Advances in quasi phase-matched optical frequency converters

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1. Abstract

Recent advances in the growth of ferroelectric crystals with low optical losses and good structural consistency in particular $KTiOPO_4$ and stoichiomethic $Mg:LiTaO_3$ allowed a vast extension of applicability of quasi-phase-matched frequency converters. In this paper we overview recent advances in structuring these materials enabling high-energy pulse generation, extremely broadband parametric gain, and some unique applications in the quasi phase-matched crystals containing submicron-periodicity 1D structures as well as in 2D-structured crystals.

2. Introduction

This year, 2011, marks the 50-th anniversary of the first experimental demonstration of frequency conversion of what was then the recently invented Ruby laser by employing second harmonic generation in crystal quartz [1]. Only one year later, in 1962, the seminal paper by J Armstrong and co-workers [2] proposed the principle of quasi phase-matching (QPM) as a generic method to increase the efficiency of second order interactions in different classes of materials. The QPM technique provides additional degrees of freedom allowing design of spatial and temporal properties of nonlinear interactions. Over the years, the additional capabilities afforded by the QPM techniques shifted focus in the field of nonlinear materials from the synthesis of new crystals with specific phase-matching and nonlinear properties towards engineering of nonlinear interactions and refining materials which can take advantage of the QPM techniques. This shift was gradually happening over the last twenty years. The result is something which can be thought of as sort of nonlinear optics engineering toolbox which could be added to the existing and developing laser engineering methods.

Regardless of apparent flexibility of the QPM techniques, its success critically depended on the initial material properties. Substantial effort over the last 10 years has been devoted to tailor the properties of the most popular oxide ferroelectrics used for QPM structure fabrication, $KTiOPO_4$ (KTP), $LiNbO_3$ (LN) and $LiTaO_3$ (LT). All these materials had different problems related to electrical properties, induced absorption, photorefraction, etc. These problems currently have been substantially alleviated or solved in principle either by appropriate doping, carefully tailored compositions of solid solutions of isomorphic compounds and/or modification of crystal stoichiometry. In our work we primarily utilize isomorphs of KTP and stoichiometric MgO-doped LT (Mg:SLT) for engineered nonlinear structures due to the high optical damage thresholds, low coercive fields and, importantly, possibility to fabricate periodic structures on the submicrometer scale. As will be shown in the presentation such structures enable realization of novel types of devices having no counterparts in the realm of birefringence phase-matched nonlinear interactions.

3. QPM structures for high energy and broadband parametric gain

High-energy mid-infrared sources are required for number of applications including spectroscopy [3], remote sensing [4], biology and medicine [5], material processing [6], standoff defense systems [7] and possibly others. Recent advances in laser technology made available efficient and robust, high energy, and good beam quality, diode-pumped Q-switched and mode-locked lasers generating around 1 μm. Optical parametric oscillators (OPO), pumped by such lasers are capable to provide high power tunable radiation in spectral regions, not covered by commercially available lasers. Using large-optical aperture PPKTP structures in such OPOs would offer high efficiency and noncritical interaction as well as flexibility in design wavelength. There are two main difficulties arising in scaling up the aperture of periodically-poled KTP (PPKTP): the high voltage needed to overcome the coercive field of the

978-1-4244-6051-9/11/$26.00 ©2011 IEEE

material, and the ferroelectric domain broadening during the growth of large aspect ratio domain structures. To address those problems the $Rb_xK_{1-x}TiOPO_4$ (RKTP) solid solution with small $x<0.01$ was chosen as the best candidate for large aperture ferroelectric domain engineering. This material has similar transmission and nonlinear properties as flux-grown KTP, however it offers orders of magnitude lower ionic conductivity which makes it an excellent choice for fabrication of high aspect ratio domain gratings [8]. Furthermore this material shows improved optical performance compared to that of flux-grown KTP [9]. Periodically poled RKTP up to 5 mm in thickness along the polar direction have been fabricated in this work using short electric field pulse poling technique. The ferroelectric domain grating quality in the fabricated crystals was evaluated employing SHG and OPO experiments. The results attest to remarkably small domain broadening and duty cycle variation in this material and homogeneous performance over the whole aperture. Such large apertures allow scaling of the ns OPO output pulse energy up to 200 mJ in 1.5μm – 3.2 μm region.

Given this energy scaling capability it would be extremely useful, at the same time, to engineer broadband interactions for ultra-broadband parametric gain in the 1 μm-3.5 μm spectral region, where oxide ferroelectrics display very low linear absorption losses. Taking into account large nonlinear coefficients in periodically structured ferroelectrics, such broadband gain would be of interest for tunable synchronously-pumped optical parametric oscillators and optical parametric chirped pulse amplifiers currently under development by different groups with the aim of few-cycle mid-infrared pulse generation.

Achieving the broadest possible parametric gain requires suitable QPM structure design so that the group velocity matching is preserved over the broad spectral range. For the collinear interaction geometry this will happen when the parametric degeneracy point is close to the zero group-velocity dispersion (GVD) point. The fact, that the spectral location of this point is specific for each nonlinear material determined by the crystal dispersion, means that each crystal will have a specific optimum pump wavelength which maximizes the parametric gain bandwidth. The first demonstration of the broadband down-conversion close to the point of zero GVD in a QPM medium was achieved with orientation-patterned GaAs, which displayed a 20 dB parametric gain bandwidth of 39 THz when pumped at around 3.3 μm [10]. Periodically poled PPKTP, PPLN and PPMg:SLT can offer alternative QPM solutions, enabling operation with pump wavelengths in the near infrared. The 10 dB bandwidths of ~150 THz and ~100 THz have been demonstrated for optical parametric generators in PPKTP and PPLN, pumped at 830 nm and 930 nm, respectively [11, 12]. PPKTP apparently has a broader gain and the pump wavelength conveniently situated where Ti:Sapphire amplifiers exhibit maximum gain. In this work we further investigated PPMgSLT and found that this material has the record-broad collinear parametric gain of 180 THz, stretching from 1.1 μm to 3.5 μm (-10 dB level) for the optimum pump wavelength of 860 nm [13]. As in the case of PPKTP this wavelength is readily accessible by standard Ti:Sapphire amplifiers as well as Cr:LiSAF lasers.

4. Novel QPM devices

Engineered QPM structures indeed allow realizing nonlinear interactions which are impossible in birefringence-phase matched media. In this work we will illustrate this by two unique QPM devices, namely, mirrorless OPO (MOPO) based on counter-propagating parametric interaction and a coherent coupling of spatially nondegenerate parametric processes in 2D hexagonal QPM lattice.

Counter-propagating nonlinear interactions allow realization of new types of parametric devices with rather unique spectral and tuning properties. One example here is a mirrorless optical parametric oscillator (MOPO) where distributed feedback, necessary for the oscillation to start, is established by the nonlinear interaction where idler and signal are mutually counter-propagating. The concept of such a device was proposed theoretically by S. Harris in 1966 [14] and demonstrated experimentally in 2007 [15]. The low natural birefringence in all known birefringence phase-matched nonlinear crystals, limits counter-propagating interactions to THz region. Due to starkly different diffraction properties of the interacting waves the MOPO has never been demonstrated in this spectral region.

To circumvent the fundamental difficulty of low natural birefringence we employed PPKTP QPM structure. Due to the large QPM grating wavevector, required for quasi-phase-matching the counter-propagating interactions in near- and mid-infrared, the task of fabricating such structures is far from trivial, considering that the periodicity should be in sub-micrometer range. From this perspective, PPKTP is an excellent material as it has been proven to allow for fabrication of sub-micrometer-periodicity ferroelectric domain gratings with large aspect ratios [16]. The realized MOPO showed thresholds for self-established oscillation low enough so that the device could, in principle, be pumped by a compact Q-switched laser generating sub-nanosecond pulses. The MOPO readily generates narrow-bandwidth, close to transform-limited mid-infrared idler pulses, without any need for spectral filtering or spectral pump conditioning. The device also exhibits quite unusual spectral properties for an optical parametric oscillator, where the phase modulation imposed on the pump wave can be transferred to the signal wave. In this work we demonstrate the compressibility of the signal pulse generated by the MOPO pumped with linearly chirped pump pulses. It would be impossible to realize such spectral properties in OPOs containing standard resonators and co-propagating parametric interactions.

The extension of poling techniques to 2D patterns has enabled purely nonlinear photonic crystals (PNPCs) based on $\chi^{(2)}$ nonlinearities, affording new degrees of freedom in the design and implementation of parametric devices [17]. $\chi^{(2)}$ PNPCs were first demonstrated in bulk PPLN [18] and more recently also in integrated formats [19]. Previous investigations have concentrated almost exclusively on frequency up-conversion processes. The few experimental studies on frequency down-conversion in PNPCs to date have concerned nearly collinear interaction geometries, in which the nonlinear response was dominated by essentially one QPM resonance [20]. On the other hand, some of the most interesting features of PNPC do stem from the interplay of their multiple nonlinear resonances, as proved by the wide gamut of novel multi-port tunable optical outputs observed in 2D up-conversion. The multiple degrees of freedom, giving rise to multiple QPM processes in parametric down-conversion are, in fact, detrimental because they give rise to poorly spatially and spectrally structured output. However, by properly designing nonlinear interaction geometry in 2D structure, it is possible to realize parametric generation where only two coherently-coupled down-conversion channels are participating, stemming from the interplay of two lowest-order QPM resonances of the 2D lattice. The experimentally observed nonlinear response, characterized by dual-beam signal outputs which share identical spectral properties, is the signature of the coherent exchange between two QPM processes, achieved through a common idler wave. Such OPG configuration has no counterpart in traditional 1D geometries and represents a first example for the increased efficiency and spectral engineering capabilities potentially affordable with PNPC structures in a variety of classical and quantum optics applications based on frequency down-conversion

5. References

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