Midinfrared optical parametric oscillator based on the wide-bandgap $BaGa_4S_7$ nonlinear crystal

Aleksey Tyazhev,[1] Dmitri Kolker,[2] Georgi Marchev,[1] Valeriy Badikov,[3] Dmitrii Badikov,[3] Galina Shevyrdyaeva,[3] Vladimir Panyutin,[1] and Valentin Petrov[1,*]

[1]Max-Born-Institute for Nonlinear Optics and Ultrafast Spectroscopy, 2A Max-Born-Strasse, D-12489 Berlin, Germany

[2]Novosibirsk State Technical University, 20 K. Marx Pr., Novosibirsk, 630072, Russia and Institute of Laser Physics (SB-RAS), 13/3 Lavren'teva Pr., Novosibirsk 630090, Russia

[3]High Technologies Laboratory, Kuban State University, 149 Stavropolskaya Street, 350040 Krasnodar, Russia

**Corresponding author: petrov@mbi-berlin.de*

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The orthorhombic biaxial crystal $BaGa_4S_7$ has been employed in a 1064 nm pumped optical parametric oscillator generating <6 ns long idler pulses with energies as high as 0.5 mJ at 6.217 μm and average power of ~50 mW at 100 Hz. Notwithstanding the relatively low nonlinearity, ~3 times above threshold operation has been achieved at pump intensities more than 5 times below the crystal surface damage limit.

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Nonoxide nonlinear optical crystals are indispensable for frequency conversion of high-power, solid-state laser systems operating near 1 μm (e.g., Nd:YAG at 1064 nm) to the mid IR above ~5 μm, the upper wavelength cutoff limit of oxide-based materials. In order to avoid two photon absorption when using short or ultrashort pulses for high efficiency, the bandgap of such crystals should correspond to <532 nm, a condition which is met by only few chalcogenide compounds. The most prominent of them is the commercially available chalcopyrite type $AgGaS_2$ (AGS). Other such crystals with relatively wide bandgap are the related defect chalcopyrite $HgGa_2S_4$, which is extremely difficult to grow; the orthorhombic $LiGaS_2$, $LiInS_2$, $LiGaSe_2$ and $LiInSe_2$, which exhibit relatively low nonlinearities; the recently developed chalcopyrite $CdSiP_2$ (CSP), which is highly nonlinear and noncritically phase matchable but transparent only up to ~6.5 μm; as well as few solid solutions whose composition is almost impossible to control during growth [1,2].

$BaGa_4S_7$ (BGS) is another chalcogenide nonlinear crystal which is a new candidate for such applications. Its noncentrosymmetric orthorhombic mm^2 structure was identified as early as 1983 [3]. The BaS-Ga_2S_3 binary phase diagram has been studied in [4]. Single crystals of BGS were grown by the Bridgman–Stockbarger technique in [5] and the SHG effect was confirmed by the Kurtz powder test. The bandgap estimated in [5] corresponds to ~350 nm (3.54 eV) and the transparency extends up to 13.7 μm at the 0 level. In [6], we measured the three refractive indices of BGS in the 0.42–9.5 μm spectral range and constructed the first set of Sellmeier equations. The correspondence between the dielectric (principal optical) axes xyz and the crystallographic axes abc of BGS, in which c coincides with the twofold symmetry axis, is $xyz = cab$ if the convention $c_0 < a_0 < b_0$ is used for the lattice parameters and $n_x < n_y < n_z$ [6]. Furthermore, the angle Ω between the optic axes and the z principal (dielectric) axis was measured to be $\Omega = 45.6°$ at 633 nm [6]. With high optical quality material, we estimated the good transmission limits of BGS, at an absorption level of 0.3 cm^{-1}, to be 0.545–9.4 μm [7]. The two nondiagonal elements of the nonlinear tensor (assuming Kleinman symmetry) were determined by SHG for a fundamental wavelength of 2.26 μm [7]: $d_{31}(5.1 \pm 0.3)$ pm/V and $d_{32} = (5.7 \pm 0.3)$ pm/V, where $d_{31}/d_{32} > 0$. Finally, BGS showed extremely high surface damage threshold at 1064 nm with 14 ns long pulses corresponding to 3.7 J/cm^2 of incident on-axis fluence (or 264 MW/cm^2 of peak on-axis intensity) [7]. Thus, while its effective nonlinearity for downconversion processes is roughly 2 times lower in comparison to the commercially available AGS, the advantage of BGS is its much higher (by an order of magnitude) surface damage threshold. This property, which is related to the large bandgap value renders BGS similar to the isostructural $LiGaS_2$ (both mm^2 point group); however, BGS is easier to grow in large sizes with good optical quality. Here we demonstrate, for the first time to our knowledge, optical parametric oscillation in the mid IR based on BGS pumped at 1064 nm.

The BGS sample employed was grown by the Bridgman–Stockbarger method using raw materials with high purity, 6Ns for Ga and S, and 99% for Ba; more details can be found in [6,7]. The as-grown crystals are normally colorless; however, the large size sample available for the present experiment (Fig. 1) had a yellow tinge.

The processed 14.05 mm long element had an aperture of 9.8×9.5 mm^2. It was cut in the x-z plane at $\theta = 9.2°$ for oo-e type I phase matching with the 9.8 mm edge parallel to the y axis (vertical in the present case), to ensure maximum effective nonlinearity d_{eff} [7]. The actual cut angle was established by Dr. K. Kato using tunable SHG for propagation near the z axis [8]. At such a small

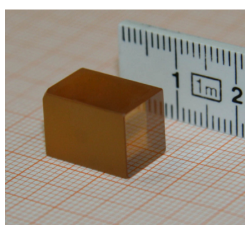

Fig. 1. (Color online) Photograph of the AR-coated BGS element.

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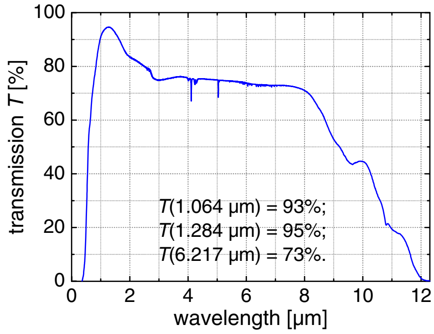

Fig. 2. (Color online) Transmission of the AR-coated BGS element measured with unpolarized light.

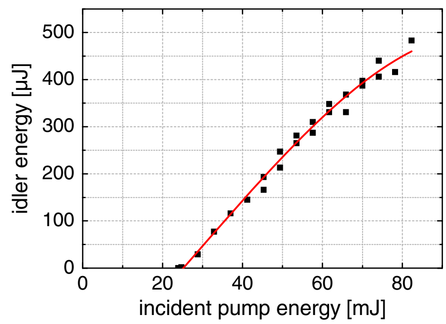

Fig. 4. (Color online) Input–output characteristics of the BGS OPO at normal incidence for a cavity length of 17 mm.

angle $d_{\mathrm{eff}} = d_{31} \cos\theta \approx d_{31}$ for normal incidence, for which the predicted idler wavelength is 6.2144 μm [8]. The walk off angle for the e-polarized pump does not exceed 10 mrad in the possible tuning range and can be neglected.

The single layer Al_2O_3 antireflection (AR) coating was specified by the supplier (ELAN Ltd.) with reflectivity of 2–3% for the pump (1064 nm) and signal (~1.3 μm) wavelengths. These values were confirmed by transmission measurements of the AR-coated sample (Fig. 2), taking into account that the crystal absorption (measured prior to coating) did not exceed 0.01 cm^{-1} at all three wavelengths. The obtained residual surface reflectivity at the idler wavelength near 6 μm (~15%) is thus totally attributed to Fresnel reflection ($n \sim 2.25$), see Fig. 2.

The optical parametric oscillator (OPO) was pumped by a diode-pumped Q-switched Nd:YAG laser/amplifier system (Innolas GmbH) delivering up to 250 mJ per pulse at 100 Hz (Fig. 3). The pump pulse duration was typically ~8 ns and the M^2 factor was measured to be ~1.4. A half-wave plate ($\lambda/2$) and a polarizer (P) were used to attenuate the pump beam and a telescope (T) was applied to expand it to a diameter of ~5.5 and ~8.8 mm in the horizontal and vertical directions, respectively. The pump beam reached the BGS crystal after reflection at a ZnSe bending mirror (BM) and passing through the plane output coupler (OC), which transmitted 82%. The OC had a transmission of 18–22% at the signal wave and ~73–84% in the idler tuning range. An Ag mirror was used as a total reflector (TR) for all three waves in a double pump pass singly resonant OPO configuration. The diaphragm (D) in Fig. 3 served to align the OPO and the filters (F) were used for suppression of the residual pump and signal pulses.

Figure 4 shows the input–output characteristics obtained at normal incidence for a minimum cavity length of 17 mm. The threshold of 25 mJ corresponds to an axial fluence of 0.134 J/cm^2 or a peak on-axis intensity of 16.7 MW/cm^2. The slope efficiency in the initial stage is ~1% with respect to the idler output but some saturation can be seen at higher powers. Nevertheless, an average idler power of ~50 mW was obtained at 100 Hz.

In the present experiment we were limited by the damage of the Ag mirror, which occurred at >50 MW/cm^2 peak pump intensity (about 5 times lower than the limit set by the BGS damage resistivity). Still this allowed us to reach 3 times above threshold pumping of the OPO. Since we tested different metallic mirrors and the situation could not be improved, the most straightforward solution to this problem seems the use of longer (>20 mm) BGS elements in the future which will reduce the OPO threshold. This should be possible with BGS, in contrast to some other nonoxide materials, due to the relatively low residual absorption losses (Fig. 2).

The idler wavelength at normal incidence was 6.217 μm, in excellent agreement with calculations [8]. The OPO linewidth, measured at the signal wavelength using a 1 mm thick Ag-coated CaF_2 Fabry–Perot etalon, was ~60 GHz(~2 cm^{-1}).

Tuning was studied by tilting the crystal at a slightly lengthened cavity (Fig. 5). The idler wavelength range extended from ~5.5 to ~7.3 μm with a pronounced enhancement at normal incidence due to the idler reflection by the crystal surfaces. This makes the OPO quasi-doubly resonant at normal incidence. The minimum idler wavelength reached at $\theta = 0°$ (noncritical interaction) was 5.47 μm while calculations predict 5.38 μm [8]. While

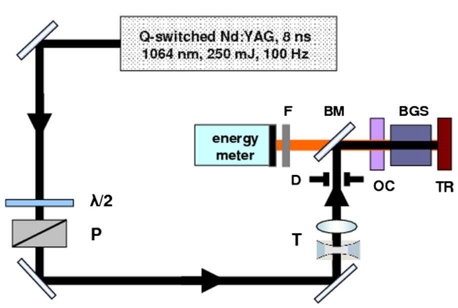

Fig. 3. (Color online) OPO experimental setup.

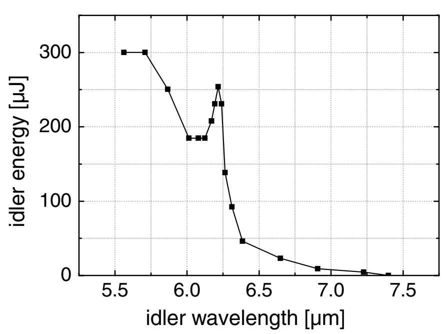

Fig. 5. Tuning characteristics of the BGS OPO for a cavity length of 20 mm recorded at an incident pump energy of 58 mJ.

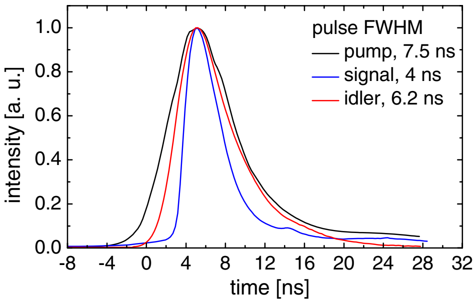

Fig. 6. (Color online) Simultaneously measured temporal pulse profiles of the pump, signal, and idler.

the refined Sellmeier equations predict a longest idler wavelength of 8.4 μm at the retracing point of $\theta = 13.2°$ [8], the longest wavelength reached for which a similar phase-matching angle could be experimentally measured was ~7.5 μm. Careful analysis of the tuning behavior in Fig. 5 showed that the decreasing d_{eff} and parametric gain alone cannot account for the strong decrease of the conversion efficiency at long idler wavelengths. The parameter that drastically changes from noncritical phase matching ($\theta = 0°$) to the retracing point ($\theta = 13.2°$) is the angular acceptance, decreasing by more than an order of magnitude to a value of ~3 mrad, comparable to the collimation accuracy of the pump beam. This effect could be easily avoided in the future by improved alignment of the pump telescope.

The temporal characteristics of the BGS OPO were measured at maximum pump level using fast photodiodes and 2 GHz oscilloscope. The (HgCdZn)Te detector used for the idler (Vigo systems model PCI-9) had a time constant of <2 ns. As can be seen from Fig. 6, both signal and idler exhibit shorter durations compared to the pump which is typical for the intensity-dependent nonlinear process in the OPO. The actual idler pulse duration should be <6 ns due to the finite detector response.

As with CSP [9], the M^2 value obtained with the BGS OPO in this short cavity was far above the value corresponding to diffraction-limited beams: for the nonresonated idler we measured $M^2 \sim 10$. This can be attributed to a combination of factors, including the large pump diameter, short pump pulse duration and cavity length, and the operation ~3 times above threshold. However, in contrast to CSP, the interaction in BGS is critical and substantial improvement of this parameter can be expected implementing the RISTRA concept [9].

In conclusion, we demonstrated that chalcogenide nonlinear crystals with relatively low nonlinearity but wide bandgap and high damage resistivity are very attractive for OPOs pumped at 1064 nm for frequency conversion to the mid IR above ~5 μm. In fact, comparing with our previous experience with crystals of AGS and $LiInSe_2$, with the present BGS crystal we reached the highest ratio of pump power above threshold (~3) without surface damage. In the present experiment, the damage limit was set by the Ag total reflector. Thus, we were able to reach with BGS the same level of idler pulse energy as with the highly nonlinear CSP [10] for which the threshold in terms of pump energy was lower by about an order of magnitude. Since there were no thermal problems with BGS and no cumulative damage occurred, we were able to pump it also at increased repetition rate of 100 Hz (most previous experiments with such 1064 nm pumped chalcogenide crystals were performed at 10 Hz [2]) achieving the highest ever average powers (~50 mW) in the 6 μm spectral range with such an OPO.

The results of the present experiment indicate that a tradeoff should be searched in the quest for new nonlinear materials for the mid IR: we reached similar OPO output energy limits with BGS and CSP, although CSP has >16 times higher effective nonlinearity since the damage resistivity of BGS is at least 10 times better. In addition, while critical phase matching has to be applied with BGS, its tuning potential is much broader. Future work will be directed toward increasing the crystal size of good optical quality BGS elements and the study of the related monoclinic $BaGa_4Se_7$, which exhibits higher nonlinearity [6].

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