7487-14, Session 4

Broadly tunable LiInSe2 optical parametric oscillator pumped by a Nd:YAG laser

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Since there are no solid state lasers in the mid-IR above 3 µm, this spectral range can be continuously covered only by nonlinear frequency down-conversion, preferably pumping by widely-used high-power diode-pumped laser systems, such as Nd:YAG, operating in the nanosecond regime. Thus, two-photon absorption (TPA) at the pump wavelength (chalcogenides) and limited transparency above 4 µm (oxides) are the two basic limitations for the nonlinear crystals applied. LiInSe2 is one of the few (only 5) non-oxide nonlinear crystals whose band-gap (2.86 eV) and transparency allowed in the past optical parametric oscillator (OPO) operation in the mid-IR without TPA for a pump wavelength of 1064 nm. Higher damage resistivity and the roughly 3 times higher thermal conductivity are the main advantages of this new orthorhombic crystal in comparison to the conventional AgGaS2. However, the first such OPO demonstration with LiInSe2 was limited to the 3.3-3.78 µm spectral range for the idler, with a maximum energy of 92 µJ at 10 Hz. In this work we employed a 5×6.5×17.6 mm3 sample of LiInSe2, grown by the vertical two-zone furnace Bridgman technique. It was cut at 41.6° in the x-y plane for type-II (eoe) interaction. Both faces were AR-coated with a single layer for the pump and signal wavelengths. The singly resonant two-mirror OPO with pump recycling was pumped by 14 ns long pump pulses from a diode-pumped and electro-optically Q-switched Nd:YAG laser, optimized for a repetition rate of 100 Hz. Only the idler was effectively out-coupled and characterized. At normal incidence, its wavelength (6.56 µm) was only slightly longer that the calculated one (6.45 µm). With an 18.5-mm-long cavity, the threshold amounted to ~6 mJ of incident pump energy (peak on-axial intensity of ~7.5 MW/cm2). The maximum idler energy obtained at 15 mJ of pump, amounted to 282 µJ, equivalent to an average power of ~28 mW. Broadly tunable operation, from 4.7 to 7.6 µm, was achieved with a slightly longer (20.5 mm) cavity by rotating the nonlinear crystal.

7487-15, Session 4

Design of near-infrared dyes for nonlinear optics: towards optical limiting applications at telecommunication wavelengths

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The rapid development of frequency-tunable pulsed lasers up to telecommunication wavelengths (1400-1600 nm) led to the design of new materials for nonlinear absorption in this spectral range. In this context, two families of NIR dyes, namely heptamethine cyanine[1] and aza-borondipyrromethene (aza-bodipy)[2] dyes were studied. In both cases they show significant two-photon cross section in the 1400-1600 nm spectral range and display good optical power limiting (OPL) properties. OPL curves were interpreted on the basis of two-photon absorption (TPA) followed by excited state absorption (ESA). Finally these systems have several relevant properties like nonlinear absorption properties, gram scale synthesis and high solubility. In addition, they could be functionalized on several sites which open the way to numerous practical applications in biology, solid-state optical limiting and signal processing.

[1] P.-A. Bouit, G. Wetzel, G. Berginc, B. Loiseaux, L. Toupet, P. Feneyrou, Y. Bretonnière, K. Kamada, O. Maury, C. Andraud, Chem. Mat. 2007, 19, 5325-5335.

[2] P.-A. Bouit, K. Kamada, P. Feneyrou, G. Berginc, L. Toupet, O. Maury, C. Andraud, Adv. Mater. 2009, sous presse.

7487-17, Session 4

Progress in ZnGeP2 and AgGaS2 crystal growth, first results on difference-frequency generation and optical parametric oscillation

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Standard oxide nonlinear materials (LiNbO3, KTP,) are widely used to implement efficient optical parametric oscillators (OPOs) in the mid-IR for many applications ranging from infrared countermeasures to trace gas monitoring and remote sensing. However, above 4.5 µm, these materials exhibit strong bulk absorption and thus become unsuitable. It is then required to implement OPOs based on less developed nonlinear materials. Among them, the chalcopyrite structure crystals like AgGaS2, ZnGeP2, AgGaSe2 or CdGeAs2 are the most popular ones thanks to of their properties (high nonlinearity, IR transparency, birefringence phase-matching schemes,).

Here, we present our last results on two of them: AgGaS2 (AGS) and ZnGeP2 (ZGP). AGS is very interesting because it can be pumped by a classic 1.064 µm diode pumped Nd:YAG laser to reach the mid IR up to 10 or 12 µm. Indeed, its transparency range is very broad from 0.5 to almost 12 µm and it can be appropriately phase-matched for such a conversion. For higher power applications, ZGP is certainly the most popular. It has a high nonlinearity and, thanks to its good thermal properties, it has a high laser induced damage threshold.

AGS and ZGP were grown by the vertical Bridgman technique in pyrolitic crucibles inside a vacuum soldered high quality quartz ampoule. Both were grown using (001) oriented seeds. The growth rate was 0.65 mm·h 1 for ZGP and 0.4 mm·h 1 for AGS.

The main problem with AGS crystals is the quite poor transparency of the as-grown samples because of micro precipitates of gallium sulfide. To eliminate those inclusions, vacuum annealings are generally carried out. In the present work, a method combining static and dynamic vacuum stage is proposed. Thus, highly transparent AGS samples were obtained. Concerning ZGP, the as-grown crystal is already quite transparent. A simple static vacuum annealing at 500 °C is necessary to obtain high transparency. Good optical quality single-crystal samples with size up to 5×5×20 mm3 were then cut and polish from our ingots to carry out nonlinear optics experiments.

Preliminary difference frequency generation (DFG) experiments are carried out on AGS: the 1.064-µm beam from a pulsed Nd:YVO4 laser is split in two parts, the first part is directly sent on the 11.6 mm long AGS crystal while the other part is used to pump a PPLN OPO emitting a 4-µm idler beam which is subsequently sent on the AGS crystal. The obtained DFG energy at 1.45 µm is consistent at a factor two with results provided by a simple model, where walk-off is neglected. This confirms the high quality of our sample.

Optical parametric oscillation is demonstrated with ZGP. The OPO cavity consists of a 20-mm ZGP crystal placed inside a singly resonant cavity. The ZGP OPO is pumped at 2.2 µm by 15-ns pulses and emits a signal radiation at 3.6 µm. Similar experiments are carried out with a commercial crystal (Moltech) The two crystals provide very comparable results, which validates the high optical quality of our sample for OPO operation.