Multiline CO$_2$ laser with Q-switching for generation of terahertz radiation

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Abstract. In this paper we consider the method of obtaining THz radiation by difference frequency generation (DFG) of multiline CO$_2$ laser. For this purpose a multiline CO$_2$ laser with Q-switching was created. The three strongest lines, 9 R(18), 9 P(20) and 9 P(22) with wavelengths $\sim$9.28, $\sim$9.55 and $\sim$9.57 $\mu$m respectively, held 85% of CO$_2$ laser power, and can be used to obtain difference frequency at a wavelength of $\sim$310 $\mu$m. DFG of other spectral lines fall within the range of 263 $\div$ 8100 $\mu$m. Different nonlinear crystals for DFG and filters to separate THz radiation were considered.

1. Introduction

The development of the terahertz frequency range is one of the most rapidly developing areas of modern applied physics. Interest in terahertz radiation is due to its following properties: non-ionizing radiation (photon energy 0.04–0.004 eV), does not harm the body, that is, it can be used to scan people and baggage; passes well through turbid environment and finely dispersed materials due to weak Rayleigh scattering.

There are many methods and installations to generate the terahertz radiation: THz quantum cascade lasers (QCL), conversion or titanium-sapphire laser radiation, free-electron lasers (FELs), semiconductor THz signal sources, and the production of THz by plasma filaments. However, almost all these methods give a sufficiently low power of radiation. For example, QCL is the most modern source of THz radiation. But at the moment their potential has not yet been fully revealed. The peak power at 5K is about 248 mW [1].

One of the ways to generate terahertz radiation is to generate the difference frequency (DFG) of CO$_2$ laser radiation in ZnGeP$_2$ [2] or GaSe crystals [3]. Two CO$_2$ lasers with the possibility of wavelength tuning were used in these studies, but with a common active medium and with separate resonators. The radiation of lasers was brought together in a crystal, where the conversion was made. In these studies, the wavelengths of these two lasers were tunable, which made it possible to obtain different wavelengths of THz radiation.
However, publications are known (see, for example, [4]), where a CO$_2$ laser operated simultaneously on several (up to 10) vibrational-rotational transitions. The aim of our work is to create a multiline CO$_2$ laser with a single resonator for DFG in nonlinear crystals. This can be achieved by Q-switching. This method is easier to implement than in the case of two lasers. There is no need to synchronize the radiation of lasers in time and space.

2. Optical scheme of measurements and experimental results

![Optical scheme](image)

**Figure 1.** The optical scheme of the installation of a multiline Q-switched CO$_2$ laser.

The optical scheme of the installation for the realization of a multiline CO$_2$ laser with Q-switch is shown in figure 1. The active medium 1 had a gas mixture of CO$_2$:N$_2$:He = 1:2:8, excited by a DC discharge at 17kV. The gas discharge tube was covered with BaF$_2$ plates mounted at the Brewster angle. A CO$_2$ resonator with a length of 255 cm is formed by a spherical mirror 3 with a radius of curvature R = 3 m and an output mirror 4. The output mirror 4 was a standard output mirror from an industrial CO$_2$ laser ILGN-704. A part of the radiation coming out of the resonator using plane-parallel plates 5 of BaF$_2$ and a spherical mirror 7 was directed to the thermoelectric sensor 10 (Ophir-3A) for measuring the energy and to the photodetector 9 (PEM-L-3, temporal resolution 0.5 ns) to measure the time behavior of the pulses, using plane-parallel plates 8 of BaF$_2$. The main part of the radiation by a mirror 11 and a spherical mirror 12 was directed into an spectrometer 13 (IKS-31) with the second PEM-L-3 photodetector. The system was adjusted with the help of a semiconductor laser 6, whose radiation was directed to resonator through a millimeter hole in the mirror 11.

CO$_2$ laser can emit in two bands: ~9 μm and ~10 μm. Since the laser tube was covered with plates of BaF$_2$, the laser emitted at a wavelength of 9.2±9.6 μm. The widest experimental spectrum consisted of 9 lines (Fig. 2). The three strongest lines 9 R(18), 9 P(20) and 9 P(22) with wavelengths ~9.28, ~9.55 and ~9.57 μm respectively, held 85% of CO$_2$ laser power. The line 9 R(18) can give DFG in mixture with 9 P(20) or 9 P(22) line at a wavelength of ~310 μm. DFG wavelengths from other lines fall within the range of 263 - 8100 μm. However, the possibilities of realizing DFG can be substantially limited by the transparent of nonlinear crystals in the terahertz range. Nonlinear crystals of ZnGeP$_2$ [5] and GaSe [6] and PbIn6Te10 [7] have fairly well transparent in the THz wavelength range.
But, for the implementation of the DFG, we needed to make sure that the generation of all these lines occurs simultaneously. For this purpose, the generation time behavior of the lines composing the output radiation spectrum was investigated. These time behaviors for the strongest lines are shown in Figure 3. The analysis showed that the generation of almost all lines occurs at the same time, and if there are deviations, the overlap in time is at least 50 percent (see Figure 3). However, the peak power of these lines can be very different.

Equally important is the stability of the generation of individual lines from pulse to pulse. Figure 4 shows pulsed-periodic generation of the full in the spectrum pulse and on individual lines, spanning ~ 40 consecutive pulses.

An analysis of the results showed that the generation of certain lines does not occur in all pulses (figure 4). For example, generation on the R (22) line is quite rare, on R (18) - more often, and on the lines P (22) and P (20) - almost all the time.
Therefore, it was advisable to focus on obtaining DFG between the lines P (20) and P (22). However, the wavelength of DFG in this case is ~ 5380 μm. Registration of such long-wave radiation is not easy. In addition, the quantum efficiency does not exceed ~ 0.1%. If we use DFG between the line R (18) and both lines P (22) and P (20) having near wavelengths, we can immediately obtain two close terahertz lines near ~ 310 μm. In this case, the quantum efficiency can exceed ~ 0.3%.

For further work, it was necessary to solve two problems: which crystal to use for DFG and which filter to use to cut off the pump CO2 laser radiation.

3. Nonlinear crystals phase-matching angles

The phase-matching angles of the DFG were calculated by the methods described in [8]. We calculated the phase-matching angles for the ZnGeP2, GaSe and PbIn10Te10 crystals.
Figures 5 and 6 show the phase-matching angle as a function of the DFG wavelength ($\lambda_3$) for the PbIn$_6$Te$_{10}$ (fig. 5) and GaSe (fig. 6) crystals. In the calculation we fixed one wavelength of the CO$_2$ laser radiation ($\lambda_1$) by 9.6 $\mu$m, and the second ($\lambda_2$) was changed in the range from 9.2 $\mu$m to 10.6 $\mu$m. Since, according to our estimates, the power of THz radiation at a wavelength of $\sim$ 310 $\mu$m is expected to be maximal, then on the phase-matching curve this point is shown by a red circle. The remaining circles (yellow) correspond to other possible wavelengths of DFG, which are obtained by using a laser of the CO$_2$ laser spectrum shown in Fig. 2. At the DFG wavelength of $\sim$310 $\mu$m, the phase-matching angle was: for a GaSe crystal - 12.5 degrees, for ZnGeP$_2$ - 25 degrees, for PbIn$_6$Te$_{10}$ - 49.6 degrees. For ZnGeP$_2$ and PbIn$_6$Te$_{10}$, large phase-matching angles are not an obstacle to the realization of DFG, since these crystals can be cut at the required angle and, as a result, pump radiation can be direct at the normal to the surface of the crystal. But in the case of GaSe this can not be done, since it stratifies. Therefore, a small phase-matching angle for a given crystal shows the possibility of implementing DFG.

4. The proposed optical scheme of DFG

In Fig. 5 the proposed installation scheme is presented. We created a multiline Q-switched CO$_2$ laser with one resonator for DFG in nonlinear crystals and selected a crystal. It remains to solve two questions: what detector to choose for recording terahertz radiation and what filter to set before it. The bottom line is that the theoretical efficiency of the conversion is of the order of $10^{-2}$, and in the experiment it is expected to be several orders lower. Therefore, THz radiation should be separated from the radiation of our laser by an optical filter.
The pump radiation has a wavelength of 9–10 μm. In figure 5 shows the transmittance and reflection of a teflon (fluoroplastic) sample of a thickness of 0.1 mm in the mid-IR and THz ranges. This material was used in work [2], but it passes a sufficiently large part of the CO$_2$ laser radiation and is not very suitable as a filter, but can be used as a material of lens 2. Another material for filter was considered crystalline quartz. Based on the analysis of figure 6, it was concluded that this material is suitable for further work as a pump radiation filter.

**Figure 7.** The proposed scheme for DFG in nonlinear crystals.

**Figure 8.** Transmittance of teflon (fluoroplastic) 0.1 mm. Mid-IR and THz range [9].

**Figure 9.** Transmittance of crystal quartz 8 mm for mid-IR (left) [10] and transmittance and reflectance of crystal quartz 1 mm. for THz range (right) [9].
Having analyzed the proposals of various manufacturers of medium-IR and THz radiation detectors, we settled on the QMC Instruments Ltd.’s Pyro-electric detector (DLA-TGS element with THz absorption coating) [11]. It has a high sensitivity (1600 V/W at a modulation frequency of the signal 80 Hz) in a wide range from 100 GHz to 100 THz at room temperature.

5. Conclusions
Multiline CO₂ laser with Q-switching of a resonator for DFG in nonlinear crystals was created. The generation spectrum contained 9 lines from a 9-micron band, since the laser tube was covered with plates of BaF₂. The three strongest lines 9 R(18), 9 P(20) and 9 P(22) with wavelengths ~9.28, ~9.55 and ~9.57 μm respectively, held 85% of CO₂ laser power. Totally, along all the generation lines, the peak power reached 1 kW. The strongest lines can give DFG at a wavelength of ~ 310 μm. DFG wavelengths from other lines fall within the range of 263 ÷ 8100 μm. The phase matching angles for DFG in the terahertz range in GaSe, ZnGeP₂, and PbIn₆Te₁₀ crystals were calculated. From these results, it was concluded that all the crystals are suitable for further work. A THz range detector and a filter were selected. The crystalline quartz is best suited for filter.

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