

BiB₃O₆ femtosecond optical parametric oscillator

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We report a femtosecond optical parametric oscillator (OPO) based on the nonlinear material BiB₃O₆. The OPO is synchronously pumped in the blue by the second harmonic of a Kerr-lens-mode-locked Ti:sapphire laser. It can provide wide and continuous tuning across the entire green–yellow–orange–red spectral range with a single crystal and a single set of mirrors. Using a 500 μm BiB₃O₆ crystal and collinear type I ($e+e \rightarrow o$) phase matching in the optical yz plane, a signal wavelength range of 480–710 nm is demonstrated with angle tuning at room temperature at average output powers of 270 mW. With 220 fs blue pump pulses, near-transform-limited signal pulses of 120 fs duration have been obtained at 76 MHz repetition rate.

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Optical sources capable of providing high-repetition-rate femtosecond pulses in the visible spectrum are of interest for a wide range of applications in time-domain spectroscopy, optical microscopy, frequency metrology, and quantum optics. A particularly difficult spectral range is 500–700 nm in the visible spectrum, which remains inaccessible to the Kerr-lens-mode-locked (KLM) Ti:sapphire laser or its frequency-doubled output.

Synchronously pumped optical parametric oscillators (OPOs) offer a practical solution for the generation of high-repetition-rate femtosecond pulses in new spectral regions, particularly the infrared (IR). When pumped directly by the KLM Ti:sapphire laser, they can readily provide femtosecond pulses throughout the 1–5 μm spectral range in the near- to mid-IR. For pulse generation in the visible spectrum, however, the direct use of the KLM Ti:sapphire laser is precluded, and additional frequency conversion schemes have to be deployed in combination with the OPO approach. There have previously been a number of attempts to provide femtosecond pulses in the visible spectrum using synchronously pumped OPOs. One technique relies on direct pumping of femtosecond OPOs with the KLM Ti:sapphire laser and subsequent second-harmonic generation (SHG) of the near-IR signal pulses into the visible spectrum internal to the OPO cavity.¹ Such systems have been based on KTiOPO₄ (KTP) or RbTiOAsO₄ (RTA) as the OPO gain material and β -BaB₂O₄ (BBO) as the SHG crystal. Because of the limited tuning capability of KTP and RTA, the visible pulses available to such OPOs cover a confined spectral range of only ~ 80 nm from 580 to 660 nm. The second method has been based on frequency doubling of a KLM Ti:sapphire laser the blue to directly pump a femtosecond OPO using noncollinear phase matching in BBO, where visible pulses over a limited range of 566–676 nm were generated at up to 100 mW average power.² The combination of the two methods has enabled the generation of femtosecond pulses in the visible spectrum, across a total tuning range of 566–676 nm. However, the remaining gaps in the 500–700 nm spectral range have so far been inaccessible to femtosecond OPOs.

Here we describe a synchronously pumped OPO that can provide femtosecond pulses with wide and continuous tunability across the entire red–orange–yellow–green (480–710 nm) spectral range using a single nonlinear crystal and a single set of mirrors. The OPO, pumped in the blue pulse by the second harmonic of a KLM Ti:sapphire laser, exploits the newly developed nonlinear material, bismuth triborate (BiB₃O₆ or BIBO), both as the doubling crystal for the Ti:sapphire pump and as the nonlinear gain medium for the OPO.

BIBO is a relatively new nonlinear material with interesting optical properties for frequency conversion in the visible and ultraviolet (UV) spectra.^{3–6} 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 very versatile phase-matching properties, large angular and spectral acceptance bandwidths, low spatial walk-off, and a broadband angle tuning at room temperature.⁵ While the UV transmission cutoff of BIBO occurs at a longer wavelength than BBO, it offers substantially larger effective nonlinearity measured to be as high as $d_{\text{eff}} \sim 3.7$ pm/V,⁶ which is comparable to that in KTP. Such a combination of properties makes BIBO highly attractive for frequency conversion in the visible and UV spectra.

The configuration of the visible femtosecond OPO based on BIBO is shown in Fig. 1. The OPO is synchronously pumped by the second harmonic of a KLM Ti:sapphire laser (Coherent, Mira 900). The 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 ($e+e \rightarrow o$) interaction in the yz plane ($\phi=90^\circ$) at an internal angle of $\theta \sim 152^\circ$ at normal incidence. This geometry yields a maximum theoretical effective nonlinear coefficient, $d_{\text{eff}} \sim 3.3$ pm/V.⁵ An average power of >1 W in the blue at $>50\%$ efficiency is available over a tunable range of 375–435 nm.^{5,7} The blue pulses have durations of ~ 220 fs.

The blue pump beam is focused to a waist radius $w_0 \sim 25$ μm inside a second BIBO crystal, the gain el-

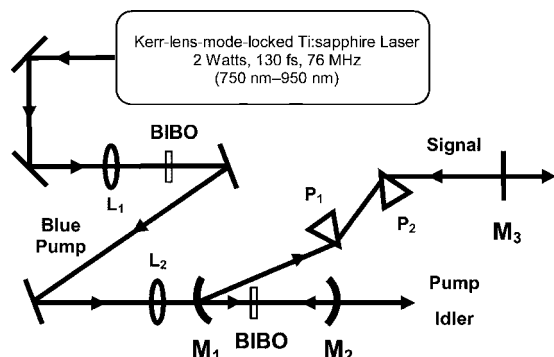


Fig. 1. Configuration of the visible BIBO femtosecond OPO synchronously pumped by the second harmonic of KLM Ti:sapphire laser in the blue. L_1 and L_2 , focusing lenses; P_1 and P_2 , prisms.

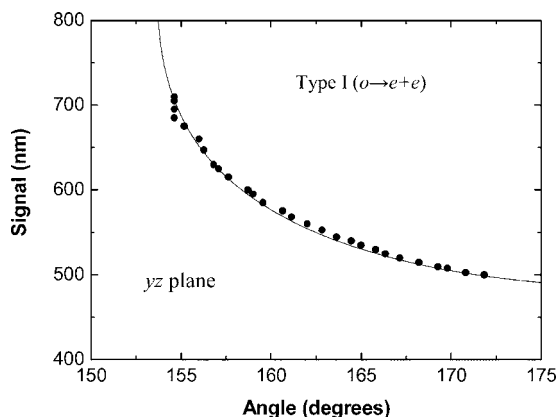


Fig. 2. Visible signal tuning range of BIBO femtosecond OPO as a function of the internal angle under phase matching in the optical yz plane. The pump wavelength is 415 nm.

ement for the femtosecond OPO. We use collinear phase matching with the crystal cut for the type I ($o \rightarrow e + e$) interaction in the yz plane ($\phi = 90^\circ$) at an internal angle of $\theta \sim 159^\circ$. From considerations of group velocity mismatch (GVM) between the blue pump and the visible signal pulses (GVM ~ 101 – 312 fs/mm over 500–700 nm), we use a crystal length of $500 \mu\text{m}$ for the OPO. The crystal end faces are antireflection coated for the signal ($R < 0.5\%$ at 500–700 nm) and have high transmission for the blue pump ($T > 95\%$ at 375–435 nm). The OPO is configured in a three-mirror, standing-wave cavity comprising two concave reflectors (M_1 and M_2) of the radius of the curvature of $r = 100$ mm and a plane output coupler (M_3). The concave mirrors are highly reflecting ($R > 99\%$) for visible signal wavelengths over 500–680 nm and highly transmitting ($T > 90\%$) for the blue pump over 380–450 nm. The mirrors also have high transmission for the idler ($T > 80\%$ at 900–3000 nm) thus ensuring singly resonant oscillation. Two uncoated Brewster-cut fused-silica prisms provide intracavity dispersion compensation.

Figure 2 shows the visible signal tuning range of the OPO at room temperature as a function of the crystal internal angle obtained at a fixed pump wavelength of 415 nm. The solid curve represents the pre-

dicted tuning range for collinear type I ($o \rightarrow e + e$) phase matching in the optical yz plane obtained using the Sellmeier relations for BIBO,⁴ where good agreement between the experimental data and theoretical calculation is evident. The OPO can be continuously tuned in the visible spectrum across the green–yellow–orange–red, from 480 to 710 nm, by changing the internal angle of the BIBO crystal between $\theta = 175^\circ$ and $\theta = 154^\circ$. 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 the OPO cavity length. We typically obtain ~ 10 nm of signal tuning for a change in the OPO cavity length of $\sim 3 \mu\text{m}$. 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 $500 \mu\text{m}$ used in our experiment or may be indicative of a longer IR transmission range in BIBO than 2700 nm.

To optimize performance, we operated the OPO under different output coupling conditions by using plane mirrors (M_3) of different reflectivities at the signal wavelength. The best performance was obtained with an 8% output coupler, where a maximum average signal power of 270 mW was extracted from the OPO at ~ 620 nm for 800 mW of blue pump power at the input to the BIBO crystal. The OPO could provide >150 mW across 500–700 nm and >200 mW across 530–650 nm. At the extremes of the tuning range toward 480 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 the OPO mirrors away from the center of the tuning curve. With the 8% output coupler, the oscillation threshold was 200 mW at the input to the OPO crystal equivalent to a fundamental Ti:sapphire laser power of 650 mW. With a high reflector plane mirror in place of an output coupler, the OPO power threshold was as low as 100 mW corresponding to a fundamental Ti:sapphire power of 420 mW.

Temporal characterization of the visible signal pulses were performed using autocorrelation measurements in a $500 \mu\text{m}$ crystal of BBO cut for type I ($o + o \rightarrow e$) phase matching at $\theta = 42^\circ$ and a UV-enhanced silicon photodiode. Without group velocity dispersion compensation, the signal pulses were strongly chirped, with corresponding broadband double-peaked spectra, characteristic of self-phase modulation (SPM). A typical interferometric autocorrelation is represented in Fig. 3(a) with the corresponding spectrum at a center wavelength of ~ 590 nm shown in Fig. 3(b). The autocorrelation profile is clearly indicative of chirped pulses with a time duration of ~ 170 fs. Due to the effects of SPM, the corresponding spectrum has a bandwidth as wide as ~ 15 nm (FWHM), resulting in a time–bandwidth product of $\Delta\nu\Delta\tau \sim 2.2$, approximately seven times the transform limit.

We therefore implemented dispersion compensation by introducing a pair of uncoated Brewster-cut

fused-silica prisms within the OPO cavity. Figures 4(a) and 4(b) show the resulting interferometric autocorrelation and spectrum of the visible signal pulses corresponding to a time duration of ~ 120 fs and a spectral bandwidth (FWHM) of ~ 3.5 nm. The time–bandwidth product is now $\Delta\nu\Delta\tau\sim 0.35$ indicating near-transform-limited pulses. The near-fivefold spectral narrowing from ~ 15 to ~ 3.5 nm is indicative of the effectiveness of intracavity dispersion compensation in combating nonlinear chirp and spectral broadening induced by SPM. We believe 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 to 700 nm. At a fundamental wavelength of 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 signal pulse duration of ~ 120 fs is likely limited by the GVM in the BBO crystal, and the pulse duration may be close to or shorter than ~ 100 fs. The use of a shorter BBO crystal autocorrelation should enable confirmation of shorter signal pulse durations.

In conclusion, we have demonstrated what is to our knowledge the first femtosecond OPO capable of covering the entire gap in the visible spectrum across 500–700 nm. Using collinear phase matching and angle tuning at room temperature, the OPO generates high-repetition-rate femtosecond pulses across

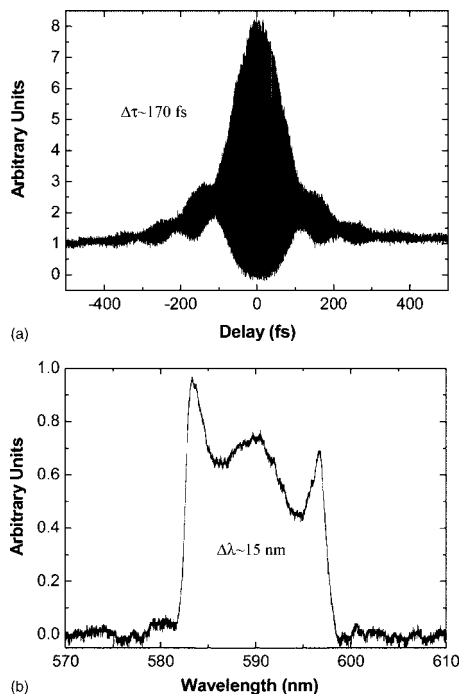


Fig. 3. (a) Typical interferometric autocorrelation and (b) corresponding spectrum of the visible signal pulses in the absence of group velocity dispersion compensation. The time duration of ~ 170 fs and the spectral bandwidth of ~ 15 nm result in a time–bandwidth product of $\Delta\nu\Delta\tau\sim 2.2$.

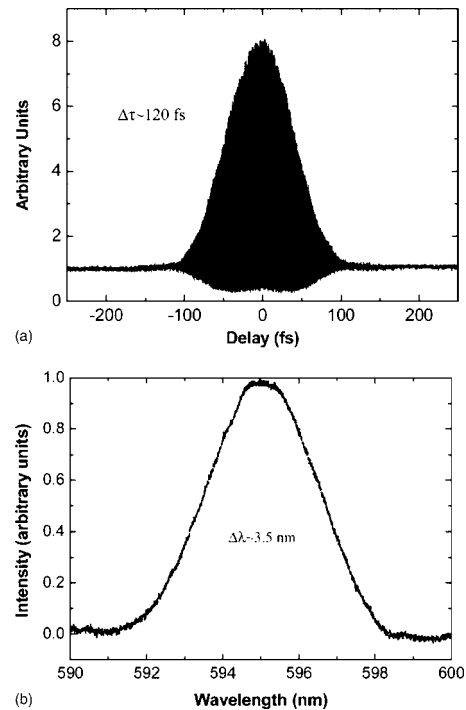


Fig. 4. (a) Typical interferometric autocorrelation and (b) corresponding spectrum of the visible signal pulses with intracavity group velocity dispersion compensation. The time duration of ~ 120 fs and the spectral bandwidth of ~ 3.5 nm result in near-transform-limited pulses with a time–bandwidth product of $\Delta\nu\Delta\tau\sim 0.35$.

480–710 nm with a single crystal of BIBO and a single set of mirrors. The wide coverage in the visible spectrum combined with practical output powers, near-transform-limited temporal characteristics, and room-temperature operation should make the OPO an attractive tool for a wide range of applications in time-domain spectroscopy, frequency metrology, and quantum optics.

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