Parametric down-conversion devices: the coverage of the mid-infrared spectral range by solid-state laser sources

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Second harmonic generation (SHG; also called frequency doubling) is a nonlinear optical process, in which photons interacting with a nonlinear material (noncentrosymmetric crystal) are effectively "combined" to form new photons with twice the energy, and therefore twice the frequency and half the wavelength of the initial photons. SHG was the first nonlinear optical process demonstrated in 1961 by Franken et al. at the University of Michigan, just one year after the discovery of the ruby laser by Maiman at Hughes Research Laboratories. It was this solid-state laser that was used in the first SHG experiment with quartz as the nonlinear optical crystal.

Soon afterwards the theory of the more general optical parametric three-wave interactions was developed. The generation of a photon at frequency ω_1 when photons at frequencies ω_3 and ω_2 are incident on a crystal with an appreciable second-order susceptibility $\chi^{(2)}$, such that $\omega_1 = \omega_3 - \omega_2$ (assuming $\omega_3 > \omega_2$), is called difference-frequency generation (DFG). In order for energy conservation to hold, this additionally implies that, for every photon generated at the difference frequency ω_1, a photon at ω_2 must also be created, while a photon at the higher frequency ω_3 must be annihilated. In addition, for the process to occur with an appreciable efficiency of frequency conversion, phase-matching must occur, so that $k_1 = k_3 - k_2$. In many situations, the field at ω_3 is an intense pump field, while the field at ω_2 is a weak signal field. DFG yields amplification of the ω_2 field (along with generation of another field at ω_1, commonly called the idler field). Thus, this process is termed parametric amplification. If this process occurs within an optical cavity with resonance frequency ω_2, the device is called an Optical Parametric Oscillator, or OPO. If the cavity has modes at both ω_2 and ω_1, the device is a Doubly Resonant OPO. DFG can also occur in the absence of an applied signal field; under these circumstances, it is called Spontaneous Parametric Fluorescence. In this case, the frequencies of the generated photons are determined by the phase-matching condition for the particular crystal orientation used.

The first OPO, based on $LiNbO_3$, was demonstrated in 1965 by Giordmaine et al. at Bell Telephone Laboratories. It utilized the high power of a short pulse (nanosecond) Nd:$CaWO_4$ laser operating in the Q-switched (giant-pulse) regime. Continuous-wave (cw) OPOs were realized in the following years: at first doubly resonant to reduce the pump threshold and later singly resonant - to avoid the instabilities and complex tuning. Another option to reduce the threshold in cw OPOs is intracavity pumping. While OPO is a feasible concept from nanosecond time scale to cw operation, the parametric down-conversion of ultrashort (picosecond and femtosecond) laser pulses can be realized in “single-pass” traveling-wave schemes, called Optical Parametric Generator (OPG) when the process is initiated by the spontaneous parametric fluorescence and Optical Parametric Amplifier (OPA) when a weak signal wave is amplified. OPG presents the simplest set-up but offers less control on the output properties and requires high pump intensities which are often close to the nonlinear crystal damage threshold. OPA is really not different from DFG except that one has in mind that the input wave at ω_2 is weak and is to be strongly amplified. DFG is applicable on any time scale (from femtosecond to cw) but requires two wavelengths and the conversion efficiency to the idler wave is rather low. OPO, OPA and OPG can produce high conversion efficiencies leading to depletion of the pump power. In these devices, Q-switched nanosecond lasers or ultrafast amplifiers are normally used for high-energy parametric down-conversion of nanosecond, picosecond and femtosecond pulses while the repetition rate varies from a few Hz up to about 1 MHz. Low energy picosecond and femtosecond pulses from mode-locked laser oscillators can be down-converted employing an OPO cavity with a length matched to the repetition rate of the pump source (from ~50 MHz to ~1 GHz). These devices are called Synchronously Pumped OPO (SPOPO).

All the above parametric down-conversion devices have their laser analogues and there are many similarities with the operation of a laser. However, there are also essential differences: (i) parametric devices require coherent pump sources, i.e. lasers, and parametric fluorescence and gain occur in the direction of the pump beam, (ii) their wavelength is determined and tuning is realized by changing the phase-matching conditions, the spectral gain-bandwidth is defined by the dispersive properties of the nonlinear crystal and its length, (iii) no heat is deposited in a nonlinear crystal unless there is some residual absorption at some of the three wavelengths involved, (iv) there is no energy storage in the nonlinear crystal because no levels are occupied and the dynamics is completely different due to the instantaneous nature of the nonlinear process: as a consequence the temporal profile of the output is determined to a great extent by the temporal profile of the pump and seed (signal) radiation as well as by the dispersive properties of the nonlinear crystal.

The tuning capability of parametric down-conversion devices is a major advantage because it can be not only much broader than any tunable laser but can cover wavelength ranges inaccessible to (solid-state) lasers.

Since the pioneering of Franken et al. on SHG, parametric down-conversion and up-conversion (i.e. sum-frequency generation with SHG being a particular case) processes as well as combinations of them have been widely used to fill in gaps in the optical spectrum where lasers do not exist or certain operational regimes are impossible: from the vacuum ultraviolet (~150 nm) to the deep mid-infrared. The main progress took place in the last two decades following the advancement in near-infrared solid-state laser technology and nonlinear optical crystals. Parametric down-conversion is more difficult to realize than up-conversion because of the higher intensities required to reach the threshold for parametric amplification but it offers wider wavelength tunability.

In this presentation I will review the development of parametric down-conversion devices operating in the mid-infrared: from 3 μm to about 15 μm, the upper wavelength limit set basically by the transparency cut-off of the existing inorganic nonlinear crystals (maximum 20-30 μm). This selection is motivated by the absence of solid-state lasers in this wavelength range. Indeed, the upper limit of practical solid-state lasers, such as Er^{3+} or Cr^{2+}, extends to about 3 μm. Other transitions in rare earth or transition metal ions at longer wavelengths exist but temperature quenching of the mid-infrared fluorescence (e.g. in Fe^{2+}) or the lack of suitable pump sources represent basic limitations. Thus operation at low temperature using pulsed pumping is required in most cases and it is difficult to imagine that all temporal regimes (from cw down to femtosecond pulses) will ever be realized with such systems which often exhibit only narrow band emission (e.g. Nd^{3+} or Pr^{3+}).

The vital element in any frequency-conversion process is the nonlinear optical crystal and this represents one of main challenges for the development of parametric down-conversion devices in the mid-infrared. Indeed, the performance of oxide based crystals is affected by multi-phonon absorption starting in the best case from about 4 μm and thus non-oxide materials have to used, such as arsenides, phosphides, sulphides, selenides or tellurides. In contrast with the oxides, which can be grown by well mastered and harmless hydrothermal, flux or Czochralski methods, the more complex Bridgman-Stockbarger growth technique in sealed (high atmosphere) ampoules, with volatile and chemically reactive starting components, is the only method used to produce large size single domain non-oxide crystals, and this certainly hampered their development all the more that special post-growth treatments are needed to restore stoichiometry and improve their optical quality. As a matter of fact such materials exhibit more defects and the residual losses (absorption and scattering) are more than one order of magnitude larger than in the best oxide crystals. Non-oxide crystals have normally smaller band-gap which is related also to their higher nonlinear coefficients. However, the figure of merit is reduced by their higher index of refraction. The band-gap value presents a serious limitation in all temporal regimes besides cw because of the onset of two-photon absorption at the shortest (pump) wavelength which normally corresponds to the highest intensity. Thus, e.g. only few materials can be used for direct conversion of femtosecond pulses from Ti:sapphire laser systems operating near 800 nm and cascaded parametric down-conversion schemes employing oxides in the first stage are required. Even for wavelengths in the 1-μm spectral range, where powerful Nd^{3+} or Yb^{3+} nanosecond and picosecond laser systems exist, limitations related to the residual loss, nonlinearity, phase-matching, thermo-mechanical properties, or simply growth and availability, do not allow one to generate high single pulse energies and/or average powers in the mid-infrared. In such cases pumping at longer wavelengths (e.g. by Er^{3+} lasers near 1.5 μm, Tm^{3+} and Ho^{3+} lasers near 2 μm or even Er^{3+} lasers near 2.9 μm) has to be used. An overview of the available mid-infrared nonlinear optical materials, emphasizing new developments like larger band-gap, engineered (mixed) crystals and quasi-phase-matched materials, will be presented.

Parametric down-conversion in the cw regime offers significantly smaller spectral linewidths with single-frequency operation possible, better frequency stability, and continuous scan possibilities at the expense, however, of lower conversion efficiency. Such parametric devices are based mainly on DFG, cw OPO operation above 3 μm has not been demonstrated with a non-oxide nonlinear material, yet. Nanosecond OPOs are interesting for military applications (countermeasures in the 3-5 μm spectral range) for jamming heat-seeking missiles. Such applications rely on the high average power (normally achieved at multi-kHz repetition rates) as well as high single pulses energy (at lower repetition rates). Another important application of nanoseconds mid-infrared OPOs is in the field of medicine – for minimally invasive surgery by selective tissue ablation. Short and ultrashort pulses in the mid-infrared possess interesting potential for processing of organic materials like resonant infrared pulsed laser deposition. Picosecond and femtosecond pulses in the mid-infrared are needed for the study of ultrafast dynamics in chemical reactions, molecular vibrations, and the application of infrared spectroscopy to problems in solid-state physics. It should be noted that the parametric gain decreases with the idler wavelength – thus above 20 μm mainly DFG with pulsed radiation is applied. The state-of-the-art of parametric down-conversion devices based on non-oxide nonlinear materials will be reviewed for all these time formats. Yet, in different parametric down-conversion device configurations and operating regimes in the mid-infrared, major limitations still exist and important challenges still remain, requiring the continued search for alternative new nonlinear materials, laser pump sources and innovative design concepts.

The author is coordinating at present the European Community project MIRSURG (www.mirsurg.eu) devoted to the development of mid-infrared OPOs for medical applications. He acknowledges financial support from the European Community's Seventh Framework Programme FP7/2007-2011 under grant agreement n°224042.