BiB$_3$O$_6$ femtosecond optical parametric oscillator

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We report a femtosecond optical parametric oscillator (OPO) based on the nonlinear material BiB$_3$O$_6$. 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 BiB$_3$O$_6$ crystal and collinear type I (e + e → 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 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. Such systems have been based on KTiOPO$_4$ (KTP) or RbTiOAsO$_4$ (RTA) as the OPO gain material and BaB$_2$O$_4$ (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. 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 (BiB$_3$O$_6$ 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. 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. 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 $d_{eff}$ = 3.7 pm/V, 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 → o) interaction in the yz plane ($\phi$=90°) at an internal angle of $\theta$=152° at normal incidence. This geometry yields a maximum theoretical effective nonlinear coefficient, $d_{eff}$ = 3.3 pm/V. An average power of >1 W in the blue at >50% efficiency is available over a tunable range of 375–435 nm. 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 el-

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plane output coupler

cavity comprising two concave reflectors (M1 and M2)

OPO is configured in a three-mirror, standing-wave

of the radius of the curvature of \( r \)

length of 500

pump and the visible signal pulses (GVM

group velocity mismatch (GVM) between the blue

lenses; P1 and P2, prisms.

length of 415 nm. The solid curve represents the pre-
crystal internal angle obtained at a fixed pump wave-

nant oscillation. Two uncoated Brewster-cut fused-
mirrors also have high transmission for the idler (\( T \)

0.5% at 500–700 nm) and have high transmission

elements 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/\( \mu \)m over 500–700 nm), we use a crys-
tal length of 500 \( \mu \)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 (M1 and M2)
of the radius of the curvature of \( r = 100 \) mm and a

plane output coupler (M3). The concave mirrors are

highly reflecting (\( R > 99\% \)) for visible signal wave-

lengths 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 reso-
nant oscillation. Two uncoated Brewster-cut fused-
silica prisms provide intracavity dispersion compensa-
tion.

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 wave-

length 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 us-

ing the Sellmeier relations for BIBO,\(^4\) where good

agreement between the experimental data and theo-

retical calculation is evident. The OPO can be con-

tinuously 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 be-

tween \( \theta=175^\circ \) 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 \( \sim 10 \) nm of signal tuning for a change

in the OPO cavity length of \( \sim 3 \) \( \mu \)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 \( \mu \)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 un-
der different output coupling conditions by using

plane mirrors (M3) of different reflectivities at the

signal wavelength. The best performance was ob-
tained with an 8% output coupler, where a maximum

average signal power of 270 mW was extracted from

the OPO at \( \sim 620 \) nm for 800 mW of blue pump power

at the input to the BIBO crystal. The OPO could pro-
vide >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 crys-
tal 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 thresh-

old was as low as 100 mW corresponding to a funda-

mental Ti:sapphire power of 420 mW.

Temporal characterization of the visible signal

pulses were performed using autocorrelation mea-
surements in a 500 \( \mu \)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 autocor-

relation is represented in Fig. 3(a) with the corre-

sponding spectrum at a center wavelength of

\( \sim 590 \) nm shown in Fig. 3(b). The autocorrelation pro-

file is clearly indicative of chirped pulses with a time
duration of \( \sim 170 \) fs. Due to the effects of SPM, the

corresponding spectrum has a bandwidth as wide as

\( \sim 15 \) 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 compensa-
tion 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 $\sim 120$ fs and a spectral bandwidth (FWHM) of $\sim 3.5$ 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 $\sim 15$ to $\sim 3.5$ 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 $\sim 120$ 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 $\sim 211$ fs in a 500 $\mu$m crystal. This implies that the measured signal pulse duration of $\sim 120$ fs is likely limited by the GVM in the BBO crystal, and the pulse duration may be close to or shorter than $\sim 100$ 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 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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