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Efficient parametric oscillation in the 8 – 10-μm range upon pumping by a femtosecond Cr : forsterite laser

V.M. Gordienko, S.S. Grechin, A.A. Ivanov, A.A. Podshivalov, E.V. Rakov

***Abstract*. Efficient parametric oscillation is obtained in the 8 – 10-μm range in a $LiInS_2$ crystal pumped by a femtosecond Cr : forsterite laser. The conversion efficiency of 0.8 % at a wavelength of 9.5 μm is achieved for the first time. The 75-% SHG efficiency for a Cr : forsterite laser in an LBO crystal is also achieved for the first time.**

Keywords: *optical parametric oscillator, femtosecond pulses, nonlinear optics.*

Progress in the development of high-power femtosecond Ti : sapphire, Cr : forsterite and diode-pumped Yb : fibre lasers of a new generation has stimulated a new research on the creation of highly efficient optical parametric oscillators (OPOs) operating in the mid-IR range. Such OPOs are required for studying the dynamics of interband transitions in superconductors and quantum-well structures [1], for controlling in real time the dynamics of intermolecular redistribution of the vibrational energy [2], for studying the peculiarities of photodissociation processes in polytiatomic molecules selectively excited by high-intensity laser radiation [3], and for studying the high harmonics generation [4]. In addition, high-power ultrashort mid-IR pulses can be used in spectroscopic femtosecond lidars [5]. Another their important application is related to the problem of creation of high-power laser systems operating in the ten-micron range when OPOs are used to form ultrashort initial pulses for their following amplification in N_2O or CO_2 amplifiers [6], which, naturally, requires high conversion efficiencies of pump radiation into the IR range.

The use of a 1.24-μm Cr : forsterite laser [$Cr^{4+} : Mg_2SiO_2$] for pumping OPOs operating in the 8 – 10-μm range, has a number of advantages compared to the use of femtosecond lasers of other types [7]. First, in a majority of nonlinear crystals two-photon absorption of pump radiation is absent and, second, according to the Manly – Row rule, a higher conversion efficiency is achieved.

Parametric oscillation in the range from 8 to 10 μm was obtained in $AgGaS_2$, $HgGa_2S_4$ and $Cd_{0.54}Hg_{0.46}Ga_2S_4$ crystals pumped by a Cr : forsterite laser [8, 9]. The conversion efficiency to the 8 – 10-μm range achieved in these experiments did not exceed 0.2 %. The analysis shows that the theoretical limit of the conversion efficiency for radiation with a Gaussian profile in space and time is 3 % – 4 %.

The aim of this paper is to create an optimal scheme for efficient optical parametric oscillation in the 8 – 10-μm range upon pumping by a femtosecond Cr : forsterite laser.

To obtain efficient optical parametric oscillation in the 8 – 10-μm range, high-power (intensity ~ 1 GW cm^{-2}) injection 1.4 – 1.5-μm radiation is required, while super-continuum generation [10] or parametric luminescence [8] allowed one to realise ~ 0.1 GW cm^{-2} intensities. The use of the OPO in the 1.4 – 1.5-μm range with a seeding of the super-continuum radiation (0.8 – 1.1 μm) will allow us to increase substantially the generation efficiency of the injection radiation.

A high efficiency of optical parametric oscillation, as in the case of harmonic generation [11], can be achieved in a crystal, which has not only a high nonlinearity but also a high spectral phase-matching width. The role of the cubic nonlinearity coefficient n_2 in processes of optical parametric oscillation has not been discussed so far in experimental works. However, it is obvious that as in the case of harmonic generation [12, 13], its influence will be one of the main factors restricting the conversion efficiency. The maximum conversion efficiency under the condition of preserving the minimum pulse duration is achieved in the case of optical parametric oscillation in a crystal in which a high spectral phase-matching width and high quadratic and small cubic nonlinearities are combined.

We performed a series of numerical experiments on the frequency conversion in different nonlinear crystals by using the spectral model [14] to find the optimal crystals for OPOs in the 8 – 10-μm range. The absence of data on the coefficient n_2 for the majority of mid-IR crystals did not allow us to take into account its influence. The highest theoretical conversion efficiency is achieved in $LiInS_2$ and $HgGa_2S_4$ crystals. It is known that among mid-IR crystals studied, the $LiInS_2$ crystal has the shortest-wavelength absorption edge (450 nm) and its two-photon absorption at 0.8 μm is two orders of magnitude weaker than that in the $AgGaS_2$ crystal [15]. These facts allow us to hope that the coefficient n_2 for $LiInS_2$ will be also the smallest. Therefore,

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V.M. Gordienko, S.S. Grechin, A.A. Podshivalov, E.V. Rakov International Laser Center, M.V. Lomonosov Moscow State University, Vorob'evy gory, 119992 Moscow, Russia;
e-mail: gord@femtosrv.phys.msu.ru, grechin@psi.phys.msu.su, rakov@femto.phys.msu.ru;
A.A. Ivanov Photochemistry Center, Russian Academy of Sciences, ul. Novatorov 7a, 117421 Moscow, Russia

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we assume that upon pumping by a femtosecond Cr : forsterite laser, the optimal crystal for OPOs in the 5 – 10-μm range will be a $LiInS_2$ crystal.

The optical scheme of the oscillator in the 8 – 10-μm range with an intermediate stage of generation of the injection 1.4 – 1.5-μm radiation is presented in Fig. 1. A 1.24-μm, 350-μJ, 140-fs Cr : forsterite laser is used as a pump source, a part of its radiation being frequency doubled and used to pump an intermediate OPO generating the required 1.4 – 1.5-μm radiation. The second harmonic is generated in a 5-mm LBO crystal ($\varphi = 0°$, $\theta = 87°$, ssf type [16]).

Figure 2 shows the experimentally measured dependence of the SHG efficiency in the LBO crystal on the incident radiation intensity. The maximum conversion efficiency was 78 %. The broadening of the second harmonic spectrum caused by the action of the Kerr nonlinearity was not observed up to the conversion efficiencies of 75 %. Note that these efficiencies exceed the efficiencies obtained by us earlier in [13], which is caused by a greater length of the crystal and its better optical quality.

In the scheme of the intermediate oscillator (OPO1) used here, generation in the 1.4 – 1.5-μm range was realised with the help of two LBO crystals ($\varphi = 0°$, $\theta = 90°$, ssf type) of length 4 mm each placed in series, which provided the maximum conversion efficiency (the optimum calculated length of the crystal was $L_{\rm opt} \sim 8$ mm). To increase the conversion efficiency in OPO1, we injected at the signal wave a part of the supercontinuum radiation (0.8 – 1.1 μm) obtained upon focusing a part of the 7-μJ second harmonic radiation into a 5-mm thick SiO_2 plate.

The supercontinuum radiation energy in the 0.8 – 1.1-μm range was 0.1 – 10 nJ. When the energy of the second harmonic was 260 μJ, the radiation energy at 1.4 μm was 7 μJ and its duration was 80 ± 20 fs. For an OPO operating in the 8 – 10-μm range (OPO2), a 3-mm $LiInS_2$ crystal ($\varphi = 35°$, $\theta = 90°$, fsf type) without an AR coating was used. The crystal was pumped by the fundamental 1.24-μm radiation and the 1.4 – 1.5-μm pulse of an idler wave of the intermediate OPO was used for injection at a signal wave. The energy was measured with a cooled (up to temperature of −200 °C) CdHgTe detector, which was calibrated at a wavelength of 1.5 μm. By placing dispersion filters in front of the detector, we estimated the IR emission spectrum and measured the dependence of the conversion efficiency on the generated radiation wavelength. A change in the SHG efficiency changed the ratio between the fundamental radiation used to pump OPOs at 8 – 10 μm and the second harmonic used for injection in the 1.4 – 1.5-μm range. In this way, the optimisation of the conversion efficiency of both OPOs was achieved.

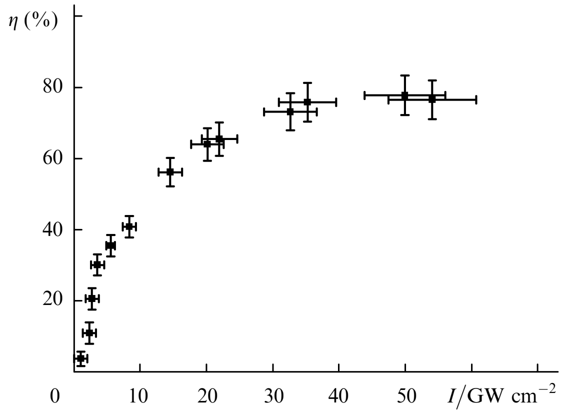

Figure 2. Dependences of the SHG efficiency for a femtosecond Cr : forsterite laser in the 5-mm LBO crystal on the pump intensity.

Figure 3 shows the wavelength dependences of the output energy in the 8 – 11-μm range and the spectrum of the 8-μm radiation. The measurements were performed for the following parameters of the pump radiation and injection: $\lambda_{\rm p} = 1.24$ μm, pulse duration $\tau_{\rm p} = 100 - 140$ fs, energy $E_{\rm p} = 160$ μJ, intensity $I_{\rm p} = 140 - 200$ GW cm^{-2}; $\lambda_{\rm in} = 1.4 - 1.5$ μm, $\tau_{\rm in} = 80$ fs, $E_{\rm in} \sim 0.2 - 1$ μJ, $I_{\rm in} = 0.2 - 1$ GW cm^{-2}. The experimental data are shown by points and the results of calculations based on the spectral model – by curves. The maximum energy $E = 1 \pm 0.5$ μJ was achieved at 9.5 μm, which corresponds to the energy and quantum conversion efficiency of 0.6 % and 4.6 %,

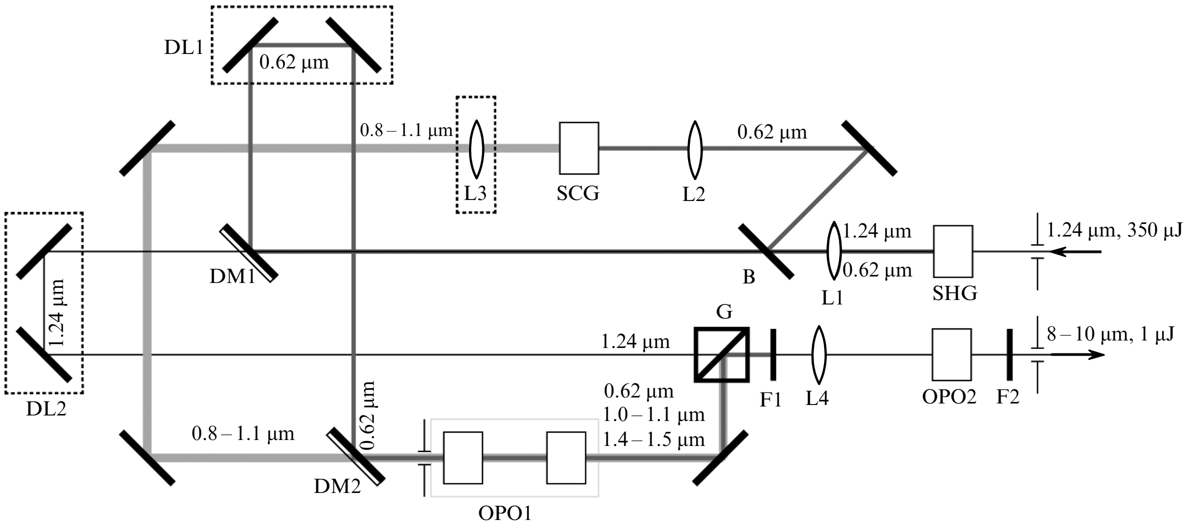

Figure 1. Scheme of the experimental setup: (DM1, 2) dichroic mirrors ($R = 100$ % at 620 nm and $T = 100$ % at 1240 nm); (L1 – L4) lenses ($f_1 = 1000$ mm, $f_2 = f_3 = 100$ mm, $f_4 = 154$ mm); (DL1, DL2) delay lines; (F1) IKS3 filter; (F2) dispersion filter; (G) Glan prism; (SHG) second harmonic generator; (SCG) supercontinuum generator; (OPO1, OPO2) nonlinear optical crystals; (B) beamsplitter.

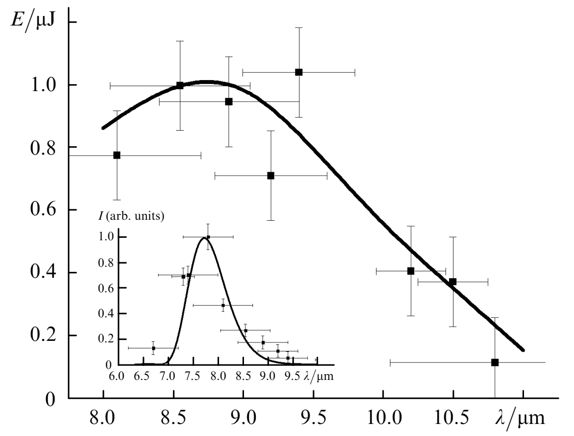

Figure 3. Experimental (points) and calculated (curve) wavelength dependences of radiation energy generated in the 8 – 11-μm range in the 3-mm $LiInS_2$ crystal pumped by a femtosecond Cr:forsterite laser (the insert shows the 8-μm emission spectrum).

respectively. Taking into account reflection losses from the faces of the crystal without AR coatings, the conversion efficiencies were 0.8 % and 5.8 %, respectively. The signal-wave gain (1.4 – 1.5 μm) was 45. The width of the spectral pulse at 8 μm was 880 nm, which corresponds to the duration of a transform-limited pulse of 130 fs. A decrease in the conversion efficiency with increasing the wavelength is caused by a rise in the crystal absorption [15] and the Manly – Row ratio.

The use of a pump Cr : forsterite laser, the scheme with an intermediate OPO and an optimal $LiInS_2$ crystal allowed us to obtain a high conversion efficiency in the 8 – 11-μm range, close to 1 %. The calculations confirmed by the experimental results obtained in this paper show that the 3-% limiting OPO efficiency at 10 μm can be achieved in a 4-mm $LiInS_2$ crystal. In a 5-mm LBO crystal, SHG is also realised in the regime of conservation of the radiation quality with the efficiency of 75 %.

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