Generation of 8 µm radiation in a ZnGeP2 single crystal at a difference frequency, with pumping by telecommunication-band radiation of 1.5 µm and radiation at a wavelength of 1.908 µm

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Abstract. This study theoretically investigates difference frequency generation in a $ZnGeP_2$ crystal pumped by a fiber laser (1.53 µm) and a Tm:YLF laser (1.908 µm) for generating mid-infrared radiation. The theoretical model incorporates phase matching conditions, nonlinear susceptibility, crystal length, and beam parameters (sum of the average power of the pump wave and signal wave and the pulse duratio)to estimate the generated pulse and average power. The impact of absorption and pulse duration on the output power is analyzed. Results indicate that increasing pump power and decreasing pulse duration enhance the average output power. The study provides a theoretical framework for designing efficient difference frequency generation-based mid-infrared sources.

1 Introduction

Sources of coherent radiation capable of generating powerful pulsed radiation in the wavelength range of 3-8 µm are crucial for developing lidar systems to monitor greenhouse gas emissions. [1] Beyond the need for high-power coherent sources in the spectral regions where greenhouse gases absorb light, there's a pressing demand for efficient sources generating radiation in atmospheric transparency windows to create differential absorption lidar systems, especially around 8 µm.

Among the vast array of nonlinear optical materials, zinc germanium phosphide ($ZnGeP_2$) single crystals stand out as one of the most effective for generating radiation in the 3-8 µm wavelength range through nonlinear processes [2,3]. $ZnGeP_2$ single crystals exhibit an absorption coefficient of <0.02 cm^{-1} in the spectral range of 2-8.3 µm [4-9]. The damage threshold of $ZnGeP_2$ at wavelengths potentially accessible by pump lasers (2.05 µm) with a pulse duration of 10 ns and a repetition rate of 10 kHz is approximately 0.074 GW/cm^2 [10]. This crystal has a high nonlinear susceptibility coefficient (d = 70-85.4

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pm/V) and high thermal conductivity (0.35 W/(cm.K)), which is particularly important for achieving high average and peak radiation power.

The most powerful coherent radiation sources in the 3-8 μm wavelength range are currently based on $ZnGeP_2$ (ZGP) nonlinear optical crystals [11-13]. These sources can generate radiation with average power up to 160 W or pulse energy up to 200 mJ with pulse durations of several tens of nanoseconds and repetition rates from a few hertz to hundreds of kilohertz [11-14]. These data indicate the promise of using $ZnGeP_2$ crystals as a nonlinear medium for direct conversion of radiation from the telecommunication band (~1.5 μm) to the mid-infrared band (~8 μm) through difference frequency generation using auxiliary coherent sources in the 1.9 μm range.

This research is dedicated to theoretical calculations of the possibility of creating a difference frequency generator based on $ZnGeP_2$ generating radiation at a wavelength of ~8 μm when pumped by radiation from 1.5 μm fiber lasers in conjunction with radiation from a 1.908 μm Tm:YLF laser.

2 Theoretical estimates and discussion

The general operating principle of a difference frequency generator involves the transfer of energy from powerful light waves - a pump wave (ω_p) and a signal wave (ω_s) - to a weak electromagnetic wave at a frequency ω_i (idler wave), satisfying the relationship $\omega_p = \omega_s + \omega_i$.

The physical cause of wave mixing is the quadratic nonlinearity of the medium's polarization, which manifests at high pump levels ($P = æ^E + \chi^{(2)E2} + ...$, where æ is the linear dielectric susceptibility, χ is the quadratic nonlinear susceptibility). Amplification of the idler wave occurs when the phase matching condition $k_p = k_s + k_i$ is met (where k_p, k_s, k_i are the wave vectors).
The phase synchronism condition can also be written as

$$\frac{n_p(\lambda_p)}{\lambda_p} - \frac{n_s(\lambda_s)}{\lambda_s} = \frac{n_i(\lambda_i)}{\lambda_i}, \quad (1)$$

where n_p, n_s, n_i are the refractive indices of the ordinary $n_o(\lambda_p)$, extraordinary $n_e(\lambda_s)$, and idler $n_i(\lambda_i)$ waves, respectively. Phase matching is most easily achieved in birefringent crystals, where phase synchronism is achieved due to the natural dispersion of the refractive indices of the ordinary $n_o(\omega)$ and extraordinary $n_e(\omega)$ waves. In accordance with the phase matching condition, it is possible to change the frequency of the amplified waveby adjusting the propagation direction of the pump and signal waves relative to the optical axis of the crystal, which in its turn can be changed by modifying the angle (θ) between the propagation direction and the optical axis of the crystal, and by modifying the wavelength of the pump and signal waves. The dependence of the refractive index of the extraordinary wave on the angle θ is determined by the following relation [14]

$$n_e(\lambda,\theta) = n_0(\lambda)\sqrt{\frac{1+tan^2(\theta)}{1+\left(\frac{n_0(\lambda)}{n_e(\lambda)}\right)^2 tan^2(\theta)}} \quad (2)$$

Table 1 presents possible wavelengths (λ_3) of radiation that can be obtained by difference frequency generation using radiation from a fiber laser at wavelengths ~ 1.53 μm and a Tm:YLF laser (1.908 μm) for various combinations of generation lines (λ_1, λ_2). Using formulas (1) and (2), the synchronism angle was calculated for difference frequency generation in a $ZnGeP_2$ single crystal for various combinations of pump wavelengths using Sellmeier's equations (3), (4) for the mid-IR wavelength range [15].

$$n_o^2(\lambda) = 4.47330 + \frac{5.26576\lambda^2}{\lambda^2 - 0.13381} + \frac{1.49085\lambda^2}{\lambda^2 - 662.55} \quad (3)$$

$$n_e^2(\lambda) = 4.63318 + \frac{5.34215\lambda^2}{\lambda^2 - 0.14255} + \frac{1.45795\lambda^2}{\lambda^2 - 662.55} \quad (4)$$

Table 1. Possible wavelengths (λ3) of radiation can be generated by difference frequency generation. λ1isthewavelength of pump wave, λ2 is the wavelength of signal wave,θ is the angle between the propagation direction and the optical axis of the crystal,φ is the azimuthal anglethe propagation direction and the optical axis of the crystal.

Type of synchronism	λ_1, μm	λ_2, μm	λ_3, μm	θ,˚	φ,˚
o-e-o	1,51	1,908	7,2	83	45
o-e-o	1,52	1,908	7.5	79	45
o-e-o	1,53	1,908	7,7	76	45
o-e-o	1,54	1,908	7,9	73	45
o-e-o	1,55	1,908	8,3	71	45
o-e-o	1,56	1,908	8,5	69	45

The pulse power of the generated radiation, taking into account absorption in the material, deviation from exact synchronism, and the presence of anti-reflection coatings on the working faces [15], is calculated using the formula (5) [17].

$$P_g = \frac{1}{2}\left(\frac{\mu_0}{\varepsilon_0}\right)^{\frac{1}{2}} \frac{4\pi^2\left(2d_{eff}\right)^2 L^2}{n_p n_s n_i \lambda_i^2}\left(\frac{P_p P_s}{\pi r^2}\right) T_p T_s T_i e^{-\alpha_i L} * \frac{1 + e^{-\Delta\alpha L} - 2e^{-\frac{1}{2}\Delta\alpha L} * \cos(\Delta k L)}{(\Delta k L)^2 + \left(\frac{1}{2}\Delta\alpha L\right)^2} \quad (5)$$

where μ_0 is the magnetic permeability, ε is the permittivity, χ is the nonlinear coefficient, *l* is the length of the nonlinear crystal, λ is the wavelength of the generated radiation, P_p and P_s are the pulse powers of the pump and signal waves, respectively, r is the beam radius at the interaction point, T_p, T_s, and T_i are the transmittances of the nonlinear crystal at the pump, signal, and idler wavelengths, respectively, $\Delta\alpha = \left|\alpha_p + \alpha_s - \alpha_i\right|$, where α_p, α_s, and α_i are the absorption coefficients of the pump, signal, and idler waves, respectively, and $\Delta k = k_p - k_s - k_i$ is the phase mismatch.

The pulse power of the radiation generated at the difference frequency, in the approximation of slowly varying amplitudes and the given field approximation, without considering the influence of absorption, is determined by the relation (6)

$$P_g = \frac{2\omega_i^2 d_{eff}^2 L^2 P_p P_s}{\varepsilon_0 c^3 n_p n_s n_i} sinc^2\left(\frac{|\Delta k| L}{2}\right), \quad (6)$$

where ω_i is the frequency of the generated radiation, deff is the effective nonlinear susceptibility of the second order, L is the crystal length, P_p and P_s are the pulse powers of the pump radiation and signal wave radiation (calculate by formula (7)), respectively, ε_o is the permittivity of free space, c is the speed of light, n_p, n_s, n_i are the refractive indices for the pump, signal, and idler waves, respectively, and Δk is the momentum mismatch.

$$P_{p,s} = \frac{P_{1,2}}{ft}, \quad (7)$$

where P_1, P_2 are the average powers of the pump and signal waves, respectively, *f* is the pulse repetition rate, and t is the pulse duration. The calculated value of pulse power obtained from formula (4), without considering the influence of absorption and with the crystal length equal to the effective length, differs from the experimentally obtained value by no more than 30% [15]. Therefore, this expression can be used to calculate the power of the radiation generated at the difference frequency. The average power of the generated radiation is calculated using the formula (8)

$$P = P_g f t, \quad (8)$$

where f is the pulse repetition rate, t is the pulse duration. From relations (6), (7), and (8), the expression for the average power of the generated radiation follows(9)

$$P = \frac{2\omega_i^2 d_{eff}^2 L^2 P_1 P_2}{\varepsilon_0 c^3 n_p n_s n_i f t} sinc^2(\frac{|\Delta k|L}{2}) \quad (9)$$

Equation (8) indicates that it is necessary to increase the average power of the pump and signal waves and decrease the duration of their radiation pulses to increase the average power of the THz radiation. At the same time, it is necessary to select the length of the nonlinear element, taking into account absorption at all three interaction wavelengths and beam walk-off, so that the output radiation power reaches a maximum value.

Figures 1 and 2 show the results of calculations, according to formula (9), of the average power of THz radiation generated in a $ZnGeP_2$ single crystal with a length of ~ 10 mm when varying the pulse duration and average pump power (pulse repetition rate ~ 10 kHz). Figure 1 shows the dependence of the average power of THz radiation on the average power of the aforementioned pump lines with a pulse duration of ~ 1 ns. Figure 2 shows the dependence of the average power of the generated radiation on the pulse duration of the pump with an average pump power of ~ 60 W at both wavelengths (λ_1 = 1.55 µm) and the signal wave (λ_2 = 1.908 µm), respectively.

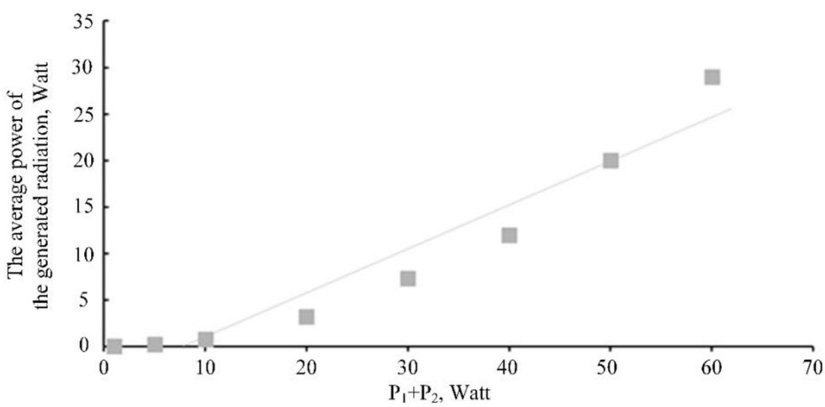

Fig. 1. Dependence of the average power of the generated radiation (λ_3 = 8.3 µm) on the average power of the pump wave (λ_1 = 1.55 µm) and signal wave (λ_2 = 1.908 µm) at a pulse duration of ~ 10 ns

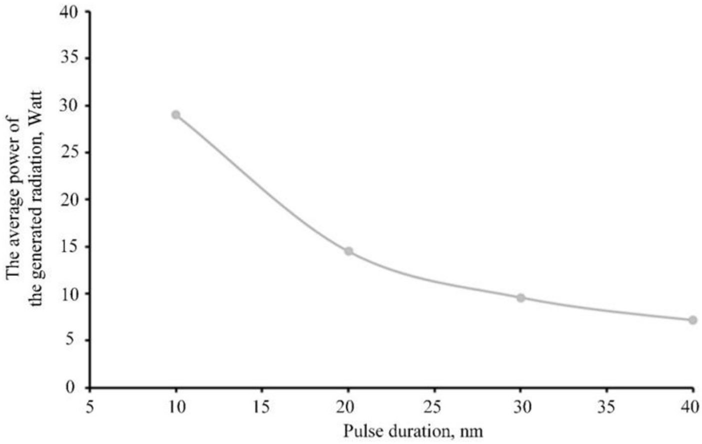

Fig. 2. Dependence of the average power of THz radiation (λ_3 = 8.3 μm) on the duration of the pump wave pulses (λ_1 = 1.55 μm) and the signal wave (λ_2 = 1.908 μm)

3 Conclusion

The analysis above indicates the promising nature of using a difference frequency generator based on a $ZnGeP_2$ crystal, pumped by telecommunication-band lasers with a wavelength of 1.5 μm and a Tm:YLF laser with a wavelength of 1.908 μm, as a radiation source in the atmospheric transparency window in the wavelength range of 8 μm. The proposed technical solutions allow for the realization of an average output power of up to 30 W at a pulse repetition rate of 10 kHz and pulse durations of several nanoseconds. It should be noted that selection of IR radiation at the output of the difference frequency generator can be achieved by changing the angle between the pump radiation direction and the optical axis of the crystal. By rotating the nonlinear crystal, continuous tuning in the wavelength range of 7.2-8.5 μm can be achieved (see Table 1).

4 Acknowledgements

This study was supported by the Ministry of Science and Higher Education of the Russian Federation (budget funds of the Institute of Atmospheric Optics SB RAS)

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