Abstract—Efficient frequency conversion of high-repetition-rate femtosecond and picosecond pulses with wide tunability throughout the visible, blue, and UV is demonstrated in the nonlinear optical crystal BiB$_3$O$_6$ (BIBO). Using direct single-pass second harmonic generation of Kerr-lens-mode-locked Ti:sapphire laser, average output powers of up to 270 mW in pulses of 120 fs at 76 MHz repetition rate have been generated across the 480–710 nm range. Output pulse durations of 220 fs and 2.8 ps have been obtained at a pulse repetition rate of 76 MHz. By implementing a synchronously pumped femtosecond optical parametric oscillator (OPO) based on BIBO, the generated spectral range is further extended to cover the entire visible spectrum, across the green–yellow–orange–red regions. Average output powers of up to 270 mW in pulses of 120 fs at 76 MHz repetition rate have been generated across the 480–710 nm range. Internal frequency doubling of the visible OPO signal pulses in the crystal of β-BaB$_2$O$_4$ has permitted high-repetition-rate femtosecond pulse generation with wide tunability across 250–350 nm in the UV at up to 100 mW of average power. The potential of BIBO for efficient frequency conversion of high-intensity pulses is also demonstrated through third harmonic generation of amplified microjoule picosecond pulses at 1064 nm. Output pulse energies of 216 μJ in 29 ps duration at 25 Hz repetition rate have been obtained at 355 nm with overall conversion efficiencies of up to 50%.

Index Terms—Blue sources, nonlinear materials, nonlinear optics, optical frequency conversion, optical parametric oscillators (OPOs), tunable lasers, ultrashort lasers, UV sources, visible sources.

I. INTRODUCTION

COHERENT ultrafast optical sources with femtosecond and picosecond pulse duration and wide tunability are of considerable interest for a wide range of scientific and technological applications in time-domain spectroscopy, optical microscopy, frequency metrology, quantum optics, biotechnology, and nanoscale science. More than 40 years after the invention of the laser, however, substantial portions of the optical spectrum from the UV to the IR remain inaccessible to conventional mode-locked laser sources. The advent of novel vibronic laser gain media, most notably Ti:sapphire, has provided a new class of practical ultrafast solid-state lasers based on Kerr-lens-mode-locking (KLM) principles, but even in such cases, the maximum spectral coverage available is limited to at best 300–400 nm. At the same time, the restricted availability of suitable solid-state laser gain materials has confined the wavelength coverage of existing ultrafast lasers, including the Ti:sapphire, mainly to limited regions in the near-IR. This has left substantial portions of the optical spectrum in the UV, visible, and IR still inaccessible to lasers.

Nonlinear optical techniques based on frequency conversion of laser light in second-order nonlinear materials offer a highly effective method to expand the spectral range of existing laser sources. Optical second harmonic generation (SHG) and sum-frequency generation (SFG) can provide spectral extension of laser sources to shorter wavelengths, while difference-frequency generation (DFG) can extend the spectral coverage to longer wavelengths. Optical parametric generation (OPG) and amplification (OPA), and the optical parametric oscillator (OPO) can, on the other hand, provide widely tunable spectral coverage at longer wavelengths using fixed-frequency laser sources with limited or no intrinsic tuning capability. At the same time, the instantaneous nature of nonlinear gain allows frequency conversion processes to retain the temporal characteristics of the input pump laser, hence enabling wavelength generation in all temporal regimes from the continuous wave (CW) to ultrafast femtosecond time scales by an appropriate choice of pump laser. These properties make nonlinear frequency conversion processes attractive and practical techniques for the generation of widely tunable radiation in spectral and temporal regions where conventional lasers or alternative technologies are not available.

The vital element in any frequency conversion process is the nonlinear optical material. Together with the input laser and pump source, it constitutes the essential component in the practical development of any frequency conversion system. In the 1980s, the emergence of a new generation of birefringent nonlinear crystals, primarily β-BaB$_2$O$_4$ (BBO), LiB$_3$O$_5$ (LBO), and KTiOPO$_4$ (KTP), with damage thresholds far exceeding those of the more classical materials and superior linear and nonlinear optical properties, provided new impetus for the advancement of frequency conversion sources from the UV to the near-IR. Combined with the availability of novel crystalline solid-state and fiber lasers with improved spectral and spatial coherence and high power, this led to the practical development of new generations of frequency conversion sources offering unprecedented performance and operating in all temporal regimes, from the CW to the femtosecond time scales. The advent of quasi-phase-matched ferroelectric nonlinear materials, particularly periodically poled LiNbO$_3$ (PPLN) and MgO-doped PPLN and also periodically poled KTiOPO$_4$ (PPKTP) and RbTiOAsO$_4$ (PPRTA), as well as periodically poled LiTaO$_3$ (PPLT) and its derivatives, stoichiometric PPLT (sPPLT) and MgO-doped
sPPLT, and orientation-patterned GaAs also continue to have a profound impact on the development of frequency conversion sources for the near- to mid-IR, particularly in the low-intensity CW operating regimes. These developments have led to remarkable advances in frequency conversion sources over the past two decades, vastly extending the wavelength range of existing laser sources to new regions from the UV and visible to the near- and mid-IR. Yet, in different device configurations and operating regimes, major limitations still exist and important challenges still remain, requiring the continued search for alternative new nonlinear materials, laser pump sources, and more innovative design concepts.

In this paper, we describe the recent developments in a new class of ultrafast frequency conversion sources, offering unprecedented wavelength coverage in the visible and UV at exceptional average powers and efficiencies. The described systems range from low-pulse-energy, high-repetition-rate femtosecond and picosecond single-pass second harmonic (SH) generators and synchronously pumped OPOs using the KLM Ti:sapphire laser to high-pulse-energy, low-repetition-rate pi-generators and synchronously pumped OPO using the KLM Ti:sapphire laser to high-pulse-energy, low-repetition-rate picosecond single-pass second and third harmonic generators based on the mode-locked Nd:YAG laser. The key to the successful realization of the described devices is the new nonlinear optical material bismuth triborate, BiB$_2$O$_6$ (BIBO), which offers large optical nonlinearity, wide transparency from the UV to near-IR, flexible phase-matching properties, high optical damage tolerance, ready availability, and low cost.

II. BIBO: NONLINEAR OPTICAL PROPERTIES

Bismuth borate (BIBO) is a relatively new nonlinear crystal [1]–[3] with unique optical properties for frequency conversion applications in the UV, visible, and near-IR. It is an optically biaxial crystal of monoclinic point group 2 symmetry. The crystal combines the advantages of UV transparency and high optical damage threshold, as in BBO and LBO, with substantially larger optical nonlinearity, comparable to KTP. The optical transmission of BIBO extends from $\sim$ 2700 nm in the near-IR down to $\sim$ 280 nm in the UV, and as an optically biaxial crystal, it offers versatile phase-matching and wavelength-tuning characteristics. The effective nonlinear coefficient of BIBO has been reported to be as high as 3.7 pm/V [4], which is nearly twice and four times larger than those of BBO and LBO, respectively, and comparable to that in KTP. BIBO also offers large angular and spectral acceptance bandwidths, low spatial walkoff, and broadband angle tuning at room temperature. Such combination of properties makes BIBO a highly attractive nonlinear material for a wide range of frequency conversion applications in the UV, visible, and near-IR.

Since the first studies of the linear optical properties of BIBO [2], several frequency conversion experiments have been reported in this material. These include internal SHG of CW radiation at 1.06 $\mu$m [5], single-pass SHG of high-intensity pulsed laser at 1.06 $\mu$m [6], Q-switched internal SHG at 1.06 $\mu$m [7], internal frequency doubling of CW 946-nm Nd:YAG laser [8], photo-induced SHG in partially crystallized BIBO glass [9], nanosecond optical parametric oscillation and harmonic generation [10], [11], UV-induced two-photon absorption (TPA) [12], and OPA of high-intensity femtosecond pulses in the near-IR pumped at 800 nm [13]. Here, we describe efficient frequency conversion of femtosecond and picosecond pulses into the visible and UV using BIBO. In the low-intensity, high-repetition-rate regime, we demonstrate continuous coverage from 250 nm in the UV to 710 nm in the visible, average powers of up to 1 W, and single-pass conversion efficiencies in excess of 50%. In the high-energy, low-repetition-rate regime, we demonstrate pulse energies of up to 216 $\mu$J at 355 nm, with overall single-pass conversion efficiencies as much as 50%.

A. Phase-Matching Directions and Effective Nonlinear Coefficients

By using the relevant Sellmeier equations and nonlinear optical coefficients [2], the effective nonlinear coefficients $d_{eff}$ of BIBO in the principal optical coordinates ($xyz$) can be obtained [3]. The $d_{ijk}$ coefficients in [2] were measured in the physical coordinates system ($XYZ$), and because of monoclinic symmetry of BIBO, the axes in this coordinates system are not parallel to the axes in optical coordinates system ($xyz$). Therefore, in order to find the nonlinear optical coefficients in $xyz$ coordinates, the $d_{ijk}$ tensor elements given in the physical coordinates [2] need to be transformed by the prescribed rotation $\phi$ ($\lambda$) around $Y$-axis, and then, renaming the axes according to $X \rightarrow z$, $Y \rightarrow x$, and $Z \rightarrow y$. The resulting values of the nonzero tensor elements are given in Table I. The calculations were performed at a wavelength of 810 nm, but given the small wavelength dependence of the $d_{ijk}$ tensor, there should be little variation in the values of nonlinear coefficients.

Calculations of phase matching for SHG can be performed in the three principal optical planes, $xy$, $xz$, and $yz$. In the $xy$ plane, type I ($o + o \rightarrow e$) phase matching is available over limited fundamental range of 540–610 nm in the visible, with an effective nonlinear coefficient

$$d_{eff} = -d_{133} \sin \phi. \quad (1)$$

The magnitude of the effective nonlinearity $d_{eff}$ varies from $\sim$ 2 to 0.3 pm/V across the tuning range from 540 to 610 nm. Type II ($o + e \rightarrow e$) phase matching is also available in the $xy$ plane, again over a limited fundamental wavelength range of 690–790 nm. The effective nonlinear coefficient in this case is given by

$$d_{eff} = -d_{123} \sin 2\phi. \quad (2)$$
The effective nonlinearity varies between $d_{\text{eff}} \sim 0.3$ pm/V at the extremes of the tuning range to 1.65 pm/V at the center of the tuning range corresponding to a fundamental wavelength of $\sim 740$ nm.

In the $xz$ plane, there are three phase-matching possibilities. For small angles of approximately $1^\circ < \theta < 11^\circ$, type I $(o + o \rightarrow e)$ phase matching over a fundamental range of $\sim 1.18$–2.33 $\mu$m is possible, with an effective nonlinear coefficient

$$d_{\text{eff}} = d_{122} \cos \theta.$$  

In this phase-matching scheme, a relatively large effective nonlinearity, $d_{\text{eff}} \sim 2.9$ pm/V, is available across the entire tuning range. For larger angles of around $35^\circ < \theta < 80^\circ$, alternative type I $(e + e \rightarrow o)$ interaction is also available for fundamental wavelengths from $\sim 610$ nm to 3 $\mu$m, but at reduced effective nonlinearity, $d_{\text{eff}} \sim 1.6$ pm/V, across most of the tuning range. The effective nonlinearity in this case is given by

$$d_{\text{eff}} = d_{213} \sin 2\theta.$$  

In addition, type II $(o + e \rightarrow o)$ phase matching is also attainable in the $xz$ plane for fundamental wavelengths between 795 nm and 3 $\mu$m, with $d_{\text{eff}} > 2$ pm/V across most of the tuning range. The effective nonlinear coefficient for this interaction is given by

$$d_{\text{eff}} = d_{212} \cos \theta.$$  

In the $yz$ plane, type I $(e + e \rightarrow o)$ phase matching is available for angles between $\theta \sim 0^\circ$ and $\theta \sim 180^\circ$ for fundamental wavelengths from 0.542 to 1.18 $\mu$m. The same phase-matching scheme is also available for angles between $\theta \sim 160^\circ$ and $\sim 180^\circ$ and for angles between $\theta \sim 0^\circ$ and $\theta \sim 20^\circ$ in this plane for fundamental wavelengths from 2.33 to 3 $\mu$m. The effective nonlinear coefficient for this scheme is given by

$$d_{\text{eff}} = -d_{122} \cos^2 \theta - d_{133} \sin^2 \theta + d_{123} \sin 2\theta.$$  

In both configurations, the effective nonlinearity can have the largest value, $d_{\text{eff}} > 3$ pm/V, across most of the tuning range compared with all other phase-matching schemes, making $yz$ the most important plane for nonlinear optical applications. Interestingly, in the $yz$ plane, there can be two possible phase-matching solutions for SHG for a given fundamental wavelength, at angles $\theta$ and $180 - \theta$, with different effective nonlinear coefficients, indicating a degenerate phase-matching behavior. However, due to the monoclinic symmetry of BIBO, type II phase matching in $yz$ plane is not possible, because of vanishing effective nonlinear coefficient. The SHG phase-matching properties and corresponding effective nonlinear coefficients in the three principal optical planes of BIBO are summarized in Table II. It is also interesting to note that type I $(o + o \rightarrow e)$ phase matching in $xz$ plane is, in fact, the continuation of type I $(e + e \rightarrow o)$ interaction in $yz$ plane, so that the phase-match directions for wavelengths shorter than $\sim 1.18$ $\mu$m lie within the $yz$ plane, and for longer wavelengths enter the $xz$ plane, until they return to the $yz$ plane for wavelengths longer than $\sim 2.33$ $\mu$m.

### III. Experiments

From the aforementioned calculations of phase-matching and effective nonlinearity, it may be concluded that the most effective phase-matching schemes in BIBO are type I $(e + e \rightarrow o)$ in the $yz$ plane and type I $(o + o \rightarrow e)$ in the $xz$ plane. The combination of these two schemes provides tuning coverage across the full transparency of the material, while at the same time maximizing the effective nonlinearity at around $2.8 < d_{\text{eff}} < 3.4$ pm/V. In implementing the different frequency conversion devices described here, we have employed type I $(e + e \rightarrow o)$ phase matching in the $yz$ plane throughout in order to achieve broad spectral coverage in the UV, visible, and near-IR, and the highest effective nonlinearity resulting in maximum conversion efficiency, output pulse energy, and average power.

### Table II

<table>
<thead>
<tr>
<th>Optical plane</th>
<th>Phase-matching scheme</th>
<th>Fundamental wavelength</th>
<th>Phase-matching angle</th>
<th>$d_{\text{eff}}$ (pm/V)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$xy$</td>
<td>Type I $(o + o \rightarrow e)$</td>
<td>540-610 nm</td>
<td>$10^\circ &lt; \phi &lt; 90^\circ$</td>
<td>$-d_{123} \sin \phi$</td>
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<tr>
<td></td>
<td>Type II $(o + e \rightarrow e)$</td>
<td>690-790 nm</td>
<td>$5^\circ &lt; \phi &lt; 85^\circ$</td>
<td>$-d_{123} \sin 2\phi$</td>
</tr>
<tr>
<td>$xz$</td>
<td>Type I $(o + o \rightarrow e)$</td>
<td>1.18-2.33 $\mu$m</td>
<td>$1^\circ &lt; \phi &lt; 11^\circ$</td>
<td>$d_{122} \cos \theta$</td>
</tr>
<tr>
<td></td>
<td>Type I $(e + e \rightarrow o)$</td>
<td>0.610-3.0 $\mu$m</td>
<td>$35^\circ &lt; \theta &lt; 80^\circ$</td>
<td>$d_{213} \sin 2\theta$</td>
</tr>
<tr>
<td></td>
<td>Type II $(o + e \rightarrow o)$</td>
<td>0.795-3.0 $\mu$m</td>
<td>$45^\circ &lt; \theta &lt; 85^\circ$</td>
<td>$d_{212} \cos \theta$</td>
</tr>
<tr>
<td>$yz$</td>
<td>Type I $(e + e \rightarrow o)$</td>
<td>0.542-1.18 $\mu$m</td>
<td>$90^\circ &lt; \theta &lt; 180^\circ$</td>
<td>$-d_{122} \cos^2 \theta - d_{133} \sin^2 \theta + d_{123} \sin 2\theta$</td>
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<tr>
<td></td>
<td>Type I $(e + e \rightarrow o)$</td>
<td>2.33-3.0 $\mu$m</td>
<td>$160^\circ &lt; \theta &lt; 180^\circ$</td>
<td>$-d_{122} \cos^2 \theta - d_{133} \sin^2 \theta + d_{123} \sin 2\theta$</td>
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<td>Type I $(e + e \rightarrow o)$</td>
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</tr>
</tbody>
</table>
A. High-Average-Power, Widely Tunable Femtosecond Pulse Generation in the Blue

Since their introduction nearly two decades ago, BBO and LBO have been established as materials of choice for nonlinear frequency conversion into the UV and visible, owing to their broad UV transmission and high optical damage threshold. However, the relatively low effective nonlinearity ($d_{\text{eff}} \sim 1$–2 pm/V) has confined the use of BBO and LBO mainly to applications involving high pulse energies or limited phase-matching geometries, and frequency conversion in the low-intensity regime is hampered by low efficiencies and average powers. On the other hand, the substantially larger effective nonlinearity of BIBO combined with versatile phase-matching properties offers the possibility to overcome such efficiency limitations in the low-intensity regime, enabling the generation of high average powers in the UV and visible. In these experiments, we demonstrate this potential and show that the generation of high-repetition-rate femtosecond pulses with wide tunability in the blue spectral range can be achieved at exceptional average powers and efficiencies using direct single-pass SHG in BIBO.

The fundamental pulses in these experiments were derived from a KLM Ti:sapphire laser (Coherent, Mira), delivering $\sim 130$ fs pulses at 76 MHz repetition rate with an average power of up to 1.9 W tunable over 750–950 nm. Several BIBO crystals of lengths 0.4, 0.7, and 1.4 mm were used in these experiments. The crystals were grown using the top-seeded technique [8] and were cut for type I $(e + e \rightarrow o)$ phase matching in $yz$ plane ($\phi = 90^\circ$) at internal angles close to $\theta = 156^\circ$ at normal incidence. The facets were uncoated. All experiments were performed at room temperature.

Fig. 1 shows typical SHG tuning data obtained with the 1.4-mm crystal and the corresponding tuning curve calculated from the Sellmeier equations [2]. We were able to achieve wavelength tuning from 375 to 435 nm, limited by the crystal aperture at larger angles.

To obtain maximum SHG efficiency and power, an important parameter, in addition to the effective nonlinearity and fundamental intensity, is spatial walkoff. For type I $(e + e \rightarrow o)$ phase matching in $yz$ plane, the calculated walkoff angle varies between 40 and 65 mrad [3] over the tuning range of our fundamental laser (750–950 nm), implying that for a focused beam waist radius $w_0 \sim 50$ µm, say, the generated SHG pulses produced at the focus will be separated from the fundamental after $\sim 2$–4 mm of propagation. However, this length will also depend on the exact position of the focus within the crystal, so that maximization of efficiency for a given crystal length will require the optimization of focusing strength and position within the crystal. Therefore, to obtain the optimum conditions for SHG output power and efficiency, we varied the focusing conditions in each BIBO sample, using different lenses with focal lengths 60–160 mm, with the beam waist location within each sample carefully optimized.

The highest SHG output power and efficiency was obtained with the longest crystal, 1.4 mm in length, with the results shown in Fig. 2. The maximum average SH power generated in this crystal was 830 mW for 1.65 W of input fundamental power, corresponding to a conversion efficiency of 50.3%. At the maximum input power, there is evidence of saturation in SHG efficiency, with the value remaining close to $\sim 50\%$.

Another important parameter in the attainment of high conversion efficiency is the phase-matching acceptance for SHG. We calculated angular and spectral acceptance for different types of phase matching in the three principal planes of BIBO. For type I $(e + e \rightarrow o)$ phase matching in the $yz$ plane, the full-width at half-maximum (FWHM) angular acceptance widths vary from 0.28 to 0.58 mrad·cm and the spectral acceptance widths vary from 1.6 to 5.4 nm·cm across the fundamental tuning range of 750–950 nm [3].

We were able to experimentally verify our calculations through measurements of SH power in the 1.4-mm crystal by using the fundamental laser in CW operation. To minimize any phase-mismatch effects due to strong focusing, diffraction, and beam divergence, we used long distance focusing with a $f = 200$ cm lens. We also apertured the input fundamental before the crystal to eliminate any residual phase mismatch due to angular spread of the beam. Using this arrangement, we were able to measure SH powers in the nanowatts range and
accurately determine the angular acceptance for the process. The results at a fundamental wavelength of 812 nm are shown in Fig. 3, together with our numerical calculation. The measured FWHM angular acceptance bandwidth is 0.34 mrad/cm, which is in excellent agreement with the calculations.

Temporal characterization of the blue pulses was also performed using cross-correlation measurements in a thin crystal of BBO. The measurements in this case were carried out in two Brewster cut BIBO samples of lengths 0.4 and 2 mm at an SH wavelength of 406 nm. From consideration of group velocity dispersion (GVD ≈ 73, 210, and 420 fs²/mm for 812, 406, and 271 nm pulses, respectively) and mismatch (GVM ≈ 315 fs/mm between 812 nm and 406 nm pulses) in BBO, we chose a 100-μm thick crystal to ensure minimal pulse broadening for 130-fs fundamental pulses in the sample. Background-free cross-correlation intensity profiles were obtained using an AlGaN detector, with the results shown in Fig. 4. For the 0.4-mm crystal, we obtained a blue pulse duration of ~220 fs, indicating significant broadening from the fundamental pulse. The result for the 2-mm crystal is also shown in Fig. 4, where a blue pulse duration of ~500 fs is obtained. The cross-correlation profile in this case has an asymmetric shape due to the increased effects of GVM in the longer crystal, which also leads to longer blue pulse duration [14].

We also compared the performance of BIBO with BBO. Using the two BIBO crystals of lengths 0.4 and 0.7 mm, we compared the SH output power with a 0.5-mm BBO crystal, under the same experimental conditions. Optimized focusing using a f = 80 mm focal length lens was used to ensure maximum output power. The results are shown in Fig. 5. The data correspond to an SH wavelength of 406 nm. The maximum average blue power generated with the 0.4-mm BIBO was 450 mW, compared to 336 mW obtained with BBO, implying ~34% power enhancement despite a shorter crystal length.

B. High-Average-Power, Widely Tunable Picosecond Pulse Generation in the Blue

The unique nonlinear optical properties of BIBO, including large effective nonlinearity combined with low spatial walkoff and large angular acceptance bandwidths, also permit efficient frequency conversion of low-intensity, high-repetition-rate picosecond pulses at unprecedented average powers and conversion efficiencies, not attainable with BBO or LBO. The high peak intensities of femtosecond pulses used in the aforementioned experiments permitted the use of crystal interaction lengths as short as 0.4–1.4 mm, without the onset of spatial walkoff or angular acceptance bandwidth limitations, thus enabling the generation of average powers as high as 830 mW in the blue at >50% efficiency. The present experiments demonstrate that such high average powers and efficiencies can also be achieved in BIBO with low-intensity, high-repetition-rate picosecond pulses. This is generally the most difficult operating regime in the pulsed format, where the low pulse energies (few nanojoules) and low intensities (few kilowatts) place several additional and more stringent demands on the material with regard to effective nonlinearity, spatial walkoff, and angular acceptance for phase matching, which hamper single-pass conversion at practical powers and high efficiencies.
As discussed in Section II, the flexibility offered by the structural symmetry of BIBO provides several phase-matching possibilities for SHG in the three principal optical planes. The most interesting planes are $xz$ and $yz$, offering the highest effective nonlinearity under type I interaction. This is shown in Fig. 6, where the calculated effective nonlinearity of BIBO in this phase-matching geometry is also compared with that of uniaxial BBO under type I $(o + o \rightarrow e)$ interaction. From the plot, it is evident that BIBO offers substantially higher effective nonlinearities than does BBO for almost all fundamental wavelengths. In particular, type I $(e + e \rightarrow o)$ interaction in the $yz$ plane yields a maximum value of $d_{\text{eff}} \sim 3.4 \text{ pm/V}$ for angles $90^\circ < \theta < 180^\circ$ [2].

In addition to the higher effective nonlinearity, BIBO also offers substantially lower spatial walkoff, which is of great advantage for low-intensity SHG. In Fig. 7, we compare the walkoff angle under type I SHG in the $xz$ and $yz$ planes of BIBO with type I interaction in BBO, where it is clear that across the entire fundamental phase-matching range, BIBO offers substantially lower spatial walkoff than does BBO. Also, interestingly, for

wavelengths near 1 $\mu$m, the walkoff angle approaches a minimum value close to zero. This makes BIBO very attractive for SHG of CW or low-intensity pulsed Nd:YAG lasers under type I near-noncritical phase-match condition.

Finally, BIBO offers increased angular acceptance for SHG. This is shown in Fig. 8, where the FWHM angular acceptance bandwidth of BIBO under type I SHG in $xz$ and $yz$ plane is compared with that of type I BBO. Again, the improved performance of BIBO over BBO is clearly evident across the entire phase-matching range. The higher angular acceptance in BIBO increases the tolerance of phase matching to tight focusing (or poor spatial beam quality) of the fundamental in the SHG process.

The combination of relatively high nonlinearity, low spatial walkoff, and large angular acceptance make BIBO an ideal candidate for SHG of low-intensity pulses, as the experimental results with high-repetition-rate picosecond pulses in this section demonstrate.

The BIBO crystal used in these experiments was 10 mm in length and 3 mm $\times$ 10 mm in aperture. It was cut for type I $(e + e \rightarrow o)$ phase matching in the optical $yz$ plane $(\phi = 90^\circ)$ at an internal angle $\theta = 152.8^\circ$ at normal incidence, corresponding to SHG at a fundamental wavelength of $\sim 820$ nm. The crystal end-faces had antireflection (AR) coating ($R < 2\%$) centered at 800 and 400 nm. The crystal was mounted on a precision rotation stage and the focusing condition, fundamental wavelength, and crystal orientation were adjusted to yield maximum SHG power.

The fundamental pulses in this experiment were also obtained from a KLM Ti:sapphire laser (Coherent, Mira 900) in picosecond operation. The fundamental pulse length, deduced from autocorrelation measurements in a 400-µm crystal of BIBO, was 2.4 ps. The laser delivered $> 1.9$ W of average power at a pulse repetition rate of 76 MHz and was tunable over 740–900 nm. Due to the low transmission loss of AR-coated optics $(R < 1\%)$, the maximum average fundamental power of $\sim 1.9$ W, corresponding to a pulse energy of $\sim 25$ nJ and peak power of $\sim 10.4$ kW, was available at the input to the crystal. All experiments were similarly performed at room temperature.
For the attainment of maximum SHG output power and efficiency, we varied the focusing conditions, with the fundamental beam waist size and location within the crystal carefully optimized. The highest SHG output power and efficiency was obtained with a focused beam waist radius \( w_0 = 50 \mu m \). The results at a SHG wavelength of 410 nm, corresponding to normal incidence, are shown in Fig. 9. The maximum obtained SHG average power was 990 mW for 1.9 W of input fundamental power, corresponding to a conversion efficiency of \( \sim 52\% \). The highest efficiency was 54\%, obtained at a fundamental power of 1.6 W, but for higher input powers, there was evidence of saturation in efficiency, with the value remaining above 50\%. We attribute the saturation behavior to the onset of TPA in the crystal. This could be alleviated by reducing the strength of focus within the BIBO crystal to allow higher power generation in the presence of increased fundamental power.

The SHG output power was characterized by excellent stability for all input fundamental powers, with a peak-to-peak fluctuation typically \(< 2\%\). We were able to tune the SH output over 370–450 nm by altering \( \theta \) in the \( yz \) plane, while varying the fundamental wavelength. We were able to obtain high SHG powers of \( > 500 \) mW at efficiencies close to 50\% across the entire tuning range, despite the increase in the reflection loss from the AR-coated crystal faces, variation in the effective nonlinearity, and reduction in the available fundamental power away from the center of the Ti:sapphire tuning range.

We also compared the performance of BIBO with BBO under type I \((o + o \rightarrow e)\) phase matching. We used a 10-mm BBO crystal cut at \( \theta = 28.5^\circ \) for SHG at \( \sim 820 \) nm at normal incidence. We measured the SHG power and efficiency under identical conditions and by using the same optimization procedure. Due to the strong back-reflection from the uncoated BBO end-faces into the laser, we were unable to deploy higher fundamental powers than 1.5 W at the crystal, even with the use of an optical isolator. As a result, the SHG data for BBO could be obtained only to the 1.5-W upper limit. Nevertheless, at all fundamental powers, we obtained higher SHG power for BIBO than for BBO of the same crystal length. At 1.5 W of input power, we obtained an SHG power of 410 mW in BBO, after correction for the reflection loss at the crystal faces, representing a conversion efficiency of 27.3\%. The SHG power generated in BIBO at 1.5 W of input power was 750 mW, corresponding to an efficiency of 50\%. The SHG power enhancement of 22.7\% is larger than that in the femtosecond regime [15], and will be further increased at the full fundamental power of 1.9 W. This confirms the superior performance of BIBO over BBO, particularly for SHG of picosecond pulses.

In separate experiments, we directly determined the magnitude of the effective nonlinear coefficient in BIBO from measurements of SHG power in the nondepleted regime at 820 nm by using the Ti:sapphire laser in CW operation. We deployed long focusing to yield an effective length of focus \((L_f = \pi b/2 = 132.7 \) cm, where \( b \) is the confocal parameter\) much greater than the crystal length and used a 4-mm crystal to have an aperture length \((L_o = \pi^{1/2} w_o/p = 10.1 \) mm\) longer than the crystal length in order to use the appropriate relation for the calculation of \( d_{eff} \) [16]. For increased accuracy, we also recorded the SHG power at different input powers and calculated a mean value for \( d_{eff} \) from the fit to the data. Using the aforementioned values, a walkoff angle \( \rho = 58 \) mrad and a measured fundamental beam waist radius of \( w_o = 332 \) \( \mu m \) inside the crystal, we obtained an effective nonlinear coefficient \( d_{eff} = 3.7 \) pm/V for type I \((e + e \rightarrow o)\) phase matching in optical \( yz \) plane. This is larger than the value of 3.2 pm/V reported previously using the Maker fringe technique [2].

Temporal characterization of the blue pulses was performed using autocorrelation measurements in a 200-\( \mu m \) BBO crystal \((\theta = 80^\circ)\) with a GaAsP detector. Background-free intensity and interferometric autocorrelations were obtained, with a typical profile shown in Fig. 10. The measurement was performed at 415 nm, due to the phase-matching cutoff in the BBO crystal near the normal incidence wavelength of 410 nm. The pulse duration deduced from the autocorrelation profile is 2.8 ps for fundamental pulses of 2.4 ps, indicating some broadening due to the combined effects of GVD (154 fs\(^2/\)mm at 830 nm, 400 fs\(^2/\)mm at 415 nm) and mismatch (380 fs/\)mm in the 10-mm BIBO crystal [3].

C. Widely Tunable Femtosecond OPO for the Visible and UV

The experiments described in Sections III-A and III-B demonstrate the potential of BIBO for efficient frequency conversion of
high-repetition-rate femtosecond and picosecond pulses into the blue through SHG of the KLM Ti:sapphire laser. However, access to broader spectral regions in the UV and visible, which are of interest for a wide range of applications, has remained challenging for many years. A particularly difficult spectral range is the 500–700 nm in the visible, which remains inaccessible to the KLM Ti:sapphire laser or its SH output. Spectral regions below 350 nm can be accessed by third harmonic generation (THG) of the Ti:sapphire laser, but practical generation is hampered by low average powers and conversion efficiencies.

In the experiments described here, we demonstrate a new source of high-repetition-rate femtosecond pulses with wide tunability and practical average powers across the entire spectral gap of 500–700 nm in the visible, as well as the 250–350 nm in the UV. This is achieved by exploiting the high average power femtosecond pulses generated by single-pass SHG of KLM Ti:sapphire laser in BIBO (see Section III-A) to achieve frequency conversion into the visible using the synchronously pumped OPO approach. Further, by internal SHG of the visible OPO signal pulses, we have generated high-repetition-rate femtosecond pulses with wide tunability and at unprecedented average powers in the UV.

Synchronously pumped OPOs offer a practical solution for the generation of high-repetition-rate femtosecond pulses in new spectral regions, particularly in the near- to mid-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, 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 using synchronously pumped OPOs. One technique relies on direct pumping of femtosecond OPOs with the KLM Ti:sapphire laser and subsequent SHG of the near-IR signal pulses into the visible internal to the OPO cavity [17]. Such systems have been based on KTP or RTA as the OPO gain material, and 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 about 80 nm from 580 to 660 nm. The second method has been based on frequency doubling of a KLM Ti:sapphire laser in 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 [18]. The combination of the two methods has enabled the generation of femtosecond pulses in the visible, 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. The synchronously pumped OPO described here 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 [20]. The device is based on BIBO, both as the SHG crystal for the Ti:sapphire pump and as the nonlinear gain medium for the OPO. Internal SHG of the OPO signal pulses is also achieved in the crystal of BBO, providing high-repetition-rate femtosecond pulses with wide tunability and practical average powers in the UV, for the first time.

The configuration of the visible femtosecond OPO based on BIBO is shown in Fig. 11. The OPO is synchronously pumped by the SH of the same KLM Ti:sapphire laser (Coherent, Mira 900), delivering 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 crystal of BIBO. The SHG crystal used in this experiment was 1 mm long and cut for collinear critical type I \((e + e \rightarrow o)\) interaction in yz plane \((\phi = 90^\circ)\) at an internal angle \(\theta \sim 152^\circ\) at normal incidence, yielding a maximum calculated effective nonlinear coefficient, \(d_{eff} \sim 3.3 \text{ pm/V}\) [3]. An average power of >1 W in the blue at >50% efficiency was available over a tunable range of 375–435 nm [3], [15]. The blue pulses had durations of ~220 fs. All experiments were performed at room temperature.

The blue pump beam was focussed to a waist radius \(w_0 \sim 25 \mu\text{m}\) inside a second BIBO crystal, the gain element for the femtosecond OPO. We employed collinear phase matching, with the crystal cut for type I \((o \rightarrow e + e)\) interaction in yz plane \((\phi = 90^\circ)\) at an internal angle \(\theta \sim 159^\circ\). From considerations of GVM between the blue pump and visible signal pulses (GVM ~101–312 fs/mm over 500–700 nm), we used a length of 500 µm for the OPO crystal. The end-faces of the crystal were AR-coated for the signal \((R < 0.5\%\) at 500–700 nm) and had high transmission for the blue pump \((T > 95\%)\) at 375–435 nm). The OPO was configured in a three-mirror, standing-wave cavity comprising two concave reflectors \((M_1\text{ and } M_2)\) of radius of curvature \(r = 100 \text{ mm}\) and a plane output coupler \((M_3)\). The concave mirrors were highly reflecting \((R > 99\%)\) for signal wavelengths over 500–680 nm and highly transmitting \((T > 90\%)\) for the blue pump over 380–450 nm. The mirrors also had high transmission for the idler \((T > 80\%)\) at 900–3000 nm), thus ensuring singly resonant oscillation. Two uncoated Brewster cut fused silica prisms provided intracavity dispersion compensation.

Fig. 12 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
\((o \to e + e)\) phase matching in the optical \(yz\) plane obtained using the Sellmeier relations for BIBO [2], where good agreement between the experimental data and theoretical calculation is evident. The OPO could 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 \(\theta = 175^\circ\) and \(\theta = 154^\circ\). The corresponding tuning range of the idler was from 3060 to 999 nm. For a given crystal angle, wavelength tuning was also available through the variation of OPO cavity length. We typically obtained \(\sim 10\) nm of signal tuning for a change in 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.

In order to optimize the 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 \(\sim 620\) 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 OPO mirrors away from the center of 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 reflectance 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 was performed using autocorrelation measurements in a 500-\(\mu\)m crystal of BBO cut for type I \((o + o \to e)\) phase matching at \(\theta = 42^\circ\) and a UV-enhanced silicon photodiode. Without GVD compensation, the signal pulses were strongly chirped, with corresponding broadband double-peaked spectra, characteristic of self-phase-modulation. The time duration deduced from autocorrelation measurements was \(\sim 170\) fs, whereas the corresponding spectrum has a bandwidth as wide as \(\sim 15\) nm (FWHM) due to the effects of self-phase-modulation, resulting in a time–bandwidth product \(\Delta\nu\Delta\tau \sim 2.2\), around seven times the transform limit.

We, thus, implemented dispersion compensation by introducing a pair of uncoated Brewster cut fused silica prisms within the OPO cavity. Fig. 13(a) and (b) shows 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. 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/\(\mu\)m for fundamental wavelengths from 500 to 700 nm. At a fundamental wavelength of 595 nm, the GVM in is 422 fs/\(\mu\)m, 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 for autocorrelation should enable confirmation of shorter signal pulse durations.

The wavelength range of the BIBO femtosecond OPO was also extended to the UV spectral range through SHG of the visible signal pulses internal to the OPO cavity. A schematic of the experimental setup is depicted in Fig. 14. The configuration of the OPO resonator in this case is identical to that used for visible pulse generation, except for the introduction of an additional pair of mirrors (M\(_4\) and M\(_5\)) to provide a second intracavity focus for SHG of signal pulses. The mirrors are of radius of curvature \(r = 100\) mm, are highly reflecting for signal wavelengths over 500–680 nm, and have \(>75\%\) transmission over 250–350 nm in the UV. The blue pump beam focusing conditions and other parameters are identical to those used in the visible OPO configuration. However, in order to maximize the intracavity signal intensity and, hence, the generated UV power, we used minimal output coupling by employing a high reflector (\(R > 99\%\) over 500–680 nm) as M\(_3\). The SHG crystal was a
BIBO sample, placed at the second cavity focus formed by M₄ and M₅. The choice of BBO as the SHG crystal instead of BIBO was governed by its deeper UV transparency (~190 nm), its broader phase-matching range in the UV, and its higher effective nonlinearity for SHG wavelengths below ~340 nm. The BBO crystal was 500-µm long and was cut for collinear type I (o + o → e) phase matching at an internal angle θ = 42°. The end-faces were AR-coated at 500–680 nm and >90% transmitting over 250–350 nm.

In this operating configuration, the OPO produced up to 100 mW of total average power in the UV in two counterpropagating beams (Fig. 14) for 800 mW of blue pump power, corresponding to an extraction efficiency of 12.5%. The use of a ring resonator for the OPO should enable the extraction of the generated UV power in a single direction. Tuning was continuous across 250–350 nm in the UV, limited by the drop in the visible signal power from the OPO at the extremes of the tuning range. Fig. 15 shows combined optimized output power performance of the OPO in the visible and UV configurations, demonstrating average output powers of up to 270 mW in the visible and up to 100 mW in the UV. Temporal characterization of the UV pulses using the cross-correlation technique, as well as implementation of a ring resonator for the OPO, is currently underway with a view to further enhancing the overall utility of the system.

D. Efficient Frequency Conversion of High-Energy Picosecond Pulses into the UV

As demonstrated in Sections III-A–III-C, the unique and versatile nonlinear optical properties of BIBO permit femtosecond and picosecond pulse generation in the visible and UV with exceptional efficiency and average power under low-intensity, high-repetition-rate operating conditions. However, in the experiments described in this section, we show that BIBO also offers great promise for efficient frequency conversion of high-energy, low-repetition-rate picosecond pulses into the visible and UV, owing to a high optical damage threshold, chemical and thermal stability, availability in high optical quality and large size, and overall favourable linear optical properties.

We demonstrate THG of microjoule picosecond pulses at 355 nm at >50% conversion efficiency. Similar recent experiments using high-energy femtosecond pulses also confirm the potential of BIBO for efficient ultrafast frequency conversion in the high-intensity pumping regime [13].

A schematic of the experimental setup is shown in Fig. 16. The fundamental pulses were derived from an amplified mode-locked Nd:YAG laser at 1064 nm (EKSPLA, PL2140), delivering picosecond pulses with energies up to 1 mJ at a repetition rate of 25 Hz. The fundamental pulse length, deduced from autocorrelation measurements in a 1-mm crystal of BIBO (φ = 90°, θ = 152°), was 35 ps. The fundamental beam was determined by direct measurements to have a TEM₀₀ spatial profile with an M² ~ 1 and a beam diameter (full width at 1/e² level) of 2w₀ = 2 mm.

For the attainment of maximum THG efficiency, we investigated three different experimental setups. We also used several different BIBO crystal samples and various focusing and collimating conditions for the fundamental and SH pulses. The best results were obtained with the optimized setup shown in Fig. 16. The design allows direct control of the temporal and spatial overlap between the fundamental and SH pulses using two separate optical paths and a delay line [21]. It also enables arbitrary variation of the energy ratio between the fundamental pulses.
and SH pulses in the third harmonic crystal, and provides an undepleted fundamental pulse for the THG process. The control of temporal and spatial overlap and energy ratio between the fundamental and SH, and the use of an undepleted fundamental, result in the highest conversion efficiency and pulse energy as well as the shortest pulse duration at 355 nm.

In the experimental setup of Fig. 16, the ratio of fundamental and SH pulse energies is varied using a half-wave-plate and polarizing cubic beam-splitter, which is AR coated at 1064 nm. The delay line is provided by four 45° polarizing cubic beam-splitters, which is AR coated at 1064 nm. The undepleted fundamental and generated SH pulses are then combined using a dichroic mirror (highly reflective at 1064 nm, highly transmitting at 532 nm) before entering the third harmonic crystal. The highest pulse energy and conversion efficiency at 355 nm were obtained with no focusing for both fundamental and SH beams.

We investigated several different crystals of various lengths and apertures. The best results were obtained with a 10-mm crystal for SHG and a 3.26-mm crystal for THG. We found that the use of longer THG crystals resulted in lower conversion efficiency at 355 nm due to the increased effects of TPA. Both crystals were uncoated and cut for type I \( (e + e \rightarrow o) \) phase matching in the optical \( yz \) plane \( (\phi = 90^\circ) \) to maximize the effective nonlinear coefficients \([3]\). For the SHG stage we used a crystal, 3 mm \( \times \) 4 mm in aperture, cut at \( \theta = 169^\circ \), with a calculated effective nonlinearity \( d_{\text{eff}} = 3.3 \, \text{pm/V} \) \([3]\). For the THG stage, we used a crystal, 5 mm \( \times \) 6 mm in aperture, cut at \( \theta = 146^\circ \), with corresponding \( d_{\text{eff}} = 2.9 \, \text{pm/V} \) \([3]\). All experiments were performed at room temperature.

In order to obtain the highest THG efficiency, at a given fundamental pulse energy, we adjusted the ratio of the fundamental and SH pulse energies at the input to the THG crystal using the half-wave-plate and beam-splitter combination. In Fig. 17, the maximum THG pulse energy and efficiency are plotted as a function of the total (fundamental plus SH) pulse energy at the input to the THG crystal. The data have been corrected for the reflection losses at the uncoated facets of the THG crystal.

For each fundamental pulse energy, the ratio of the fundamental and SHG pulse energies was optimized to yield the maximum THG output. Over the range of measurements, the ratio of the fundamental to SH pulse energy varied between 1 and 1.5. As is evident from the plot, single-pass conversion efficiencies in excess of 50% and maximum pulse energies of up to 216 \( \mu \text{J} \) at 355 nm were obtained for a total pulse energy of 441 \( \mu \text{J} \) at the input to the THG crystal.

Temporal characterization of the 355-nm pulses was performed using noncollinear cross-correlation between the fundamental and third harmonic pulses in a 2-mm BBO crystal \( (\theta = 20^\circ) \), and the generated pulse energy at 266 nm was recorded as a function of delay between the two pulses. A typical background-free cross-correlation trace, at the maximum THG pulse energy, is shown in Fig. 18. To obtain the THG pulse duration, we performed deconvolution of the cross-correlation result in Fig. 18 with that of the fundamental pulse, and assuming a Gaussian temporal profile, this leads to a THG pulse duration of 29 ps (FWHM). The corresponding SHG pulse duration was also determined using the experimental setup of Fig. 16, through collinear cross-correlation between the fundamental and SH pulse in the same third harmonic crystal \( (\phi = 90^\circ, \theta = 146^\circ) \). This resulted in an SHG pulse length of 40 ps.

E. Two-Photon Absorption (TPA)

An important parameter in frequency conversion under high-energy, high-intensity conditions is multiphoton absorption. In BIBO, with a UV absorption edge near 280 nm, TPA may be a significant factor in the attainment of the highest conversion efficiency in our experiments. To ascertain the role of TPA, we performed absolute measurements of TPA coefficient in BIBO at 355 nm using the transmittance approach \([19]\) in an uncoated sample under nonphase-matched conditions. The crystal was cut at \( \theta = 155^\circ (\phi = 90^\circ) \) at normal incidence and had dimensions of 5 mm \( \times \) 10 mm \( \times \) 10 mm long. The generated third harmonic beam was focused inside the crystal, and we recorded the transmittance as a function of input pulse intensity at 355 nm by varying the input pulse energy. The diameter (full width at \( 1/e^2 \) intensity level) of the 355-nm beam at the center of the crystal was \( 2w_o \sim 770 \, \mu\text{m} \). The TPA coefficient was determined for two orthogonal linear polarizations of the input beam at 355 nm, an ordinary \( (o) \) polarization parallel to the crystallographic b-axis (optical x-axis) and an extraordinary \( (e) \) polarization in the crystallographic c-plane (optical yz-plane).

The results of measurements for the \( o \) and \( e \) polarizations are shown in Fig. 19, where the percentage crystal transmittance at...
355 nm is plotted against the input pulse intensity. It is evident from the plots that BIBO exhibits higher absorption for the e polarization than for the o polarization.

Assuming a Gaussian profile for the input pulses in time and space, the energy transmittance through the nonlinear crystal of length $l$ in the presence of linear absorption is given by [12], [19]

$$T = \frac{\alpha T_0}{\sqrt{\pi} \beta I_0 (1 - R)(1 - e^{-\alpha l})} \times \int_{-\infty}^{+\infty} \ln \left( 1 + \frac{\beta}{\alpha} I_0 (1 - R)(1 - e^{-\alpha l}) e^{-\kappa^2} \right) d\kappa \quad (7)$$

where $\alpha$ is the linear absorption coefficient, $T_0 = (1 - R)^2 e^{-\alpha l}$ is the initial transmittance, $\beta$ is the TPA coefficient, $I_0$ is the maximum on-axis intensity, $R = (n - 1)^2/(n + 1)^2$ is the Fresnel reflectivity of crystal faces, and $\kappa = 2\tau/\tau_p$. The variable $\tau_p$ is the pulse width at $1/e$ level (the FWHM pulse width $\tau$ is given by $\tau = \tau_p \sqrt{\ln 2}$). To obtain the TPA coefficient, we first evaluated the linear absorption coefficients at 355 nm from the experimental transmission values by correcting for the Fresnel reflection loss at the uncoated crystal faces. Using the aforementioned equation, we calculated the TPA coefficient $\beta_e$ corresponding to the theoretical curve passing through this point and the initial transmittance value ($T_o = 0.812, T_e = 0.759$). The overall TPA coefficient value was determined as arithmetic average of all $\beta_i$ related to the corresponding curve in Fig. 4. For o polarization, we obtained $\alpha_o = 0.022$ cm$^{-1}$ and $\beta_o = 0.71$ cm/GW. For e polarization, we obtained $\alpha_e = 0.067$ cm$^{-1}$ and $\beta_e = 1.37$ cm/GW. These values are substantially lower than $\beta_o = 7.1$ cm/GW and $\beta_e = 16$ cm/GW reported previously [11] under nanosecond pulse radiation at 355 nm in the same optical plane (yz), but at $\theta = 37^\circ$ ($\phi = 90^\circ$). The origin of this discrepancy is not clear, but may be due to the more complicated nature of intensity-dependent absorption with nanosecond pulses compared with picosecond pulses.

In addition, our measurements confirm that TPA is not an important factor in limiting the maximum THG conversion efficiency at $\sim 50\%$. At the THG pulse energy of 67 $\mu$J ($I_0 \sim 0.18$ GW/cm$^2$), corresponding the onset of saturation in conversion efficiency (Fig. 2), the calculated TPA loss at 355 nm in 3.26-mm BIBO crystal is $\sim 1.3\%$. At the maximum THG pulse energy of 216 $\mu$J ($I_0 \sim 0.57$ GW/cm$^2$), the corresponding TPA loss is $\sim 4\%$. These imply that the saturation in conversion efficiency in Fig. 17 may be more significantly due to pump–depletion effects of fundamental and SH pulses than TPA.

We also investigated the possible presence of TPA at the SH wavelength of 532 nm in the same 10-mm BIBO crystal, but did not observe any significant absorption at focused intensities of up to 3.5 GW/cm$^2$. We also did not observe any sign of optical damage to the BIBO crystals under picosecond pulse radiation in our experiments at 1064, 532, and 355 nm at optical intensities of up to 1 GW/cm$^2$, confirming the promise of BIBO for frequency conversion of high-intensity pulses into the visible and UV.

**IV. CONCLUSION**

In this paper, we have described efficient generation of femtosecond and picosecond pulses with wide tunability across the UV and visible spectral regions using optical frequency conversion techniques in the new nonlinear material BIBO. We have demonstrated the generation of high-repetition-rate femtosecond and picosecond pulses with wide tunability across the 370–450 nm in the blue spectral range at average powers of nearly 1 W with conversion efficiencies in excess of 50% using simple single-pass SHG of the KLM Ti:sapphire laser. The limit to the demonstrated tuning range was set by the tuning range of the fundamental laser or the limited aperture of the BIBO crystal, and so could be readily extended to cover the 350–500 nm range corresponding to the full tuning potential of the Ti:sapphire laser. We have reported the generation of widely tunable high-repetition-rate femtosecond pulses across the entire visible range of 480–710 nm by developing a synchronously pumped OPO based on BIBO, and achieved wavelength extension to 250–350 nm in the UV using intracavity SHG of the visible signal pulses in BBO. The combination of single-pass SHG, synchronously pumped OPO, and internally frequency-doubled OPO provide continuous coverage across the entire 250–710 nm spectral range at room temperature. Combined with the tunability of the fundamental Ti:sapphire laser, the approach provides an exceptionally versatile and continuously tunable source of high-repetition-rate femtosecond pulses from the UV to near-IR across 250–1000 nm. We have also demonstrated the promise of BIBO as a viable material for frequency conversion in the high-intensity regime through THG of microjoule picosecond pulses at 355 nm, where overall conversion efficiency of as much as 50% have been achieved without the onset of optical damage or TPA limitations.

The uniquely versatile nonlinear optical properties of BIBO combined with the frequency conversion methods described here provide efficient and widely tunable ultrafast sources across the UV and visible, offering the advantages of simplicity,
practicality, high average power and pulse energy, and convenient operation at room temperature.

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