Simulation of parametric generation of light pulses in nonlinear crystals pumped by 2–3 μm lasers

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Received April 9, 2003

A method is developed to solve the system of shortened equations describing parametric generation in a one-frequency OPO cavity under nanosecond pulsed pumping. The frequency conversion coefficient for $ZnGeP_2$, $AgGaSe_2$, and $HgGa_2S_4$ crystals and the optimal reflection coefficient R^2_{opt} of the output mirror are estimated versus the pump energy density W_p at different values of the OPO nonstationarity parameter γ. It is found that for any W_p an optimal value of the pump pulse duration τ_p exists stipulated by the parametric oscillation threshold. It is concluded that OPO with high (up to tens of watt) output power can be developed.

Introduction

The available optical parametric IR oscillators (OPO) are rarely used in lidar systems because of their low power characteristics and, sometimes, limited spectral tuning range. In recent years the situation has somewhat changed. Impressive results on OPO with the mean power higher than 16 W in $ZnGeP_2$ crystals pumped by Ho:YAG laser radiation (λ = 2.05 μm, τ = 6–8 ns) were obtained,[1,2] but up to now they have not found their application in atmospheric studies. The aim of this paper is to perform comparative analysis of possibilities of the high power OPO development based on $ZnGeP_2$, $AgGaSe_2$, and $HgGa_2S_4$ crystals as most promising for this purpose.

Solid-state lasers of 2–3 μm region are considered as pump sources. In this region, generation at some transitions of rare-earth ions in different activated crystals was obtained. The most efficient generation with the output parameters of radiation suitable for OPO pumping was observed at the transitions $Ho^{3+}\ {}^5I_7 \to {}^5I_8$ ($\lambda_{Ho} = 2$ μm) and $Er^{3+}\ {}^4I_{11/2} \to {}^4I_{13/2}$ ($\lambda_{Er} \approx 3$ μm) in activated YAG ($\lambda_{Ho} = 2.1$ μm, $\lambda_{Er} = 2.94$ μm), YLF ($\lambda_{Ho} = 2.05$ μm, $\lambda_{Er} = 2.81$ μm), and YSGG ($\lambda_{Ho} = 2.09$ μm, $\lambda_{Er} = 2.79$ μm) crystals pumped by diode lasers.[3] All these factors together open a possibility of creation of a simple and reliable, fully solid-state radiation source, attractive for lidar systems.

1. Mathematical model of pulsed OPO

When three waves propagate through a square-nonlinear medium: the pump wave $\mathbf{E}_p$ and two weak ones referred to as idler $\mathbf{E}_i$ and signal $\mathbf{E}_s$, the nonlinear interaction between them produces a re-emitted wave at the frequency $\omega_p - \omega_s$ with the wave vector $\mathbf{k}_p - \mathbf{k}_s$ and a wave at the frequency $\omega_p - \omega_i$ with the wave vector $\mathbf{k}_p - \mathbf{k}_i$. If the conditions $\omega_s + \omega_i = \omega_p$ and $\mathbf{k}_s + \mathbf{k}_i = \mathbf{k}_p$ are met, there arises an effective parametric interaction of the three light waves in the crystal, which leads to intensification of the signal and idler waves due to the pump wave energy. The phase matching condition $\mathbf{k}_s + \mathbf{k}_i = \mathbf{k}_p$ can be met in anisotropic nonlinear crystals through selection of the waves' polarization and the angle between the optical axis and the direction of wave propagation.

Parametric intensification of light pulses in the approximation of slowly varying amplitudes is described by the following system of "shortened" equations[4]:

$$\begin{aligned} &\mathrm{d}A_s^* / \mathrm{d}z + \delta_s A_s^* = i\sigma_s A_p^* A_i, \\ &\mathrm{d}A_i / \mathrm{d}z + \delta_i A_i = i\sigma_i A_p A_s^*, \\ &\mathrm{d}A_p / \mathrm{d}z + \delta_p A_p = -i\sigma_p A_i A_s, \end{aligned} \tag{1}$$

where A_j is the complex field amplitude connected with the real amplitude a_j and phase φ_j as $A_j = a_j \exp(i\varphi_j)$, $\sigma_j = 4\pi\omega_j d_{\mathrm{eff}} / [cn(\omega_j)]$ are the nonlinear link coefficients; d_{eff} are the effective nonlinear susceptibilities; δ_j are the absorption coefficients. Restrict our consideration to the nanosecond region of pump pulse durations. In this case we neglect the group detuning of pulses, since the calculations of the dispersion dependences show that at the pump pulse duration longer than 10^{-9} s the group lengths far exceed crystal lengths used in practice.[5,6] System (1) can be easily reduced to the system of equations for real amplitudes and the generalized phase $\Psi = \varphi_p - (\varphi_s + \varphi_i)$. In this case, the solution for $\Psi = \pi/2$ corresponds to the highest efficiency of the parametric conversion. Under this condition, from Eq. (1) we obtain the system of shortened equations for real amplitudes:

$$\begin{aligned} &\mathrm{d}a_s / \mathrm{d}z + \delta_s a_s = \sigma_s a_p a_i, \\ &\mathrm{d}a_i / \mathrm{d}z + \delta_i a_i = \sigma_i a_p a_s, \\ &\mathrm{d}a_p / \mathrm{d}z + \delta_p a_p = -\sigma_p a_i a_s. \end{aligned} \tag{2}$$

0235-6880/03/07 582-07 $02.00

Consider a nonlinear crystal with faces normal to the phase matching direction for the waves with the frequencies ω_s and ω_i. The crystal is placed in a cavity, whose axis also coincides with the phase matching direction. The cavity mirrors are fully transparent for the pump and signal waves, but for the idler wave the reflection coefficient is equal to unity for one mirror and R^2 for another. The pump wave is incident on the crystal from the side of the mirror reflecting totally the idler wave. In such a cavity, in addition to the waves propagating in the direction of the pump wave, a back idler wave arises because of reflection from the R^2 mirror. We denote amplitudes of direct waves by "+", the back wave – by "–". Assume that the back idler wave does not interact with the direct waves; therefore, it can be described by the equation

$$\mathrm{d}a_i^- / \mathrm{d}z + \delta_i a_i^- = 0. \tag{3}$$

The process of generation in OPO can be represented as a series of steps, each corresponding to a single bypass of the cavity including the following stages: propagation of direct waves, reflection from the mirror with R^2, propagation of the back wave, reflection from the totally reflecting mirror. For every step with the number N we have to solve the system of equations

$$\begin{aligned} \mathrm{d}a_{sN}^+ / \mathrm{d}z + \delta_s a_{sN}^+ &= \sigma_s a_{pN}^+ a_{iN}^+, \\ \mathrm{d}a_{iN}^+ / \mathrm{d}z + \delta_i a_{iN}^+ &= \sigma_i a_{pN}^+ a_{sN}^+, \\ \mathrm{d}a_{pN}^+ / \mathrm{d}z + \delta_p a_{pN}^+ &= -\sigma_p a_{iN}^+ a_{sN}^+ \end{aligned} \tag{4}$$

with the boundary conditions for the interaction zone boundaries (0 and L_z)

$$a_{sN}^+(0) = a_{sn}^+;\ a_{iN}^+(0) = a_{in}^+ + a_{iN-1}^-(0);\ a_{pN}^+(0) = a_{pN}^+ \tag{5}$$

for the forward pass through the cavity and

$$\mathrm{d}a_{iN}^- / \mathrm{d}z + \delta_i a_{iN}^- = 0 \tag{6}$$

with the boundary condition

$$a_{iN}^-(L_z) = R\, a_{iN}^+(L_z) \tag{7}$$

for the backward pass.

In Eq. (5) we use the following designations: a_{pN}^+ is the amplitude of the pump wave, a_{sn}^+ and a_{in}^+ are the noise amplitudes at the frequencies ω_s and ω_i at the input into the crystal. The crystal field always has fluctuations in the form of weak chaotic signals. Thanks to nonlinear interaction of these signals with the pump wave, propagation of the latter in the crystal is accompanied by the parametric luminescence, namely, re-emission of radiation at the frequencies lower than the pump frequency. The initial conditions for the signal and idler waves correspond to quantum fluctuations caused by the parametric luminescence noise and are determined by the energy density of zero states of photons $E_j^{(0)} = h\nu / 2L_r S$, here S is the beam cross section area, and L_r is the cavity length. Consequently, the noise amplitude can be written as

$$a_{jn}^+ = (2E_j^{(0)} / n_j \varepsilon_0 c)^{1/2}.$$

Assume that OPO is pumped by the Gaussian pulse with the field amplitude

$$a_{pe}^+(r, t) = a_p^0 \exp[-2(t / \tau_p)^2 - 0.5(r / w)^2], \tag{8}$$

where τ_p is the $1/e$ (power) duration of the pump pulse; w is the $1/e$ (intensity) radius of the cross distribution; a_p^0 is the maximum pump amplitude. The time $t = 0$ corresponds to the maximum pump amplitude. Usually, the duration of the pump pulse τ_p exceeds the time τ_0 needed for bypassing the OPO cavity, and in our calculations this requirement is automatically met. Therefore, Eq. (8) can be approximated by a step-wise function with the step width equal to τ_0 and the pump field constant at a step.[7] These steps correspond to the time steps in evolution of generation. For every time step N ($\tau_N = N \cdot \tau_0$) we have to solve the problem (4)–(8).

2. Calculation technique for OPO energy characteristics

The calculating technique is close to that described in Ref. 8. For practical implementation of the computational algorithm, pass on to dimensionless variables and introduce the following designations:

$$\zeta = z / L_z;\ u_{jN} = a_{jN} / a_p^0;\ \delta_j^* = \delta_j L_z;\ l_{nl} = (a_p^0 \sigma_p)^{-1}; \tag{9}$$
$$\mu = \omega_i / \omega_p;\ g = L_z / l_{nl};\ \rho = r / w;\ \gamma = 2(\tau_0 / \tau_p)^2.$$

When introducing l_{nl} and g, we believe that in the region of crystal's transparency the normal dispersion of the refractive indices is small ($n_p \approx n_s \approx n_i$) and l_{nl} includes only the refractive index at the pump wavelength n_p.

In accordance with the selected form of the pulse (8), consider the axially symmetric problem that is close to realistic situations. In this case, we take into account the dependence of the functions u_{jN} only on one cross coordinate ρ. The values of $u_{jN}(\rho)$ are specified and calculated on the grid ρ_k. The pump amplitude on the radial layer with the number k and at the time step with the number N is calculated as

$$u_{pNe}^{k+} = \exp[-\gamma N^2 - (1/2)\rho_k^2]. \tag{10}$$

For each layer and time step, the system describing the direct waves

$$\begin{aligned} \mathrm{d}u_{sN}^{k+} / \mathrm{d}\zeta + \delta_s^* u_{sN}^{k+} &= (1 - \mu) g u_{pN}^{k+} u_{iN}^{k+}, \\ \mathrm{d}u_{iN}^{k+} / \mathrm{d}\zeta + \delta_i^* u_{iN}^{k+} &= \mu g u_{pN}^{k+} u_{sN}^{k+}, \\ \mathrm{d}u_{pN}^{k+} / \mathrm{d}\zeta + \delta_p^* u_{pN}^{k+} &= -g u_{iN}^{k+} u_{sN}^{k+} \end{aligned} \tag{11}$$

is integrated numerically with the boundary conditions

$$u_{sN}^{k+}(0) = u_{sn}^{k+},\ u_{iN}^{k+}(0) = u_{in}^{k+} + u_{iN-1}^{k-}(0),$$

$$u_{pN}^{k+}(0) = u_{pN}^{k+}. \tag{12}$$

The equation describing the back wave can be integrated analytically

$$u_{iN}^{k-}(0) = Ru_{iN}^{k+}(1)\exp(-\delta_i^*). \tag{13}$$

System (11) was integrated numerically with allowance for Eqs. (10), (12), and (13) by the Runge–Kutta method with automatic step control. The calculated parameters were $S_p(t)$, $S_s(t)$, and $S_i(t)$, i.e., the shapes of the pump pulse, the signal and idler waves at the OPO output, as well as η_s and η_i the energy efficiencies of conversion into radiation with the wavelengths λ_s and λ_i. The calculation was performed by the following equations:

$$S_i(t_N) = (1 - R^2)\sum_k [\rho_k (u_{iN}^{k+})^2] / \sum_k \rho_k,$$

$$S_s(t_N) = \sum_k [\rho_k (u_{sN}^{k+})^2] / \sum_k \rho_k, \tag{14}$$

$$S_p(t_N) = \sum_k [\rho_k (u_{pN}^{k+})^2] / \sum_k \rho_k;$$

$$\eta_s = \sum_N S_s(t_N) / \sum_N S_p(t_N),$$

$$\eta_i = \sum_N S_i(t_N) / \sum_N S_p(t_N) \tag{15}$$

at different values of the system parameters g, γ, μ, R, and δ_j^*.

3. Results of calculation

To obtain a highly efficient conversion at three-frequency parametric interactions, it is necessary that the phase matching conditions were met. The calculated phase matching curves for OPO of the I type of interaction pumped by Ho, Er, and Nd laser radiation are depicted in Fig. 1. Note that for the $HgGa_2S_4$ crystal the Nd laser can be used as a pump source,[9] while the $ZnGeP_2$ and $AgGaSe_2$ crystals are opaque for radiation with the wavelength of 1 μm.

A very interesting feature is the possibility of obtaining noncritical spectral matching, when it becomes possible to generate wide-spectral radiation with a wavelength of 2.9–6.2 μm (curve *2*) in the $HgGa_2S_4$ crystal and 4.1–7.8 μm in the $ZnGeP_2$ crystal (curve *7*).

Estimate the ranges of possible variations of the input system parameters. The current level of coating allows one to produce OPO mirrors with the reflection coefficient for R^2 from 2 to 98.5%. At λ_p = 2 μm parameter μ varies from 0.35 to 0.55 and at λ_p = 3 μm – from 0.55 to 0.82.

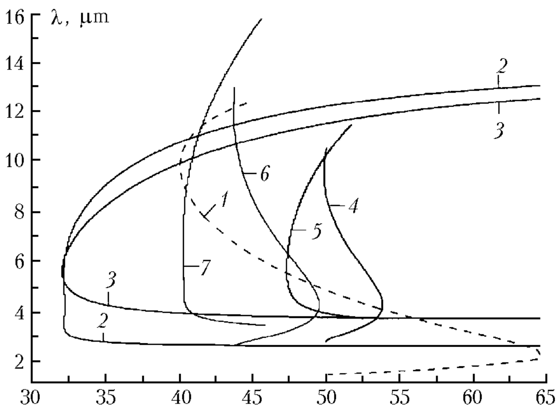

Fig. 1. Conversion phase matching curves for OPO in $HgGa_2S_4$ crystal pumped by 1.06 μm (*1*), 2.1 μm (*2*), and 2.8 μm (*3*) radiation; $ZnGeP_2$ crystal pumped by 2.1 μm (*4*) and 2.8 μm (*5*) radiation; and $AgGaSe_2$ crystal pumped by 2.1 μm (*6*) and 2.8 μm (*7*) radiation.

The optical loss coefficient 2δ in $HgGa_2S_4$ and $AgGaSe_2$ samples produced by a reproducible technology is, respectively, no more than 0.1 (Ref. 10) and 0.01 cm^{-1} (Ref. 11) for the spectral region under consideration. Reference 12 reports the growing of $ZnGeP_2$ crystals with the optical loss coefficient very small for this type of crystals: 2δ ~ 0.01 cm^{-1} in the region of 3–8.5 μm and 2δ ~ 0.05 cm^{-1} at λ_p = 2 μm (Ref. 13).

In OPO designing, a general tendency is to decrease the length of its cavity for more complete filling of the crystal with the pump radiation and abatement of losses. Assuming that in the limiting case the cavity length is equal to the crystal length, we can estimate τ_0 by the equation

$$\tau_0 = 2L_z n / c. \tag{16}$$

For L_z = 2 cm, taking into account that the values of the refractive indices of the crystals considered are about 3, we obtain $\tau_0 \approx 0.4$ ns. In our consideration the duration of the pump pulse τ_p can vary from a few nanoseconds to a few microseconds depending on the particular laser design, parameters of its active medium, Q-switching method, etc. Therefore, γ can vary widely from 10^{-2} to 10^{-8} and even smaller. From Eq. (9) and taking into account Eq. (8), we have for g

$$g = \frac{4\pi\omega_p d_{eff}}{cn_p} 0.0915 \left(\frac{W_p}{\tau_p n_p}\right)^{1/2} L_z, \tag{17}$$

where $W_p = E_p/(\pi w^2)$ is the mean energy density on the crystal surface; E_p is the pump pulse energy, the coefficient 0.0915 arises when we go from the field amplitudes written in the CGSE system to the intensity usually measured in W/cm^2. The value of W_p is restricted by the surface damage threshold equal to 1–4 J/cm^2 for the crystals under consideration. Assuming that the minimal duration of the pump pulse is 5 ns and the maximal pump energy density W_p on the crystal surface is 1 J/cm^2 and taking into account the values of d_{eff}, L_z, and n_p, we obtain that the nonlinearity parameter g can achieve the maximum value of 22.

For calculations, we took the experimentally realizable lengths of crystals (L_z) and OPO cavities (L_r): for $HgGa_2S_4$ and $ZnGeP_2$ $L_z = 1$ cm and $L_r = 1.5$ cm, for $AgGaSe_2$ $L_z = 2$ cm and $L_r = 2.5$ cm. For the fixed values of W_p and γ, the optimal reflection coefficient of the exit mirror R^2_{opt} was determined, at which the coefficient of energy conversion into the given spectral range η_i reached its maximum.

Figures 2 and 3 depict the conversion coefficients η_i and the optimal reflection coefficient of the exit mirror R^2_{opt} vs. the pump energy density W_p for different values of OPO nonstationarity parameter γ based on $HgGa_2S_4$ and $AgGaSe_2$ crystals pumped at $\lambda_p = 2.1$ μm. Analysis of dependences in Figs. 2 and 3 shows that for any value of W_p there is a value of γ that is optimal with respect to the value of η_i and, consequently, the optimal duration of the pump pulse τ_p. As γ decreases (τ_p increases), R^2_{opt} and the generation threshold increase. The threshold energy increases, because the input pump intensity decreases at the given value of W_p. At higher threshold energy, parametric generation develops more efficiently due to the increase of the total interaction length with increasing τ_p.

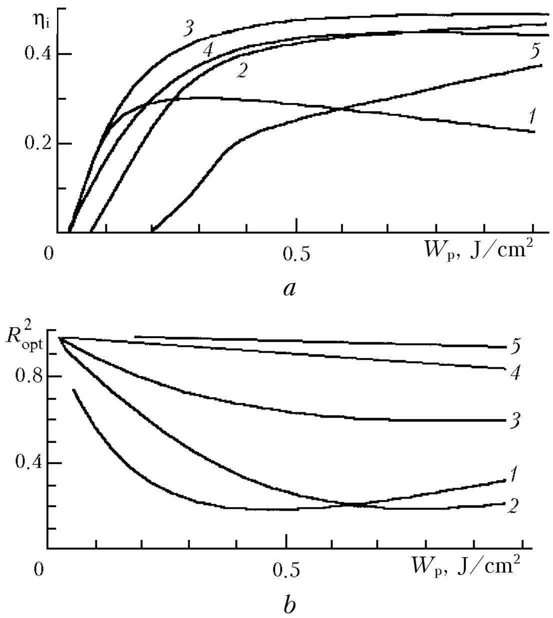

Fig. 2. The conversion coefficient (*a*) and the optimal reflection coefficient of the exit mirror (*b*) for OPO based on $HgGa_2S_4$ pumped by radiation at $\lambda_p = 2.1$ μm vs. the pump energy density at different values of γ: 10^{-3} (*1*), 10^{-4} (*2*), 10^{-5} (*3*), 10^{-6} (*4*), and 10^{-7} (*5*).

At $W_p = 0.5$ J/cm^2 that certainly guarantees no damage of the crystal surface, $\gamma = 10^{-5}$ is optimal in the OPO operating mode close to degenerate. In this case, η_i is ~ 0.47 for OPO based on $HgGa_2S_4$ and ~ 0.42 for OPO based on $AgGaSe_2$. Despite the absorption coefficient for the pump radiation in the $HgGa_2S_4$ crystal ($2\delta \sim 0.1$ cm^{-1}) is higher than in $AgGaSe_2$ ($2\delta \sim 0.01$ cm^{-1}), η_i is somewhat higher.

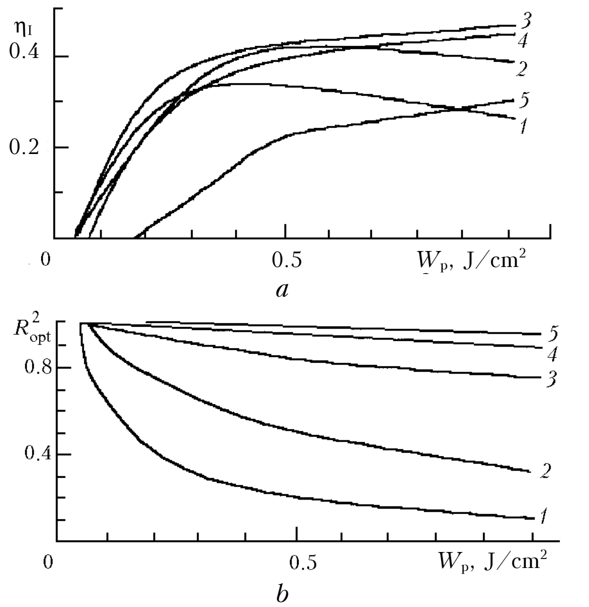

Fig. 3. The conversion coefficient (*a*) and the optimal reflection coefficient of the exit mirror (*b*) for OPO based on $AgGaSe_2$ pumped by radiation at $\lambda_p = 2.1$ μm vs. the pump energy density at different values of γ: 10^{-3} (*1*), 10^{-4} (*2*), 10^{-5} (*3*), 10^{-6} (*4*), and 10^{-7} (*5*).

In addition, note that in the comparative experiment on studying the surface damage threshold the optical damage threshold of $HgGa_2S_4$ crystals turned out to be 2.45 times higher than that of $AgGaSe_2$ crystals and 2.2 times higher than in $ZnGeP_2$ crystals.[14] Since the pump radiation with $\lambda_p = 2.8$ μm is almost identically absorbed by the $ZnGeP_2$ and $AgGaSe_2$ crystals and $ZnGeP_2$ nonlinearity is higher than that of $AgGaSe_2$, the calculations were performed only for the former. For this crystal, at $\lambda_i = 4.6$ μm for $W_p = 0.5$ J/cm^2 the optimal value is $\gamma = 10^{-4}$ and in this case η_i ~ 0.3.

Figure 4 illustrates variation of η_s and η_i at OPO detuning from the mode close to degenerate. As λ_s increases and λ_i decreases, the value of η_s drops down, while η_i grows, and $\eta_s + \eta_i$ keeps roughly the same. At the pre-selected crystal and cavity lengths, the values of $\gamma = 10^{-4} - 10^{-5}$ correspond to the pump duration $\tau_p = 30$–160 ns, which is readily achievable, since there exist various methods to change pulse duration in the region of 10 ns–4 μs without significant energy losses with conservation of the pulse shape, for example, through the use of specialized cavities with a modulator employing the effect of violation of total internal reflection.[15]

Of significant interest are Nd:YAG-laser pumped OPOs frequency tunable in the entire mid-IR region. New nonlinear crystals $AgGaGeS_4$ and $HgGa_2S_4$ can be used in them, since their birefringence is sufficient to meet the phase matching conditions and they are transparent for the pump radiation.[10] The table

Parameters of OPO based on widely used and new nonlinear crystals

Crystal	λ_p, μm	$\Delta\lambda_s$, μm	$\Delta\lambda_i$, μm	$\Delta\theta_p$, deg	Δd_{eff}, pm/V	Type of interaction, plane	I_d, rel. units
$ZnGeP_2$	2.1	2.53–4.3	4.312.4	44–49.4	51–58	(o–ee)	1
	2.8	3.9–5.6	5.6–12.4	46–55	53–62.9	(o–ee)	1
$AgGaSe_2$	1.06	1.17–1.18	12.5–13.5	75–90	35.0–32.0	(e–oo)	0.9
	2.1	2.42–4.1	4.1–13.5	43–48.6	22.5–25.0	(e–oo)	0.9
$HgGa_2S_4$	0.53	0.55–0.58	6.51–13.0	49–90	24.0–18.0	(e–oo)	2.2
	1.06	1.16–2.13	2.13–13.0	38.5–61.5	21.0–15.0	(e–oo)	2.2
	2.1	2.43–4.1	4.1–13.0	35.4–42.3	16.0–14.5	(e–oo)	2.2
GaSe	2.8	3.3–5.7	5.7–19	29–40	41.4–47.2	(e–oo)	0.85
$AgGaGeS_4$	1.06	1.2–2.13	2.13–11.5	33.4–48.1	9.0–6.5	XZ, (e–oo)	1.6
	1.06	1.2–2.13	2.13–11.5	35.6–38.3	9.4–9.5	XY, (e–oo)	1.6

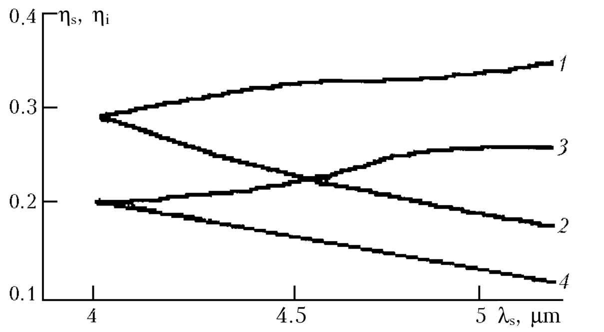

Fig. 4. Coefficient of conversion in $AgGaSe_2$ [λ_s (*1*), λ_i (*2*)] and $ZnGeP_2$ crystals [λ_s (*3*), λ_i (*4*)] for $\gamma = 10^{-4}$, $R^2 = 0.65$, $W_p = 0.5$ J/cm^2, $\lambda_p = 2.1$ μm.

presents some of our calculated data on OPO based on the crystals considered, as well as $AgGaGeS_4$, $HgGa_2S_4$, and GaSe crystals. The GaSe crystals are considered here, since, as follows from our calculations and some experiments,[1] they allow ultrawide spectral tuning. The main difficulty associated with the use of these crystals is their very low thermal and mechanical properties (cleavage) and, as a consequence, impossibility of mechanical treating and cutting optical elements at the needed angles to the optical axis. However, this difficulty can be overcome by doping the initial In crystal or growing mixed GaSe:InSe crystals.[16]

In the table $\Delta\theta_p$ is the range of variation of the phase matching angles; I_d is the optical damage threshold of the crystals relative to $ZnGeP_2$ crystals. It also follows from tentative estimates that, besides the possibility of Nd:YAG-laser pumped OPO, new $AgGaGeS_4$ and $HgGa_2S_4$ crystals (with ignoring of their thermal properties to be determined) allow one to realize OPO with the same and, possibly, even higher efficiency than in the high-quality $ZnGeP_2$ crystals.

4. Estimation of the OPO output beam line width

In solving some problems of atmospheric optics, an important parameter is the OPO line width $\Delta\nu$, which can be estimated by the following equation:

$$\Delta\nu = \Delta\nu_c / p^{1/2}, \tag{18}$$

where $\Delta\nu_c$ is the parametric gain line width for a given crystal; p is the number of bypasses of the OPO cavity. To calculate $\Delta\nu_c$, let us use the well-known equations[17]:

$$\Delta\nu_c = \sqrt{2(\ln 2)^{1/2} g^{1/2} / (L_z \vartheta_{12})} \tag{19}$$

for the mode close to degenerate and

$$\Delta\nu_c = 2(\ln 2)^{1/2} [g\mu(1-\mu)]^{1/2} / (L_z \nu_{12}) \tag{20}$$

far from degeneration, where

$$\nu_{12} = |\partial k_i / \partial\nu_i - \partial k_s / \partial\nu_s|;$$

$$\vartheta_{12} = |\partial^2 k_i / \partial\nu_i^2 + \partial^2 k_s / \partial\nu_s^2|. \tag{21}$$

Figure 5 depicts $\Delta\nu_c$ as a function of the OPO radiation wavelength for the crystals considered. Specifying the duration of the pump pulse τ_p, we can estimate p as

$$p = \tau_p / \tau_0. \tag{22}$$

For $\tau_p = 100$ ns the OPO line width is 4–5 cm^{-1} at pumping at $\lambda_p = 2.1$ μm in the mode close to degenerate; it is ~ 1.2 cm^{-1} at $\lambda_i = 4.6$ μm for $AgGaSe_2$ and ~ 2.5 cm^{-1} for $ZnGeP_2$; and it equals ~ 1.3 cm^{-1} for $AgGaSe_2$ and ~ 0.4 cm^{-1} for $ZnGeP_2$ when pumping at $\lambda_p = 2.8$ μm and $\lambda_i = 4.6$ μm.

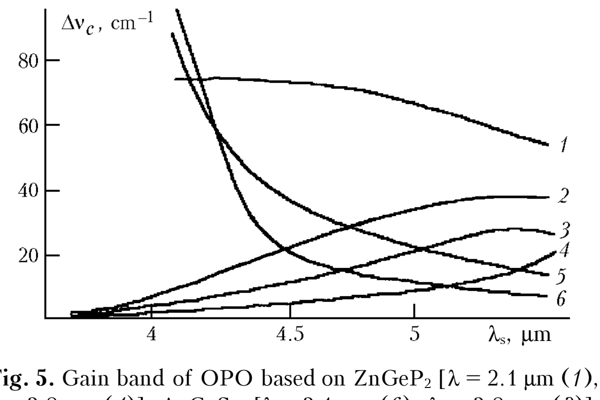

Fig. 5. Gain band of OPO based on $ZnGeP_2$ [$\lambda = 2.1$ μm (*1*), $\lambda = 2.8$ μm (*4*)]; $AgGaSe_2$ [$\lambda = 2.1$ μm (*6*), $\lambda = 2.8$ μm (*2*)]; $HgGa_2S_4$ [$\lambda = 1.06$ μm (*5*), $\lambda = 2.1$ μm (*3*)].

5. Analysis of correctness of the calculations presented

Consider the correctness of the simplifying assumptions lying in the basis of the system of equations (2) that describes the three-frequency parametric interaction. With the mean output power of the order of several watts and the pulse repetition frequency of 1 kHz, the pump pulse energy should be 10–20 mJ. At the fixed value of $W_p = 1$ J/cm^2 this condition determines the minimal radius of the pump beam $w = 0.07$–0.1 cm. At this value of w the diffraction length $l_d = 2\pi w_0^2 n/\lambda$ more than an order of magnitude exceeds the lengths of the crystals used, therefore in this case we can neglect the diffraction blooming of the beam. The aperture length $l_w = 2w/\beta$, where β is the anisotropy angle, several times exceeds the length of the existing elements for $HgGa_2S_4$, $ZnGeP_2$, and $AgGaSe_2$, therefore the neglect of the drift effect in the system of equations (2) is also justified when considering these types of crystals. The need to take into account the wavelength detuning Δk at parametric generation is determined by the divergence $\Delta\Theta_p$ and nonmonochromaticity of the pump radiation $\Delta\nu_p$, as well as by the temperature inhomogeneity of the nonlinear medium ΔT. The dependence of Δk on these parameters in the linear approximation is described by the equation

$$\Delta k = \partial(\Delta k / \partial T)\Delta T + \partial(\Delta k / \partial\Theta)\Delta\Theta_p + +(\partial(\Delta k / \partial\nu)\Delta\nu_p. \tag{23}$$

The presence of the detuning $\Delta k = \pi/L_z$ almost halves the conversion efficiency.[4] Based on the dispersion and temperature dependences of the refractive indices, we have estimated the angular, spectral, and temperature widths of the phase matching using the known equations.[18]

The spectral width $\Delta\nu$ of the phase matching for the crystals considered is 0.4–3.2 cm^{-1}, while spectral width of solid-state lasers does not exceed 10^{-2} cm^{-1} when using longitudinal mode selectors, therefore the wavelength detuning caused by the spectral composition of the pump radiation can be neglected in calculations. The angular width $\Delta\Theta$ of phase matching in the crystals considered at $\lambda_p = 2$–3 μm is 2–8 mrad, which is several times higher than the divergence realized for solid-state lasers.

The inhomogeneous component of the radial temperature distribution ΔT, which causes formation of the thermal lens and the wavelength detuning that cannot be compensated by the crystal turn, was estimated in Ref. 19. The thermal lens causes an additional divergence of the pump radiation. The focal length of the induced thermal lens f_T and the related divergence Θ_T can be estimated as[20]

$$\Theta_T = w / f_T = w(\partial n / \partial T)W_p f_r \delta L_z / \kappa. \tag{24}$$

where κ – is the coefficient of thermal conductivity.

The estimates for $ZnGeP_2$ at $W_p = 1$ J/cm^2 and the pulse repetition frequency of $f_r = 1$ kHz give $\Theta_T = 40$–45 mrad. Thus, in the case of pumping at $\lambda_p = 2.1$ μm, the noticeable absorption at λ_p and the resulting heating of the crystal restrict the maximal mean power convertible without loss in efficiency to the level of 5–7 W because of formation of the thermal lens and temperature-caused phase mismatch. These values follow from the comparison of Θ_T, angular width of phase mismatch $\Delta\Theta = 5$ mrad, temperature width of phase mismatch $\Delta T \approx 15°$, and the inhomogeneous component of the radial temperature distribution equal to 60° (Ref. 19). However, there is a way to decrease the 2-μm absorption in $ZnGeP_2$ due to improved technology of the crystal growing and after-growth treatment.[13] For pumping at $\lambda_p = 2.8$ μm, $\Theta_T = 1.6° \ll \Delta\Theta = 7.2°$, and the conversion is performed without a loss in efficiency. Unfortunately, the data on thermo-optical properties of the $HgGa_2S_4$ crystal are lacking in the scientific literature, and this fact did not allow us to calculate its Θ_T. The similar analysis of results of estimation for $AgGaSe_2$ shows that the main cause for the conversion efficiency decrease at a high mean pump power is an appearance of the thermal lens. The level of the mean power convertible in $AgGaSe_2$ without some loss in efficiency is restricted to 8–10 W and can be increased through the cryogenic cooling, since in this case the heat conductivity increases 5 to 6 times, while the value of dn/dT decreases 2 to 3 times.[21]

Conclusion

The possibilities of creating high-power OPO pumped by 2–3-μm lasers have been comparatively analyzed. The technique developed has allowed a calculation of the dependence of the maximum conversion coefficient η_i and the optimal reflection coefficient of the exit mirror R^2_{opt} on the pump energy density W_p for different values of the nonstationarity parameter γ for OPO pumped at $\lambda_p = 1.06$, 2.1, and 2.8 μm. It was found that for any value of W_p there exists a value of γ (pump pulse duration τ_p) optimal with respect to η_i. As γ decreases (τ_p increases), R^2_{opt} and the generation threshold increase. The line width of the output beam has been estimated and the assumptions used when drawing the system of related shortened equations, describing the parametric interaction in the cavity, partly filled with a nonlinear medium, have been checked for correctness. If the optical loss level in the $ZnGeP_2$ crystal is decreased down to 0.01 cm^{-1} in the region of 2 μm, this crystal will be preferable for creation of efficient OPO as compared to $AgGaSe_2$. At the same time, the $HgGa_2S_4$ crystal is the best for creation of wide spectral OPO pumped by the Nd:YAG-laser radiation.

Acknowledgments

I would like to express my gratitude to M.M. Makogon for useful criticism and discussions.

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