STIMULATED FOUR-FIELD PARAMAGNETIC SCATTERING

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The possibility of transforming the frequency of powerful lasers by means of stimulated four-field scattering in transparent isotropic media is considered theoretically. The calculations show that for the frequency dependence of the refractive index encountered in these media the spatial relations can be satisfied in the range between $0.1\ \omega_1$ and $1.9\ \omega_1$. The scattering threshold for interaction between plane monochromatic waves in an ideal resonator is approximately 10^8 W/cm^2 provided that the nonlinearity is 10^{-13} CGSE. Possible nonlinearity mechanisms and their effect on the transformation process are discussed. Stimulated parametric scattering without a change of frequency due to the interaction of two opposite light beams from a ruby laser in a medium is observed experimentally. Cylindrical focusing optics and a resonator perpendicular to the axis of the initial beams were employed in the experiments. The power density of the scattered radiation was of the same order of magnitude as in the initial beam. Scattering was observed in a liquid, glasses, and an air-spark plasma.

1. INTRODUCTION

As is well known, the interaction of four waves in a nonlinear medium is described by a tensor of the fourth rank $\chi^{(3)}_{ijkl}(\omega_\alpha, \omega_\beta, \omega_\gamma, \omega_\delta)$. This tensor is responsible for a number of interesting effects, such as frequency tripling,[1,2] change of refractive index in a strong field,[3,4] Raman scattering,[1,2] and "scattering of light by light."[5] In this paper we shall consider only the process of transforming two initial "strong" waves with frequencies ω_1 and ω_2 into two "weak" waves with frequencies ω_3 and ω_4. This effect can in principle be used for the realization of four-field tunable parametric amplifiers and oscillators.

The possibility of such a transformation has not been discussed thoroughly in the literature before, evidently because of the assumed smallness of the inertialess electronic part of $\chi^{(3)}_{ijkl}$, which is equal approximately 10^{-15} to 10^{-16} CGSE.[2] Recent experimental[4,6] and theoretical[7] investigations show that in certain media it can be of the order of 10^{-12} to 10^{-13} and even 10^{-10} CGSE. This permits the hope that the threshold conditions for obtaining stimulated four-field parametric scattering may be fulfilled at reasonable flux densities of the initial laser radiation.

In certain special, particularly favorable cases four-field transformation of frequencies has already been observed experimentally: in [8], where three initial strong waves interacted, and in [9], where all four waves were resonant. Also, in [10] spontaneous scattering was observed in the interaction of only two initial strong waves.

In the case we are considering, the choice of frequencies ω_3 and ω_4 is limited by the conditions of temtemporal and spatial synchronism:

$$\omega_1 + \omega_2 = \omega_3 + \omega_4, \tag{1.1}$$

$$\mathbf{k}_1 + \mathbf{k}_2 = \mathbf{k}_3 + \mathbf{k}_4 + \Delta, \tag{1.2}$$

where the $\mathbf{k}_i$ are wave vectors and Δ is the wave detuning. A four-field oscillator would differ advantageously from the three-field one already realized[11] by the fact that the condition of spatial synchronism would be fulfilled over a rather wide range of tuning in isotropic media because of only one normal dispersion. If the dispersion of the refractive index $n(\omega_\alpha)$ is given analytically in the form[12]

$$n^2(\omega_\alpha) \approx 1 + f_0\omega_0 / (\omega_0^2 - \omega_\alpha^2), \tag{1.3}$$

where f_0 is a quantity proportional to the oscillator strength at frequency ω_0 (we are limited to only one such oscillator), then, by solving together Eqs. (1.1), (1.2), and (1.3) under the condition $\omega_1 = \omega_2$ and $\Delta = 0$, we can find, for given $\mathbf{k}_1$ and $\mathbf{k}_2$, all allowable directions $\mathbf{k}_3$ and $\mathbf{k}_4$ and the tuning range of the frequencies ω_3 and ω_4. In the most general case, when the angle $2\theta_0$ between vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ is arbitrary, the point of coincidence of vectors $\mathbf{k}_3$ and $\mathbf{k}_4$ slides in space over the surface of some body of revolution of the "dumbbell" type (tuning characteristic). The intersection of this surface with a longitudinal plane is described in polar coordinates (ω_3, θ) by the equation

$$\omega_3^2 n^2(\omega_3) + 4\omega_1^2 n^2(\omega_1)\cos^2\theta_0 - 4\omega_1\omega_3 n(\omega_1)\cos\theta_0\cos\theta = \omega_4^2 n^2(\omega_4). \tag{1.4}$$

If, for example, we use the experimental data on the dispersion in the cubic crystal NaCl, then for $\omega_1 \approx 2.7\times10^{15}$ sec^{-1} (frequency of the ruby laser) and $2\theta_0 = 0$, the allowable tuning range of the frequency ω_3 is between $0.1\ \omega_1$ and $1.9\ \omega_1$. The angle θ does not exceed 1–2° in this case. If $2\theta_0 = \pi$, the tuning range is zero, i.e., $\omega_1 = \omega_2 = \omega_3 = \omega_4$, and the angle θ can be anything. We therefore have the degenerate case of a parametric oscillator.

If a nonlinear change of the refractive index in a strong field is allowed for, the considered curves should be only slightly deformed. It is of significance in this case that when $2\theta_0 = 0$, degenerate scattering should even now be observed at some small angle $\theta_{\rm opt}$, the magnitude of which depends on the density of the strong field. This case of scattering has been observed, for example, in [5].

2. PARAMETRIC OSCILLATION IN A SEMIBOUNDED MEDIUM

Let the medium occupy the half-space $z \geq 0$. From the Maxwell equations in steady state it is easy to find, using standard procedures,[1] the reduced equations for the real, slowly-varying amplitudes A_α and phases φ_α of all four plane waves:

$$\frac{\partial A_{1,2}}{\partial z}+\delta_{1,2}A_{1,2}-\sigma_{1,2}A_{2,1}A_3A_4\sin\Phi=0,$$
$$\frac{\partial A_{3,4}}{\partial z}+\delta_{3,4}A_{3,4}+\sigma_{3,4}A_{4,3}A_1A_2\sin\Phi=0,$$
$$\frac{\partial\Phi}{\partial z}-\Delta_z+\Delta_z^{nl}-[\sigma_1A_1^{-1}A_2A_3A_4+\sigma_2A_1A_2^{-1}A_3A_4-\sigma_3A_1A_2A_3^{-1}A_4 - \sigma_4A_1A_2A_3A_4^{-1}]\cos\Phi=0. \tag{2.1}$$

In these equations

$$\Phi=\varphi_3+\varphi_4-\varphi_1-\varphi_2+\Delta_z z, \tag{2.2}$$
$$\Delta^{nl}=\Delta_3^{nl}+\Delta_4^{nl}-\Delta_1^{nl}-\Delta_2^{nl}, \tag{2.3}$$
$$\delta_\alpha=\alpha_\alpha/n(\omega_\alpha)\cos(\widehat{\mathbf{k}_\alpha\mathbf{z}_0}), \tag{2.4}$$
$$\sigma_\alpha=e^{i(\Delta_x x+\Delta_y y)}\beta_\alpha\omega_\alpha c/n(\omega_\alpha)\cos(\widehat{\mathbf{k}_\alpha\mathbf{z}_0}), \tag{2.5}$$
$$\Delta_\alpha^{nl}=\omega_\alpha c\sum_\beta\gamma_\alpha^{(\beta)}A_\beta^2/n(\omega_\alpha)\cos(\widehat{\mathbf{k}_\alpha\mathbf{z}_0}). \tag{2.6}$$

Here $\mathbf{z}_0$ is a unit vector along z; Δ_i is the i-th projection of the detuning vector; α_α (cm^{-1}) is the attenuation coefficient for wave ω_α;

$$\beta_\alpha=\frac{2\pi}{c^2}\mathbf{e}_\alpha\cdot\hat{\chi}^{(3)}(\omega_\alpha,\ \omega_\beta,\ \omega_\gamma,\ \omega_\delta)\vdots\mathbf{e}_\beta\cdot\mathbf{e}_\gamma\cdot\mathbf{e}_\delta; \tag{2.7}$$
$$\gamma_\alpha^{(\beta)}=\frac{2\pi}{c^2}\mathbf{e}_\alpha\cdot\hat{\chi}^{(3)}(\omega_\alpha,\ \omega_\alpha,\ \omega_\beta,\ \omega_\beta)\vdots\mathbf{e}_\alpha\cdot\mathbf{e}_\beta\cdot\mathbf{e}_\beta(2-\delta_{\alpha\beta}), \tag{2.8}$$

where $\mathbf{e}_\alpha$ is the unit polarization vector for wave ω_α; $\delta_{\alpha\beta}$ is the Kronecker symbol.

Equations (2.1) are suitable when all $|\cos(\widehat{\mathbf{k}_\alpha\mathbf{z}_0})|$ are sufficiently large. Otherwise, it is necessary to use an equation of the second order in z or to introduce derivatives with respect to x and y. We shall meanwhile assume that all $\cos(\widehat{\mathbf{k}_\alpha\mathbf{z}_0}) > 0$, and that the detuning Δ_z is compensated due to a nonlinear addition to the refractive index, i.e.,

$$\Delta_z=\Delta_z^{nl}, \tag{2.9}$$

which corresponds to optimum amplification. In the degenerate case, when $\omega_1 = \omega_2 = \omega_3 = \omega_4$ the optimum detuning

$$\Delta_z\approx\frac{12\pi\omega_1}{cn(\omega_1)}\chi(A_1^2-A_3^2)\approx\frac{12\pi\omega_1}{cn(\omega_1)}\chi A_1^2, \tag{2.10}$$

where

$$\chi=\mathbf{e}_1\cdot\hat{\chi}^{(3)}(\omega_1,\omega_1,\omega_1,\omega_1)\vdots\mathbf{e}_1\cdot\mathbf{e}_3\cdot\mathbf{e}_3.$$

From (2.10) and for $\chi > 0$, we obtain the angle for optimum amplification

$$\Theta_{\text{opt}}\approx\sqrt{12\pi\chi A_1^2}, \tag{2.11}$$

which agrees with [5].

It can be shown that for the given fields ($A_{1,2} \gg A_{3,4}$) the phase Φ tends to the stable value $-\pi/2$. Under these conditions it is easy to find the threshold condition from (2.1):

$$A_1A_2>\sqrt{\frac{\delta_3\delta_4}{2\sigma_3\sigma_4}}=\frac{c}{2\pi\chi}\sqrt{\frac{\alpha_3\alpha_4}{2\omega_3\omega_4}}. \tag{2.12}$$

For $\chi = 10^{-13}$ CGSE, $\omega_3 = \omega_4 = 2.7\times10^{15}$ sec^{-1}, $\alpha_3 = \alpha_4 = 10^{-2}$ cm^{-1}, $A_1 = A_2$, the intensity of initial radiation necessary for stimulated scattering will be of the order 100 MW/cm^2. We introduce the "numbers of quanta" $m = A_1^2 = A_2^2$, $p = A_3^2 = A_4^2$. Setting $\omega_1 = \omega_3$, we find the first integral of the system (2.1)

$$m(z)+p(z)=m(0)+p(0)=K. \tag{2.13}$$

From this it is easy to find also another integral

$$C|\cos\Phi|\rho(1-\rho)=1, \tag{2.14}$$

where $\rho = m/K$, and C is a constant of integration. The phase plane of the system is given by Eqs. (2.13) and (2.14). From the basic system of equations (2.1) it is seen that a change in fields occurs in a characteristic length

$$L=1/2\sigma_3A_1^2. \tag{2.15}$$

For a flux density of 100 MW/cm^2, $\chi = 10^{-13}$ CGSE, $\omega_1 = 2.7\times10^{15}$ sec^{-1}, we find from (2.5), (2.7), and (2.15) $\sigma_3 = 4.3\times10^{-8}$ CGSE and $L \approx 60$ cm. Since in practice it is difficult to produce such an interaction length, it is necessary to place a nonlinear medium in the resonator for effective transformation.

3. PARAMETRIC OSCILLATION IN A RESONATOR IN THE CASE $\cos(\mathbf{k}_\alpha\mathbf{z}_0) > 0$

Let a medium transparent to the pump wave at frequency $\omega_{1,2}$ be placed in a Fabry–Perot resonator of length l. We shall investigate the equations of the system, using the method of successive steps.[1] Since it is sufficiently well known, we present only the final results. For the designated fields the condition of self-excitation in the degenerate case for $\Phi = -\pi/2$ will be

$$R(0)R(l)\exp[\sigma_3p(0)l-2\delta_3l]>1, \tag{3.1}$$

where R(0) and R(l) are the coefficients of reflection of the mirrors. The time required for the buildup of the scattered waves to some specified level $m_N(l) = g_N m_0(0)$ will be

$$\tau_N=\tau_0\frac{\lg g_N}{\lg[R(0)R(l)]+(2\sigma_3p(0)-2\delta_3)l}, \tag{3.2}$$

where

$$\tau_0=ln(\omega_3)/c. \tag{3.3}$$

We consider also some special cases.

1. The resonator has identical parameters at frequencies ω_3, ω_4, i.e., $A_3(0) = A_4(0)$. The condition of self-excitation is

$$R(0)R(l)\exp[(\sqrt{\sigma_3\sigma_4}A_1A_2-2\delta_3)l]\geqslant1. \tag{3.4}$$

2. The resonator at frequency ω_4 is absent, i.e., $A_4(0) = 0$. Then

$$R(0)R(l)e^{-2\delta_3l}\operatorname{ch}(\sqrt{\sigma_3\sigma_4}A_1A_2l)\geqslant1. \tag{3.5}$$

Using the results of (2.13) and (2.14) we find that in the "degenerate" steady state (when $\delta_3 = \delta_4 = 0$), $m(l) = p(0)$. For excitation of oscillations it is necessary that the conditions for self-excitation be fulfilled and the pulse length of the pump be greater than τ_N. Calculations show that in the case of development of parametric oscillation from spontaneous thermal noise, the latter condition can be extremely important, since the length of the pulses of present-day powerful lasers—the pumps—is of the order of 10^{-8} sec.

We have been considering four-field parametric scattering in a nonresonant medium, i.e., the tensor $\chi^{(3)}_{ijkl}(\omega_\alpha, \omega_\beta, \omega_\gamma, \omega_\delta)$ was assumed real. In the case of resonance, for example $2\omega_1 = \omega_{10}$, where ω_{10} is a transition frequency, the magnitude of the tensor can increase by a factor of 10^4, as the simplest estimates show.[13] Such an increase, of course, lowers the threshold by a corresponding factor. We note that resonant four-field parametric scattering in this case arises as the interference of two two-photon processes (for $2\omega_1 = \omega_{10}$) and the generation is qualitatively different from generation in the nonresonant case. As estimates show,[13] the yield of radiation in the most favorable conditions can reach 20% with a relatively low threshold.

We remark that we assume the phase of the radiation of lasers to be stable, although it is random to some degree. Calculations in the limit of random phase (we omit them here) carried out by the method of Kirsanov, Selivanenko, and Tsytovich[14] or using the probabilities calculated from quantum transition theory[15] show that phase instability leads to an elevation of the generation threshold.

4. PARAMETRIC OSCILLATION IN A PLANE-PARALLEL PLATE WITH $\cos(k_4 z_0) < 0$

In the given case the waves A_3 and A_4 propagate in opposite directions. We consider the steady state of oscillation in a medium bounded by the planes $z = 0$ and $z = l$, with boundary conditions

$$A_3(0) = A_4(l) = A_0. \tag{4.1}$$

The behavior of the system will be described by the same equations (2.1), in which we now have to choose the appropriate signs in front of δ_4, σ_4, and Δ^{nl}_4. As before, we mutually compensate Δ and Δ^{nl}. Because of the different signs of Δ^{nl}_3 and Δ^{nl}_4, the magnitude of the detuning can now be zero. For the given fields A_1 and A_2 the phase Φ tends to the value $-\pi/2$. For $\delta_3 = \delta_4 = 0$ the solution of the system (2.1) has the following simple form:

$$A_3 = A_0 \frac{\cos[(z-l)\lambda] + \sqrt{|\sigma_3/\sigma_4|}\sin lz}{\cos \lambda l}, \tag{4.2}$$

where

$$\lambda = \sqrt{\sigma_3\sigma_4}\, A_1 A_2. \tag{4.3}$$

The solution for A_4 is similar.

We now turn our attention to the fact that for a certain critical length the denominator in (4.2) goes to zero. This means that the system is unstable in time and it is possible to excite oscillations in it, as in the case of a resonator. The threshold fields necessary to obtain amplification for $\delta_3, \delta_4 \neq 0$ can be found from the inequality

$$A_1 A_2 \geqslant \sqrt{(\delta_3 - \delta_4)^2 / 4(\sigma_3\sigma_4)}. \tag{4.4}$$

5. STIMULATED PARAMETRIC SCATTERING OF OPPOSITE WAVES AT HIGH ANGLES

Let us direct $\mathbf{k}_1$ and $\mathbf{k}_2$ along the z axis and $\mathbf{k}_3$ and $\mathbf{k}_4$ along the y axis (generally speaking, $\mathbf{k}_3 \uparrow\downarrow \mathbf{k}_4$ can be in any direction). Scattering will be described by the same system (2.1), in which the coordinate z in the equations for A_3 and A_4 is replaced by y. As in the preceding section, the difference from previous results will be only in the signs of δ_4, σ_4, and Δ^{nl}_4. Thus, the investigation of this case for the given fields with boundary conditions (4.1) was in fact already done in Sec. 4. The estimates are given in [16].

In the presence at the scattering frequency of a resonator with axis along y and quality Q, the waves A_1, A_2 and A_3, A_4 will form two standing waves $\tilde{A}_1$ and $\tilde{A}_3$. We shall assume both waves to be identically polarized along the x axis, and the wave $\tilde{A}_1$ as given. Then the equations for the real amplitude $\tilde{A}_3$ and phase Φ are

$$\frac{\partial \tilde{A}_3}{\partial t} + \frac{\omega_1}{2Q}\tilde{A}_3 = -2\pi\omega_1\chi^{(3)}_{1111}(\omega_1, \omega_1, \omega_1, \omega_1)\tilde{A}_1^2\tilde{A}_3 \sin\Phi,$$
$$\tilde{A}_3 \frac{d\Phi}{dt} = -4\pi\omega_1\chi_{1111}(\omega_1, \omega_1, \omega_1, \omega_1)\tilde{A}_1^2\tilde{A}_3\cos\Phi. \tag{5.1}$$

From these equations it is seen that Φ quickly tends to the stable value $-\pi/2$. The system will be self-excited at

$$\tilde{A}_1^2 > 1/4\pi\chi^{(3)}_{1111}Q. \tag{5.2}$$

For $\chi^{(3)}_{1111} = 10^{-13}$ CGSE, and $Q = 10^7$, this corresponds to a power density of ~ 50 MW/cm^2.

6. EXPERIMENTAL OBSERVATION OF STIMULATED FOUR-LEVEL PARAMETRIC SCATTERING

It has been possible to observe only degenerate scattering experimentally. The results of preliminary observations of such scattering in a direction transverse to the axis of the initial light beam were published in [16]. The anomalies of the frequency shifts of the scattered radiation reported in that paper indicate that four-level parametric processes were participating. Scattering without change of frequency was also seen in a similar geometry by Emmett and Schawlow[17] in the study of Mandel'shtam–Brillouin scattering at 90°. However, the authors of this paper explain the occurrence of the unshifted component by thermal Rayleigh scattering of the dissipative type,[18] not taking into account that for the excitation of this scattering large absorption coefficients are required. Possible mechanisms of nonlinearity leading to the excitation of degenerate parametric scattering will be discussed below.

In Sec. 4 it was shown that for the observation of degenerate scattering it is necessary to establish those optimal experimental conditions that permit the standing pump wave required by the theory to have been already present before, and not appear as a result of the mechanism of successive amplification of the 180° components of stimulated Mandel'shtam–Brillouin scattering (SMBS), as in [16]. This experimental configuration is shown in Fig. 1. It consists of two identical ruby lasers with saturable filters, which are coupled together by the radiation via focusing cylindrical lenses CL and CL′. The common focal line of these lenses is inside of

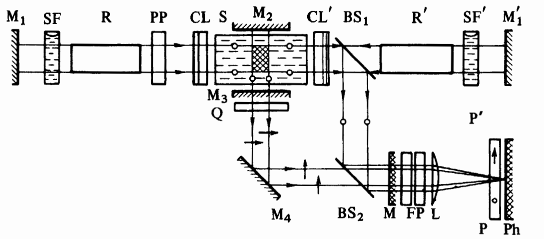

FIG. 1. Experimental arrangement: M_1, M_1', and M_2 –dielectric mirrors with 98% reflection at 6943 Å; SF and SF′–saturable filters; R and R′–ruby rods, 12 cm long; CL and CL′–cylindrical lenses; S–sample (in cuvette); M_3 –output mirror of the transverse resonator, reflection 50%; Q–quartz plate; M_4 –rotatable mirror; BS_1 and BS_2 –beam splitters; M–ground glass; FP–Fabry-Perot etalon; L–lens; P and P′–polaroid filters with mutually perpendicular directions of transmission (polarization of radiation is indicated by arrows and circles); Ph–photographic film.

a cuvette containing liquid (shown by the dashed lines). The left half of this two-branched laser could also work independently with the resonator formed by mirror M_1 and the plane-parallel plate PP, by screening off the right half. Thus, it was possible to produce both a standing and a traveling strong wave in the cuvette containing the nonlinear medium. For radiation scattered in the transverse direction there was a resonator formed by the mirrors M_2 and M_3, which could be rotated by a certain angle about an axis normal to the plane of the figure. The spectra of the scattered and initial radiation in a single pulse were compared in the Fabry–Perot etalon FP. The well-known polarization method was used to distinguish the spectra on the photographic plate.[17] Since in this configuration the polarizations of the radiations coincided (both polarizations perpendicular to the plane of the figure), a quartz plate Q was used to rotate one of them by 90°.

In pumping with a standing wave, stimulated scattering without a frequency shift was obtained in many transparent (to the ruby laser radiation) liquids and glasses: water, acetone, ethyl alcohol, glycerine, carbon disulfide, benzene, toluene, nitrobenzene, quinoline, the glasses K-8 and TF-6, fused quartz, etc. The most characteristic spectra are shown in Fig. 2. In the upper half of each of the frames (a, b, c, d, e) is shown the emission spectrum of the laser with its companion SMBS components; the spectrum of the stimulated scattering is in the lower half. Frames a and b were obtained with water; c, d, and e with the TF-6 glass. Pumping in cases a and d was by a traveling wave; in b, c, and e, by a standing wave.

It is seen in Fig. 2 that scattering at the fundamental frequency of the laser is missing when pumping is done with a traveling wave, as happened also in [16]. It appears only at the frequencies of the Stokes 180° components of SMBS (frames a and d). The scattered radiation is so intense that it excites in the transverse resonator its own 180° SMBS component (the next Stokes component in scattering).

In pumping with the standing wave, on the other hand, scattering occurs both at the fundamental frequency and at the frequencies of all the Stokes and anti-Stokes components of SMBS (frames b, c, and e). In certain cases, depending on the pump power, 90° components appear in the scattering in addition to the 180° SMBS components (frame b); these bear no relation to the four-field mechanism we are considering.

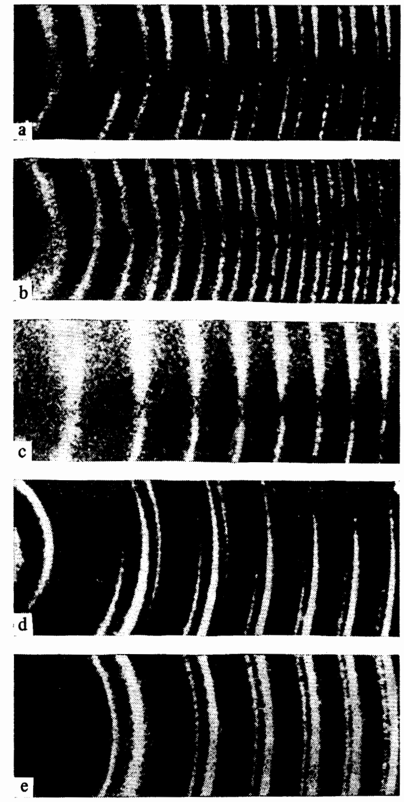

FIG. 2. Emission spectra: upper half of each frame is the pump spectrum, lower half, scattering spectrum; a–water pumped by traveling wave, etalon thickness 1 cm; b–the same, pumped by standing wave; c–glass TF-6, pumped by standing wave, etalon 1 cm; d–glass TF-6, pumped by traveling wave (higher radiation density), etalon 0.2 cm; e–the same, pumped by standing wave.

The most characteristic frame, which underlines the basic idea of the experiment, is frame c, which differs from frames d and e by higher pump power. SMBS is absent in this case. In pumping with a traveling wave at the same power, scattering was not observed in the transverse direction with this sample. The sample in both cases was destroyed in the interaction region. In the glass samples, in addition to destruction in the interaction region, there were also observed threadlike tracks of destruction along the axis of the transverse resonator up to 2 cm long; these are evidently associated with self-focusing. Scattering was also excited in the plasma flare from a breakdown in air which was formed by focusing the standing wave onto the surface of the glass sample directed along the axis of the transverse resonator. The spectrum of the scattering in this case was similar to the spectrum in Fig. 2c. Thus, the results of this experiment confirm the four-field mechanism of the observed stimulated scattering.

In using the transverse resonator the power density of the pump in the interaction region required to excite scattering was estimated to be about 10^8 to 10^9 W/cm^2. It was lowest in benzene, nitrobenzene, and acetone; highest in water. This agrees with the theoretical estimates made in Sec. 5. The maximum power we had available ($\sim$5 GW/cm^2 in the interaction region) was sufficient to excite scattering in the media at a low threshold without a resonator. There was no significant change in the character of the spectrum in this case (see also [16]).

Compared to the results of [16], the divergence of the scattered radiation with the use of the transverse resonator was greatly less, of the order of 1 to 2°. As the resonator was rotated about an axis perpendicular to the plane of Fig. 1 up to an angle of 10° (the maximum angle obtainable in the setup), the direction of the radiation maximum moved along with it. There were no noticeable changes in the spectrum and threshold as this was done.

In acetone, with maximum pumping, the total energy and pulse length of the scattered radiation were equal respectively to 0.4 J and 12 nsec. The laser pulse was about 16 nsec in length. The pulses coincided in time. An estimate of the flux density of the scattered radiation in the interaction region gave the magnitude $\sim 2-3$ GW/cm^2. For this estimate we used the area of the destroyed portion of the output mirror of the transverse resonator. Thus the transformation efficiency was approximately 40–50%.

7. ATTEMPTS TO OBSERVE SCATTERING WITH FREQUENCY CHANGE. DISCUSSION OF RESULTS

The most convenient variant of a parametric oscillator for the experimental observation of frequency transformation is evidently the one described in Sec. 4, when the pump waves $\mathbf{k}_1$ and $\mathbf{k}_2$ ($\omega_1 = \omega_2$) propagate in opposite directions at some angle $180° > 2\theta_0 > 90°$. The Stokes and anti-Stokes scattered waves $\mathbf{k}_3$ and $\mathbf{k}_4$ ($\omega_3 > \omega_1 = \omega_2 > \omega_4$) should propagate in this case toward each other along the z axis, and for them it is possible to use a single common resonator. In the limit, when the angle $2\theta_0 \to 180°$, we have a degenerate type of oscillator.

In the experiment the radiation of the ruby laser was reflected at some angle near to 180° from a mirror which was situated in the nonlinear medium under investigation. The direct and reflected (from the mirror) waves were in this case the pump waves $\mathbf{k}_1$ and $\mathbf{k}_2$. Observation of the scattering was conducted in the plane of incidence of the beams along the reflecting surface of the mirror. The mirrors of the transverse resonator were set up perpendicularly to this direction. To increase the density of the field in the interaction region the radiation of the laser was focused on the mirror by a cylindrical lens. In this it was presumed that the surface destruction observed at high power did not hinder the development of the scattered pulse, since the destruction usually sets in at the end of the laser pulse or even after it.[19]

As nonlinear media we used the liquids enumerated in the preceding section with highest $\chi^{(3)}_{ijkl}$, measurements of which were made in [4]. In spite of the fact that we used pump power densities an order of magnitude greater than that required by theory (10^9-10^{10} W/cm^2 compared to 10^8 W/cm^2, see Sec. 4), we did not see the expected scattering with frequency change. Only degenerate scattering with a spectrum corresponding to pumping by a traveling wave was excited in the transverse resonator. For certain angles $2\theta_0$ only an increase in the output and number of Stokes and anti-Stokes components of Raman scattering was noted. These angles are, evidently, the optimum ones for such scattering. It is known that Raman scattering may also be considered as a four-field process.

Let us analyze possible reasons for an elevation of the threshold for the excitation of stimulated four-field parametric scattering.

Spatial inhomogeneity of the amplitude and phase of the pump fields. In the theoretical calculations it was assumed that the pump is a plane monochromatic wave source. This is not actually true. As was shown in [20] the instantaneous intensity of the radiation of a pulsed ruby laser varies over the cross section of the generated beam by more than an order of magnitude, and the phase by more than 2π. In addition, they vary strongly during the course of the pulse. The average frequency of oscillation also changes. Even if we could remove all these instabilities, the ideal wave front would nevertheless be distorted by self-focusing.

Small magnitude of the inertialess part of $\chi^{(3)}_{ijkl}$. It is easily shown that the magnitude of the expected frequency change $\Delta\nu \le 1/\tau_r$, where τ_r is the relaxation time of the nonlinear mechanisms that participate in the frequency transformation. For a striction mechanism this time is of the order 10^{-10} sec, for an orientation mechanism, 10^{-11} sec.[6] Hence the limits of the change with the participation of these mechanisms are limited by the frequencies of the SMBS components and of the wing of the Rayleigh line. The temperature (entropy) mechanism of nonlinearity can give a frequency change only within the limits of the line width because of the slow velocity of the entropy waves.[21]

In Sec. 5, for the estimate of the threshold, we used the results of measurements of $\chi^{(3)}_{ijkl}$ accomplished earlier.[4] The fact that in the case of degenerate scattering, when all nonlinear mechanisms can participate, the estimate of the threshold was more or less correct, is evidence of the validity of the results of these measurements. However, the question about the magnitude of the nonlinear electronic susceptibility still remains open, evidently. The experimental conditions in [4] were such that the striction part of the nonlinear susceptibility was excluded from the measurements. The static-field experiment described there also says that the orientational part is small. In the arrangement, however, the entropy part could have been registered along with the electronic part.

Competition of other types of scattering with a lower threshold. SMBS and stimulated Raman scattering have a lower threshold. In the excitation of SMBS up to 80% of the pump radiation can be reflected from the interaction region,[22] which will inhibit the concentration of a sufficiently high pump field density.

For these reasons, nonresonant excitation of parametric four-field generation with a frequency transformation is obviously a rather difficult experimental problem.

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