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### PARAMETRIC AMPLIFIERS AND GENERATORS OF LIGHT

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# 1. INTRODUCTION. THE PROBLEM OF CREATING TUNABLE GENERATORS OF COHERENT OPTICAL RADIATION

ONE of the most important problems of laser physics is to extend the frequency range of generators of coherent optical oscillations. Many possibilities uncovered by the invention of lasers remain unrealized because coherent optical emission generators, which employ single-photon transitions in inverted quantum systems, can operate in principle only at a relatively small number of rigidly fixed frequencies. The foregoing pertains in particular to powerful generators, which make it possible to carry out research in probably one of the most interesting field of scientific research and application, namely nonlinear optics. Indeed, power on the order of 10-100 MW, at which strong nonlinear effects are produced in solids, liquids, and gases, can be obtained at the present time only with the aid of ruby lasers ( $\lambda \approx 0.7 \mu$ ) and with neodymiumglass lasers ( $\lambda \approx 1.06 \,\mu$ ). Although many important researches on the features of the interaction between powerful radiation matter have been carried out with these lasers, they must obviously be regarded only as the first step in the development in this field of physics. So far there has been very little study of resonant nonlinear interactions in the visible, ultraviolet, and especially in the infrared. The latter is of particular interest, for it is precisely in the infrared range where the resonant vibrational frequencies of molecules lie. Therefore we can expect that powerful generators of coherent infrared radiation will exert very strong action on substances. Some progress in the extension of the range of powerful coherent optical oscillations was reached by means of nonlinear optics itself. Mention should be made here first of work on the generation of harmonics [1-4] and on the stimulated Raman scattering (see, for example, [5,6] and also the reviews of [47]), which have made it possible (by using various combinations of these effects) to cover by means of a set of discrete lines the range from 0.26 to  $1.1 \mu$  at powers not lower than 100 kW-5 MW in individual lines. The possibility of using these lines for physical research was demonstrated, for example, in [4,6-9]. At the same time it must be emphasized that such frequency converters for laser energy are far from capable of solving the problem. Indeed, the problem of covering the optical band with generators of greater oscillations can be regarded as solved only when the frequencies

of the coherent oscillations will be made continuously variable just as, for example, in the microwave band of the electromagnetic spectrum. It is obvious that only if tunable generators of powerful coherent radiation become available will it be possible to investigate fully and to realize the capabilities of the strong action of magnetic radiation on matter. It must also be emphasized that the development of continuously tunable generators of coherent optical radiation (especially continuously-tunable continuous-emission generators) can revolutionize also the experimental techniques used in linear optics: it can greatly increase the accuracy of the absorption spectral analysis. light measurements, etc.

An effective method for producing continuously tunable generators of coherent optical radiation is the use of the so-called parametric interactions of light waves in an optically transparent medium. The possibility of producing parametric light generators that can be continuously tuned in a wide range, and data on the tuning devices, were predicted by the authors<sup>[10]</sup> and by Kroll<sup>[11]</sup> in papers published in 1962; questions concerning parametric amplification and generation in the optical band were discussed also by  $Kingston^{[12]}$ . Somewhat later Siegman<sup>[13]</sup> proposed to use a parametric generator as an active amplitude limiter for optical oscillations. The theory of parametric amplifiers and light generators was developed in greater detail in papers  $[14-16,48]^*$ . The first result of experiments in which it was possible to observe parametric amplification and generation of optical waves was reported at the All-union Symposium on Nonlinear Optics in Minsk  $(4-11 \text{ June, } 1965^{[17]})$  and the Conference on Quantum Electronics in San Juan, Puerto Rico (28-30 June 1965; see [8, 19-21]). The operating principle of parametric amplifiers and light generators produced to date consists in the following. In an optically transparent nonlinear medium, whose polarization **P** is

$$\mathbf{P} = \mathbf{x}\mathbf{E} + \hat{\mathbf{y}}\mathbf{E}\mathbf{E} \tag{1}$$

(here  $\kappa$  is the linear susceptibility and  $\hat{\chi}$  is the nonlinear susceptibility of lower order), the energy of a powerful light wave (so-called pump of frequency  $\omega_p$ ) can be transferred by means of weak oscillations of frequencies  $\omega_1$  and  $\omega_2$ , satisfying the relation

<sup>\*</sup>We note that the theory of wave interactions of the parametric type as applied to microwave devices was developed in [<sup>33,36</sup>].

(2)

(3)

$$\omega_{\mathbf{p}} = \omega_1 + \omega_2.$$

This can be readily verified by representing, in accordance with (1), the action of the pump wave on the nonlinear medium as a modulation of its dielectric constant in accordance with the traveling-wave law:

$$\begin{split} \boldsymbol{\varepsilon} \left( \boldsymbol{\omega}, \ \mathbf{r} \right) &= \boldsymbol{\varepsilon}_0 \left\{ 1 + m \left( \boldsymbol{\omega}_{\mathbf{p}} \right) \left[ \exp \left( i \left( \boldsymbol{\omega}_{\mathbf{p}} t - \mathbf{k}_{\mathbf{p}} \mathbf{r} \right) \right) \right. \\ &+ \exp \left( - i \left( \boldsymbol{\omega}_{\mathbf{p}} t - \mathbf{k}_{\mathbf{p}} \mathbf{r} \right) \right) \right] \right\}, \end{split}$$

In a nonstationary medium, whose properties are described by formula (3), the waves at frequencies  $\omega_1$  and  $\omega_2$  satisfy (2)

$$\mathbf{E}_{i} = \mathbf{A}_{i} \exp \left[ i \left( \omega_{1} t - \mathbf{k}_{i} \mathbf{r} \right) \right] + \mathbf{c.c.},$$

$$\mathbf{E}_{2} = \mathbf{A}_{2} \exp \left[ i \left( \omega_{2} t - \mathbf{k}_{2} \mathbf{r} \right) \right] + \mathbf{c.c.}$$

$$(4a)$$

no longer propagate independently, as would be the case in a linear medium with constant parameters, but interact with each other. Indeed, the induction  $D(\omega_1)$  at the frequency  $\omega_1$  is

$$\mathbf{D}(\omega_{1}) = \varepsilon_{0}\mathbf{E}_{1} + \varepsilon_{0}m\mathbf{A}_{2}^{*}\exp\left\{i\left[\omega_{1}t - (\mathbf{k}_{p} - \mathbf{k}_{2})\mathbf{r}\right]\right\}, \quad (5a)$$
  
$$\mathbf{D}(\omega_{2}) = \varepsilon_{0}\mathbf{E}_{2} + \varepsilon_{0}m\mathbf{A}_{1}^{*}\exp\left\{i\left[\omega_{2}t - (\mathbf{k}_{p} - \mathbf{k}_{1})\mathbf{r}\right]\right\}. \quad (5b)$$

The second terms in (5) characterize the interaction of the waves in a medium with variable parameters; it is obvious that the interaction will be maximal (will accumulate with increasing distance) if these terms have the same spatial periodicity as the first terms in formulas (5), that is, if

$$\mathbf{k}_{\mathbf{p}} = \mathbf{k}_{\mathbf{i}} + \mathbf{k}_{\mathbf{2}}.\tag{6}$$

The meaning of the last condition can be easily understood: it is equivalent to stipulating that the phase relations (and consequently also the character of the energy relation) between the waves be maintained constant over the entire extent of the nonlinear medium. Condition (6) is frequently also called the synchronism condition.

When conditions (2) and (6) are satisfied, the nonstationary medium performs work on the waves at frequencies  $\omega_1$  and  $\omega_2$ , whose amplitudes then increase. To determine the growth rate of these waves, let us consider for simplicity the scalar problem. (A more accurate and more consistent analysis will be deferred to Sec. 2.) We consider a semibounded medium with variable parameter (3), direct the z axis along the pump-wave vector  $\mathbf{k}_p$  and along the normal to the separation boundary; then, obviously, the amplitudes  $A_1$  and  $A_2$  can depend only the coordinate z. Substituting in Maxwell's equations

$$\Delta E - \frac{1}{c^2} \frac{\partial^2 D}{\partial t^2} = 0, \qquad (7)$$
$$D = \varepsilon E \qquad (8)$$

the field E in the form

$$E = A_1(z) \exp \left[i \left(\omega_1 t - \mathbf{k}_1 \mathbf{r}\right)\right]$$

$$+ A_2(z) \exp \left[i \left(\omega_2 t - \mathbf{k}_2 \mathbf{r}\right)\right] + \text{c.c.}$$
(9)

and using (2), (3), and (6), we obtain a system of two

coupled wave equations for the fields  $E_1$  and  $E_2$ . The latter can be greatly simplified by using the fact that for the cases of practical interest the coefficient of modulation of the dielectric constant is small (m =  $4\pi\chi A_p/\epsilon_0 \approx 10^{-6}-10^{-6}$ ) and consequently the variations of the complex amplitudes  $A_1$  and  $A_2$  over the wavelength are small. This enables us to write

$$\frac{d^2A_i}{dz^2} \ll k_i \frac{dA_i}{dz} , \qquad (10)$$

and consequently, the system of second-order equations can be replaced by a simplified system of first order equations

$$\frac{dA_1}{dz} = -\frac{imk_1}{2\cos\left(\widehat{\mathbf{k}_1\mathbf{k}_p}\right)}A_2^*, \quad \frac{dA_2^*}{dz} = \frac{imk_2}{2\cos\left(\widehat{\mathbf{k}_2\mathbf{k}_p}\right)}A_1, \quad (11)$$

which reduces to a single second-order equation

$$\frac{d^2A_1}{dz^2} = \frac{m^2k_1k_2}{4\cos\left(\widehat{\mathbf{k}_1\mathbf{k}_p}\right)\cos\left(\widehat{\mathbf{k}_2\mathbf{k}_p}\right)} A_1.$$
(12)

Equation (12) has obviously solutions which increase exponentially with the coordinate z

$$A_1(z) = A_1(0) e^{\Gamma z}, \qquad A_2(z) = A_2(0) e^{\Gamma z}, \tag{13}$$

where the growth increment is

$$\Gamma = \frac{1}{2} \sqrt{\frac{m_1 m_2 k_1 k_2}{\cos\left(\widehat{\mathbf{k}_1 \mathbf{k}_p}\right) \cos\left(\widehat{\mathbf{k}_2 \mathbf{k}_p}\right)}}$$

The foregoing signifies that the stationary medium, whose properties are described by (3), acts as an amplifier for the waves at frequencies  $\omega_1$  and  $\omega_2$ . However, if we place mirrors in the paths of these waves (with amplitude reflection coefficients  $R_1$  and  $R_2$ ) in such a way that the corresponding optical cavities resonate at frequencies  $\Omega_1 \cong \omega_1$  and  $\Omega_2 \cong \omega_2$ , then oscillations can become self-excited in the medium, by virtue of (13) at frequencies  $\omega_1$  and  $\omega_2$  if (the case of exact resonance  $\Omega_1 = \omega_1$  and  $\Omega_2 = \omega_2$ )

$$R_1(\omega_1) R_2(\omega_1) e^{\Gamma d} > 1, \quad R_1(\omega_2) R_2(\omega_2) e^{\Gamma d} > 1.$$

For  $\Gamma d \ll 1$  the last conditions can be written in the form of a single inequality which has a very lucid form

$$\frac{m}{2} > \frac{1}{\sqrt{Q_1 Q_2}}, \qquad (14)$$

where

$$Q_i = \frac{k_i l_i}{1 - R_1(\omega_i) R_2(\omega_i)}$$

are the Q-factors of the resonators at frequencies  $\omega_{1}$  and

$$l_i = \frac{d}{\cos{(\mathbf{k}_i \cdot \mathbf{k}_p)}}$$

are the lengths of the corresponding resonators.

A very important circumstance here is that at a fixed pump frequency, as follows from the foregoing analysis, the frequencies  $\omega_1$  and  $\omega_2$  can generally speaking be arbitrary. Thus, use of parametric interactions makes it possible in principle to convert co-



herent oscillations at a fixed frequency (for example, coherent oscillations of a ruby laser, a neodymiumglass laser, or their harmonics) into coherent oscillations with tunable frequency.

Using (14) we can readily estimate the pump power necessary to excite parametric oscillations. Putting  $\chi \cong 3 \times 10^{-9}$  cgs esu (this value is characteristic of the KH<sub>2</sub>PO<sub>4</sub> crystal which is extensively used in non-linear optics) and  $Q_{1,2} \cong 10$ , and recognizing that m =  $4\pi\chi A_p/\epsilon_0$ , we find the threshold pump field corresponding to satisfaction of the self-excitation condition a value  $A_p = 10^2$  cgs esu, that is, the pump power flux  $P_p = \frac{c}{8\pi} A_p^2$  corresponding to the threshold of parametric excitation should be several MW/cm<sup>2</sup>. Such power levels can be readily obtained with modern lasers.

The phenomenon of amplification of waves at frequencies  $\omega_1$  and  $\omega_2$  in a medium with variable parameters has much in common with parametric resonance in a system of two cavities tuned to frequencies  $\Omega_{1,2}$ and coupled by a capacitance that varies like C =  $C_0[1 + m \cos \omega_p t]$ , where  $\omega_p \cong \Omega_1 + \Omega_2$ , a case very thoroughly studied in the theory of oscillations. The time variation of the oscillation amplitudes of such resonators is described by equations of the type (11) (see, for example, [22]), so that the process of amplification of the waves (4), which develops in space, has a perfectly clear temporal analog. Such an analogy can make it easier for the radiophysicist and the radio engineer, who operate with circuits with lumped parameters, to understand the nonlinear optical processes; for a more detailed discussion see <sup>[23]</sup>. Therefore, if we use the language of radiophysics, a nonlinearity of the type (1) can be called reactive; it is analogous to a nonlinear capacitance or inductance, justifying the use of the term "parametric" for the optical phenomena considered here. We note, finally, that condition (14) has the same form as the condition of parametric excitation of a two-loop circuit with lumped constants. Yet attention must be called to an important circumstance, which noticeably distinguishes optical parametric phenomena from the corresponding phenomena in circuits with lumped constant. In optics,

FIG. 1. One-dimensional parametric interaction in a uniaxial crystal and diagram of tunable parametric light generator using such an interaction.

Figure a) shows, in the first quadrant of the z', x' plane (z' is the optical axis), the cross sections of the surfaces of the wave numbers  $\mathbf{k}_1^{(0)}$ ,  $\mathbf{k}_2^{(0)}$ (solid circles), and  $\mathbf{k}_p^{(e)}$ . The point of intersection of circle radius  $\mathbf{k}_1^{(0)} + \mathbf{k}_2^{(0)}$  (dash) with the curve  $\mathbf{k}_p^{(e)}$  ( $\theta$ ) defines the direction along which  $\mathbf{k}_1^{(0)} + \mathbf{k}_2^{(0)} = \mathbf{k}_p^{(e)}$ . Figure b) shows schematically a tunable generator and two methods of frequency tuning (by rotating the crystal or with a static electric field).

parametric excitation has a wave character, and therefore its variation is determined essentially not only by the temporal (frequency) but also by the spatial relations: to obtain self-excitation of parametric oscillations in the optical band, it is necessary to have not only "optical" tuning of optical resonators, but also to satisfy relation (6) ("wave tuning") between the wave vectors, thus imposing rather stringent requirements on the dispersion properties of the medium. Satisfaction of the synchronism condition (6) during the frequency tuning of an optical parametric generator is in general one of the technical problems which arise when working with such generators. Of course, even in frequency tuning of resonators of an optical parametric generator there are certain problems specific of this band and connected with the multimode character of the pump (actually, usually one excites in place of (3) the entire spectrum of waves of the dielectric constant), and the multimode nature of the oscillations in the optical resonators.

The requirement imposed by (6) on the dispersion properties of the medium can be easily determined, considering for simplicity the so-called degenerate regime of the parametric generator, in which  $\omega_1 = \omega_2$  $= \omega = \omega_p/2$ . We can then conclude from (6) that for effective interaction of the waves the medium should have anomalous dispersion. We can write for the refractive indices, if (6) is satisfied,

$$n_1(\omega) > n_2(2\omega). \tag{15}$$

This, as is well known, is also the condition for effective generation of optical harmonics (see  $[1^{-4}]$ ). In the region of optical transparency, the anomalous dispersion can be imitated by allowing waves of different polarization to interact in an anisotropic medium. This circumstance was established by Giordmaine<sup>[24]</sup> and by Terhune and co-workers<sup>[25]</sup>; its use is essentially the basis of many successes attained in experimental nonlinear optics. Figures 1 and 2 show possible variants of accumulating parametric interactions in uniaxial negative crystals (z' and x' are the symmetry axes of the crystal) and the circuits corresponding to the parametric light generators.

The interaction shown in Fig. 1,a can be called ac-

FIG. 2. Two-dimensional parametric interaction in a uniaxial crystal and diagram of tunable parametric generator employing such an interaction.

Figure a) (plotted for the case of a degenerate parametric interaction  $\omega_1 = \omega_2 = \frac{\omega_p}{2}$ ) shows the sections of the surfaces of the wave vectors  $k_1^{(0)}$  and  $k_p^{(e)}/2$  in the first quadrant of the x', z' planes. In figure b) is shown the diagram of the parametric generator with two-dimensional interaction. The frequency is tuned here by varying the positions of the optical resonators relative to the crystal.

cumulating one-dimensional parametric interaction  $(\mathbf{k}_1 \parallel \mathbf{k}_2 \parallel \mathbf{k}_D)$ . If the double refraction in the crystal is sufficiently large, and the dispersion is relatively weak, then anomalous dispersion may be simulated, for example, upon interaction of the extraordinary waves at the higher frequencies with the ordinary ones at the lower ones. Figure 1,a shows a method of graphically determining the directions in which the ordinary waves at frequencies  $\omega_1$  and  $\omega_2$  can increase exponentially in the field of an extraordinary pump wave; along this direction  $\mathbf{k}_1^O + \mathbf{k}_2^O + \mathbf{k}_p^O$  (the indices o and e will denote here and throughout the ordinary and extraordinary waves). Figure 1,b shows a tunable parametric generator and light amplifier in which such a one-dimensional interaction is used. The frequency and direction of the wave vector of the pump wave are fixed here. The mirrors R<sub>1</sub> and R<sub>2</sub> have simultaneously a high reflection coefficient at frequencies  $\omega_1$ and  $\omega_2$ ; the resonance at the pump frequency is not essential (at sufficiently high values  $m \sim Q^{-1/2}$  one can forego also the use of a resonator at one of the generated frequencies).

The generated frequencies can be tuned by rotating the crystal in the manner shown in Fig. 1,b (of course, this should not bring the resonator out of adjustment; the corresponding technical details are not shown in the figure), by applying a static electric field which changes, as a result of the electro-optical effect, the optical properties of the crystal (this method was proposed in [10]; it was used in [26,27] for modulation of optical harmonics), and finally, by changing the temperature of the crystal (see [20]). Figure 2, a shows the diagram of an accumulating parametric interaction which can be called two-dimensional, while Fig. 2,b shows the corresponding parametric generator. As before, the waves at frequencies  $\omega_1$  and  $\omega_2$  are ordinary waves, and the pump wave is extraordinary, but the vectors  $\mathbf{k}_1^{O}$ ,  $\mathbf{k}_2^{O}$ , and  $\mathbf{k}_p^{O}$  are not parallel. We note that this choice of wave types is not obligatory. In many crystals the synchronism condition (6) can be satisfied also in the case when the wave at frequency  $\omega_1$  is ordinary and extraordinary waves are excited at frequencies  $\omega_2$  and  $\omega_p$ , that is, the satisfaction of the



condition  $\mathbf{k}_1^{O} + \mathbf{k}_2^{e} = \mathbf{k}_p^{e}$  is possible. In the case of twodimensional action the resonators for the frequencies  $\omega_{1,2}$  are separated in space and the tuning is effected by varying the position of the resonators relative to the position of the crystal (the frequency and position of the wave vector of the pump, as in Fig. 1, are assumed fixed). In the papers cited above, both indicated types of parametric interaction were effected.

One-dimensional parametric interactions in the crystals KH<sub>2</sub>PO<sub>4</sub> (KDP) and LiNbO<sub>3</sub> (lithium niobate) are reported in [17,19,20], and two-dimensional parametric interaction in a KDP crystal in <sup>[18]</sup>. The amplification realized in <sup>[17,20]</sup> was sufficient to satisfy the self-excitation condition (14) for parametric oscillations and, consequently, to trigger the tunable light generator. In the parametric generator described by Giordmaine and Miller<sup>[20]</sup>, using the highly efficient</sup>nonlinear crystal LiNbO<sub>3</sub>, the wavelength of the parametric oscillations varied from 0.97 to  $1.15 \mu$ ; the pump wavelength was  $\lambda_{\rm D} = 0.529 \,\mu$  (the pump was the second harmonic of a calcium-tungstate laser activated with neodymium). The experimentally determined threshold power of the pump, corresponding to the self-excitation of the oscillation was  $P_p = 4$  $imes 10^5$  W/cm<sup>2</sup>, and the efficiency of the generator was  $\eta = P_{1,2}/P_p \approx 0.1\%.$ 

The parametric generator operating in our laboratory, based on a scheme described in <sup>[17]</sup>, used a KDP crystal; by virtue of the lower value of  $\chi$  in this crystal, the threshold pump power (the pump generator used in <sup>[17]</sup> had  $\lambda_p = 0.529 \,\mu$ ) was approximately 12  $\times 10^6$  W/cm<sup>2</sup>; the frequency tuning was by rotating the crystal and had a range ~ 600 Å; the powers P<sub>1,2</sub> reached 200-300 W; the range of frequencies was limited by the properties of the mirrors. Parametric interaction was observed in <sup>[18,19]</sup> in a field of a pump generator with  $\lambda_p = 0.35 \,\mu$  (second harmonic of ruby-laser emission) and, in <sup>[21]</sup> with the aid of gas lasers (to be sure, the pump power in these experiments remains still appreciably below threshold). The results of these investigations thus offer evidence that the present-day level of laser technology makes feasible tunable generators of coherent optical emission on an

experimental basis. At the same time, these results should be regarded as preliminary, merely illustrating the possibilities of experimental realization of the principle of parametric amplification and generation in the optical band, but by no means exhausting all the possibilities mentioned here. A discussion of these possibilities, based on a more complete and more consistent theory, will be presented in Sec. 2, where we shall also discuss in greater detail the experimental results.

It should be noted that in a strong pump field the parametric interaction can occur not only between the light waves themselves, but also between light waves and acoustic waves, light waves and spin waves, etc. Processes of this kind are called induced scattering. The wave picture of the processes of induced scattering is perfectly analogous to the wave picture of the parametric amplification discussed above. Therefore, many results of the theory of wave parametric interactions are directly applicable to induced scattering. An examination of the induced-scattering phenomena from the point of view of "parametric" concepts, on the other hand, makes it possible to analyze the possibility of creating tunable generators for hypersound, using optical pumping (using induced Mandel'shtam-Brillouin scattering in crystals cooled to liquid-helium temperature), parametric generators for millimeter and submillimeter wavelengths with optical pumping, etc. It is interesting that in some cases intense coherent oscillations in a scattering medium, excited in the course of induced scattering of intense optical radiation, can themselves serve as a pump for lowerfrequency of electromagnetic waves, acoustic waves, etc. (see, for example, <sup>[28]</sup>).

A brief discussion of these questions is the subject of Sec. 3 of the present article.

#### 2. AMPLIFICATION AND GENERATION OF SUB-HARMONICS IN THE OPTICAL BAND

In this section we discuss first in greater detail the theory of parametric interactions in the optical band. In the general case, we must take into account here quantum effects; we note, incidentally, that when (2) and (6) are multiplied by Planck's constant they acquire a clear quantum meaning-the first of these relations becomes the energy conservation law and the second the momentum conservation law. Therefore, in quantum language, the process of parametric amplification can be treated as the process of coherent decay of the pump photons following interaction with the photons of frequencies  $\omega_1$  and  $\omega_2$ . However, most problems connected with optical parametric processes can be solved in the quasiclassical approximation, in which the field is not quantized and quantum theory is used only to investigate the nonlinear polarization of the medium. Exceptions are problems involving the noise properties of parametric amplifiers and the

pre-oscillation noise of parametric generators; for lack of space, however, we shall not discuss these questions. An analysis of quantum fluctuations in parametric processes is given in <sup>[29-32]</sup>.

# 2.1. Principles of the Theory of Parametric Light Generators

We first write out the equations for the parametric action of traveling light waves in a more general form than in Sec. 1. The need for such a generalization is connected with the following circumstance. The ideas concerning a medium with variable parameters, used in Sec. 1, have obviously a limited region of application. They can be used only so long as the amplitude of the pump wave greatly exceeds the amplitude of the wave at frequencies  $\omega_1$  and  $\omega_2$  (A<sub>p</sub>  $\gg$  A<sub>1</sub>, A<sub>2</sub>). At the same time, the exponential growth of the amplitudes  $A_1$  and  $A_2$  in the medium with polarization of the type (1) can be limited only by the reaction of the waves  $A_1$ and  $A_2$  on the pump wave. Therefore, in constructing a theory of parametric light generator, one of the most important problems of which is the determination of the efficiency of the generator, the waves at the frequencies  $\omega_1$ ,  $\omega_2$ , and  $\omega_p$  should be considered on a par. The presence of feedback, which is determined by the mirrors, makes it necessary, in general, to take into account the change in the complex amplitudes both in space and in time. Finally, in constructing a more exact theory it is necessary to take into consideration the redistributed losses in the nonlinear medium (connected with the imaginary part of the linear susceptibility  $\kappa$ ) and the possible deviation from the exact synchronism condition (6). Taking the foregoing into account, we seek the solution of the nonlinear Maxwell's equations in an anisotropic medium (as shown in Sec. 1, this is precisely the case of greatest interest)

where

$$\mathbf{P}^{(1)} = \int_{0}^{\infty} \hat{\mathbf{x}}(t') \mathbf{E}(t-t') dt', \quad \hat{\mathbf{x}}(\omega) = \hat{\mathbf{x}}^{0}(\omega) - i \frac{\hat{\sigma}(\omega)}{\omega},$$
$$\mathbf{P}^{(n1)} = \hat{\mathbf{x}} \mathbf{E} \mathbf{E},$$

 $\operatorname{rot}\operatorname{rot}\mathbf{E} + \frac{1}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} + \frac{4\pi}{c^2} \frac{\partial^2 \mathbf{P}^{(1)}}{\partial t^2} + \frac{4\pi}{c^2} \frac{\partial^2 \mathbf{P}^{(n1)}}{\partial t^2} = 0,$ 

(16)\*

in the form

$$\mathbf{E} = \mathbf{E}_{1} + \mathbf{E}_{2} + \mathbf{E}_{p} = \mathbf{e}_{1}A_{1}(t, \mathbf{r}) \exp \left[i\left(\omega_{1}t - \mathbf{k}_{1}\mathbf{r}\right)\right]$$
$$+ \mathbf{e}_{2}A_{2}(t, \mathbf{r}) \exp \left[i\left(\omega_{2}t - \mathbf{k}_{2}\mathbf{r}\right)\right]$$
$$+ \mathbf{e}_{p}A_{p}(t, \mathbf{r}) \exp \left[i\left(\omega_{p}t - \mathbf{k}_{p}\mathbf{r}\right)\right] + \text{c.c.;}$$
(17)

here  $\mathbf{e}_1$ ,  $\mathbf{e}_2$ , and  $\mathbf{e}_3$  are unit vectors describing the polarizations of the waves; in the first approximation the nonlinear interaction does not change the polarizations of the proper waves of the anisotropic medium.

<sup>\*</sup>rot  $\equiv$  curl.

In accordance with the foregoing we shall now write in place of (6) a more general relation in the form

$$\mathbf{k}_1 + \mathbf{k}_2 = \mathbf{k}_p + \Delta, \tag{18}$$

where  $\Delta$  is the so-called detuning vector ("wave" detuning). Then, substituting (17) in (16) and discarding, as before, the second derivatives (the changes in the complex amplitudes in space and in time can be regarded as slow), we arrive at a system of first-order simplified equations

$$\mathbf{s}_{1} \left\{ \mathbf{e}_{1} \left[ \mathbf{k}_{1} \mathbf{e}_{1} \right] \right\} \frac{\partial A_{1}}{\partial t} + \left[ \mathbf{e}_{1} \left[ \mathbf{k}_{1} \mathbf{e}_{1} \right] \right] \nabla A_{1} + \left( \mathbf{e}_{1} \hat{\mathbf{a}}_{1} \mathbf{e}_{1} \right) A_{1} + i \beta \omega_{1}^{2} e^{i \Delta \mathbf{r}} A_{p} A_{2}^{*} = 0, \qquad (19a)^{*}$$

$$\mathbf{s}_{2} \left[ \mathbf{e}_{2} \left[ \mathbf{k}_{2} \mathbf{e}_{2} \right] \right] \frac{\partial A_{2}}{\partial t} + \left[ \mathbf{e}_{2} \left[ \mathbf{k}_{2} \mathbf{e}_{2} \right] \right] \nabla A_{2}$$
$$+ \left( \mathbf{e}_{2} \hat{\boldsymbol{\alpha}}_{2} \mathbf{e}_{2} \right) A_{2} + i \beta \omega_{2}^{*} e^{i \Delta \mathbf{r}} A_{\mathbf{p}} A_{1}^{*} = 0, \qquad (19b)$$

$$\mathbf{s}_{\mathbf{p}}[\mathbf{e}_{\mathbf{p}}[\mathbf{k}_{\mathbf{p}}\mathbf{e}_{\mathbf{p}}]] \frac{\partial A_{\mathbf{p}}}{\partial t} + [\mathbf{e}_{\mathbf{p}}[\mathbf{k}_{\mathbf{p}}\mathbf{e}_{\mathbf{p}}]] \nabla A_{\mathbf{p}} + (\mathbf{e}_{\mathbf{p}}\hat{\boldsymbol{\alpha}}_{\mathbf{p}}\mathbf{e}_{\mathbf{p}}) A_{\mathbf{p}} + i\beta\omega_{\mathbf{p}}^{2}e^{-i\Delta\mathbf{r}}A_{\mathbf{1}}A_{\mathbf{2}} = 0;$$
(19c)

here

$$s=\frac{\partial \omega /\partial k}{(\partial \omega /\partial k)^2}$$

are the ray vectors of the waves, whose absolute values are equal to the reciprocals of the group velocities at corresponding frequencies,

$$\hat{\alpha}_i = \frac{2\pi\omega_i\hat{\sigma}(\omega_i)}{c^2}$$

are the attenuations and

$$\beta = \frac{2\pi}{c^2} \mathbf{e}_1 \hat{\chi} (\omega_p - \omega_2) \mathbf{e}_p \mathbf{e}_2 = \frac{2\pi}{c^2} \mathbf{e}_2 \hat{\chi} (\omega_p - \omega_1) \mathbf{e}_p \mathbf{e}_1$$
$$= \frac{2\pi}{c^2} \mathbf{e}_p \hat{\chi} (\omega_1 + \omega_2) \mathbf{e}_1 \mathbf{e}_2$$
(20)

are the coupling coefficients, while  $\hat{\chi}(\omega_i \pm \omega_k)$  are the spectral components of the nonlinear susceptibility tensor (of third rank); relation (20) is valid by virtue of the special symmetry properties of the tensor  $\chi$ (see, for example, <sup>[14,37]</sup>). The case of parametric amplification of weak unmodulated waves at frequencies  $\omega_1$  and  $\omega_2$  in the field of a strong unmodulated pump wave is described by equations (19a) and (19b) in which  $\partial A_1/\partial t = \partial A_2/\partial t = 0$ . If we regard a boundary-value problem similar to that of Sec. 1, then

$$\begin{array}{l} [\mathbf{e}_{1}[\mathbf{k}_{1}\mathbf{e}_{1}]]\nabla \\ = k_{1}\cos\left(\widehat{\mathbf{k}_{1}\mathbf{s}_{1}}\right)\cos\left(\widehat{\mathbf{s}_{1}\mathbf{z}_{0}}\right)\frac{d}{dz}, \\ [\mathbf{e}_{2}[\mathbf{k}_{2}\mathbf{e}_{2}]]\nabla \\ = k_{2}\cos\left(\widehat{\mathbf{k}_{2}\mathbf{s}_{2}}\right)\cos\left(\widehat{\mathbf{s}_{2}\mathbf{z}_{0}}\right)\frac{d}{dz}, \end{array} \right)$$

$$(21)$$

and, using standard methods for stability analysis of the system of equations (exponential growth of the amplitudes  $A_1$  and  $A_2$  with the coordinate can be regarded as "instability in space"), we can determine

$$*[\mathbf{k}_1 \mathbf{e}_1] \equiv \mathbf{k}_1 \times \mathbf{e}_1.$$



FIG. 3. Regions of parametric amplification of traveling light waves: The abscissas represent the wave detuning  $\Delta$ , and the ordinates the pump amplitude  $A_p$ . In a lossless medium the region of amplification lies between the two straight lines drawn from the origin; in a lossy medium, for the same detuning, larger values of  $A_p$  are necessary; the corresponding region is shown shaded.

in terms of the coordinates  $A_p$  and  $\Delta$  the region in which parametric amplification of traveling light waves is possible. The regions of amplification are shown in Fig. 3 for the case of zero losses and nonzero losses\*. It is seen from the diagrams in Fig. 3 that the larger the modulus of the wave detuning and the larger the damping decrement the more pump power is necessary to obtain the exponential-growth regime.

Equations (19), but now written out in full, should also be used to construct a theory of the processes occurring in a parametric generator; to this end they must be solved with the boundary conditions on mirrors located at z = 0 and z = d. The characteristic parameters of the problem are in this case:

1) the nonlinearity of the medium defined by the parameter  $\beta$ ,

- 2) the pump amplitude A<sub>p</sub>,
- 3) the damping decrements  $e_i \hat{\alpha}_i e_i$ ,
- 4) the wave detuning  $\Delta$ , defined by formula (18), and 5) the relative frequency detuning

$$\xi = \frac{\omega_p - \Omega_1 - \Omega_2}{h_1 + h_2}, \qquad (22)$$

where  $\Omega_1$  and  $\Omega_2$  are the resonant frequencies of the modes of the optical resonator in which the parametric oscillations are excited, and  $h_1$  and  $h_2$  are the widths of the spectrum of these modes:  $h_i = \Omega_i / Q_i$ .

It should be noted that a solution of the system (19) with boundary conditions defined at z = 0 and z = d is in general a very complicated problem, since even the simplified equations (19) are partial differential equations. The problem is also much more complicated because practical interest attaches primarily to an investigation of the transients in the parametric generator. We recall that the pulse duration in the high-

<sup>\*</sup>It is easy to see that the amplification regions shown in Fig. 3 have the same form as the instability regions of a linear oscillating circuit with variable capacitance or inductance; in the latter case the abscissas represent the frequency detuning.

power lasers customarily used as the pump generator in optical parametric devices does not exceed  $\tau_p = 2 \times 10^{-8}$  sec, so that the stationary parametric oscillations may not have time to settle within a time  $\tau_p$  in those cases when the threshold of the parametric selfexcitation is exceeded by a small margin. We describe here briefly two possible variants of the solution of the equation of a parametric generator.

# 2.2. Transient and Stationary Processes in a Parametric Generator—The Method of Successive Steps

One of the simplest methods, one especially convenient for digital-computer calculations, is the method of successive steps, in which the establishment of the parametric oscillations can be broken up into a series of successive stages, described by equations for a semi-bounded medium. The time derivatives in (19) can then be disregarded. Indeed, in such an approach inclusion of the time derivatives is essential only when the time of the group delay of the pump pulses and of the parametric oscillations

$$T = Nd\left(\frac{1}{u_{p}} - \frac{1}{u_{1,2}}\right)$$
(23)

(here N is the number of reflections,  $u_p$  and  $u_{1,2}$  are the group velocities at the corresponding frequencies) becomes comparable with the duration of the pump pulse  $\tau_p$ . In the cases of practical interest  $\tau_p \approx T$ only when N  $\cong$  200–300, so that this effect can certainly be neglected.

Let us illustrate the calculation procedure and some of the results, using a very simple example of a degenerate parametric generator ( $\omega_1 = \omega_2 =$  $= \omega_p/2$ ) and one-dimensional interaction (see Fig. 1). Going over in (19) to real amplitudes and phases, we arrive for the case in question at a system of equations for the amplitude of the subharmonic A, the pump amplitude A<sub>p</sub>, and the phase  $\Phi$ :

$$\frac{dA}{dz} + \sigma A_{\rm p}A\sin\Phi + \delta A = 0, \qquad (24a)$$

$$\frac{dA_{\mathbf{p}}}{dz} - \sigma A^2 \sin \Phi + \delta_{\mathbf{p}} A_{\mathbf{p}} = 0, \qquad (24b)$$

$$\frac{d\Phi}{dz} + \Delta + \sigma \left( 2A_{p} - \frac{A^{2}}{A_{p}} \right) \cos \Phi = 0.$$
 (24c)

Here  $\Phi = 2\varphi - \varphi_p - \Delta z$ , where in turn  $\varphi$  is the phase of the subharmonic,  $\varphi_p$  is the phase of the pump,  $\Delta$ the z component of the wave detuning vector  $\Delta$ ,  $\delta$  and  $\delta_p$  are the damping decrements of the corresponding wave, and  $\sigma$  is the coupling coefficient. For the case considered here we have  $\sigma = \frac{2\omega^2}{k_1}\beta$ .\* The calculation procedure consists of breaking up the process of establishment of the parametric oscillations into a sequence of steps described with the aid of the equations in (24). The boundary conditions for each succeeding step are determined by the results of the preceding step and by the properties of the mirrors. Introducing the real amplitude reflection coefficients of the mirrors  $R_1$  and  $R_2$  and the phase shifts  $\psi_{1,2}$  of the reflection from the mirrors, we can establish the connection between the amplitudes and the phases for the successive steps. It should be noted that even for the simplest variant of the parametric generator considered here the problem can be formulated in several ways. In particular, a distinction is made, generally speaking, between processes in a system in which the optical cavity resonates simultaneously at the pump frequency and at the subharmonic frequency, and in a system in which the optical cavity is practically transparent to the pump wave. Comparison of the different generator schemes can be found in <sup>[16]</sup>. We shall present here the results of a calculation for the scheme of a generator which has been experimentally realized (see [17] and [20]) and for which the reflection coefficients  $R_1$  and  $R_2$  are small at the pump frequency:  $R_1(\omega_p) \approx R_2(\omega_p) \approx 0$ . The boundary conditions on the mirrors for the amplitude of the subharmonic and the phase  $\Phi$  are

$$A_{N}^{(-)}(d) = R_{2}(\omega) A_{N}^{(+)}(d), \ A_{N}^{(+)}(0) = R_{1}(\omega) A_{N-1}^{(-)}(0), \ (25a)$$
  
$$\Phi_{N}^{(-)}(d) = \Phi_{N}^{(+)}(d) + \Psi_{2}, \ \Phi_{N}^{(+)}(0) = \Phi_{N-1}^{(-)}(0) + \Psi_{1}; \ (25b)$$

here N is the number of the step, the plus sign denotes quantities pertaining to the waves moving from left to right (forward), and the minus sign quantities pertaining to the backward waves.

The conditions for self-excitation of the generator and the transient time can be determined by using only Eqs. (24a) and (24b), in which the pump amplitude can be regarded as a parameter;  $A_p = A_{p0}$ , where  $A_{p0}$  is the amplitude of the forward pump wave at the entrance to the resonator. It must be borne in mind that when  $\Delta \neq 0$  and  $\delta \neq 0$  the variation of the amplitude of the subharmonic with the distance has a more complicated form than for the simplest case considered in Sec. 1. When  $\Delta/\sigma A_{p0} \ll 1$  and  $\delta < \sigma A_{p0}$  we have

$$A(z) = A_0 \exp\left[\sigma A_{\mathbf{p}\,0} \sqrt{1 - \frac{\Delta}{2\sigma A_{\mathbf{p}\,0}}} - \delta\right] z. \quad (26)$$

Thus, for  $\Delta > 2\sigma A_{p0}$  (see also Fig. 3) the exponential amplification gives way to damped oscillations. Using (26) we can obtain a more accurate self-excitation condition (cf. Sec. 1); for the settling time  $\tau_s$  we have approximately

$$\tau_{s} \simeq \frac{2 dn}{c} \left[ \sigma A_{p0} d \sqrt{1 - \frac{\Delta}{2\sigma A_{p0}}} - \delta d \right]^{-1} \ln \frac{A_{s}}{A_{0}}, \quad (27)$$

where  $A_S$  is the steady-state amplitude; the larger the ratio  $A_S/A_0$ , the more accurate is formula (27). (If the generator is self-excited by fluctuations, then  $A_S/A_0\approx 10^4-10^5.)$ 

<sup>\*</sup>Here  $\sigma$  is a scalar and of course has no connection with the conductivity tensor  $\sigma$  used in [<sup>16</sup>].



FIG. 4. Theoretical transient curves of degenerate parametric oscillations in a resonator transparent to the pump. The curves were plotted for the following values of the parameters:  $\sigma = 1 \text{ cm}^{-1}$ ,  $\delta = \delta_p = 0$ ,  $R_1(\omega) = R_2(\omega) = 0.99$ , d = 1 cm,  $A_0 = 10^{-3}$ ,  $A_{po} = 1$ . Curves 1, 4, and 9 represent the quantities  $A_N^{(+)}(d)$ ,  $A_{p(N)}^{(-)}(0)$ , and  $A_{p(N)}^{(+)}(d)$  for  $\Delta/2\sigma = 5 \times 10^{-3}$ ; curves 2,5 and 8-for  $\Delta/2\sigma = 0.5$ ; curves 3,6 and 7-for  $\Delta/2\sigma = 1$ .

To determine the stationary amplitude, it is necessary to solve the complete system (24). The results of such a solution, carried out by numerical means, are shown in Fig. 4, which contains plots of the amplitude of the forward wave of the subharmonic at the output mirror of the resonator  $A_N^{(+)}(d)$ , the amplitude of the forward pump wave at the output mirror  $A_{p(N)}^{(+)}(d)$ , and the amplitude of the backward pump wave at the input mirror  $A_{p(N)}^{(-)}(0)$  (this wave occurs even when  $R_1(\omega_p)$ =  $R_2(\omega_p) = 0$  as a result of the frequency doubling of the backward wave of the subharmonic), normalized to the value of  $A_{\texttt{p0}}.$  We see from Fig. 4 that the efficiency of the parametric generator can be quite high; the generator in question is simultaneously an active limiter of the amplitude of the pump wave passing through the resonator. An interesting fact is that the parametric generator is a unique nonlinear mirror for the pump wave; as can be seen from the plots in Fig. 4, the nonlinear reflection coefficient  $R^{(nl)} = A_p^{(-)}(0)/A_{p0}$ can be quite high. A more detailed exposition of the results of the parametric-generator theory based on the method of successive steps can be found in [16].

A similar analysis can be carried out for a nondegenerate generator. A qualitative picture of the processes considered above remains the same here, and the power distribution over the frequencies is described by the simple relation

$$\frac{A_1^2}{\omega_1} = \frac{A_2^2}{\omega_2} \,. \tag{28}$$

## 2.3. On the Theory of the Stationary Regime of a Parametric Light Generator. Parametric Amplification and Generation in Real Beams

Another possible approach to the theory of the parametric generator, one particularly fruitful in the analysis of the stationary generator regime, is to represent the oscillations of the subharmonic in the form of a standing wave with complex amplitude, which depends only on the time. A basis for this is the fact that the amplification of the subharmonic during one passage is small and the amplitudes of the forward and reflected waves of the subharmonic can be assumed equal in the succeeding steps. In this case, considering the degenerate mode, we can seek the subharmonic field in the form (we use real notation):

$$\mathbf{E} = \mathbf{e}A(t)\sin\left(\frac{n\pi}{d}z\right)\sin(\omega t + \varphi), \qquad (29a)$$

where n is the number of the longitudinal mode excited in the optical resonator, and the pump field, as follows from the results obtained above, must be sought in the form of a superposition of the forward and the reflected waves, with amplitudes that depend generally speaking on both the coordinate and on the time:

$$\mathbf{E}_{\mathbf{p}} = \mathbf{e}_{\mathbf{p}} A^{(+)}_{\mathbf{p}}(t, z) \cos \left[ \omega_{\mathbf{p}} t - k_{\mathbf{p}} z + \varphi_{\mathbf{p}}^{(+)} \right] + \mathbf{e}_{\mathbf{p}} A^{(-)}_{\mathbf{p}}(t, z) \cos \left[ \omega_{\mathbf{p}} t + k_{\mathbf{p}} z + \varphi_{\mathbf{p}}^{(-)} \right].$$
(29b)

Substituting (29a) and (29b) in (16) we arrive at a system of equations of the type (19). In view of the length of these equations, we shall not write them out here; we shall merely indicate that for the stationary case  $(\frac{\partial}{\partial t} = 0)$  it is possible to analyze them in the phase plane and obtain analytic results for some particular

cases. We note that the nonlinear-loss mechanism, connected with the frequency doubling of the backward wave of the subharmonic, has no analog in circuits with lumped constants; therefore if  $A_{p0}\sigma > \delta$ , allowance for the linear losses in the parametric generator of light is immaterial also from the fundamental point of view. Using such an approach, we can analyze the dependence of the amplitude of parametric oscillations on the pump amplitudes, on the wave and frequency detunings, etc.

In a nondegenerate parametric generator the threshold pump intensity depends on  $\Delta$  like  $\Delta^2 d^2/(1 - \cos \Delta d)$  and on  $\xi$  like  $1 + [2\xi(\Omega_1 + \Omega_2)/h_1 + h_2]^2$ . A similar analysis can be carried out also for a nondegenerate parametric generator. One of the interesting questions here is that of the frequency stability of the parametric oscillations. Indeed, in the nondegenerate regime (un-like the degenerate one, in which the relation  $\omega = \omega_p/2$  is exactly satisfied) the frequencies  $\omega_1$  and  $\omega_2$  can vary with the resonator parameters, etc. If the stationary amplitudes of the parametric oscillations are determined by the final pump power, then the frequencies  $\omega_{1,2}$  are equal to

$$\omega_1 = \Omega_1 + \xi h_1, \ \omega_2 = \omega_p - \omega_1 = \Omega_2 + \xi h_2,$$

where  $\xi$  is given by (22).

Using the foregoing formulas, we can analyze the dependence of the frequencies of the tunable parametric generator on the resonator parameters, that is, determine the technical width of the spectral line of the generator. It is interesting that under certain conditions unilateral deviations of the optical-resonator parameters do not influence the generated frequencies; the frequency stability of the parametric generator of light can exceed the stability of the partial frequencies of the optical resonators.\* In this sense, a parametric light generator can be similar to two-loop parametric generators used for frequency stabilization in the radio<sup>[38,39]</sup> and microwave bands<sup>[40]</sup>. In this connection it is appropriate to turn attention to the fact that one of the essential difficulties that arise in realization of tunable parametric generators in the radio band, connected with the mutual synchronization of the oscillations at frequencies  $\omega_1$  and  $\omega_2$  (see, for example,<sup>[40]</sup>) does not appear in the optical band. The generation of the harmonics  $n\omega_1$  and  $m\omega_2$ , which is responsible for the mutual synchronization, is ineffective here because of the large wave detunings. Exceptions are near-degenerate regimes for which difficulties arise during the course of frequency tuning near  $\omega \cong \omega_{\rm D}/2$ ; this has apparently taken place in the experiments described in [20].

To determine the tuning range of a parametric light generator and the corresponding orientations of the crystal (in one-dimensional interaction) or optical resonators (in two-dimensional interaction) there is no need to use the equation of the generator. These quantities are determined by the linear dispersion properties of the working crystal. By specifying the form of the nonlinear interaction (as already indicated, the synchronism conditions in typical nonlinear crystals can be satisfied for the interaction  $k_1^O + k_2^O = k_p^O$ and sometimes for the interaction  $k_1^O + k_2^e = k_p^e$ ) and the values of the frequencies  $\,\omega_1,\,\omega_2,\,\text{and}\,\,\omega_p$  (in practice it is convenient to specify  $\omega_p$  and the ratio  $\kappa$ =  $\omega_1/\omega_p$ ), and by using the dispersion properties of the crystals, it is possible to determine from the specified value of the angle  $\theta_p = (\mathbf{k}_p \mathbf{z}'_0) (\mathbf{z}'_0)$  is a unit vector along the optical axis) the values of the angles  $\theta_1$ =  $(\mathbf{k}_1 \mathbf{z}_0')$  and  $\theta_2 = (\mathbf{k}_2 \mathbf{z}_0')$ . Figure 5 shows such a construction for the interaction  $k_1^O + k_2^O = k_p^O$  in a KDP crystal at  $\lambda_p = 0.35 \,\mu$  (second harmonic of a ruby laser). The vertices of the curve correspond to the direction of the one-dimensional interaction for a given  $\kappa = \omega_1 / \omega_p$ . It follows from the presented curves that for given  $\theta_p$  and  $\kappa$  there exists only one pair of angles  $\theta_1$  and  $\hat{\theta_2}$  satisfying the synchronism condition.



FIG. 5. Tuning curves of optical parametric generator using the interaction  $\mathbf{k}_1^0 + \mathbf{k}_2^o = \mathbf{k}_p^o$  in a KDP crystal. The abscissas represent the angle  $\theta_p = (\mathbf{k}_p \mathbf{z}_0)$  and the ordinates the angles  $\theta_1 = (\mathbf{k}_1 \mathbf{z}_0^i)$  and  $\theta_2 = (\mathbf{k}_2 \mathbf{z}_0^i)$ . The parameter of the curves is the quantity  $\kappa = \omega_1/\omega_p$ . The curves were plotted for  $\lambda_p = 0.35 \ \mu$ .

Of course, for effective utilization of a nonlinear crystal as a working medium in a parametric light generator, the crystal must have not only suitable dispersion characteristics in the frequency band of interest, but also ensure satisfaction of conditions for parametric excitation at not too high pump powers. The latter imposes limitations on the magnitude of the nonlinear-susceptibility tensor and on the damping decrement.

At present there are four sources of powerful coherent optical oscillations that are suitable for use as pump generators in optical parametric generators. These are the ruby laser, the neodymium-glass laser, and their second harmonic generators. Typical data on pump generators and a list of nonlinear materials, suitable for production of the appropriate parametric generators, based on the results of papers published up to November 1, 1965, are listed in the Table (we present here also the values of the nonlinear susceptibilities for KDP, ADP, and LiNbO<sub>3</sub> crystals).

In concluding this section, we must emphasize that the parametric-generator theory presented above was developed for the case of plane quasi-monochromatic waves. It is advantageous to discuss, albeit qualitatively, the question of the corrections that must be introduced in this theory to interpret data obtained with real laser beams. If we deal with unfocused beams (such beams are customarily used in experiments with solid-state pump generators) we use the geometricoptics approximation.\* In this case the most important

<sup>\*</sup>Moreover, a regime is also possible in which the frequency stability of the parametric generator exceeds the stability of the pump frequency.

<sup>\*</sup>At the same time, allowance for the diffraction effects is of interest in connection with experiments in which the pump generators used are gas lasers<sup>[21]</sup>; to satisfy the self-excitation conditions, focusing of the pump is necessary in this case. A procedure for calculating nonlinear optical effects in focused beams is given, for example, in<sup>[41]</sup>.

#### PARAMETRIC AMPLIFIERS AND GENERATORS OF LIGHT

Pump generator	Nonlinear material	Average wavelength of parame- tric genera- tor	Parameters of nonlinear materials
Ruby laser $\lambda_{\mathbf{p}} \approx 0.7 \ \mu$ , $P_{\mathbf{p}} = 100 \ \text{MW}$	$\begin{array}{c} {\rm LiNbO_3}\\ {\rm NH_4D_2PO_4}\\ {\rm (DADP)} \end{array}$	$\lambda \simeq 1.4 \ \mu$	1) KDP: $d_{36} = 3.40^{-9} \text{ CGSE}$ at $\lambda = 1.06 \mu$
$ \begin{array}{l} \text{Nd}^{3} ^{+}\text{-glass laser} \\ \lambda_{p} = 1.06 \ \mu, \\ R_{p} = 100 \ \text{MW} \end{array} $	LiNbO3	$\lambda \simeq 2$ μ	2) LiNBO <sub>3</sub> <sup>20</sup> : $d_{31}=3\cdot10^{-8}$ CGSE 3) ADP: $d_{36}=2\cdot10^{-9}$ CGSE
Second harmonic of ruby laser $\lambda_{\mathbf{p}} \approx 0.35 \ \mu$ ,	KH <sub>2</sub> PO <sub>4</sub> (KDP) NH <sub>4</sub> H <sub>2</sub> PO <sub>4</sub> (ADP)	λ=0.7 μ	
$P_{p}^{P} = 7 - 8 \text{ MW}^{[18]}$			Damping decrements for ADP:
Second harmonic of glass laser $\lambda_{p} \approx 0.53 \ \mu$ , $P_{p} \approx 25 \ MW[^{17}]$	KH <sub>2</sub> PO <sub>4</sub> (KDP) NH <sub>4</sub> H <sub>2</sub> PO <sub>4</sub> (ADP) LiNbO <sub>3</sub>	$\lambda = 1.06 \mu$	at $\lambda = 1.06 \ \mu$ $2\delta = 0.151 \ \text{cm}^{\text{-1}}$ at $\lambda = 0.53 \ \mu$ $2\delta = 0.024 \ \text{cm}^{\text{-1}}$ [42]

differences between the beam generated by the laser and the model considered above are its finite aperture, divergence, and mode structure. Within the framework of the geometric-optics approximation, the effect of the finite aperture of the pump beam is connected with the difference in the directions of the wave and ray vectors in an anisotropic medium. If we denote by  $\gamma$  the anisotropy angle,  $\gamma = (\mathbf{ks})$ , the effects of the finite aperture will come into play at lengths  $l_a = ML\gamma^{-1}$ , where L is the beam diameter and M is a dimensionless coefficient, the magnitude of which is determined by the properties of the spatial coherence of the pump. If the pump generator operates at the lowest transverse mode,  $M \cong 1$ , and for ordinary conditions realized in experiments, then  $l_a \approx 10-15$  cm. Using the value of  $l_{\rm a}$  calculated in this manner, we can then estimate the number of reflections in the resonator of a parametric generator N, for which the influence of the aperture effect is still immaterial:  $l_a \approx Nd$ .

In order to analyze the question of influence of the divergence of the pumping, it is necessary to take into account the fact that the wave detuning  $\Delta$  depends on the angle between the ray in question and the optical axis (see, for example, Fig. 1,a); for small deviations from the synchronism condition (6) we have

$$\Delta = K \left( \theta - \theta_0 \right). \tag{30}$$

Using (26), we can determine the critical divergence angle (it is sometimes called the capture angle), at which exponential growth of the subharmonics is still possible:

$$\alpha_{\rm cr} = \frac{2\sigma A_{\rm p_0}}{K} \,. \tag{31}$$

Thus, if we deal with amplification of traveling waves, the influence of the beam divergence  $\alpha_0$  can be neglected if  $\alpha_0 < \alpha_{cr}$ .

In an optical resonator, waves can be regarded as plane if  $\alpha_0 < \alpha_r$ —the angular aperture of the central maximum of the resonator. Thus, the beam divergence

does not influence processes in parametric light generators if

$$\alpha_0 < \alpha_{cr} < \alpha_r$$
.

Processes in parametric light generators are strongly influenced by the mode structure of the time spectrum of the pump. For a generator with one-dimensional interaction (see Fig. 1), the condition for existence of resonance simultaneously at the frequencies  $\omega_1$  and  $\omega_2$  is equivalent, obviously, to the condition of synchronism of the pump-generator modes and the modes of the parametric-generator optical resonator. Therefore a pump in the form of a solid-state laser is as a rule inefficiently used in a parametric generator.

The angular dependence of the signal with  $\lambda = 1.06 \mu$ amplified in a degenerate parametric amplifier of traveling wave ( $\lambda_p \approx 0.53 \mu$ ) is shown in Fig. 6. A KDP crystal 3 cm long and a pump-power flux P<sub>p</sub>  $\approx 100 \text{ MW/cm}^2$  were used in these experiments [17]. The angular width of the amplification curves was determined by the value of  $\alpha_{cr}$ . Different curves correspond to different experiments (their different position is connected with the phase selectivity of the degenerate parametric amplifier); the average power gain was 2-3, which exceeded the threshold of parametric excitation. The introduction of mirrors led to regenerative amplification and self-excitation of the oscillations (the characteristics of the generator are given in Sec. 1).

The threshold of parametric excitation is always clearly pronounced. A study of the spectrum of the angular structure of parametrically excited oscillations offers evidence of a high degree of their spatial and time coherence. In the experiments described in <sup>[20]</sup>, the divergence of the pump was  $\alpha_p \approx (2-3) \times 10^{-3}$  rad, and the divergence of the radiation of the parametric generator was  $\alpha_{1,2} \approx 3 \times 10^{-3}$  rad. The width of the pump spectrum was  $\delta\lambda_p \approx 1.54$ Å, and that of the parametric was in some cases not worse than  $\delta\lambda_{1,2} = 0.3$ Å.

The frequency tuning of the parametric generator in



FIG. 6. Experimental plots (taken from  $[1^{7}]$ ) of the intensity of the signal (in relative units) amplified in an optical parametric amplifier vs. the direction. The angle  $\theta_0$  corresponds to the synchronism direction. The different experiments, and the horizontal line drawn through unity corresponds to the pump turned off.

<sup>[20]</sup> was realized by changing of the temperature of LiNbO<sub>3</sub> crystal used as the nonlinear medium. The length of the crystal amounted in this case to d = 0.53 cm and the reflecting coatings were deposited directly on the crystal. Experimental tuning curves obtained in <sup>[20]</sup> are shown in Fig. 7.

It must be noted that in the experiments carried out to date, no attempt was made to optimize the efficiency of the parametric generators, therefore the efficiencies of ~ 0.1-0.05%, obtained at the present time, cannot be regarded as the limits.

In experiments with  $\lambda_p = 0.35 \,\mu$ , described in <sup>[18,19]</sup>, the threshold of parametric excitation was not exceeded; we note that the registration of the different frequencies, carried out in <sup>[19]</sup>, does not of necessity prove the existence of exponentially growing waves (for more details see <sup>[17]</sup>). A distinguishing feature of <sup>[18]</sup> is the fact that it is devoted to a study of two-dimensional interaction. It is of interest to note that in two-dimen-



FIG. 7. Experimental dependence of the frequencies generated by a tunable parametric generator with  $\lambda_p = 0.529 \ \mu$ , on the temperature of the working LiNbO<sub>3</sub> crystal (the plot is borrowed from[<sup>20</sup>]).

sional interaction the phase selectivity of degenerate parametric amplification drops out; this effect had no analog in the radio band.

## 3. PARAMETRIC INTERACTION AND INDUCED SCATTERING

The parametric interactions of light waves, considered in Secs. 1 and 2 were but one of the examples of a broad class of wave interactions of a similar type. Acoustic and light waves, spin and light waves, plasma and light waves, etc. can also interact effectively in the field of an intense light pump. Here, exactly as in the interaction of light waves and interactions of waves of different nature, exponentially growing waves can occur (instability in space; see Fig. 3), and hence, if suitable feedback is introduced, parametric excitation of acoustic, plasma, etc. waves can occur under the influence of light pumping. To explain the foregoing let us write out, for example, the equations for the parametric interaction of acoustic and light waves in an isotropic medium in the field of a powerful optical pump (called induced Mandel'shtam-Brillouin scattering). The connection between the acoustic and light fields is determined, on the one hand, by the presence of an electrostiction force

$$f = \frac{1}{8\pi} \left[ E^2 \varrho \frac{\partial \epsilon}{\partial \varrho} \right], \qquad (32)$$

where  $\rho$  is the density (action of electromagnetic waves on sound waves), and by the presence of a term in the polarization of the form

$$\mathbf{P} = \frac{1}{4\pi} \frac{\partial \varepsilon}{\partial \varrho} \frac{\partial \varrho}{\partial p} \mathbf{E} p, \qquad (33)$$

where p is the sound pressure (this term describes the action of sound waves on light waves). Then, in the field of a powerful pump wave

$$\mathbf{E}_{\mathbf{p}} = \mathbf{A}_{\mathbf{p}}(t, \mathbf{r}) \exp\left[i\left(\omega_{\mathbf{p}}t - \mathbf{k}_{\mathbf{p}}\mathbf{r}\right)\right]$$
(34)

the interaction of the acoustic wave

(35)

$$p = p(t, \mathbf{r}) \exp \left[ \iota \left( \Omega t - \mathbf{k}_{\Omega} \mathbf{r} \right) \right]$$

and the light wave

$$\mathbf{E}_{1} = \mathbf{A}_{1}(t, \mathbf{r}) \exp\left[i\left(\omega_{1}t - \mathbf{k}_{1}\mathbf{r}\right)\right], \qquad (36)$$

whose frequencies and wave numbers satisfy the relations

$$\omega_1 + \Omega = \omega_p, \quad \mathbf{k}_1 + \mathbf{k}_\Omega = \mathbf{k}_p + \Delta, \tag{37}$$

are described by the equations

$$(\mathbf{k}_{1}\nabla) \mathbf{A}_{1} + \frac{k_{1}}{u_{1}} \frac{\partial \mathbf{A}_{1}}{\partial t} + \delta_{1} \mathbf{A}_{1} - i \frac{\partial e}{\partial \varrho} \frac{\partial \varrho}{\partial p} \omega_{1}^{2} \frac{[\mathbf{k}_{1}[\mathbf{k}_{1}\mathbf{A}_{p}]]}{2k_{1}^{2}c^{2}} p^{*}e^{i\Delta \mathbf{r}} = 0,$$
(38a)

$$(\mathbf{k}_{\mathbf{p}} \nabla) \mathbf{A}_{\mathbf{p}} + \frac{k_{\mathbf{p}}}{u_{\mathbf{p}}} \frac{\partial \mathbf{A}_{\mathbf{p}}}{\partial t} + \delta_{\mathbf{p}}^{*} \mathbf{A}_{\mathbf{p}} - i \frac{\partial \varepsilon}{\partial \varrho} \frac{\partial \varrho}{\partial p} \omega_{\mathbf{p}}^{2} \frac{[\mathbf{k}_{\mathbf{p}} [\mathbf{k}_{\mathbf{p}} \mathbf{A}_{\mathbf{i}}]] p}{2k_{\mathbf{p}}^{2} c^{2}} e^{-i\Delta \mathbf{r}} = 0,$$
(38b)

$$\mathbf{k}_{\Omega} \nabla p + \frac{k_{\Omega}}{u_{\Omega}} \frac{\partial p}{\partial t} + \delta_{\Omega} p + i \frac{1}{8\pi} \varrho \frac{\partial e}{\partial \varrho} (\mathbf{k}_{\Omega} - \Delta)^2 \mathbf{A}_{\mathbf{p}} \mathbf{A}_{1}^{*} e^{-i\Delta \mathbf{r}} = 0.$$
(38c)

It is easy to see that Eqs. (38) have the same form as (19); to be sure, unlike (19), they are vector equations, for in an isotropic medium the polarizations of the natural waves are not defined.

Just as the system (19), the system (38) has an exponentially growing solution at pump power above threshold. Therefore nonlinear medium in which acoustic and light oscillations interact can serve as a parametric generator of hypersound with optical pump-ing. It should be noted that although induced Mandel'-shtam-Brillouin scattering has already been observed many times experimentally (for example, see [43]), intense hypersonic oscillations were not registered directly. It can be expected that irradiation of optically transparent crystals (for example, quartz) with powerful laser pulses at helium temperatures, will make it possible to excite intense sound oscillations with power

 $P_{\Omega} \simeq \frac{\Omega}{\omega_p} P_p$ . (We note that similar amounts of power

are transferred to hypersonic oscillations also in liquid, but there, owing to the large attenuation of the hypersound, they are dissipated in the form of heat.)

An interesting possibility of recording intense hypersonic oscillations arising during the course of induced scattering is the use of the piezoeffect in a scattering medium. In this case the scattering medium becomes an optically-pumped microwave generator; it then becomes possible to generate oscillations in the millimeter band. Parametric submillimeter generators with optical pumping can be constructed, apparently, by using induced scattering by spin waves. We note, finally, that intense oscillations of a scattering medium, arising during the course of induced scattering, can themselves serve as a pump for lower-frequency oscillations (for example, acoustic oscillations). Interesting possibilities are uncovered by the use of coherent molecular oscillations produced during the course of induced Raman scattering (induced Raman scattering is described by equations of the type (38), if the damping decrement for the sound is assumed to be

sufficiently large). V. T. Platonenko and one of the authors of this article have shown<sup>[28]</sup> that coherent molecular oscillations at frequency  $\Omega$  can excite infrared waves at frequencies  $\omega_1$  and  $\omega_2$  satisfying the relation  $\Omega = \omega_1 + \omega_2$  at a pump-laser power not much higher than the threshold for induced Raman scattering. Recently V. Akanaev observed such an interaction experimentally in our laboratory in strongly compressed hydrogen.

#### 4. CONCLUSIONS

The material presented in this review offers evidence of the possibility of experimental realization of the principle of parametric amplification and generation in the optical band. We can expect further progress in the development of nonlinear materials, resonator systems, and pump sources to lead to the creation of parametric generators in different regions of the visible and infrared bands. Especially promising is research aimed at creating tunable optical parametric generators of continuous action, using gas lasers as pump generators.\*

It must be noted that parametric interaction in a medium with polarization of the quadratic type and with a pump frequency exceeding the frequency of the parametric oscillations ("high-frequency" pump), are not the only parametric interactions which is possible in the optical band. In principle, there are definite prospects for observing parametric effects with "low-frequency" pumping (see, for example,  $[^{44}]$ ); also interesting are parametric interactions in a medium with nonlinear polarization of the cubic type  $\mathbf{P} = \hat{\chi} \mathbf{EE} \cdot \mathbf{E}$ . In such a medium (for example, in CaCO<sub>3</sub> crystals), the energy of the powerful pump oscillations of frequency  $\omega_p$  can be transferred to parametric oscillations at frequencies  $\omega_1$  and  $\omega_2$  satisfying the relation  $2\omega_p = \omega_1 + \omega_2$  (see  $[^{14}]$ ). With such an interaction it is especially easy to satisfy the synchronism conditions.

It is of course too early to speak of all the applications of parametric generators; it is appropriate to note, however, that they can apparently be used effectively in optical information-processing systems, in analogy with their radio-frequency counterparts. In conclusion, it should be noted that recently definite prospects have arisen for constructing tunable optical generators using multi-photon processes† in inverted quantum systems. A. M. Prokhorov and A. S. Selivanenko have proposed using such transitions for the construction of powerful lasers with tunable frequencies.<sup>[46]</sup> At the already mentioned Quantum-electronics Conference in Puerto Rico it was reported that such a transition, in which a photon and a phonon took part, was experimentally observed.

<sup>\*</sup>Most promising from this point of view are argon and CO<sub>2</sub> lasers.

 $<sup>^{\</sup>dagger}A$  comprehensive review of the literature on multi-phonon processes was presented by A. M. Bonch-Bruevich and V. A. Khodovoï[ $^{45}$ ].

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