Advances in Ultrafast and Continuous-Wave Optical Parametric Oscillators

(*Invited Paper*)

M. Ebrahim-Zadeh
ICFO-Institut de Ciencies Fotoniques, Mediterranean Technology Park, 08860 Castelldefels (Barcelona), Spain
Instictucio Catalana de Recerca i Estudis Avancats (ICREA), Passeig Luis Companys 23, 08010 Barcelona, Spain

Abstract-**Effective strategies for the generation of widely tunable coherent radiation in the visible and ultraviolet using synchronously-pumped and continuous-wave (cw) optical parametric oscillators are described. Spectral regions from 250 to 710 nm are accessed and output powers of hundreds of milliwatts with him spectral, temporal and spatial beam quality are obtained in ultrafast femtosecond and cw operation.**

I. INTRODUCTION

The development of coherent solid-state light sources for new spectral regions has long been a major goal of research in laser science and technology, since Maiman demonstrated the first experimental laser in 1960. Many approaches to deliver coherent light in difficult spectral regions have been explored using conventional laser techniques, but access to extended regions in the ultraviolet (UV), visible and infrared (IR) have remained difficult because of the absence of suitable solid-state gain materials.

Optical parametric oscillators (OPOs) offer attractive alternatives for the generation of coherent radiation in spectral regions inaccessible to lasers. While the first experimental demonstration of an OPO was reported in 1965, for nearly two decades thereafter there was little progress in practical development of OPO devices, owing to the absence of suitable nonlinear materials and solid-state laser sources. With the advent of a new generation of birefringent nonlinear crystals such as β-BaB_2O_4 (BBO), LiB_3O_5 (LBO) and $KTiOPO_4$ (KTP) in the mid-1980s and quasi-phase-matched (QPM) materials, particularly periodically-poled $LiNbO_3$ (PPLN), in the mid-1990s, there began renewed interest in OPOs as practical alternatives to lasers for coherent light generation in new spectral regions. In the intervening period, OPOs have been transformed into viable, state-of-the-art light sources capable of accessing difficult spectral regions and addressing real applications beyond the reach of lasers.

Advances in OPO devices have been more deliberate in the 1-5 μm spectral range, driven by the availability of practical near-IR solid-state laser pump sources and suitable nonlinear materials. As a down-conversion process, optical parametric generation extends the pump to longer wavelengths. As such, the widespread use of solid-state Nd-doped crystalline and fiber lasers near 1 μm or the Ti:sapphire laser near 800 nm as the pump source has permitted OPO access to wavelengths above 1 μm. The main exception has been in the nanosecond pulsed regime, where the use of Q-switched frequency-doubled and tripled Nd-based solid-state lasers at 532 nm and 355 nm has enabled wavelength extension of pulsed OPOs into the visible and near-UV. In other operating regimes, advancement of OPO devices to wavelengths below 1 μm has been challenging because of a lack of laser pump sources of sufficient intensity and high beam quality at shorter wavelengths in the visible and UV, and so new techniques have to be devised to overcome this obstacle.

Here, we describe practical approaches to spectral extension of ultrafast and cw OPOs into the visible and UV. In the ultrafast femtosecond regime, we exploit efficient single-pass second harmonic generation (SHG) of the Kerr-lens-mode-locked (KLM) Ti:sapphire laser in novel birefringent nonlinear crystal BiB_3O_6 (BIBO) to achieve wavelength coverage across the entire visible range of 480-710 nm with an average power of 270 mW. By deploying additional frequency doubling of the signal pulses internal to the OPO cavity, we provide further access to the 250-355 nm spectral range in the UV at up to 225 mW average power. In the cw regime, by deploying frequency-doubled cw Nd-based solid-state lasers at 532 nm in combination with the QPM nonlinear crystal MgO:sPPLT, and by using internal frequency doubling of the OPO signal in BIBO, we demonstrate wavelength generation down to 425 nm in the blue at up to 450 mW of power.

II. EXPERIMENTS

A. Synchronously-Pumped Femotsecond Optical Parametric Oscillator for the Visible

The configuration of the femtosecond OPO is shown in Fig. 1. The OPO is synchronously-pumped in the blue by the second harmonic of a KLM Ti:sapphire laser and exploits BIBO, both as the doubling crystal for the pump and as the gain medium for the OPO. The nonlinear crystal, BIBO, is a relatively new material with attractive optical properties for frequency conversion in the visible and UV [1]. It has an optical transmission from ~2700 nm in the IR down to ~280 nm in the UV. As a biaxial crystal, BIBO also exhibits versatile phase-matching properties, large angular and spectral acceptance bandwidths, low spatial walkoff and broadband angle tuning at room temperature [2]. While the UV transmission cutoff in BIBO is at a longer wavelength than BBO, it offers substantially larger effective nonlinearity, as high as d_{eff}~3.7 pm/V [3], which is comparable to that in KTP.

The Ti:sapphire pump laser delivers pulses of ~130 fs at 76 MHz, with an average power of up to 1.9 W over a tunable range of 750-950 nm. Frequency doubling of the laser is achieved in a single pass in a 1-mm crystal of BIBO. The crystal is cut for collinear critical type I (e+e→o) interaction in *yz* plane (ϕ=90°) at an internal angle θ~152° at normal incidence. This geometry yields a maximum theoretical effective nonlinear coefficient, d_{eff}~3.3 pm/V [2]. An average power of >1 W in the blue at >50% efficiency is available over a tunable range of 375-435 nm [4]. The blue pulses have durations of ~220 fs. The blue pump beam is focused to a waist radius w_0~25 μm inside a second BIBO crystal, the gain element for the femtosecond OPO. We use collinear phase-matching, with the crystal cut for type I (o→e+e) interaction in *yz* plane (ϕ=90°) at an internal angle θ~159°. From considerations of group velocity mismatch (GVM) between the blue pump and visible signal pulses (GVM~ 101-312 fs/mm over 500-700 nm), we use a 500-μm crystal for the OPO. The crystal faces are AR-coated for signal (R<0.5% at 500-700 nm) and have high transmission for pump (T>95% at 375-435 nm). The OPO is configured in a three-mirror cavity with 2 concave reflectors (M_1, M_2: r=100 mm) and a plane output coupler (M_3). The concave mirrors have >99% reflectivity for signal wavelengths over 500-680 nm and >90% transmission for the pump over 380-450 nm. The mirrors also have >80% transmission for the idler over 900-3000 nm, thus ensuring singly-resonant oscillation. Two uncoated Brewster-cut fused silica prisms provide intracavity dispersion compensation.

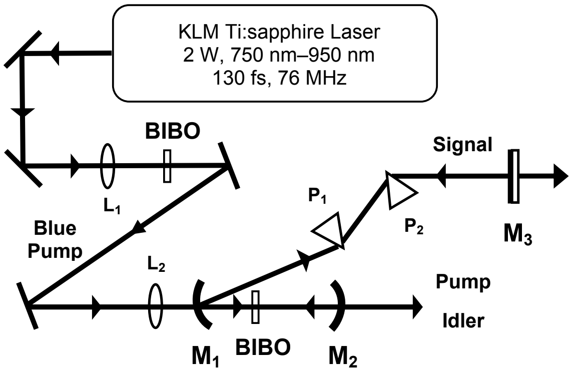

Fig. 1. Configuration of the visible BIBO femtosecond OPO synchronously-pumped by the second harmonic of KLM Ti sapphire laser in the blue.

Figure 2 shows the visible signal tuning range of the OPO at room temperature, as a function of crystal internal angle obtained at a fixed pump wavelength of 415 nm. The solid curve represents the predicted tuning range for collinear type I (o→e+e) phase-matching in the optical *yz* plane obtained using the Sellmeier relations for BIBO [1], where good agreement between the experimental data and theoretical calculation is evident. The OPO can be continuously tuned in the visible across the green-yellow-orange-red, from 480 to 710 nm, by changing the internal angle of the BIBO crystal between θ=175° and θ=154°. The corresponding tuning range of the idler is from 3060 to 999 nm. For a given crystal angle, wavelength tuning is also available through the variation of OPO cavity length. We typically obtain ~10 nm of signal tuning for a change in OPO cavity length of ~3 μm. Interestingly, the mid-IR idler wavelength of 3060 nm generated by the OPO is well beyond the nominal 2700 nm absorption cutoff in BIBO. This could be due to the short crystal length of 500 μm used in our experiment or may be indicative of a longer IR transmission range in BIBO than 2700 nm.

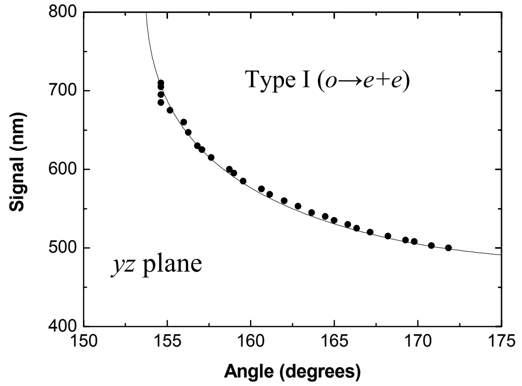

Fig. 2. Visible signal tuning range of BIBO femtosecond OPO as a function of internal angle in the optical *yz* plane. The pump wavelength is 415 nm.

We generated a maximum visible average power of 270 mW at ~620 nm for 800 mW of blue pump power with an 8% output coupler. The OPO could provide >150 mW across 500-700 nm, and >200 mW across 530-650 nm. At the extremes of the tuning range towards 480 nm and 710 nm, a visible signal power >100 mW was still available. The reduction in the signal power at the extremes of the tuning range was attributed to the increase in the transmission of OPO mirrors away from the centre of tuning curve. With the 8% output coupler, the oscillation threshold was 200 mW, equivalent to a Ti:sapphire laser power of 650 mW. With a high reflector plane mirror in place of an output coupler, the OPO threshold was as low as 100 mW, corresponding to a Ti:sapphire pump power of 420 mW.

Temporal measurements were performed using interferometric autocorrelation technique in a 500-μm crystal of BBO cut for type I (o+o→e) phase-matching (θ=42°) and a UV-enhanced silicon photodiode. Figures 3(a) and 3(b) show the autocorrelation trace and spectrum of the output pulses, indicating a duration of ~120 fs and a bandwidth (FWHM) of ~3.5 nm. The time-bandwidth product is $\Delta\nu\Delta\tau$~0.35, implying near-transform-limited pulses. The measured pulse durations may in fact be shorter than ~120 fs due to the large GVM in the BBO autocorrelation crystal. The calculated GVM for SHG of visible pulses in BBO varies from 690 fs to 275 fs/mm for fundamental wavelengths from 500 nm to 700 nm. At 595 nm, the GVM is 422 fs/mm, resulting in a mismatch of ~211 fs in a 500-μm crystal. This implies that the measured pulse duration of ~120 fs is likely limited by GVM in the BBO crystal and the pulses may be close to or shorter than ~100 fs. This could be confirmed by using a shorter BIBO autocorrelation crystal.

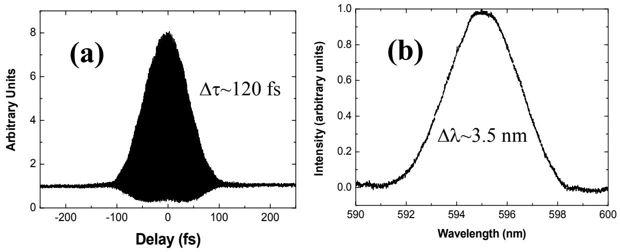

Fig. 3. Typical interferometric autocorrelation (a), and Spectrum (b), of the visible signal pulses with intracavity dispersion compensation, indicating a time-bandwidth product ΔνΔτ~0.35.

B. *Synchronously-Pumped Femotsecond Optical Parametric Oscillator for the Ultraviolet*

To achieve wavelength extension into the UV, we perform internal frequency-doubling of the visible signal pulses in the BIBO femtosecond OPO describe above. The OPO configuration is shown in Fig. 4. The resonator is now a bifocal ring, comprising four concave reflectors (r=100 mm) and two plane mirrors. The concave mirrors, M_1 and M_2, provide the focus for the OPO crystal, whereas M_5 and M_6 allow focusing into the doubling crystal. All mirrors are >99% reflecting for the visible signal wavelengths over 500-700 nm. M_1 and M_2 are also >90% transmitting for the blue pump over 380-450 nm. The ring resonator allows the generation of the UV output in one direction through M_6. To allow maximum UV extraction, M_6 also has high, but variable transmission (T~70% to 90%) over 250-350 nm. For frequency-doubling, we used BBO, because of its deeper UV transparency (~180 nm) compared to BIBO (~280 nm) and higher nonlinear efficiency for fundamental wavelengths below ~650 nm [3]. The BBO crystal was 500-μm, cut at θ=42° for type I ($o+o\rightarrow e$) SHG, providing an effective nonlinear coefficient, d_{eff}~1.4-1.8 pm/V, across the fundamental signal tuning range. The crystal faces were AR-coated (R<1%) over 500-700 nm and had <8% reflectivity over 250-350 nm.

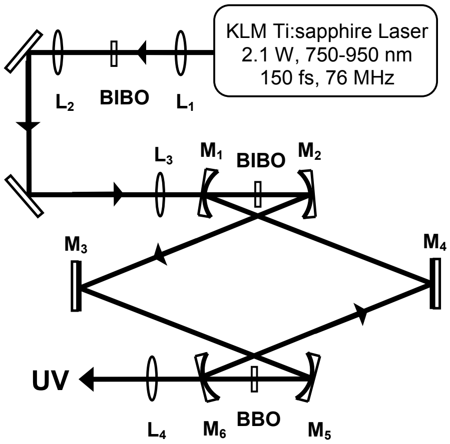

Fig. 4. Configuration of the intracavity frequency-doubled BIBO femtosecond OPO for pulse generation in the UV.

Wavelength tuning in the UV was achieved by continuous tuning of the OPO signal across the visible through rotation of the BIBO crystal and simultaneous angular tuning of the BBO doubling crystal. The OPO signal could be tuned across 500-710 nm by changing the internal BIBO crystal angle from θ=171.5° to θ=154.5°. The corresponding UV wavelength range of 250 to 355 nm was generated for a change in the internal angle of the BBO crystal from θ=52.3° to θ=33.1°. The limit to the obtained tuning range in the UV was set by the overall reflectivity of the cavity mirrors at the signal wavelength. By using mirrors with broader reflectivity band and shorter pump wavelengths near 400 nm, full coverage across 230-360 nm should be attainable.

We were able to generate UV average powers of >175 mW over ~70% of the tuning range (275-350 nm) and >100 mW over ~80% of the tuning range (270-355 nm). In the wavelength range of 255-270 nm, practical powers of 25 to 100 mW were still generated, with 5 mW available at 250 nm. The highest UV average power was obtained at 323 nm, reaching 225 mW at the maximum blue power of 1.15 W, representing a conversion efficiency of 19.7%. This is close to the maximum signal power of 270 mW extracted directly from the visible OPO previously, implying that the intracavity doubling acts as an almost optimized nonlinear output coupler. The UV output beam had close to a circular profile with a M^2<1.1. The blue pump power threshold for the frequency-doubled OPO was 150 mW, equivalent to a fundamental Ti:sapphire laser power of 600 mW.

Temporal measurements were performed using cross-correlation between the UV pulses and Ti:sapphire input pulses at 830 nm in a 500-μm BBO crystal cut for type I ($o+o\rightarrow e$) phase-matching (θ =26°). Background-free intensity profiles were obtained using a GaAsP detector. A typical profile and spectrum at 323 nm are shown Fig. 5, confirming near-transform-limited pulse with Δν.Δτ~0.34. Across the UV tuning range, pulse durations of 132 fs to 250 fs were measured, with corresponding time-bandwidth products varying from ~0.34 to ~0.6.

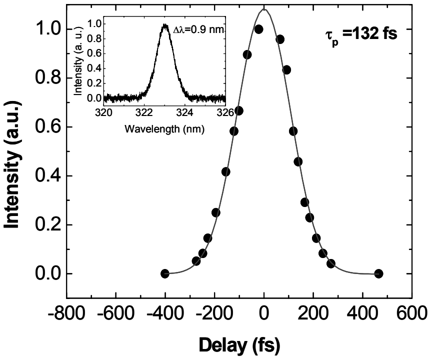

Fig. 5. Cross-correlation trace, and (inset) spectrum of the generated UV pulses at 323 nm. The time–bandwidth product is Δν.Δτ~0.34.

C. *Continuous-Wave Optical Parametric Oscillator for the Blue*

The application of internal frequency doubling technique to the BIBO femtosecond OPO, described above, and the effective generation of ultraviolet pulses at practical powers provided new impetus for the extension of this approach to the cw regime. To achieve this goal, we exploit intracavity SHG of a green-pumped, cw, singly-resonant OPO (SRO) with MgO:sPPLT as the nonlinear crystal [5-7]. The configuration of the cw SRO is shown in Fig. 6.

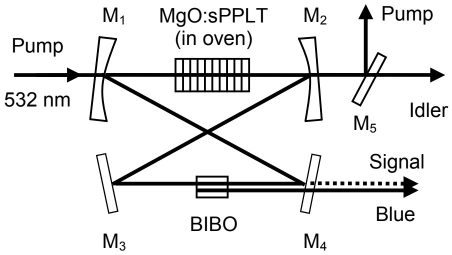

Fig. 6. Schematic of the intracavity frequency-doubled cw SRO based on MgO:sPPLT for the generation of tunable cw radiation in the blue.

The SRO cavity is formed in a ring, with two concave reflectors, M_1, M_2: r=50 mm) and two plane mirrors (M_3, M_4). M_1, M_2 and M_3 are >99.9% reflecting for the signal (840-1000 nm). M_4 is also highly reflecting (>99%@850-920 nm, >99.9%@920-1000 nm), and 85-90% transmitting over 425-500 nm. All mirrors also have 85-90% transmission at the idler (1100-1400 nm). The nonlinear crystal is MgO:sPPLT (d_{eff}~10 pm/V). It is 30-mm long with a single grating (Λ=7.97 μm). The crystal faces are <0.5% reflecting for the signal (840-1000 nm) and >98% transmitting at 532 nm. The residual reflectivity of the coating is 0.6% to 4% per face for the idler (1100-1400 nm). The pump sources is a 10-W frequency-doubled, cw diode-pumped Nd:YVO_4 laser at 532 nm with a linewidth <25 MHz. For internal doubling, we used BIBO, 5-mm long and 4×8 mm in aperture, cut at θ=160° (φ=90°) for type I SHG *(e+e→o)* in *yz*-plane (d_{eff}~3.4 pm/V). The crystal faces have a reflectivity of <0.5% over 850-1000 nm and <0.8% over 425-500 nm. For the SRO, we use a relatively strong focusing (ξ_{SRO} =2), corresponding to a pump beam radius of w_{op}=24 μm inside the MgO:sPPLT crystal [5,6]. The signal waist is ~31 μm in the OPO crystal (b_s=b_p) and ~160 μm (ξ_{SH}~0.015) in the BIBO crystal.

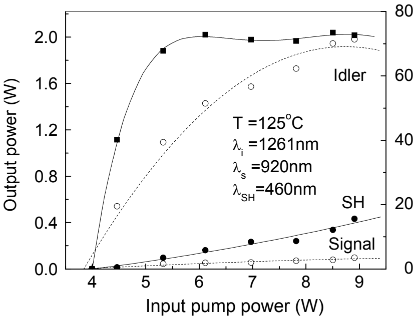

Fig 7. Single-frequency blue, signal and idler power, and pump depletion versus input pump power to the intracavity frequency-doubled cw SRO.

By temperature tuning the signal from 978 to 850 nm (idler from 1167 to 1422 nm), we could tune the SH blue output from 489 to 425 nm by varying the internal angle of the BIBO crystal from 163.8° to 155.2°. We obtained blue powers from 45 mW at 425 nm to 300 mW at 489 nm, with as much 448 mW available at 459 nm. We extracted >300 mW of blue power over 53% of the tuning range and >100 mW over 90% of the tuning range. From the transmission of mirror M_4 and the out-coupled signal power, we calculate the intracavity signal power to vary from ~170 W at 978 nm to ~35 W at 850 nm, representing a maximum single-pass SHG efficiency of 0.29%. In addition to the blue and signal, the SRO also simultaneously generates up to 2.6 W of idler power across 1167-1422 nm. The pump depletion varies from ~73% to ~48% across the tuning range. Figure 7 shows power scalability of the cw SRO near the maximum SHG power (460 nm). At the highest pump power of 8.9 W, we obtained 432 mW of blue, 97 mW of out-coupled signal, and ~1.98 W of idler power, with a pump depletion of ~73%. The threshold for the frequency-doubled SRO is 4 W (2.4 W without the BIBO crystal).

We analyzed the spectrum of the generated blue light using a confocal scanning interferometer (FSR=1 GHz, finesse=400). A typical transmission fringe pattern at maximum blue power at 460 nm is shown in Fig. 8, confirming single-frequency operation with a linewidth of ~8.5 MHz. We also recorded far-field energy distribution of the blue output beam, confirming a Gaussian spatial distribution, with an ellipticity of 0.69.

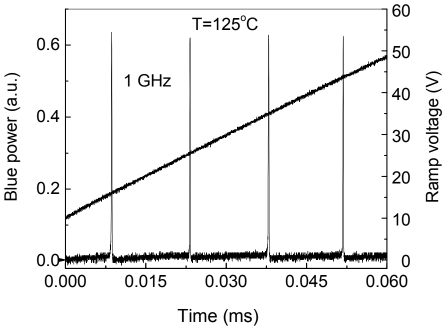

Fig. 8. Single-frequency spectrum of the generated blue light recorded by a scanning confocal interferometer.

III. CONCLUSIONS

We have demonstrated that the application of frequency upconversion techniques in combination with OPOs can provide a viable solution to coherent light generation in the visible and UV, offering exceptionally broad tunability, practical powers, and high spectral, temporal, spatial quality in the ultrafast femtosecond and cw operating regimes.

ACKNOWLEDGMENT

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