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Mercury thiogallate nanosecond optical parametric oscillator continuously tunable from 4.2 to 10.8 μm

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Abstract

We demonstrate an optical parametric oscillator based on two $HgGa_2S_4$ crystals with an extremely wide tuning range from 4.2 to 10.8 μm. The $HgGa_2S_4$ optical parametric oscillator was pumped by a Q-switched nanosecond Nd:YLF laser at 1.053 μm. The absorption spectrum of ammonia was presented to demonstrate the feasibility of the developed optical parametric oscillator system for spectroscopic measurements and gas detection.

Keywords: mercury thiogallate ($HgGa_2S_4$), optical parametric oscillator (OPO), ammonia absorption spectrum

(Some figures may appear in colour only in the online journal)

1. Introduction

Widely tunable mid-infrared laser sources are of great interest for various scientific applications such as trace gas detection, spectral analysis, remote sensing, environment monitoring and others. Optical parametric oscillation is one of the most widespread ways to generate tunable coherent radiation in the spectral range from visible to mid-IR.

Usually a nonlinear crystal used in a mid-IR optical parametrical oscillator (OPO) pumped at about 1 μm is silver thiogallate $AgGaS_2$ (AGS). However, this crystal has a relatively low surface damage threshold of 10 MW cm^{-2} (20 ns, 10 Hz) [1]. In terms of the surface damage threshold, mercury thiogallate $HgGa_2S_4$ (HGS) is a more promising nonlinear material than AGS. As compared with AGS, HGS has a relatively high surface damage threshold of 40 MW cm^{-2}, (30 ns) [2] and a relatively high nonlinear coefficient $d_{36} = 31.5$ pm V^{-1} [3] and against $d_{36} = 17.5$ pm V^{-1} for AGS [3]. The main feature of both crystals is that HGS and AGS can be pumped at about 1 μm by a commercially available laser (e.g. Nd:YAG, Nd:YLF) without two-photon absorption due to its wide band-gap (2.73 eV for AGS and 2.79 eV for HGS) [4]. In spite of the fact that AGS is a standard non-oxide mid-IR material, HGS is a more attractive nonlinear optical material for OPO due to its high nonlinearity, high damage threshold and better heat conduction. HGS is a negative uniaxial crystal and belongs to the $\bar{4}$ symmetry class. The effective nonlinearities of HGS are given by $d_{eoe} = (d_{36}\sin 2\varphi + d_{31}\cos 2\varphi)\sin\theta$ for the type I interaction and by $d_{eoe} = d_{oee} = (d_{36}\cos 2\varphi - d_{31}\sin 2\varphi)\sin 2\theta$ for the type-II interaction. HGS has a transmission range extending from 0.55 to 13 μm [5]. This crystal was introduced for nonlinear optical applications as early as 1976 [5]. However, the use of this material for OPO was restricted due to the poor optical quality of the grown samples. Suitable-sized HGS samples with good optical quality were grown by the Bridgeman–Stockbarger technique in High Technologies Laboratory, Kuban State University; and nanosecond OPO based on HGS was first demonstrated in 2003 [6]. The idler tuning range of HGS OPO described in [6] was 2.3–4.4 μm. Later, the same authors presented HGS OPO with tuning range from 3.7 to 5.7 μm [7]. The tuning range was restricted due to the crystal dimensions and OPO cavity configuration.

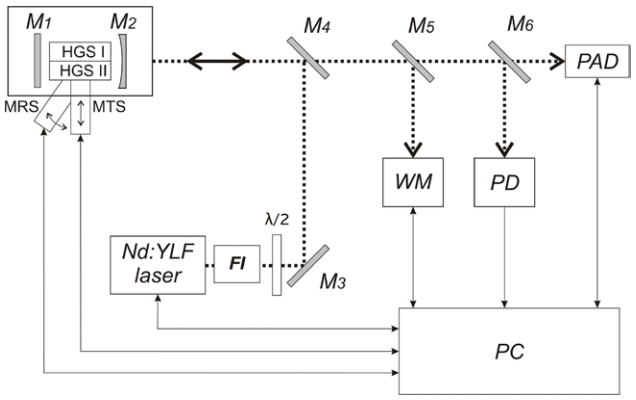

Figure 1. Experimental set-up: Nd:YLF laser—pump laser, FI—Faraday isolator; $\lambda/2$—half-wave plate; M1–M6—mirrors; MTS—motorized translation stage; MRS—motorized rotation stage; WM—wavelength meter; PD—pyroelectric photodetector; PAD—photo-acoustic detector; PC—computer.

The maximum idler energy was 3.3 mJ at 4 μm (67 mW, 5 Hz). OPO was pumped at 1.064 μm by a Nd:YAG laser. The pulse duration was 4.5 ns. The laser pulse energy was about 100 mJ.

The nanosecond OPO based on HGS, which was described in [4], had a high idler energy of 6.1 mJ at 4.03 μm (610 mW, 100 Hz). This OPO was pumped by a Q-switched Nd:YAG laser/amplifier system delivering up to 250 mJ per pulse at 100 Hz with a pulse duration of 8 ns.

In the present work we demonstrate a nanosecond OPO based on HGS with an exceedingly wide tuning range from 4.2 to 10.8 μm. We propose a new conception of a widely tunable nanosecond optical parametric oscillator based on two HGS crystals installed into the same cavity. Each crystal operates in its own spectral range, but the switching of HGS crystals shifts the wavelength tuning range to the other spectral range. The developed OPO combined with a resonant photoacoustic detector allows the creation of a laser gas analyzer for the detection of different gases in a wide spectral range.

2. Experimental set-up

The HGS samples used in the present study were grown by the Bridgeman–Stockbarger technique in High Technologies Laboratory, Kuban State University. The samples were cut for the type-II (eoe) phase-matching. One sample (sample A) was cut at $\theta = 60^\circ$ and $\varphi = 0^\circ$ for an idler wavelength of about 4.8 μm at normal incidence. The second sample (sample B) was cut at $\theta = 47^\circ$ and $\varphi = 0^\circ$ for an idler wavelength of about 7.5 μm at normal incidence. Both samples were 13 mm-long with an aperture of 5 mm × 5 mm. These samples were polished and antireflection coated at the same technological block. The surfaces of the crystals were coated with antireflection (AR) dielectric layers so that the transmission 1.053 μm was ~99%.

The experimental set-up of the developed OPO based on two HGS crystals is shown in figure 1. The experimental set-up consists of the following components: a pumping Nd:YLF laser, a Faraday isolator (FI), a half-wave plate ($\lambda/2$), mirrors (M_1–M_6), a pyroelectric detector (PD), a wavelength meter (WM), a photo-acoustic detector (PAD), a motorized translation stage (MTS), a motorized rotation stage (MRS) and a computer (PC).

The pump source is a diode-pumped Q-switched Nd:YLF laser (TECH-1053-N, Laser-compact Group, Russia). The pump wavelength is 1.053 μm and the pulse duration is 5–10 ns. The maximum laser pulse energy at repetition rate of 100–5000 Hz is about 1300 μJ. The pump pulses that pass through the FI and half-wave plate are reflected by mirrors M3 and M_4 into the OPO cavity, which is formed by mirrors M1 and M2.

The OPO cavity with a length of 23 mm is formed by the semitransparent M2 (Layertec) and reflecting M1 (Thorlabs) mirrors. The output coupler M2 is transparent for the pump and idler waves and has $R = 90\%$ for the signal wave. The signal wave is not totally reflected by the output coupler to avoid extreme intracavity fluence that could damage the crystals. The HGS crystals are placed on the OPO cavity inside a thermostat. A Peltier element is used to maintain the optimal temperature of both crystals at a level of 20–30 °C with an accuracy of 0.1 °C. The OPO wavelength tuning is performed via the precise rotation of the HGS crystals relative to the optical cavity axis using the MRS and switching between the crystals by MTS.

The signal and idler waves were extracted from the OPO cavity through the output coupler M2. Subsequently, the idler wave passed through the dichroic mirrors M4, M5 and splitting mirror M6, and then arrived to the PAD, which was described in [8]. The latter was used to record the absorption spectra of the various gas mixtures. The signal wave was reflected by dichroic mirror M5 to the wavelength meter WM (Angstrom LSAL IR, Russia). The signal wavelength was measured by the WM, and then the idler wavelength was determined by recalculation. Mirror M5 was transparent for the idler wave. The splitter M6 reflected a part of the idler beam to the pyroelectric detector PD (MG-30, Russia), which was used to measure the OPO power and normalize the PAD data. Electrical signals from the WM, PD and PAD were sent to the PC for processing and display. The pulse repetition rate, the pulse energy of the pump laser, the temperature of the crystals, the movement of the MTS (for switching between two crystals) and the MRS were controlled by the PC.

3. HGS OPO: experimental results

The obtained idler wavelength tunability was 4.2–5.71 μm for the first crystal and 5.7–10.8 μm for the second crystal. The total scan time in a spectral range 4.2–10.8 μm was less than 2 min.

The OPO pulse energy was measured using Ophir Vega PE-10C power/energy meter (Israel) placed in front of mirror M6. The measured idler energy was 4–10 μJ in the spectral range of 4.2–5.71 μm (sample A), 1–8 μJ in the spectral range of 5.7–8.5 μm (sample B) and less than 1 μJ at 8–10.8 μm. The experiments were carried out under the following conditions: the pump energy was 900 μJ; the pulse repetition rate was 1000 Hz; and the temperature of the HGS crystals was 25 °C. The average OPO radiation power was ~10 mW (1000 Hz)

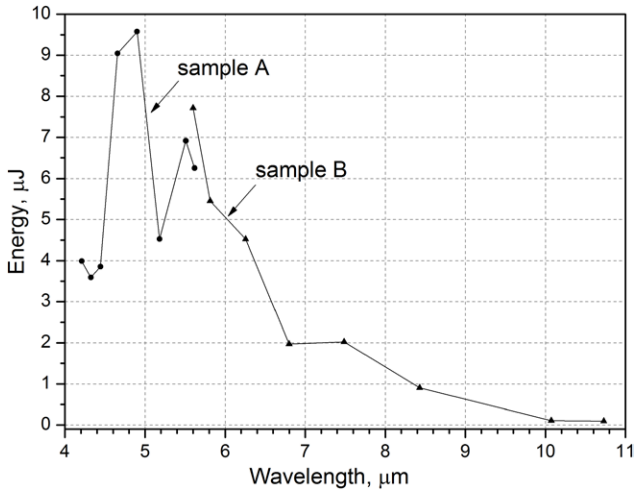

Figure 2. Idler energy versus idler wavelength: sample A ($\theta = 60\,°$, $\varphi = 0°$); sample B ($\theta = 47\,°$ and $\varphi = 0\,°$).

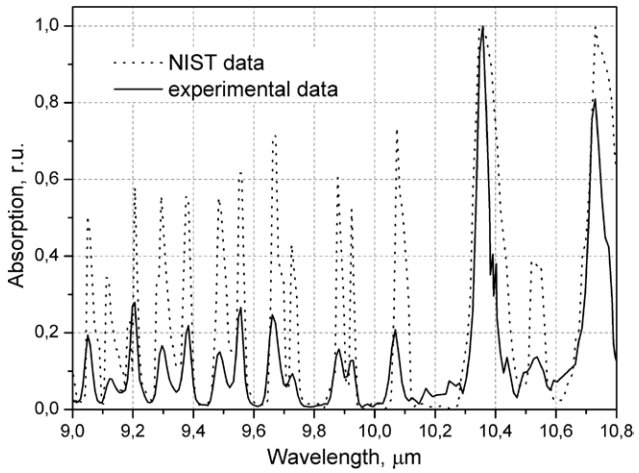

Figure 3. Absorption spectra of ammonia (gas mixture of 0.1% NH_3 in N_2). Solid line—experimental data, dashed line—data from NIST.

at 4.5 μm. The OPO energy monotonically decreased with increasing the wavelength in a spectral range of 8–10.8 μm. The OPO threshold was 11 mJ cm^{-2}. This value agrees with the 10 mJ cm^{-2} given in [7]. The energy dependence is not symmetric, with a stronger decrease at longer wavelengths (figure 2) due to an increase of absorption level in mercury thiogallate at wavelengths of more than 9 μm.

4. Analysis of the NH_3 absorption spectrum

To demonstrate the feasibility of the OPO system for spectroscopic measurements and gas detection we took the absorption spectra of NH_3 gas with distinct rotational–vibrational features in the mid IR. A test gas mixture containing 0.1% ammonia in nitrogen was used. The PAD used in this experiment was 90 mm in length and had a fundamental resonance frequency of ~1800 Hz, and the quality factor was ~40. The PAD was filled with the test gas mixture via gas purging. The pulse repetition rate of the pump laser was equal to the PAD resonance frequency. The OPO wavelength was continuously tuned in the spectral range of 9–10.8 μm (1110–926 cm^{-1}). The ammonia absorption spectrum was measured with the PAD. The data from the PD were used to normalize the PAD signals with respect to the idler energy. The normalized NH_3 absorption spectrum was recorded by the PC in real time.

Figure 3 compares the NH_3 absorption spectrum obtained with our HGS OPO system with the National Institute of Standards and Technology (NIST) data (4 cm^{-1} resolution). As seen in figure 3, the experimental absorption spectra and the NIST data agree with each other, thus confirming a high degree of reliability of the obtained results.

5. Conclusion

The developed OPO provides continuous wavelength tuning in the spectral range from 4.2 to 10.8 μm. An extremely wide tuning range was obtained by a precise linear switching between the crystals and a precise rotation of two HGS crystals. The ammonia absorption spectrum was investigated using the developed OPO. The experimental absorption spectra and the NIST data agree with each other. This device can be used for industrial, medical and special applications.

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