Application Note 35

Spectral Broadening and Temporal Compression of Ultrashort Pulses in a Gas Cell

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Introduction

Spectral broadening, also known as continuum generation, arising from the interaction of intense ultrashort laser pulses with nonlinear materials is a well known, highly exploited phenomenon.[1] White light generated from focusing ultrashort pulses into transparent media or by the propagation of high intensity pulses through fiber is commonly used as a coherent broadband source for applications such as time-resolved spectroscopy[2], seeding optical parametric amplifiers[3,4], and optical coherence tomography[5], just to name a few. The effect of spectral broadening can also be used for generating pulses significantly shorter than the original input pulse (i.e. pulse compression). Utilizing this technique for pulse compression was demonstrated by Ippen et al.[6] for the compression of picosecond pulses down to the femtosecond time regime using a single mode fiber and a grating pair. Spectral broadening and the subsequent recompression of high intensity pulses in rare gases was demonstrated by Krausz and coworkers[7], and has since become a proven technique for the generation of pulses as short as a few optical cycles (5.1 femtoseconds, FWHM).[8]

In this note, we evaluate the performance of a 125 cm long argon filled gas cell used in conjunction with the Spectra-Physics Tsunami® seeded Spitfire® Pro amplified laser. Spectral broadening increases the bandwidth of the Spitfire by a factor of 3 before the onset of dramatic spectral modulation. Compression of such broadband pulses, although beyond the scope of this note, can in general be accomplished by using commercially available negative group delay dispersion (GDD) mirrors. Here we determine the time profile of the output pulse by calculating the Fourier transform of the broadened spectrum with zero phase. This represents the shortest obtainable pulse for a given bandwidth.

Theory

Spectral broadening can be described as a combination of nonlinear parametric processes including stimulated Raman, four-wave mixing, self-phase modulation as well as potentially higher order processes. The principle mechanism for spectral broadening varies for the different media described above. For supercontinuum generation in fibers, a combination of nonlinear processes leads to complex soliton dynamics. In rare gases, the mechanism of self-phase modulation dominates. Self-phase modulation is a third order process arising from the intensity dependence of the refractive index. It can be easily understood by first expressing the electric field of an ultrashort pulse as a traveling wave,

$$E(z,t) = A(z,t)e^{-i(\omega_o t - kz)} + c.c. \quad ,(1)$$

where A is the envelope function, ω_o is the carrier frequency, t is time, k is the wave-vector, and z is the direction of propagation. After propagating through a length of material, L, the total phase shift of the field is given by,

$$\phi(t) = \omega_o t - \frac{[n_o + n_2 I(t)]\omega_o L}{c} \quad , (2)$$

where n_o is the refractive index of the material, n_2 is the nonlinear refractive index of the material, I(t) is the intensity profile of the pulse and c is the speed of light (The following two relations were used in deriving equation 2: $k = n\,\omega_o / c$ and $n = n_o + n_2 I(t)$). The instantaneous frequency of the pulse, $\omega(t)$, can be determined by taking the derivative of the phase shift with respect to time,

$$\omega(t) = \omega_o - \frac{n_2 \omega_o L}{c} I'(t) \quad . (3)$$

Equation (3) leads to a time dependent frequency shift, $\Delta\omega(t)$, given by,

$$\Delta\omega(t) = \frac{n_2 \omega_o L}{c} I'(t) \quad [9] \quad .(4)$$

Equation 4 is the origin of self-phase modulation which depends not only on material parameters (i.e. the nonlinear refractive index and length of material), but also on the temporal profile of the pulse.

From equation 4, it is clear that given enough intensity, interactions between light and matter will result in spectral broadening (all real materials have an n_2). It is not clear however, that the additional bandwidth generated from this interaction can be utilized to obtain a shorter pulse. Figure 1a illustrates a Gaussian intensity profile as well as $\Delta\omega$

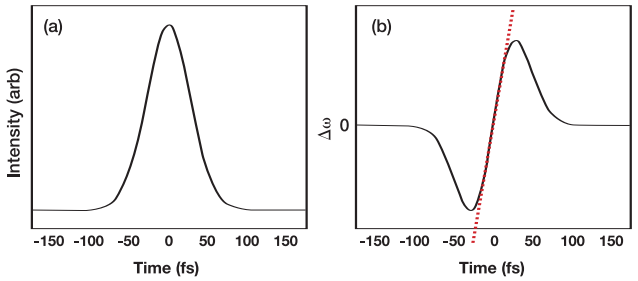

Figure 1: (a) A Gaussian pulse in time. (b) Modification of the carrier frequency as a function of time for self-phase modulation

as a function of time (figure 1b). $\Delta\omega$ is the alteration to the carrier frequency, ω_o, due to self-phase modulation. Negative values of $\Delta\omega$ correspond to a red-shift of the pulse and positive values of $\Delta\omega$ correspond to a blue-shift. Figure 1b shows that for a Gaussian pulse, the leading edge of the

pulse is red-shifted while the trailing edge is blue-shifted. This corresponds to positive dispersion; moreover, for the central portion of the pulse, the dispersion can be attributed to GDD as illustrated by the linearity of the dashed red line. The wings of the pulse do exhibit higher order dispersion; however, the bulk of the pulse is compressible simply by adding negative GDD. The analysis presented above holds for well-behaved (Gaussian-like) pulses at moderately high intensities. At extremely high intensities, self-phase modulation leads to complex spectral profiles which are not necessarily compressible in time.

Experimental Setup

A block diagram of the experimental setup is illustrated in figure 2. The laser source is the Spectra -Physics Spitfire Pro

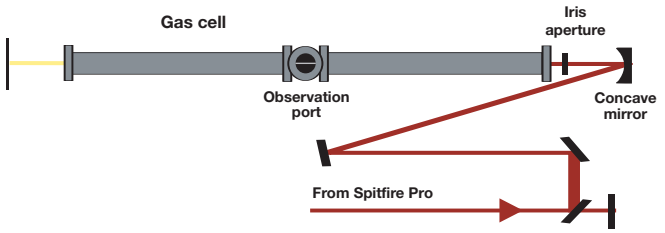

Figure 2: Block diagram for the experimental setup used to produce continuum pulses. The laser source is the Spectra Physics Spitfire laser. Two inch ultrafast mirrors as well as one ultrafast beamsplitter are used to route the beam to a two inch 2000 mm radius of curvature concave mirror. The concave mirror focuses the pulse in the center of the gas cell filled with one atmosphere of argon.

regenerative amplifier seeded with the Spectra-Physics Tsunami oscillator. The Spitfire is capable of more than 3.5W of output power at 1 kHz with pulse duration as short as 35 femtoseconds, FWHM. The choice of a gas in the cell, pressure, radius of curvature of the focusing mirror and the length of the cell depend on a combination of parameters including pulse energy, pulse width, mode quality and stability of the laser. For more information, we refer the reader to numerous publications in this field.[10,11,12] In this note, we provide a set of parameters as well as components that are optimized for our system. For these experiments, the bandwidth of the Tsunami was narrowed to produce 330 cm^{-1} out of the amplifier in order to create a smoother spectral profile. This corresponds to a 45 femtosecond transform limited pulse.

The beam emerging from the Spitfire was routed to a concave mirror (2000 mm radius of curvature) using several high power two inch diameter ultrafast mirrors as well as one ultrafast beamsplitter in order to attenuate the beam by 50%. The angle between the concave mirror and the last routing mirror was kept small in order to minimize astigmatism introduced by operating the concave mirror off axis. To further attenuate the beam, an iris was introduced into the beam path just prior to the gas cell. The average power of the beam after the iris was 1 W corresponding to a pulse energy of 1 mJ.

The gas cell is based on standard UHV parts that can be purchased from several suppliers. The detailed description of how to assemble the cell can be found in the appendix. Briefly, the cell consists of a stainless steel tube, 125 cm in length, with an observation port located at the center of the tube. Two millimeter thick fused silica windows were used at the entrance and exit ports of the cell, and the overall clear aperture of the system is 35 mm. For these measurements, the pressure of argon in the cell was about two atmospheres.

The 2000 mm radius of curvature mirror focuses the beam approximately in the center of the gas cell. Figure 3 shows a picture of the filament seen through the observation port of

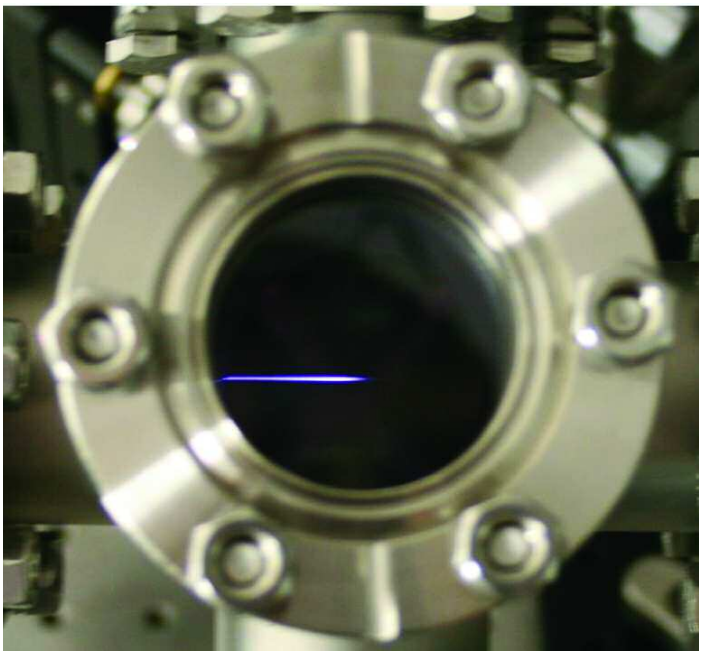

Figure 3: The filament produced by focusing the high intensity pulse into Ar gas cell at 2 atmospheres

the cell. The bluish streak is the plasma channel formed due to the ionization of the argon gas by the femtosecond laser pulse. The plasma channel extends well beyond the Rayleigh range of the beam which under these focusing conditions is approximately 5 mm. This effect, also termed filamentation, occurs when self-focusing is balanced with the defocusing effect of diffraction and plasma produced by multiphoton ionization of the argon gas. The result of this interplay is a self-guided high intensity field that can extend distances much greater than the Rayleigh range. This self-guiding action eliminates the need for a waveguide to maintain high intensities over long distances necessary for effectively driving self-phase modulation in gases.

Figure 4 is a picture of the output beam from the gas cell projected against a wall. To the eye, the center of the beam appears as continuum (white light) surrounded by a bluish hue. The beam profile of the continuum was similar to that of the input beam, both being quite round. The pointing

Figure 4: The beam emerging from the argon gas cell. It appears as white light to the naked eye

stability of the continuum was similar to the pointing stability of the pump laser with the argon cell removed from the beam path. In fact, with the exception of the spectral broadening, it appears that the continuum takes on the excellent beam characteristics of the Spitfire laser.

The spectrum of the continuum (figure 5b), as well as the spectrum of the bandwidth limited Spitfire (figure 5a), were recorded on the OSM-100-VIS/NIR spectrometer. The spectrum of the laser pulse produced by the Spitfire appears to be approximately Gaussian with 330 cm^{-1} of bandwidth, FWHM. After emerging from the gas cell, the bandwidth of the pulse increases by almost a factor of three to 850 cm^{-1}; however, the spectral profile is far from Gaussian. There appears to be a "hole" in the central region of the spectrum. This is a characteristic of continuum generation due to self-phase modulation and can lead to 100% modulation of the spectrum at high enough intensities.

Figure 5c and 5d show the calculated transform limited temporal profiles corresponding to the spectral output of the Spitfire and continuum respectively. At transform limit, the pulse duration of the Spitfire is 45 fs while for the continuum, the pulse duration is 14 fs. This represents a compression ratio of more than a factor of three. While increasing the intensity of the pulse in the gas chamber results in more bandwidth, the pulse does not necessarily become shorter in time. As mentioned in the previous paragraph, increasing the intensity beyond a certain point results in a deeply structured spectrum where the additional bandwidth generated through the nonlinear interaction cannot be utilized to further compress the pulse. In order to further compress the pulse, additional stages (argon cells) can be added in series with pulse compression between each stage. This strategy has been shown to produce pulses as short as 5.1 fs in the 200 μJ range.[8]

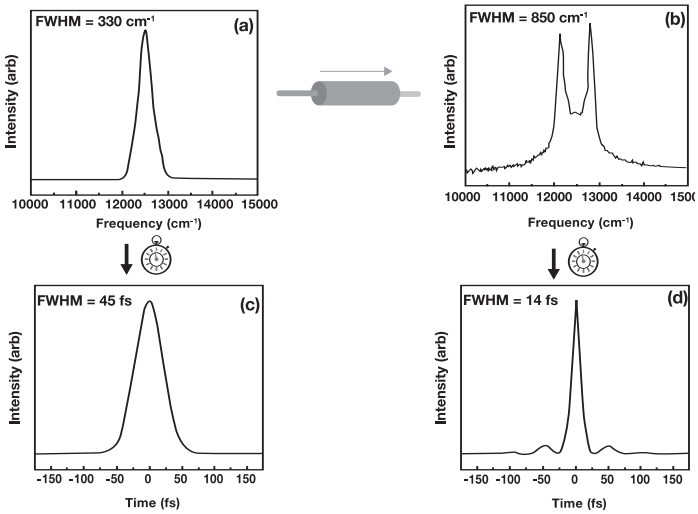

Figure 5: The pulse, in frequency and time, before and after traveling through the argon filled gas cell. Time profiles are Fourier transforms of the corresponding spectra.

Finally, it is worth commenting on pulse compression. The pulse emerging from the gas cell has approximately -100 fs^2 to - 200 fs^2 of group delay dispersion. The sources of this dispersion are a combination of the windows, the argon gas, as well as the self-phase modulation process itself. The high intensity of the pulses forbids the use of refractive compressors such as a prism compressor, and the dispersion is too small to take advantage of a grating compressor. More complicated compression arrangements can be designed such as a grating stretcher/compressor combination; however, it is far more straightforward to simply use negative GDD mirrors. For this application, it is critical that the negative GDD mirror introduce a constant amount of GDD (in the vicinity of -20 fs^2/bounce to -40 fs^2/bounce) over a broad spectral range (600 nm – 1000 nm). Typically, these types of mirrors are produced in pairs, where the GDD of the individual mirror is highly oscillatory, but when functioning as a pair, the GDD is quite flat.

Appendix: Construction of the Gas Cell

The assembled gas cell is shown in Figure 6. The cell is entirely based on standard UHV components. The central portion of the gas cell shown in Figure 7 is a 5-way cross with CF flanges that allows for attachment of input and output valves, a glass observation port, and user defined lengths of tubing along the optical axis to determine the overall length of the cell. Depending on the parameters of the laser to be used, the length of the cell can be varied by selecting standard or custom lengths of tubing. For this note, the total length of the cell was 125 cm requiring custom 22.15" lengths of tubing. The fused silica input and output end ports are also mounted on flanges for easy assembly. The two lengths of tubing and the two valve assemblies are mounted to the 5-way cross with copper gaskets between the flanges. By gradually and evenly tightening the bolts, the knife edges on the counterbores of the flanges dig into the copper gasket and create an air tight seal (consult manufacturer's instructions for torque specifications). As this equipment is designed for UHV applications, the observation window and two end ports need to be flipped and mounted to the cell using Viton® gaskets, rather than the copper gaskets (see Figure 8). This arrangement allows the glass or fused silica windows to seat against a seal under positive pressure. Take care not to over tighten the end ports and observation window when using the Viton® gaskets.

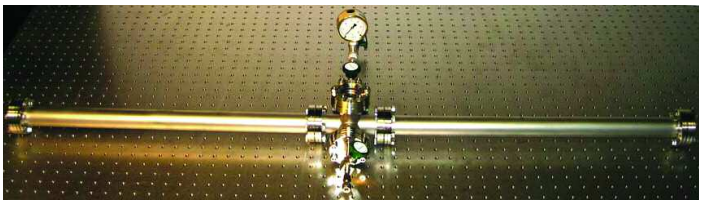

Figure 6: Assembled gas cell

Fill the cell with inert gas to test for leaks and tighten screws if necessary. UHV component manufacturers have no maximum positive pressure specifications, so use caution when increasing the pressure and never exceed two atmospheres. The addition of an accurate pressure gauge to monitor the cell pressure is recommended. Always use personal protective equipment when operating the cell under pressure. The cell is equipped with inlet and outlet valves to control gas flow. To purge the cell, close the outlet valve,

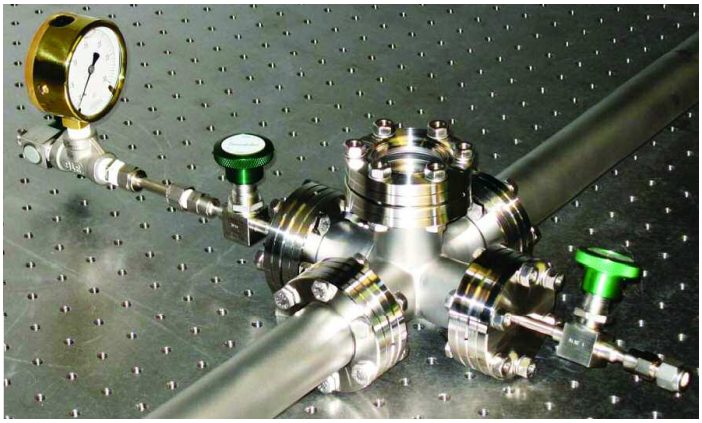

Figure 7. The 5 way cross with CF flanges, inlet and outlet valves, pressure gage

open the inlet valve to pressurize the cell, close the inlet valve and open the outlet valve to relieve most of the pressure. Repeat this until the gas contained within the cell meets your purity requirement. Fill the cell to the desired pressure and close the inlet valve. With a proper seal, constant pressure inside the cell can be preserved for several days. Fine adjustments can be made through the use of the inlet and outlet valves.

Figure 8. The end port with Viton gasket

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