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Tunable third-harmonic probe for non-degenerate ultrafast pump–probe measurements

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Abstract. In this article, we report a method to achieve a precisely tunable highly stable probe beam generation for performing pump–probe experiment around a given wavelength by tilting a sum frequency generation (SFG) crystal angle. The width of the generated third-harmonic beam is of the order of 2 nm throughout the tunable range. This method of probe beam generation has its application in isolating contributions from closely separated excitation states.

Keywords. Third harmonic generation; pump–probe spectroscopy.

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Subpicosecond ultrashort pulses are used for studying ultrafast processes in materials [1–3]. The time resolution is achieved by using pump–probe geometry. Often it is required to vary the pump and/or probe wavelength across a material resonance, which requires some wavelength tuning of the laser. For example, the contribution from closely separated energy levels in semiconductor nanostructures can be isolated by tuning the probe or excitation wavelength [2]. For such studies, wavelength tunability is usually achieved by an optical parametric amplifier (OPA) or a non-collinear OPA pumped by a femtosecond oscillator–amplifier combination. This allows for a wide tunability range as by using frequency conversion in nonlinear crystals, the wavelength can be tuned from ultraviolet (UV) to infrared (IR). This tunability, however, comes with a disadvantage. Most amplifier–OPA systems work at KHz rep-rates as compared to the MHz rep-rates available with femtosecond oscillators. The power stability of the oscillator output is much better, allowing for very sensitive pump–probe measurements. Furthermore, since the spectral width of a femtosecond pulse is large, frequency selective elements (e.g. etalons) need to be inserted in the beam to reduce the spectral width for studying materials having an energy level separation less than the pulse spectral width [4, 5]. We demonstrate an alternate method that addresses all these issues. It would be useful where a limited range of tunability along with high sensitivity measurements is required. In this article, we report a method to achieve a precisely tunable, highly stable UV probe source by using sum frequency generation (SFG).

The experimental set-up used to generate the tunable UV pulses is shown in figure 1. Ti:sapphire laser oscillator provides ∼80 fs, 500 mW pulses at 800 nm with 82 kHz repetition rate. A beam splitter was used to split the beam into two with the power ratio of 20 : 80. The stronger beam was used to generate the second harmonic of the fundamental 800-nm pulse. To achieve a broad second harmonic spectrum and to have high conversion efficiency, a 0.2-mm thick beta-barium borate (β-BaB_2O_4 or BBO) crystal was used for second harmonic generation (SHG). The unconverted fundamental beam was blocked by colour glass fitters. The 400-nm beam from the SHG crystal was collimated using a 10-cm focal length lens. The 400-nm beam was focussed on another BBO crystal with a 15-cm focal length lens. The power of frequency-doubled beam at the second crystal was measured to be 20 mW. The weaker 800-nm beam was also brought to the same crystal after passing through a variable delay line in order to time match the 800-nm and 400-nm beams at the crystal. The two beams interacted in the crystal to generate UV radiation at the sum frequency that corresponds to a wavelength of 267 nm. For the SFG, we have used type I non-collinear geometry. Since the fundamental and second-harmonic beams have orthogonal polarizations, a half-wave plate–polarizer combination was used to rotate the polarization of the fundamental beam. The BBO crystal used for SFG of 400 nm and 800 nm has a cut angle of 44.3° and was mounted on a rotating stage. The mounting was done in such a way that the position of the beams do not shift on the SFG crystal during rotation. The axis of crystal rotation is perpendicular to the plane of incidence.

To achieve temporal matching between the 800-nm and 400-nm beams, we have at first matched the fundamental beams at the SFG position using another SHG crystal cut at 29°. For doing this, the unconverted 800-nm beam was allowed to pass through, while the 400-nm beam was blocked by suitable filters. After this time matching, the filter was changed to allow only 400-nm beam and the SHG crystal was replaced by the SFG crystal. Now polarization of the 800-nm beam is made to be parallel with the 400-nm beam. The delay between the pulses is now corrected for the difference in the path length of 400 nm and 800 nm. The final spatial overlap is confirmed using a CCD camera. The optical generated ∼266 nm beam was monitored with a fibre-based spectrograph (Ocean Optics HR4000).

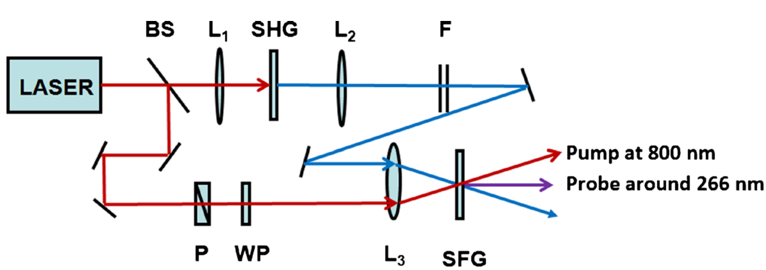

Figure 1. The schematic of the pump and probe generation set-up. BS – Beam splitter, L – lenses, SHG – second-harmonic crystal, F – colour filters, P – polarizer, WP – wave plates and SFG – sum frequency crystal.

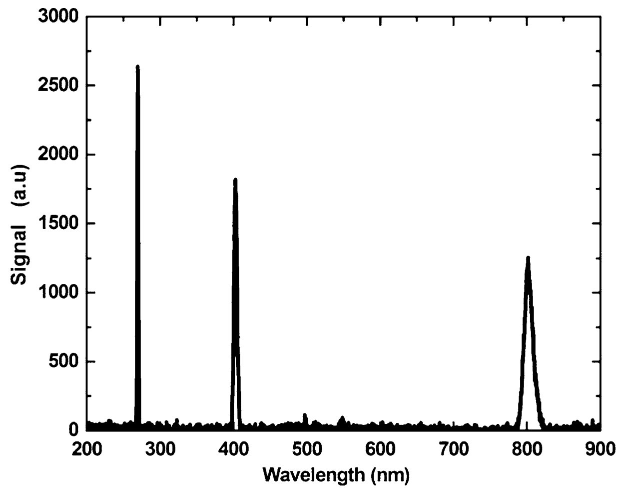

Figure 2. The complete spectrum showing the fundamental, second-harmonic and third-harmonic beams.

Figure 2 shows the recorded spectrum of all the three beams: the fundamental, the second-harmonic and the third-harmonic beams. The recorded spectrum clearly shows that the spectral width of the 400-nm pulse is less than that of the 800-nm beam. The ∼266 nm pulse is even narrower. The full-width half-maximum (FWHM) of the fundamental, second-harmonic and third-harmonic beams were found to be 14 nm, 5 nm and 2 nm, respectively. Figure 3 shows the variation of the peak power and energy of the third-harmonic beam as a function of the crystal tilt angle. Both peak at the theoretically calculated phase-matching angle for 800 nm. The width of the angle tuning curve is due

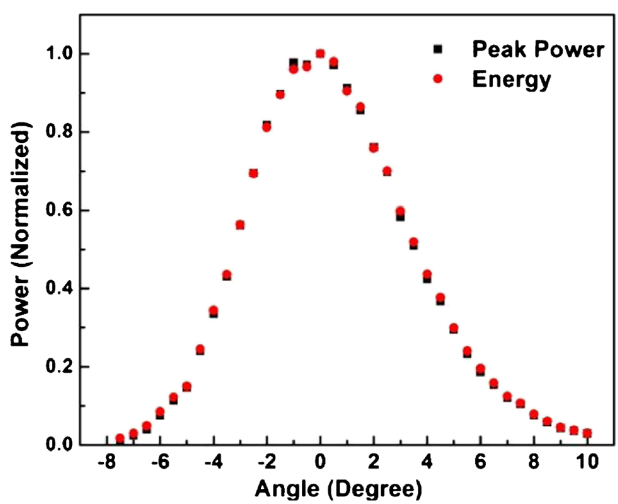

Figure 3. Variation of the peak power and integrated energy of the generated third-harmonic beam with the tilt angle.

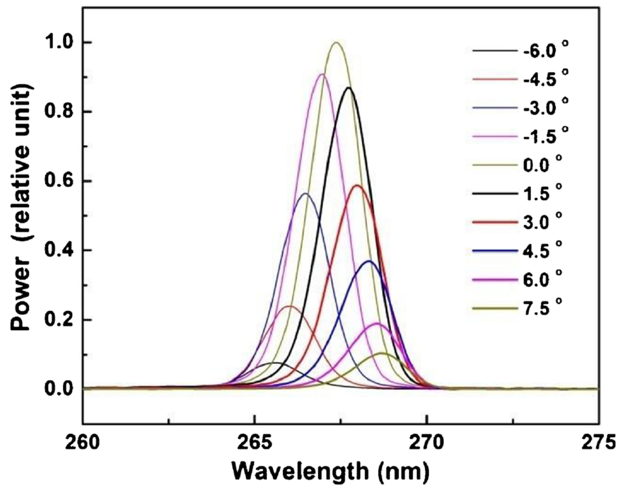

Figure 4. The variation of spectrum of THG pulses with the crystal angle.

to the large spectral width of the femtosecond pulse. At different tilt angles, different spectral components could be producing the harmonic generation. Therefore, the third-harmonic spectrum was recorded as a function of the crystal tilt angle, some of which are shown in figure 4. A continuous shift of the peak wavelength is observed in the third-harmonic spectrum. Interestingly, the width of the third-harmonic beam remains of the order of 2 nm. Thus, we are able to generate a narrow spectrum pulse and fine-tune the wavelength over a short range (∼ 4 nm). Figure 5 shows the smooth wavelength shift as the crystal angle is changed.

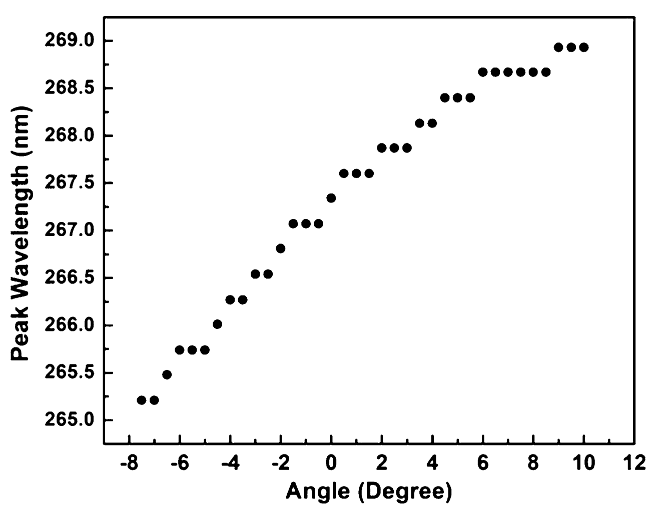

Figure 5. Variation of peak wavelength of third-harmonic beam as a function of tilt angle.

The tunability of wavelength of the SFG pulse by changing the crystal tilt angle is achieved with femtosecond pulses due to their broad spectral width. When a beam of finite spectral width is incident on the SFG crystal at a tilt angle, only a part of the spectrum is converted to third-harmonic due to the variation in the phase-matching condition for different wavelengths [6]. For a small range of tilt angles, the range of wavelengths for which phase-matching condition can be achieved lies within the spectrum of the fundamental pulse. As the angle changes, the phase matching ($\Delta K = 0$) at the central frequency is not the best choice for the maximum conversion efficiency but shifts to other neighbouring wavelengths. There is an additional angle dependence caused by the focussing of the two incident beams. The net effect is to generate a narrow-band tunable beam.

In conclusion, we have shown that tuning of the peak wavelength of non-collinear third-harmonic is obtained by tilting the SFG crystal and can provide a suitable UV source for high-resolution ultrafast pump–probe experiments. It is much easier to extend this method to tune the wavelength of the laser around its second-harmonic, which will have broader tunability. This method of generating short-duration, highly stable, short-range tunable probe beams has its application in isolating contributions from closely separated excitation states.

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