We use a compact chirped-pulse amplified system to harmonically generate ultra-short pulses for aerosol fluorescence measurements. The seed laser is a compact, all-normal dispersion, mode-locked Yb-doped fiber laser with a 1050-nm center wavelength operating at 41 MHz. Average powers of more than 1.2 W at 525 nm and 350 mW at 262 nm are generated high-dispersion fiber, amplified by a high-power, large-mode area fiber amplifier, and recompressed using a chirped volume holographic Bragg grating. The resulting high-peak-power pulses allow for highly efficient harmonic generation. We also demonstrate, for the first time to our knowledge, the use of a mode-locked ultra-violet source to excite individual biological particles and other collaboration particles in an inlet air flow as they pass through an optical chamber. The repetition rate is ideal for bio-fluorescence measurements as it allows faster sampling rates as well as the higher peak powers as compared to previously demonstrated Q-switched systems while maintaining a pulse period that is longer than the fluorescence recovery time. Thus, the fluorescence excitation can be considered to be quasicontinuous and required no external synchronization and triggering.
Green and Ultraviolet Pulse Generation with a Compact, Fiber Laser, Chirped-Pulse Amplification System for Aerosol Fluorescence Measurements

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Abstract—We use a compact chirped-pulse amplified system to harmonically generate ultra-short pulses for aerosol fluorescence measurements. The seed laser is a compact, all-normal dispersion, mode-locked Yb-doped fiber laser with a 1050-nm center wavelength operating at 41 MHz. Average powers of more than 1.2 W at 525 nm and 350 mW at 262 nm are generated with <500-fs pulse durations. The pulses are time-stretched with high-dispersion fiber, amplified by a high-power, large-mode-area fiber amplifier, and recompressed using a chirped volume holographic Bragg grating. The resulting high-peak-power pulses allow for highly efficient harmonic generation.

We also demonstrate, for the first time to our knowledge, the use of a mode-locked ultra-violet source to excite individual biological particles and other calibration particles in an inlet air flow as they pass through an optical chamber. The repetition rate is ideal for bio-fluorescence measurements as it allows faster sampling rates as well as the higher peak powers as compared to previously demonstrated Q-switched systems while maintaining a pulse period that is longer than the fluorescence recovery time. Thus, the fluorescence excitation can be considered to be quasi-continuous and requires no external synchronization and triggering.

Index Terms—Fiber lasers, Mode-locked lasers, Nonlinear optical devices, Harmonic generation, Ultrafast spectroscopy, Aerosol detection, Laser-induced fluorescence

I. INTRODUCTION

The generation of green and ultra-violet wavelength, mode-locked pulses are of great interest for fluorescence measurements of biological media [1], micro-machining, and many other applications that require either high-peak power or quasi-continuous ultra-violet lasers. Bio-threat detection sensors developed recently have used laser induced fluorescence measurement to provide an initial rapid alert, or sensors developed recently have used laser induced fluorescence measurement to provide an initial rapid alert, or...
significantly reduces the size and complexity of the pulse compressor as compared to the conventional Treacy compressor [7] as well as increases the power handling capability.

We have recently demonstrated the application of a mode-locked green laser to two-photon fluorescence cross-section studies of bio-aerosols [8]. To our knowledge, there has not been a demonstration of the use of mode-locked ultra-violet pulses for single-photon fluorescence measurements. We will present fluorescence measurement results for both dye-doped polystyrene latex (PSL) particles and biological particles.

III. LASER CONFIGURATION

To achieve the high peak powers in the fundamental wavelength pulses that are necessary for efficient conversion and ultimately, significant power levels in the ultra-violet wavelength, we implement a fiber CPA technique. In order to create a compact, lightweight, and fieldable system, we develop a normal-dispersion Yb-doped fiber laser, which is followed by a single-mode Yb-doped fiber preamplifier, high-dispersion fiber for temporal stretching of the pulses, a multi-stage, large-mode-area, Yb-doped fiber power amplifier, and finally a chirped volume holographic Bragg grating to recompress the pulses. Frequency conversion to a wavelength of 525 nm is achieved with a lithium triborate (LBO) crystal and further conversion to a wavelength of 262 nm is achieved with a beta barium borate (BBO) crystal. A schematic diagram of the overall system is illustrated in Fig. 1.

![Schematic diagram of system.](image)

The chirped pulse amplification segment of the setup is illustrated in Fig. 3, where insets show the temporal pulse shape and spectrum. The stretched pulse, as detected by a Thor Labs D400FC InGaAs detector and Agilent Infiniium 86109A optical detector plug-in module, is also shown in Fig. 3. First the laser output is amplified using an Yb-doped fiber preamplifier (Pre-Amp 1) with ~1.1 m of gain fiber that has a core size of 7 µm (INO Yb708). The gain fiber is end-pumped with 550 mW of 976-nm pump in a counter-propagating configuration. The output power is ~55 mW. Stretching the pulse to ~125-ps duration is achieved with the combination of a 30-m length of Corning high-dispersion fiber (D=100ps/nm/km) and ~28-m length of Corning HI-1060 fiber (D=35ps/nm/km). The loss of the fiber stretcher is almost 6 dB because the high-dispersion fiber is designed for a wavelength of 980 nm. A single-mode, large-mode-area, Yb-doped fiber (Liekki Yb1200-10/125DC) preamplifier (Pre-Amp 2) is used to amplify the stretched pulse to ~155-mW average power. The 0.85-m length gain fiber has a core size of 9.7 µm and is co-propagating, end-pumped with 500 mW of 976-nm pump. This is then followed by a double-clad fiber amplifier with a 1.65-m length, 20-µm core gain fiber (Liekki Yb1200-10/125DC).
Fig. 3: Chirped-pulse amplification setup. The stretched pulse is detected by a fast photodetector and measured with an oscilloscope. The compressed pulse is measured with an intensity autocorrelator and the spectrum is characterized with an optical spectrum analyzer. Yb = ytterbium, HDF = high-dispersion fiber, CVBG = chirped volume Bragg grating, and PC = polarization controller.

Yb1200-20/125DC). This amplifier has four 976-nm wavelength pump laser diodes (Alfalight) that each have a maximum output power of 4 W, which are coupled via a multimode coupler (SIFAM) and counter-propagating to the signal. The output power of the amplifier versus the total pump power is plotted in Fig. 4, and the maximum output power is ~6.5 W for ~15 W of pump power, indicating over 43% pump efficiency. The power amplifier output is collimated with a 25-mm lens, anti-reflection coated for the wavelength range of 650 to 1050 nm. Finally, the pulses are recompressed using a chirped volume holographic Bragg grating (Optigrate) with a reflection bandwidth of 20 nm. The characteristic spectral reflection response function is illustrated in Fig. 5. The reflected power is ~69% of the total incident power, as shown in Fig. 6, and we achieve a maximum output of ~4.5 W. The size of the grating is 4x8x25mm and both end facets are anti-reflection coated.

4x8x25mm and both end facets are anti-reflection coated. The recompressed pulse, shown in Fig. 3, has an autocorrelation width of ~670 fs with some pedestals, which are likely due to higher order dispersion effects. The expected transform-limited pulse width for a normal-dispersion laser with this spectral width is ~454 fs [2].

Fig. 4: Power amplifier performance. The plot shows the measured output power versus the input pump power. The input pump power is calculated based on the drive current and efficiency of the laser diode.

The harmonic generation segments comprise the final stages of the system. The crystals and lenses are all anti-reflection coated for the fundamental and/or the second harmonic frequencies. The polarization controller, located between Pre-Amp 2 and the power amplifier, is used to align the amplifier output polarization to the axis of the crystals. A 100-mm lens is used to focus into the 5x5x20-mm length LBO crystal (Coherent) and a 100-mm lens is used to collimate the output. The spot size inside the crystal is approximated by using a knife-edge measurement technique in free-space at the approximate center location of the crystal. The beam is measured to be ~64-μm FWHM (~109-μm “1/e²” width). Estimating the crystal refractive index as 1.56, a calculated 45-
4 mm confocal parameter implies a fairly uniform intensity within the LBO crystal. A cold mirror is used to separate the 1050-nm and 525-nm wavelengths, and the latter is focused into the 4x4x6-mm length BBO crystal (Optical Planz) with a 60-mm lens for 262-nm wavelength generation. Again, using the knife-edge measurement technique, the spot size in free-space is \( \sim 75 \mu m \) FWHM \((\sim 127 \mu m \text{ "}1/e^2\text{" width})\). And again using a refractive index of 1.56, the calculated 120-mm confocal parameter is much longer than the crystal, thereby insuring a uniform intensity beam inside the crystal. The output of the BBO crystal is followed by a fused silica prism for separating the green and ultra-violet wavelengths.

**IV. RESULTS**

Figure 7 illustrates the second harmonic generation results. Figure 7(a) represents the measured power conversion from 1050-nm to 525-nm wavelength. Figure 7(b) shows the calculated conversion efficiency as a function of the input power. The efficiency improves with increased power until an input power level of \( \sim 3 \text{ W} \) when it reaches the maximum conversion efficiency of \( \sim 30\% \). No saturation in output power is observed up to our maximum average input power of 4.5 W, resulting in an output power of 1.25 W and a pulse energy of \( \sim 30.5 \text{ nJ} \). The spectrum of this second harmonic pulse is shown in Fig. 8, and the FWHM width is \( \sim 0.8 \text{ nm} \). This is narrower than expected and is probably due to a combination of effects including the chirp on the input pulse as well as the LBO acceptance angle and phase matching characteristics.

![Fig. 7: Performance of the second harmonic generation process. (a) Measured power conversion from 1050 nm to 525 nm wavelengths, (b) Calculated conversion efficiency versus input power.](image)

Figure 9 illustrates the measured performance of the conversion from 525 nm to 262 nm (fourth harmonic of the fundamental wavelength). Figures 9(a) and 9(b) represent the power conversion from the 525 nm to the 262 nm wavelength, and the calculated conversion efficiencies, respectively. In this case, the peak conversion efficiency is \( \sim 28\% \). Again, no power saturation effects are observed and a maximum average output power \( \sim 350 \text{ mW} \) is achieved, providing a maximum pulse energy of \( \sim 8.5 \text{ nJ} \). The spectrum of this fourth harmonic is shown in Fig. 10, and the FWHM width is \( \sim 0.6 \text{ nm} \).

![Fig. 8: Spectrum of the 525-nm pulses. The FWHM width is \( \sim 0.8 \text{ nm} \).](image)

![Fig. 9: Performance of the uv generation process. (a) Measured power conversion from 524 nm to 262 nm wavelengths, (b) Calculated conversion efficiency versus input power.](image)
Because of phase matching and acceptance angle requirements, the harmonic generation processes leads to an elliptically shaped output beam. Figures 11(a) and 11(b) shows the beam intensity profiles of the green and ultra-violet output beams, respectively. The minor to major axis ratio is ~0.6 in both cases.

Finally, another parameter of interest in these systems is the electrical-optical conversion efficiency. Based on the DC voltage and current of all the laser diode drivers and thermoelectric coolers, we calculate the electrical to ultra-violet (optical) conversion efficiency to be ~0.4% and ~1.5% for the conversion to the green wavelength.

Note that the multimode coupler used for the power amplifier pumps has 6 possible inputs. Since we are currently using just 4 pumps, there are still 2 remaining, unused, ports. Thus, the system could achieve even higher output powers by simply adding pumps to those ports.

V. FLUORESCENCE EXCITATION

We conduct several proof-of-principle experiments to show that mode-locked ultra-violet pulses can be used effectively for fluorescence excitation and particle detection. The Aerosol Interrogation Module breadboard used for our mode-locked laser studies has been described previously elsewhere [9]. The aerosol particles are pumped through a 700-μm aerosol nozzle into the focal volume of the aerosol chamber where the individual micron-sized particles are interrogated. The scattered and fluorescence light are collected by an elliptical reflector and sampled in several broad bandwidth detection bands. The ultra-violet beam is focused to form a 730-μm by 350-μm beam in the focal volume of the elliptical collection mirror. The particles flow at a median velocity of roughly 4.3 m/s, which translates to a transit time through the beam of 83 μs. The entire signal is integrated by setting the electronic gate width for the integrator to be 100 μs.

The fluorescence and elastic scatter are measured from a number of calibration PSL particles and biological agent simulant particles. The fluorescence signal and elastic scatter signal as a 10-μm NaCl particle doped with tryptophan transits through the laser beam is shown in Fig. 12. The quasi-continuous nature of this excitation is evident in these plots. The particle is continuously excited by different pulses as it traverses the beam. The fluorescence emission from the...
tryptophan in the 300 to 340-nm band is shown in the top trace and the elastic scatter signal is shown in the bottom trace. The DC background level seen in the elastic scatter signal is due to the stray scattered light from the aerosol chamber. The signals are integrated, corrected for the various instrument responses, and expressed as number of photons emitted by the particle.

The fluorescence of a size series of green dye-doped PSL spheres ranging in size from 2 to 5 μm is plotted in Fig. 13.

Roughly 10,000 particles are sampled for each particle size. The fluorescence emission measured in the 400 to 500-nm band is plotted versus the fluorescence emitted in the 500 to 600-nm band. As expected, the fluorescence signal scales with particle size and the distribution of the particles is much tighter for the bigger size particles. In fact, the fluorescence signal shows an approximately cubic dependence on the particle size as one would expect from homogenous particles.

The fluorescence measured from the bio-warfare agent simulants Bacillus Globigii (BG) and ovalbumin is shown in Fig. 14. BG is a simulant for anthrax spores and ovalbumin is a common simulant for biological toxins. The particles are generated using a Sono-tek generator and the mass median size for BG and ovalbumin are 4.7 μm and 5.8 μm, respectively, with a FWHM of less than 2 μm. The fluorescence in the 300 to 340-nm band is plotted versus the 360 to 400-nm fluorescence band. These fluorescent bands are the prominent protein signature bands for biological materials. As one can observe from the fluorescence emission from the bacterial spores (Fig. 14), the BG signature is well separated from that of ovalbumin. The fluorescence signature shows wider distribution due to the wider range of particle sizes in the sample. These preliminary measurements on bioaerosols from mode-locked ultra-violet excitation are encouraging.

VI. CONCLUSION

We demonstrate ultra-short pulsed harmonic generation to 525-nm and 262-nm wavelengths starting with a 41-MHz mode-locked, all-normal-dispersion, Yb-doped fiber laser with a center wavelength of 1050 nm. Nonlinear effects during amplification are reduced through the implementation of chirped-pulse amplification. Frequency doubling results in average powers of greater than 1.2 W (30.5 nJ/pulse) for the 525-nm wavelength and 350 mW (8.5 nJ/pulse) for the 262-nm wavelength with <500-fs pulse durations. We also demonstrate that mode-locked ultra-violet pulses generated by a fiber laser are viable sources for fluorescence excitation of biological particles.

REFERENCES