Model studies of THz-range generation by down-conversion in GaSe and GaSeS crystals

D.M. Lubenko[1,2], V.F. Losev[1,2,3], Yu.M. Andreev[2,4], G.V. Lanskii[2,4]
[1] High Current Electronics Institute SB RAS, 2/3, Akademicheskii Ave., 634055, Tomsk, Russia, lideru@gmail.com;
[2] National Research Tomsk State University, 36, Lenina Ave., 634050, Tomsk, Russia;
[3]National Research Tomsk Polytechnic University, 30,LeninaAve., 634050, Tomsk, Russia;
[4]Institute of Monitoring of Climatic and Ecological Systems SB RAS, 10/3,Akademicheskii Ave.,634055, Tomsk, Russia;

ABSTRACT

Model study of not phase matched and phase matched optical rectification or down-conversion of Ti:Sapphire laser pulses at 950 nm into THz and far-IRrange in pure and S-doped GaSe single crystals is carried out. First, the ordinary and extraordinary wave dispersions of the GaSe refractive indices were measured by terahertz time-domain spectroscopy (THz-TDS). Measured data were approximated in the form of Sellmeier dispersion equations for 0.62 – 2000 μm range with using available shorter wave data.

Keywords: GaSe crystal, model study, nonlinear optics, THz

1. INTRODUCTION

Over the past decades, frequency conversion through using phase matchable (anisotropic) nonlinear crystals has found more and more applications in producing THz coherent sources. The key to this technique lies in the nonlinear crystals, as they have to possess low optical loss, high damage threshold and large nonlinearity coefficient leading to high output power and hence can meet the increasing power demand in various applications. Besides, crystals should be hard for easy access and processing at arbitrary direction with fine finish. However, there is limited number of anisotropic inorganic nonlinear crystals suitable for THz applications that always possess much lower optical loss, higher damage threshold and larger hardness to that for organic crystals. Small optical loss and higher pump intensity is resulting in higher conversion efficiency. Larger hardness made easy access and processing at arbitrary direction.

Most suitable inorganic acentrosymmetric crystals for THz frequency converters are $ZnGeP_2$[1,2,3],ε-GaSe[4,5] (herein after GaSe), $AgGaSe_2$[6], $AgGa_{1-x}In_xSe_2$[6]and Tl_3AsSe_3[7]crystals. Among them, GaSe crystal is of extra interest due to extreme physical properties that allowed broadband highly efficiency parametric frequency conversion. For example, DFG within 2.7-38.4 & 58.2-3540 μm[8]and further up to 5640 μm[4]is realized[6]. In the total, frequency conversion is realized within 0.7895-5640 μm[4,8,9]; output power THz pulse power reached is up to 2 kW[10]; up to 10-50 MW is predicted. Interesting way for generation of THz emission, by realizing of parallel sum frequency generation of one non-selective CO laser, and difference frequency generation of sum frequencies and rest pump emission in a single GaSe sample at fixed angular position was recently proposed[11].

Many of the unique physical properties of GaSe are associated with its layered structure. The basic four-fold layer consists of two monoatomic sheets of Ga sandwiched between two monoatomic sheets of Se. The strong covalent interaction within the atomic layers and week, Van-der-Waals type bonding between basic layers, renders GaSe as a highly anisotropic material. On the other hand, the layer structure results in extreme low hardness (almost zero by Mohs scale) and easy cleaving along planes parallel to the atomic layers, and finally in hampering of out-of-door large-area crystals applications.

Fortunately, an originalε-polytype structure of GaSe is strengthening doping with different elements, as well other physical properties responsible for frequency conversion efficiency are modifying. It allowed easier processing at arbitrary directions and improves frequency conversion efficiency. Modified properties and improved efficiencies are

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reported for a number of doped crystals: light (GaSe:S) and heavely S-doped GaSe crystals that also referred to as solid solution crystals GaSe:GaS ($GaSe_{1-x}S_x$, where x is mixing ratio)[12,13,14,15,16], GaSe:In and $Ga_{1-x}In_xSe$[17,18,19,20,21], GaSe:Te and $GaSe_{1-x}Te_x$[19,22,23], doped GaSe:Er[24,25,26],GaSe:Al[27,28,29], GaSe:Ag[30], and GaSe:InSe or $Ga_{1-x}In_xSe_{1-y}S_y$[31] crystals. Increased frequency conversion efficiency is recorded for frequency conversion into both mid-IR[13,17,18,20] and THz[32,33,34,35]range. Recently, summarized data were presented in few papers[36-40], which consider some other double element doped GaSe crystals. Strengthened structuregives opportunity of the application in out-of-door applied systems[41].

Due to a set of modified parameters: increased damage threshold, decreased phase matching angle, lower absorption and refraction, short-wavelength shifted transparency and phase matching ranges etc. the highest frequency conversion efficiency was recorded for solid solution $GaSe_{1-x}S_x$ crystals[13,33,34]. In particular, for Er^{3+}:YAG laser SHG conversion efficiency in optimally composition $GaSe_{1-x}S_x$ crystal was of 2.4 times higher to that for pure GaSe crystal, as well as for THz generation by Ti:Sapphire laser frequency down-conversion. In contradiction, negative effects of S-doping on the optical damage threshold and on frequency conversion efficiency were also reported[16,42] that reflect doping-induced degradation in optical quality.

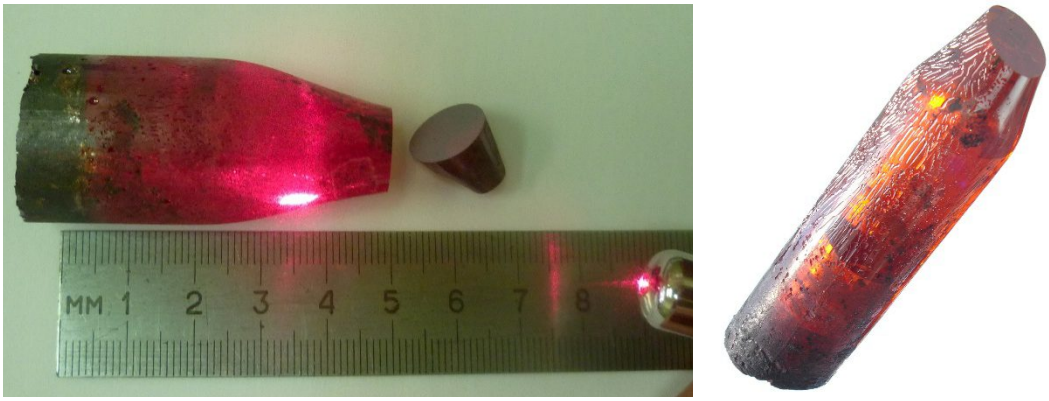
In fact, differences in the state-of-the-art of growth technology, limited distribution of doped GaSe crystals and still problematic cut and high optical quality polishing of pure and doped GaSe crystals are reasons of paucity and highly scattered data on optical properties of pure and doped GaSe crystals in THz range. Until recently Due to limited distribution and hard processing absorption spectra for e-wave in $GaSe_{1-x}S_x$ crystals (i.e. absorption anisotropy properties) in the THz range have only been studied for two solid solution compositions: $GaSe_{0.74}S_{0.26}$[15] and $GaSe_{0.71}S_{0.29}$[16]. From data in these studies and measurements at fixed frequencies[4,34] it was established that the absorption coefficient α_o exceeds $\alpha_e$at THz frequencies as it does in the pure GaSe crystal. This difference in absorption loss leads to a higher efficiency of THz e-wave generation[8,34]. It was also predicted and confirmed experimentally that the uncommon ee-e type of interaction can be realized in pure and S-doped GaSe crystals[15,16].

Successful design of THz sources calls for adequate data on PM possibilities and potential efficiencies for all possible three frequency interactions. In turn, it needs in correct data on dispersion properties and absorption spectra over the entire transparency range for pure and S-doped GaSe and solid solution $GaSe_{1-x}S_x$ crystals as a function of the mixing ratio. Correct data are a crucial factor in the selection of the most efficient type of three frequency interactions and in maximizing the frequency conversion efficiency. Original processing technology allowed fabrication of a range of high optical quality S-doped crystals at θ=90°. Recently, by using these and cleaved samples of the same composition, refractive indices for ordinary (o) and extraordinary (e) waves were successfully measured[43]. However, analyses of phase matching were not carried out.

In the present work, we report model study of phase matching for different typesof three frequency interactions in GaSe and $GaSe_{1-x}S_x$crystals by using designed dispersion equations.Two Ti:Sapphire lasers that are available in our lab (operating, respectively, at 950 nm and 980-1080 nm) are considered as pump sources.

2. CRYSTAL GROWTH AND CHARACTERIZATION

A modified synthesis of polycrystalline material and the vertical Bridgman single crystal growth method were employed to grow single crystals of solid solution $GaSe_{1-x}S_x$. The starting materials for the synthesis were Ga 99.9997, Se 99,99 and S 99.95. The stoichiometric charge of Ga and Se, and the nominal 0, 1.1, 2.5, 5, 7 mass.% S (x=0, 0.05, 0.11, 0.22, 0.29, 0.44) was weighed out. Synthesis ampoules were loaded up to 65% in volume to minimize the quantity of residual gases and cosequent interaction so as to improve optical quality. Other details on the synthesis process are reported elsewhere[44]. After several hours of melt homogenization during the synthesis process, the temperature was slowly decreased to 40 K below the melting point of 1238 K of the compound at the rate of ~10 K/h. For the growth process, the polycrystalline material was loaded into a single wall cylindrical ampoule. The internal surface had a layer of pyrolytic carbon which protected the melt from reaction with the ampoule wall material and impurities. The unseeded crystal growth was performed by the vertical Bridgman method with heat field symmetry change and the symmetry center moving all over the oven space that is described elsewhere[45,46]. The sealed growth ampoule was loaded into a furnace having a temperature gradient of ~15 K/cm at the estimated level of crystallization front. After homogenization of the melt at the temperature 30 K above the melting point, the ampoule was mechanically lowered at the speed of 10 mm/day. No eutectic was found by visual examination. The grown boule allows easy sample cleaving with high optical quality surfaces up until the end section.Photos of doped GaSe crystals are shown in Fig. 1.



a b

Figure 1.External view on as-grown high quality GaSe:Scrystals: (a) 1 mass% and (b) 11 mass% S-doped.

Two types of $GaSe_{1-x}S_x$ samples were fabricated for the present study. The first type was cleaved from as-grown boules, i.e. it had faces orthogonal to the c-axis, so that a beam traversing the sample travelled parallel to the c-axis. The high optical quality of these samples can be estimated by the naked eye, evident in their transparency and homogenity. The second type was mechanically cut and polished. These samples were made by first immersing a section of the $GaSe_{1-x}S_x$ boule in monomer (polymetil crylate) mixed with a thermoinitiator and placed in an oven for polymerization for 2 hours. The produced samples of both types were free from precipitates, voids or micro bubbles, or other visual defects.

Optical properties in mid-IR range were studied by using homemade spectrophotometers but THz-TDS (time-domain spectroscopy) measurements of o- (α_o) and e-wave (α_e) absorption coefficient spectra and absorption anisotropy in the 0.3-4.0 THz range for solid solution crystals $GaSe_{1-x}S_x$ as it is described in details elsewhere[39,40]and commerce Z-3 (Zomega, USA) spectrometer. It was found that grown GaSe crystals possesses from 2 to 3 times lower absorption coefficient that crystals grown by common syntheses and single crystal growth technology. In line, optimally 2-3 mass% S-doped GaSe crystals also demonstrated 2-3 times lower absorption coefficient to that for pure GaSe crystals grown by modified technology. So such, it seems us that measurement results are quite adequate.

3. MODEL STUDY

Dispersion properties for GaS and GaSe were measured in THz range and approximated in the form of dispersion equations all over the entire transparency range. Available data for from visible to through mid-IR were also used in the approximation[43].

Phase matching angles for difference frequency generation (DFG) were calculated by using well known relations:

$$\frac{1}{\lambda_{pump}} = \frac{1}{\lambda_{signal}} - \frac{1}{\lambda_{idle}}, \quad (1)$$

considering phase matching condition as k_s-k_i-$k_p \leq 10^{-4}$. In relation (1)

$$k_0 = \frac{n_o}{\lambda}, \quad (2)$$

$$k = \frac{\frac{n_0 n_e}{\sqrt{n_o^2 sin^2\theta + n_e^2 cos^2\theta}}}{\lambda}. \quad (3)$$

Dispersion equations for the entire transparency range of GaSe[43]:

$$n_o^2 = 10.6409 + \frac{0.3788}{\lambda^2 - 0.1232} + \frac{6963.32}{\lambda^2 - 2198.85} + \frac{0.017\lambda^2}{\lambda^2 - 262177.5577}, \quad (4)$$

$$n_e^2 = 5.76 + \frac{3879}{\lambda^2} - \frac{0.2288}{\lambda^4} + \frac{0.1223\lambda^2}{\lambda^6} + \frac{0.4206\lambda^2}{\lambda^2 - 1780.3}. \quad (5)$$

Dispersion equations for the entire transparency range of GaS[47]:

$$n_o^2 = -\frac{0.01129}{\lambda^6} + \frac{0.03648}{\lambda^4} + \frac{0.51402}{\lambda^2} + 6.59624 + \frac{2.71047\lambda^2}{\lambda^2 - 1025.42116}, \quad (6)$$

$$n_e^2 = \frac{0.0113}{\lambda^6} + \frac{0.10569}{\lambda^4} - \frac{0.44573}{\lambda^2} + 4.92144 + \frac{0.315\lambda^2}{\lambda^2 - 720.12225}. \quad (7)$$

Phase matching estimation for solid solution crystals $GaSe_{1-x}S_x$ can be carried out by using relationship proposed in[48] that is, in particular, adapted for solid solution GaSe:GaS in[49] as follows:

$$n_{o,e}^2(GaSe_{1-x}S_x) = (1-x)n_{o,e}^2(GaSe) + xn_{o,e}^2(GaS) \quad (8)$$

4. RESULTS AND DISCUSSION

Calculated dispersions and birefringence for pure and S-doped GaSe crystals are presented in the Fig. 2-5.

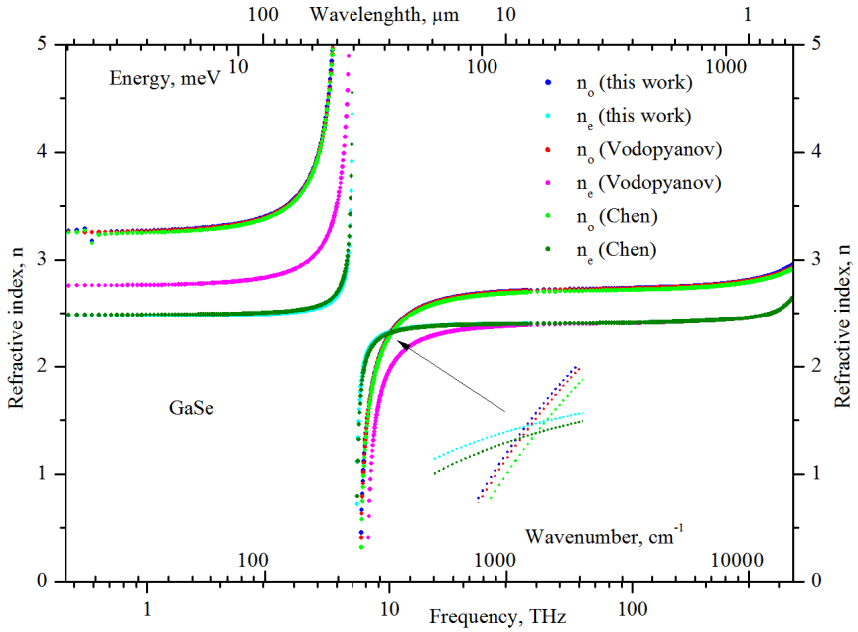


Figure 2. Comparison calculated refraction indices for GaSe crystal.

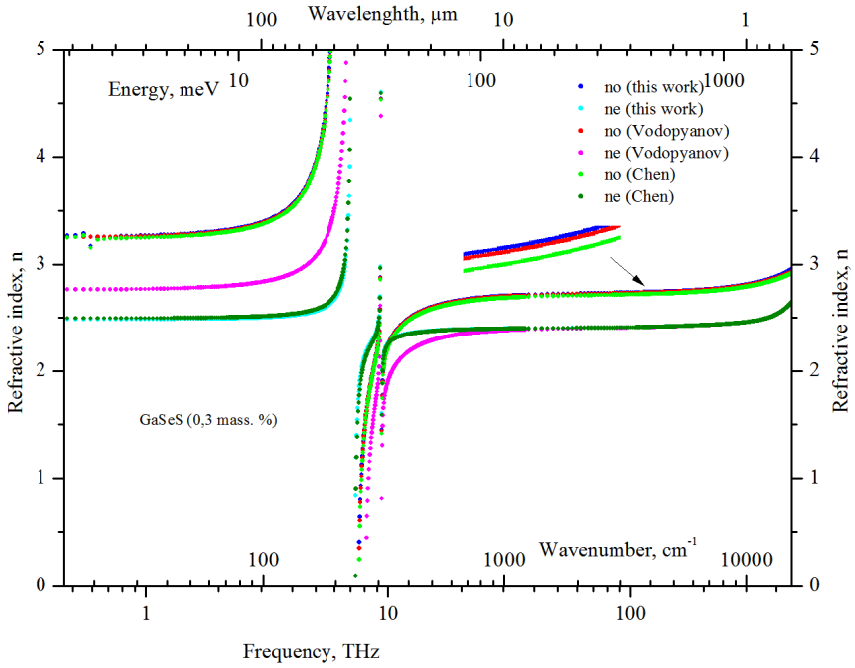


Figure 3. Comparison calculated refraction indices for GaSeS (0.3 mass/%) crystal

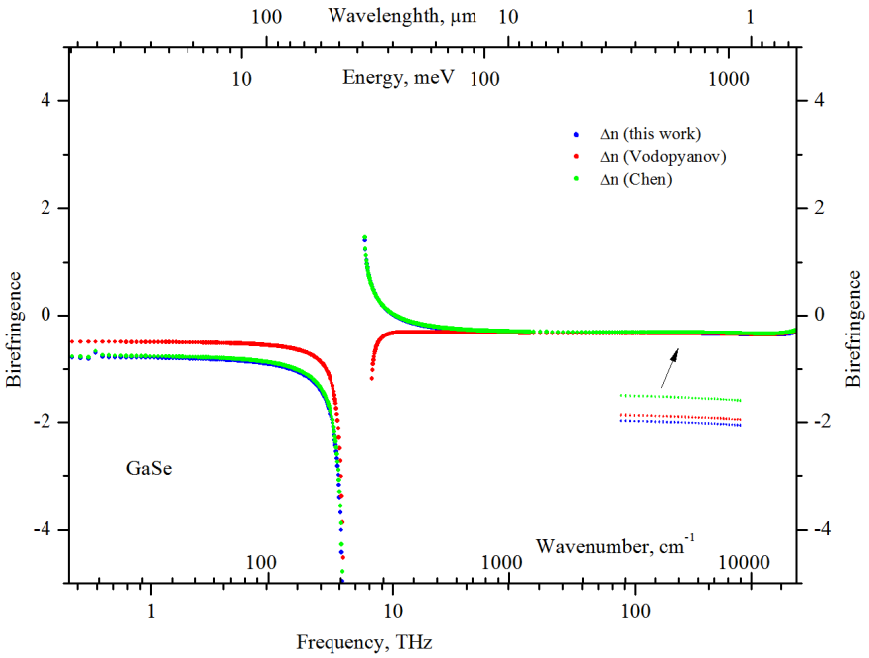


Figure 4. Comparison calculated birefringence for GaSe crystal

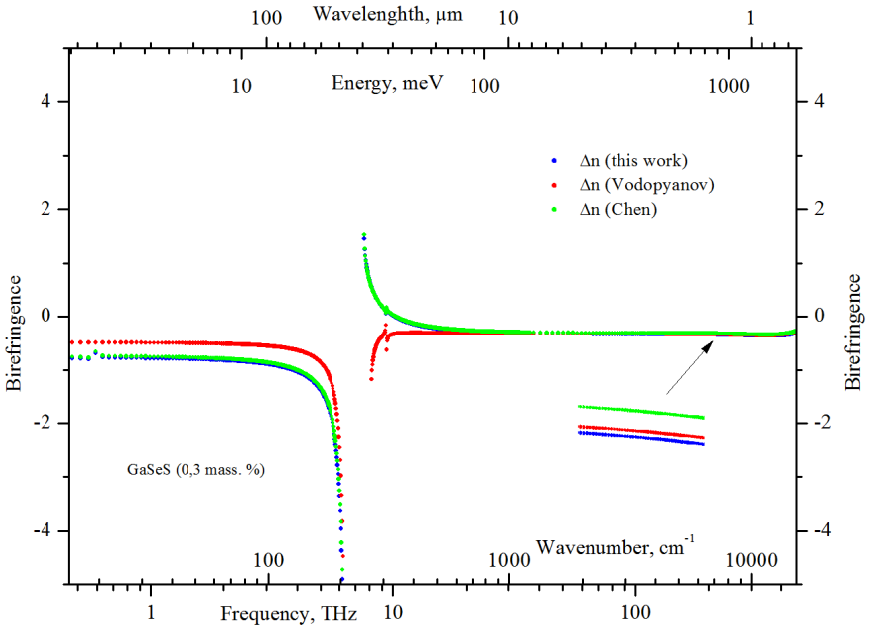


Figure 5. Comparison calculated birefringence for GaSeS (0.3 mass.%) crystal

In Fig. 2-5 it is seen that n_e dispersions in the THz range recorded in this study are huge, almost for 2 times, different as estimated from widely used dispersion equations from Vodopyanov et all. On the one hand, recorded data are close to that determined by Chen and earlier measured data[50].

DFG PM estimated by using different dispersion equations[51,52]for interactions resulting in e-wave THz generation are plotted for comparison in Fig. 6,7.

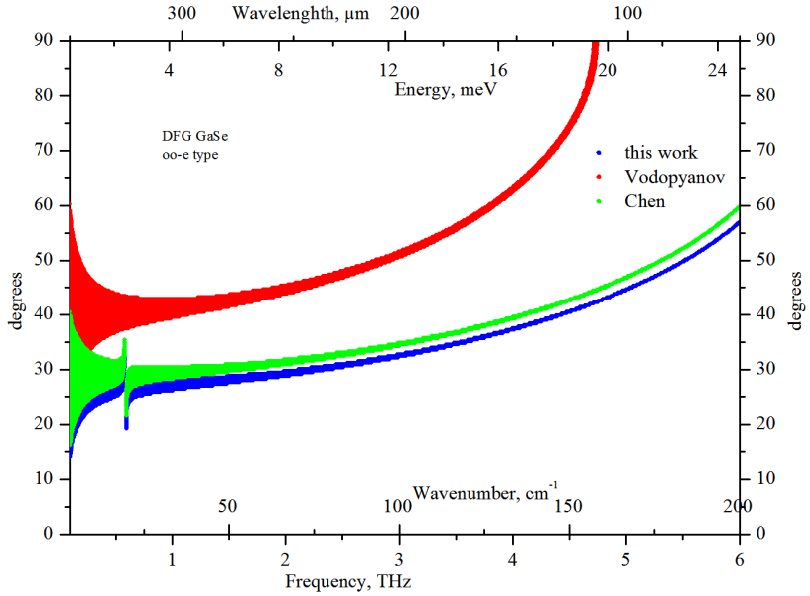


Figure 6. Comparison of calculated PM angles for GaSe crystal foroo-e interaction type

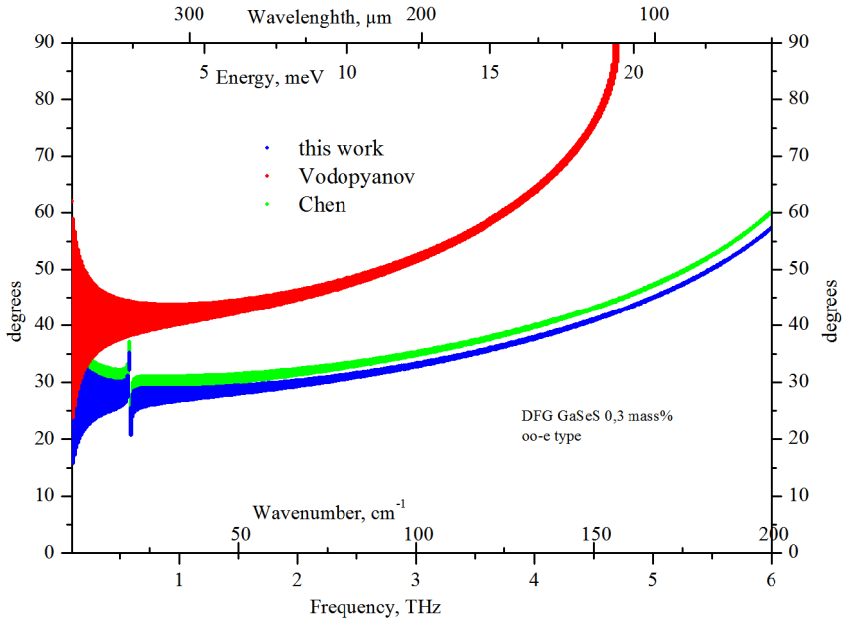


Figure 7. Comparison of calculated PM angles for GaSeS (0.3 mass/%) crystal for oo-e interaction type.

In Fig.6-7 it is seen that, as it should be bearing in mind differences in n_e dispersions in Fig.2-5, PM conditions for down-conversion of 930 and 970 nm Ti:Sapphire lasers estimated by using dispersion equations by Vodopyanov et all. differ significantlyto that estimated with new proposed[47] and Chen's et all.[51] equations. On the other hand, it is in contradiction

with the fact of wide using of Vodopyanov's dispersion equations and well matching of experimental and estimated PM angles. It can be explained by differences in the magnitudes of PM angles for considered down-conversion processes. Common down-conversion process in published papers arerelated to down-conversion of 1.064 μm Nd:YAG laser and OPO operating at close wavelengths. It occurs at small internal PM angles anything like 2-4 degrees. In this case, n_e magnitudes are close to n_o magnitudes that are identical for all dispersion equations presented in Fig. 2-5. For down conversion of Ti:Sapphire lasers PM angles are large and difference in the n_e dispersion became important. It resulted in significant differences of PM angles.

This result is very important for our prospective study on down-conversion of available 950 nm as and tunable 780-1080 nm Ti:Sapphire lasers.

5. CONCLUSION

Improved quality crystals were grown by modified technology and used in these measurements. Dispersion properties of o- and e-wave refractive indices and absorption coefficients for GaSe and $GaSe_{1-x}S_x$ crystals were preliminary measured by THz-TDS, approximated in the equation form and then used in the study. Model study of THz generation in pure and solid solution $GaSe_{1-x}S_x$ crystals by Ti:Sapphire laser is carried out.

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