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Optical parametric oscillator on an $Hg_{1-x}Cd_xGa_2S_4$ crystal

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***Abstract*. Lasing was obtained for the first time in an optical parametric oscillator (OPO) on an $Hg_{1-x}Cd_xGa_2S_4$ crystal pumped by a nanosecond Nd : YAG laser. Due to the cadmium concentration gradient along the crystal axis, the OPO could be tuned under noncritical phase-matching conditions by a linear displacement of the crystal. The tuning range was 2.85 – 3.27 μm, with the maximum slope conversion efficiency equal to 6.6 %.**

Keywords: *optical parametric oscillator, cadmium-mercury thiogallate, Nd : YAG laser.*

1. Introduction

One of the possible methods for developing tunable lasers in the mid-IR range (3 – 12 μm), which are of great practical interest at present, is the elaboration of optical parametric oscillators (OPOs). Only a few nonlinear crystals are known that can be used in OPOs operating in this spectral range; the most promising are $ZnGeP_2$, $AgGaSe_2$, $AgGaS_2$, and $HgGa_2S_4$ [1 – 5]. All these crystals have different disadvantages, which restrict their use in OPOs. Because of this, the growth technology of already known crystals is constantly being improved, and the search for new nonlinear optical media is being continued.

Note that of special interest are crystals that can be used in mid-IR OPOs pumped by Nd lasers, which are the most widespread and efficient. Most often, silver thiogallate ($AgGaS_2$) and mercury thiogallate ($HgGa_2S_4$) are used in such OPOs. $HgGa_2S_4$ crystals can be more promising in the future due to their high nonlinearity, high damage threshold and good heat conduction. In addition, the admixture of cadmium allows one to obtain solid solutions of cadmium – mercury thiogallate ($Hg_{1-x}Cd_xGa_2S_4$, which possess a number of advantages.

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First, doping with cadmium at different concentrations x leads to a change in birefringence and the principal values of refractive indices in crystals. As a result, by varying x, one can control the phase-matching conditions. In particular, at a certain concentration of cadmium in a crystal, the conditions of noncritical phase matching can be realised.

Second, the natural gradient of the cadmium concentration along a boule length appears during the crystal growth. This circumstance permits the fabrication of elements for nonlinear-optical converters with a continuously changing x in the phase-matching plane. By using such elements, an OPO can be tuned by linearly moving the crystal with respect to the pump beam (or moving the pump beam with respect to the crystal). In this case, the conditions of noncritical phase matching are not violated.

Recently, due to the development of the growth technology of single crystals at the High Technologies Laboratory of Kuban State University, the $Hg_{1-x}Cd_xGa_2S_4$ crystals of good optical quality with the cadmium concentration continuously varying in the phase-matching plane were grown. In this paper, we demonstrate for the first time parametric lasing in these crystals pumped by a Q-switched Nd : YAG laser and present the results of the study of the basic characteristics of the OPO.

2. Growth of $Hg_{1-x}Cd_xGa_2S_4$ crystals

Single crystals of solid $Hg_{1-x}Cd_xGa_2S_4$ solutions were grown by the Bridgman – Stockbarger technique in quartz ampoules with the internal diameter of 18 mm and the wall thickness of 4.5 mm. A quartz ampoule was filled with a preliminary synthesised mixture of the required chemical composition consisting of high-purity Hg, Cd, Ga, and S (99.9999 %). Before synthesis, the ampoule with a mixture was evacuated down to the residual air pressure 2×10^{-5} Torr and sealed off. The volume ratio of the gas phase over the melt and the melt in the growth ampoule was 1 : 1. Crystals were grown on seeds oriented perpendicular to the (110) crystallographic plane at a rate of 2 – 6 mm per day at different temperature gradients (from 2 to 10 °C cm^{-1}) in growth furnaces. The crystallisation temperature changed from 1000 to 918 °C depending on the chemical composition of the melt.

During the growth of $Hg_{1-x}Cd_xGa_2S_4$, the chemical composition changes over the boule height along the crystal axis. In the lower part of the boule, the crystal is enriched by cadmium whose concentration gradually decreases upward over the boule. This occurs because cadmium – mercury thiogallate is a solid solution of a variable chemical composition and the distribution coefficient of cadmium is smaller than unity. In these crystals, twins can be formed in the (112) direction. The (101) plane is the cleavage plane.

From the grown crystal, an element with the transverse size 30.6×8 mm and length 11 mm was cut, which was oriented to realise the type I noncritical phase matching (e $\rightarrow$ oo) ($\theta = 90°$, $\varphi = 45°$). Weak change in colour was observed along the crystal axis from one crystal face to another. The optical surfaces of the element were covered with AR coatings. The reflectivity from each surface was 0.2 % – 0.8 % in the spectral range from 1 to 2 μm.

3. Experimental study of the OPO

The OPO was pumped by a *Q*-switched, travelling-wave Nd : YAG laser with a ring semi-confocal resonator. Lasing at the TEM_{00} fundamental mode was provided with a 1.7-mm aperture placed inside the ring resonator. The FWHM of the output pulse was 22 ns and the maximum pulse energy was 12 mJ. The laser operated at a pulse repetition rate of up to 10 Hz.

The OPO resonator was formed by two plane dielectric mirrors. The input mirror was made of the KV fused silica with the AR coating (at the pump wavelength) on the front surface, while the output mirror was made of calcium fluoride. The reflectivity of the input and output mirrors was 95 % – 99 % in the wavelength range 1.4 – 1.9 μm. Therefore, the *Q* factor of the resonator for the signal wave was maximal, whereas the idler wave was coupled out of the resonator with minimal losses (the transmission coefficient of the input mirror was 80 % – 90 %). The reflectivities of the input and output mirrors for the pump wave were 10 % and 30 %, respectively, and the distance between the OPO resonator mirrors was 3 cm. The OPO was located in the pump beam waist formed by a focusing mirror ($f = 2$ m). The beam waist diameter at the $1/e^2$ level in the interaction region with the crystal was 2 mm. The OPO was tuned by moving linearly the crystal with respect to the pump beam in the direction parallel to its axis.

The pump pulse energy and the intensity distribution over the beam cross section were measured by splitting a fraction of radiation with an optical wedge and detecting it with a PE-10-BB energy meter (Ophir) and a C4915 camera (Cohu). The output OPO energy was measured with the same energy meter. The idler wave was separated using a set of calcium fluoride filters with a dielectric coating (with the transmission coefficient for pump radiation less than 1 %) and a Ge plate. The wavelength was measured with a Digikrom CM112 monochromator. A signal transmitted through the monochromator was detected with an MG-30 pyroelectric detector and a TDS-1012 oscilloscope. The OPO radiation was focused by a barium fluoride lens with the focal distance 60 mm. The accuracy of the wavelength measurement was 0.01 μm. The spatial distributions of the radiation intensity over the beam cross section for the idler and signal waves were controlled with a Pyrocam-III camera (Spiricon).

We measured the output energy E_i of the OPO at the idler-wave frequency, the pump energy E_p, and the wavelength λ_i of the idler wave. The energy conversion coefficient $\eta = E_i/E_p$ was determined from the measured energies E_i and E_p. The slope conversion coefficient η_s was determined by the slope of the approximating dependence $E_i(E_p)$.

The following results were obtained. Tunable parametric lasing was obtained for the first time in a cadmium – mercury thiogallate crystal in the spectral range from 2.85 to 3.27 μm for the threshold pump energy from 2.5 to 4 mJ (for different positions of the crystal). Figure 1 shows the dependence of the OPO output energy for the idler wave on the pump energy. The maximum energy conversion coefficient for the idler wave ($\lambda_i = 3.03$ μm) was 4.5 % and the slope conversion coefficient was 6.6 %.

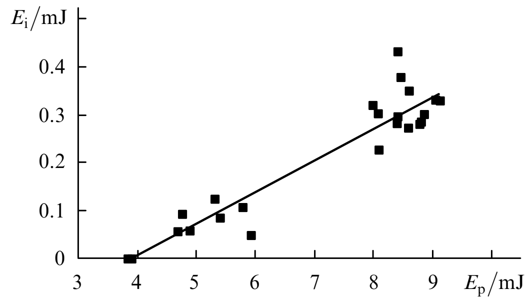

Figure 1. Dependence of the OPO output energy at the idler-wave frequency on the pump energy ($z = 11$ mm).

Figure 2 shows the dependences of the measured energy conversion coefficient η and the slope conversion coefficient η_s on the pump-beam position. For $z = 0 - 25$ mm, they vary within a comparatively narrow range, which demonstrates the homogeneity of the cadmium distribution along the crystal axis. The conversion efficiency decreases noticeably at the crystal edge (~ 3 mm), which is probably explained by considerable phase distortions in this region.

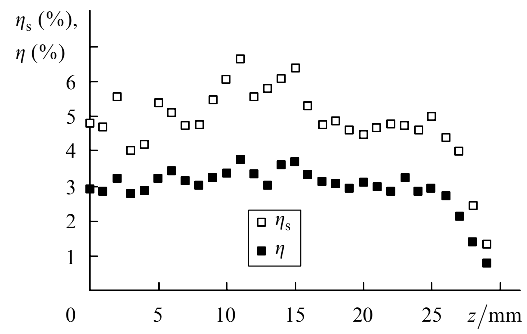

Figure 2. Dependences of the maximum and slope energy conversion coefficients on the displacement of the pump beam with respect to the side surface of the crystal (in the phase-matching plane).

We measured the spatial distributions of the radiation intensity over the idler-wave cross section in the near- and far-field zones (Fig. 3). These distributions characterise the optical quality of the crystal. As a whole, the obtained OPO radiation distributions show that the inhomogeneities of the refractive index in the crystal are very small at least in the interaction region of the waves. The prolate shape is probably caused by the radiation 'spreading' in the phase-matching plane. The divergence of the idler wave of the OPO estimated from Fig. 3 was $\sim 2 \times 10^{-2}$ rad at the $1/e^2$ level.

Figure 3. Distributions of the radiation intensity over the beam cross section at the OPO output at an the idler-wave frequency (λ = 3.03 μm) for $z = 11$ mm in the near-field (the distance from the OPO output mirror is 160 mm) (a) and far-field (at the focus of a lens with $f = 100$ mm) (b) regions.

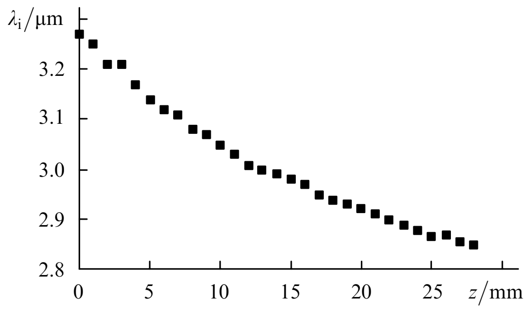

Figure 4. Dependence of the idler-wave wavelength at the OPO output on the displacement of the pump beam with respect to the side surface of the crystal.

Figure 4 presents the experimental dependence of the idler-wave wavelength on the position of the pump beam in the crystal. Upon the displacement of the pump beam in the phase-matching plane by 28 mm, the wavelength changed by 0.42 μm.

4. Dispersion dependences for the $Hg_{1-x}Cd_xGa_2S_4$ crystals

To find the relation between the chemical composition of the $Hg_{1-x}Cd_xGa_2S_4$ crystal and its optical properties, we measured the dispersion of refractive indices. For this purpose, crystal prisms were fabricated with $x = 0.27$ and 0.33 and the angle of refraction $\sim 20^\circ$ (with the 10×10-mm operating faces). The refractive indices were measured in the spectral range 0.55–10 μm by the autocollimation method using the setup described in [6]. The absolute measurement error for good-quality crystals did not exceed 0.001. The results of these measurements at room temperature are presented in Table 1.

The measured values of the refractive index were approximated by the Sellmeyer equation [5, 7]

$$n^2 = A_1 + \frac{A_3}{A_2 - \lambda^2} + \frac{A_5}{A_4 - \lambda^2}, \quad (1)$$

where λ is the radiation wavelength in μm. The Sellmeyer coefficients at other concentrations of cadmium were determined from the values of the refractive indices found by interpolating by x the dependences $n_o(\lambda)$ and $n_e(\lambda)$ for $x = 0.27$ and 0.33 and also similar dependences measured in papers [7, 8]. The Sellmeyer coefficients found in this way for crystals with different concentrations of cadmium are presented in Table 2.

Table 1. Principal values of the refractive indices for $Hg_{1-x}Cd_xGa_2S_4$ crystals.

λ/mμ	$x = 0.33$		$x = 0.27$	
	n_o	n_e	n_o	n_e
0.550	2.5659	2.5342	2.5810	2.5445
0.600	2.5319	2.5018	2.5437	2.5077
0.650	2.5053	2.4726	2.5160	2.4811
0.700	2.4838	2.4557	2.4948	2.4611
0.800	2.4556	2.4260	2.4662	2.4337
0.900	2.4374	2.4094	2.4481	2.4161
1.000	2.4253	2.3976	2.4344	2.4039
1.100	2.4163	2.3873	2.4258	2.3956
1.200	2.4110	2.3815	2.4189	2.3882
1.300	2.4051	2.3765	2.4134	2.3829
1.400	2.4003	2.3727	2.4095	2.3803
1.500	2.3965	2.3689	2.4057	2.3765
1.600	2.3922	2.3649	2.4027	2.3738
1.700	2.3895	2.3621	2.4003	2.3713
1.800	2.3873	2.3610	2.3984	2.3689
1.900	2.3848	2.3591	2.3970	2.3670
2.000	2.3834	2.3578	2.3958	2.3654
3.000	2.3756	2.3501	2.3873	2.3582
4.000	2.3695	2.3441	2.3805	2.3525
5.000	2.3656	2.3397	2.3749	2.3473
6.000	2.3587	2.3322	2.3690	2.3406
7.000	2.3515	2.3253	2.3589	2.3304
8.000	2.3444	2.3171	2.3493	2.3204
9.000	2.3332	2.3059	2.3381	2.3104
10.000	2.3211	2.2938	2.3259	2.2953

Table 2. Sellmeyer coefficients for $Hg_{1-x}Cd_xGa_2S_4$ crystals.

x	n	A_1	A_2	A_3	A_4	A_5
0.20	n_o	7.17117	639.04	−914.648	0.0979835	−0.220006
	n_e	7.03365	653.431	−943.36	0.0770492	−0.210144
0.25	n_o	7.42819	749.29	−1295.98	0.0880175	−0.219608
	n_e	7.3628	783.563	−1411.87	0.0801083	−0.207095
0.30	n_o	7.62356	836.474	−1642.64	0.0717384	−0.220695
	n_e	7.41107	805.268	−1512.04	0.0714881	−0.207339
0.35	n_o	8.47402	1167.86	−3327.19	0.0635593	−0.2180798
	n_e	8.40374	1171.46	−3389.17	0.0807452	−0.2007114
0.40	n_o	8.47346	1184.47	−3414.44	0.0406186	−0.220462
	n_e	8.33498	1143.19	−3255.20	0.0705351	−0.20029
0.45	n_o	8.50139	1213.14	−3569.69	0.0180232	−0.221991
	n_e	8.45613	1182.89	−3536.20	0.0653473	−0.197853
0.50	n_o	8.47455	1222.13	−3599.99	−0.00379508	−0.222829
	n_e	8.42368	1168.94	−3479.53	0.0602277	−0.19524

The maximum deviation of the approximated values of the refractive index from experimental values was 3×10^{-3}.

By using the obtained Sellmeyer equations and the 90° phase-matching wavelengths calculated from them, we determined the concentrations of cadmium in the crystal. For $z = 0$ and 28 mm, the cadmium concentration was $x =$ 0.25 and 0.21, respectively. Figure 5 presents the dependence of the cadmium concentration on the displacement of the pump beam in the phase-matching plane obtained by

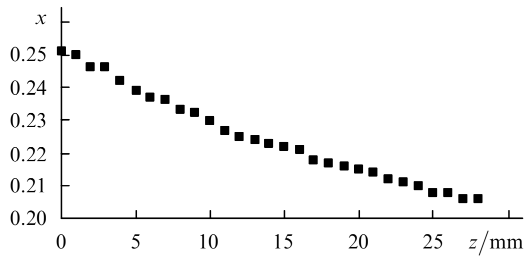

Figure 5. Dependence of the calculated concentration of cadmium in the crystal on the distance from the side surface of the crystal.

using the experimental values of the idler-wave wavelengths. One can see that the cadmium concentration continuously, almost linearly, varies along the element length. This also confirms good optical quality of the crystal.

5. Conclusions

We have developed the growth technology and grown a new nonlinear-optical crystal, cadmium – mercury thiogallate ($Hg_{1-x}Cd_xGa_2S_4$), which has good optical quality and the homogeneous distribution of the Cd impurity along the phase-matching axis of the crystal. The crystal was used to built a Nd : YAG-pumped OPO continuously tunable in the mid-IR range from 2.85 to 3.27 μm with the maximum slope conversion efficiency 6.6 %. The Sellmeyer coefficients were determined for solid $Hg_{1-x}Cd_xGa_2S_4$ solutions. The cadmium concentration in the crystal was determined over the converted radiation wavelength. Thus, we found that the parameter x changed along the crystal axis from 0.21 to 0.25. It should be emphasised that the crystal was oriented to realise noncritical phase matching and the wavelength was tuned by moving the laser beam linearly along the crystal axis perpendicular to the phase-matching axis. The continuous tuning range of the OPO was limited only by the properties of the crystal determined by the cadmium concentration. By varying the parameter x from 0.14 to 0.56, we obtained OPO lasing under the conditions of noncritical phase matching in the spectral range from 2.7 to 9 μm.

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