## High-resolution terahertz spectrometer with up to 110 m single-pass base

Z.-M. Huang<sup>a</sup>, J.-G. Huang<sup>a</sup>, Y.-Q. Gao<sup>a</sup>, Q.-J. Yang<sup>a</sup>, J. Wu<sup>a</sup>, Y. Qu<sup>a</sup>, Yu.M. Andreev<sup>b,c,d</sup>,

K.A. Kokh<sup>c,d,e</sup>, G.V. Lanskii<sup>b,c,d</sup>, A.A. Lisenko<sup>f</sup>, V.A. Svetlichnyi<sup>c,d</sup>

<sup>a</sup> Shanghai Institute of Technical Physics CAS, Shanghai, China
<sup>b</sup> Institute of Monitoring of Climatic and Ecological Systems SB RAS, Tomsk, Russia
<sup>c</sup> Siberian Physical-Technical Institute of Tomsk State University, Tomsk, Russia
<sup>d</sup> High Current Electronics Institute SB RAS, Tomsk, Russia
<sup>e</sup> Institute of Geology and Mineralogy SB RAS, Novosibirsk, Russia
<sup>f</sup> Institute of Atmospheric Optics SB RAS, Tomsk, Russia

**Abstract:** Terahertz spectrometer up to 110 m single-pass measurement trace was realized by room-temperature GaSe:S down-converter operating at 0.2 - 0.6 THz range with  $0.1 \text{ cm}^{-1}$  high resolution. *Ab-initio* measurements were carried out in the real atmosphere.

Keywords: THz; GaSe:S; Down-converter; High-resolution.

## I. INTRODUCTION

Last decade searching for new low optical loss, high nonlinear and damage threshold, suitably anisotropic materials resulted in rediscovery of heavily S-doped GaSe (GaSe:S) or solid solution crystals  $GaSe_{1-x}S_x$ , where x is the mixing ratio [1]. Those crystals demonstrated impressive results in usability due to the lattice strengthening, as well as in efficiency of down-conversion into the mid-Infrared & Terahertz ranges due to improved optical properties. The best optical quality possessed optimally (2-3 mass %) S-doped GaSe or GaSe\_{1-x}S\_x, x=0.09-0.133. Small amount (0.01-0.05 at. %) of Al-doping in GaSe significantly strengthened the lattice and improved the optical quality. Accumulative effect was interested in this crystal after optimal double element doping, special by S and Al [2-6].

Last year, we reported first application (spectroscopic tool) of the designed monochromatic ( $\Delta v < 0.1 \text{ cm}^{-1}$ ) THz source based on GaSe<sub>1-x</sub>S<sub>x</sub>:Al down-converter or difference frequency generator (DFG) [7]. A liquid He cooled Si bolometer was exploited as a standoff detector located at 10 m distance from the down-converter with response time 0.8 msec. Now, the single-pass base of the spectrometer is extended to up to 110 m with high pressure PE lens assembly for THz beam uptake and fast THz detectors based on Schottky diode operated at room temperature.

## II. RESULTS

In this investigation single crystal of solid solution GaSe<sub>91</sub>S<sub>0.09</sub> doped by 0.03 at. % of Al is used as the down-converter from near IR into the THz range. Preferably type II of three-wave interaction is realized due to simplicity of the optical setup alignment. A tunable narrow-bandwidth,  $\Delta v=0.075$  cm<sup>-1</sup>, idler or signal wave of a KTP OPO SUNLITE EX, model PR 8000/OPO and its residual pump emission of a Nd:YAG laser (Continuum Electro-Optics Inc., USA) ( $\Delta v=0.003$  cm<sup>-1</sup>) operating at 10 Hz PRF are used as the pump sources. Pump beam parameters are not optimized. Multimode beam cross sections of the OPO (pulse duration 3.8 ns, 40 mW average)

power, horizontal polarization) and pump Nd:YAG laser (10 ns, PRF 10 Hz, 100 mW average power) are different shape and coincide maximum for 50%. Peak powers match well in time by optical delay line. No focusing lens is used for pump beams. Two periscopes are used to adjust both pump beams onto the crystal input facet independently. DFG optical setup is shown in Fig. 1. Spectral bandwidth of the generating THz emission is estimated to be less than 0.1 cm<sup>-1</sup>.



Fig.1. Optical setup of the down-converter: P<sub>1,2</sub>, periscopes; D<sub>1,2,3</sub>, diaphragms; WP<sub>1,2</sub> Wollaston prisms; L, THz beam collimating lens;  $\lambda/2$ , half wavelength plate; F, PE filters to block pump beams; arrows show beam polarization.

To detect the generated THz signal, both liquid He cooled Si bolometer with sensitive area  $\emptyset 2$  mm and noise-equivalent power ~  $4.3 \times 10^{-14}$  W/Hz<sup>1/2</sup>, and room temperature THz detector based on Schottky diode are used for comparison. Homemade lens of high pressure PE are used to collect THz beam by standoff detectors. The first collimating lens with focus lengths of 12.5 cm is installed after the crystal output. Other focus length is between 0.6 to 5 m (low fabrication quality is found). The Schottky diode detector is mounted with a silicon lens on the surface, and integrated with a low-noise signal amplifier: (effective THz range 50-1200 GHz, electric bandwidth 10-10<sup>6</sup> Hz, responsivity 27000 V/W @ 70 GHz and 1400 V/W @ 1000 GHz at 25 °C). Its noise-equivalent power is ~ 6 pW/Hz<sup>1/2</sup>. Teledyne-Lecroy WaveRunner 610Zi scope (4 GHz, 1 mV noise level) is used to record detector signals. The designed THz source (Fig. 2) is simple in construction, easy to alignment, but not optimized and can be further improved.



Fig. 2. Top view on the down-converter: 9V (upper left corner inset) and strong saturated (upper right corner inset) signal on the output of Si bolometer.

The insets in Fig. 2 show the recorded waveforms of the two detectors. The peak voltage is over 9 V of the power supply and strongly saturated on the output of Si bolometer, which confirms a high efficiency of THz generation in this crystal. For the Schottky diode, the up-limitation of output THz signal is 3 V set by the manufacture. The signal response sensitivity of the room temperature detector is demonstrated about 10 times higher than the liquid He cooled Si bolometer at wavelength  $\sim 1$  mm.

Estimations and preliminary test of THz spectrometer operating at 0.5-1 mm range with a single pass distance of 20, 50 and 80 m respectively in lab conditions show little attenuation by the atmosphere except for the well resolved separated water absorption lines. As large as 824 mV signal with 80 m standoff distance is recorded by A1 detector. It is found that the number of lens could be noticeable reduced due to the insignificantly decrease signal.

Finally, complicated single THz pass trace with 8 flat Al mirrors and up to 16 high pressure PE lens is established in the lab surrounding by two close crossing corridors. Compared for the large signal at 80 m, significant signal decrease (down to 25-30 mV) over 110 m occurs due to improper adjustment with last two lens of 5 m long focus distance. However, it still allows us to record the absorption spectra with SNR $\leq$ 30.

Designed spectrometer can be considered as the first step in design of long (up to 1 km) trace out-of-door system because of several reasons: (1). Thin (2.5 mm) GaSe<sub>1-x</sub>S<sub>x</sub>, x=0.09-0.133 crystal doped with 0.02 at. % of Al seems to be good choice as the down-converter into mid-IR and THz range. Its lattice is significantly strengthened (demonstrates ability to cracking), and suitable for out-of-door applications. Absorption coefficient  $\alpha \approx 0.2$  cm<sup>-1</sup> at 1 THz is minimal amongst known nonlinear crystals. Its efficiency can be improved by optimization geometrical parameters, operation temperature, antireflection coating, etc.; (2). Manufactured DFG prototype is simple, easy alignment and portable device (Fig.1a) that is

suitable for out-of-door (mobile system) applications; (3). Down-conversion efficiency can be further improved by using longer crystal (now only 2.5 mm length crystal is used), optimization of the parameters of pump beams and other optical parts or by utilizing of lidar type transmitting/receiving optical systems; (4). Presence of suitable transparency windows in the atmospheric 0.3-0.6 THz spectra is experimentally confirmed in the study.

Besides, not only unpolished Al and copper plates, but also strong crumpled and spread Al foils can be used as reflectors in this THz region. Further optimization of output electronic parts of THz detector – Schottky diode will give us the ability to control time shape-form of the recorded signal.

As a conclusion, we have realized a prototype of up to 110 m single-pass measurement trace terahertz spectrometer in GaSe:S down-converter with 0.1 cm<sup>-1</sup> high resolution. The distance can be further extended to 1 km for remote practical out-of-door applications.

This work is supported by China Science Found, China, Grant No. 61274138, 16JC1403400, 15ZR1445500; Russia Science Found, Grant No.15-19-10021 (crystal growth, modeling, co-operative experimental study).

## REFERENCES

- K.R. Allakhverdiev, R.I. Guliev, E.Yu. Salaev, V.V. Smirnov, "Investigation of linear and nonlinear optical properties of GaSe<sub>x</sub>Se<sub>1-x</sub> crystals", Soviet J. Quant. Electron., **12**(1982), 947-948.
- [2] J. Guo, J.J. Xie, D.J. Li, G.L. Yang, F. Chen, C.R. Wang, L.M. Zhang, Yu.M. Andreev, K.A. Kokh, G.V. Lanskii, V.A. Svetlichnyi, "Doped GaSe crystals for laser frequency conversion (Review)", Light: Science & Applications 4(2015) e362.
- [3] K.A. Kokh, J.F. Molloy, M. Naftaly, Yu.M. Andreev, V.A. Svetlichnyi, G.V. Lanskii, I.N. Lapin, T.I. Izaak, A.E. Kokh, "Growth and optical properties of solid solution crystals GaSe<sub>1-x</sub>S<sub>x</sub>", Materials Chemistry and Physics, 154(2015), 1 52-157.
- [4] J. F. Molloy, M. Naftaly, Yu. Andreev, K. Kokh, G. Lanskii, V. Svetlichnyi, "Absorption anisotropy in sulfur doped gallium selenite crystals studied by THz-TDS", Optical Materials Express, 4(2014), Issue 11, 2451-2459.
- [5] M. Naftaly, J.F. Molloy, Yu.M. Andreev, K.A. Kokh, G.V. Lanskii, V.A. Svetlichnyi, "Dispersion properties of sulfur doped gallium selenide crystals studied by THz TDS", Optics Express 23(2015), No.25, 32820– 32834.
- [6] J. Guo, D.-J. Li, J.-J. Xie, L-M Zhang, Z.-S. Feng, Yu.M. Andreev, K.A. Kokh, G.V. Lanskii, A.I. Potekaev, A.V. Shaiduko, V.A. Svetlichnyi, "Limit pump intensity for sulfur-doped gallium selenide crystals", Laser Phys. Lett., 11(2014), No.5, 055401.
- [7] Z.-M. Huang; J-G Huang; Y-Q Gao; Yu M. Andreev; K. A. Kokh; G. V. Lanskii; I. N. Lapin; V. A. Svetlichnyi, "Long-wave IR source based on GaSe<sub>1-x</sub>S<sub>x</sub>" Proc. of 40th Int. Conf. on Infrared, Millimeter, and Terahertz Waves (IRMMW-THz 2015), Hong Kong, 23-28.08.2015, (2015) 1-2.