Long-wave IR source based on $GaSe_{1-x}S_x$

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***Abstract*—$GaSe_{1-x}S_x$ crystals were used for long-wave mid-IR generation by KTP OPO down-conversion. Optimal composition $GaSe_{1-x}S_x$, shown 6 times higher efficiency than that of GaSe. Significant structure strengthening was achieved with Al-doping.**

I. INTRODUCTION

A tunable long-wave mid-IR source is the key to chemical identification, reflection spectroscopy of solids and isotope separation of heavy elements [1].

The narrow-bandwidth tunable source operating over the entire mid-IR and THz ranges was demonstrated by mixing (down-conversion) of MOPO idler waves and residual Nd:YAG emission in layered GaSe [2,3]. This monochromatic source can be used, for example, as a seed oscillator to extend spectral range & scaled-up power mid-IR laser system that includes cryogenic CO laser and unique multiprocess frequency converter [4]. On the other hand, the layered structure resulted in extreme low mechanical properties that limits its application. Fortunately, GaSe is an excellent matrix material for doping with different elements. HeavilyS-doped (from 2 to 3 mass %) or solid solutions $GaSe_{1-x}S_x$, x=0.09-0.133, shown impressive results in usability and frequency conversion efficiency due to, relatively, strengthened structure and improved optical properties [5]. Although $GaSe_{1-x}S_x$ are still layered structures, they are much useful for processing and suitable for out-of-lab applications. Tunable wide-spectral-bandwidth mid-IR OPG [6] and narrow bandwidth THz mixer [7] using selected (not optimal composition) $GaSe_{1-x}S_x$ demonstrated up to 3 times higher frequency conversion efficiency at fixed and up to 15 times at maximal possible pump intensity. The efficiency of the mixing can be further improved by optimization of S-content but further lattice strengthen for wider utilization by doping with second element. In particular, Al-doping in GaSe demonstrated highest increase in the hardness [8].

In this study $GaSe_{1-x}S_x$ compositions (x=0.09, 0.112, and 0.133) that are within the optimal range [5], and (0.01-0.75 at. %) Al-doped $GaSe_{1-x}S_x$ were tested for down-conversion into long-wave mid-IR and THz range in comparison with that for GaSe. It is well known that GaSe doping with isovalent elements allow someone to control a range of its optical properties within broad limits on the growth stage by chemical composition control. Used dopants should form isostructural binary compounds. Among them S-doping demonstrates best results in the improvement of optical properties. Doping with isovalent Al that does not form isostructural binary compound allowed someone to change hardness up to 3 times [9]. Al concentrations were selected within optimal doping limits of 0.01-0.05 for pure GaSe crystals and larger that, relatively, optimizes optical quality and maximizes hardness at still suitable optical quality for application. A tunable narrow-bandwidth ($\Delta\nu$=0.075 cm^{-1}) idler wave of KTP OPO SUNLITE EX, model PR 8000/OPO and residual pump emission ($\Delta\nu$=0.003 cm^{-1}) of Nd:YAG laser (Continuum Electro-Optics, Inc.) were used as the pump sources.

2. RESULTS

Studied samples were manufactured by cleaving from the as-grown boules and not processed. Solid solutions $GaSe_{1-x}S_x$ samples were high (α<0.1 cm^{-1}) optical quality. To the best of our knowledge additive effect from double-element (S and Al) doping was clearly observed for the first time. In the result, both optical and mechanical properties were improved additively.

Fig.1 presents phase matching conditions for down-conversion (mixing) into mid-IR.

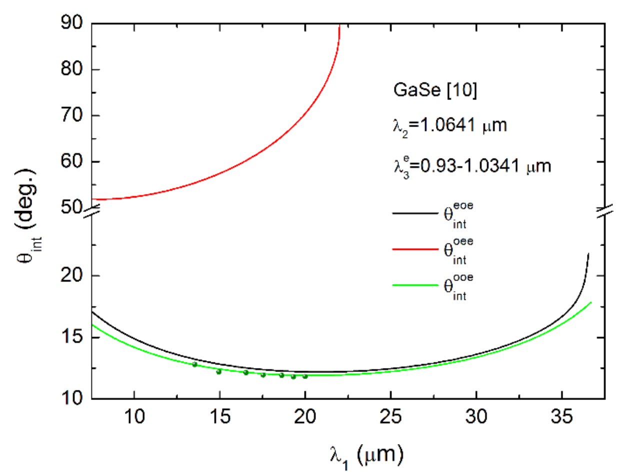

Fig. 1. Estimated (lines) and measured (points) PM angles for mixing of KTP OPO (λ=0.9835 -1.0103 μm) and residual Nd:YAG emissions (points) in GaSe.

Simple optical setup for designed oe-e type mixer is shown in Fig.2.

Beam cross sections of the OPO (pulse duration 8 ns, 20 mW average power) and its pump Nd:YAG laser (10 ns, PRF 10 Hz, 2 times higher average power) were different shape and coincide only for about 50%. Anyway, generated mid-IR emission was detected by Gollay cell (Tydex GC-PS/1, Russia) operating at room temperature. Main attention was paid to generation of 13-20 μm emission (OPO wavelength range (λ=0.9835-1.0103 μm) useful for heavy isotope separation, gas and solid state matter monitoring. Total spectral range covered by generated THz emission is 75-3774.8 μm that was monitored by LHe-cooled Si bolometer.

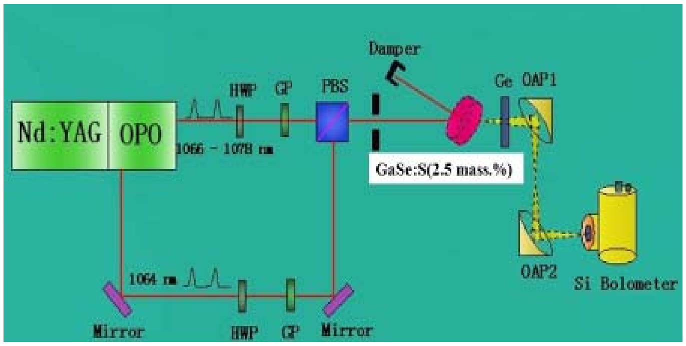

Fig.2. Optical setup at the down-converter of OPO and Nd:YAG emissions into mid-IR and THz range.

Beam cross sections of the OPO (pulse duration 8 ns, 20 mW average power) and its pump Nd:YAG laser (10 ns, PRF 10 Hz, 2 times higher average power) were different shape and coincide only for about 50%. OPO and Nd:YAG pulse durations mismatch for 20%. Anyway, generated mid-IR emission was detected by Gollay cell (Tydex GC-PS/1, Russia) with SNR up to few hundreds. Output signal of LHe-cooled Si bolometer shown saturation at about reached output signal of 9 Volts during detection of the near field short wave (75-300 μm.) THz emission (Fig.2b).

The $GaSe_{0.888}S_{0.112}$ or 2.5 mass% S-doped GaSe was of the best optical quality and shown over 3 times higher down-conversion efficiency to that for GaSe at identical experimental conditions. Doping with 0.05 at.% Al-doped increases efficiency for 15-20% but at pre-damage pump intensity it increases efficiency up to 5 times. It seems that double element doping results in accumulative effect of each single element doping. This crystal pumped into mechanical surface defect shown for the first time in our experience cracking similar to a glass. Optical quality of $GaSe_{0.888}S_{0.112}$ doped with 0.75 at.% of Al was noticeably degraded but still estimated as optical grade by naked eye. It feels as of a “metal-grade” hardness.

Remote detection of THz emission at wavelength up to 1 mm was tested. A sample of the measurement trace containing six 50-mm reflecting mirrors is shown in Fig.3.

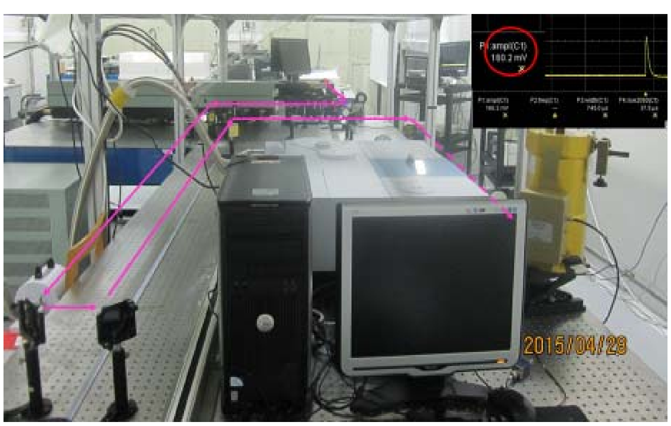

Fig.3. External view on remote (10.1 m) detection of the THz emission with Si bolometer.

One (120 mm on the crystal output) or two (120 mm on the crystal output and 240 mm before Si bolometer) was also used to collect THz emission onto sensitive area of the detector detector disposed at over 10 m distance from down-converter. As large as up to 0.4 V signal was recorded during detection of 1.55 THz emission. Detected signal dropped down few tens of mV with the wavelength increasing due to divergence increasing.

Designed monochromatic source can be used as a spectroscopic tool or as a seed source in design of long-wave scaled-up power laser system with extended operation range. Optical properties of some products manufactured from bio objects (paper, different fabrics of natural fibers, wood) were successfully tested, as well as atmospheric water vapor absorption. No attempt was made yet to optimize crystal lengths as well to pump maximal pump intensity or use larger aperture optics. Therefore, there is a serious potential to enlarge SNR ratio or measurement trace.

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