Self-Trapping of Supercontinuum Generated by Femtosecond Pulses in a Noble Gas

Trenton R. Ensley, Dmitry A. Fishman, Lazaro A. Padilha, Claudiu M. Cirloganu, Scott Webster, David J. Hagan, and Eric W. Van Stryland

CREOL & FPCE, The College of Optics and Photonics, Univ. of Central Florida, USA
Email: tensley@creol.ucf.edu

INTRODUCTION. The observation of ultra-broadband supercontinuum generation in gases was first reported by Corkum et al. [1]. The possibility to produce high energy white-light continua (WLC) using ultra-short pulses has sparked interest in applications such as remote sensing in the atmosphere [2] and nonlinear spectroscopy [3]. In contrast to white-light generation in solid-state fibers, the nature of WLC generation in gases is not fully understood. The opportunity of having a broadband source of radiation confined in one beam with high spectral energy density has paved the way for replacing conventional sources of radiation such as optical parametric generators/amplifiers (OPGs/OPAs) [3]. The use of parametric devices requires phase matching of nonlinear crystals to generate and amplify certain frequencies which lead to the repeated realignment of optical experiments. In addition, the importance of having good spatial quality beams for nonlinear optical experiments often requires spatial filtering. The spatial properties of WLC from gases have been shown to be excellent over most of their bandwidth. Thus, having a single white-light filament as a tunable source requires only the use of narrow band-pass filters in order to perform nonlinear spectroscopic characterization of materials [3]. Based on this, there is an ongoing effort to understand the mechanisms and limitations involved in producing a high energy ultra-broadband source of radiation.

In this work, we have investigated the two main nonlinear mechanisms, namely self phase modulation and the production of plasma, that are thought to be involved in the generation of a stable WLC in gases in order to achieve high spectral energy output. The calculations presented in the current work confirm the extent of the nonlinear index change due to the plasma production as well as the conditions necessary to attain a self guided filament through self focusing of femtosecond optical pulses in a pressurized noble gas.

RESULTS AND DISCUSSION. To determine the magnitude of the nonlinear refractive index change due to the production of plasma and self-focusing, the free-electron density arising from ionization and the nonlinear susceptibility of the gas was incorporated into our model. Assuming a Drude-like contribution to the refractive index , the change due to the plasma $\Delta npl = -N_e e^2/(2m\varepsilon_0\omega^2)$ where e is the charge of the electron, m is the electron mass, ε_0 is the permittivity of free space, and ω is the pump frequency. The number density of free electrons, N_e, is found by $dN_e/dt = \Gamma(N_0-N_e)$ where Γ is the ionization rate, plotted in Fig. 1a, and N_0 is the number density of initial electrons, related to the pressure of the gas medium. The ionization rate of krypton as a function of incident pulse energy was calculated using the model by Liu et al. [4] and shown in Fig. 1a. The model shows that the ionization rate changes by nearly ten orders of magnitude with respect to one order of magnitude change of the incident pulse energy for our experimental conditions. This result is in good agreement compared to the ionization rate of air calculated in Ref. [5] using a different model, which has an atomic field strength similar to that of krypton.

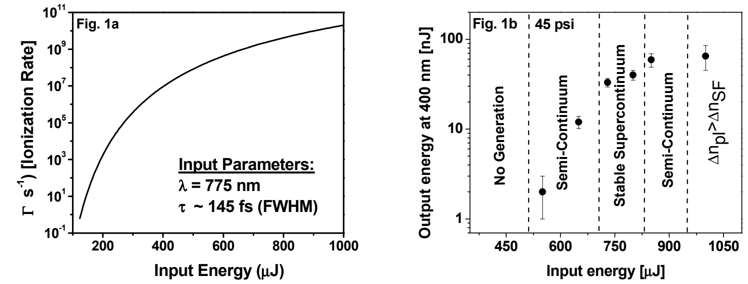

Fig. 1 a) Ionization rate and **b)** output energy at 400 nm versus input energy in krypton.

The model used to determine the energy E_C required in order to initiate self-trapping is given in Ref. [6] utilizing both contributions to the refractive index: self focusing, Δn_{SF}, which is equal to $n_2 I$, with n_2 being the nonlinear refractive index, I the irradiance, and plasma, Δn_{PL}. The change in refractive index may be represented by a positive contribution alone due to self focusing which is given by Eq. {1}

$$E_{C_{SF}} \propto \frac{\tau\lambda^2}{n_2 n_0} \quad \{1\} \qquad \text{and} \qquad E_{C_{SF+Plasma}} \propto \frac{\tau(\lambda^2 + \Delta n_{PL} w_0^{\,2} n_0)}{n_2 n_0} \quad \{2\}$$

where τ is the pulse-width, λ is the wavelength of radiation, n_0 represents the linear refractive index of the gas medium, and w_0 is the minimum beam waist, or by both a positive and negative contribution (Eq. {2}) from self focusing and plasma production, respectively. By incorporating both contributions, one may model the conditions necessary to sustain filamentation.

We experimentally characterized the WLC for different gas pressures and incident pulse energies, employing a Clark-MXR Ti:Sapphire laser system (775nm, 145 fs, 1 mJ @ 1 kHz) as the incident pump source. In Fig. 1b, the spectral energy output at 400nm (± 5nm) of the supercontinuum is plotted as a function of incident pulse energy. This particular wavelength is the cutoff of our anti-Stokes shift which is why we chose to conduct our studies here. For a particular pressure (45 psi) there are four distinctive regions of energy input: energy less than that required to induce self-trapping, flickering white light which we refer to as semi-continuum, stable supercontinuum, and multiple filamentation due to catastrophic self-focusing and/or large plasma formation. The optimum incident pulse energy to sustain a stable WLC was determined experimentally for different gas pressures and is plotted in Fig. 2a (open circles). The experimental data in Fig. 1b can be fit with the self-trapping model outlined by Eq. {2} using the negative contribution Δn_{pl} as a fitting parameter. We find that the phase accumulation from both self-focusing and plasma production is nearly the same in value. Calculations of the spectral broadening due to the phase accumulation from self-phase modulation due to self focusing and plasma production were made based upon a Fourier transformation of our incident pulse and are shown in Fig. 2b. Assuming a temporally Gaussian pulse, with a central frequency ω_0, the Fourier transform $S(\omega)$ takes the form:

$$S(\omega) \sim \left| \int_{-\infty}^{\infty} g(\omega,t) e^{i\phi_m(t)} dt \right|^2 \qquad \{3\}$$

where $g(\omega,t) = \exp(-t^2/\tau^2)\exp(i(\omega-\omega_o)t)$ and $\phi_m(t) = k\Delta n_m z R_m(t)$, where k is the free space wave-vector, the subscript m denotes the type of nonlinear phase response: self-phase modulation or plasma, Δn_m is the change in refractive index due to each contribution, and $R_m(t)$ is the phase response function of the given nonlinearity m which we assume as having shapes $R_{SPM}(t) = \exp(-t^2/\tau^2)$ and $R_{Plasma}(t) = erf(t/\tau)+1$.

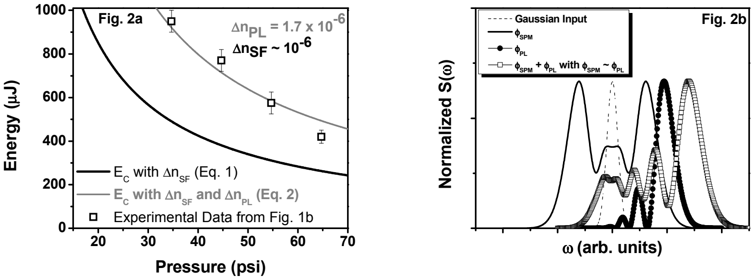

Fig. 2 **a)** Incident energy as a function gas pressure needed for stable filamentation.
b) Spectral broadening due to contributions from SPM and plasma production.

Fig. 2b shows the spectral broadening due to self-phase modulation, plasma production, and the combination of both. Including the phase accumulation just from self-phase modulation, we calculate a symmetric broadening around our incident Gaussian pulse. An anti-Stokes broadening is observed by only including contributions from the phase accumulation of the plasma. By incorporating both nonlinearities and setting their phases nearly equal, we observe a strong anti-Stokes broadening followed by a relatively weak Stokes broadening which is typically observed in continuum generation in gases [3].

CONCLUSIONS. We have presented a theoretical and experimental study of supercontinuum generation and characterization. The self-trapping model of Ref [6] includes refractive index contributions from two nonlinear mechanisms: self-focusing and plasma production. The input energy required to sustain a self-guided filament was experimentally determined and used to calculate the change in refractive index due to plasma formation. Utilizing the ionization rate and the nonlinear refractive index of krypton, we have experimentally and theoretically determined that for stable supercontinuum generation the phase accumulation due to self-phase modulation and ionization are nearly the same in value, as was suggested in Ref [5].

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