Pulse propagation in hollow core fiber at high pressure regime: application to compression of tens μJ pulses and determination of nonlinear refractive index of Xe at $1.03\mu\text{m}$

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Classical expression for propagation constant and absorption coefficient derived by Narcatilli and Schmeltzer for hollow core fiber (HCF) has been slightly modified to account for gas material dispersion at high gas pressure. As a proof of concept, nonlinear refractive index of xenon gas has been investigated by numerically fitting to experimentally obtained spectral broadening in HCF under intense high repetition rate pulses. By varying the xenon pressure inside the HCF, a pressure dependent nonlinear refractive index value of $\kappa_2 = (50.1 \pm 0.3) \times 10^{-20} \text{ cm}^2/\text{W atm}$ at $1.03 \mu\text{m}$ is obtained, which compared favorably with literature reported values. Finally, temporal compression of $50 \mu\text{J}$, 320 fs pulses at 0.6 MHz repetition rate to 61 fs with 0.29 GW peak power in a HCF filled with 5 bars of Xe gas has been demonstrated using a single HCF compression stage.

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1. Introduction

Recent development of Yb-doped rigid fiber amplifier has made available a promising alternative to the Ti:Sapphire amplifier [1,2] as light source for femtosecond scale photochemistry and electron dynamics experiments. While the gain bandwidth of Yb-doped fiber is not as broad as Ti:Sapphire, Yb-doped glass offers the potential of high average power with tunable repetition rate up to megahertz while still delivered sufficient peak intensity for light-matter interaction of interest. Sub-picosecond pulses at repetition rate of hundreds of kilohertz and average power of hundreds of Watts have already been demonstrated [3-8]. Combining with spectral broadening via noble gas-filled HCF, the shortcoming of narrower bandwidth can be overcome. In a recent paper, Tünnemann's group [9] demonstrated a setup with above 100 W average power and near single cycle pulse via two stages xenon-filled HCF compression. Application of Yb-doped amplifier to EUV generation has also been recently demonstrated [9-13]. While spectral broadening and temporal compression of Yb-doped laser are easily achievable at hundreds of microjoules to millijoules level [4-8,13], spectral broadening in tens of microjoules and megahertz level is relatively harder to realize. Given that most commercially available Yb-doped laser are operated in this regime, an understanding in the temporal compression in this regime is highly beneficial for further application such as optical pump-probe spectroscopy, higher-order harmonic generation and high-power terahertz generation. Experimental demonstration of spectral broadening and temporal compression has been demonstrated in $\sim 0.1 \text{ GW}$ regime 100 KHz Ti:Sapphire amplifier system [14], however the role of noble gas material

dispersion at the higher pressure needed for spectral broadening in this regime has never been discussed. In this paper, we investigate self-phase modulation and pulse compression in a xenon-filled HCF at such regime by means of experiments and numerical simulations. Through our investigation, we have found that the classical expressions in [15] for propagation constant and absorption coefficient need to be modified to accommodate the fact that noble gas material dispersion is comparable to the waveguide induced dispersion for pressure commonly used for pulse compression in this regime. While a similar noniterative approach based on the broadened bandwidth have been reported [16] and has the advantages of simple and direct feedback, an iterative approach such as the one presented here is not constrained by input pulse temporal profile and phase and can be easily adapted to account for other dispersive components in beam path and higher order dispersive and nonlinear effects.

Recently Kagome photonic crystal fibers (PCF) [17] and bulk unguided compression schemes [18,19] have been demonstrated with much lower attenuation for pulse compression in the high average power, moderate pulse energy ($1 - 10 \mu\text{J}$) regime. However, in the regime of tens of μJ reported here, a conventional HCF with bore radius of $\sim 100 \mu\text{m}$ still delivers reasonable spectral broadening with less complicated alignment, vibrational sensitivity while offering long term repeatability and survivability, which could in turn allows for less complication, better predictability and ease of operation in further spectral generation, manipulation and detection stages downstream.

2. Experiment

Fig. 1. (a) Schematic of the measurement and compression scheme. Pulses from the Yb-doped fiber laser is coupled to a 1 m-long hollow-core fiber with a bore radius of 100 μm and filled with 5 bars of xenon for spectral broadening. The output is then temporally compressed by chirped mirrors. **(b)** FROG extracted temporal intensity and phase of the direct laser output pulses with FWHM width of ~ 320 fs. Blue arrows indicated satellite pulses from residue higher order dispersive effect inside the laser. **(c)** Corresponding extracted spectrum with a FWTM width of 11 nm before compression.

In Fig. 1, we show the experiment setup of our nonlinear refractive index extraction and pulse compression scheme. The laser source is a commercial Yb-doped fiber amplifier from Amplitude Systèmes with a pulse energy of 50 μJ at a repetition rate of 600 kHz. With a full-width tenth maximum (FWTM) bandwidth of ~ 11 nm, the output pulse width of this laser is 320 fs (transform limited pulse duration is 293 fs) after optimizing the laser built-in grating compressor. At the immediate output of the laser, the $1/e^2$ beam radius is measured to be 1.53 mm with an M^2 value of 1.1 and a divergence angle of 0.5 mrad. After a propagation path length of 2.76 m, the $1/e^2$ beam radius expanded to 2.88 mm. The laser beam is then focused with a $f = 500$ mm fused silica plano-convex lens into a 1 m-long xenon-filled fused silica HCF with bore radius of 100 μm . The measured beam waist of 63 μm is optimal for coupling into EH_{11} fundamental mode of the HCF [20]. To improve the thermal stability of the HCF output, the V-groove that holds the HCF and the gas cell housing the V-groove and HCF are water cooled. After collimation with a $f = 500$ mm plano-convex lens, the output power is measured to be > 18 W. The overall transmission efficiency is hence calculated to be $> 60\%$, which is quite close to the theoretical transmission efficiency of 67.2% of a 1 m-long, 100 μm bore radius fiber [15,21]. Within the HCF, the peak power of the laser pulse is estimated to be ~ 0.10 GW, which is relatively small compared to the threshold intensity of 2.5 GW for self-focusing, hence the main nonlinear contribution in the fiber is spectral broadening via self-phase modulation. Furthermore, the Intensity at the entrance of the HCF is calculated as $1.6 \times 10^{12} \text{ W/cm}^2$ which is one or two orders of magnitude lower than typical laser induced ionization threshold intensity of $10^{13} - 10^{14} \text{ W/cm}^2$ for noble gas indicated that scattering and absorption effect due to plasma generation are negligible.

Fig. 2. Comparison between the measured SPM-broadened spectra (solid lines) at selected Xe pressures with the corresponding fitted spectra (dashed lines), obtained from least-squares fitting of the experimental spectra using Eq. 1 and 2. Comparisons at other fitted pressures can be found in Fig. 7 in appendix B.

The dependence of the spectral broadening on the xenon pressure is measured (Fig. 2) with an Ocean Optics USB2000+ spectrometer equipped with a Silicon CCD detector. While Silicon detector has a cutoff wavelength at 1080 nm, we do not observe artificial truncation of broadened spectra up to the pressure of 6 bars. As quick rolloff of the detector sensitivity around this wavelength could affect the accuracy of our measurement, the spectrometer with its coupling optical fiber had been carefully calibrated with a stabilized tungsten lamp and neon arc lamp. In general, the output spectral width increased from a FWTM value of 40 nm at 3.4 bars up to FWTM value of 56 nm at 6 bars with no significant degradation in output power. A few sharp spikes in the central portion of the output spectra that became more complicated at higher Xe pressure are observed and can be traced back to small satellites (indicated by blue arrows in Fig. 1) in the laser output. These satellites are due to residual high-order dispersion terms in the laser output that cannot be compensated by the laser built-in grating compressor.

Using the extracted temporal pulse profile and phase of the direct output from the laser shown in Fig. 1 and assuming that the main contribution of the discrepancy between theoretical transmission coefficient and measured transmission coefficient is due to coupling losses while higher nonlinear effects such as self-steepening and Raman resonances are negligible, we are able to match the experimentally measured spectra at different xenon pressures with theoretical pulse propagation calculated using the split-step Fourier method [22,23]. Dispersion of fused silica with thickness of 3.4 mm ($GVD = 18.95 \text{ fs}^2/\text{mm}$ and $TOD = 41.06 \text{ fs}^3/\text{mm}$) are added before and after the split-step Fourier method propagation calculation to account for the material dispersion of lenses and windows in front and at back of the HCF. At the measured Xe pressures, the material

dispersion of Xe gas ($GVD = 0.339 \text{ fs}^2/\text{mm}$ and $TOD = 0.208 \text{ fs}^3/\text{mm}$ at 5 bars) is comparable or higher than the waveguide induced dispersion ($GVD = -0.028 \text{ fs}^2/\text{mm}$ and $TOD = 0.046 \text{ fs}^3/\text{mm}$ for a bore radius of $100 \text{ }\mu\text{m}$), hence the equations of propagation constant β and attenuation α for the fundamental EH₁₁ mode in Ref [15] are modified to incorporate material dispersions and are shown in Eq. 1 and 2 (Derivation in appendix A. A comparison between uncompensated FROG extracted result and its corresponding fitted spectrum and spectral phase is shown in Fig. 8 in appendix B).

$$\beta = \frac{2\pi n_{Xe}}{\lambda} \left[1 - \frac{1}{2} \left(\frac{2.405\lambda}{2\pi n_{Xe} a} \right)^2 \left(1 + \text{Im}(v_1) \frac{\lambda}{\pi n_{Xe} a} \right) \right] \quad (1)$$

$$\alpha = \left(\frac{2.405}{2\pi n_{Xe}} \right)^2 \frac{\lambda^2}{a^3} \text{Re}(v_1) \quad (2)$$

Here λ is the wavelength of the input beam, a is the bore radius of the HCF, $v_1 = \frac{1}{2}(\nu^2 + 1)/\sqrt{\nu^2 - 1}$, $\nu = n_{\text{SiO}_2}/n_{\text{Xe}}$, n_{SiO_2} and n_{Xe} are refractive index of fused silica [24] and Xe gas [25] respectively. Global least-squares fitted error between measured and calculated spectra, $\text{err} = \sum_{\lambda,i} (S_{\text{meas},i}(\lambda) - a_i \cdot S_{\text{theo},i}(\lambda, n_{2,i}))^2$ is used for extracting the best-fit pressure dependent refractive index $\kappa_2 = (50.1 \pm 0.3) \times 10^{-20} \text{ cm}^2/\text{W atm}$. $S_{\text{meas},i}(\lambda)$ and $S_{\text{theo},i}(\lambda, n_{2,i})$ are the experimentally recorded spectra and the theoretical spectra reconstructed for a given Xe pressure, respectively, $n_{2,i} = \kappa_2 p_i$ is the nonlinear refractive index at a given pressure p_i , and a_i is a scaling factor to match theoretically generated spectra and the corresponding experimentally obtained spectra. The extracted value compares favorably with previously reported κ_2 values obtained from hyperpolarizability measurements that employ third-harmonic generation [26,27], electric field-induced second-harmonic generation [28,29] and Kerr effects [30,31] (Table 1). We note that potential sources of error in our value are thermal expansion and distortion of HCF via heating from leakage laser energy and the potential overestimation of coupling losses and associated underestimation of scattering losses. Indeed, when using the laser built in external pulse picker to select 1 pulse out of every 60 pulses, which ensure the same pulse energy, temporal and spatial profile is generated but with 1/60 of the average power, we noticed a slight increase ($\sim 10\%$) in the spectral broadening effect, a fact that suggests that the thermal expansion of the HCF affects or plasma built up effect between pulses coupling behavior of the HCF. As the coupling efficiency does not changed between Xe filled and vacuum purged tubing, plasma built up effect is ruled out as a potential contribution to this effect.

Fig 3. A comparison between best fitted results taken at the Xe pressure of 6 bars using Eq. 1&2 with consideration of gas material dispersion (thick black line), their corresponding equations in Ref [15] that do not take account of gas material dispersion (thin red line) and the measurement (blue dashed line).

Temporal compression of the HCF output (FWTM 48.5 nm) at a xenon pressure of 5 bars is achieved via two reflections off of chirped mirrors (Ultrafast Innovation HD1455), which offer a GDD compensation of -1000 fs^2 and reflectivity of **99.80 %** per reflection over the spectral range of 1000-1060 nm. After compression, the output pulse width is measured by a home built second harmonic generation frequency-resolved optical gating (SHG-FROG) [32] to be 61 fs FWHM, close to the transform-limited pulse duration of 51 fs (Fig. 4). Beam quality factor of $M^2 \approx 1.05$ and ellipticity of ~ 0.95 are measured at the output of the HCF owing to the guiding nature of the HCF acting as a spatial filter for the laser beam.

Fig 4. (a) FROG measurement and reconstruction of the compressed output at Xe pressure of 5 bars. The reconstruction G-value is 5.795×10^{-3} . (b) Extracted temporal and (c) Spectral profiles and phases. Small satellites -500 fs before and 500 fs after main peak in temporal profile (indicated by blue arrows) are due to uncompensated high order dispersion from laser itself (Fig. 1b). Inset in (b) shows the output spatial profile from the HCF. (d) Knife-edge measurement of the HCF output after collimation with a $f=500\text{mm}$ fused silica lens.

Table 1. κ_2 value calculated from our work, hyperpolarizability value measured via third harmonic generation [26,27], electric field induced

second harmonic generation [28,29] and kerr effects [30,31] after wavelength scaling [33] to $\lambda = 1030$ nm. ^aMean and standard deviation of our global fitted result is calculated from extracted results from two interleaved sets of pressure dependent spectra from our measurement. ^bHyperpolarizability value in Shelton is from a fitting function to a series of ESHG measurements at different wavelengths, the maximum error among these measurements is 0.4 %.

Source	Method	κ_2 ($\times 10^{-20}$ cm ² /W atm)
This work	Kerr eff.	50.1 ± 0.3 ^a
Lehmeier et. al. [26]	THG	68.1 ± 6.8
Ward & New [27]	THG	51.0 ± 1.2
Shelton [28]	ESHG	59.2 ^b
Finn & Ward [29]	ESHG	77.2 ± 15.0
Nibbering et. al. [30]	Kerr eff.	77.1 ± 7.6
Wahlstrand et. al. [31]	Kerr eff.	55.08 ± 10.45

Further spectral broadening in a second Xe-filled HCF was attempted (Fig. 5). In this case, the FWTM of the output spectrum at Xe pressure of 3 bars is broadened to 125 nm (transform limited pulse width <15 fs) and the output power is >8.5 W. (Note that the detectable signal range is extended by using a calibrated Horiba iHR320 spectrometer equipped with an InGaAs photodiode). FROG measurements suggest that the resultant spectral phase is highly nonuniform, mainly due to uncompensated higher order dispersion from previous stage and laser. Given that the characteristic length for optical shockwave generation given by $z_s \approx 0.39(L_{NL}/s) \approx 2.7$ m in [22] is close to the length of the HCF used in this experiment, it is also possible that it might take a role in causing the complicated spectral phase. Nonetheless, the large deviation in spectral phase from simple 2nd or 3th orders dispersion behavior at the central of the spectrum, which also covered a significant portion of spectral power show that good pedestal free temporal compression is not possible with chirp mirrors. Temporal compression to achieve higher peak power would require techniques such as multiphoton intrapulse interference phase scan (MIIPS) [34], which allow compensation of higher-order spectral phase distortions.

Fig. 5 (a) Spectral broadening of the compressed HCF output with a second HCF stage at different Xe pressures. (b) FROG measurement and reconstruction of second stage direct output at the pressure of 3.0 bars. (c) Extracted uncompensated temporal profile and phase. Blue arrows indicated satellites from laser output (d) Extracted uncompensated spectral amplitude and spectral phase.

3. Conclusion

The classical expression from [15] has been extended to include the effect of material dispersion of Xe gas at high pressure. As a proof of concept, we obtained pressure-dependent nonlinear refractive index κ_2 of xenon gas at 1.03 μ m via a combination of experimental spectral broadening in a xenon-filled HCF at various gas pressures and numerical pulse propagation simulations extracted with the modified equations and literature values. Our result shows that the modified equations provided better prediction of spectral broadening at high pressure at the few atmospheric pressure regime, which is necessary for compression of optical pulses of μ J regime using conventional HCF configuration.

We and others [16] demonstrated that a conventional HCF based compression scheme is suited for compression of pulses with peak power on the order of 0.1 GW. At a pressure of 5 bars, the spectrally broadened pulse is compressed to a FWHM duration of 61 fs. Such ultrashort pulses are well-suited for femtosecond pump-probe spectroscopy, given their ability to resolve sub-100-fs dynamics, the output stability of fiber lasers, their ease of operation and maintenance free operation. Their high repetition rates also make these ultrashort pulses well-suited to photoemission spectroscopy and microscopy where high peak power might need to be avoided for elimination of coulombic repulsion induced resolution degradation.

The significant of this work is hereby summarized again as followed: 1. We extended the classical expression for propagation constant and

absorption coefficient in HCF [15] to account for material dispersion. 2. Using the modified equations and split-step Fourier method, we extracted nonlinear refractive index for Xe gas which compared favorably with literature value. The accuracy of our method is demonstrated by comparison with measured pressure dependent spectra and FROG extraction of spectrum and spectral phase. 3. We successfully applied our finding on spectral broaden of tens of μJ pulses. Pulse compression at this regime for high repetition rate Yb-doped fiber laser are still infrequently reported but yet important as this is the regime where commercialized high repetition rate Yb-doped fiber amplifier commonly operated.

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Appendix A. Derivation of Eq. 1 & Eq. 2

We begin our derivation by solving for the Maxwell equations for a hollow core waveguide as shown in Fig. 6.

Fig. 6 Geometry of the hollow core waveguide. (left) Side view of the waveguide. (Right) Front view of the waveguide.

To derive the complex propagation constant $\gamma = \alpha + i\beta$, it is important to realize that our case is similar to [15] with the exception of internal relative permittivity $\epsilon/\epsilon_0 = 1$ is replaced by $\epsilon/\epsilon_0 = n_{Xe}^2 \approx 1$. Hence the solutions of Maxwell equations are similar to Eq. 2-4 in [15] with the form:

$$\begin{aligned} E_{znm}^i, H_{znm}^i &\propto J_n(k_i r) \exp(i(\gamma z - \omega t)) \\ E_{znm}^e, H_{znm}^e &\propto J_n(u_{nm}) \exp(i(k_e(r-a) + \gamma z - \omega t)) \end{aligned} \quad (3)$$

Where $J_n(x)$ is the Bessel function of the first kind with order n , u_{nm} is the m th root of $J_{n-1}(x)$, E_{znm}^i, H_{znm}^i are z -component of the internal electric and magnetic mode within the Xe filled core, E_{znm}^e, H_{znm}^e are z -component of the external electric and magnetic mode at the fused silica cladding, γ, ω are complex propagation constant and angular frequency of the propagating mode, r, z are transverse and longitudinal position and k_i, k_e satisfied the following relationships:

$$\begin{aligned} k_i^2 &= k^2 - \gamma^2 \approx k'^2 \\ k_e^2 &= v^2 k^2 - \gamma^2 \approx (v^2 - 1)k'^2 \end{aligned} \quad (4)$$

Where $v = n_{SiO_2}/n_{Xe}$ and $k' = n_{Xe}k = 2\pi n_{Xe}/\lambda$.

Eq. 4 and the assumption $k'a = 2\pi n_{Xe}a/\lambda \gg |v|u_{nm}$ essentially indicated that we are only interested in the situation when good propagation is ensured by glazing incident reflections at the

core/cladding interface. At $r = a$, boundary condition gave an approximated expression k_i similar to Eq. 16 of [15],

$$J_{n-1}(k_i a) = i v_n \left(\frac{k_i}{k'} \right) J_n(k_i a) \quad (5)$$

Where,

$$v_n = \begin{cases} 1/\sqrt{v^2 - 1} & \text{for TE}_{0m} \\ v^2/\sqrt{v^2 - 1} & \text{for TM}_{0m} \\ \frac{1}{2}(v^2 + 1)/\sqrt{v^2 - 1} & \text{for EH}_{nm} \end{cases} \quad (6)$$

From Eq. 4, it is easy to realized that $k_i/k' \ll 1$, hence Eq. 5 can be further simplify by Taylor expansion of $J_{n-1}(k_i a)/J_n(k_i a)$ from $k_i a \rightarrow u_{nm}$,

$$\frac{J_{n-1}(k_i a)}{J_n(k_i a)} = \frac{\partial}{\partial(k_i a)} \left[\frac{J_{n-1}(k_i a)}{J_n(k_i a)} \right]_{k_i a \rightarrow u_{nm}} \times (k_i a - u_{nm}) + \dots \quad (7)$$

Using Bessel function identity $J_n(x) = \pm J'_{n\pm 1}(x) + ((n \pm 1)/x)J_{n\pm 1}(x)$, Eq. 7 becomes

$$\begin{aligned} k_i a &\approx \frac{u_{nm}}{1 + i \frac{v_n}{k' a}} \\ &\approx u_{nm} \left(1 - i \frac{v_n}{k' a} \right) \end{aligned} \quad (8)$$

Hence the complex propagation constant can then be derived from Eq. 4 and Eq. 8,

$$\begin{aligned} \gamma &= k' \sqrt{1 - \left(\frac{k_i}{k'} \right)^2} \\ &= k' \left(1 - \frac{1}{2} \left(\frac{k_i}{k'} \right)^2 + \dots \right) \\ &\approx k' \left\{ 1 - \left(\frac{u_{nm}}{k' a} \right)^2 \left[1 - 2i v_n / k' a \right] \right\} \end{aligned} \quad (9)$$

Separation of real and imagine parts,

$$\begin{aligned} \beta &= \frac{2\pi n_{Xe}}{\lambda} \left\{ 1 - \frac{1}{2} \left(\frac{u_{nm}}{2\pi n_{Xe} a} \right)^2 \left[1 + \frac{\lambda}{\pi a n_{Xe}} \text{Im}(v_n) \right] \right\} \\ \alpha &= \left(\frac{u_{nm}}{2\pi} \right)^2 \frac{\lambda^2}{a^3} \text{Re}(v_n) \end{aligned} \quad (10)$$

Here we note that the expression in Eq. 10 reduced to the classical propagation and attenuation constant in Eq. 21 of [15] when λ/a is small but β is still dominated by modal dispersion and further reduced to $\beta \approx 2\pi n_{Xe}/\lambda$ when $\lambda/a \ll 1$ or when material dispersion is much higher than the modal dispersion. In the specific case of the fundamental mode EH_{11} , $u_{11} = 2.405$, which together with Eq. 6 and Eq. 10, we derived Eq. 1 and Eq. 2.

Appendix B. Additional supporting figures

Fig. 7 Additional comparisons between global fitted spectra and their corresponding measurement.

Fig. 8 Comparison between the best fitted (dashed lines) spectrum (black lines) and spectral phase (red lines) with FROG extraction of uncompensated output (solid lines) of HCF at the Xe pressure of 3.4 bars.

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