More than 40 years after the invention of the laser, the generation of femtosecond pulses in the visible and ultraviolet (UV) remains beyond the reach of mode-locked, solid-state lasers. The advent of novel vibronic gain media has provided a new generation of femtosecond sources, most notably the Kerr-lens-mode-locked (KLM) Ti:sapphire laser. Nevertheless, the spectral range available to such systems remains confined to the near-infrared. Nonlinear frequency conversion can provide spectral extension to shorter wavelengths, but important regions in the visible and UV remain uncovered. A particularly difficult spectral range is the 500 to 700 nm in the visible, which cannot be accessed by the KLM Ti:sapphire laser or its frequency doubled output. Spectral regions below 350 nm can be reached by frequency tripling of the Ti:sapphire laser. However, practical generation is hampered by low average powers and conversion efficiencies.

Synchronously pumped optical parametric oscillators (SPOPOs) represent viable sources of high-repetition-rate femtosecond pulses in spectral regions that are inaccessible to lasers. When pumped directly by the KLM Ti:sapphire laser, SPOPOs can provide femtosecond pulses across the 1 to 5 µm range in the near- to mid-infrared. However, pulse generation in the visible and UV has remained similarly difficult due to a lack of suitable nonlinear materials and mode-locked pump lasers at shorter wavelengths.

To overcome this obstacle, two techniques have been previously deployed with the SPOPO approach based on the direct use of the KLM Ti:sapphire laser as the pump source and subsequent up-conversion of the femtosecond signal pulses into the visible, or the use of a frequency-doubled KLM Ti:sapphire laser to pump a visible SPOPO. The combination of these methods has enabled femtosecond pulse generation over a total tuning range of 566 to 676 nm. However, the limited phase-matching capabilities of KTP, RTA and BBO deployed in these systems have prevented femtosecond pulse generation outside this range.

We have now developed a new source of broadly tunable femtosecond pulses for the visible and UV based on a single SPOPO system. The oscillator, which is pumped by the second harmonic of a KLM Ti:sapphire laser, can provide wide and continuous wavelength coverage across the entire green-yellow-orange-red (480 to 710 nm) using a single nonlinear crystal and a single set of mirrors, and can deliver average powers of 270 mW in 120-fs pulses at a 76 MHz repetition rate. The key to the successful realization of the device is the new nonlinear material, bismuth triborate (BiB₃O₆ or BBO), which is used both as the doubling crystal for the Ti:sapphire pump and as the gain element for the SPOPO.

Moreover, by internal SHG of the visible signal pulses, we have for the first time generated tunable femtosecond UV pulses across 240 to 355 nm at practical average powers of up to 100 mW. The combination of the KLM Ti:sapphire pump laser and the SPOPO system provides a unique source of femtosecond pulses across the entire 240 to 1,000 nm spectral range. Such a source will be of practical interest for a wide range of scientific and technological applications in time-domain spectroscopy, frequency metrology, quantum optics, biophotonics and nanotechnology.

The author would like to thank M. Ghotbi, A. Esteban-Martin, A. Majchrowski, I.V. Kityk and E. Michalski for their contributions leading to the work described here.

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Photographs of the synchronously-pumped BiB₃O₆ femtosecond optical parametric oscillator operating across the entire green-yellow-orange-red regions of the visible spectrum.