# Estimation of Possibility to Create a Parametric Generator of THz Radiation on Base of $ZnGeP_2$ Crystals Modified by Fast E-Beam[1]

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**Abstract – The possibility of singly-resonant parametric generation in the submillimeter wavelength range is analyzed for a $ZnGeP_2$ crystal synchronously pumped by a 100-ps pulse train of $CO_2$ laser second harmonic. Calculations were performed for the $ZnGeP_2$ crystals with different post-growth treatment: after thermal annealing as well as after fast e-beam irradiation.**

**Calculated results showed that by using the second harmonic of a $CO_2$ laser with energy density 1.8 J/cm$^2$ submillimeter radiation peak power values from 0.3 to 8.5 MW in the range of 0.94–3.3 THz (320–90 μm) are achievable. The calculated radiation peak power values greatly exceed known experimental data achieved by other nonlinear optical methods.**

## 1. Introduction

Generation of coherent radiation in the submillimeter (SMM) range using high intensity laser pulses is a practical goal of nonlinear optics. The highest SMM radiation peak power achieved so far was obtained by difference frequency generation (DFG) of near-IR radiation in GaSe and $ZnGeP_2$ nonlinear crystals [1]. DFG scheme using a GaSe crystal permitted tuning of SMM radiation in the range of $\lambda = 66.5 \div 5664$ μm to produce a maximum peak power of 389 W. SMM radiation tuning in the range of $83.1 \div 1642$ μm produced a maximum peak power 134 W from a $ZnGeP_2$ crystal.

Numerical simulation of DFG in a $ZnGeP_2$ crystal pumped by 10-μm radiation has shown the possibility of achieving much higher powers (up to 1 GW) of $\lambda = 800$ μm radiation using a 2.5-ps pump pulse intensity of 550 MW/cm$^2$ [2].

In the present article, a new scheme is considered: it is proposed to produce the SMM radiation by parametric generation (PG) in a $ZnGeP_2$ crystal pumped by a train of high power 100-ps laser pulses. Two cases of crystal pumping by $CO_2$ laser second harmonic (SH) at $\lambda = 5.14$ μm are analyzed: a) the crystal after thermal annealing, b) the crystal after fast e-beam irradiation. The second case supposes the reduced both free carrier concentration and SMM absorption coefficient.

The SH generation efficiency for short pulses of $CO_2$ laser is high enough (up to 50% according to [3]) moreover absorption coefficient of 5-μm radiation in this crystal is more than an order of magnitude [2, 4] less compared with values for 10-μm radiation.

## 2. Estimate of maximum allowable pump energy density for nonlinear crystal

The absence of experimental data about maximum pump values for nonlinear crystal $ZnGeP_2$ pumped by 100-ps radiation pulses in the 5–10 μm range required us to make corresponding estimates. These estimates were carried out using the data from two independent sources. Linear regression of the $ZnGeP_2$ damage threshold data obtained for $CO_2$ laser radiation pulses with varied duration [5] gives the following expression for the maximum permissible intensity $I_{thr}$ (W/cm$^2$) as a function of pulse duration τ (s):

$$I_{thr} = 10^{3.82} \cdot \tau^{-0.59} . \quad (1)$$

This approximation includes the data for the pulse duration range of $\tau = 2 \cdot 10^{-9}$-1s. Extrapolation of equation (1) for $\tau = 100$ ps gives $I_{thr} = 5.25 \cdot 10^9$ W/cm$^2$, corresponding to a laser pulse energy density $E_{thr} = 0.53$ J/cm$^2$.

At the same time it is known [6] that $ZnGeP_2$ laser damage results in the destruction of the surface layer only, with micro-defects and Ge-enriched inclusions of metal-type conductivity playing the main role in the optical breakdown process. This is due to enhanced material evaporation near defects with an increased optical absorption coefficient and consequently higher local temperature. The development of optical damage in the $ZnGeP_2$ is very similar to the process occurring on polished metal surfaces, where micro-defects are a major cause of laser induced breakdown. The threshold intensity for plasma formation at the surface of polished metal mirrors in the range of $\lambda = 2.9$–$10.6$ μm is inversely proportional to λ according to [7]. If we assume that this dependence is applicable for $ZnGeP_2$ and use the value of $I_{thr}$ determined for 110-ps laser pulse at $\lambda = 2.94$ μm in reference [4], then one can estimate the value of the threshold energy density at $\lambda = 10$ μm as $E_{thr} = I_{thr}\tau(2.94/10.27) = 0.94$ J/cm$^2$.

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Thus, we propose that the nonlinear crystal damage threshold for a 100-ps 10.27-μm radiation will be in the range of 0.5–0.9 J/cm$^2$. For a pulsed pump of the same duration and $\lambda = 5.14$ μm, the similar estimate gives the range 1.1–1.9 J/cm$^2$.

Considering the case of crystal interaction with a sequence of pulses, we must take into account a temperature decay time constant ($\tau_r$) for the absorbing centers located in the layer adjacent to crystal surface, since the thermal explosion of such centers leads to material damage [8]. In view of the absence of experimental data on $\tau_r$ for $ZnGeP_2$, it seems reasonable to consider the most unfavorable situation, when temperature relaxation between pulses will be negligible. In such a case we can consider that the interaction of a pulse train with crystal to be equivalent to the interaction of a single pulse, with energy equal to the pulse train energy. Then, for further calculations with a pulse train, we can use the values of train energy density not exceeding the upper boundary of the ranges estimated above.

## 3. The calculation model

Calculations were performed for the pump parameters corresponding to radiation of the "Picasso-2" laser system [9]. In particular, the central part of laser radiation spot with energy ~ 2 J, selected by the diaphragm with certain cross-section $S$, provides a homogeneous spatial distribution of energy density across the beam aperture. In experiment, a keeping of radiation plane wave front and selected energy density can be realized by optical telescopes and calibrated attenuators. In calculations, energy density was 1.8 J/cm$^2$ for 5-μm pump ($S = 0.56$ cm$^2$).

The temporal radiation structure in calculations corresponded to the experimental one: the train of 15 pulses separated by 9.3 ns while each pulse of the train has a flat top with duration $\tau = 100$ ps and a steep rise and fall (1–2 ps). Such temporal shape allowed us to use a rectangular approximation for each pump pulse in our calculations.

The pump radiation parameters also allowed us to use some simplifications for calculations of PG, assuming interaction between the pump wave $\lambda_3$ and parametric waves $\lambda_1$, $\lambda_2$. If the nonlinear crystal length satisfies the inequality $L < L_{qs}$ (where the quasi-static length $L_{qs} = \tau/\nu_{31}$, and the group mismatch $\nu_{31} = 1/u_3 - 1/u_1$ for group velocities $u_1$, $u_3$, while group mismatch $\nu_{32} < 10^{-13}$ s/cm can be neglected due to proximity of $\lambda_3$ and $\lambda_2$) then we can use a quasi-static approximation for PG process simulation [10]. The calculated dependence of length $L_{qs}$ on wavelength $\lambda_1$ is showed in Fig. 1. As it seen the quasi-static approximation is valid for relevant SMM range if $L < 6$–13 cm.

The use of a ring cavity for $\lambda_1$ wave (Fig. 2) having a round trip time equal to pump train interval reduced on time of group delay permits us to neglect the accumulating delay of pulse $\lambda_1$ with respect to pulse $\lambda_3$ and to increase accordingly, the total interaction length for successive round trips through the cavity.

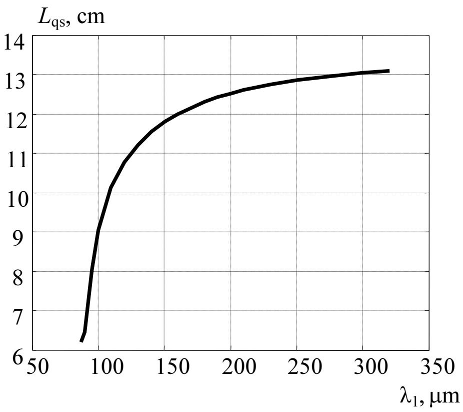


Fig. 1. Dependence of quasi-static length $L_{qs}$ for SMM wave $\lambda_1$ in nonlinear crystal $ZnGeP_2$ pumped by radiation of wavelength $\lambda_3 = 5.14$ μm

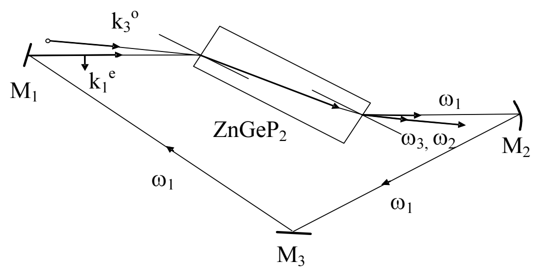


Fig. 2. Optical scheme for a parametric generator with nonlinear crystal $ZnGeP_2$ and ring cavity ($M_1$, $M_2$, $M_3$ are turning mirrors)

To decrease SMM wave diffraction losses, one of the cavity mirrors must have a determined radius of curvature which will provide compensation of the SMM wave divergence (estimated diffraction divergence angle is 20 mrad for $\lambda_1 = 100$ μm). Spatial separation of pump and SMM beams can be achieved by combination of the selected incidence beam angle on the crystal surface and the difference in refraction coefficients.

At the first pass through the crystal, the parametric gain will be high only for those components of the thermal noise at SMM and IR wavelengths with frequencies $\omega_1$ and $\omega_2 = \omega_3 - \omega_1$, for which wave vectors correspond to phase-matching conditions. In subsequent passes, the IR wave will be automatically slaved to the phase matching. Using known approximations of refraction index of $ZnGeP_2$ in IR and SMM ranges [4, 11] the angular tuning curves of PG were calculated for the relevant values of pump wavelength $\lambda_3 = 5.14$ μm (Fig. 3).

The calculations showed that phase matching of PG can take place for the collinear scheme: $k_1^e + k_2^o = k_3^o$, $\omega_1 + \omega_2 = \omega_3$. For the 5-μm pump values $\theta s = 40°$–$19°$ apply in the range $\lambda_1 = 90$–320 μm. An effective nonlinearity $d_{eff}^{(eo-o)} = d_{36}\sin\theta_s\sin 2\phi$ (where $d_{36}$ is the nonlinear coefficient of the crystal, $\theta_s$ is the phase-matching angle, $\phi$ is the azimuthal angle of axes orientation). We used a value of $d_{36}$ = 75 pm/V in our calculations [4], which does not account for the contribution of ionic and ionic-electronic polarizabilities since phonon resonances in $ZnGeP_2$ are 7 $cm^{-1}$ away from the investigated SMM region [11].

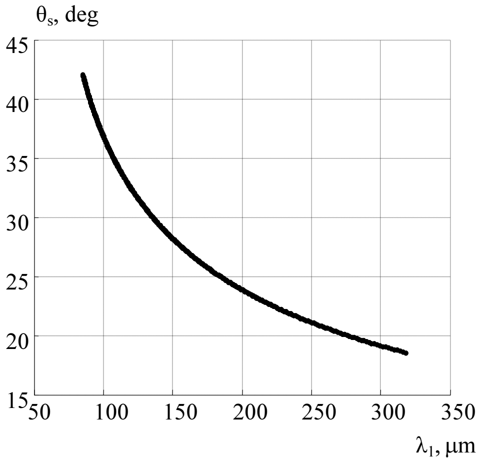


Fig. 3. The dependence of PG phase-matching angle $\theta_s$ in $ZnGeP_2$ on wavelength $\lambda_1$ for pump radiation wavelength $\lambda_3$ =5.14 μm

The interaction of three plane waves in $ZnGeP_2$ nonlinear crystal was considered on the basis of a singly-resonant PG model. The system of four truncated differential equations (2) for real parts of electric field amplitudes $A_i$ and generalized phase $\psi$ were used for the description of wave interaction for a single pass through the nonlinear crystal [10]:

$$
\begin{aligned}
&dA_1/dz + 0.5\cdot\alpha_1\cdot A_1 = \sigma_1\cdot A_2\cdot A_3\cdot\sin\psi,\\
&dA_2/dz + 0.5\cdot\alpha_2\cdot A_2 = \sigma_2\cdot A_1\cdot A_3\cdot\sin\psi,\\
&dA_3/dz + 0.5\cdot\alpha_3\cdot A_3 = -\sigma_3\cdot A_j\cdot A_s\cdot\sin\psi, \qquad (2)\\
&d\psi/dz = \Delta k + (\sigma_3\cdot A_1\cdot A_2/A_3 - \sigma_1\cdot A_2\cdot A_3/A_1 -\\
&\qquad -\sigma_2\cdot A_1\cdot A_3/A_2)\cdot\cos\psi,
\end{aligned}
$$

where $\sigma_i = 8\pi^2 d_{eff}/n_i\lambda_i$ is the nonlinear interaction coefficients; $n_i$, $\alpha_i$ are the refraction and absorption coefficients, and $i = 1, 2, 3$. The generalized phase $\psi = \varphi_3 - \varphi_1 - \varphi_2 - \Delta k{*}z$. According to phase-matching conditions it was assumed that $\Delta k = k_3^o - k_1^e - k_2^o = 0$. Argument $z$ varied from 0 to a fixed value $L$, which was chosen in a range of 0.3–6 cm.

Samples of $ZnGeP_2$ grown at IMCES SB RAS usually have absorption $\alpha_3^o \leq 0.02$ $cm^{-1}$ in the 3–8 μm range, so the value 0.02 $cm^{-1}$ was used in the calculations for the 5-μm pump. The work [11] has shown that the values of absorption coefficient range from $1.84 > \alpha_1^e > 0.62$ $cm^{-1}$ over the wavelength range 90–320 μm. Fig. 4 shows both absorption coefficient spectrum (1) for annealed $ZnGeP_2$ crystal [11] and (2) calculated in absence of free carriers absorption. The second case is realized under fast e-beam irradiation of the crystal according [12, 13].

Besides linear absorption, we included reflection losses at an inclined incidence of radiation on non-AR-coated optical surfaces of the nonlinear crystal in the total losses for the three waves. In particular, for all interacting waves, the single-surface transmission coefficients were ~ 0.7. The practical crystal parameters (aperture, length, absorption) chosen represent "state-of-the art" technology[1].

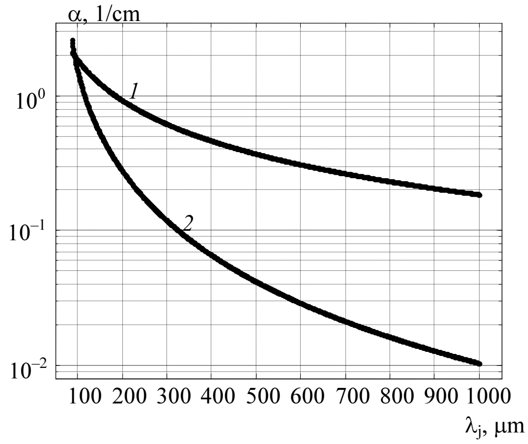


Fig. 4. The absorption coefficient spectrum of $ZnGeP_2$ crystal after thermal annealing (*1*) and calculated without free carrier losses (*2*)

Integration of equation system (2) was carried out by the Runge–Kutta numerical method in series for each of the pump pulses which pass through the crystal synchronously with amplified SMM radiation. Initial values of amplitudes, both $P_{01}$ for SMM and $P_{02}$ for IR wave, were determined according to the Plank formula for thermal radiation. The values are within intervals: $P_{01} = 5\cdot10^{-6}$–$2.8\cdot10^{-7}$ W, $P_{02} = 1.1\cdot10^{-6}$–$2.5\cdot10^{-6}$ W for the relevant ranges of $\lambda_1$ and $\lambda_2$ respectively.

For subsequent passes through the crystal the initial amplitude of the SMM wave was determined by the value calculated at the exit of the crystal after the previous pass. It was assumed that the initial generalized phase for interacting waves has the optimum value $\psi_o = \pi/2$ as the next pump pulse enters the crystal, i.e., at the beginning of each calculation cycle.

The main output parameter of the calculations was the maximum peak power $P_1$ of SMM radiation at the exit from nonlinear crystal after the $N$ passes required to reach this maximum. The value $N$ depends on crystal length, pump intensity and wavelength $\lambda_1$. $N$ is 10–13 for the laser pulse train used.

The computation showed that for $\lambda_3$ = 5.14 μm, high pump intensities and a long crystal length $L$, after

[1] The original technology for experimental manufacture of high optical quality $ZnGeP_2$ single crystals with dimensions up to 3 cm in diameter and up to 12 cm long was developed at IMCES SB RAS (Tomsk) and is now in operation.

certain number $N$ of crystal passes determined by $\lambda_1$, the variation of the interacting waves' power, $P$i along the propagation direction in the crystal developed as oscillation. It was showed in [14], that such phenomena can occur during the interaction of three constrained waves in a nonlinear dielectric. To exclude the PG development in this way and also to obtain the maximum value of $P_1$ at the nonlinear crystal exit, we had to choose the $L$ value depending on value of $\lambda_1$ by using an additional condition: a decrease of the pump power $P_3$ inside the crystal due to nonlinear interaction with parametric waves must be less than 20% of it is initial value.

## 4. Calculation results

The calculations for the case of $CO_2$ laser radiation SH pumping the nonlinear crystal at energy density 1.8 J/cm$^2$ and aperture S = 0.56 cm$^2$ showed that for the ordinary $ZnGeP_2$ crystal the $P_1$ value can reach $0.3–8.5 \cdot 10^6$ W in the range $\lambda_1$ = 87–320 μm at crystal lengths from 1.6 to 4.3 cm. For the $ZnGeP_2$ crystal without free carrier's losses the $P_1$ value can reach $3 \cdot 10^5–8.5 \cdot 10^6$ W in the same range at identical lengths (Fig. 5). As it was indicated for each SMM wave the individual optimum crystal length takes place. The optimal length $L$ for the second crystal was used in the calculations and one is shown on Fig. 6. The data obtained enable us to optimize the nonlinear crystal length for a particular SMM wavelength.

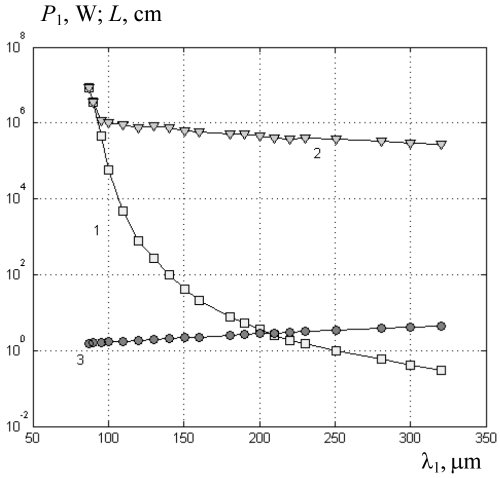


Fig. 5. The dependencies of maximum peak power $P_1$ for a 5.14-μm pump with an energy density of 1.8 J/cm$^2$ with the $ZnGeP_2$ crystal of ordinary type (1) and without free carrier losses (2). Crystal length $L$ (3) was optimized for case (2)

We propose to extract the SMM pulse from the ring cavity by means of stimulated reflection from a semiconductor plate installed inside the resonator at the Brewster angle. A SMM reflection coefficient of 60%, induced in a silicon plate by laser irradiation, was realized in [15]. Thus, the proposed PG scheme should convert the 5-μm pulse train into a single SMM radiation pulse with peak power from 0.3 to 8.5 MW, depending on its wavelength.

## 5. Conclusions

Our computations of singly-resonant PG within nonlinear crystal of $ZnGeP_2$ pumped by a train of high power 100-ps radiation pulses from a $CO_2$ laser SH lead to the following conclusions:

To get high peak power (0.3–8.5 MW) radiation in the SMM range of 0.94–3.3 THz (320–90 μm) one can use as a pump, the SH of $CO_2$ laser with energy density up to 1.8 J/cm$^2$ in a nonlinear crystal with the length ranging from 1.6 to 4.3 cm depending on the SMM wavelength. The $ZnGeP_2$ crystal has not to have free carriers losses i.e., it is necessary to use the crystal after fast e-beam irradiation.

The predicted values of SMM radiation peak power far exceed the values currently obtained experimentally by DFG using near-IR lasers.

## References

[1] Y.J. Ding and W. Shi, Laser Physics **16**, 562–570 (2006).

[2] V.V. Apollonov, Yu.A. Shakir, and A.I. Gribenyukov. J. Phys. D: Appl. Phys. **35**, 1477–1480 (2002).

[3] Yu.M. Andreev, V.Yu. Baranov, V.G. Voevodin et al., Kvant. Elektron. 14, 2252–2254 (1987).

[4] V.G. Dmitriev, G.G. Gurzadyan, and D.N. Nikogosyan, *Handbook of Nonlinear Optical Crystals*, Berlin, Springer-Verlag, 1997.

[5] J.H. Churnside, Yu.M. Andreev, A.I. Gribenyukov et al., *NOAA Technical Memorandum ERL WPL-224 WPL*, Boulder, Co., USA, 1992, p. 1–18.

[6] Yu.M. Andreev, V.G. Voevodin., A.P. Vyatkin et al., *Nonlinear optical crystals A2B4C52 for IR laser radiation conversion*, Tomsk, Rasko, 1992, pp. 46–100.

[7] A.V. Bessarab, V.I. Novik, L.V. Pavlov et al., Zh. Tekh. Fiz. **59**, 886–888 (1980).

[8] M.F. Koldunov, A.A. Manenkov, and I.L. Pokotilo. Izv. Akad. Nauk, Fiz. **59**, 72–82 (1995).

[9] V.V. Apollonov, N.V. Pletnyev, V.R. Sorochenko et al., Proc. SPIE **5120**, 291–296 (2003).

[10] V.G. Dmitriev and L.V. Tarasov. *Applied nonlinear optics*, Moscow, Fizmatlit, 2004, p. 290.

[11] V.V. Voitsekhovsky, A.A. Volkov, G.A. Komandin, and Yu.A. Shakir, Fizika Tverdogo Tela **37**, 2199–2202 (1995).

[12] V.N. Brudnyi, V.G. Voevodin, and S.N. Grinyaev, Fizika Tverdogo Tela **48**, 1949–1961 (2006).

[13] G. Verozubova, A.I. Gribenyukov, A.Yu. Trofimov et al., *in Proc. of Symp. Progress in Semiconductors II – Electronic and Optoelectronic Applications* **744**, Mat. Res. Soc., Boston, MA, 2003, p.315–320.

[14] J.A. Armstrong, N. Bloembergen, J. Ducuing et al., Phys. Rev. **127**, 1918–1939 (1962).

[15] M.F. Doty, B.E. Cole, B.T. King, and M.S. Shervin, Rev. Sci. Instrum. **75**, 2921–2925 (2004).