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Generation of tunable mid-IR radiation by second harmonic in a $CdGeAs_2$ crystal

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Abstract. **Tunable mid-IR radiation is obtained during second harmonic generation of tunable CO_2-laser radiation using a $CdGeAs_2$ crystal. Its angular tuning characteristics at the CO_2-laser wavelength, angular acceptance angle and spectral acceptance are measured. For second harmonic generation at 10.6 μm, the conversion efficiency in the $CdGeAs_2$ crystal is 90 times higher than that in the $ZnGeP_2$ crystal.**

Keywords: *tunable CO_2 laser, second harmonic generation, nonlinear $CdGeAs_2$ crystal.*

1. Introduction

Nonlinear frequency mixing is now a well-established technique for obtaining tunable radiation in the spectral range where laser source as such is not available. Among the known different techniques, second harmonic generation is the simplest and the conversion efficiency achieved can be much higher than in any other interaction. Since the discovery of second harmonic generation by Franken et al [1] in 1961, numerous nonlinear crystals have been grown and tested for efficient application in laser devices. But till now, no ideal crystal has been grown for laser application from ultraviolet (UV) to infrared (IR) spectral range and search for efficient crystals is still going on.

Optical radiation in the spectral range from 3 to 5 μm is of particular interest for such applications as atmospheric monitoring, trace gas detection in the atmospheric window, laser spectroscopy, Auger recombination, etc. Because a tunable laser source for this spectral range is still absent, use can be made of either difference frequency generation or optical parametric oscillation to obtain lasing in this range. The above-mentioned processes have both advantages and disadvantages. Second harmonic of tunable CO_2-laser radiation can also serve as a tunable source in the 4.5-to-5.5 μm spectral range, thereby providing the higher efficiency of generated radiation than any other process because it is the simplest process of optical frequency conversion. Frequency doubling of a CO_2 laser has been studied separately by different investigators in different IR transmitting crystals such as $AgGaSe_2$ [2 – 6], $AgGaS_2$ [7], $ZnGeP_2$ [8 – 11], Tl_3AsS_3 [12, 13], GaSe [14, 15], $AgGa_xIn_{1-x}Se_2$ [16 – 20], $HgGa_2S_4$ [21 – 23], and $CdSiP_2$ [24]. Nevertheless, each crystal possesses advantages and disadvantages.

Cadmium germanium arsenide ($CdGeAs_2$, CGA) is a positive uniaxial crystal having the point group symmetry ^{42}m and optical transmission from 2.4 to 18 μm. This crystal can be a suitable candidate for a second harmonic generator of CO_2-laser radiation. Cadmium germanium arsenide, a ternary chalcopyrite crystal, has the highest second order nonlinearity (236 pm V^{-1}) in the inorganic nonlinear crystal group and has been known since 1967 [25]. The crystal is a perfect candidate for second harmonic generation (SHG) of CO_2-laser radiation and also for IR radiation of the second atmospheric window (8 – 12 μm spectral range) through optical parametric processes. Its optical properties were described by Byer et al [26] and Boyd et al [27]. Second harmonic generation of TEA CO_2-laser radiation and difference frequency generation were reported by Kildal et al [28]. Though the device based on the CGA crystal was demonstrated in the early 1970s, its application was restricted due to nonavailability of a crystal of big size and good optical quality. Nowadays, with the development of the growth technology, large crystals of high optical quality have been grown for laser device applications [29, 30]. Such laser devices as efficient parametric frequency generators for the spectral range 7 – 20 μm [31, 32] as well as second harmonic and sum frequency generators [33 – 36] have also been demonstrated. The temperature dependence of the SHG efficiency has been considered by Zakel et al [37]. In this paper, we demonstrate, perhaps for the first time, formation of tunable mid-IR radiation by second harmonic generation from cw CO_2-laser radiation.

2. Experimental

We used in the experiment a 6×10×5.7-mm type-I CGA crystal cut at an angle of 41°. Its transmission characteristics throughout its spectral range were measured with a FTIR spectrophotometer and in the spectral region from 9 to 11 μm – with a cw CO_2 laser (without taking into account the Fresnel reflection loss, Fig. 1; see also [25, 31]). Using the relation given in [38] the measured transmission coefficient made it possible to determine the absorption of this 5.7-mm-thick crystal. Thus, the absorption coefficient of the crystal was equal to 0.05 – 0.07 cm^{-1} throughout its usable transmission range.

A sealed cw CO_2 laser tunable from 9.2 to 10.78 μm was used as a pump source in our experiment. A chopper was placed inside the laser cavity to increase the peak power, which also helped to detect the generated radiation. The cw CO_2-laser radiation was focussed onto the crystal by a ZnSe lens with a focal length of 12 cm. The crystal was placed not

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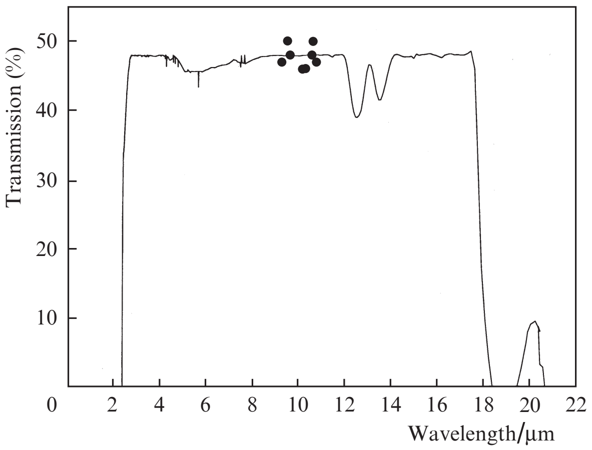

Figure 1. Transmission characteristics of a 5.7-mm-thick $CdGeAs_2$ crystal. The curve is spectrophotometric measurement, dots (●) are the values experimentally measured with a tunable CO_2 laser in the 9–11-μm spectral range.

in the focal point of the lens but within the confocal length (6.5 cm) to avoid any surface damage (the aperture length was 1.5 cm). The CO_2-laser wavelength was monitored with a spectrum analyser. The generated second harmonic radiation was detected with a liquid-nitrogen-cooled MCT detector operating without a preamplifier. The residual unconverted fundamental radiation was blocked by a 3-mm-thick sapphire plate. The electrical signal from the detector was displayed on a 100-MHz storage oscilloscope.

3. Result and discussion

The wavelength dependences of phase matched tuning characteristics for second harmonic generation in the crystal are shown in Fig. 2. The curves correspond to theoretical calculations obtained from different Sellmeier coefficients of Kato, as reported by Vodopyanov et al. [32]. One can see that the experimental points are in close agreement with the predicted phase matched angles obtained from the Sellmeier coefficients of Kato [32]. The maximum departure of the experimental values from the theoretical results obtained from Kato's Sellmeier coefficients [32] is ±0.5°. This departure may be caused by the difficulty of ascertaining the phase matched peak arising due to broad angular and spectral bandwidths. Using other available Sellmeier coefficients [28, 39], we have found that the predicted phase matched angles differ by more than 1°; however, there is a large departure (about 10°) of the measured phase matched angle from that obtained theoretically by using Sellmeier coefficients, as reported by Byer at al. [26]. The difference of the measured phase matched angles from the theoretical values obtained with the help of other Sellmeier coefficients [26, 28, 39] may be due to inaccurate dispersion values at the desired wavelengths obtained from the Sellmeier coefficients derived by the investigators [26, 28, 39] mentioned earlier. The angular phase matched bandwidth in this crystal was also measured at 10.6 μm. It has been found that the measured full width at half maximum (inside the crystal) is 0.9°, while the theoretical value is 0.597° (obtained using Kato's Sellmeier coefficients [32]); the total angular (zero-to-zero detector reading) bandwidth is 1.3°, while the corresponding theoretical value is 1.35°. These differences of measured values from the theoretical results may be explained by the measurement error due to fluctuations of detector output readings as well as by the inaccuracy of the dispersion value obtained from the Sellmeier equations. A similar angular phase matched bandwidth is also observed in other wavelengths. In addition, we have found that with changing the wavelength of fundamental radiation from 9.2 to 10.7 μm, the changes in the output readings of the detector are virtually absent. The observed variation in the output signal is due to a change in the laser beam power at the fundamental frequency. One can also see from Fig. 3 that there is a broad phase matched angle in the spectral range of CO_2-laser radiation.

Because of the low output power (1 W) of cw radiation at the fundamental frequency, we failed to measure the output power at 10.6 μm with a power meter; however, we were able to compare the conversion efficiency of a 5.7-mm-thick CGA crystal with that of a 3-mm-thick $ZnGeP_2$ (ZGP) crystal by

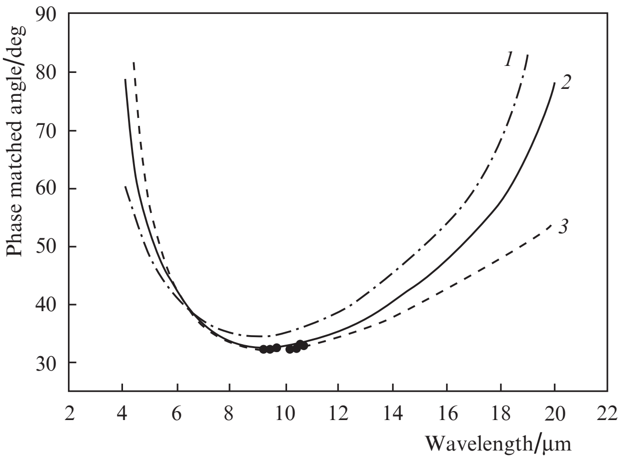

Figure 2. Wavelength dependences of phase matched tuning characteristics for type-I (ee→o) second harmonic generation in the $CdGeAs_2$ crystal. The curves correspond to calculations obtained from different Kato's Sellmeier coefficients [26] (*1*), [28, 39] (*2*) and [32] (*3*); dots (●) correspond to the measured values.

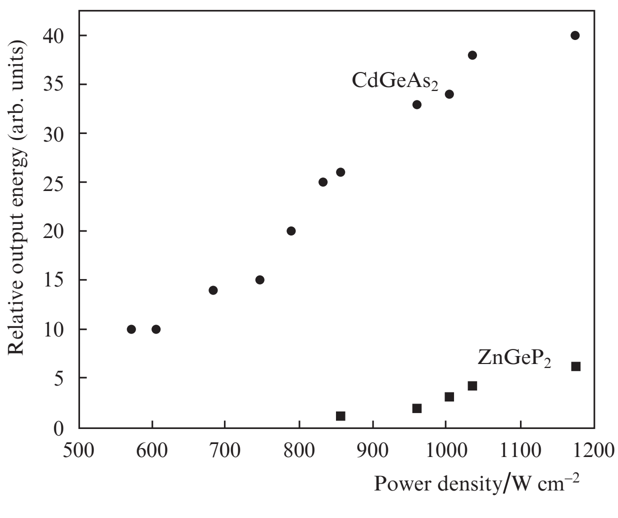

Figure 3. Dependences of the relative output energy on the power density during the second harmonic generation of CO2-laser radiation in a 5.7-mm-thick $CdGeAs_2$ crystal and a 3-mm-thick $ZnGeP_2$ crystal at 10.6 μm.

measuring the output energy of the generated second harmonic radiation of the incident CO_2 radiation at 10.6 μm at different power densities. This was done just changing the focal position of the lens such that the crystal lies within the confocal distance (6.5 cm). The measurement data indicates that the conversion efficiency of this CGA crystal is about 6.25 times higher than that of the $ZnGeP_2$ (ZGP) crystal although consideration of the figure of merit (d_{eff}^2/n^3) has shown that at 10.6 μm the CGS crystal is about 60 times more efficient that the $ZnGeP_2$ crystal. It is still not clear why the signal level obtained with the CGA crystal is 14 times lower. It may be caused by a considerable amount of incident power loss (about 32% from the front surface of the incident beam and about 35% from the back surface of the generated beam) because of Fresnel reflection from the crystal surfaces due to large rotation angle of the crystal (~30°) compared to the ZGP crystal whose output surface is almost normal to the incident radiation. It is also worth mentioning here that Kildal et al. [28] reported the 15% conversion efficiency for type-II SHG of 10.6-μm radiation from a TEA CO_2 laser with a pulse duration of 160 ns in a 9-mm-thick crystal. Vodopyanov et al. [32] reported the 62% conversion efficiency for SHG of 10-μm radiation from a free electron laser with a pulse duration of 3.9 ps in a 7-mm-thick crystal.

4. Conclusion

Thus, we have found that the CGA crystal is a perfect candidate for mid-IR generation by different frequency interaction processes. For SHG the advantage of this crystal is that due to flat phase matched angular tuning characteristics for the CO_2-laser radiation, and due to broad angular acceptance, it is not necessary to realign the crystal to the phase matched peak when changing the incident radiation wavelength. From the point of view of the figure of merit, the CGA crystal is far more efficient than the ZGP crystal.

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