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Optical parametric generation in CGA crystal

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Abstract

We have demonstrated optical parametric generation (OPG) in CGA crystal known for its extremely high second-order nonlinearity (236 pm/V). A single-pass OPG was pumped by 10 μJ, 600 fs pulses at $\lambda = 5$ μm and the output was tunable in the 7–18 μm range. Efficient second harmonic generation of 9–11 μm radiation with an internal conversion efficiency of 62% has also been demonstrated. © 2002 Elsevier Science B.V. All rights reserved.

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$CdGeAs_2$ (CGA) crystal is known for its highest second-order nonlinearity ($d_{eff} = 236$ pm/V) among any crystals in practical use (for example, its nonlinear-optical figure of merit, d_{eff}/n^3, is 42 times larger than that of periodically poled lithium niobate). It transmits light between 2.4 and 18 μm and is a perfect candidate for second-harmonic generation (SHG) and nonlinear-optical frequency conversion via optical parametric oscillation (OPO), specifically into the second atmospheric window (8–12 μm). In the past, the linear absorption of as-grown CGA crystals was high ($\sim$1 cm^{-1}) which precluded its wide use in nonlinear optics.

Lately, it became possible, owing to the advancements in the crystal growth and after-growth processing, to produce large size CGA crystals with improved transmission [1–3]. As a result, highly efficient frequency doubling of CO_2 laser radiation (conversion efficiency 28% at room temperature and 50% at 77 K) [1] and difference frequency generation (at room temperature) with photon conversion efficiency >20% in the 7–20 μm spectral range [3] have been reported in CGA. Despite the fact that pulsed thresholds as low as 30 μJ are expected in a singly resonant CGA OPO, the great potential of this crystal as an OPO/OPG material has not been materialized so far. In this paper, we report the first demonstration of optical parametric generation (OPG) in CGA (which we regard as a logical step to achieving an OPO in this crystal), as well as efficient SHG operation.

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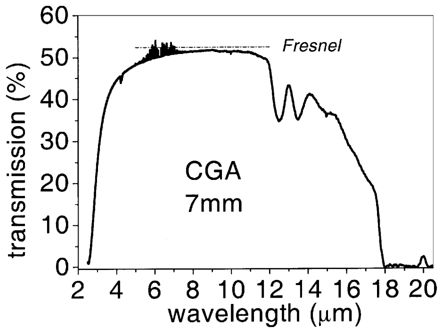

Fig. 1. Measured transmission spectrum of the uncoated CGA crystal. Dash–dotted curve corresponds to Fresnel reflection losses (refractive index $n \approx 3.5$).

The CGA crystal was grown at Inrad, by the horizontal gradient freeze technique, and was subjected to post-growth treatment (irradiation and annealing) [2] to decrease its room temperature optical absorption. The 6×8 mm^2 cross-section, 7 mm long sample designed for type I phase matching was cut at $\theta = 33°$ and $\varphi = 0°$. The crystal faces were polished but not anti-reflection coated. Fig. 1 shows the measured transmission spectrum. The values of room-temperature absorption coefficient of less than 0.1 cm^{-1} were achieved in the whole range $\lambda = 5$–12 μm. This is comparable to the results of Schunemann et al. [4] reported recently.

As a pump source for the OPG, we have used $\lambda \approx 5$ μm pulses from the Free-Electron Laser FELIX in Nieuwegein, Netherlands [5]. Schematic of the OPG experiment is shown in Fig. 2. The laser beam consisted of a train of short pulses (micro-pulses), which are separated by intervals of 40 ns. The duration of the train (macro-pulse) was approximately 4 μs ($\sim$100 micro-pulses) with the total energy $\sim$1 mJ and the overall laser repetition rate was 10 Hz. The laser micro-pulse duration was $\tau \approx 600$ fs at a FWHM spectral width of 24 cm^{-1}. (It has been found in the previous experiments [6], using autocorrection technique with a GaSe crystal that the time-bandwidth product for the FELIX pulses is close to 0.44, i.e. near the value for the Fourier-transform-limited Gaussian pulses.) The pump laser beam was focused into the CGA crystal using a telescope made of two BaF_2 lenses with $f = +15$ and $f = -5$ cm and had a beam size ($1/e^2$ intensity radius) of $w_0 \approx 450$ μm (which is larger than the birefringent walk-off in the CGA crystal: 160 μm at $L = 7$ mm and $\theta = 33°$). The OPG output was monitored using a long-pass ($\lambda > 5.5$ μm) filter and MCT (77 K) or pyroelectric detector. An OPG threshold was observed at the incoming peak laser intensity of approximately $I_L = 3$ GW/cm^2. This is close to the calculated value (2.2 GW/cm^2) [7] obtained from our numerical modeling of the three-wave interaction process. The modeling assumed Gaussian pulse shape in time and transverse coordinate and neglected group velocity mismatch between the three interacting waves (the time walk-off between any of the three beams was less than 55 fs at 7 mm crystal length, which is much shorter than the laser pulse duration).

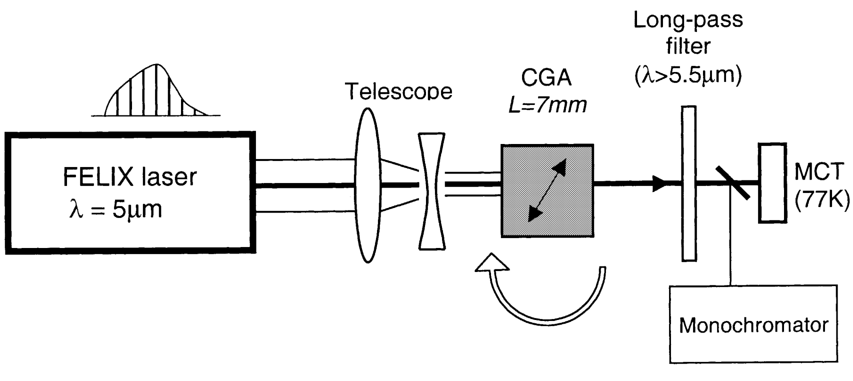

Fig. 2. Schematic of the OPG experiment.

The OPG output was easily detectable when it was angular-tuned within the range 7–18 μm. The experimental OPG angular tuning curve is shown in Fig. 3. The n_o and n_e dispersion relations of Kato [8], provide the best agreement between the calculated curve (Fig. 3, dash–dotted line) and experimental tuning data. The spectral width of the OPG output (measured at the signal wave, λ_s, with a TRIAX 550 Jobin Yvon monochromator) was typically 250 cm^{-1}-away from degeneracy ($\lambda_s \sim 8$ μm, $\lambda_i \sim 13$ μm), and 600 cm^{-1}-near degeneracy at $\lambda_s \sim \lambda_i \sim 10$ μm. These values are on the same order as the calculated OPG acceptance bandwidth for the 7 mm CGA crystal: 150 cm^{-1} (away from degeneracy) and 590 cm^{-1} (at the degeneracy).

Fig. 4 shows the FELIX laser macro-pulse envelope, along with the OPG pulse envelope, both recorded with slow (~100 ns) integrating detectors. The width of the OPG pulse envelope is much narrower since the OPG conversion efficiency is strongly dependent on peak power. At the peak pump intensity of $I_L = 6$ GW/cm^2, and typical OPG internal photon conversion efficiency was 3%.

Taking advantage of the broad tunability of the FELIX laser, we have also performed type-I, SHG experiments in the range of fundamental wavelengths 9–11 μm. In, this case, we have used FELIX pulses with $\tau = 3.9$ ps duration ($\Delta\nu = 3.8$ cm^{-1}) focused to a beam size of $w_0 \approx 400$ μm. The inset to Fig. 5 represents the SHG phase-matching angular curve 10 μm → 5 μm, which is peaked, at $\theta = 32.0°$. This was used to get the missing data point in the OPG tuning curve of Fig. 3 at exactly the OPG degeneracy. SHG phase-matching angles for other wavelengths were found to be $\theta = 32.2°$ ($\lambda_\omega = 9$ μm), and $\theta = 32.46°$ ($\lambda_\omega = 11$ μm). Fig. 5 represents internal SHG conversion efficiency 10 μm → 5 μm as a function of the incident peak intensity, from which one can see that an efficiency as high as 62% can be reached at 380 MW/cm^2.

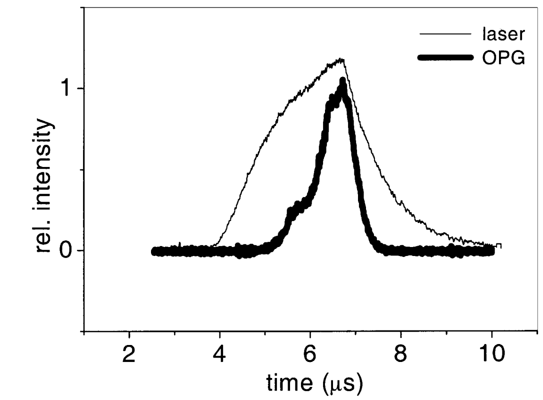

Fig. 4. Pump laser macro-pulse envelope (thin line) and the OPG pulse envelope (thick line).

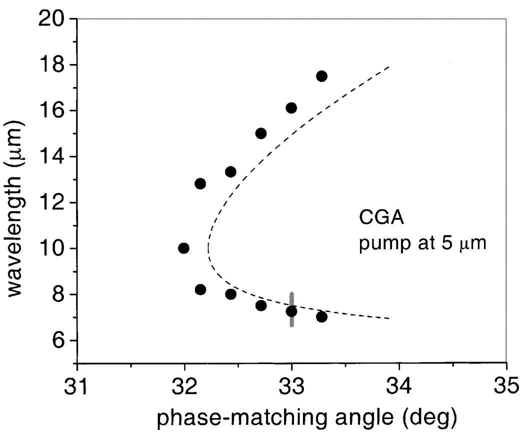

Fig. 3. Angular tuning curves of type-I OPG pumped at $\lambda = 5$ μm. Vertical bar corresponds to the typical FWHM linewidth. Dashed line – calculated from known dispersion relations [8].

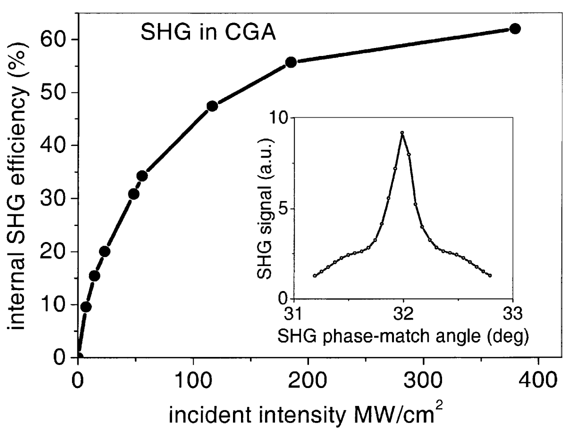

Fig. 5. SHG (10 μm → 5 μm) internal conversion efficiency in CGA, as a function of the incident peak laser intensity. Inset: the SHG phase-matching angular curve.

In conclusion, we have demonstrated the first CGA-based OPG which was pumped by sub-picosecond pulses at 5 μm and which was continuously tunable between 7 and 18 μm. We regard this as a step to achieving a, low-threshold OPO pumped by nanosecond pulses in this crystal. In particular, we have measured experimental OPG/OPO angular tuning curves for CGA, in a large range of wave lengths, which is essential for future OPO designs. We have also demonstrated efficient SHG, with a 62% internal conversion efficiency, using 3.9 ps pulses in, the 9–11 μm range.

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[7] The OPG threshold in this case corresponds to the threshold of detection of the OPG signal which we assumed to be approximately 10^{-5} of the level of the pump power.

[8] K. Kato, Private Communications, 1997 ($n_o^2 = 12.4008 + 2.1603/(\lambda^2 - 2.0617) - 0.00133 \cdot \lambda^2$; $n_e^2 = 13.0079 + 3.2613/(\lambda^2 - 2.8382) - 0.00125 \cdot \lambda^2$).