2015 International Siberian Conference on Control and Communications (SIBCON)

A comparison of terahertz electro-optic sampling in ZnTe, ZnSe, GaP and $GaSe_{1-x}S_x$ crystals

R. A. Red'kin, S. A. Bereznaya,
Z. V. Korotchenko, S. Yu. Sarkisov
Functional Electronics Laboratory
Tomsk State University, TSU
Tomsk, Russia
ruseg89@mail.ru

Abstract — **Efficiency of terahertz electro-optic sampling in a number of II-VI, III-V and III-VI crystals have been experimentally compared and analyzed. The terahertz pulses generated in femtosecond laser plasma filament were electro-optically detected in the crystals under investigation and recorded using conventional THz-TDS setup. Influence of crystal natural properties and chemical compound on the recorded spectra is discussed. In particular, influence of sulfur concentration in $GaSe_{1-x}S_x$ mixed crystals on efficiency of terahertz detection was tested.**

Keywords — electroptic-sampling; terahertz radiation; terahertz time-domain spectroscopy; gallium selenide

I. Introduction

Electro-optic sampling is one of the most widely used methods for terahertz detection in terahertz time-domain spectroscopy (THz-TDS). The process is based on polarization changing of probing optical pulse in an electro-optic medium under action of terahertz field. The resulting appearance of orthogonal to initially linearly polarized beam polarization component can be provided by Pockels or Kerr effects as well as by nonlinear sum or difference frequency generation. For efficient interaction of the optical and terahertz beams in the detecting medium the well-known group velocity matching condition must be satisfied. For birefringent nonlinear crystals also phase-matching conditions are important. Basically the requirements to the electro-optical materials are close to those for terahertz emitters via optical rectification and the materials used for both applications are, in many cases, the same [1].

One of the best nonlinear crystals for terahertz generation and detection in THz-TDS is gallium selenide. It is also widely used for nonlinear frequency conversions in IR [2, 3] and terahertz ranges [4, 5]. Gallium selenide possesses unique properties for nonlinear generation of high-intensity (peak electric fields up to 100 MV/cm [6, 7]) and ultrabroadband terahertz pulses for terahertz time-domain spectroscopy.

In order to modify the optical properties of GaSe and to improve its intrinsically low mechanical properties extensive theoretical and experimental studies of doped crystals have been performed [8-12]. One of the most studied dopant in GaSe is sulfur. The mixed $GaSe_{1-x}S_x$ crystals are known to become harder with increasing sulfur content. The nonlinearity and refractive indexes are changed as well. This influences the terahertz generation and detection efficiency. Applications of $GaSe_{1-x}S_x$ crystals with relatively low and high sulfur content for terahertz detecting is less studied compared to generation processes.

In the present study we compare terahertz detection in Bridgman grown GaSe, GaSe:S 0.9 mass % and GaSe:S 7 mass % crystals and in widely used electro-optic sensors ZnTe [13], as well as some other crystals. Zinc telluride is the most widely used terahertz electro-optic sensor. Gallium selenide has advantage in high terahertz frequencies (>8 THz) and when very broad spectral range is required [6, 14].

II. Experimental details

Zinc telluride and selenide had zinc blende structures and (110) and (111) orientations respectively. The trigonal $LiNbO_3$ and hexagonal $GaSe_{1-x}S_x$ and $CdSe_{1-x}S_x$ were z-cut. The GaP crystal, available in our experiments was with unknown orientation. All the crystals had thicknesses of 1-2 mm.

The experiments were done at conventional THz-TDS setup, sketched at Fig. 1. To obtain terahertz pulses we used laser pulses of Spitfire Pro (Spectra-Physics, USA) regenerative amplifier with 800 nm central wavelength, 35 fs pulse width and 3.5 mJ pulse energy. Terahertz pulses were generated in air plasma and BBO (θ=29°) crystal with 200 μm thickness was placed 2.5 mm before filament to generate second harmonic at 400 nm wavelength. It is required to accelerate the charges in air plasma [15].

This work was supported by The Ministry of Education and Science of the Russian Federation (Project No 3.1206.2014)

978-1-4799-7103-9/15/$31.00 ©2015 IEEE

All the detector crystals were placed normally to the probe and terahertz beams and were optimized on azimuth angles. The optical transmission spectra were recorded at room temperature using USB 4000+ (Ocean Optics, USA) spectrometer with incandescent lamp as a light source.

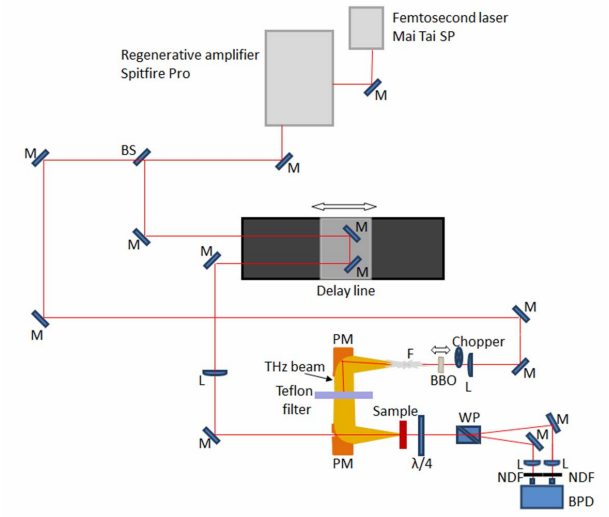

Fig. 1. Sketch of the experimental setup, M – mirror, L – lens, PM – parabolic mirror, WP – Wollaston prism, NDF – neutral density filter, λ/4 – quarterwave plate, BPD – balance photodiodes, BS – beam splitter, F – filament.

III. RESULTS

The measured transmission spectra are given in Fig. 2.

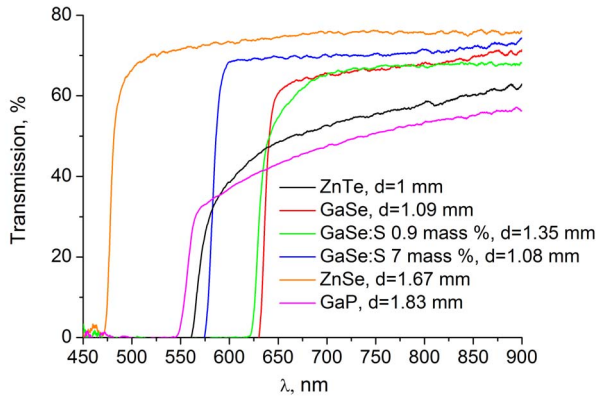

Fig. 2. Optical transmission spectra of the studied crystals. Thicknesses of the samples are given on the plot.

It can be seen that ZnSe possesses the highest transparency, while in ZnTe and GaP the transmission losses are remarkably higher. The shift of the fundamental absorption edge of GaSe:S crystals with increasing sulfur content is also clearly seen (Fig. 2).

The highest signal exceeding noise level up to about 5.5 THz was registered with ZnTe (Fig. 3). It is seen that in pure GaSe the signal exceeds the noise level up to 2.5 THz. A dip at 0.59 THz present at the spectrum detected in GaSe is connected with well-known phonon absorption resonance [12]. Gallium selenide possesses restrahlen band in frequency region 4–8 THz that explains the absence of the registered signal in this region. The dip at 0.59 THz vanishes in GaSe:S 0.9 mass % but the detection efficiency is lower at frequencies higher than 0.8 THz compared to pure GaSe. In the case of GaSe:S 7 mass % the efficiency is remarkably lower.

The thicknesses of ZnSe and GaP crystals were probably too large and dips which can be attributed to the anti-synchronism can be observed at 0.7 and 1.3-1.5 THz.

In $LiNbO_3$ very low signal was detected, only five times exceeding noise level (not shown in Fig. 3). We checked also $CdSe_{1-x}S_x$ mixed crystals in the wide range of x and no electro-optic sensing was observed.

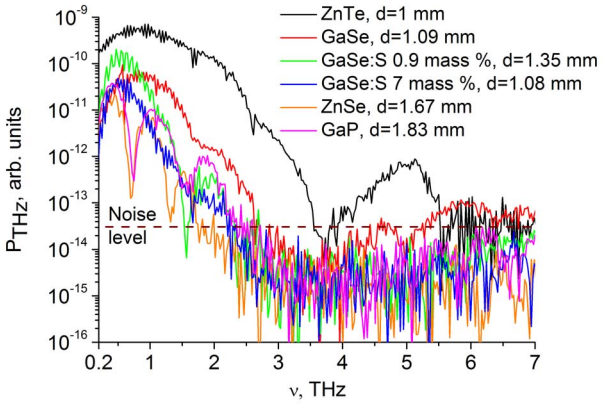

Fig. 3. Terahertz spectra registered in the studied crystals. Thicknesses of the samples are given on the plot.

While Pockels effect is the mechanism of terahertz interaction with the probe optical pulse in ZnTe, GaP and ZnSe crystals possessing no birefringence, in the $GaSe_{1-x}S_x$ crystals difference and sum frequency mixing can be more important [14].

In the case of GaSe sum and difference frequency mixing can result in appearance of orthogonal polarization component of the probe beam. The following phase-matching conditions can be written for these processes

$$\omega_{opt} n(\omega_{opt}) \pm \omega_{THz} n(\omega_{THz}) = (\omega_{opt} \pm \omega_{THz}) n(\omega_{opt} \pm \omega_{THz}), \quad (1)$$

where ω_{opt} and ω_{THz} are cyclic optical and terahertz frequencies and n represents refractive indexes.

The group velocity of the optical beam must be equal to phase velocities of terahertz spectral components that gives the following condition [1]

$$\Delta k(\nu, \lambda) = [n_{gr}(\lambda) - n_{THz}(\nu)] \frac{2\pi\nu}{c} = 0, \quad (2)$$

where Δk is phase mismatch, ν is a terahertz frequency and λ is optical probe pulse central wavelength. Group refractive index n_{gr} can be calculated by formula

$$n_{gr}(\lambda) = n(\lambda) - \lambda \frac{dn(\lambda)}{d\lambda}\Big|_{\lambda}. \quad (3)$$

For all the crystals the detection efficiency was sensitive to rotation on azimuth angle φ around the surface normal. In

the case of GaSe the efficient nonlinearity depends on the internal phase-matching angle θ and azimuth angle φ when both ordinary and extraordinary waves interact as

$$d_{eff}=d_{22}\cos^2\theta\cos 3\varphi \qquad (3)$$

or

$$d_{eff}=-d_{22}\cos\theta\sin 3\varphi. \qquad (4)$$

This in particular explains the fact that in many cases the terahertz detection in GaSe is most efficient at normal incidence. According to (3) and (4) at rotating on azimuth angle 3 maximums and 3 minimums of signal were observed in $GaSe_{1-x}Se_x$ crystals.

IV. Conclusion

The efficiency of terahertz detection in different spectral regions was compared in ZnTe, GaP, ZnSe and GaSe:S crystals. It was found that ZnTe is efficient up to frequencies higher than 5 THz. The detection efficiency in GaSe:S 0.9 mass % is lower at frequencies higher than 0.8 THz compared to pure GaSe. The effect is determined by phase-matching conditions and nonlinearity change. At high sulfur content (GaSe:S 7 mass %) the efficiency is remarkably lower due to lower nonlinearity.

References

[1] M. M. Nazarov, S. A. Makarova, A. P. Shkurinov, O. G. Okhotnikov, "The use of combination of nonlinear optical materials to control terahertz pulse generation and detection," Applied Physics Letters, vol. 92, pp. 021114-1–021114-3, 2008.

[2] R. Hegenbarth, A. Steinmann, S. Yu. Sarkisov, H. Giessen, "Milliwatt-level mid-infrared (10.5-16.5 μm) difference frequency generation with a femtosecond dual-signal-wavelength optical parametric oscillator" Optics Letters, vol. 37, pp. 3513-3515, 2012.

[3] R. Hegenbarth, A. Steinmann, S. Mastel, S. Amarie, A.J. Huber, R. Hillenbrand, S.Yu. Sarkisov, H. Giessen, "High-power femtosecond mid-IR sources for s-SNOM applications"// Journal of Optics, vol. 16, pp. 094003-1–094003-7, 2014.

[4] W. Shi, Y.J. Ding, "Tunable coherent radiation from terahertz to microwave by mixing two infrared frequencies in a 47-mm-long GaSe crystal," International Journal of High Speed Electronics and Systems, vol. 16, pp.589-595, 2006.

[5] A. A. Angeluts, V. V. Bezotosnyi, E. A. Cheshev, G. N. Goltsman, M. I. Finkel, S. V. Seliverstov, M. N. Evdokimov, M. V. Gorbunkov, G. K. Kitaeva, A. L. Koromyslov, P. V. Kostryukov, M. S. Krivonos, Y. V. Lobanov, A. P. Shkurinov, S. Yu. Sarkisov, V. G. Tunkin, "Compact 1.64 THz source based on a dual-wavelength diode end-pumped Nd:YLF laser with a nearly semiconfocal cavity," Laser Physics Letters, vol. 11, p. 015004, 2014.

[6] A. Sell, A. Leitenstorfer, R. Huber, "Phase-locked generation and field-resolved detection of widely tunable terahertz pulses with amplitudes exceeding 100 MV/cm" Optics Letters, vol. 33, pp. 2767-2769, 2008.

[7] O. Schubert, M. Hohenleutner, F. Langer, B. Urbanek, C. Lange, U. Huttner, D. Golde, T. Meier, M. Kira, S.W. Koch, R. Huber, "Sub-cycle control of terahertz high-harmonic generation by dynamical Bloch oscillations" Nature Photonics, vol. 8, pp. 119 -123, 2014.

[8] N.B. Singh, D.R. Suhre, V. Balakrishna, M. Marable, R. Meyer, N. Fernelius, F.K. Hopkins, D. Zelmon, "Far-infrared conversion materials: Gallium selenide for far-infrared conversion applications," Progress in Crystal Growth and Characterization of Materials, vol. 37, pp. 47-102, 1998.

[9] V.V. Atuchin, S.A. Bereznaya, N.F. Beisel, Z.V. Korotchenko, V.N. Kruchinin, L.D. Pokrovsky, A.I. Saprykin, S. Yu. Sarkisov, "Growth, chromium distribution and electrical properties of GaSe:Cr single crystals," Materials Chemistry and Physics, vol. 146, pp. 12-17, 2014.

[10] V.N. Brudnyi, A.V. Kosobutsky, S.Yu. Sarkisov, "Charge neutrality level and electronic properties of GaSe under pressure," Semiconductors, vol. 44, pp. 1158-1166, 2010.

[11] A.V. Kosobutsky, S. Yu. Sarkisov, V.N. Brudnyi, "Structural, elastic and electronic properties of GaSe under biaxial and uniaxial compressive stress," Journal of Physics and Chemistry of Solids, vol. 74, pp. 1240-1248, 2013.

[12] M. M. Nazarov, S. Yu. Sarkisov, A. P. Shkurinov, O. P. Tolbanov, "$GaSe_{1-x}S_x$ and $GaSe_{1-x}Te_x$ thick crystals for broadband terahertz pulses generation," Applied Physics Letters, vol. 99, pp. 081105-1–081105-3, 2011.

[13] C. Winnewisser, P. U. Jepsen, M. Schall, V. Schyja, H. Helm, "Electro-optic detection of THz radiation in $LiTaO_3$, $LiNbO_3$, and ZnTe" Applied Physics Letters, vol. 70, pp. 3069-3071, 1997.

[14] K. Liu, J. Xu, X-C. Zhang, "GaSe crystals for broadband terahertz wave detection*"* Applied Physics Letters, vol. 85, pp. 863–865, 2004.

[15] M. Kress, T. Loffler, S. Eden, M. Thomson, H. Roskos, "Terahertz-pulse generation by photoionization of air with laser pulses composed of both fundamental and second-harmonic waves" Optics Letters, vol. 29, pp. 1120–1122, 2004.