INFLUENCE OF THE ELECTRO-OPTICAL EFFECT ON THE FREQUENCY OF A PARAMETRIC LASER WITH A KDP CRYSTAL

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Submitted April 8, 1968

Zh. Eksp. Teor. Fiz. 55, 802–812 (September, 1968)

The theoretical and experimental investigation of the feasibility of electro-optical tuning of a parametric laser with a KDP crystal is reported. It is shown that such a tuning is most effective near the Curie point of the KDP crystal where the electro-optical coefficient r_{63} is three orders higher than at room temperature. Tunability within the range of 1410 Å is attained.

INTRODUCTION

THE development of parametric lasers[1-4] offers new possibilities of scientific and technological application of lasers because of their tuning capability.

We know that the parametric laser emits a set of frequencies ω_1 and ω_2, satisfying the relations

$$\omega_p = \omega_1 + \omega_2, \tag{1}$$

$$k_p = k_1 + k_2, \tag{2}$$

where ω_p is the pump frequency and k_i is the corresponding wave vector, $|k_i| = \omega_i n_i / c$ where n_i is the index of refraction of the medium. It follows from (1) and (2) that the frequencies emitted by the parametric laser depend on the dispersive properties of the crystal. By changing its refractive indices by some method we can obtain frequency-tunable coherent oscillations within the optical region, provided the threshold conditions are exceeded.

Along with the existing methods of tuning parametric lasers (mechanical rotation of the crystal within the parametric resonator[1] and temperature variation of the refractive indices of a nonlinear crystal[2]) the electro-optical method of tuning is of definite interest. This method consists of varying the output frequencies of the parametric laser by changing the indices of refraction of the crystal through the application of an external electric field (electro-optical effect). The method was proposed in a very general sense in[5], was developed in more detail in[6,7], and was realized experimentally in[8,9] for the KDP crystal and in[10] for $LiNbO_3$.

The present paper contains a more detailed discussion of the experimental methodology and theoretical and experimental results reported in[9], and offers new experimental data.

COMPUTATION OF THE INFLUENCE OF THE ELECTRO-OPTICAL EFFECT ON THE TUNING CURVES OF THE PARAMETRIC LASER

Condition (2), called the phase-matching condition, is equivalent to the requirement that phase relations among the interacting waves be maintained along the entire nonlinear medium. It follows from the dispersive properties of the nonlinear medium and from (1) and (2) that fixed-frequency waves are excited in the medium (in a crystal for example) at completely determined phase-matching angles. The electro-optical method of frequency tuning consists of varying the phase-matching angles (and thus varying frequency) by the application of an external electric field to the nonlinear crystal:

$$\Delta\omega \sim \Delta n \sim rE, \tag{3}$$

where Δn is a change in the index of refraction due to field E, and r is the so-called linear electro-optical coefficient. The influence of the Kerr effect on frequency tuning is discussed below. The influence of the electro-optical effect on frequency tuning in a parametric laser was investigated by performing an analysis of the KH_2PO_4 (KDP) crystal pumped at $\lambda_p = 0.53\ \mu$ (second harmonic of a Nd^{3+} glass laser) for the case of a one-dimensional parametric interaction of two types:

$$k_p^e = k_1^o + k_2^o \quad (eoo), \tag{4}$$

$$k_p^e = k_1^o + k_2^e \quad (eoe). \tag{5}$$

The superscripts o and e refer to the ordinary and extraordinary rays respectively.

The constant external electric field E is considered directed along the optical axis Z of the crystal, because the effect exerted by the field on the index of refraction is an order of magnitude lower in the X and Y directions (X and Y being the crystallographic axes). A plane monochromatic pumping wave is assumed.

Relations (4) and (5) can be rewritten as

$$\omega_p n_p^e = \omega_1 n_1^o + \omega_2 n_2^o, \tag{4'}$$

$$\omega_p n_p^e = \omega_1 n_1^o + \omega_2 n_2^e. \tag{5'}$$

Since the expression for n^e is a complicated function of several parameters (θ, ω, T, E; θ is the angle between the direction in the crystal and the optical axis Z, and T is crystal temperature), an exact analytical solution is not possible. Therefore the expressions for the extraordinary indices of refraction were expanded in a Taylor series using only the first two terms of the expansion. The values of the indices of refraction were taken or computed from[11]. All the computations were performed on the EVM M-20 computer.

According to the computation the application of the electric field shifts the tuning curves and consequently varies the frequency of the parametric laser along a fixed direction in the crystal determined by the angle θ.

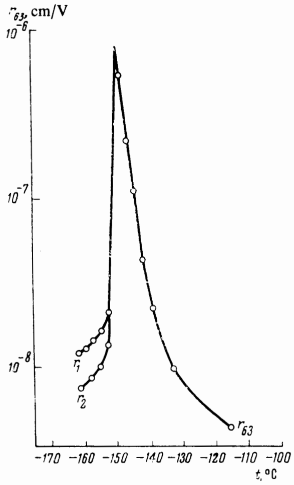

FIG. 1. Electro-optical coefficients r_i as functions of temperature for the KDP crystal. r_{63} – at $T > T_c$; r_1, r_2 – at $T < T_c$.

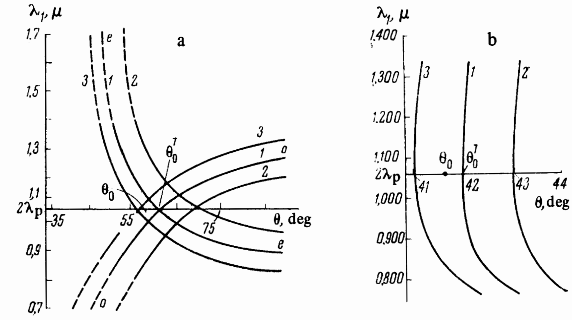

FIG. 2. Shift of the tuning curves following the application of an external field. a – for the (eoe) interaction; b – for the (eoo) interaction. θ_0 and $\theta_0{}^T$ are phase-matching angles for the degenerate case at T = 300°K and T = T_c respectively.

However this variation is small in KDP crystal at room temperature even if the fields are large (~ 100 kV/cm). In order to obtain a significant frequency variation with reasonable electric fields it is necessary to increase the electro-optical coefficient r_{63} by cooling the crystal down to the temperatures of the order of the Curie point (T_c = 123°K for the KDP crystal). The electro-optical coefficient r_{63} sharply rises (by about three orders) when the crystal temperature varies from room temperature to the Curie point[12]. Figure 1 shows an experimental graph of the thermal dependence of r_{63} for KDP specimens used in our experiment.

Data on the thermal variation of the indices of refraction of KDP necessary for the computation were taken from[13]. The dispersion of r_{63} is neglected because of its low value in the optical range. The computation results are given in Fig. 2. Figure 2 a shows the tuning curves of the parametric laser for the (eoe) interaction at T ~ T_c when E = 0 (curve 1) and when E = ± 10 kV/cm (curves 2 and 3). These curves indicate the possibility of a fairly broad tuning range of the parametric laser upon the application of the field in the forward and reverse directions. For example the wavelength change is $\Delta\lambda_1 \approx \Delta\lambda_2 \approx 1200$ Å for the angle θ_0^T with an initial wavelength of $\lambda_1 = \lambda_2 = 1.06\ \mu$ at T ~ T_c and E = ± 10 kV/cm. The tuning curves for the (eoo) interaction are much steeper and offer a greater opportunity for electro-optical tuning (see Fig. 2b, notation is the same as in Fig. 2a). Thus according to the computation the wavelength change for the angle θ_0^T is $\Delta\lambda_1 \approx 7000$ Å and $\Delta\lambda_2 \approx 3000$ Å with a field of E = ± 10 kV/cm. Consequently the tuning range can be large up to the crystal absorption limit.[1)]

Of considerable interest from the viewpoint of the electro-optical frequency tuning is also the two-dimensional parametric interaction which, for given pumping and signal frequencies, permits us to optimize θ with respect to electro-optics and nonlinear interaction.

[1)]However, the steepness of the tuning curves hampers the realization of a parametric laser with a KDP crystal having the above type of interaction [1].

EXPERIMENTAL PART

Experimental Setup. Method of Measurements.

The setup shown schematically in Fig. 3 was assembled for the experimental verification of the possibility of electro-optical tuning of a parametric laser with a KDP crystal.

The beam of a Q-switched Nd^{3+} glass laser (rotating prism with an auxiliary reflector prism) passes through an amplifier and enters a KDP crystal (l = 3 cm) serving as a frequency doubler in which the primary emission at $\lambda_0 = 1.06\ \mu$ generates a second harmonic at $\lambda_p = 0.53\ \mu$; the latter is the pump radiation for the parametric laser. An additional glass plate is placed at the Brewster angle in the laser resonator to achieve a more complete polarization of the emission. Such a disposition of the plate with respect to the end faces of the active rod yields a more effective polarization than a stack of plates parallel to the end faces (see also[14]). The liquid filter F_1 (Koblentz mixture) effectively cuts off the primary radiation and passes about 90% of the pumping radiation when the cell length is l = 1 cm.

The KDP parametric crystal (l = 3.3 cm) is housed in an optical cryostat that has two mirrors (m – m) serving as the input and output windows and forming a parametric resonator. The mirrors have a transmission coefficient of about 85% at the pump wavelength and a reflection coefficient R = 86% at the wavelength $\lambda_0 = 1.06\ \mu$ with a plateau near the degenerate case of 500–700 Å at the level R > 90%. The KDP parametric crystal was

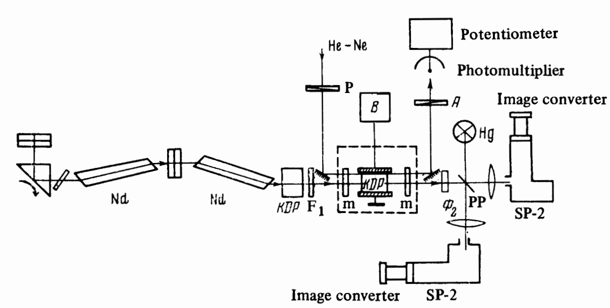

FIG. 3. Diagram of the experimental setup. F_1 – liquid filter; F_2 – KS-10 filter; B – high-voltage supply; PP – plane-parallel glass plate; P – polarizer; A – analyzer.

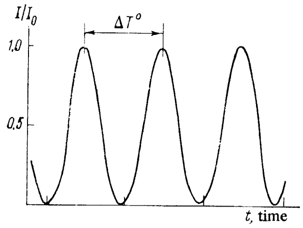

FIG. 4.

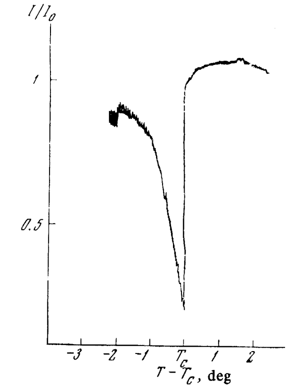

FIG. 5.

cut in the following manner: the input and output faces are perpendicular to the direction determined by the angles $\theta = 57°$ (phase-matching angle for the (eoe) interaction) at $T = 300°K$ and $\varphi = 15°$. The azimuthal angle φ was selected to meet two conditions simultaneously: $\Delta n(E) \neq 0$ and $\beta \neq 0$, where β is a nonlinear coupling coefficient for modes with the above polarizations[15]. In our cut $\beta = 0.87\ \beta_{max}$ and $\Delta n(E) = 0.5\ \Delta n(E)_{max}$.

A vacuum of $\sim 10^{-5}$ mm Hg was maintained in the optical cryostat. The electrodes, to which pulsed or constant electric fields were applied, served at the same time as cold fingers. The design of the cryostat permitted the electrodes to hold the crystal in a flexible manner during the passage through T_c when rigid electrodes would crack the crystal. Crystal temperature was measured by the phase method described in[16] since a thermocouple cannot be introduced into the crystal because of the effect of a powerful light radiation on the thermocouple junctions. The beam of a helium-neon laser passed through the polarizer-crystal-analyzer system and entered a photomultiplier whose signal was recorded by a potentiometer. When crystal temperature varies the transmission curve assumes the form (see Fig. 4)

$$\frac{I}{I_0} = \sin^2\frac{\pi\Delta n(T)l}{\lambda}. \qquad (6)$$

Since in a general case $\Delta n(T)$ can be represented as $\Delta n(T) = \alpha(T)(T - T_0)$, where $\alpha(T)$ is the thermal birefringence coefficient, the temperature interval $\Delta T = T - T_0$ between two neighboring minima (or maxima) of transmission curve (6) is expressed as follows:

$$\Delta T = \lambda / l\alpha(T).$$

Since within the temperature range $T > T_c$ the relation $\alpha(T) = \text{const} = 0.11649 \times 10^{-4}\ \text{deg}^{-1}$[13] holds with high precision in the KDP crystal, when $\lambda = 0.6328\ \mu$ and $l = 3.3$ cm we have $\Delta T = 1.73°$. In the range $T < T_c$ the graph of the function $\alpha = \alpha(T)$ is plotted and used to determine ΔT.

A strong scattering of light, or critical opalescence, typical of phase transitions of the second kind occurs directly next to or at T_c. Crystal transmission near T_c is shown in Fig. 5. The shape of this curve is in a qualitative agreement with theoretical considerations developed in the phase transition theory[17]. This scattering increases the threshold of parametric generation

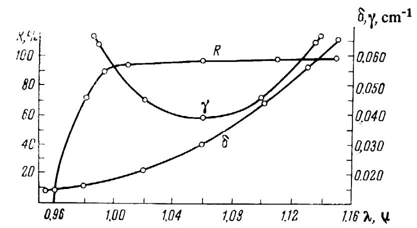

FIG. 6. Dependence of R, δ, and γ on the wavelength. δ – is damping decrement of the KDP crystal and γ is a quantity proportional to $\sqrt{P_p^{thr}}$ [1].

directly next to T_c and must be taken into account during the experiment. In our experiment the maximum pump power density was 45 MW/cm² when the threshold power for the degenerate case was 18–20 MW/cm². The corresponding calculated value (12 MW/cm²) was obtained from data given in Fig. 6. The divergence of the pumping beam was $2\alpha_\theta = 5'$, $2\alpha_\varphi = 16'$. The pump pulse length was 50 nsec. As in[1] a powerful pump beam was observed in reverse operation of the laser.

Experimental Results

Figures 7 a-c show the emission spectrum of the parametric laser obtained with the ISP-51 spectrograph (mean dispersion in the $\lambda = 1\ \mu$ range is D = 300 Å/mm) and an image converter. Mercury lines are given for comparison. Figures 7 d-g show emission spectrograms

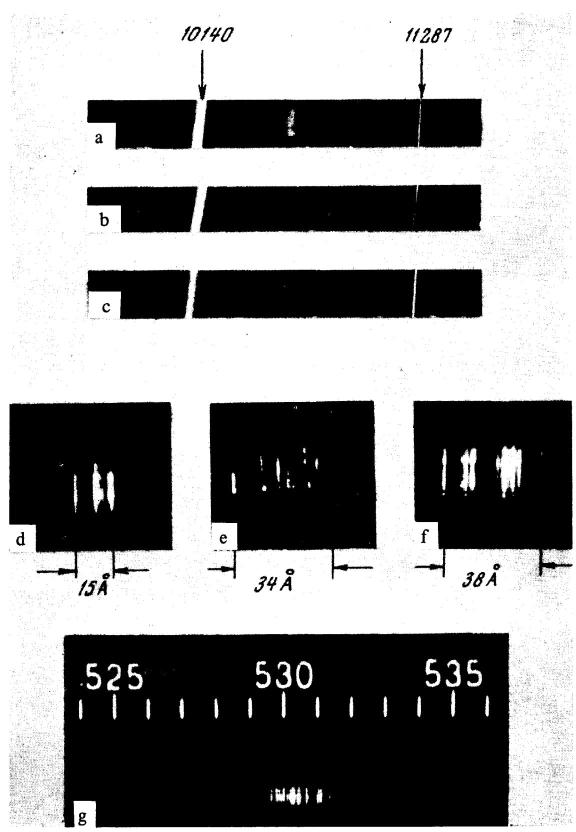

FIG. 7. Spectrograms of parametric laser and pump emission. a – T = 300°K, E = O; b – T = 124°K, E = O; c – T = 124°K, E = 5 kV/cm; d – T = 300°K, E = O; e – T = 124°K, E = 5 kV/cm; f – T = 109°K, E = E_{coerc}; g – pump spectrum.

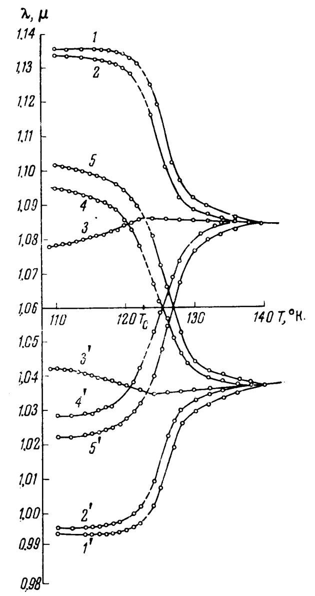

FIG. 8.

of the parametric laser and pump obtained with the SP-2 high-resolution spectrograph (PGS-2 with a dispersion D = 7 Å/mm). As we see the pump and parametric signals had a multimode character in our experiments.

Figure 8 shows the results of measuring the wavelength tuning range of the parametric laser. According to curves 3—3′ (E = 0 and only T varies) the maximum total thermal tuning reaches a value $\Delta\lambda = \Delta\lambda_1 + \Delta\lambda_2 = 510$ Å or 2.8 Å/deg. When the crystal temperature alone is varied only those branches of the tuning curves are excited for which λ_1 is the extraordinary and λ_2 the ordinary rays and $\lambda_1 > \lambda_2$. Curves 2—2′ correspond to the case[2)] of E = 3.33 kV/cm, 4—4′ to E = −3.33 kV/cm, 1—1′ to E = 5 kV/cm, and 5—5′ to E = −5 kV/cm. These curves show that the electro-optical effect begins to take hold (for the above fields) only at temperatures within 20° of T_c ($T > T_c$). In the region of $T < T_c$ the influence of the field is small due to spontaneous polarization. We were not able to achieve parametric generation directly next to T_c ($\Delta T \sim 1-2°$) because of the sharp threshold rise due to the scattering discussed above. The maximum change in wavelength due to T and E (for E = 5 kV/cm) is $\Delta\lambda_1 = 750$ Å, $\Delta\lambda_2 = 660$ Å which amounts to a total tuning range of 1410 Å.

The obtained tuning range is not the limit because the crystal cut used in this experiment yields only $0.5\ \Delta n(E)_{max}$. A different cut with increased threshold pump power may permit us to span a larger wavelength range. We must note however that according to careful measurements of $\Delta n(E)$ that we carried out in a series of electro-optical experiments[3)] there is a pronounced nonlinearity near T_c of the type

$$\Delta n(E) = aE - bE^2 - cE^3,$$

Table I

E_z, kV/cm	$\Delta\lambda_1$ exp, Å	$\Delta\lambda_1$ theor, Å
0	0	0
1	160	165
3,33	300	372
5	384	618

i.e., the linear electro-optical effect is masked by various other effects, such as the quadratic effect (Kerr effect), reducing the values of $\Delta n(E)$ and consequently of $\Delta\lambda(E)$ as compared to those ascribed only to the linear electro-optical effect. This is indeed observed experimentally when $T = T_c + 1°$ (see Table I). Computation shows that saturation of $\Delta\lambda(E)$ occurs with fields stronger than 5 kV/cm. In the region of $T < T_c$ the variation in wavelength of the parametric laser with constant temperature and varying field reveals a hysteresis as shown in Fig. 9. When the field E_z slowly varies from 5 to −5 kV/cm, jumps in $\Delta\lambda$ are observed simultaneously with the well-known Barkhausen jumps in the high-voltage rectifier ($\Delta U \sim 300$ V) circuit. The $\Delta\lambda$ jumps are due to domain rearrangement accompanying crystal polarization reversal that causes a sharp change in its optical properties (in particular, in its refraction indices) over the cross section of the light beam (see below). After two consecutive polarization reversal

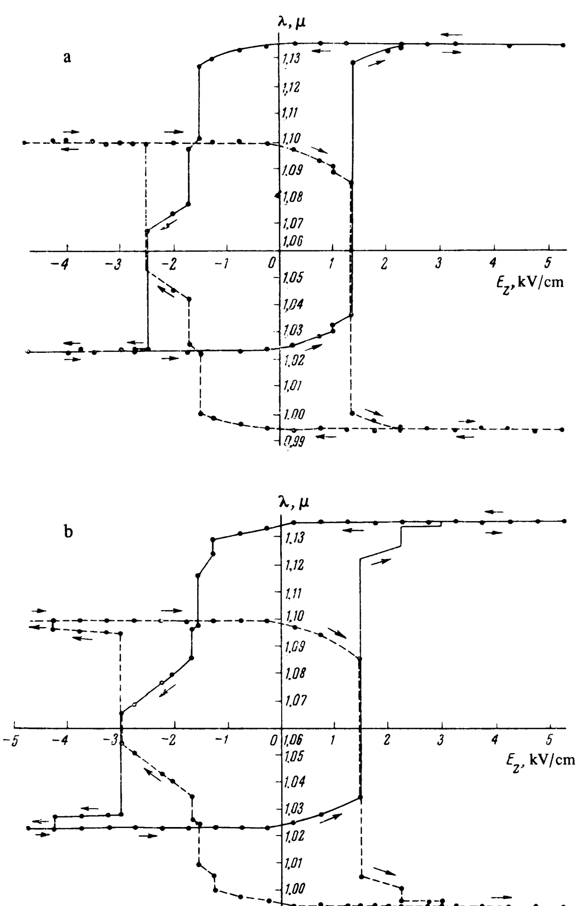

FIG. 9. Wavelength tuning of the parametric laser for two consecutive polarization reversal cycles of the crystal at $T < T_c$ (T = 109°K). a — lst cycle; b — 2nd cycle. Arrows indicate the cycling direction.

[2)]The sign of the field is arbitrary. The change in the field sign is equivalent to a 180° rotation of the crystal within the resonator in the initial field.

[3)]Yu. N. Polivanov. Thesis, Novosibirsk State University, 1967.

cycles the output wavelengths of the parametric laser fall on different, if similar, curves (see Fig. 9; cycles I and II differ in the shape of the tuning curves that show a statistical spread for the same values of the external field because of the above polarization jumps). It seems that exact (controlled) frequency tuning of the parametric laser within the range $T < T_c$ is difficult when polarization reversal of the crystal is present because the polarization jumps are of a random nature. Controlled tuning with the application of the field is obtained only in the region $T > T_c$ where the tuning hysteresis disappears.

The average power of the parametric output was 17 kW in the degenerate case and decreased toward the tuning limit. Strong fluctuations of the parametric output intensity were observed when T varied in the range $T > T_c$, and especially when $T < T_c$, in both polarization reversal cycles. In individual cases the intensity of the parametric signal exceeded that of the degenerate oscillations 5—6 times. This phenomenon seems to be due to the multimode pumping regime (mode interaction in parametric transformation of light[1]) over the entire temperature range under investigation, and also to the domain effect in polarization reversal within the $T < T_c$ range. The crystal is known to divide into two volumes with opposed domain polarization[18] during a zero-field passage through T_c in the $T < T_c$ range and also during polarization reversal under a field strength of the coercive value. The effect of the multi-domain structure on the parametric signal output appears to be due to an increase of the coherent length of the side rays of the pump beam caused by a compensation of the phase mismatch between neighboring domains (or entire domain regions) that have antiparallel polarization[15].

During the passage through T_c in the presence of a field the intensity fluctuations of the parametric output markedly decreased in the region $T < T_c$. Similar phenomena were observed during the generation of the second harmonic[19].

In the vicinity of T_c ($\pm 1°$—$2°$) parametric generation vanished while beyond this region but still near T_c the mean intensity of parametric oscillations remained approximately constant during phase transition both when $E_z = 0$ and when E_z was applied in various directions. This result is in a disagreement with second harmonic data[19] according to which the maximum intensity $I_{2\omega}$, depending on the sign of the applied field, increases or decreases about 4 times at $T < T_c$ relative to its value at $T > T_c$. This seems to be due to the difference between the interaction types. The (eoo) type interaction was used in[19] in contrast to the (eoe) type interaction used in our experiment.

According to the computation the coupling coefficient β determining the efficiency of parametric interaction and consequently the intensity of parametric signals varies in a different manner in different interaction types at the T_c symmetry jump. In the case of the (eoo) interaction $\beta_0 = -\frac{1}{2}\chi_{36} \sin\theta \sin 2\varphi$ (symmetry D_{2d} at $T > T_c$) changes either into $\beta_1 = -\frac{1}{2}\chi_{36} \sin\theta \cos 2\varphi'$ or into $\beta_2 = -\frac{1}{2}\chi_{32} \sin\theta \cos 2\varphi'$ when $T < T_c$ (symmetry $C_{2\nu}$) depending on the sign of the applied field. Since $\varphi' = \varphi \pm 45°$, $\sin\varphi = \cos 2\varphi'$ and $\beta_0 : \beta_1 : \beta_2 = \chi_{36} : \chi_{31} : \chi_{32}$, and since $\chi_{31} = \chi_{36} + \Delta\chi$, $\chi_{32} = \chi_{36} - \Delta\chi$, where $\Delta\chi$ is the change in nonlinear susceptibility during phase transition[19], then $I_{2\omega}$ undergoes a jump in value during the passage through T_c.

In the case of the (eoe) interaction $\beta_0 = \frac{1}{2}\chi_{36} \sin 2\theta \times \cos 2\varphi$ (at $T > T_c$) changes (at $T < T_c$) with the applied field of any sign into the expression

$$\beta_{1,2} = \frac{1}{4}[\chi_{24} - \chi_{15}] \sin 2\theta \sin 2\varphi'.$$

Since $\chi_{24} = \chi_{36} - \Delta\chi$, $\chi_{15} = -(\chi_{36} + \Delta\chi)$[19], $\frac{1}{4}[\chi_{24} - \chi_{15}] = \frac{1}{2}\chi_{36}$ and $\beta_0 = \beta_{1,2}$, i.e., there is no jump change in the intensity of parametric oscillations during phase transition, which is in agreement with the observed facts.

The width of the spectrum of parametric oscillations was on the average $\Delta\lambda_{par} = 13$ Å for a pumping spectrum width of $\Delta\lambda_p \approx 20$ Å. The value of $\Delta\lambda_{par}$ markedly increased near T_c reaching the value of 34 Å at $T = T_c + 3°$ for example. The broadening of the emission spectrum of the parametric laser near T_c ($T > T_c$), with constant $\Delta\lambda_p$ and pump divergence α_p, is probably due to a temperature gradient ∇T over the cross section of the pumping beam within the crystal, leading to a significant gradient ∇r_{63} (due to the strong dependence of r_{63} on T near T_c) and consequently to a spread of the values of n over the cross section of the beam, i.e., to an increase of $\Delta\lambda_{par}$.

According to computations $\nabla T = 0.5°$ in the region $T_c - (T_c + 5°)$ causes a spread in the values of $\Delta r_{63} = 0.2 \times 10^{-6}$ cm/V, which in turn, in a constant external field $E_z = 5$ kv/cm, leads to $\Delta\lambda_{par} = 36$ Å. A similar broadening to 38 Å was also observed in both polarization reversal cycles at constant temperature $T < T_c$ ($T = 109°$K). This seems to be due to the inhomogeneity of the refraction index in the pumping beam cross section resulting from domain rearrangement in separate regions of the crystal and also to increased pumping beam divergence caused by scattering at domain boundaries. The latter also leads to spectral broadening of the parametric signal. The optimization of the parametric laser and especially of its output power and emission spectrum width was not the object of this work. These parameters however can be improved by single-mode pumping.

It is of interest to compare the quantities characterizing the accuracy of wavelength tuning in various existing types of parametric lasers (see Table II). While the electro-optical tuning is the more precise, other methods offer a broader bandwidth[1,2]. Therefore a precise and rapid wavelength tuning in any range available to a given parametric laser may require the combination of electro-optical, mechanical, and thermal tuning.

CONCLUSION

The feasibility of electro-optical frequency change in a parametric laser with a KDP crystal is theoretically

Table II

Crystal used in parametric laser	type of interaction	tuning		
		mechanical, Å/min of arc	thermal, Å/degree	electro-optical Å-cm/V
KDP	*eoe*	3	1,5	0,13
$LiNbO_3$	*eoo*	8.5	260	$6{,}5 \cdot 10^{-3}$

investigated and experimentally proved in this paper. Other nonlinear materials such as $LiNbO_3$, ADP, DADP, and DKDP have no anomalously high electro-optical coefficients at T_C. DKDP is of interest since its T_C is 100° higher than that of KDP. The new material $Ba_2NaNb_5O_{15}$[20] is also of considerable interest with respect to electro-optical frequency tuning of parametric lasers since it has large nonlinear and electro-optical constants. The advantage of the electro-optical method of frequency tuning over the mechanical and thermal methods consists in its low inertia; it will be fully appreciated with the arrival of continuous parametric lasers or pulse parametric lasers with high repetition rate. Such lasers are feasible according to estimates[20,21].

In conclusion the authors consider it their pleasant duty to thank V. V. Aborin for help with the experiment, N. D. Lizunov for the high quality of preparation of KDP crystals, and M. S. Drobynina for help with the graphs.

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Translated by S. Kassel
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