Difference frequency generation of CO and CO_2 lasers in $PbIn_6Te_{10}$ crystal

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***Abstract*—$PbIn_6Te_{10}$ is a new nonlinear material with main transparency window 1.7-31 µm. For the first time optical properties were studied in the THz region, as well as DFG of CO and CO_2 laser theoretically and experimentally. The simulation indicated that DFG spectrum can contain dozens of strong lines within 10-13 µm at fixed phase-matching angle. As well CO and CO_2 laser lines can be down-converted, respectively, into 110-1000 and 200-1000 µm range. CO laser and CO_2 laser conversion efficiency into the mid-IR domain was up to ~10^{-6}.**

I. INTRODUCTION

Frequency mixing of CO and CO_2 lasers radiation in nonlinear crystal allows one to obtain thousands of spectral lines in the very broad wavelength range from 2.5 to ~17 µm [1]. Unfortunately, the most of known mid-IR crystals such as $CdGeAs_2$, $AgGaSe_2$ or GaSe have long-wave transparency edge below 18 µm [2]. Thus, an extension of frequency converted CO or CO_2 laser radiations to longer wavelength of up to 30 µm requires a development of nonlinear crystals transparent in the above mentioned range. The crystal $PbIn_6Te_{10}$ (PIT) was claimed as a new mid-IR broadband material for nonlinear applications [3]. It has very wide main transparency window extending from 1.7 to 31 µm, high nonlinear coefficient of 51 pm/V and adequaty birefringence of $\Delta n \approx 0.05$ for phase-matching that makes this crystal very attractive for three-wave mixing in the mid-IR [3]. In this research, for the first time, we studied optical properties of PIT crystal in the THz region, as well as examined difference frequncy generation (DFG) of CO and CO_2 lasers into the mid-IR and THz region by modeling, and also frequency conversion of CO laser into mid-IR experimentally.

II. RESULTS

PIT crystal of 20x13x14.5 mm^3 size , was exploited in the experimental study of DFG under the pump by a hybrid laser system consisted of multiline Q-switched CO and CO_2 laser described in details elsewhere [1]. The FWHM pulse duration of each laser was about 1 µs at Q-switching frequency of ~100 Hz. The peak power reached 2 kW for CO laser and 0.3 kW for CO_2 laser. The CO laser spectrum consisted of about 100 emission lines in the wavelength range from 5 to 7 µm. The CO_2 laser spectrum consisted of two strong lines at wavelengths of 9.3 and 9.6 µm, which contained ~85% of the total laser power. A few weak CO_2 laser lines were also recorded. The both laser beams were focused onto the PIT crystal by ZnSe lens with a focal length of 18 cm. To select DFG radiation an optimized set of long-wave pass spectral filters (Spectrogon LP-11450) were applied, which attenuated pump radiation more than 10^8 times.

A maximum DFG power on wavelengths longer than 11.5 µm was obtained at phase-matching angle of 33.1°. The peak power of DFG radiation reached 1.2 mW. Frequency conversion efficiency in power is estimated at about 10^{-6}.

Simulation of DFG was carried out under plane-wave and low frequency conversion efficiency approximation. The simulation for frequency conversion of not selective CO laser and two-wavelength CO_2 laser into the mid-IR accounted experimental conditions, with fixed angular position of the crystal used. The result on estimated efficiency is plotted in Fig. 1. In this figure it is seen that DFG spectrum consisted of dozens of strong frequency converted lines in the wavelength range from 10 to 13 µm. It means that noncritical phase-matching takes place in PIT crystal. For wavelengths longer than 13 µm the power of DFG lines is reducing sooner due to significant reducing in output power of CO laser lines at wavelengths exceeded 5.6 µm. To extend DFG spectrum to longer wavelengths, the power of high vibrational-rotational CO laser lines should be increased.

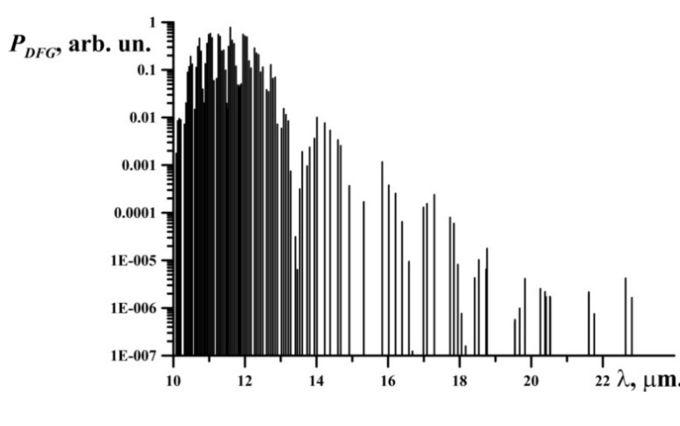

Fig. 1. Calculated spectrum of DFG at phase-matching angle 33.1°.

Optical properties of PIT in the THz range were studied by homemade time domain spectrometer (TDS) in the limited spectral range 0.2-1.3 THz due to high, few tens cm^{-1} absorption coefficient of non-identified origin. Adequate data were selected from recorded data by using visual criterion as quality of etalon patterns described in [4]. Selected dispersion

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curves are presented in Fig. 2.

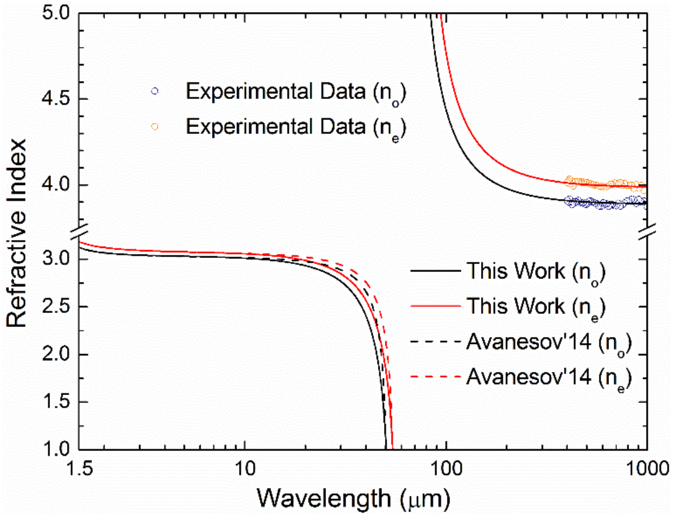

Fig.2. Refractive index dispersion in PIT samples: points are experimental data; solid lines are approximations; $\theta_{int.}$ is internal phase matching angle.

By using dispersion equations from [3] and recorded data, new dispersion equations were approximated for the entire transparency range of PIT as:

$$n_o^2 = 15.1089 + \frac{0.60530}{\lambda^2 - 1.18239} + \frac{25844.6}{\lambda^2 - 4370.34}$$

$$n_e^2 = 15.8779 + \frac{0.92709}{\lambda^2 - 0.84640} + \frac{33220.0}{\lambda^2 - 5176.38} \quad (1)$$

By using equations (1) phase matching curves for down-conversion of CO (Fig. 3) and CO_2 laser lines into the THz region were calculated.

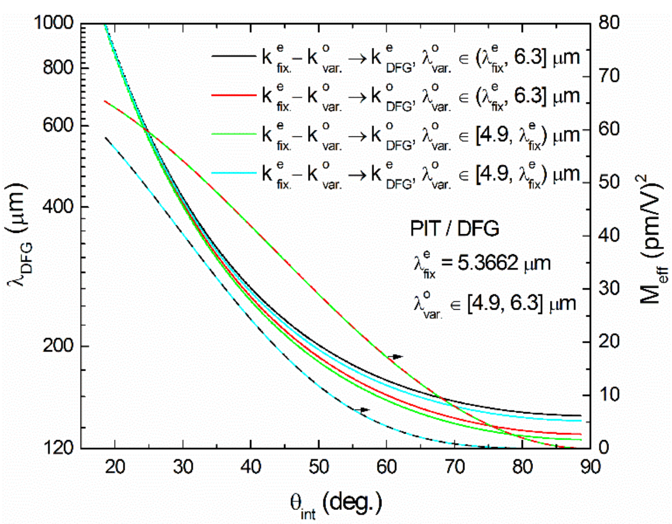

Fig. 3. Phase matching curves for I and II type down-conversion of nonselective CO laser lines in PIT: 5,3662 μm is the most powerful emission line; longer (up to 6.3 μm) and shorter (down to 4.9 μm) wavelengths pump was considered. Presented data, not possibility, are limited by 1000 μm.

In Fig. 3 it is seen that down conversion is possible into the spectral range from 110 to over 1000 μm. Longer wavelengths are generating at smaller phase matching angles that favorable due to angular dependences of effective nonlinearities for I and II type of interactions $d_{\text{eff}}(eo \rightarrow o) = d_{11}\cos\theta\cos 3\varphi$ and $d_{\text{eff}}(eo \rightarrow e) = d_{11}\cos^2\theta\sin 3\varphi$, respectively, those are maximal at $\theta_{int.} = 0$.

Down-conversion of CO_2 laser is found possible from 200-240 μm depending on interaction type to over 1000 μm. To estimate figure of merit for THz generation second order nonlinear susceptibility coefficients from [3] were used.

III. Summary

Thus, the promise of the PIT crystal for long-wave frequency mixing was shown. In the first examination of PIT crystal the peak power of DFG lines of CO and CO_2 laser radiation reached 1.2 mW. Optical properties and phase-matching of PIT crystal in the THz range were studied. We do believe that both improvement of molecular gas lasers and PIT crystal will enable us to cover long-wave domain of the mid-IR range up to 30 μm and the THz range over 1.3 THz.

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