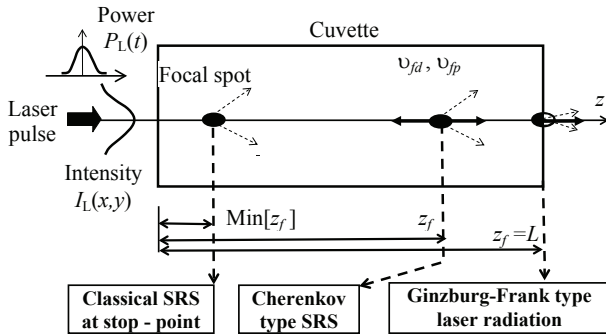
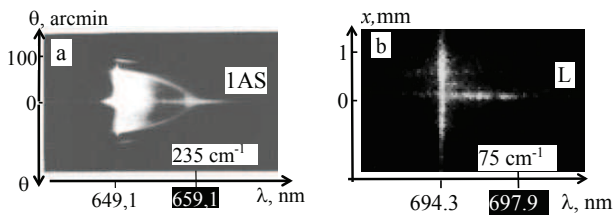**Fig. 1.** Simplified scheme for describing the processes

Fig. 2. Experimental frequency-angular spectrum of the Cherenkov-type scattering (a) in the coordinates angel (θ) – wavelength (λ) and frequency-spatial spectrum of the transition-type scattering (b) in the coordinates: lateral coordinate (x) – wavelength (λ) for a ruby laser at 20 ns, 0.5 J pulse in toluen

indicatrix for the parametric scattering. However, the indicatrix asymmetry is another question.

When the focal spot is closer to the exit of a cuvette, the Cherenkov-type superluminal scattering can be observed for parametric SRS. The experimental frequency-angular spectrum of the Cherenkov-type scattering is presented in Fig. 2, a for the first anti-Stokes (1AS) SRS in toluene under the excitation by a ruby laser in the coordinates: scattering angle (θ) – wavelength (λ). As the focal spot crosses the exit boundary of a cuvette, Ginzburg–Frank transition-type scattering is observed for the laser radiation. An experimental frequency-spatial spectrum of the transition-type scattering is presented in Fig. 2, b for the ruby laser radiation (L) in the coordinates: lateral coordinate (x) – wavelength (λ). The maximum Stokes frequency shifts in reverse centimeters (cm^{-1}) are indicated in Fig. 2.

A general qualitative similarity for the axial scattering is observed. This is a result of the mutual SPM-effect of the laser radiation. The maximum Stokes frequency shift $\Delta\nu_{1AS}$ for the first anti-Stokes SRS is about 3 times more than the laser shift $\Delta\nu_L$, because

$\nu_{1AS} = 2\nu_L - \nu_S$ and $\Delta\nu_{1AS} = 2\Delta\nu_L + \Delta\nu_S = 3\Delta\nu_L$ (here ν_S is the Stokes component frequency). The maximum Stokes frequency shift $\Delta\nu_{2AS}$ for the second anti-Stokes SRS is about 7 times more than $\Delta\nu_L$, seeing $\nu_{2AS} = 2\nu_{1AS} - \nu_L$, $\Delta\nu_{2AS} = 2\Delta\nu_{1AS} + \Delta\nu_L = 7\Delta\nu_L$.

For the maximal value $\Delta\nu_L$ of the frequency Stokes-shift caused by the transition effect for the laser radiation, it is possible to derive the analytical expression:

$$\Delta\nu_L \approx \nu_L \Delta n_{\mathrm{f}}\{z = L\} \frac{\upsilon_{\mathrm{fd}}\{z = L\}}{c},$$

where z is a longitudinal coordinate, $\Delta n_{\mathrm{f}}\{z = L\}$ is an increment of the refractive index at the focal point at the distance $z = L$ (at the medium boundary), $\upsilon_{\mathrm{fd}}\{z = L\}$ – a velocity of the focal point at the medium boundary (without considering the difference between times, which are necessary for the pulse fragments to reach the focal point), and c – the speed of light.

The maximum energy density of axial ($\theta = 0$) 1AS radiation is located at the frequency determined by conditions, which are similar to those for the Cherenkov radiation: equality of the phase velocities of electromagnetic waves $\upsilon_{\mathrm{ph}}\{\omega\}$ at the frequency ω and the phase velocity υ_{ap0} of a polarization at the anti-Stokes Raman frequency ω_a. The axial frequency shift in toluene is $(\omega\{\theta = 0\} - \omega_a)/2\pi c = -197\ cm^{-1}$.

For $\theta \neq 0$ and $\upsilon_{\mathrm{fp}} = \upsilon_{\mathrm{ap0}}$, the frequency-angular branches are related by the expression

$$\cos\theta \approx \upsilon_{\mathrm{ph}}(\omega)/\upsilon_{\mathrm{ap0}}$$

that gives a parabola for $\cos\theta \approx 1 - \theta^2/2$.

3. Conclusions

The Ginzburg–Frank (transition effect) – type and Cherenkov (superluminal effect) – type radiations are analyzed at the nanosecond laser pulse excitation in the spectra of a laser and SRS.

1. At the transition effect, the maximum frequency shift of the laser radiation appears, when the focal point of self-focusing intersects the exit boundary of the medium, and the phase delay of SFM before and after the focal point is uncompensated.

2. The SRS Cherenkov-type radiation appears at the coincidence of self-focusing focal point speed, the

phase velocity of a nonlinear polarization at the anti-Stokes Raman frequency and the phase velocity of scattered SRS-radiation. This is a result of the superluminal speed of the focal point and the velocity of a nonlinear polarization.

It will be if interest to analyze the mutual impact of the Ginzburg–Frank and Cherenkov effects on SRS.

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РОЗСІЮВАННЯ ГІНЗБУРГА–ФРАНКА ТА ЧЕРЕНКОВСЬКОГО ТИПІВ ЗА САМОФОКУСУВАННЯ НАНОСЕКУНДНИХ ЛАЗЕРНИХ ІМПУЛЬСІВ У РІДИНАХ

Р е з ю м е

Досліджено динаміку нелінійно-оптичних процесів (самофокусування, фазова самомодуляція, вимушене комбінаційне розсіювання) у керівських рідинах за дії наносекундних лазерних імпульсів. Результати доводять наявність перехідного ефекту типу Гінзбурга–Франка, який породжує нові спектральні компоненти лазерного випромінювання на межі середовища. Пояснено генерацію протяжних частотно-кутових смуг вимушеного комбінаційного розсіювання. У випадку збігання швидкості фокальної точки з фазовою швидкістю нелінійної поляризації на антистоксовій комбінаційній частоті та фазовою швидкістю розсіяного аксіального випромінювання виникають найінтенсивніші смуги, які описуються рівняннями, характерними для черенковського випромінювання.