Optical Parametric Oscillation in Hg_{1-x}Cd_xGa_2S_4

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Abstract-- In this work, parametric interaction is investigated with the account for phase change of interacting pump, signal and idler waves in the materials of infrared range of spectrum in case of Hg_{1-x}Cd_xGa_2S_4 crystal. We give an analysis of the threshold intensity of pumping for Hg_{1-x}Cd_xGa_2S_4 crystal under conditions of the existing experiment. The values of refractive indices, angle of phase matching and angular dispersion coefficient of the second order have been calculated for Hg_{1-x}Cd_xGa_2S_4 at 90-degree phase matching.

Index Term-- parametric interaction, mid-IR region, constant-intensity approximation, 42.65.-k; 42.65.Yj; 42.70.Mp; 42.79.Nv

1. INTRODUCTION

Tunable parametric sources of coherent radiation in combination with the effects of frequency mixing permit to extend considerably the field of tunable wavelengths of laser radiation. The optical multipliers of frequency serve this task too. The optical parametric generators of the mid-infrared region of spectrum are successfully used for the tasks of spectroscopy, on their base LIDARs, in particular for investigation of the Earth and atmosphere are being worked out.

There is a few nonlinear crystals used for the mid-infrared range of a spectrum. Among them it is possible to note ZnGeP_2, AgGaSe_2, AgGaS_2 and HgGa_2S_4. Nowadays, non-oxide crystals of HgGa_2S_4 and Hg_{1-x}Cd_xGa_2S_4 are used actively. These crystals are applied successfully at the mid-infrared spectral range for parametric generation. That means the interest in using these kind of chalcogenide crystals are increased due to technological elaboration of similar structures, namely because of obtaining this crystals with the perfection in optical quality and large size [1-2].

Choosing mercury thiogallate crystals are because of some considerations. First of all based on this crystal we can produce parametric interaction where technologically developed and popular Nd:YAG lasers could be used as pumping resource. The other reason is the high transmission of radiation in the 12-15 mcm range. Furthermore these crystals have high nonlinear susceptibility, radiation resistance and higher mechanical hardness [3]. As it is shown in [2] the Hg_{1-x}Cd_xGa_2S_4 crystals, depending on the merit of non-oxide birefringent nonlinear crystals versus transparency range, are at better positions compared to CGA, CSP, ZGP and GaSe.

As we can see from [1-5], the admixture of cadmium in a mercury thiogallate changes the main values of refractive index and nonlinear coefficients in Hg_{1-x}Cd_xGa_2S_4 which means, for example, if the nonlinear coefficient in HgGa_2S_4 is 15.57 pm/V, then it is 1.6 times higher for Hg_{1-x}Cd_xGa_2S_4 and is equal to 24.94 pm/V. According to the experience carried our by the mentioned authors of Refs. 1 and 5, the value for double refraction is very sensitive to the parameter x. From here, by changing the Cd part in crystal (parameter x) we can set double refraction, the condition for phase matching, and it is possible to realize uncritical phase matching regime. Resulting from works [1, 5] during the process of crystal growth change of concentration of cadmium results in change of parameter x along a boule length (in growth direction). This causes the conversion in frequency by a continuously change of parameter x. The last is carried out by linear displacement of crystal in relation to a laser beam. According to what authors have stated the conditions for uncritical phase matching are not violated in this case.

The theoretical investigation of nonlinear interaction mainly are carried by numerical analysis for nonlinear interaction of optical waves. But the analytical method is more appropriate for the sake of better physical clarification. The most known analytical method is constant field approximation [6].

At parametric interaction it is necessary to take into account the threshold character of parametric generation to which the losses and phase mismatch of interacting waves make considerable contribution. The simultaneous account of these two factors may be provided by theoretical analysis of wave interaction in the constant intensity approximation [7, 8], which work well for reverse reaction of excited wave to exciting waves.

Purpose of this work is to study the parametric generation of laser radiation in Hg_{1-x}Cd_xGa_2S_4 crystal based on theoretical consideration of nonlinear interaction of waves while phase variation of all waves are taken into account. The comparison of the obtained results has been made on conversion efficiency with experimentally measured values. Some recommendations are offered to increase efficiency of frequency conversion.

2. THEORY

Now we want to look at the parametric generation in Hg_{1-x}Cd_xGa_2S_4 crystal addressed in [1-5]. A nanosecond Nd:YAG laser (\lambda_p=1.064 mcm) with 12mJ energy and 22 ns pulse duration is used as pumping source. According to experience a crystal with 11 mm in length is placed in 3 cm resonator which has two flat mirrors in both sides. In [1-5] singly-resonant optical parametric oscillation (SRO) is carried out with “two-pass” pumping. The mirrors at the entry of resonator (left) and its exit (right) have reflectivity of 95-99%
at the signal wavelength $\lambda_s=1.5772$ mcm. Pumping of crystal was done where the reflectivity of left mirror was 10% and the reflection of right mirror was 30% for pumping waves. The diameter beam waist is 2 mm at the interaction range of crystal. The output for idler wave occurs on right mirror which has 80-90% of transmission ($\lambda_i=3.27$ mcm). As result “two-pass” state for pump and signal waves occur in resonator and idler wave get “single-pass” resonator state, i.e. according to [2] OSO type of schemes takes place.

In experiment an uniaxial crystal of Hg$_{1-x}$Cd$_x$Ga$_2$S$_4$ cut-off with the cross-section of 30.6x8 mm$^2$ and length of 11 mm is used. Uncritical phase matching with $\theta = 90^\circ$ and $\varphi =45^\circ$ of first type takes place. Let’s take a look at $e^{\rightarrow 00}$ scalar interaction which is characterized by nonlinear parametric generation in a crystal with length $l$. In this crystal the optic wave with frequency of $\omega_p$ generates idler wave with frequency of $\omega_s$ and signal waves $\omega_i$ ($\omega_p = \omega_s + \omega_i$). At this stage we can use the known system of reduced equations [6].

When we solve this system using the constant intensity approximation at the exit of crystal, we get the following expression for idler wave intensity [9]:

$$I_i(\ell_1) = I_{i_0} \exp(-2\delta_1 l_1) \left[ \cosh^2 q_3 l_1 + \left( \frac{\Delta}{2} + \frac{\gamma A_{i_0} A_{p_0}}{A_{i_0}} \right)^2 \sinh^2 q_3 l_1 \right]$$

where

$$q_1^2 = \Gamma_p^2 - \Gamma_s^2 - \frac{\Delta^2}{4}, \quad \delta = \delta_s + \delta_i + \delta_p, \quad p = \delta - i\Delta,$$

$$\Gamma_s^2 = \frac{p^2}{4} - q, \quad \gamma = \Gamma_s - (\Gamma_p)^2 + \delta_i (\delta_s + \delta_p - i\Delta),$$

$$\Gamma_p^2 = \gamma' \Gamma_i^2, \quad q_2^2 = q - p^2 / 4, \quad P = (\delta_i - \delta_s - \delta_p + i\Delta) / 2.$$  \hspace{1cm} (2.1)

Here $A_{i_0,i_0,p_0}$ are initial values for complex amplitude of signal, idler and pump waves at respective frequencies along the z-axis (at the left entry of the crystal). For j-th wave (j=s, i, p) the nonlinear and linear absorption coefficients are shown by $\gamma_j$ and $\delta_j$, and $\Delta = k_p - k_s - k_i$ is the phase mismatching between interacting waves.

The values of amplitudes of pump and signal waves are calculated at the input by upcoming considerations in single resonator while the parametric amplification of idler wave is considered. The standard technique [9] is used to theoretically investigate the parametrical three-wave interaction for two pass of optical resonator by pump and signal waves. For this purpose the reduced system of equations have been solved in the constant intensity approximation for each pass of resonator with corresponding boundary conditions. At this time the conditions of experiments are taken into account. To be exactly correct, dissipation for all interacting waves include: effective dissipation in unit length of crystal while scattering is considered; dissipation of diffraction ($a^d_{i,s,p} = \frac{1}{L} \ln \left( \frac{a^2 - \lambda_{i,s,p}^2 L}{a^2 - 2\lambda_{i,s,p}^2 L} \right)$) where $a$, $L$ are the radius of plane-parallel circular mirror and length of optical resonator respectively); losses on mirrors due to reflection from mirrors;

Fresnel losses for each surface ($a_{Fresnel} = \left( \frac{n_{i,p} - 1}{n_{i,p} + 1} \right)^2$ for waves which are perpendicular to the crystal). In experiment reflection coefficients that are mentioned above for laser mirrors will make the resonator to have high quality for signal wave than idler wave. The high value of transmission coefficient in output laser mirror (80 %-90 %) for idler wavelength signifies the minimal dissipation for output radiation at idler wavelength.

The sides of the sample were made transparent in the experiment but the value of Fresnel dissipation is not diminished even when these waves are perpendicular to the crystal. For interacting waves in [3] the absorption coefficients are taken from experimental measurements and they are equal to 0.2 cm$^{-1}$ for pumping waves, 0.3 cm$^{-1}$ for signal wave and 0.4 cm$^{-1}$ for idler wave.

It should be noted that at the analysis in constant intensity approximation not only the phases and losses of interacting waves are considered but also partial reduction of pumping and signal waves is taken into account which is the result of energy transfer from excited waves to exciting one, that is $I_p(\ell_1) < I_{p_0}, I_s(\ell_1) < I_{s_0}, I_i(\ell_1) < I_{i_0}$. So the constant intensity approximation is used for each of two transitions of a crystal.

As is known, the parametric process has threshold character. It is possible to determine threshold value of pumping wave amplitude in the constant intensity approximation. At greater powers of pumping and lengths of interaction, the threshold amplitude of pumping looks as ($\delta_s = \delta_p + \delta_i$) [9]

$$A_{p_0,thresh}(\ell) = \sqrt{\frac{\Gamma_s^2 + \delta_i^2 + \Delta^2 / 4}{\gamma_\ell}}$$  \hspace{1cm} (2.2)

From (2.2), it is seen that with the increase of losses the condition of parametric amplification, i.e., $I_i(\ell) \geq I_{i_0}$ takes place at greater values of pumping amplitude. As seen from (2.2) the $A_{p_0,thresh}$ value increases with an increase in mismatch and losses ($\delta$). It is seen from the result that the
account of the depletion effects \( \Gamma_s = \gamma_p' / I_{\mu s} \), i.e. of the reverse reaction of excited waves on the pump wave, leads to a raise in the threshold pumping amplitude [10].

As we know, Hg\(_1\).Cd\(_{0.3}\).Ga\(_{2.7}\)S\(_4\) in mixed structure is affected by cadmium composition. Let’s identify the wavelength of idler and signal waves in Hg\(_1\).Cd\(_{0.3}\).Ga\(_{2.7}\)S\(_4\) crystal at the wavelength of \( \lambda_i = 1.064 \) mcm at 90\(^°\) synchronism. By choosing \( \alpha = 0.25 \) for cadmium concentration, the experimental value of wavelength of signal and idler waves are specified for this case which are presented in Refs. [1, 5]. Here the equation of \( \frac{1}{\lambda_p} = \frac{1}{\lambda_i} + \frac{1}{\lambda_s} \) should be established for the sake of parametric generation of the radiations at pumping wavelength of \( \lambda_p = 1.064 \) mcm and \( \lambda_i = 3.274 \) mcm. From here we can get the proper value of 1.5772 mcm for wavelength of signal wave. The main values of refraction indices for wavelength of waves under study are derived from Sellmeyer equation [1, 5], and \( n_{p, e}^{o, e}, n_{s, e}^{o, e}, n_{i, e}^{o, e} \) are taken from tables.

In the experiment for real frequency converters, it is impossible to ensure a condition of phase agreement (phase matching \( \Delta = 0 \)). An error in following the condition of phase matching determines the width of phase matching. Spectral width of pump radiation line, deviation from phase matching angle due to divergence of laser radiation, and instability of temperature for a crystal converter all contribute to mismatch. Hence received information, in particular, on angular width of phase matching, will permit to calculate the maximum divergence of light beam for pumping. Moreover, determination of conditions for realizing uncritical phase matching at chosen length of pump wave is important for exclusion of taking down the influence of birefringent walkoff on generation efficiency. This fact allows one to take away the restriction of length of the used crystals [11]. As it is known, birefringent walkoff effect is possible to be removed by using of 90-degree phase matching. For the majority of nonlinear crystals 90-degree phase matching does not take place for wavelengths of practical interests [12]. In the case under consideration of Hg\(_1\).Cd\(_{0.3}\).Ga\(_{2.7}\)S\(_4\) crystal the given condition is carried out in the middle IR spectrum range that does these crystals attractive for application.

Let us estimate deviation angle from the direction of phase matching \( \Delta \theta \) for the negative single-axis Hg\(_0.7\).Cd\(_{0.3}\).Ga\(_{2.7}\)S\(_4\) crystal in case of parametric generation radiation at wavelength of 3.03 mcm (phase matching of the first type, ee \( \rightarrow \) oo interaction). The calculation was carried out using the coefficients in the Sellmeier relation for the main values of refractive indices [1]. To determine the angular width of phase matching we will calculate the angular disperse coefficient according to Ref. 12.

In case of 90-degree phase matching the angular dispersion coefficient of the second order in our case (e-oo scalar interaction at parametrical generation) looks like

\[
\frac{\partial^2 \Delta}{\partial \theta^2} = -\frac{2\pi \left( n_p^{o} \right)^2 - \left( n_p^{e} \right)^2}{\lambda_p \left( n_p^{o} \right)^2}
\]

(2.3)

In the Table I, the values of refractive indices, the angle of phase matching and angular dispersion coefficient of the second-order are provided.

<table>
<thead>
<tr>
<th>Crystal</th>
<th>( \lambda ) mcm</th>
<th>( n_{p,oo}^{o} )</th>
<th>( n_{s,oo}^{o} )</th>
<th>( n_{i,oo}^{o} )</th>
<th>Phase matching type</th>
<th>( d_{\text{eff}} ) pm( \sqrt{\text{V}} )</th>
<th>( \theta_\text{e} ) degree</th>
<th>Angular dispersion coefficient of the second order, cm(^{-1}) ang. min.(^{-2})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hg(<em>0.7).Cd(</em>{0.3}).Ga(_{2.7})S(_4)</td>
<td>1.064 (pump)</td>
<td>2.430288</td>
<td>2.39899</td>
<td></td>
<td>e ( \rightarrow ) oo</td>
<td>24.94</td>
<td>90</td>
<td>0.0000634955</td>
</tr>
<tr>
<td>Hg(<em>0.7).Cd(</em>{0.3}).Ga(_{2.7})S(_4)</td>
<td>1.5772 (signal)</td>
<td>2.405065</td>
<td>2.367763</td>
<td></td>
<td>e ( \rightarrow ) oo</td>
<td></td>
<td>90</td>
<td>0.0000634955</td>
</tr>
<tr>
<td>Hg(<em>0.7).Cd(</em>{0.3}).Ga(_{2.7})S(_4)</td>
<td>3.27 (idler)</td>
<td>2.386262</td>
<td>2.35255</td>
<td></td>
<td>e ( \rightarrow ) oo</td>
<td></td>
<td>90</td>
<td>0.0000634955</td>
</tr>
</tbody>
</table>

3. RESULTS AND DISCUSSION

To study the parametrical frequency conversion in middle IR-range, we will make the numerical calculation of the analytical expression (1), which is received in the constant-intensity approximation. The parameters of the task are chosen according to conditions of existing experiments for the given Hg\(_1\).Cd\(_{0.3}\).Ga\(_{2.7}\)S\(_4\) crystal [1-5].
Fig. 1. Dependences of gain coefficient of idler wave in Hg$_{0.75}$Cd$_{0.25}$Ga$_2$S$_4$ crystal $I_i(I)$ as a function of the pump intensity $I_{po}$ ($\lambda_p=1.064$ mcm) (experimental dots from Ref. 1 and curves 1-5) calculated in the constant-intensity approximation for $\delta_i = \delta_p = 0.15$ cm$^{-1}$, 0.2 cm$^{-1}$ (curve 3) and $\delta_i = 0.15$ cm$^{-1}$, $\delta_p = 0.2$ cm$^{-1}$ (curve 4) at $\Delta/2\Gamma_p=0.1$ (curves 2-5), 0.3 (curve 1), crystal length of $\ell=1.1$ cm [1, 2, 5].

In Fig. 1 the dynamic process of parametrical amplification of idler wave $I_i(I)$ is shown on cited pumping intensity $I_{po}$ at the condition of phase mismatching for two variants of $\Gamma_s/\Gamma_p$ and losses. As is seen from figure, parametric gain at different losses starts at various values of pumping intensity (curves 1–5). With an increase of pumping intensity the first horizontal section of dependence is observed, i.e. parametric reinforcement is absent. Then at the definite value of pumping intensity, i.e. at threshold amplitude of pumping, the notable raising of dependence begins. With the increase of losses, the growth of dependence takes place at greater values of threshold intensity of pumping (compare curves 4–5). So, from the numerical analysis (1) (for $\Delta/2\Gamma_p=0.1$) it follows that in case of parametric interaction in Hg$_{1-x}$Cd$_x$Ga$_2$S$_4$ crystal, threshold intensity of pumping is equal to $I_{po}=0.37$ MW/cm$^2$ (0.25 mJ) at $\delta_i=0.2$ cm$^{-1}$, $\delta_p=0.2$ cm$^{-1}$ (curve 3); at $\delta_i=0.15$ cm$^{-1}$, $\delta_p=0.2$ cm$^{-1}$, it is equal to $I_{po}=0.3$ MW/cm$^2$ (0.2 mJ) (curve 4); and at $\delta_i=0.15$ cm$^{-1}$, $\delta_p=0.15$ cm$^{-1}$ reaches the value $I_{po}=0.15$ MW/cm$^2$ (0.1 mJ) (curve 5).

Also parametrical gain grows with increasing of parameter $\Gamma_s/\Gamma_p$ (compare curves 2 and 5) and decreasing of phase mismatching (compare curves 1 and 2).

Here in addition to the numerical calculation of the expression (1) (curves 1–4) experimental points are used from Ref. 1. More the best agreement between theoretical and experimental results is observed at value of parameter $\Gamma_s/\Gamma_p=0.0004$ in the range of experimentally used values of pump intensity from 3 mJ up to 10 mJ. As is seen in Fig. 1 it is observed both theoretically and experimentally that monotonous increase of $I_i(I)$ occurs when pump intensity grows.

By using the data provided in the table, dependencies of parametric conversion efficiency on angular mismatching $\Delta\theta$ are presented in Fig. 2 for Hg$_{0.75}$Cd$_{0.25}$Ga$_2$S$_4$ crystal at two values of pump intensity ($\Gamma_s/\Gamma_p=0.005$ and 0.0004). Just as one should expect, the dependencies are of nonmonotonous character. With the increase in pump intensity, conversion efficiency also increases (compare curves 2–3).
From the numerical analysis of the analytical expression (1) for idler wave intensity, obtained in the constant-intensity approximation, it follows that the width of angular phase matching is being changed by pump intensity. As we see in this figure, when pump intensity increases 1.31 times, the idler intensity increases 1.45 times (curves 2 and 3) and at the same time the width of curve for phase matching $I_i(\Delta \theta)$ decreases to 9%. For comparison the curve 1 calculated for other value of relation $\Gamma_s/\Gamma_p=0.0004$ is given here. The increase in the given relation, i.e. intensity of signal wave, conducts to growth of parametrical gain (to compare curves 1 and 2).

As is seen from dependence of $I_i(\Delta \theta)$ on angular width of phase mismatching chosen at 1/2 of maximum efficiency (Fig. 3), angular width is equal to 7.24 mrad, when we calculate it for Hg$_{0.75}$Cd$_{0.25}$Ga$_2$S$_4$ crystal length of 11 mm and pump intensity equal to 9 MW/cm$^2$. To compare, in case of 3 mm long CGA crystal, angular width of phase matching makes up 4.24 mrad [13], when pump intensity is equal to 0.0012 MW/cm$^2$. By comparing the behavior of curves 1 and 2 (Fig. 3), it is seen that the change of $I_i(\Delta \theta)$ by 5.1 % takes place in the angular range of changes from $-30$ mrad to $+30$ mrad. The curve 1 becomes more flat in comparison with curves 2 and 3. It testifies the transition to the regime of uncritical character of crystal towards following condition of phase matching; thus, for instance, in AgGaSe$_2$ crystal the change of conversation efficiency by 0.036% takes place in the angular range of changes from $-0.6$ mrad to $+0.6$ mrad [11]. Hence the weak changes of efficiency dependence on angular mismatch from phase matching direction takes place at small values of pump intensity (compare curves 2 and 3). Similar slight changes of conversion efficiency under deviation from direction of phase matching, i.e. the uncritical regime of the considered crystals can be realized at low levels of intensity for pump and signal waves.

Fig. 2. Dependences of gain coefficient of idler wave in Hg$_{0.75}$Cd$_{0.25}$Ga$_2$S$_4$ crystal on phase mismatching $\Delta/2\Gamma_p$ for $\delta_p=0.2$ cm$^{-1}$, $\delta_s=0.15$ cm$^{-1}$ (curves 1-3), at pump intensity $I_p=9\text{ MW/cm}^2$ (curves 1-2), 11.8 MW/cm$^2$ (curve 3) and $\Gamma_s/\Gamma_p=0.0004$ (curve 1), 0.005 (curves 2-3).
Fig. 3. Dependences of gain coefficient of idler wave in Hg$_{0.75}$Cd$_{0.25}$Ga$_2$S$_4$ crystal on angular mismatching $\Delta \theta$ for $\delta_p = 0.2$ cm$^{-1}$, $\delta_i = 0.15$ cm$^{-1}$ (curves 1-3), at pump intensity $I_p = 9$ MW/cm$^2$ (curves 1-2), 10 MW/cm$^2$ (curve 3) and $\Gamma_i / \Gamma_p = 0.0004$ (curve 1), 0.005 (curves 2-3).

4. **CONCLUSION**

Thus, from the results of the analysis of nonlinear interaction of optical waves with account for phase effects and comparison of them with existing experimental data, it is possible to state that by the choice of optimum values of parameters (pumping intensity, linear tuning with account for impact of linear losses in a medium) an efficiency of parametric conversion in the considered crystals of middle IR range may be increased and the condition for increasing degree of uncritical angular phase matching is possible to choose. Therefore, based on these crystals we can produce parametric generation where technologically developed and popular Nd:YAG lasers could be used as pumping resource. This will draw nearer the creation of the efficient sources of coherent radiation, which is tunable by frequency in the middle-IR region of spectrum.

The developed method can be used to study other nonlinear optical processes, e.g. sum frequency and harmonic generation, etc. in other perspective crystals.

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