Quasi-phase-matched gallium arsenide for versatile mid-infrared frequency conversion

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Abstract: Progress in processing low-loss quasi-phase-matched gallium arsenide crystals makes it possible to benefit from their excellent nonlinear properties in practical mid-infrared sources. This paper addresses both crystal growth aspects and the most recent device demonstrations.

OCIS codes: (190.4400) Nonlinear optics, materials; (140.3070) Infrared and far-infrared lasers.

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1. Introduction

Powerful coherent laser sources are needed throughout the mid-infrared region for a number of civilian or defense applications, exploiting either the atmospheric transmission windows, or the fingerprint molecular absorption. Nonlinear optical materials play a key role in this respect

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as they permit the frequency down-conversion of mature high power near-infrared solid-state lasers into the mid-IR, where few direct laser solutions exist.

Gallium arsenide (GaAs) has excellent characteristics for parametric frequency conversion and is potentially one of the most attractive mid-IR nonlinear optical materials. It has a large second-order nonlinear optical coefficient, a wide transparency range, excellent mechanical properties, and a high thermal conductivity [1]. The crystal is optically isotropic precluding birefringent phase-matching, however with the appropriate quasi-phase-matching (QPM) capability, it can be used for numerous nonlinear optical applications. The drawbacks of previous QPM GaAs devices based on diffusion bonding of thin GaAs wafers with periodic orientation reversal [2], have been eliminated by the use of wafer-scale processing techniques to fabricate periodically inverted, or Orientation-Patterned Gallium Arsenide (OP-GaAs), template substrates suited to Hydride Vapor Phase Epitaxy (HVPE) thick-film regrowth. Atmospheric pressure HVPE enables growth rates of about 30 µm/h and low doped layers with excellent optical properties. Careful selection of the growth parameters can preserve the periodic orientation of the template substrate on millimeter thicknesses, thus enabling free space propagation of pump and frequency-converted infrared beams.

After a brief review of the most relevant properties of OP-GaAs crystals for QPM infrared generation, including damage threshold values for various pump pulses, the main fabrication steps are presented below. The emphasis has been made on the control of the optical losses and recent progress is put in perspective with practical devices and applications described in the last section.

2. Material properties

Table 1 compares important properties of OP-GaAs with another well known QPM material, periodically poled lithium niobate (PPLN), and with $ZnGeP_2$ (ZGP), suited to mid-IR frequency conversion by birefringent phase matching.

Table 1. Comparison of Material Properties

	PPLN	ZGP	**OP-GaAs**
Transmission (µm)	0.35-4.5	1-12	**1-16**
Nonlinear coeff. d (pm/V)	27	75	**96**
Thermal conductivity (W/m.K)	5	35	**52**

OP-GaAs thus enables wavelength conversion with a figure of merit d^2/n^3 (where *n is* the refractive index) four times superior to PPLN over a huge wavelength range, and even regardless of the orientation of pump beam polarization owing to the tensor properties of the crystal [3]. Its moderate optical index dispersion in the IR range translates into longer QPM periods as compared to PPLN. Figure 1 gives the dependence of signal and idler wavelengths parametrically generated from various pump wavelengths and grating periods, showing the potential of OP-GaAs for widely tunable sources.

In practice, OP-GaAs crystals are most often anti-reflection coated on their input and output facets. The damage threshold of coated crystals has been measured with several pulsed pumping lasers around 2 µm (a pump wavelength above 1.7 µm is recommended to avoid two-photon absorption). In the nanosecond regime, the 2 J/cm^2 value is similar to the threshold for ZGP [4]. Recent tests carried out with a 1064 nm pumped PPLN source delivering 500 ps pulses at 1.9 µm gave a 0.2 J/cm^2 value, indicating that a square law can reasonably be used to predict damage thresholds down to this pulse duration.

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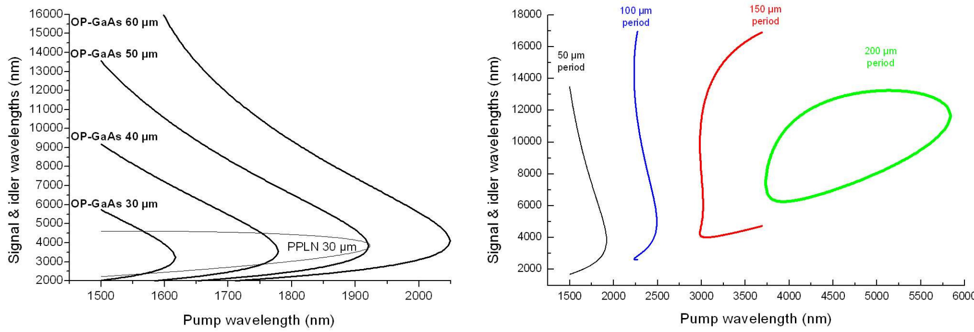

Fig. 1. OP-GaAs tuning curves.

3. OP-GaAs fabrication and characterization

The need for thick structures requires fast epitaxial growth procedures with excellent selectivity. The method of choice for achieving fast growth rates is the HVPE technique.

The epitaxial growth on orientation-patterned semiconductor crystals suitable for QPM conversion first requires templates with modulated crystalline orientation, constituting the seeds for the epitaxial regrowth. In a compound III-V semiconductor with zincblende structure, the orientation reversal consists of the exchange of the atoms between the two sublattices (Ga and As), which is equivalent to a reversal of the III-V bond stacking. The templates can be fabricated on crystals with the two crystal orientations, [001] and [00-1], that can be obtained by an all-epitaxial MBE process enabling sublattice reversal [5,6] or formed by the wafer bonding method [7]. In this method, the authors grow by MOVPE a stop layer and a thin GaAs layer on a 2 inches [001] wafer. This wafer is then bonded with opposite crystal axis orientation on another [001] wafer. Next, the [00-1] side is etched until only a thin [00-1] layer remains on the [001] substrate. Finally, the domain periods and duty cycles are defined by photolithography, and the patterned template is etched to reveal the orientation-patterned gratings (see Fig. 2).

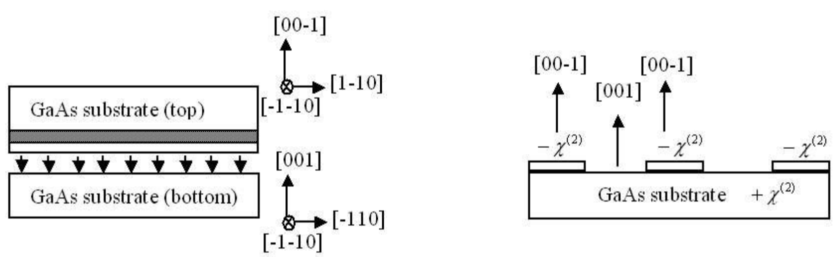

Fig. 2. Template fabrication (light eventually propagates along [-110]).

The second stage of the fabrication consists of the regrowth on the OP-GaAs template to obtain the thick OP-GaAs crystal required for bulk optical pumping. In order to optimize the QPM the crystallographic orientations must be preserved all along the growth process, keeping the duty cycle of the domains equal to the one defined on the template, usually 1:1. Atmospheric pressure HVPE can provide efficient growth of high quality GaAs layers with hundreds of µm thickness at high growth rates (see Fig. 3). The HVPE growth is mainly limited by the adsorption of the gaseous molecules onto the surface, decomposition of the ad-species and desorption processes. HVPE growth is perfectly orientation-selective as it depends on the intrinsic growth anisotropy of the crystal, which can be controlled by the growth temperature and the precursor gas composition [8]. Figure 3 shows some examples of

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OP-GaAs structures. Sample (a) is 20 µm thick and its 3.6 µm period is suited to second harmonic generation of 1.55 µm lasers. Sample (b) is 0.5 mm thick and has a 63 µm period and propagation losses as low as 0.016 cm^{-1}. It has been implemented in a mid-IR source described in the next section. Sample (c) is discussed below.

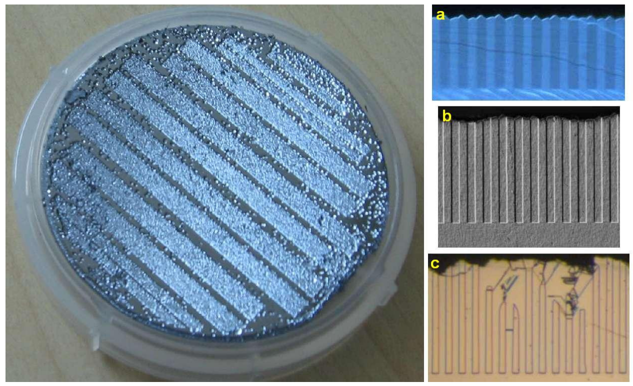

Fig. 3. Left: a 2 inches wafer after the HVPE growth. Right: side views of periodic orientation patterning after revelation by chemical etching (see text).

The typical HVPE growth rate stands close to 30 µm/h for GaAs in an atmospheric HVPE reactor due to the high decomposition rate of the growth precursors. However, the high growth rate results in parasitic nucleation on the reactor walls, which depletes the nutrients, changes the effective gas flow rates, and decreases the crystal growth rate. To maintain a constant vapor composition, growth interruptions formerly had to be made every few hours in earlier experiments for cleaning the reactor. Those interruptions generally appear as bright luminescence planes in cathodoluminescence images such as the one shown in Fig. 4 [9].

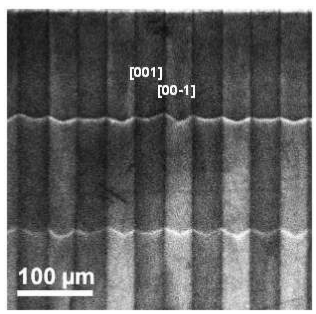

Fig. 4. Panchromatic CL image showing growth interruptions (compared to previous figures, the template is on top).

Recent improvements to the HVPE machine (flux control and sample holder) enabled us to carry out much longer growth cycles, up to 30 hours without cleaning interruptions. This in practice led to an increase in the final OP-GaAs layer thickness from typically 0.5 mm up to more than 1 mm and paves the way toward samples with still lower losses. As an example, Fig. 3(c) shows a very recent 1.2 mm thick sample with a 146 µm period suited to future mid-IR generation from a 3 µm pumping laser and obtained with a single growth interruption.

4. Applications: examples and prospects

During the last decade, non linear frequency conversion has witnessed an unprecedented leap with regard to both versatility and affordability with the emergence of periodically poled LiNbO3 (PPLN) and related ferroelectric materials, allowing efficient QPM wavelength conversion of various continuous wave or pulsed lasers. However, the mid-IR transmission cut-off of such oxide crystals has limited their applicability to wavelengths below about 4.5

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µm. The extension of the QPM technique to GaAs has enabled to fill numerous gaps in terms of applications. After a first proof of concept in 2001 [10], the expected breakthrough occurred in 2004 when the first OPO based on OP-GaAs was demonstrated. In a paper co-authored by Stanford University and Thales TRT, a 2.3 to 9.1 µm tuning range was reported [11]. Since then, the implementation of similar crystals in various configurations has been contemplated and three examples of applications are given below.

As far as continuous wave operation is concerned, local gas sensing with a broadly tunable single-frequency mid-infrared source based on difference frequency generation has been shown [12]. A milliwatt-level output was obtained in the 7.6-8.2 µm range from a 8 W fiber-amplified 1.5 µm diode laser and a 0.5 W Tm-doped fiber laser at 1.9 µm. Figure 5 presents a measured and a computed tuning curves. The excellent agreement between the two curves indicates that the OP-GaAs sample was close to a perfect 50% duty cycle grating.

The available mid-infrared power was appropriate for a methane sensing experiment, but other applications may require more powerful sources. The obvious solution for that is to use similar crystals in a continuous wave optical parametric oscillator (OPO) configuration. The reduction of propagations losses is then the key point to obtain reasonable thresholds and the first demonstration of such a device has indeed been reported very recently [13].

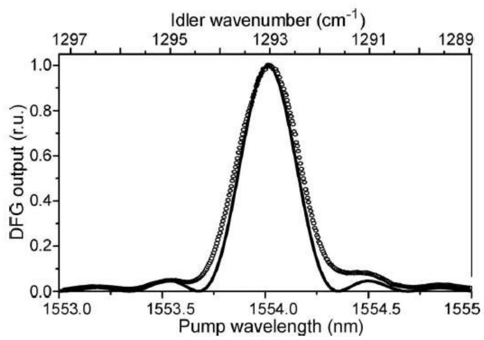

Fig. 5. Measured Π°| and calculated (solid curve) difference frequency generation output versus signal wavelength.

In nanosecond pulsed regime, the 0.02 cm^{-1} loss level is perfectly suited to efficient OPO operation. Applications such as directed infrared countermeasures demanding multi-watt level mid-IR sources thus strongly benefit from the advent of the OP-GaAs technology. To demonstrate its versatility, a compact fiber laser-pumped OPO has thus been built [14].

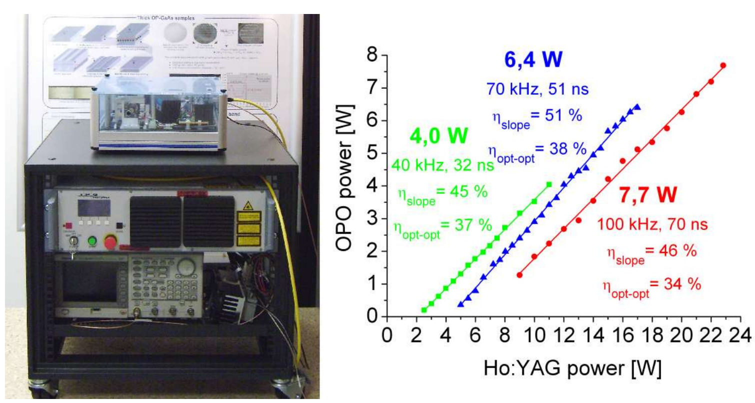

Fig. 6. Left: compact fiber-laser pump OPO module. Right: power scaling of the mid-IR output from the OP-GaAs optical parametric oscillator.

Starting from a remote commercial continuous wave fiber laser, a 25x30x6 cm module integrating a 50 kHz 40 ns Q-switched Ho:YAG laser and an OPO based on OP-GaAs was

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fabricated. A 3 W level output was obtained from this tunable OPO in the 3-5 µm range (signal + idler) with a 53% conversion efficiency and an unprecedented beam quality (M^2 = 1.4). The output beam from the OPO also offers more than 3 W of additional power at the 2.1 µm pump wavelength. This compares very favorably with former designs based on PPLN crystals, both in terms of power and beam quality. Figure 6 shows what the device looks like in addition to more recent results obtained by Hildenbrand et al. using a more powerful pump [15] and demonstrating that a 10 W level mid-IR source is a most probable prospect.

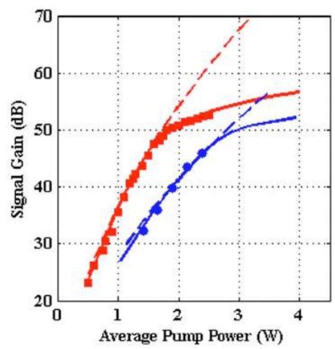

Fig. 7. Gain versus pump average power. Squares (Circles), experiment with a 41-mm-long (32-mm-long) crystal. Solid curves, theoretical fits with SNLO calculation; dotted curves, with non-depleted pump approximation.

Last but not least, both continuous wave and pulsed regimes have been merged in an experiment targeting mid-infrared remote sensing by optical parametric amplification of a distributed feedback quantum cascade laser (QCL) in OP-GaAs [16]. Using a 3 mW DFB QCL at 4.5 µm and less than 3 W of average pump power from a 20 kHz 30 ns Q-switched Ho:YAG laser at 2.1 µm, it was possible to demonstrate a gain up to 53 dB (see Fig. 7). The amplified beam had a very good beam quality (M^2 = 1.3) and its peak power reached 600 W.

5. Conclusion

The fabrication of Orientation-patterned Gallium Arsenide crystals has lately witnessed significant progress and this non-linear optical material has now reached maturity for several applications requiring mid-infrared photons with power or wavelength ranges not easily available through other sources.

OP-GaAs is routinely grown on 2-inch wafers at TRT with a simple process for initial wafer periodic patterning, resulting in the capability to produce 0.5 mm thick samples with several centimeters length. Thanks to the optimization of the growth process, absorption losses have been reduced down to 2%/cm or less and the sample thickness recently increased to the millimeter level. Such material improvements have enabled to demonstrate nanosecond pulsed OPOs with several watts of average output power in the 3 to 5 µm range and optical to optical conversion efficiencies of about 50%. They may also soon permit the realization of milljoule level sources and continuous wave devices with large tunability

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