

and  $h_0$  agrees with the assumption that the experimentally observed dependence of  $\alpha$  on the direction of the light polarization is due to the magnetic action of the light wave on the quasiequilibrium system of magnetic moments of the film, which retains its detecting properties also in the optical band. It is possible that these properties are retained also at higher frequencies.

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#### OBSERVATION OF ULTRASHORT RADIATION PULSES IN STIMULATED SCATTERING OF LIGHT IN THE RAYLEIGH LINE WING

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We report in this paper the first experimental observation of ultrashort or picosecond radiation pulses (PP) produced by stimulated scattering of light in the Raleigh-line wing (SRW).

The possible occurrence of PP in the SRW process, with a duration on the order of the anisotropy relaxation time  $\tau$ , was pointed out earlier [2] by Fabelinskii and the present authors. This possibility has now been experimentally confirmed.

The SRW was excited by a giant pulse from a ruby laser [1] of power  $\sim 150 - 200$  MW, duration 10 - 15 nsec, and spectral width  $\sim 2 \times 10^{-2} \text{ cm}^{-1}$ . The exciting light was focused into a vessel with nitrobenzene 10 cm long, with focal distances  $f = 3$  and 1.5 cm.

Feedback was produced between the laser and the scattering medium. The scattered light was observed in the forward and backward directions.

The method of two-photon luminescence was used to observe the PP in the SRW [3]. A narrow beam of the scattered light passed through a vessel containing a solution of rhodamine-6G in ethyl alcohol and was reflected backwards from the mirror M (Fig. 1). Maxima of the two-photon luminescence should occur at the points of encounter of the forward PP and those reflected from the mirror. If there is only one such pulse, then the maximum luminescence should be observed at the mirror; if there are several, then a corresponding number of maxima are observable also at a definite distance from the mirror. The  $T = 2l/c$  between two PP can be determined from the distance  $l$  between the mirror and the first luminescence maximum, and the width  $\Delta l$  of this luminescence maximum yields the PP duration  $t_s = \Delta l/c$ , where  $c$  is the velocity of the light in the medium.

Ultrashort pulses in SRW were observed both in the direction of propagation of the exciting radiation, and in the opposite directions. Such pulses could be registered only

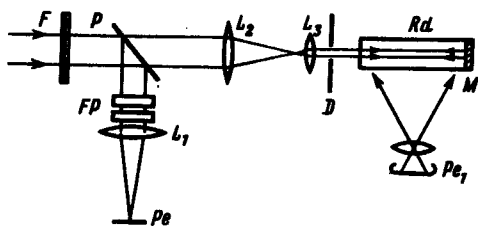


Fig. 1

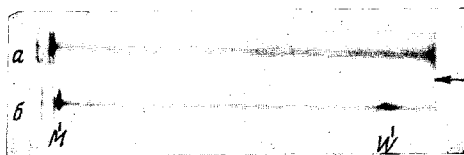


Fig. 2

Fig. 1. Setup for recording the spectrum and the picosecond pulses in stimulated scattering of light in the Rayleigh-line wing: F - interference optical filter ( $\lambda_{\max} = 6943 \text{ \AA}$ ), Rd - vessel with solution of rhodamine-6G in ethyl alcohol, M - mirror ( $R \sim 100\%$ ), Pl<sub>1</sub> - photocamera with green light filter, FP - Fabry-Perot interferometer.

Fig. 2. Photograph of traces of two-photon luminescence: a - exciting light ( $\lambda = 6943 \text{ \AA}$ ), b - light scattered by nitrobenzene. W - maximum of two-photon luminescence, showing the existence of picosecond pulses in the stimulated scattering of light in the Rayleigh line wing.

under experimental conditions such that the interferometer revealed an intense SRW spectrum in the form of a band with total width  $\sim 0.5 - 1.5 \text{ cm}^{-1}$ , adjacent on the Stokes side to the line of the exciting radiation (cf., e.g., [1, 4]).

Figure 2a shows part of the trace of the exciting radiation in the luminescent solution, while Fig. 2b shows part of the trace of the light scattered by nitrobenzene. We see on Fig. 2b a maximum of the two-photon luminescence produced at the point of encounter of at least two PP propagating in the direction of the mirror M and reflected from it. Consequently, the scattered light contained not less than two ultrashort pulses of duration  $t_s \leq 10^{-11} \text{ sec}$ .

When the exciting radiation was focused in the scattering medium by a lens with  $f = 3 \text{ cm}$ , the time between two neighboring pulses was  $T \approx 7 \times 10^{-10} \text{ sec}$ . When  $f$  was decreased by one-half,  $T$  also decreased by approximately one-half. Consequently, under the conditions of our experiment, when  $f$  is decreased by one-half the length  $L$  of the nonlinear interaction region in nitrobenzene, which is essential for the occurrence of PP in SRW, also decreased by one-half, since  $T \sim 2L/c$ . In this case we obtain  $L \sim 6 - 7 \text{ cm}$  for  $f = 3 \text{ cm}$  and  $L \sim 3 - 3.5 \text{ cm}$  for  $f = 1.5 \text{ cm}$ . It follows from these results that although the formation of the PP occurs principally in the focal region of the lens, much larger lengths  $L$  may be effective in the production of such a nonstationary effect, and in our case  $L \sim 2f$  or somewhat larger.

When PP is produced in SRW, the scattering process should have a nonstationary character, and for the case of plane waves and time intervals  $t \gg \tau$  it is described by the system of equations

$$\frac{1}{c} \frac{\partial I_0}{\partial t} + \frac{\partial I_0}{\partial x} = -D I_0 I_1, \quad (1)$$

$$\frac{1}{c} \frac{\partial I_1}{\partial t} - \frac{\partial I_1}{\partial x} = D I_0 I_1, \quad (2)$$

where  $I_0$  and  $I_1$  are the intensities of the beams of exciting and scattered light propagating towards each other, and  $D$  is the nonlinear-coupling constant

$$D = \frac{64\pi^2 \omega_0}{135n^4 c^2} (n^2 + 2) N \frac{(\alpha_1 - \alpha_2)^2}{kT} \frac{\Omega r}{1 + \Omega^2 r^2}. \quad (3)$$

Here  $\Omega = \omega_0 - \omega_1$ ,  $\omega_0$  and  $\omega_1$  are the frequencies of the exciting and scattered light,  $\alpha_1$  and  $\alpha_2 = \alpha_3$  are the principal polarizabilities of the molecule, and  $N$  is the number of molecules per  $\text{cm}^3$ .

The system (1) - (2) is identical with the system of equations describing stimulated Raman scattering of light [5], differing from the latter only in the expression for  $D$ . An analysis of this system of equations shows that at large values of the gain, when  $I_1$  becomes of the order of  $I_0$ , PP can be produced in the scattered light; these were observed earlier by Maier and Kaiser [5], and now by us in SRW.

It follows from the general solution of the system (1) - (2), when the boundary conditions a)  $x = 0$ ,  $I_0 = I_0(0)$  and b)  $x = L$ ,  $I_1 = I_1(L)$  are used, that the stimulated scattering may have a nonstationary character if the inequality  $I_1(L) \exp[DI_0(0)L] > (1/2) \times I_0(0)$  is satisfied. In this case several PP can be produced in the scattered light in SRW, their minimum duration is limited to the value  $(t_s)_{\min} \sim \tau$ , and their spectral broadening is  $(\Delta\Omega)_{\max} \sim 1/\tau$ .

The occurrence of PP explains why the SRW spectrum usually has the form of a broad band rather than the narrow line expected from the stationary theory. This explains also the competition between the SMBS and SWR [2, 4].

Short intense SRW pulses can themselves give rise to secondary SRW scattering (but not SMBS) in the nonlinear-interaction region, and this leads to an additional, possibly multiple, broadening of the scattered-radiation spectrum.

We note also that the occurrence of PP in scattered radiation may exert a strong influence on the kinetics of the development of self-focusing, by limiting the lifetime of the self-focusing filaments to the value  $T_1 \sim 2L/c$ , where  $L$  is the length of the filament.

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