Contribution to the theory of stimulated Raman scattering in focused light beams

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Emission of the first anti-Stokes and second Stokes components of stimulated Raman scattering is considered for the case of focused beams of the exciting and first Stokes components. Conditions are found under which emission occurs predominantly along the generators of the cones. Expressions are obtained for the angles of predominant emission and for the respective angular widths. The dependence of the emission intensity on the layer thickness of the scattering layer or on lengths of the focal regions of the exciting and first Stokes components is investigated. It is found that in the limit of a strongly elongated focal-region caustic the emission cones of the first anti-Stokes and second Stokes components go over to cones of the Cerenkov type. It is shown that the so-called class-II emission of these components, which is observed experimentally, can be interpreted as emission from the foci of a multifocus structure.

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INTRODUCTION

It is well known that parametric excitation of anti-Stokes and higher Stokes components of stimulated Raman scattering (SRS) is due to their interaction with the wave of the exciting radiation and the first Stokes component of the scattering (see, e.g., [1-3]). In the approximation in which the exciting and Stokes fields are given, the radiation of each of the considered components is described as the radiation of extraneous currents (extraneous polarization) with a specified distribution of the amplitudes and the phases. Inasmuch as under ordinary conditions, the scale of the spatial variation of the complex amplitudes of the currents is much smaller than the dimensions of the scattering region, the fields radiated by the different sections of this region can interfere with one another, leading to a specific directivity pattern in the summary radiation. Under these conditions, the angular distribution of the intensity of the considered components far from the scattering region is sensitive even to relatively small changes in the form of the current distribution. If at the same time the field of Stokes component (or of the exciting radiation) is random, then the angular distribution of the intensity of the anti-Stokes or higher Stokes component will be correspondingly strongly dependent on the statistical properties of this field.

The essential dependence of the angular distribution of the intensity of the indicated components on the form of the distribution of the fields of the exciting radiation and of the first Stokes component was noted in papers by one of us.[2,4] In those papers, the angular distribution of the intensity of the anti-Stokes and higher Stokes components of SRS was calculated for the case when the exciting radiation is a plane monochromatic wave, whereas the first Stokes component is generated by individual pointlike bare sources of the first Stokes frequency in a medium with large gain (see[2,5]). It was shown in a paper[6] devoted to the influence of the statistical properties of the field of the first Stokes component on the angular distribution of the intensity of the considered components, that the plane-wave method used for the calculations in the earlier papers (without account of the interference of the different plane waves with one another) leads generally speaking, with respect to the components of higher orders, to qualitatively incorrect results. This seems to explain also the contradictory character of the later studies [7,8], which deal with the radiation of higher-order components and which use similar methods to simplify the analysis.[1)]

Experimentally, starting with the work of Terhune[9] and Stoicheff[10], radiation of anti-Stokes and higher Stokes components was observed along the generators of the cones, a distinction being made between cones of two types. The first is characterized by a definite apex angle (the so-called class-I radiation). The cones of the second type can multiply, diffuse, or randomly varying in the apex angle from experiment to experiment (class II radiation, see [11]). In his experimental study, Garmire [11] has shown that radiation of class II is connected with the onset of thin optical "filaments" in the medium, which in turn were interpreted as waveguide propagation of the light.

A consistent theory of class-I radiation as radiation from individual Stokes pointlike sources was developed in [2,4]. In these studies, in particular, relations were established for the wave vectors of the interacting waves, from which the apex angles were established for the radiation cones of the higher-order components. The radiation angles of the considered components, as calculated in the indicated theory, coincide with high accuracy with the experimentally observed quantities. On the basis of this theory, a new type of radiation cone, connected with the interaction of ordinary and extraordinary waves, was also predicted[12]. Cones of this type were then observed experimentally in[13,14]. The observed pictures of the radiation of the anti-Stokes and second-Stokes components, which depend essentially on the orientation of the crystal, turn out to be in good quantitative agreement with the theoretical predictions at all possible orientations. Thus, radiation of class I has by now been sufficiently thoroughly investigated both experimentally and theoretically.

As to the radiation of class II, its initial interpretation[11] as radiation from an optical waveguide filament (i.e., as radiation of Cerenkov type[15]) encountered a difficulty in that the calculated cone-apex angles were fixed for each given substance, always exceeding to one degree or another the measured values in the anti-Stokes region, but smaller than the observed values in the Stokes region.[11,16] Since it was established recently (see the review[17]) that the optical filaments observed in substances are in fact the results of the motion of foci of a multifocus structure, it is of interest to consider the radiation of the anti-Stokes and higher Stokes SRS from foci, i.e., under those conditions when the exciting radiation (which we shall henceforth call the principal component), or else the radiation of the first Stokes component, is focused. The present paper is devoted to this question. Using as an example the first anti-Stokes and the second Stokes components, we shall show below that the properties of the radiation of class II are explained on the basis of the multifocus structure. In addition, a new type of radiation cone is predicted for the anti-Stokes and higher Stokes components in focused beams.

We note right away that the obtained radiation cones are produced as radiation of a polarization wave whose velocity exceeds that of light in the medium at the same frequency. From the point of view, an analogy can be drawn with Cerenkov radiation. However, unlike radiation of the Cerenkov type, which appears for example in calculations for an unbounded filament, in the case of focused beams the values of the angles themselves differ from the Cerenkov angles. Therefore the picture obtained by us can be regarded only as modified Cerenkov radiation.

1. DERIVATION OF INITIAL RELATIONS

Thus, we assume that the fundamental and first Stokes components are specified focused axially-symmetrical beams (for the sake of argument, Gaussian) propagating along a common axis in a medium that is active in the Raman spectrum. We confine ourselves to the case of small angles of convergence (divergence) of the beams, which is of greatest interest from the practical point of view. The complex amplitudes of the fields of the first anti-Stokes $v_1^*(\mathbf{r})$ and of the second Stokes $u_2(\mathbf{r})$ components are connected with the complex amplitudes of the fundamental $E_0(\mathbf{r})$ and first Stokes $u_1(\mathbf{r})$ components by the equation [1-3]

$$-\Delta v_1^*-k_1^2v_1^*=\alpha_1F(\mathbf{r})u_1^*E_0^2,\quad -\Delta u_2-k_{-2}^2u_2=\alpha_{-2}F(\mathbf{r})u_1^2E_0^*,\qquad (1)$$

where

$$\alpha_1=\frac{4\pi\Gamma}{\Delta-i}k_1^2,\quad \alpha_{-2}=\frac{4\pi\Gamma}{\Delta+i}k_{-2}^2,\quad \Gamma=\frac{3\lambda_{-1}^4NQ_0}{2^8\pi^5\hbar\Delta\omega},$$
$$k_l=k_l\varepsilon^{1/2}(\omega_l),\quad k_l=\omega_l/c,\quad \omega_l=\omega_0+l(\omega_0-\omega_{-1});\qquad (2)$$

ω_0, ω_{-1}, ω_1, and ω_{-2} are the frequencies of the fundamental, first-Stokes, first-anti-Stokes, and second-Stokes components, respectively, $\lambda_{-1} = 2\pi/k_{-1}$ is the length of the Stokes wave, Q_0 is the cross section, per molecule, of the spontaneous Raman scattering in the transition under consideration, $\Delta\omega$ is the line width of the spontaneous Raman scattering, $\Delta = 2(\omega_{-1} + \omega_r - \omega_0)/\Delta\omega$, ω_r is the frequency of the considered transition of the material, N is the density of the molecules, $F(\mathbf{r}) = 0$ outside the volume V of the active medium and $F(\mathbf{r}) = 1$ inside this volume.

In our case of focused Gaussian beams, the expressions for E_0 and u_1 can be written in the form (see, e.g., [17])

$$E_0=\frac{\tilde{e}_0}{1+i\zeta/l_0}\exp\left(ik_0z-\frac{1}{2a_0^2}\frac{1}{1+i\zeta/l_0}r_\perp^2\right),$$
$$u_1=\frac{\tilde{u}_1}{1+i(\zeta+\Delta\zeta)/l_{-1}}\exp\left(ik_{-1}z-\frac{1}{2a_{-1}^2}\frac{1}{1+i(\zeta+\Delta\zeta)/l_{-1}}r_\perp^2\right),\qquad (3)$$

where $\zeta = z - z_0$. The notation introduced in (3) has the following meaning: a_0 and a_{-1} are the radii of the focal regions of the beams of the fundamental and first Stokes components, $z = z_0$ and $z = z_0 - \Delta\zeta$ are the cross sections corresponding to these radii; l_0 and l_{-1} are the lengths of the diffraction divergence of the considered beams. The conditions for the weak convergence of the beams, under which expressions (3) are valid, take the form

$$\theta_\alpha\ll1,\qquad (4)$$

where $\theta_\alpha = (k_\alpha a_\alpha)^{-1}$, $\alpha = 0, -1$.

We shall henceforth be interested in the fields v_1^* and u_2 at large distances from the scattering volume. In this case Eqs. (1) yield

$$v_1^*(\mathbf{r})=\frac{\alpha_1}{4\pi}\frac{\exp(ik_1r)}{r}\int\limits_V d\mathbf{r}'\,u_1^*E_0^2\exp(-ik_1\mathbf{n}\mathbf{r}'),$$
$$u_2(\mathbf{r})=\frac{\alpha_{-2}}{4\pi}\frac{\exp(ik_{-2}r)}{r}\int\limits_V d\mathbf{r}'\,u_1^2E_0^*\exp(-ik_{-2}\mathbf{n}\mathbf{r}'),\qquad (5)$$

where $\mathbf{n} = \mathbf{r}/r$. We calculate the integrals in (5) in analogy with [18]. Bearing further in mind the case of small observation angles $\theta = (n_x^2 + n_y^2)^{1/2}$, i.e., putting $n_z = 1 - \theta^2/2$ and taking (3) into account, we obtain

$$v_1^*(\mathbf{r})=\frac{\alpha_1\exp(ik_1r)}{4\pi r}\int\limits_V d\mathbf{r}'\Phi_1(\zeta')\exp\Big[i\Big(-\Delta k_0+\frac{k_1\theta^2}{2}\Big)z'-ik_1\mathbf{n}_\perp\mathbf{r}_\perp'+\gamma_1(\zeta')r_\perp'^2\Big],$$
$$u_2(\mathbf{r})=\frac{\alpha_{-2}\exp(ik_{-2}r)}{4\pi r}\int\limits_V d\mathbf{r}'\Phi_{-2}(\zeta')\exp\Big[i\Big(-\Delta k_{-1}+\frac{k_{-2}\theta^2}{2}\Big)z'-ik_{-2}\mathbf{n}_\perp\mathbf{r}_\perp'+\gamma_{-2}(\zeta')r_\perp'^2\Big],\qquad (6)$$

where

$$\zeta'=z'-z_0,\quad \mathbf{n}_\perp=(n_x,n_y),\quad \Delta k_0=k_1+k_{-1}-2k_0,\ \Delta k_{-1}=k_0+k_{-2}-2k_{-1},$$
$$\Phi_1(\zeta)=\frac{\tilde{e}_0^2\tilde{u}_1^*}{(1+i\zeta/l_0)^2(1-i(\zeta+\Delta\zeta)/l_{-1})},$$
$$\Phi_{-2}(\zeta)=\frac{\tilde{e}_0^*\tilde{u}_1^2}{(1-i\zeta/l_0)(1+i(\zeta+\Delta\zeta)/l_{-1})^2},\qquad (7)$$
$$\gamma_1(\zeta)=-\frac{1}{a_0^2}\frac{1}{1+i\zeta/l_0}-\frac{1}{2a_{-1}^2}\frac{1}{1-i(\zeta+\Delta\zeta)/l_{-1}},$$
$$\gamma_{-2}(\zeta)=-\frac{1}{2a_0^2}\frac{1}{1-i\zeta/l_0}-\frac{1}{a_{-1}^2}\frac{1}{1+i(\zeta+\Delta\zeta)/l_{-1}}.$$

For an active-region layer $0 < z < l$ we go over in (6) from the Cartesian variables x' and y' to the polar variables $r_\perp'$ and φ, and integrate with respect to φ. Then, for example, the first expression in (6) takes the form

$$v_1^*(\mathbf{r})=\frac{\alpha_1\exp(ik_1r)}{2r}\int\limits_0^l dz'\Phi_1(\zeta')\exp\Big[i\Big(-\Delta k_0+\frac{1}{2}k_1\theta^2\Big)z'\Big]\times\int\limits_0^\infty r_\perp'J_0(k_1\theta r_\perp')\exp[\gamma_1(\zeta')r_\perp'^2]dr_\perp'.\qquad (8)$$

Subsequent integration with respect to $r_\perp'$ yields

$$v_1^*(\mathbf{r})=-\frac{B_1\exp(ik_1r)}{4r}\int\limits_{-z_0}^{l-z_0}d\zeta'\frac{\Phi_1(\zeta')}{\gamma_1(\zeta')}\exp\Big[i\Big(-\Delta k_0+\frac{1}{2}k_1\theta^2\Big)\zeta'+\frac{k_1^2\theta^2}{4\gamma_1(\zeta')}\Big]$$
$$u_2(\mathbf{r})=-\frac{B_{-2}\exp(ik_{-2}r)}{4r}\int\limits_{-z_0}^{l-z_0}d\zeta'\frac{\Phi_{-2}(\zeta')}{\gamma_{-2}(\zeta')}\exp\Big[i\Big(-\Delta k_{-1}-\frac{k_{-2}\theta^2}{2}\Big)\zeta'+\frac{k_{-2}^2\theta^2}{4\gamma_{-2}(\zeta')}\Big]\qquad (9)$$

where

$$B_1=\alpha_1\exp[i(-\Delta k_0+{}^1/_2k_1\theta^2)z_0],\quad B_{-2}=\alpha_2\exp[i(-\Delta k_{-1}+{}^1/_2k_{-2}\theta^2)z_0].\qquad (10)$$

2. RADIATION CONES OF THE FIRST ANTI-STOKES AND SECOND STOKES COMPONENTS

We consider initially the case of a focused wave of the fundamental and plane wave of the first-Stokes component, i.e., we put $a_{-1}^2 = \infty$ and $l_{-1} = \infty$.

In this case expression (9), for example for the fields $u_2(\mathbf{r})$ of the second Stokes component, can be easily integrated and yields

$$u_2(\mathbf{r})=\frac{A_{20}}{r}\frac{\sin[{}^1/_2\varkappa_2(\theta)l]}{\varkappa_2(0)l}\exp\left(-\frac{k_{-2}^2a_0^2}{2}\theta^2\right), \tag{11}$$

where

$$A_{20}=B_{-2}a_0^2l\tilde{e}_0^*\tilde{u}_1^2\exp[i(k_{-2}r-\varkappa_2z_0+\varkappa_2l/2)],$$
$$\varkappa_2(\theta)=-\Delta k_{-1}+{}^1/_2k_{-2}(1+k_{-2}a_0^2/l_0)\theta^2. \tag{12}$$

The quantity Δk_{-1} in (12) is determined by the dispersion of the refractive index, and for liquids and solids in the visible region of the spectrum it is usually of the order of $\Delta k_{-1}\sim\Delta k_0\sim 30\ \mathrm{cm}^{-1}$. At $\Delta k_{-1}l\gg\pi$ expression (11) yields a sharply pronounced maximum of the radiation intensity of the second Stokes component for the angle $\theta=\theta_{-2}$, where

$$\theta_{-2}=\left[\frac{2\Delta k_{-1}}{k_{-2}(1+k_{-2}a_0^2/l_0)}\right]^{1/2}. \tag{13}$$

The total angle width $\Delta\theta_{-2}$ of this maximum is equal to

$$\Delta\theta_{-2}\approx\frac{2\pi}{\theta_{-2}k_{-2}l(1+k_{-2}a_0^2/l_0)}. \tag{14}$$

It is of interest to note that, according to (11) and (12), the radiation intensity at $\theta=\theta_{-2}$ is proportional to l^2, whereas the angle width $\Delta\theta_{-2}$ is proportional to l^{-1}. By the same token, with increasing width l of the active layer, the total power of the second Stokes component, radiated into the cone under consideration, increases in proportion to l.[2)] Of course, this growth can continue only until the reaction of the second Stokes component on the fundamental and on the first Stokes component appears. We note also that according to (11), outside the limits of the considered radiation cone, the angular distribution of the intensity has a structure in the form of oscillations in θ.

At $\theta_{-2}\gg\theta_0$, the systematic component of this structure has a maximum corresponding to $\theta=0$ (i.e., in the exact forward direction) with an angle width $\Delta\theta\sim\theta_0$. We see that the width of this maximum is determined by the angle of convergence of the rays in the beam of the fundamental component. An interesting feature of the considered angular distribution of the intensity is that it does not depend on the location of the focal region relative to the layer of the active medium. This distribution remains unchanged, for example, also in the case when only a converging or diverging part of the beam is situated in the active-medium layer.

As to expression (9) for the field $v_1^*(\mathbf{r})$ of the first anti-Stokes component, in this case, taking (7) into account, this expression takes the form

$$v_1^*(\mathbf{r})=\frac{A_{10}}{2\pi rl_0}\exp\left(-\frac{1}{4}k_1^2a_0^2\theta^2\right)\int\limits_{-z_0}^{l-z_0}d\zeta'\frac{\exp[i\varkappa_1(\theta)\zeta']}{1+i\zeta'/l_0}, \tag{15}$$

where

$$A_{10}={}^1/_2\pi B_1a_0^2l_0\tilde{e}_0^2\tilde{u}_1^*e^{ik_1r},$$
$$\varkappa_1(\theta)=-\Delta k_0+{}^1/_2k_1\theta^2(1-k_1a_0^2/2l_0). \tag{16}$$

For simplicity we consider the expression (15) under conditions when the active medium is infinite, i.e., $z_0=\infty$ and $l-z_0=\infty$. In this case

$$v_1^*(\mathbf{r})=\frac{A_{10}(r)}{r}\exp\left[-\frac{1}{4}k_1^2a_0^2\theta^2-\varkappa_1(\theta)l_0\right]\quad\text{at}\quad\varkappa_1(\theta)>0, \tag{17}$$

and $v_1^*(\mathbf{r})=0$ at $k_1(\theta)<0$. According to (17), the radiation of the first anti-Stokes component occurs in the angle region $\theta>\theta_1$, where

$$\theta_1=\left[\frac{2\Delta k_0}{k_1(1-k_1a_0^2/2l_0)}\right]^{1/2}. \tag{18}$$

At $\Delta k_0l_0\gg 1$, this radiation is directed mainly at an angle θ_1 (i.e., along the generators of a cone with apex angle $2\theta_1$), and the total angle width $\Delta\theta_1$ of the corresponding distribution of the intensity is

$$\Delta\theta_1\approx\frac{\ln 2}{2\theta_1k_1l_0(1-k_1a_0^2/2l_0)}. \tag{19}$$

In contrast to the case of the second Stokes component considered above, the radiation intensity of the first anti-Stokes component at $\theta=\theta_1$, as seen from (17), has a finite limit as $l\to\infty$. This means that the considered radiation cone of the first anti-Stokes component is formed in the region of the focus of the fundamental component. At $\theta<\theta_1$, according to (17), there is no radiation of the first anti-Stokes component. This conclusion, however, is a consequence of taking the limit of an infinite active medium. Of course, when finite values of l are taken into account, the corresponding step in the intensity distribution becomes "smeared out," and therefore at $l\gg l_0$ the considered angular distribution will be only strongly asymmetrical with respect to the value $\theta=\theta_1$. Taking into account the finite values of l and the condition $\theta_1\gg\theta_0$ there appears in general, besides the indicated maximum at $\theta=\theta_1$, also a noticeable maximum at $\theta=0$ with angle width $\Delta\theta\sim\theta_0$.

We consider now the case of a focused first Stokes wave and a plane fundamental-component wave, i.e., we put $a_0^2=\infty$ and $l_0=\infty$. Here, obviously, we can put without loss of generality $\Delta\xi=0$, which in turn means that the cross section of the minimal radius of the beam of the first Stokes component is $z=z_0$.

In this case expression (9), for example for the field $u_2(\mathbf{r})$ of the second Stokes component takes, with allowance for (7), the form

$$u_2(\mathbf{r})=\frac{A_{21}}{2\pi rl_{-1}}\exp\left(-\frac{1}{4}k_{-2}^2a_{-1}^2\theta^2\right)\int\limits_{-z_0}^{l-z_0}d\zeta'\frac{\exp[i\chi_2(\theta)\zeta']}{1+i\zeta'/l_{-1}}, \tag{20}$$

where

$$A_{21}={}^1/_2\pi B_{-2}a_{-1}^2l_{-1}\tilde{e}_0^*\tilde{u}_1^2\exp(ik_{-2}r),$$
$$\chi_2(\theta)=-\Delta k_{-1}+{}^1/_2k_{-2}\theta^2(1-k_{-2}a_{-1}^2/2l_{-1}). \tag{21}$$

We see that expression (20) is of the same form as expression (15). Therefore the singularities of the angular distribution of the radiation intensity of the second Stokes component will be the same as those considered above for the first anti-Stokes component. In particular, for an unbounded active medium ($z_0=\infty$, $l-z_0=\infty$) we have

$$u_2(\mathbf{r})=\frac{A_{21}}{r}\exp\left[-\frac{1}{4}k_{-2}^2a_{-1}^2\theta^2-\chi_2(\theta)l_{-1}\right]\quad\text{at}\quad\chi_2(\theta)>0, \tag{22}$$

$u_2(\mathbf{r})=0$ at $\chi_2(\theta)<0$. This expression, at $\Delta k_{-1}l_{-1}\gg 1$, defines a radiation cone with apex angle $2\varphi_{-2}$, where

$$\varphi_{-2}=\left[\frac{2\Delta k_{-1}}{k_{-2}(1-k_{-2}a_{-1}^2/2l_{-1})}\right]^{1/2}, \tag{23}$$

and with total angle width $\Delta\varphi_{-2}$ corresponding to the intensity distribution

$$\Delta\varphi_{-2}\approx\frac{\ln 2}{2\varphi_{-2}k_{-2}l_{-1}(1-k_{-2}a_{-1}^2/2l_{-1})}. \tag{24}$$

In analogy with the preceding case of the first anti-Stokes component, this radiation cone is formed in the region of the focus of the first Stokes component.

Finally, for the field $v_1^*(\mathbf{r})$ of the first anti-Stokes component, starting from (9) and taking (7) into account, we obtain in the case under consideration

$$v_1^*(\mathbf{r})=\frac{A_{11}}{r}\frac{\sin[{}^1/_2\chi_1(\theta)l]}{\chi_1(\theta)l}\exp\left(-\frac{1}{2}k_1^2a_{-1}^2\theta^2\right), \tag{25}$$

where

$$A_{11}=B_1la_{-1}^2\tilde{e}_0^2\tilde{u}_1^*\exp[i(k_1r-\chi_1z_0+{}^1/_2\chi_1l)],$$
$$\chi_1(\theta)=-\Delta k_0+{}^1/_2k_1\theta^2(1+k_1a_{-1}^2/l_{-1}). \tag{26}$$

Expression (25) is analogous to (11). Therefore the angular distribution of the intensity of the first anti-Stokes component has in this case the same singularities as the angular distribution of the intensity of the second Stokes component in the preceding case. At $\Delta k_0 l \gg \pi$, the distribution of $|v_1(\mathbf{r})|^2$ has a sharply pronounced maximum of intensity for the value of the angle $\theta = \varphi_1$, where

$$\varphi_1=\left[\frac{2\Delta k_0}{k_1(1+k_1a_{-1}^2/l_{-1})}\right]^{1/2}. \tag{27}$$

The total angle width $\Delta\varphi_1$ of this maximum is equal to

$$\Delta\varphi_1\approx\frac{2\pi}{\varphi_1k_1l(1+k_1a_{-1}^2/l_{-1})}. \tag{28}$$

We see that the radiation intensity at $\theta = \varphi_1$ is proportional to l^2, the angle width is of the order of l^{-1}, and accordingly the power radiated in the considered cone is proportional to l. Outside the limits of the radiation cone, the angular distribution of the intensity has a structure in the form of oscillations in θ, the systematic component of which at $\theta_1 \gg \theta_{-1}$ has the maximum for $\theta = 0$ with total angle width $\Delta\theta \sim \theta_{-1}$.

3. POSSIBLE EXPLANATION OF RADIATION CONES OF CLASS II

Let us examine in greater detail expressions (13), (18), (23), and (27), which determine the angles at the apices of the cones of the predominant radiation of the considered components. In these expressions, besides the values of the wave vectors k_l, there enter also values of the ratios[3)] a_0^2/l_0 and a_{-1}^2/l_{-1}. For a Gaussian beam in a homogeneous medium we have $l_0 = k_0a_0^2$ and $l_{-1} = k_{-1}a_{-1}^2$, from which, recognizing that $\Delta k_\alpha \ll k_\alpha$, we get

$$\theta_{-2}=(k_0\Delta k_{-1}/k_{-2}k_{-1})^{1/2}\quad \theta_1=2(k_0\Delta k_0/k_1k_{-1})^{1/2},$$
$$\varphi_{-2}=2(k_{-1}\Delta k_{-1}/k_{-2}k_0)^{1/2},\quad \varphi_1=(k_{-1}\Delta k_0/k_0k_1)^{1/2}. \tag{29}$$

The values (29) for φ_{-2} and φ_1 coincide formally with the class-I radiation angles[2], and the values of (29) for θ_{-2} and θ_1 are seen to be different. If, however, the medium is nonlinear, then the field distribution in the region of the focus can in some cases be close to Gaussian, but generally with a different value of a_α^2/l_α as compared with the case of a linear medium, so that $l_\alpha = \nu_\alpha k_\alpha a_\alpha^2$, where ν_α in general differs from unity. A similar situation is realized in the region of the foci of a multi-focus structure.[17] It is therefore of interest to consider the expressions for θ_{-2}, φ_{-2}, θ_1, and φ_1 at values of ν_α different from unity.

As $\nu_\alpha \to \infty$ (strongly elongated caustic) we obtain the following expressions for the considered angles:

$$\theta_{-2}=\varphi_{-2}=\bar{\theta}_{-2}\equiv(2\Delta k_{-1}/k_{-2})^{1/2},\quad \theta_1=\varphi_1=\bar{\theta}_1\equiv(2\Delta k_0/k_1)^{1/2}. \tag{30}$$

These values differ from those defined by (29) by an approximate factor $\sqrt{2}$. If it is recognized that $\bar{\theta}_\alpha^2 \ll 1$, then we see that the radiation determined by (30) is of the Cerenkov type, analogous to that considered[6,15] from an active volume in the form of a filament of sufficiently large length.

For foci of a multifocus structure, the values of ν_α under different conditions can assume different values, such as $\nu_\alpha = 1-5$.[17] Consequently, the values of the angles of predominant radiation of the considered components, as seen from (13), (18), (23), and (27), can be different, depending on the concrete observation conditions.[4)] The intervals on the possible variations of φ_1 and φ_{-2} are in this case

$$\bar{\theta}_{-2}<\varphi_{-2}<2(k_{-1}\Delta k_{-1}/k_0k_{-2})^{1/2},\quad (k_{-1}\Delta k_0/k_0k_1)^{1/2}<\varphi_1<\bar{\theta}_1. \tag{31}$$

As seen from (31), the considered radiation has an intermediate character between the radiation of class I and radiation of the Cerenkov type. The obtained picture for the first anti-Stokes and second Stokes components in the considered case of a focused first Stokes component (see (31)) agrees with the experimentally observed picture of class II radiation. This picture was attributed in[11] to the presence of "filaments" in the fundamental component. The theory developed above shows that radiation of class II can be attributed to the presence of a multifocus structure in the first Stokes component. We note also that at $\nu_\alpha > 1$ the predicted values of the angles θ_{-2} and θ_1 corresponding to the multifocus structure in the fundamental component (for a plane first-Stokes component wave) fall in the intervals

$$(k_0\Delta k_{-1}/k_{-2}k_{-1})^{1/2}<\theta_{-2}<\bar{\theta}_{-2},\qquad \bar{\theta}_1<\theta_1<2(k_0\Delta k_0/k_1k_{-1})^{1/2} \tag{32}$$

and by the same token do not correspond to the experimentally observed quantities. The apparent reason for the the absence of cones of predominant radiation at angles θ_{-2} and θ_1 is that, as shown in[17], the multifocus structure in the fundamental component is usually produced under essentially nonstationary conditions (with respect to the SRS process) whereas the theory developed above is directly applicable only to the case of an SRS process that is stationary (quasi-stationary) in time. It should also be noted that for a focused wave of the fundamental component, even under conditions of quasistationary SRS processes, the validity of the results is limited to the case of a plane first-Stokes wave. In the case of a multifocus structure in the fundamental component, the last condition is usually likewise not satisfied, since the spatial coherence of the first Stokes component (in the absence of a multifocus structure in it) is as a rule insufficiently large. To realize the predicted radiation at angles θ_{-2} and θ_1 we can, for example, introduce into the investigated volume of the medium, from the outside, both the fundamental (focused) wave and the first Stokes component in the form of a plane wave with frequency close to the first Stokes frequency.

We note in conclusion, that the results obtained above provide also a number of examples (in addition to the results of[2,4,6]), which show the extent to which the angular distribution of the intensity of the anti-Stokes and higher-Stokes components of the SRS is sensitive to the concrete form of the distribution of the phases of the fields of the fundamental and first Stokes components, and by the same token afford an additional possibility of verifying that such simplified calculation methods as the plane-wave method are not justified.

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1)We note that the results of [6] are not quite correctly reflected in [7]. We note also that in [8] then considered parametric ("coherent four-photon") process with participation of an exciting, first Stokes, first anti-Stokes, and second Stokes components, as distinguished from similar processes with participation of only the exciting, first-Stokes and first anti-Stokes components, or else the exciting, first Stokes and second components. However, the main contribution ot the SRS intensity in liquids and solids is made by the last two processes (see [2]), whereas the first process and a number of other similar processes, interfering with the last two, give rise to corrections that do not play a principal role. We therefore, as usual, take into account the laser two processes.

2)We see that there is an analogy with the Cerenkov radiation of a particle passing through a layer of a medium of thickness l. However, the angle θ_{-2} differs in general from the Cerenkov value (see also Sec. 3). By the same token, the obtained picture can be regarded as modified Cerenkov radiation.

3)We note here that the radiation of anti-Stokes and higher Stokes components from a bounded active region with longitudinal dimension l and transverse dimension a, at different ratios of l/a, was considered in a simplified model in [19]. No focused beams, however, were considered in [19].

4)Simultaneous radiation of the considered component from several foci of a multifocus structure can lead to multiplicity of the radiation cones. As seen from (22) and (25), at $\Delta k_\alpha l_\alpha \lesssim 1$ the radiation can also be "diffuse" (i.e., without clearly defined cones). In some cases (for example in the case of simultaneous focusing of the fundamental and first Stokes component), radiation of "diffuse" type is possible also between distinctly defined external and internal cones.

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9