Powerful Optical Parametric Oscillator (OPO) on GaSe Crystal Pumped by an 3-Micron Laser

EOARD OPO
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This report results from a contract tasking General Physics Institute of the Russian Academy of Sciences as follows: The contractor will investigate the generation of powerful, tunable mid-IR radiation by developing a 60 - 100 mJ, Q-switched, 150-200 ns pulse length, mode-locked 5 Hz rep rate pump using a YSGG:Cr:Yb:Ho laser operating at three microns, and then using this to pump a GaSe nonlinear crystal cut to produce tunable radiation in the mid-IR.
The proposed research should demonstrate an extension our project on resonator optical parametric oscillation (OPO) in a GaSe crystal pumped by an Q-switched three micron laser. Our previous work demonstrated feasibility of obtaining of resonator-type OPO on GaSe. At this phase we were concentrating on obtaining powerful tuneable IR radiation. The work should consist of improving of pump laser system and architecture of GaSe OPO in order to have ~10 mJ tuneable IR near 6 micron with 20% conversion efficiency.

INTRODUCTION

In the last few years the greater interest has returned to mid-infrared optical parametric oscillators (OPO) pumped by solid state lasers. This was a result of considerable progress in laser and non-linear crystal growth technologies. A new class of IR solid state lasers based on Ho\(^{3+}\) and Er\(^{3+}\) - doped crystals have been developed. Now both these types of lasers can be pumped by flash lamps or laser diodes. The latest laboratory versions of these lasers have efficiencies similar to Nd-doped lasers and erbium lasers are the longest wavelength high power solid state lasers currently available. These laser sources are very well suited for generating continuously tuneable intense and ultrashort IR light pulses using non-linear optical frequency conversion in optical parametric oscillators based on IR non-linear crystals. The GaSe crystal has large transparency range (0.65-18 μm) and higher non-linear figure of merit than other crystals such as CdSe, AgGaSe\(_2\), which were used for optical parametric oscillators. In our previous work we reported on obtaining tuneable IR radiation from resonator OPO on GaSe crystal pumped by YSGG:Cr:Er laser. In this project we had proposed to develop our achievement toward to obtain powerful resonator OPO pumped by Q-switched and mode locked 3-micron laser.
Comparison with ZGP crystal and CdGeAs:

CdGeAs$_2$ does not suit for OPO purposes since it is transparent from 4 micron and could be now only used for SHG of CO$_2$-laser.

ZGP crystal is the only and the major competitor to GaSe because of great achievements of research lab at Lockheed-Martin(LM), Sanders(Nashua,NH) both in improvement of pump laser technique and crystal growth. Some Advantage of GaSe lower cost. Binary semiconductors not triple-comments on that, maybe higher average power, however it is very difficult to compare now with LM because of retard in high tech in Russia. Except paper on GaSe - ZGP comparison for SHG CO$_2$ there is no data on average power. Now LM crystals are very good but very expensive.

Our first experience in working with GaSe OPO pumped by Q-switched pulses shows that pumping of GaSe by 7 $\mu$m laser radiation (a holmium laser) is also possible, while the tuning range in this case is better fit to the window of transparency of atmosphere(3.5-4.5 $\mu$m). For this purpose the best is to use existed pump laser like at LM or at Wright Lab(WPAFB, Dayton,OH).

The tuning range of 3-18 $\mu$m is of a great interest for spectroscopy because most of the frequencies of molecular vibrations as well as the sub-band transition energies in semiconductor quantum well structures lie in this spectral range. High intensity tuneable picosecond laser light could be used for time-resolved saturation spectroscopy in this spectral band. There are several windows of transparency of the atmosphere (the widest is between 3.5-4.2 $\mu$m) in the tuning range of such OPOs and these OPO could be used for remote sensing and atmosphere pollutant detection. Recently, dramatic improvement of cornea ablation by 6.45 $\mu$m laser radiation was reported. The 6.45 micron radiation corresponded to the bending vibration mode of water and obtained from free electron laser (FEL).
performance characteristics of OPOs (pulse energy, peak intensity, average power) are comparable with those of a free electron laser. The OPO however is a table top instrument compared with the warehouse sized installation needed to accommodate the electron beam accelerator and magnetic wiggler for a FEL.

TECHNICAL CONDITIONS

The work on the project consists of two major parts. One of them is analysis and realisation of powerful pumping laser which will be used to pump OPO. We had to use new YSGG:Cr:Yb:Ho (λ = 2.92 μm) laser crystal grown at the General Physics Institute, working as a 4-level system and lasing at 2.85-3.05 μm in free running mode. This new laser medium has low lasing threshold, larger laser cross-section than other media (YSGG:Cr:Er, YAG:Er) and as results has high single-pass amplification at room temperature.

The goals was to have this laser working at 10 Hz repetition rate, Q-switching with 15-20 mJ at 100 ns, TEM$_{00}$ - mode. An approach that combines high conversion efficiency of a picosecond travelling wave OPO and high output energy of a resonator OPO is an OPO synchronously pumped by a train of picosecond pulses from a mode-locked laser.

A synchronous pump means that the optical length of the OPO cavity is multiple of pump laser cavity. In this optical set-up modes in the OPO cavity are locked resulting in low generation threshold and high peak power of tuneable IR pulses.

The pumping laser in our project will be actively Q-switched, mode-locked YSGG:Cr:Yb:Ho, generating at one or several lines around 2.92 μm. We intend to use all our previous technical developments obtained in the previous work. I is HV electronic driver (output voltage 3-5 kV, pulse duration 10-20 μs, f=60 MHz) for HF electro-optical modulation of intracavity losses. Output laser pulse will have
energy $E=20$ mJ in 150-200 ns envelope consisting of 20-30 pulses with single pulse duration 100 - 200 ps, repetition rate 1-5 Hz. We will try to amplify this laser pulse in an amplifier. It should be noted that repetition rate like 10 Hz seems to us now too high because of thermoconductivity of the YSGG crystal.

In experiments on non-linear OPO we had tested GaSe crystals which are available from crystal growth laboratories in Tomsk, Russia. We had bought two GaSe crystals with aperture 10 mm and thickness 10 mm also along the optical axis. The crystals with aperture up to 15-20 mm and length up to 30-40 mm are available but costs approximately 10,000 $ (Voevodin, Tomsk).

Since GaSe crystal is very soft and has layered structure perpendicular to the optical axis, it can not be polished at a certain angle to the optical axis. However, birefringence in this crystal is high enough to obtain both types of phasematching with 3 micron pump by rotating the crystal at a moderate angle ($\theta_{\text{internal}}<20^\circ$). The crystal itself should be put in a special holder. It was planned to study both type I and II phase matching. With the 3 $\mu$m pump at type I (e-oo) phase matching a GaSe OPO generates a wide spectrum IR signal (pseudo-continuum at 4-8 $\mu$m). For type II (e-oe) interaction, the external phase matching angle at the degenerate point is close to the Brewster angle that decreases Fresnel reflection on the faces of the crystal and thus decreases threshold.

We had planed to use a cylindrical lenses in the telescope (optical focusing system) in order to enlarge size of the pump laser beam in the plane of the optical walk-off and focusing in the perpendicular direction (cylindrical lenses have been ordered).

Regarding this peculiarity of GaSe we were going to consider different setups of a pump laser and OPO: two different cavities matched by a lens; intracavity OPO in case of pure Q-switched pump; output coupling due to Fresnel reflection; double crystals scheme with rotation of each of them in opposite direction to compensate walk-off of the parametric signal from the pumped area, etc.
To obtain tuning in the whole transparency range, different sets of OPO mirrors should be needed. Mirrors are available from the 'KVANT' company in Nizhny Novgorod and we have bought the series of the OPO mirrors.

We had tried to deposit more soft one-layer antireflected coating onto GaSe surface (like LiF, Al₂O₃), but unsuccessful because of too different the lattice constants of this crystals.

**EXPERIMENTAL PART**

Under all value used in preceding project of Er-laser on the base crystalline matrixes YSGG it possesses one defect - rather small single-pass gain (approximately twice on the length 10 cm). Besides, in the course of performing a preceding project was discovered essential absorption of laser radiation on the wavelength \( \lambda = 2.79 \text{ mkm} \) in the atmospheric air; the absorption coefficient was found approximately \( 4 \times 10^{-3} \text{ cm}^{-1} \). This rather large absorption coefficient prevents realisation of some schemes of resonant parametric oscillator, for instance, a scheme so called "synchronous pumping".

In 1996 in General Physics Institute was originated new laser crystal – an Yttrium-Scandium-Gallium Garnet doped with Holmium (YSGG:Cr:Yb:Ho, in drawings below marked as YSGG:CYH), lasing at selflimited transition. This laser allows in the free-running mode of oscillation to be tuned within the wavelengths range of 2.84 - 3.05 mkm and possesses in 2 - 3 times more high single-pass gain factor [1–3].

In the process of performing a given project was reasonable research new Ho-laser in the Q-switch mode, taking into account of its using as a pumping laser of the parametric oscillator on GaSe and other non-linear crystals for mid infrared.

We were conducted studies to obtaining of giant pulses in this new efficient laser crystal - YSGG:CYII. Its oscillation in the Q-switched mode was obtained with help of an electrooptical shutter on the LiNbO₃ crystal with the ends which
were cut at the Brewster angle and without linear polarisers (or polarizers). In the giant pulse mode we observed a lasing wavelength equals 2.92 mkm, while in the free-running mode a lasing wavelength occupied the spectral range from 2.84 mkm to 3.05 mkm[2]. In the Q-switched mode pulse duration was ~150 ns with the energy of pulse ~15 mJ. In the future we will measure the absorption coefficient of the laser radiation on the wavelength 2.92 mkm in the room air, however our preceding perennial experience of work with YAG:Er laser, oscillating on the wavelength 2.94 mkm – close to 2.92 mkm – did not found any problems with sufficient absorption in the air.

The second and fourth harmonic generation was also obtained in LiNbO₃ crystal with efficiency ~20% of each cascade. Thereby, new Ho-laser (2.92 mkm) side by side with Er-laser can be used as the laser for the pumping of parametric oscillators in the IR spectral range.

Until June this year we had (borrowed) only one sample of the YSGG:Ho crystal of the length and diameter equal 55 mm and 4 mm accordingly with the plane-parallel polished ends. Lasing parameters of this crystal are shown below.

At the end of June 1998 we had two more samples got, but both of them were without fabricated and polished ends.

The optical facility of GPI was working with this samples, but slowly as usual in summer because of vacation time and because of the August-98 financial crisis. We hoped to get both crystals back at the end of August 1998, but had got them in the October 1998.

One of this crystals has the length and diameter equal 77 mm and 4 mm, another one – 95 mm and 5 mm accordingly.

After that we have started to search the gain parameters of the single pass amplifier on each of this samples in the Q-switch mode regime. We were using the active Q-switching regime of master-oscillator with help of the electrooptical shutter on the LiNbO₃ crystal driving by a high-voltage power supply.
The scheme of laser resonator of the master-oscillator on the YSGG:Cr$^{3+}$:Yb$^{3+}$:Ho$^{3+}$ crystal is shown on the Fig.1. (In the lower text we will write YSGG:CYH instead of YSGG:Cr$^{3+}$:Yb$^{3+}$:Ho$^{3+}$ crystal.)

![Diagram of laser resonator]

On the Fig.1 is shown plane-parallel laser resonator with:

- Laser rod: 4x55 mm, no AR coating,
- OM: output dielectric mirror with reflection R=55%, HR: high reflection mirror,
- Length of resonator $L_R = 55$ cm,
- Free running with LiNbO$_3$ crystal inside resonator: two lines with $\lambda_1 = 2.84 \text{ mkm}$, $\lambda_2 = 2.92 \text{ mkm}$
  with intensity ratio as 1:10; pump energy threshold was $E_{\text{thresh}} = 12 \text{ J}$,
- Q-switching: one line $\lambda = 2.93 \text{ mkm}$, output energy $E = 15 - 17 \text{ mJ}$, TEM$_{00}$-mode;
  pump energy threshold was $E_{\text{thresh}} = 18 \text{ J}$, pulse duration was $\tau_{1/2} = 140 \text{ ns}$ as can see on the Fig.2.
Fig. 2.

The trace of the oscillogram, which is shown on the Fig. 2, has the time rate equals to 100 ns per large division of scale.

We have got also the second and fourth harmonic cascade generation with help of two LiNbO$_3$ crystals. Both of the LiNbO$_3$ crystals has size 5x5x20 mm and was cut with angle equals to $\theta = 52^0$ between the optical axis and geometrical axis lying along the largest size.

The angle of synchronism for the type oo-e parametric interaction for the second harmonic generation $\theta(2\omega) = 53^0$ and the angle of synchronism for the fourth harmonic generation $\theta(4\omega) = 50^0$. This fact allows very easy to make adjustment for each of the crystals to get the maximum of the possible harmonic intensity. It should be noted that optical axes of this LiNbO$_3$ crystals was rotated each other on the $90^0$, of course. The scheme of the cascade harmonic generation is shown on the Fig. 3.
The silica quartz lens with focal length equals to 300 mm was placed in front of the both LiNbO₃ crystals. The fundamental frequency of Ho-laser radiation was equal to 2.92 mkm, the giant pulse energy was equal to 10 mJ in TEM₀₀-mode and its pulse duration was equal to \( \tau(\omega) \sim 160 \text{ ns} \) (piroelectric photodetector was saturated), as it can see on the Fig.4 (the lowest trace of the oscillogram). The vector of polarisation \( \mathbf{e}(\omega) \) of the fundamental radiation was normal to the plane of the picture, hence the first LiNbO₃ crystal has the optical axis lying in the plane of the picture to generate the second harmonic radiation with the vector of polarisation \( \mathbf{e}(2\omega) \) lying in the plane of the picture (\( oo-e \) type of parametric interaction). The similar situation take place with the second LiNbO₃ crystal which is rotated on 90° to the first one. We have obtained the second harmonic radiation with \( \lambda(2\omega) = 1.46 \text{ mkm} \) and conversion efficiency

\[ \omega \rightarrow 2\omega \quad \eta = 23 \% \text{ at pulse duration } \tau(2\omega) = 92 \text{ ns} , \]

as it can see on the Fig.4 (the middle trace of oscillogram). The fourth harmonic radiation with \( \lambda(4\omega) = 0.73 \text{ mkm} \) has conversion efficiency \( \omega \rightarrow 4\omega \) approximately 2% and pulse duration \( \tau(4\omega) = 64 \text{ ns} \) (the upper trace of the oscillogram on the Fig.4).
The next part of investigations was connected with experimental realization of the renewed pump source for mid-IR OPO.

This pump is based on the new laser crystal YSGG:Cr:Yb:Ho with concentration of Ho$^{3+}$ about $5 \times 10^{19}$ cm$^{-3}$ and energy transfer process

$$\text{Cr}^{3+} \rightarrow \text{Yb}^{3+} \rightarrow \text{Ho}^{3+}$$

with calculated efficiency [4] over 90%. Lasing of Ho$^{3+}$ ions is possible on $^5I_6 \rightarrow ^5I_7$ ($\lambda=2.92 \mu$m) and on $^5I_7 \rightarrow ^5I_8$ ($\lambda=2.1 \mu$m) transitions both separately and simultaneously [5].

The lifetimes of the $^5I_6$ and $^5I_7$ levels of Ho$^{3+}$ are 0.47 and 9.8 ms, respectively, so that the 2.92 $\mu$m lasing channel is self-terminated and can work
only in pulse regime. It means besides that we can use only one half of the inverse population of upper laser level $^5I_{16}$ to get the laser oscillation.

We would like in future to insert the Ho-crystal-oscillator into resonator with high reflection mirrors for wavelength $\lambda=2.1$ $\mu$m to sufficiently increase the decay of the lower laser level $^5I_{17}$ population because of free running oscillation on the laser transition $^5I_{17} - ^5I_{18}$. It could improve the oscillation parameters for the transition $^5I_{16} - ^5I_{17}$ on the wavelength $\lambda=2.92$ $\mu$m.

In the first interim report we mentioned about the absorption coefficient of the laser radiation on the wavelength $2.92$ mkm in the room air. We have measured this absorption coefficient, which is estimated as $\alpha(\lambda=2.92$ mkm) = $3.3\times10^{-3}$ m$^{-1}$ and for our experiments is negligible.

Unfortunately during carrying out this task we waste much time (more then two months):

firstly, we hoped to get fabricated samples of Ho-crystals from our optical facility at the end of August, but the crystals was gotten at the first week of October because financial crises in Russia (the optical facility of GPI doesn’t work this period);

secondly, in October we were starting with our laser experiments, but some days after we had gotten some problems with our experimental equipment which were repairing more then two weeks.

We had carried out the experiments right after repairing equipment to measure gain coefficient, but we had not enough time before sending the second quarter interim report and have got only preliminary results, which is shown on the Fig.5 as the dependence of the single-path gain in YSGG:Cr$^{3+}$:Yb$^{3+}$:Ho$^{3+}$ crystal of the diameter 4 mm and the length 77 mm on amplifier pump energy in J. Input energy of giant pulse of master-oscillator is equal 4 mJ. It should be noted that illuminated length of the Ho-crystal-amplifier at this measurements was just 50 mm because of the illuminating cavity which we had those time.
Giant pulse gain in Ho-crystal
of diameter 4 mm and length 77 mm

![Graph showing single path gain vs. amplifier pump energy]

We were continuing our experiments to get a giant pulse gain coefficients at different input energies from 1 mJ to 10-15 mJ and output energy from amplifier. This set of curves needs us to determine the main amplification parameters of YSGG:Cr:Yb:Ho-crystal.

We have repeated later the measurements of the amplifier output energy vs. amplifier pump energy with another Ho-crystal. This holmium crystal-amplifier has
diameter 5 mm, the total length of the crystal equals to 95 mm and the illuminated length of the crystal 80 mm.

The results of our measurements is shown on the Fig.2.

![Graph](image)

Illuminated length of the crystal-amplifier is 80 mm, input energy of the amplifier is 10 mJ in Q-switch TEM00 mode with pulse duration ~150 ns and gaussian beam diameter 2w~2 mm.

Fig.2.

We have so the output energy of the single pass amplifier up to almost 60 mJ in Q-switch mode operation on the wavelength 2.93 mkm. Thus we have made a part of the contract.

The next part concern with the possibility of OPO realisation.

1. There are two OPO resonator schemes on GaSe crystals to reduce an influence of Fresnel’s losses on threshold power of pumping radiation:
   a) one GaSe sample, one of the its sides coated with reflective layer of gold or aluminium; in this case it is redoubled possible length of parametric interaction, but without compensation of birefringence angle for e-waves, Fresnel’s losses take
place on the input-output GaSe proofs only; increment of amplification of a parametric waves is almost by four times higher (if the losses are rather low) at the same Fresnel's losses. (Fig.1).

![Reflection coating (Au or Al)](image)

**Fig.1**

b) two GaSe samples under phasematched angle, but with the compensation of birefringence angle for e-waves. (Fig.2). In this case is not required too much radius of gaussian beam of the pumping radiation, but Fresnel's losses are twice more, than for the one sample, but the increment of amplification of a parametric waves is almost by four times higher (if the losses are rather low) at the same Fresnel's losses, as in the case (a).

In fact, if we have one GaSe crystal of the length $l$ inserted between two mirrors and the Fresnel's losses on the both sides of GaSe are $\delta$, then the single path parametric gain is

$$G(l) = \Gamma^2 l^2 - \delta l.$$  

In the case (a) we have $G(2l) = (\Gamma \cdot 2l)^2 - \delta \cdot 2l = 4[\Gamma^2 l^2 - (\delta/2) l]$ and the ratio
G(2l)/G(l)= {4[Γ^2 - (δ/2)l ]}/( Γ^2 - δ.l)- 4 at δ.l << Γ^2. In the case (b) we have G(2l) = (Γ·2l)^2 - 2(δ.l) = 4[Γ^2 - (δ/2)l ] and the ratio G(2l)/G(l) is exactly the same as in the case (a) but in the case (b) is there the birefringence angles compensation of two GaSe crystals as it was written above.

![Fig. 2](image)

The increment of parametric amplification Γ without losses can be written by the next formula (the case of low single path gain and gaussian beams of interacting waves):

\[
Γ^2L^2 = (256\pi^2/c^3)(ω_s ω_i/n_{p,n_0,n_i})[(d_{eff}^2L^2)/(w_p^2+w_s^2)]P_p ,
\]

where \(P_p\) is the pumping power, \(L\) is the length of parametric interaction, c - light velocity in the vacuum, \(n_i\) - refractive index, \(w_j\) - radius of gaussian beam, \(ω_j\) - frequencies of interacting waves, \(d_{eff}\) is the effective quadratic susceptibility, which is equal (for GaSe at different types of interaction) to

\[
d_{eff}(e-oo) = -d_{22}\cosθ\sin3φ \\
d_{eff}(e-e0) = -d_{22}\cos^2θ\cos3φ
\]

and \(θ\) - an angle between the wave vector \(k_p\) of pumping radiation and optical axis (lying along the crystallographic axis Z) of the non-linear crystal; \(φ\) - an angle between two crystallographic planes (XZ) and (kp Z). We need (and can), of course,
to choose the angle $\varphi$ so that $\sin 3\varphi = 1$ and $\cos 3\varphi = 1$ (must be equal to 1) for e-oo and e-eo types accordingly to maximise $d_{\text{eff}}$.

A calculated tuning curves for GaSe crystal pumped by radiation with $\lambda = 2.92 \, \text{mkm}$ are shown on the Fig.3a. and Fig.3b. We can estimate $d_{\text{eff}}(\theta)$ for both types of interactions from this tuning curves by taking an angle $\theta \sim 11^0$ for type I and $\theta \sim 15^0$ for type II.

The estimations of $d_{\text{eff}}$ for this matchangles of both types of interactions give:

\[
d_{\text{eff}}(\text{e-oo}) = -d_{22}\cos\theta\sin 3\varphi = -1.27 \times 10^{-7} \, \text{cm/dyna}^{1/2}.\cos 11^0 = -1.25 \times 10^{-7} \, \text{cm/dyna}^{1/2}
\]

\[
d_{\text{eff}}(\text{e-eo}) = -d_{22}\cos^2\theta\cos 3\varphi = -1.20 \times 10^{-7} \, \text{cm/dyna}^{1/2}.\cos^2 15^0 = -1.12 \times 10^{-7} \, \text{cm/dyna}^{1/2}
\]

![Fig.3a.](image)

If we take for simplicity $|d_{\text{eff}}| = 1.2 \times 10^{-7} \, \text{cm/dyna}^{1/2}$ for both types of interactions and take into account the value $n_p n_s n_i = 17.5$ and the values of $w_p = w_s = 0.1 \, \text{cm}$ (plan-parallel resonator),

we can write the expression for the increment of parametric single path.
gain $\Gamma$:
\[
\Gamma^2 = \frac{(256\pi^2/c^3)(\omega_s \omega_i)[(d_\text{eff})/(n_p n_s n_i)(w_p^2 + w_s^2)]P_p =}{(256\pi^2/c^3)(\omega_s \omega_i)(0.82 \times 10^{-15} \text{ cm}^2/\text{dyna})P_p/(w_p^2 + w_s^2)} =
= 3.85 \times 10^{-13} \text{ [s.cm}^{-2/\text{erg}]xP_p[\text{erg/s}]},
\]
where $c = 3.1 \times 10^{10} \text{ cm/s}, \omega_s = \omega_i = 2\pi c/\lambda \text{degeneracy} = 5.1 \times 10^{13} \text{ s}^{-1}, (\omega_s \omega_i) =
= 9.87 \times 10^{28} \text{ s}^{-2} \sim 10^{29} \text{ s}^{-2}, (w_p^2 + w_s^2) = 0.02 \text{ cm}^2$.

If we take output energy after Ho-amplifier $E_{\text{out}} = 50 \text{ mJ} = 5.1 \times 10^5 \text{ erg}$, pulse duration $\tau = 1.5 \times 10^{-7} \text{ s}$, then $P_p = E_{\text{out}}/\tau = 3.33 \times 10^{12} \text{ erg/s} = 3.3 \times 10^5 \text{ W}$ and it gives us
\[
\Gamma^2 = 3.85 \times 10^{-13} \text{ [s.cm}^{-2/\text{erg}]xP_p[\text{erg/s}]} = 1.28 \text{ cm}^2, \Gamma = 1.13 \text{ cm}^{-1}.
\]

Now we take into account a losses of resonator OPO. The expression for the threshold increment of parametric single path gain $\Gamma_{\text{th}}$ with the losses of resonating parametric "signal" wave $\Delta_s$ and not steady-state regime of parametric oscillation with pump-pulse duration $\tau$ is:

Fig. 3b.
\[ \Gamma_{th}^2 \Gamma^2 = \Delta_s + 60[ L + l(n-1)]/c \tau = \Delta_s + \Delta \tau , \]

where \( L \) is a total length of OPO-resonator, \( l \) is the length of parametric interaction inside of our non-linear crystal (GaSe), \( n \) is the refractive index of non-linear crystal, \( c \) - light velocity in vacuum, \( \Delta \tau \) is the non-steady-state part of losses. The "signal" (or the same for "idler") parametric wave losses \( \Delta_s \) for a good optical quality GaSe crystal are only Fresnel's losses and they could be chosen as the Fresnel' losses of p-component of polarisation, which lie in the plane, containing the wave vector \( k \) of laser radiation and optical axis \( C \) of the GaSe crystal.

In this case \( \Delta_s = 4(\sim 10\%) = 0.4 \), i.e. the radiation reflects four times (total path inside a resonator) with \(-10\%\) reflection on each surface of GaSe crystal. The non-steady-state part of losses \( \Delta \tau \) depends on pump-pulse duration \( \tau \) (which equals to \(-150\) ns for Ho-laser) and on \( L \sim 10\) cm, \( l \sim 1\) cm, \( n \sim 2.4 \), so that \( \Delta \tau \sim 0.15 \). The total loses are \( \Delta_s + \Delta \tau = 0.55 \) and at \( l = 1\) cm we have \( \Gamma_{th}^2 = 0.55 \) cm\(^2\). The estimation of the threshold pump power \( P_{p,th} \) gives us

\[ P_{p,th} = 0.55 \text{ cm}^2/3.85 \times 10^{-13} \text{ [s.cm}^{-2}\text{/erg]} = 1.43 \times 10^{12} \text{ [erg/s]} \text{ or } 1.43 \times 10^5 \text{ J/s} = 143 \text{ kW}. \]

It means that \( E_{p,th} = (1.43 \times 10^5 \text{ J/s}) \times (1.5 \times 10^{-7} \text{ s}) = 21.5 \text{ mJ} \).

If we have \( E_{out} = 50 \text{ mJ} \), then at \( l_{GaSe} = 1\) cm we get \( \Gamma^2 \Gamma^2 = 1.28 \).

It should be noted that the energy \( E_{out} = 50 \text{ mJ} = 5 \times 10^5 \text{ erg} \) at the pulse duration \( \tau = 150 \text{ ns} = 1.5 \times 10^{-7} \text{ s} \), gives the pump peak power \( P_p = E_{out}/\tau = 3.33 \times 10^{12} \text{ erg/s} \), and surface power density \( P_p/S \) at the gaussian beam radius \( w = 0.1 \text{ cm} \) (then \( S = \pi w^2 / 2 = 1.6 \times 10^{-2} \text{ cm}^2 \)) will have the rather high value

\[ P_p/S = 21.2 \text{ MW/cm}^2 . \] This value of \( P_p/S \) is very close to damage of the surface of GaSe.

We guess that is the reason of our unsuccessful experiments with the operation the resonant GaSe OPO, when almost each laser pump-pulse gives us the
pulse of parametric oscillation, but also gives simultaneously the surface damage of GaSe crystal.

2. We had observed suddenly unexpected reduction of gain factor in the amplifier, which was reduced until zero. Firstly it was could not understand, what is wrong. We have accidentally called attention that was changed fase matched angle of the second and fourth harmonic generation, at the same value of the output power of fundamental laser radiation. Then we have understood that is changed wavelength of laser generations.

In this contract we had at first only one Ho-crystal (4mm in diameter and 55 mm in length) for master-oscillator in Q-switch TEMoo mode on the wavelength 2.92 mkm.

We have got later two more Ho-crystals, that allow us to get with help of single gain amplifier on Ho-crystal-amplifier (5 mm in diameter and 95 mm in length with illuminated length equals to 80 mm) output energy up to 58 mJ and fulfil a part of contract.

We would like to note that the Ho-crystal-amplifier was fabricated just in October 1998. But during the work the Ho-crystal of master oscillator had changed his lasing properties, namely had changed spectral line of oscillation: instead of one laser line 2.92 mkm this Ho-crystal operates now on several laser lines. It is necessary to understand a reason of this phenomena.

The reason is as we think that long time operation in hard regime had led to appearance of colour centres, which absorption bands had changed oscillations properties of Ho-crystal.

We have observed 6 spectral lines oscillating simultaneously: 2.84 mkm (strong), 2.91 mkm (week), 2.93 mkm (strong), 2.95 mkm (week), 2.96 mkm (week). Week lines is more than an order of value less in intensities then strong lines. Accuracy of measuring was not so good because of rather low spectral
resolution of our monochrometer MDR-4, which has the focal length equals to 300 mm and the grating with 150 lines per mm and size 40x40 mm.

We may suppose however that strong line 2.93 mkm is almost the same that strong line (in former) 2.92 mkm and has no difference for pump source of optical parametric oscillator on the GaSe crystal.

We continue to research an amplifier on the Ho-crystal-amplifier (5 mm in diameter and 95 mm in length with illuminated length equals to 80 mm) with one more crystal of 4 mm in diameter and illuminated length equals to 50 mm as the master-oscillator.

It is obviously now to us that it is necessarily to make a dispersive resonator for the master-oscillator of Ho-laser to select only one strong laser line. It should be better for the Ho-amplifier operation, since the inversion population of Ho-amplifier-crystal will not separate into several lasing lines.

Comment: The value of gaussian beam radius \( w = 0.1 \) cm was measured by means of moving sharp edge and was calculated with help of integral of errors.

CONCLUSION

YSGG:Cr\(^{3+}\):Yb\(^{3+}\):Ho\(^{3+}\) -crystal as the new active medium for solid-state laser of the middle infrared is more complicated, than we thought before. It is required to continue experiments with it and to search the properties of this new laser crystal in detail.

References:
2. A.F.Umyskov, Yu.D.Zavartsev, A.I.Zagumennyi, V.V.Osico, P.A.Studenikin, Cr\textsuperscript{3+} : Yb\textsuperscript{3+} : Ln\textsuperscript{3+} : YSGG crystal laser with a continuously tuneable emission wavelength in the range 2.84 – 3.05 mm, Quantum Electronics, 26(7), 563–564 (1996).

3. A.F.Umyskov, Yu.D.Zavartsev, A.I.Zagumennyi, V.V.Osico, P.A.Studenikin, Efficient 3-mm Cr\textsuperscript{3+} : Yb\textsuperscript{3+} : Ln\textsuperscript{3+} : YSGG crystal laser, Quantum Electronics, 26(9), 771–772 (1996).


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