Selected Recent Advances in Optical Parametric Oscillators

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Introduction

This report will first review the basic concepts behind optical parametric oscillation followed by selected recent topics related to optical parametric oscillators (OPOs) that may be of interest to the Phys 555 class, such as: novel quasi-phase matched materials for OPOs, semiconductor-based OPOs, spectroscopic applications of OPOs, and entangled cavity OPOs.

The fundamentals of optical parametric generators were first laid out by Armstrong, et al.[1] and Kroll [2] in 1962. The first demonstration of the operation of an OPO by Giordmaine and Miller dates back to 1965 [3]. Early experience with these devices was not satisfactory due to the unavailability of suitable nonlinear optical materials and poor pump beam characteristics.

Optical parametric oscillation is a nonlinear process in which a pump beam is converted into two lower energy beams known as the signal beam ($\omega_s$) and the idler ($\omega_i$) beam. As the pump passes through the nonlinear medium, the spontaneous breakup of pump photons occurs through spontaneous parametric emission. This arises from mixing of the zero point flux of the electromagnetic field at $\omega_s$ and $\omega_i$ with the incoming pump photons, through the nonlinear polarization. The field of the intense pump beam mixes with a signal field and the idler field mixes back with the pump to produce additional signal. This regenerated signal remixes with the pump to produce more idler. This process continues until power is transferred from the pump to the signal and idler field. The wavelengths of the three beams must satisfy: $1/\lambda_p = 1/\lambda_s + 1/\lambda_i$ or equivalently $\omega_p = \omega_s + \omega_i$ illustrating conservation of energy. For a more detailed explanation of the principles of parametric generation and amplification, the reader is referred to [8].

OPOs may be operated continuously, with Q-switched pulses, or with mode-locked lasers. In the latter case, synchronous pumping is required where the length of the OPO cavity is adjusted so that the cavity round-trip frequency matches the repetition rate of the pump, or is an integer fraction of that. Since two downconverted waves (the signal and the idler) are generated within an OPO, there is the choice of resonating one of these two waves (singly resonant oscillator (SRO)), or to resonate both simultaneously (doubly resonant oscillator (DRO)). The DRO results in two intense fields within the nonlinear crystal at the same time, thus reducing the threshold as compared to the SRO. However, stability and smooth tuning are compromised in a DRO. An intra-cavity OPO may be employed to alleviate the reduced threshold in the SRO where the nonlinear crystal is situated inside the cavity of the pump laser, allowing the large circulating laser field to be accessed and hence reducing the external pump threshold.

A parametric oscillator is similar to a laser, but based on optical gain from parametric amplification rather than on stimulated emission. Although OPOs are in many respects similar to lasers, there are also several of important differences. For instance, the parametric amplification process requires phase matching to be efficient. Also, no heat is deposited in the nonlinear crystal, assuming that there is no parasitic absorption. Energy is not stored in the nonlinear crystal for an OPO as in a laser, thus, gain is present only as long as the pump wave is there, and pump fluctuations directly affect the signal power.

In current OPO devices, the generated wavelengths are limited by the availability of nonlinear materials that can simultaneously satisfy the phase-matching, energy conservation and optical transmission conditions. Given a material that meets these criteria, a periodic reversal of the sign of
the nonlinear susceptibility can be introduced every coherence length (typically achieved by electric field poling), and the generated fields do not cancel. This quasi-phase matching (QPM) principle allows a cumulative growth of the generated field. For a given crystal temperature and propagation direction, phase matching may be satisfied only for one combination of frequencies. By changing the temperature, and thus the refractive index of the crystal, the frequency of the output from the crystal may be tuned. Tuning by angle results in restricted angular acceptance and walk-off, which restricts the interaction length. In the case of QPM, additional tuning flexibility is also possible using several uniform gratings on a single crystal, thus, it is possible to achieve frequency tuning through simple mechanical translation of the crystal.

**Advances in QPM Materials for OPO Applications**

Although LiNbO$_3$ is well established as a good QPM material, including a desirable $d_{eff}$, it also exhibits several undesirable properties, including thermal lensing, susceptibility to optical damage, and photorefractive degradation. The last of these precludes its room temperature operation. Despite having a $d_{33}$ coefficient approximately 60% that of periodically poled (PP) LiNbO$_3$, PP arsenates and phosphates of the potassium titanyl phosphate (KTP) type lack the photorefractive effects and have a reduced susceptibility to thermal lensing, making RbTiOAsO$_4$ (RTA) and KTiOAsO$_4$ (KTA) attractive alternatives for the development of practical OPOs [5,6].

Additionally, much attention has been directed toward QPM, PP, magnesium oxide doped, LiNbO$_3$ (MgO:PPLN), due to its much higher damage thresholds, lower coercive field, larger nonlinear susceptibility, and wider transparency range as compared to non-doped LN [7]. These materials can now be grown as thick as 3 mm [8] and as long as 70 mm [9]. A high energy (total output energy: 22 mJ, pump energy: 46 mJ), room temperature OPO has been developed using this material with a pump threshold of ~3 mJ, and total slope efficiency of 51% [8]. Another MgO:PPLN OPO, pumped by a 50 W ytterbium laser, produced over 10 W of near diffraction limited output at 3 µm [10].

Tandem nonlinear crystals have become common in OPO research over the past several years. Although schematically (see Fig. 1) the tandem OPO is represented as consisting of two separate crystals, in practice, only a single monolithic crystal is used, and the conversion takes place in two differently QPM gratings [11]. In this scenario, pump pulses are converted in the first grating to non-resonant signal pulses and resonant idler pulses. In the second grating, the non-resonant signal pulse acts as a second pump for a further parametric interaction, in which the idler pulses remain resonant and a second, non-resonant idler (referred to in Fig. 1 as “idler(2)” ) is generated. Hence, the idler pulses experience gain twice with the possibility (under strong pumping) to obtain more than 100% quantum efficiency in the conversion from the pump to the idler [11].

![Fig. 1: Tandem nonlinear QPM grating [11]](image-url)
### Semiconductor-based OPOs

Recently, semiconductors have been exploited for use in OPOs due to their excellent characteristics for parametric frequency conversion. For example, GaAs is potentially one of the most attractive mid-IR nonlinear optical materials. It has an extremely large second order nonlinear coefficient of $d_{14} = 5 \times d_{33} (\text{LiNbO}_3) = 94 \text{ pm/V}$ (near 4 $\mu$m), a wide transparency range of 0.9 - 17 $\mu$m, and high thermal conductivity [12]. The drawbacks of previous QPM-GaAs devices (based on diffusion bonding of GaAs wafers) were eliminated in recent work by combining two crystal growth techniques known as molecular beam epitaxy and hydride vapor phase epitaxy [12].

GaAs can be pumped by near-IR lasers to achieve tunability in the entire region of common molecules (2 - 17 $\mu$m) [12]. Powerful IR light that aircraft can potentially employ to divert heat-seeking missiles can be generated by a GaAs OPO. Moreover, it can potentially generate the far-IR light suitable for terahertz imaging at airport security as well as trace gas detection [13].

Vodopyanov, et al., reported the first demonstration of an OPO based on GaAs and, in fact, the first demonstration of an OPO based on an optically isotropic $\chi^{(2)}$ material [12,13]. This OPO is based on a 0.5 x 5 x 11 mm$^3$ GaAs sample, with a domain reversal period of 61.2 $\mu$m. The pump source for the GaAs OPO was the signal wave output of a PPLN OPO, tunable over the range 1.8 - 2 $\mu$m. The PPLN OPO was pumped by a Q-switched Nd:YAG laser. Tuning either the PPLN OPO or the temperature of the GaAs crystal, allows the mid-IR output to be tuned between 2.28 - 9.14 $\mu$m, which is limited only by the spectral range of the OPO mirrors. The pump threshold of the GaAs OPO is 16 mJ, and the photon conversion slope efficiency reaches 54% [12]. The highest average power (0.45 W) and efficiency (20% slope) achieved to date from a GaAs OPO was obtained by Schunemann, et al. [14].

Another semiconductor with a very high nonlinear optical coefficient, ZnGeP$_2$ ($d_{\text{eff}} = 75 \text{ pm/V}$), has been utilized in an efficient, high average power (> 10 W), angle tuned, SRO OPO. The output was continuously tunable from 3.8 -12.4 $\mu$m with a pump threshold less than 1 mJ over the entire 4 - 12 $\mu$m range of the output, and the quantum conversion efficiency reached 35%. With a single intra-cavity etalon, the linewidth was narrowed to 0.5 cm$^{-1}$ [15].

### Spectroscopic Applications of OPOs

OPOs are useful tunable sources of narrowband coherent light for high resolution spectroscopy and related sensing applications due to their ability to possess a narrow linewidth, wide spectral coverage, good stability, and broad tunability. CW OPOs have now been established as practical and efficient sources of broadly tunable mid-IR radiation for monitoring trace amounts of volatile organic compounds in biology, environmental analysis, and medicine [16].

As a first example, an all solid state, transportable, photoacoustic spectrometer for highly sensitive mid-IR trace gas detection was developed using a PPLN-based CW OPO [16]. The frequency tuning qualities of the OPO allow reliable scan over gas absorption structures. A detection limit of 110 ppt for ethane was achieved [16]. In a similar manner, the absorption spectrum of CO$_2$ was recorded [18]. In another experiment, an OPO based on PP-RbTiOAsO$_4$ was used to detect methane
gas in concentrations as low as 30 ppm.m [18]. Low pressure absorption lines of N$_2$O, were observed with a pulsed, entangled cavity, OPO. This source runs at a high repetition rate (>10 kHz) with a low threshold of oscillation (< 8 mJ), is tunable over 5 cm$^{-1}$, and displays single frequency Fourier transformed limited operation with a linewidth of 0.005 cm$^{-1}$ [19]. The details of the entangled cavity OPO will be discussed more thoroughly in the following section of this report.

To achieve a narrow linewidth, injection seeding by a low-power tunable coherent source, such as a tunable diode laser, is often employed to achieve single mode operation of a nanosecond pulsed OPO. In many narrowband pulsed OPO designs, injection seeding is preferred to the traditional intracavity tuning using a grating or etalon, which is damage prone owing to optical losses that cause the operating threshold of the OPO to approach the damage threshold of the NLO medium. However, continuous single mode tuning of a pulsed OPO usually requires active feedback control of the OPO cavity length. A design that comprises a self-adaptive, injection seeded OPO cavity that is automatically resonant and tunable with no mechanical control in the OPO cavity was developed. One regular cavity reflector was replaced with a phase conjugate reflector, with its wavelength selectivity controlled adaptively by the same light that is used to injection seed the OPO [20].

**Entangled Cavity OPOs**

PPLN has been utilized as a source of entangled photon pairs [10,19]. Polarization entangled states can be generated using one dimensional PPLN, and frequency entangled states using two dimensional PPLN [21].

GaAs, with its high-symmetry nonlinear optical susceptibility tensor (the only non-zero elements are $d_{14} = d_{25} = d_{36}$), offers possibilities for parametric amplification of circularly polarized or even non-polarized light. Thus, randomly polarized reference frames can be generated to create entangled photon pairs which can be used for quantum cryptography [12].

Entangled-cavity OPOs (ECOPOs) have been developed that are single mode with high output power [19]. In such a device the signal and idler fields experience two different cavities (see Fig. 2) and thus display different free spectral ranges. The optical length of the signal and idler cavities can be engineered so that, a single exact coincidence occurs between signal and idler modes on which the OPO oscillates and the mode overlap over partial coincidences is too small to allow other pairs of modes to oscillate. The ECOPO is a unique device that displays both single frequency operation and a low threshold of oscillation, due to the double resonance effect [19].

![Fig.2: Schematic of an entangled cavity OPO [19]](image-url)
REFERENCES