Advances in the spectral coverage of tunable continuous-wave Optical Parametric Oscillators

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ABSTRACT

We discuss principles, design challenges, performance highlights as well as current limitations of state-of-the-art widely tunable continuous-wave optical parametric oscillators sought to be practical for implementation as turn-key systems. Employing a flexible two-stage design concept that can be adapted to several single-frequency laser pump sources, we demonstrate how a wavelength range from 450 nm up to 3500 nm can be covered almost seamlessly. Emerging key-applications in the realm of quantum technology, like fundamental studies of novel color centers in diamond, are presented in an illustrative manner.

Keywords: Tunable laser, optical parametric oscillator, quantum emitter, color center

1. INTRODUCTION

Light sources based on optical parametric oscillator (OPO) technology offer a remarkable versatility of operational wavelengths compared to conventional lasers¹. This is because the OPO principle relies on a process referred to as parametric conversion in a nonlinear optical material rather than on stimulated emission in a suitable laser gain medium. As illustrated in Figure 1, the OPO process can be perceived as splitting of an incoming pump photon of high energy into two photons of lower energy, the latter usually referred to as signal and idler photons, respectively. It is subject to the conservation principles of photon energy and photon momentum (phase-matching condition), but otherwise, at least in theory, not limited by fundamental restrictions. In other words, as long as the two conditions are met, the operational wavelength of the signal (respectively the idler) can be freely chosen.



Figure 1. Left: Schematics of lasing in a four-level gain medium. The process is based on population inversion (maintained, e.g. by optical pumping) and stimulated emission. Right: Schematics of optical parametric conversion in a nonlinear medium. In an intuitive picture, pump laser photons are split into pairs of photons of lower energy, commonly referred to as signal (ω_s, k_s) and idler photons (ω_i, k_i). This so-called three-wave mixing of pump, signal, and idler waves is intimately linked to the second order nonlinearity of the employed nonlinear crystal. The process is subject to conversion of photon energy ($\omega_p = \omega_s + \omega_i$) and photon momentum ($k_p = k_s + k_i$), but otherwise the operational wavelengths of a signal/idler-pair can be in principle freely chosen and continuously tuned spectrally.



Figure 2. Schematic beam path inside the first ever commercially available cw OPO design covering the visible spectral range. In a first step (OPO), a 532 nm laser pumps a nonlinear crystal to generate signal (900-1050 nm) and idler (1080-1300 nm) photons in the near infrared spectral range. Wavelength selection using a two-stage etalon process and subsequent second harmonic generation (SHG) converts either signal or idler photons into the visible range of the spectrum. The green arrows depict the pump laser beam, red arrows depict the signal/idler beams (arbitrary assignment), blue arrows depict the beam path of frequency-doubled signal/idler. (b) Output power vs wavelength.

While the OPO concept has been experimentally demonstrated very soon after the invention of the laser itself², practicable devices have been slow to come, which is especially true for systems operating in continuous wave mode (cw OPOs). Only quite recently, realization of widely tunable cw OPOs has been spurred by the emergence of novel types of nonlinear crystals, and, on the other hand, the increasing availability of suitable high performance pump lasers. As we show in the following, cw OPOs have thereby matured into widely tunable sources of coherent radiation with unprecedented wavelength coverage and excellent operational characteristics³ - enabling experiments that would have been otherwise hampered by the technical complexity of suitable alternatives or even the lack thereof⁴.

2. WIDELY TUNABLE CW OPOS: PRACTICAL DESIGN CONSIDERATIONS

We note that OPOs, unlike lasers, require coherent light for pumping, and that by the nature of the parametric process any of the generated output wavelengths will be longer than the pump wavelength. As a consequence, cw OPO devices sought to operate across the visible spectral range do either require cw ultraviolet (UV) pump light sources or, alternatively, need to employ an additional frequency conversion stage. Despite the appealing simplicity that pumping at UV wavelengths would offer, so far only two-stage implementations have been proven practical, combining long wavelength pumping with second-harmonic conversion (SHG) of the primary OPO output. Thereby, requirements on pump laser light sources are stringent in terms of preferential single mode operation, noise characteristics, beam quality, and beam pointing stability. As of today, we resort to either high performance diode pumped solid state (DPSS) lasers (for lower pump powers), or to high performance fiber laser based solutions (for higher pump powers).

In order to achieve sufficient conversion efficiency, the nonlinear medium is operated inside a resonator cavity (typically a bow-tie cavity, named after its characteristic shape), to ensure multiple paths increase the gain at each round trip. It appears fair to say that only so-called "singly resonant" designs are feasible for operationally stable turnkey systems: For a particular operational wavelength of the entire system, the primary OPO cavity is operated "on resonance" at either a particular signal wavelength or a particular idler wavelength. While keeping one wave (e.g. signal) resonantly circulating inside the OPO cavity, its counterpart (e.g. idler) can be extracted for subsequent wavelength conversion. In practice, the effective OPO cavity length needs to be actively stabilized (to a multiple integer of the circulated wavelength). To this end, a Pound-Drever-Hall (PDH) or a side-fringe locking stabilization scheme can be utilized, in conjunction with a piezo element mounted cavity mirror that allows control of the effective OPO cavity length.



Figure 3. Schematic beam path inside a cw OPO system tailored for gap-free tuning in the visible range at optimized output power. A 780 nm laser is employed to pump a nonlinear crystal to generate a signal wave in the range 1000 - 1540 nm and an idler wave in the range 1580-3540 nm. The primary OPO cavity is kept on resonance for the signal wave. Wavelength selection by a two-stage process (Etalon, birefringent filter) and subsequent second harmonic generation (SHG) converts signal photons into the range 500 - 765 nm. Red arrows depict the pump laser beam, dark red (orange) arrows the signal (idler) beams, green arrows depict the beam path of frequency-doubled signal. (b) Output power vs wavelength.

To ensure that only a single (signal or idler) OPO cavity mode is supported for resonant oscillation, a combination of mode-selective intra-cavity optical elements needs to be employed. We find best results when using a combination of either two Etalons (with distinctly different free spectral ranges) or combining an Etalon with a birefringent filter element - in either case the optical elements being aligned in series along the intra-cavity beam path.

Figure 2 shows the schematics of a widely tunable cw OPO system covering the visible spectral range and designed along the outlined considerations - and notably the first of such kind available as commercial turnkey system. In the first (singly resonant) cavity, 532 nm cw laser light is used to pump a nonlinear crystal (periodically poled lithium niobate or tantalate) for generating signal and idler waves in the near infrared spectral range (900-1050 nm and 1080-1300 nm, respectively). The system design encompasses coarse pre-selection of the operational signal/idler wavelength through mechanical selection of the crystals' poling period and control of the crystal temperature, and mode filtering using a combination of two Etalons. The primarily generated signal or idler wave is converted into the visible range in a SHG cavity, to cover a tuning range of 450-525 nm respectively 540-650 nm. Except for the so-called degeneracy gap around 532 nm, which essentially reflects the need to switch cavity mirrors from highly-reflective to highly-transmissive when tuning the OPO cavity across 1064 nm, the wavelength coverage is truly continuous. Depending on the particular choice of pump laser, output powers in the visible range of up to 500 mW can be achieved (cf. Figure 2).

While the clear key performance characteristic of the platform presented in Figure 2 is to provide convenient access to wavelengths as short as 450 nm, we do not hesitate to stress that the inherent degeneracy-gap is a drawback for experimental settings that call for gap-free tuning protocols. Along the same lines is the notorious demand for higher output powers for applications like mastering diffractive optical elements⁵. This motivates the accordingly adapted novel scheme presented in Figure 3, in which a fiber- laser based pump source delivering 7 Watt at 780 nm is used to drive the primary OPO process efficiently up to multi-Watt output levels. In essence, the resonator layout for both OPO and SHG is designed to match optimum OPO pump threshold and to maximize SHG conversion rates up to more than 60% ⁶. Thereby, and in contrast to the scheme presented in Figure 2, the main conceptual idea is to operate the primary OPO cavity *always* on resonance for the signal wave (1000 - 1540 nm), and to feed solely this signal wave into the SHG process. This not only avoids the need of switching OPO cavity mirrors but also provides gap-free tuning across the 500 - 765 nm wavelength range, at a typical output power of 1 W. To the best of our knowledge, the design delivers unprecedented tunable cw OPO performance with regard to combined wavelength coverage and output power. We point out that the system layout is general enough to be further adaptable, e.g. by power up-scaling or wavelength shifting of the employed pump laser.



Figure 4. (a) Power fluctuation of 600 nm output of the cw OPO design shown in Figure 3, measured on a timescale of 30 min. (b) Long-term frequency stability of the cw OPO design shown in Figure 3 in closed-loop operation. Employing a software module in conjuction with a high performance wavemeter, a frequency stability of +/-1 MHz over hours can be achieved.

3. PERFORMANCE HIGHLIGHTS AND TUNING CHARACTERISTICS

Since each of the nonlinear processes involved in a two-stage cw OPO design tends to increase power fluctuations, it is the system power stability that is often critically examined. To give a practical example, when using a state-of-the-art 532 nm DPSS laser with a long term power stability of better than <2% over 8 hours, an amplitude stability of the SHG output of <5% can be achieved, which is quite sufficient for many applications. Given that some of the SHG output power can be sacrificed, this performance can be significantly improved by invoking external or internal power stabilization schemes. As to former, e.g. external closed-loop amplitude modulators might be employed in a straight forward manner, to typically achieve <1% power fluctuation (while sacrificing typically 25% of the available SHG output power). Internal power stabilization schemes can give even better results, but are technically considerably more sophisticated. For the sake of brevity, here, we resort to highlight Figure 4a, showing experimentally measured < 0.5% SHG peak-to-peak power fluctuation achievable with the cw OPO design presented in Figure 3.

As to frequency stability, both designs presented deliver high quality cw output with typical linewidths of < 500 kHz throughout both the visible and the near infrared tuning range, corresponding to typical coherence lengths well above 100 m. Thereby, a long term frequency stability of < 150 MHz over hours is routinely observed at typical lab conditions. For applications with highest demands, the performance characteristics can be further improved by operating the system in closed-loop mode, i.e. in conjunction with an external wavelength measurement device (wavemeter). In this operation mode, the achievable long term stability essentially approaches the measurement resolution of the external wavemeter device itself, which can be as low as on the order of a few MHz, as illustrated in Figure 4b.

In Figure 5, we illustrate application-tailored tuning protocols that can be realized with the tunable cw OPO designs discussed above. Not least, the shown data reflects the challenging requirements of typically fundamental research applications in terms of simultaneous wavelength coverage and flexibility in wavelength step-sizes: On the one hand, due to broad and unstructured spectral bands of a particular sample under study (at room temperature), a thorough spectral characterization might require wavelength tuning over a wide spectral range. On the other hand, identification and resonant excitation of single electronic transitions - typically on the order of a few GHz width at cryogenic temperatures - requires sufficiently narrow laser light emission linewidths and tuning in the (sub-) picometer range. Widely tunable cw OPOs offer a variety handles for wavelength tuning and can be adapted to such demands in most cases. As illustrated in Figure 5, the commonly applied wavelength tuning mechanisms are coarse tuning (essentially by crystal temperature), stepwise tuning (by stepping of the intra-cavity etalon), and truly continuous mode-hop free tuning (by piezo-scanning the OPO cavity length) - cf. caption of Figure 5 for more details.



Figure 5. (a) Power of the signal output of the cw OPO design shown in Figure 3 recorded for a coarse scan. (b) Frequency tuning of the cw OPO shown in Figure 2 by stepping the intra-cavity Etalon at a central wavelength of 940 nm. The frequency of the laser light output can be changed in discrete steps down to 2 GHz (in the IR range). (b) Truly continuous (mode-hop free) scan at a central wavelength of 940 nm. The scan range is >10 GHz and can reach up to more than 20 GHz depending on the central wavelength. After frequency conversion, this results in mode-hop free scans over more than 40 GHz in the visible wavelength range.

4. APPLICATION EXAMPLE: COLOR CENTERS IN DIAMOND

In quantum technology, many of the envisaged device architectures rely on quantum systems generating exactly one photon per excitation event, so-called single-photon emitters. Implementing such single-photon emitters in solid state is widely recognized to offer technologically appealing advantages. Among most promising candidates are "color centers", microscopically localized impurities in diamond crystals. Early attention has focused on the nitrogen vacancy (N-V) center in diamond, where a nitrogen atom together with a vacant site replace two adjacent carbon atoms of the diamond lattice. While the N-V center arguably is the most extensively studied quantum emitter, another class of defects based on group-IV elements (Si-, Ge-, Sn-, Pb-V) has been attracting considerable interest recently⁷, due to potentially further improved properties like lower susceptibility to external noise.

Though the inventory of optically active centers in diamond is remarkably diverse, their spectral characteristics are governed by similar physical principles. A crucial factor is the extent of coupling of the emitters electronic transitions to lattice vibrations of the surrounding crystal host. The strength of this phonon coupling can be inferred by the appearance of phonon sidebands (PSBs) in absorption and/or emission spectra, in addition to the sharp zero-phonon-line (ZPL) of the purely electronic transition. For an ensemble of single-photon emitters, in turn, the spectrum is additionally broadened by inhomogeneous static variations in the local microscopic environments of individual sites.

The spectral characterization of single quantum systems by traditional direct absorptions measurements is hardly feasible. Researchers typically rather rely on photoluminescence excitation (PLE) spectroscopy, in which the photon emission intensity of a quantum emitter is measured while tuning the excitation (laser) frequency. Given the distinct spectral features, the identification and characterization of single emitters requires a profound design of frequency tuning mechanisms. For illustration, Figure 6 shows experimental data compiled from two studies concerned with the germanium-vacancy center in diamond (Ge-V)⁸. It has been recently proposed to offer a variety of attractive properties for quantum technology applications, like single-photon emission under room temperature conditions. Notably, these defects possess inversion symmetry and therefore are not sensitive to local fluctuation in electric fields.

Figure 6a shows a PLE spectrum of an ensemble of Ge-V color centers recorded at room temperature conditions. It has been recorded by exploiting the full coarse tuning range of a cw OPO scheme discussed above, straight-forward accessible by automated cavity optics and crystal selection along with temperature tuning. The measurement clearly reveals a maximum count rate when resonantly driving the ZPL at approximately 602 nm. The data is adapted with permission from a comparative study on Si-V and Ge-V color centers⁸.



Figure 6. (a) Photoluminescence excitation spectrum of an ensemble of Ge-V centers recorded at 0.25 mW excitation power. The spectrum is recorded at room temperature under excitation wavelength tuning from 450 nm to 640 nm. Red arrows indicate the wavelength interval shown in figure b). Adapted with permission from reference 8. (b) Normalized photoluminescence spectrum of a single Ge-V center recorded at a temperature of 5 K. The purple line is a fit to four Gaussian peaks. The red arrow marks zero excitation detuning as shown in figure c). Adapted with permission from reference 9. (c) Photoluminescence excitation spectrum of the most prominent transition in b), recorded under tuning the resonant excitation. The orange line represent the signal when an additional 532 nm gating laser is switched on, the blue line represent the signal when the gating laser is off. Adapted with permission from reference 9.

As shown in Figure 6b, the photoluminescence spectrum of a single Ge-V center at cryogenic temperature conditions reveals a four-line fine structure of the ZPL emission (because of strong spin-orbit coupling, the ground state and the first excited state are split into a pair of energy levels with twofold spin degeneracy). Note that the spectral window shown in Figure 6b is just a tiny cutout of Figure 6a around the ZPL, and is showing the signal of a single Ge-V emitter rather than an ensemble.

In Figure 6c, the resonant fluorescence characteristics of the most prominent feature in the spectrum of the single Ge-V emitter is precisely characterized. For this purpose, the cw OPO excitation wavelength is tuned within an interval of approximately +/- 5 GHz across the spectral resonance, by truly continuous frequency tuning in conjunction with a wavemeter device. It should be emphasized that the resonant PLE signal is only detectable at additional non-resonant excitation, i.e. when the Ge-V center is additionally excited with a gating laser (at 532 nm). The role played by the 532 nm light is that of a switch controlling the onset and decrease of resonant fluorescence. The experimental findings are discussed in detail in reference 9, where the authors also quantitatively explain the observed dynamics by the presence of a dark state.

5. OUTLOOK

The widely tunable cw OPO designs presented here provide unprecedented performance with regard to combination of wavelength coverage and output power. Thereby, the basic system design is general enough to be further adaptable, e.g. by power up-scaling or wavelength shifting of the employed pump laser. As we hope to have illustrated in this contribution, cw OPO technology can be expected to mature into a choice well recognized among laser light sources that accompany experimental studies in fundamental and application-oriented research.

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