ISSN 1062-8738, Bulletin of the Russian Academy of Sciences: Physics, 2019, Vol. 83, No. 3, pp. 237–241. © Allerton Press, Inc., 2019.
Russian Text © Y.M. Andreev, A.A. Ionin, I.O. Kinyaevskiy, Y.M. Klimachev, 2019, published in Izvestiya Rossiiskoi Akademii Nauk, Seriya Fizicheskaya, 2019, Vol. 83, No. 3, pp. 292–296.

Numerical Study of Multiline CO Laser Frequency Conversion in a $ZnGeP_2$ Crystal to the THz Range

Y. M. Andreev[a, b], **A. A. Ionin**[c], **I. O. Kinyaevskiy**[c, *], **and Y. M. Klimachev**[c]

[a] *Institute of Monitoring of Climatic and Ecological Systems, Siberian Branch, Russian Academy of Sciences, Tomsk, 634055 Russia*

[b] *Siberian Physicotechnical Institute, National Research Tomsk State University, Tomsk, 634050 Russia*

[c] *Lebedev Physical Institute, Russian Academy of Sciences, Moscow, 119991 Russia*

**e-mail: kigor@sci.lebedev.ru*

Received September 3, 2018; revised September 10, 2018; accepted October 22, 2018

Abstract—A numerical simulation was performed for frequency conversion of multiline CO laser radiation to the THz range by difference frequency generation in a ZnGeP2 crystal. It was shown that the difference frequencies spectrum can be tuned in the wavelength interval of 70–1300 μm by tuning the phase-matching angle in the range of 11° to 55°. The maximal peak power of difference frequencies radiation (integral over the spectrum) was ~0.23 W at 45° phase-matching angle, which corresponds to a peak power conversion efficiency of 6×10^{-5}.

DOI: 10.3103/S1062873819030043

INTRODUCTION

Research on developing systems for monitoring the atmosphere in the terahertz (THz) band is now under way around the world, for many reasons. The first of these is the presence of a window of transparency in the shortwave end of the atmosphere and the absence of the absorption spectra of water vapor as the main perturbing factor in its longwave end. The long wavelength of THz radiation minimizes scattering by dust particles and precipitation, while the high power of its penetrating into nonpolar liquids and nonconducting solids minimizes its attenuation in them. The safety for living organisms resulting from the low energy of radiation quanta allows the widespread use of systems based on the sources of THz-radiation in actual practice.

The introduction of highly sensitive detectors with subnano- and nanosecond resolution (cryogenic superconducting bolometers and room-temperature Schottky diodes in particular) are advantageous technical factors. These detectors have stronger (by 1–2 orders of magnitude) voltage responses when detecting nanosecond probe pulses than the widely used cryogenic silicon bolometers with response constants in the millisecond range, and allow the development of lidar-type systems based on nanosecond sources of radiation and having kilometer-long metering paths [1]. The optical qualities of the atmosphere also allow the creation of such measuring instruments [2]. Using highly efficient generators of picosecond pulses of THz radiation with pumping by a high-power Ti:sapphire laser system [3, 4] did not allow us to go beyond kilometer-long metering atmospheric paths of the atmosphere. Neither were we able to demonstrate the possibility of measuring small components of the atmosphere, or to create simple and easy to use sources of radiation for systems to monitor them [3, 5]. It is therefore of great interest to search for ways of creating powerful, reliable, and simple nanosecond sources of terahertz (THz) radiation.

CHOOSING A NONLINEAR CRYSTAL AND A PUMPING LASER

The use of nonlinear crystal optics with second-order nonlinearity is a promising way of generating THz radiation. However, the choice of the nonlinear process (parametric frequency conversion (PFC)), the nonlinear crystal and its parameters, and the type of three-wave interaction between waves in the crystal are key elements that determine the efficiency of PFC. The limiting efficiency of PFC is determined by the Manley–Rowe relation (i.e., the ratio of the energy of light quanta, converted to the frequency of pumping radiation) [6]. It follows that using high-energy molecular gas lasers (CO_2 and CO lasers) and the most efficient so-called standard of nonlinear mid-IR crystals ($ZnGeP_2$ crystal, or ZGP) [7] would be the best way of developing powerful sources of THz radiation. This has not, however, proven to be true in practice, at least with reference to CO_2 lasers [8–11]. Note that ZGP crystals exhibit a high degree of optical loss at the wavelengths of a CO_2 laser, and a low degree of optical

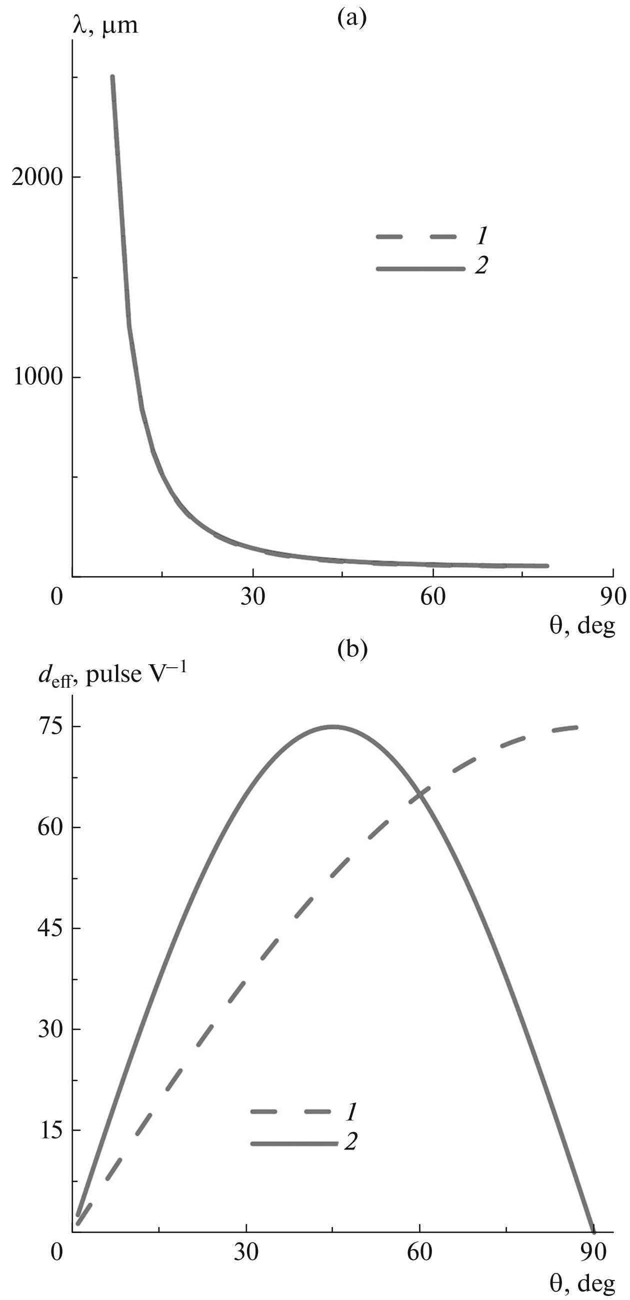

Fig. 1. Dependence of (a) the wavelength of THz radiation and (b) effective nonlinearity on phase-matching angle in a ZGP crystal: (*1*) *ooe*; (*2*) *eoe*.

loss in the THz band [12]. However, the record-breaking efficiency of PFC in the middle infrared band (more than 80% for SHG nanosecond CO_2 lasers [13]), and the record-breaking efficiency of the SHG nanosecond radiation of CO lasers [14], for which the optical losses are minimal in the transmission window of ZGP, were obtained in it. Another promising crystal for the parametric conversion of the frequency of laser radiation to the THz band is GaSe, which is also transparent in the middle infrared and THz bands. However, GaSe crystals have been found to be less efficient than $ZnGeP_2$ by a factor of 7 for frequency conversion of the CO laser radiation under the same conditions [14].

The possibility of creating a difference frequency generator (DFG) or a down converter of CO laser radiation to the THz band, was noted in [15], but the potential and possibilities of CO laser PFC for obtaining THz radiation were not considered in detail.

In this work, we performed a numerical simulation for the frequency conversion of multiline carbon monoxide laser radiation to the THz band via the collinear DFG in a ZGP crystal to examine the potential opportunities in detail. Our choice of the object of investigation seemed promising because of the rich spectrum of laser radiation in the wide range of possibilities of scaling the power of output radiation and the minimal optical loss in the crystal. Since it is a process with no threshold, the collinear generation of difference frequencies does not require the use of dielectric mirrors or antireflection coatings on the working faces of the crystal.

ANALYZING THE CONDITIONS OF PHASE-MATCHING AND EFFECTIVE NONLINEARITY

Obtaining the phase-matching of waves interacting inside a nonlinear crystal is one of the key conditions for achieving the maximum efficiency of frequency conversion [16]. Analysis of all possible three-wave interactions corresponding to DFG of CO laser radiation to the THz band shows that two types of collinear phase matching are possible in a ZGP crystal: Type I (*ooe*) and Type II (*eoe*). The notations *o* and *e* correspond to ordinary and extraordinary waves, and interacting waves (or waves denoted by indices 1, 2, and 3) are listed in descending order of their wavelength.

Our calculated dependences of phase-matching (PM) angle θ on difference frequency wavelength λ are presented in Fig. 1a. The methodology for PM angles calculation was described in, e.g., [16]. Dispersion equations from [16] for the middle infrared band and from [17] for the THz band were used in our calculations.

We can see from Fig. 1a that the PM curves overlap because of a minor difference between them. Calculations show the DFG in a ZGP crystal can be used to overlap the spectral interval of wavelengths from 60 μm (5 THz) to 3000 μm (0.1 THz) corresponding to the THz and shortwave edge of millimeter range.

To obtain a high PFC efficiency under PM, a high nonlinear response of polarization (described numerically by coefficient d_{eff} of second-order quadratic nonlinear susceptibility) must be observed. Since a ZGP crystal has point symmetry group $\bar{4}m2$, for PFC to the THz-band when the Kleiman conditions are not fulfilled, d_{eff} is calculated with the formulas [6]

$$\text{Type I: } d_{eff}(ooe) = d_{36}\sin\theta\sin 2\varphi,$$
$$\text{Type II: } d_{eff}(eoe) = (d_{14} + d_{36})\sin\theta\cos\theta\cos 2\varphi,$$

where θ is the angle of phase synchronism; φ is the azimuth angle of the direction of interaction (which is optimized with the direction of the flat of the crystal faces); and d_{ij} denotes components of the tensor of the coefficient of second-order quadratic nonlinear susceptibility.

The effective coefficients of nonlinear susceptibility for Type I and Type II interactions were calculated with coefficient $d_{36} = d_{14} = 75$ pm V^{-1} [16] and are given in Fig. 1. We can see that for the PM angles in the range of 0° to 55°, corresponding to that of difference frequency generation ~70 μm (~4.3 THz) to 3000 μm (0.1 THz), Type II PFC is more effective than Type I . Note that in [17], the DFG of CO_2 laser radiation to the THz band was done in accordance with only Type I interaction.

MODELING

PFC to the THz band was simulated numerically for the parameters of CO laser radiation of used in [18] for wide-band two-stage PFC in a ZGP crystal. The spectrum of each pulse of CO laser radiation contained 150 rotational-vibrational components in the wavelength interval of 5.0 to 7.5 μm. The peak power of radiation (integral along the spectrum) was 4 kW. The simulation was conducted in a plane-wave approximation and a predetermined amplitude of the field of pumping radiation following the procedure in, e.g., [16] and [18]. The ZGP crystal was 10 mm long and was sheared at the angle φ = 0° optimized for Type II PFC. The radius of the laser beam was 0.1 mm and was kept constant over the length of the crystal. Losses due to Fresnel reflection at the faces of crystal were not considered in our calculations.

The calculated spectra of DFG radiation at different PM angles θ are shown in Fig. 2. It was found that the spectrum of the CO laser (150 lines in the wavelength interval from 5.0 to 7.5 μm) allowed us to obtain more than five thousand lines of DFG in the wavelength interval of 50 to 5000 μm. The possibility of tuning the DFG spectrum in the THz area of the spectrum in the wavelength interval of 70 (~4.3 THz) to 1300 (~0.23 THz) μm is demonstrated in Fig. 2 for tuning the PM angle in the range from 11° to 55°.

Note that because the spectrum of the CO laser is discrete and the wavelengths of its spectral components are fixed, the spectrum of DFG radiation also has the discrete structure of a fixed set of closely spaced narrow spectral lines. The DFG spectrum is in this case tuned by optimizing the PM conditions for any specified spectral region.

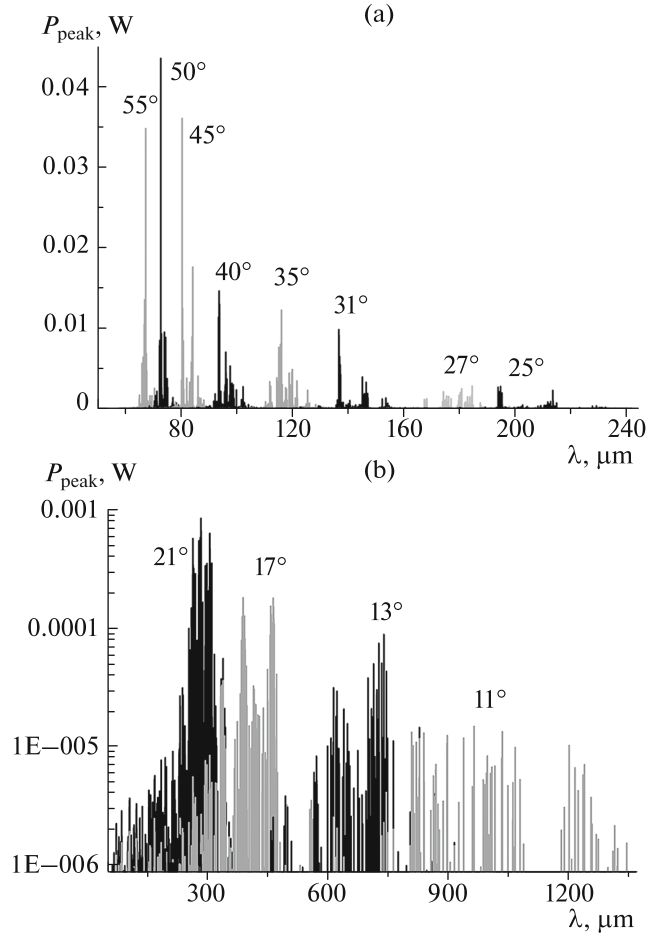

Fig. 2. Calculated spectra of a CO laser's difference frequency radiation in a $ZnGeP_2$ crystal at different phase-matching angles: (a) 25°–55°; (b) 11°–21° (logarithmic scale).

It is evident from Fig. 2 that the power of the DFG radiation lines fell sharply upon tuning to the long-wave region of the spectrum. This was due to two factors: a reduction of the ratio between the energy of a quantum of frequency converted light and the pumping radiation (a drop in the Manley–Rowe relation), and a reduction of the effective nonlinearity factor upon decreasing PM angle from 55° to 0° (see Fig. 1b).

The dependence of the power of DFG radiation (integral along the spectrum) on the PM angle is presented in Fig. 3.

The maximum power of DFG radiation (integral along the spectrum) in the THz band is observed at PM angles in the range of 35°–60°, which agrees with the calculation results presented in Fig. 2. The maximum of the dependence has a peaked structure because of the discreteness of the pumping radiation spectrum. The maximum DFG peak power of 0.23 W corresponds to a peak power efficiency of 6×10^{-5}.

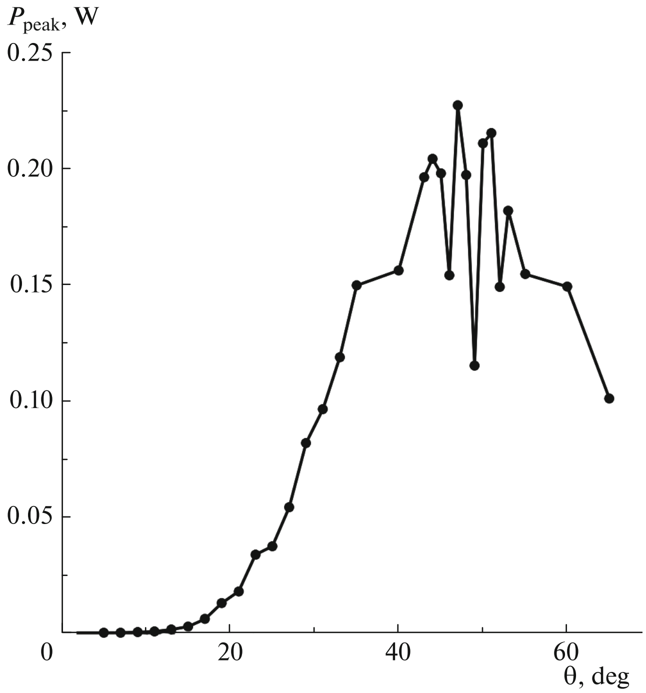

Fig. 3. Dependence of the DFG radiation power (integral along the spectrum) on the PM angle in a ZGP crystal.

Our calculations of power and efficiency were performed using the experimental conditions of [18]. The radiation intensity was in this case ≈13 MW cm^{-2}, which was lower than the optical damage threshold of a ZGP crystal by a factor of 6 (78 MW cm^{-2} [16]). The efficiency of conversion and the power of the difference frequency radiation will therefore be higher by factors of 6 and 36, respectively, when using a more powerful CO laser for pumping a ZGP crystal. Note that the efficiency of conversion can be even higher when using high-power nanosecond pulses of a CO laser (see, e.g., [14]).

Negative factors affecting the efficiency of the considered laser system should also be noted. The investigation performed in [19] showed that inside a pulse of a Q-switched multiline CO laser, the dynamics of its spectral components differ. However, the considerable time shift at the beginning and end of generation is characteristic only of faint spectral lines, which ultimately results in a small (by ~15–20%) reduction in the efficiency of conversion (integral along the spectrum). Note too that since crystal pumping is done with multiwavelength radiation, only some of the frequency components participate efficiently in the generation of difference frequencies at a fixed angle of the crystal. The efficiency of conversion can in this case be improved by concentrating the power of CO laser radiation at predetermined wavelengths in the quasi-selective mode of CO laser operation [14].

CONCLUSIONS

Frequency conversion of multiline CO laser radiation to the THz-band by difference frequency generation in a $ZnGeP_2$ crystal was simulated numerically. It was shown that collinear phase matching is possible for two types of frequency conversion: I (*ooe*) and II (*eoe*). However, Type II frequency conversion is of greater interest because of the higher coefficient of effective second-order nonlinear susceptibility. Numerical experiments on difference frequency generation of multiline CO laser radiation in a $ZnGeP_2$ crystal were also performed for Type II conversion. The possibility of tuning of the difference frequency spectrum to the THz area of the spectrum in the wavelength interval of 70 μm (~4.3 THz) to 1300 μm (~0.23 THz) was demonstrated by phase-matching angle tuning from 11° to 55°. The maximum peak power of the difference frequencies (integral along the spectrum) was 0.23 W at a near 45° phase-matching angle, which corresponds to the efficiency of conversion at a peak power of 6×10^{-5}. The conversion efficiency and power of difference frequency radiation can be improved considerably by scaling the power of nanosecond pulses of a quasi-selective CO laser.

ACKNOWLEDGMENTS

This work was supported by the Russian Foundation for Basic Research, project no. 18-52-16019.

REFERENCES

1. Huang, Z.-M., Huang, J.-G., Zhou, W., et al., *Proc. 42nd Int. Conf. on Infrared, Millimeter, and Terahertz Waves*, Cancun, 2017. https://doi.org/10.1109/IRMMW-THz.2017.8067110.
2. Ding, Y.J. and Shi, W., *Laser Phys.*, 2006, vol. 16, p. 562.
3. Stepanov, A.G., Henin, S., Petit, Y., et al., *Appl. Phys. B*, 2010, vol. 101, p. 11.
4. Hoffmann, M.C. and Fülöp, J.A., *J. Phys. D*, 2010, vol. 44, p. 083001.
5. Kim, G.-R., Jeon, T.-I., and Grishkowsky, D., *Opt. Express*, 2017, vol. 25, p. 25422.
6. Zernike, F. and Midwinter, J.E., *Applied Nonlinear Optics*, New York: Wiley, 1973.
7. Schunemann, P., *Laser Focus World*, 1999, vol. 35, p. 85.
8. Tochitsky, S.Ya., Sung, C., Trubnick, S.E., et al., *J. Opt. Soc. Am. B*, 2007, vol. 24, p. 2509.
9. Xie, J.-J., Guo, J., Zhang, L.-M., et al., *Fundam. Probl. Sovrem. Materialoved.*, 2012, vol. 9, p. 486.
10. Shi, W. and Ding, Yu.J., *Appl. Phys. Lett.*, 2003, vol. 83, p. 848.
11. Ding, Y.J., *IEEE J. Sel. Top. Quantum Electron.*, 2007, vol. 13, p. 705.

12. Andreev, Yu.M., Verozubova, G.A., Gribenyukov, A.I., and Korotkova, V.V., *J. Korean Phys. Soc.*, 1998, vol. 33, p. 356.

13. Andreev, Yu.M., Geiko, P.P., Baranov, V.Yu., et al., *Sov. J. Quantum Electron.*, 1987, vol. 17, p. 1435.

14. Andreev, Yu.M., Budilova, O.V., Ionin, A.A., et al., *Opt. Lett.*, 2015, vol. 40, p. 2997.

15. Lai-ming, Zh., Ji-Jiang, X., Jin, G., et al., *Opt. Precis. Eng.*, 2012, vol. 20, p. 277.

16. Dmitriev, V.G., Gurzadyan, G.G., and Nikogosyan, D.N., *Handbook of Nonlinear Optical Crystals*, Berlin: Springer, 1999, 3rd ed.

17. Kumbhakar, P., Kobayashi, T., and Bhar, G.C., *Appl. Opt.*, 2004, vol. 43, p. 3324.

18. Andreev, Yu.M., Ionin, A.A., Kinyaevskiy, I.O., et al., *Quantum Electron.*, 2013, vol. 43, p. 139.

19. Ionin, A.A., Kinyaevskiy, I.O., Klimachev, Yu.M., et al., *Appl. Phys. B*, 2017, vol. 123, p. 234.

Translated by G. Levina