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Parametric amplification of quantum cascade laser radiation at 4.6 μm in a nonlinear $ZnGeP_2$ crystal

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Amplification of pulsed quantum cascade laser radiation at $\sim 4.6\,\mu$m through nonlinear conversion in a $ZnGeP_2$ crystal has been demonstrated experimentally. The peak power at the output of a nonlinear crystal was 373 W at input peak power of the quantum cascade laser of 0.4 W.

Keywords: Quantum cascade laser, Ho:YAG laser, parametric nonlinear conversion.

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One of the atmospheric transparency windows is located in the $4.5-5.2\,\mu$m spectral region. This heightens the interest in development of lasers radiating in this wavelength range. A quantum cascade laser (QCL) is one of the radiation sources of this kind that, owing to its unipolarity and independence of interband transitions, has the capacity to operate in middle and far IR ranges. This makes such lasers competitive in ecological monitoring [1], non-invasive medical diagnostics [2], remote sensing [3], and wireless optical communications [4].

A significant progress in QCL technology resulted in a considerable enhancement of their output power. Specifically, an output power in excess of 10 W has been achieved in the pulsed mode at room temperature at a wavelength of $4.5-4.6\,\mu$m [5]. However, formidable barriers to raising the QCL output power still exist. Lateral widening of the output aperture translates into deterioration of the beam quality due to multimode generation, generally providing no gain in brightness. An increase in the number of cascades in the active region leads to an increase in operating voltage, a reduction in efficiency, the emergence of higher-order transverse modes, and deterioration of the beam quality. If the pump current is raised well above the lasing threshold, the voltage drop across the QCL active region increases inevitably, inducing the misalignment of laser and injection quantum cascade levels and inhibiting the rise of output power.

Several ways to raise the output power of QCL systems have been discussed in literature. A monolithic combination of a master oscillator and an amplifier in a MOPA (master oscillator power amplifier) configuration [6,7], which is commonly used to enhance the power of distributed-feedback QCLs supporting single-frequency lasing, is of particular interest. An array of QCLs synchronized in phase may provide a peak pulse radiation power of several tens of watts [8,9]. Specifically, an array of 100 QCLs has demonstrated lasing with a central wavelength of $4.76\,\mu$m and a peak power of 40 W [8]. The efficiency of an array with a triple-digit number of elements is relatively low compared to the efficiency of a single emitter [5]. This is largely attributable to complications in the removal of heat from a chip at higher powers. In addition, destructive interference, which reduces the overall efficiency of an array, may arise when the radiation of individual elements is combined. However, far-field measurements performed in [8] have confirmed that, impressively, more than 73.3% of radiation power were contained within the central spot even at high pump currents.

Parametric amplification of QCL radiation is a technically simpler way to achieve high radiation powers in the pulsed mode. Specifically, a peak power of 580 W has been reported in [10] for a distributed-feedback QCL that outputs 3 mW in the continuous-wave mode (a 53 dB gain) at a wavelength of $4.5\,\mu$m. A pulsed Ho:YAG laser producing pulses with a width of 30 ns at a frequency of 20 kHz with a wavelength of $2.09\,\mu$m was used as a pump source. A periodically polarized nonlinear OP-GaAs element 41 mm in length was the nonlinear crystal. The results of parametric amplification of QCL radiation in the $8-10\,\mu$m spectral range have also been reported in [11,12]. A maximum gain of 20.5 dB (by a factor of 111) and 26 dB, respectively, was achieved.

In the present study, we report the results of experiments on amplification of QCL radiation with wavelength $\lambda \approx 4.6\,\mu$m via parametric conversion in a nonlinear $ZnGeP_2$ crystal. A quantum cascade laser (amplified radiation) and a Ho:YAG pump laser were used for parametric conversion. Both sources were operated in the pulse-

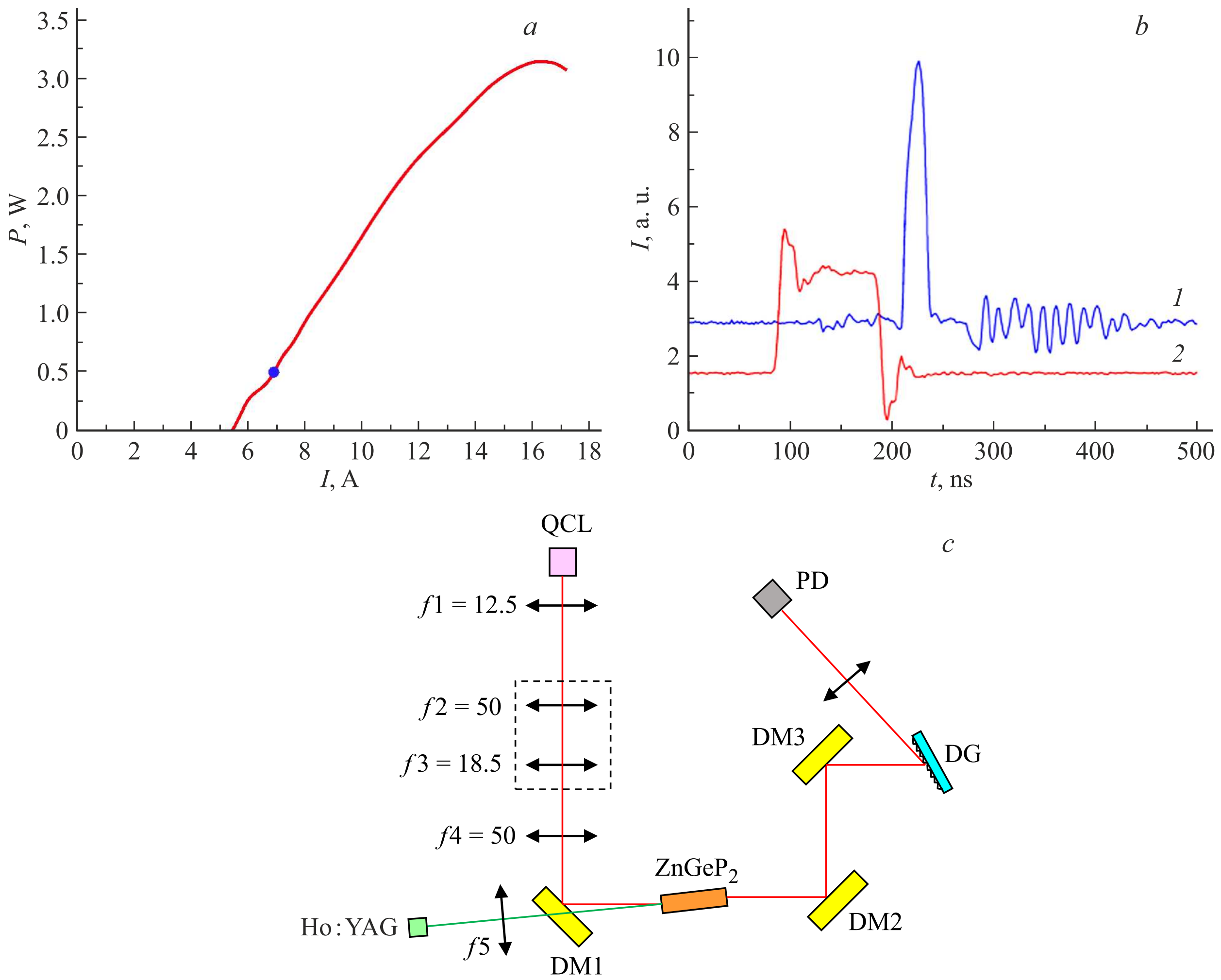

Figure 1. *a* — Watt–ampere characteristic of the QCL. The dot indicates the operating point of this laser. *b* — Oscilloscope record of a QCL radiation pulse (*1*) and a drive generator pulse (*2*). *c* — Diagram of the experimental setup.

periodic mode at a frequency of 10 kHz; thus, temporal synchronization of amplified and pump pulses was required. The use of a nonlinear $ZnGeP_2$ crystal, wherein phase synchronicity is achieved without periodic polarization, enabled the amplification of multimode radiation of a wide-stripe QCL with a Fabry–Pérot cavity with a broad lasing spectrum. It should be noted that we failed to find any other published studies reporting the amplification of QCL radiation with a wavelength of 4.6 μm in a nonlinear $ZnGeP_2$ crystal.

The QCL was operated near the lasing threshold (the QCL pump current was 1.2 times greater than the threshold value), and the radiation pulse power was around 0.5 W (Fig. 1, *a*) for an approximately 20-ns-wide pulse (see the oscilloscope record in Fig. 1, *b*). The QCL bandwidth was approximately 18 nm with a central peak near 4585 nm (Fig. 2, *a*).

A Ho:YAG laser radiating at a wavelength of 2.091 μm ($\Delta\lambda \approx 0.5$ nm), which was the fundamental mode of a stable cavity ($M^2 \sim 1.1$) formed by planar and spherical mirrors, was used for pumping. Its radiation spectrum is shown in Fig. 2, *b*. An acoustooptical modulator inside the cavity was used for Q switching, which enabled the generation of pulse-periodic radiation. The FWHM of a pump laser pulse was approximately 20 ns.

The diagram of the experimental setup is shown in Fig. 1, *c*. The QCL radiation, which had a large angular divergence, was collected into a parallel beam by lens $f1$, narrowed down by telescopic pair $f2{-}f3$, and focused by lens $f4$ to the input facet of the $ZnGeP_2$ crystal. This crystal was $6 \times 6 \times 15$ mm in size. Its optical axis was oriented at angle $\theta = 54.5°$ to the direction of propagation of pump radiation. The principal plane of the $ZnGeP_2$ crystal is parallel to the picture plane. The QCL polarization corresponded to an extraordinary ray. The Ho:YAG laser pump radiation corresponded to an ordinary ray and was fed to the input crystal facet via dichroic mirror DM1, which reflected the QCL radiation with $\lambda \approx 4.6$ μm and transmitted the Ho:YAG laser radiation with a wavelength of 2.091 μm. The needed power density of pump radiation was achieved by

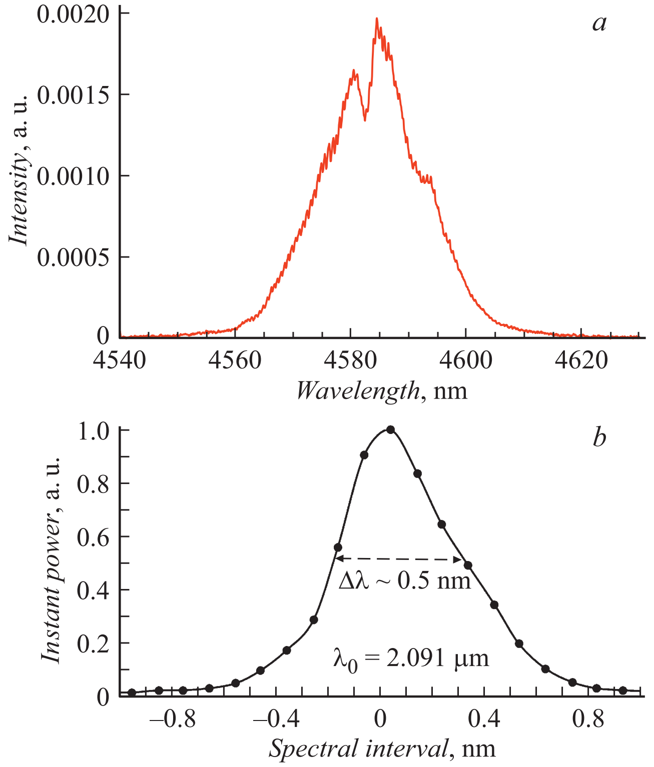

Figure 2. *a* — QCL radiation spectrum; *b* — Ho:YAG laser radiation spectrum.

focusing it at the input crystal facet with the use of lens $f5$. The phase synchronicity was tuned by rotating the crystal in the picture plane. Following conversion in the crystal, the output beam, which had several frequency components, was reflected off two dichroic mirrors DM2 and DM3 in order to separate high-power pump radiation with $\lambda \approx 2.1\,\mu$m from relatively weak radiation with $\lambda \approx 4.6\,\mu$m. The beam was directed to diffraction grating DG, which sent the fraction with $\lambda \approx 4.6\,\mu$m to photodetector PD, to perform additional selection of the QCL wavelength.

The width of an amplified radiation pulse with $\lambda \approx 4.6\,\mu$m was 6 ns. The observed reduction in width (compared to the initial QCL pulse) is attributable to the fact that the temporal profile of an amplified pulse is shaped by both initial and pump pulses. It should be taken into account that the relation between amplified and pump pulses is exponential, since amplification is governed by pumping. This is the reason why the temporal profile of an amplified pulse is narrower than the profiles of both initial QCL and pump Ho:YAG laser radiation pulses.

The mean power of a radiation pulse 6 ns in width after the diffraction grating was 22.4 mW. The corresponds to a peak power of 373 W at a pulse repetition rate of 10 kHz. Since the peak QCL radiation power at the input facet of the nonlinear crystal was 0.4 W, the parametric gain was approximately 30 dB (without regard for the losses at lenses $f1-f4$).

Figure 3 shows the theoretical and experimental time dependences of the instantaneous power of a QCL radiation pulse amplified in the nonlinear $ZnGeP_2$ crystal. The calculation of parametric amplification of radiation was performed in SNLO [13]. We used the function of this software package that solves numerically a system of coupled equations for intensity magnitudes in the process of three-wave interaction. Both crystal parameters (refraction indices and absorption coefficients, slip angles, nonlinearity coefficient, and length) and the characteristics of QCL and pump laser pulses (energies, widths, and beam diameters) served as input data. It follows from Fig. 3 that the theoretical and experimental values of width of an amplified radiation pulse (~ 8.5 and ~ 6 ns, respectively) agree closely. At the same time, the calculated absolute peak power of a radiation pulse is close to 425 W, which is approximately 15% greater than the experimental value. In our view, these discrepancies are attributable to numerous simplifying assumptions made in the theoretical model where pulses of pump and QCL radiation are regarded as Gaussian ones and are assumed to be matching perfectly in time and space.

In view of this, several opportunities for further enhancement of parametric gain may be noted. First, a more accurate temporal alignment of pump pulses and QCL radiation pulses needs to be achieved. The width of both pulses is close to 20 ns. Owing to the specifics of the current design of the Ho:YAG pump laser, pump pulses are generated with an uncertainty on the order of 10 ns, while the stability of operation of the quantum cascade laser is much higher (the QCL pulse jitter is below 1 ns). Consequently, instead of the entire QCL pulse, only a faction of it overlapping with a pump pulse is subjected to nonlinear conversion; this results in losses.

Second, the fraction of QCL energy involved in conversion may be increased by optimizing the system of QCL focusing to a nonlinear crystal. The Ho:YAG pump laser produces a near-Gaussian beam with $M^2 \approx 1.1$. Such parameters allow one to focus pump radiation into a beam with a waist of 0.7 mm that diverges only weakly over the

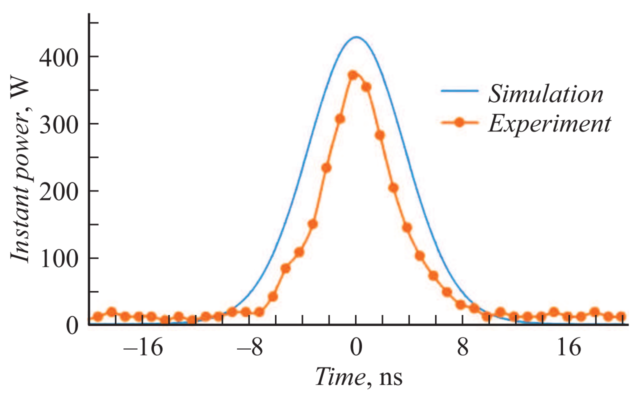

Figure 3. Theoretical and experimental time dependences of the instantaneous power of a QCL radiation pulse amplified in the nonlinear crystal.

nonlinear crystal length (15 mm). The quantum cascade laser is designed so that its divergence is inherently higher (and differs from one axis to the other). In the present study, plane-spherical lenses, which do not allow one to correct the elliptical shape of the QCL beam section, were used to obtain a collimated beam. The initial high divergence of the QCL beam is retained in the course of shaping of a small-diameter waist for coupling with pump radiation in a nonlinear crystal. Consequently, only a fraction of the QCL beam is overlapping and collinear with pump radiation, and this leads to losses. Cylindrical lenses and a more thorough approach to the design of the entire system of QCL radiation input into a nonlinear crystal need to be introduced in order to reduce these losses.

It also appears possible to raise the output power of converted radiation by increasing the QCL radiation power. In the present study, the QCL was supplied from a current source that could be operated only slightly above the QCL lasing threshold. The use of a higher-power source should provide an opportunity to raise the QCL radiation power and, consequently, the power of converted radiation at the crystal output. An increase in pump power will have the same effect on the power at the crystal output, but the radiation power density in the crystal should be kept below the damage threshold.

Thus, experiments on amplification of quantum cascade laser radiation with a wavelength of 4.6 μm via nonlinear conversion in a $ZnGeP_2$ crystal produced the following results: the peak power achieved at the crystal output was 373 W at a gain close to 30 dB. These results open up the prospects for production of a radiation source with a peak power of $\sim$ 1 kW based on parametric amplification of narrowband QCL radiation. Such a source could extend considerably the operating range of gas analyzers and advanced optical communication systems.

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Conflict of interest

The authors declare that they have no conflict of interest.

References

[1] A. Kosterev, G. Wysocki, Y. Bakhirkin, S. So, R. Lewicki, M. Fraser, R.F. Curl, Appl. Phys. B, **90** (2), 165 (2007). DOI: 10.1007/s00340-007-2846-9

[2] A. Schwaighofer, M. Brandstetter, B. Lendl, Chem. Soc. Rev., **46** (19), 5903 (2017). DOI: 10.1039/c7cs00403f

[3] P.Q. Liu, *Mid-infrared quantum cascade lasers with novel active core and laser cavity*, PhD Thesis (Princeton, 2012).

[4] X.Pang, O. Ozolins, L. Zhang, R. Schatz, A. Udalcovs, X. Yu, S. Lourdudoss, Phys. Status Solidi A, **218** (3), 2000407 (2020). DOI: 10.1002/pjtf.202000407

[5] V.V. Dudelev, D.A. Mikhailov, A.V. Babichev, G.M. Savchenko, S.N. Losev, E.A. Kognovitskaya, A.V. Lyutetskii, S.O. Slipchenko, N.A. Pikhtin, A.G. Gladyshev, D.V. Denisov, I.I. Novikov, L.Ya. Karachinsky, V.I. Kuchinskii, A.Yu. Egorov, G.S. Sokolovskii, Quantum Electron., **50** (11), 989 (2020). DOI: 10.1070/QEL17396.

[6] B. Hinkov, M. Beck, E. Gini, J. Faist, Opt. Express, **21** (16), 19180 (2013). DOI: 10.1364/oe.21.019180

[7] M. Bertrand, A. Shlykov, M. Shahmohamadi, M. Beck, S. Willitsch, J. Faist, Photonics, **9** (8), 589 (2022). DOI: 10.3390/photonics9080589

[8] F.-L. Yan, J.-C. Zhang, Z.-Z. Jia, N. Zhuo, S.-Q. Zhai, S.-M. Liu, F.-Q. Liu, Z.-G. Wang, AIP Adv., **6** (3), 035022 (2016). DOI: 10.1063/1.4945383

[9] W. Zhou, D. Wu, Q.Y. Lu, S. Slivken, M. Razeghi, Sci. Rep., **8**, 14866 (2018). DOI: 10.1038/s41598-018-33024-7

[10] G. Bloom, A. Grisard, E. Lallier, C. Larat, M. Carras, X. Marcadet, Opt. Lett., **35** (4), 505 (2010). DOI: 10.1364/ol.35.000505

[11] Q. Clément, J.-M. Melkonian, J. Barrientos-Barria, J.-B. Dherbecourt, M. Raybaut, A. Godard, Opt. Lett., **38** (20), 4046 (2013). DOI: 10.1364/ol.38.004046

[12] F. Gutty, A. Grisard, C. Larat, D. Papillon, M. Schwarz, B. Gérard, R. Ostendorf, J. Wagner, E. Lallier, Adv. Opt. Technol., **6** (2), 95 (2017). DOI: 10.1515/aot-2016-0062

[13] http://www.asphotonics.com/SNLO

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