A route to supercontinuum generation in gas-filled hollow-core anti-resonant fibers is demonstrated through the creation of a broad vibrational Raman frequency comb followed by continuous broadening and merging of the comb lines through either rotational Raman scattering or the optical Kerr effect. The demonstration experiments, utilizing a single pump pulse with 20 ps duration at 532 nm in a nitrogen-filled fiber, produce a supercontinuum spanning from 440 to 1200 nm, with an additional deep ultraviolet continuum from 250 to 360 nm. Numerical results suggest that this approach can produce even broader supercontinuum spectra extending from the ultraviolet to mid-infrared.

1. Introduction

Stimulated Raman scattering (SRS)—a process of cascaded inelastic scattering of laser light from molecules—can create broad sets of discrete spectral lines from narrowband driving lasers. [1–3] Such Raman frequency combs have been widely studied in both free-space [4,5] and waveguide geometries [6,7] and found application in the synthesis of trains of light-field transients [8–10] as well as for efficient wavelength conversion. [11,12] By tightly confining both the gas molecules and laser light inside hollow-core photonic-crystal fibers (HC-PCF), such as anti-resonant guiding fibers, Raman frequency combs can be efficiently generated directly from quantum noise fluctuations and do not require multicolor pumping. [6,7,13–18] However, the Raman combs produced to date, while spanning multiple octaves, have always consisted of discrete spectral lines with at most moderate spectral broadening or shifting. [17,19]

In this paper we demonstrate, both experimentally and numerically, that SRS can create a broadband and smooth supercontinuum from a single narrowband pump laser. The key mechanism is a two-step process. At the start of the fiber, an extremely broad frequency comb is generated through a large vibrational frequency shift. At larger propagation distances this comb is subsequently broadened into a smooth supercontinuum by the influence of either rotational SRS, the optical Kerr effect (instantaneous nonlinear refractive index), or both of them combined. Interestingly, numerical simulations suggest that by combining the vibrational and rotational response, it is possible for the continuum to form by the Raman effect alone. This is an alternative regime of supercontinuum formation in optical fibers—conventional routes, such as soliton self-compression and fission dynamics, [20] modulational instability, [21] or self-phase modulation (SPM) and self-steepening, are all dominated by the contribution of the optical Kerr effect. Our numerical results suggest that our approach can produce supercontinuum spectra spanning from the ultraviolet to the mid-infrared (270 to beyond 2000 nm). Experimentally we generate a flat supercontinuum spanning 440 to 1200 nm and an additional deep ultraviolet continuum from 250 to 360 nm. These results are achieved by pumping a nitrogen-filled anti-resonant fiber with 20 ps pump pulses at 532 nm.

2. Theory and Numerical Simulations

We model the propagation of the electric field through the HC-PCF using a unidirectional pulse propagation equation [22,23]

\[ \partial_z E(z, \omega) = i \left[ \beta(z, \omega) - \frac{\omega}{v(z)} \right] E(z, \omega) + \frac{i \omega^2}{2c^2 \varepsilon_0 n(z, \omega)} \tilde{P}(z, \omega) \tag{1} \]

S.-F. Gao, Y.-Y. Wang
Guangdong Provincial Key Laboratory of Optical Fiber Sensing and Communication
Institute of Photonics Technology
Jinan University
Guangzhou 511443, China
E-mail: dearyingyingwang@hotmail.com
S.-F. Gao, Y.-Y. Wang, P. Wang
National Center of Laser Technology
Institute of Laser Engineering
Beijing University of Technology
Beijing 100124, China
F. Belli, C. Brahms, J. C. Travers
School of Engineering and Physical Sciences
Heriot-Watt University
Edinburgh EH14 4AS, UK

The ORCID identification number(s) for the author(s) of this article can be found under https://doi.org/10.1002/lpor.202100426

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where $\tilde{E}(z, \omega) = \mathcal{F}[E(z, t)]$ is the Fourier transform of the linearly polarized, mode-averaged electric field $E(z, t)$. $z$ is axial propagation distance through the fiber, $\omega$ is angular frequency, $\beta(z, \omega)$ is the frequency- and pressure-dependent propagation constant of the fundamental fiber mode, which we model using the hollow-capillary model [23-24] with the pressure-dependent dispersion of the gas, v = (2/3) n(z)c. $n(z)$ is the group velocity of the reference frame, c is the vacuum light speed, $\varepsilon_0$ is the permittivity of free space, and $\tilde{P}(z, \omega)$ is the Fourier transform of the nonlinear polarization

$$P(z, t) = \tilde{P}(z, \omega) + P^i(z, t) + P^r(z, t) + P^v(z, t)$$

The Kerr polarization is $P^k(t) = N\gamma^k E^3$ where $\gamma^k$ is the third-order hyperpolarizability and $N$ is the number density of molecules. The polarization due to ionization and the plasma response, $P(t)$, is calculated using the method of Geissler, [26] using the tunneling limit of the Peremolov–Popov–Terent’ev ionization rate [27] with an ionization potential of 15.38 eV. The vibrational polarization $P^v$ and the rotational response $P^r$ will be discussed below. The numerical parameters we use are given in Table 1. We neglect the loss of the fiber and the dispersion spikes arising due to the fiber resonances [28].

Table 1. Parameters for numerical simulations.

| Parameter | Unit | Value | Ref. |
|-----------|------|-------|------|
| $\gamma^k$ | 10⁻⁴⁴ cm³V⁻¹ | 95.2 | [29] |
| $\nu_c$ | THz | 69.85 | [30] |
| $\Delta \nu$ | THz | $\approx 0.04$ | [31] |
| $\Delta Q / Q$ | 10⁻²⁵ m² | 1.75 | [32] |
| $\mu$ | 10⁻¹⁶ kg | 1.16 | [33] |
| $B$ | m⁻¹ | 199 | [30] |
| $D$ | 10⁻⁶ m⁻¹ | 5.76 | [30] |
| $q_{\text{odd}}$ | 1 | | |
| $q_{\text{even}}$ | 2 | | |
| $\Delta a$ | 10⁻¹¹ m³ | 6.7 | [34] |
| $b_i$ | GHz amagat⁻¹ | $\approx 3.3$ | |

The generation of a Raman frequency comb from a single pump pulse can be initiated spontaneously from noise [35,36] and subsequently amplified through SRS. [7] The (nonlinear or inelastic) coupling of the light field with the stretching and alignment of homonuclear diatomic molecules is described through

Figure 1 shows numerical simulation results when including only the Kerr effect (only $P^k$ in the model) for a 26 μm diameter hollow fiber and pulses with 20 ps duration, 80 μJ energy and a wavelength of 532 nm. We considered a nitrogen pressure gradient from 1 to 40 bar, corresponding to our experimental parameters, which are detailed later. Additional numerical simulations (see Figure 5 and discussion around it) show that none of the dynamics we describe here depend on the pressure gradient, and would also occur in a statically filled fiber. Pure Kerr propagation without the Raman contribution results in negligible broadening, because the pump pulse and fiber parameters do not lead to either significant self-phase modulation or modulation instability. The spectral width induced by SPM is approximately given by $\Delta \omega \approx \phi / T_\tau$, where $\phi = (2/3)n_0P_0L/cA_{eff}$, $n_0$ is the average number density, and the factor $2/3$ accounts for the pressure gradient. For our parameters this gives $\Delta \omega = \Delta \omega / 2 \pi \approx 2.6$ THz, and as a consequence the spectrum barely changes.

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first-order perturbations of a nonrigid rotor model. As the vibrational frequency \( \nu_v \) is typically much larger than the rotational frequencies \( \nu_r \), and large enough that at room temperature the ground vibrational state is thermally populated, Raman scattering from molecular stretching is captured and modeled by a single vibrational transition. Therefore the time-dependent nonlinear polarization due to the vibrational coupling is described by:

\[
P^\nu = E(t) N(4\pi \varepsilon_0)^2 \kappa_v \int_0^\infty h(\nu_v, T_v, r) E(t - \tau)^2 d\tau
\]

where the Raman response function is \( h(\nu, T_v, r) = \sin(2\pi \nu v) \exp(-t/T_v) \), \( T_v = (\nu \Delta \nu)^{-1} \) is the dephasing time with \( \Delta \nu \) the full-width at half-maximum (FWHM) linewidth of the Raman transition, \( \partial \alpha / \partial Q \) is the isotropic averaged polarizability derivative with respect to the ensemble-averaged molecular stretch \( Q \), and \( \mu \) is the reduced molecular mass.

In Figure 1b, we include only the vibrational Raman response \( P^\nu \). Our parameters correspond to pumping close to the transient gain regime, and because the ratio \( \Delta \nu / \nu_v \) is relatively small (Table 1) the Raman response gives rise to a vibrational frequency comb with well-separated peaks at both lower (Stokes) and higher (anti-Stokes [AS]) frequencies separated by \( \nu_v \). Note that each line results from scattering from the previous line using the same (fundamental) vibrational transition, and hence the Stokes and AS lines are equally spaced in frequency. This is similar to previous results in light homonuclear gases such as hydrogen.[6,7,15–18]

As described in some of those previous studies, the smooth dispersion profile of gas-filled fibers allows for phase-matching of the anti-Stokes components of the comb.[6] For the parameters used here, vibrational comb formation occurs at around 80 cm, pure vibration: the spectrum appears very smooth and continuous. Over 20 rotational levels with \( \Delta J = 2 \). In nitrogen we can neglect the centrifugal contribution (\( B/D > 10^5 \)), and the rotational line spacing is \( \nu_r = (\nu_1 + \nu_0)/2 \), where \( \nu_1 \) and \( \nu_0 \) are the frequencies of the current and previous AS orders, and \( \Delta \nu_r \) is the propagation constant difference between the pump and first Raman Stokes frequency. The spectral evolution obtained for pure rotational SRS is shown in Figure 1c. It shows different behavior to the case of pure vibration: the spectrum appears very smooth and continuous, and mostly down-shifted to longer wavelengths; the overall spectral coverage is also much reduced. The frequency down-shift results from the slower rotational SRS response, resulting in an asymmetric, positive, phase-shift as described in detail elsewhere.[41] The vibrational comb is indeed formed when the phase-mismatch for each AS order, calculated as \( \Delta \nu = \nu_{AS1} - \nu_{AS2} + \Delta \nu_r \), is typically much larger than the rotational separation between the AS and previous AS order. a) The phase-mismatch for each AS order, calculated from (a). For all orders the dephasing length is longer than the distance over which each comb line is generated.

\[
P^\nu = E(t) N(4\pi \varepsilon_0)^2 \sum_j k_j \int_0^\infty h(\nu_j, T_v, r) E(t - \tau)^2 d\tau
\]

where \( k_j \) is Boltzmann’s constant, \( T \) is the temperature, and \( \eta_j \) is a statistical weighting factor that varies with odd and even \( J \) to account for nuclear spin (see Table 1). Selection rules limit the rotational transitions to pairs of levels with \( \Delta J = 2 \). For nitrogen at high pressure, the rotational Raman gain spectrum is continuous: at 40 bar, \( \Delta \nu_r = 0.15 \text{ THz} \), which is similar to the line spacing \( \nu_r/\nu_0 \). The rotational Raman polarization is approximated by a sum over all allowed rotational transitions:

\[
P^\nu = E(t) N(4\pi \varepsilon_0)^2 \sum_j k_j \int_0^\infty h(\nu_j, T_v, r) E(t - \tau)^2 d\tau
\]

where \( \Delta \alpha \) is the molecular polarizability anisotropy.

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where the pulse experiences an almost pure sinusoidal phase modulation, resulting in a frequency comb.

When combining the vibrational Raman and Kerr effects, the vibrational comb lines broaden into each other (Figure 1d), forming a broadband continuum. Figure 3 shows spectrograms of the key stages of evolution: vibrational comb formation, followed by spectral broadening of the comb lines into a supercontinuum. As noted previously, the pump pulse does not drive significant SPM. However, in the time domain the vibrational frequency comb corresponds to an ultrashort pulse train. Figure 4 shows that after 0.91 m propagation, the vibrational comb consists of a train of 2.1 fs pulses with a peak power of 20 MW. The formation of such ultrafast pulse structures, due to the coherent combination of all the Raman comb lines, is consistent with previous studies. The combination of shorter duration and higher peak power enhances self-phase modulation by a factor of $\approx 50\,000$ compared to the 20 ps pump pulses with a peak power of 3.8 MW, leading to spectral broadening of the comb. Furthermore, the individual comb lines broaden into each other to form a continuum. Previous Raman frequency comb experiments have either worked with much lower peak power and longer pump pulses, producing a pure frequency comb in which SPM never occurs and no continuum is formed, or with much shorter pulses, leading to immediate SPM broadening of the pump pulse itself. With our parameters, in contrast, SPM occurs only after the formation of the frequency comb.

While broadening of the vibrational comb due to the Kerr effect leads to a continuum, it is not the only mechanism that can achieve this. Remarkably, combining both vibration and rotation, but not the Kerr effect, as shown in Figure 1e, leads to a pure Raman supercontinuum spanning multiple octaves achieved via spectral broadening and merging of several vibrational lines. This works well in nitrogen, because the spacing and linewidth of the rotational levels are similar, which creates a near-continuous rotational Raman gain spectrum. However, we expect that this is a universal feature of molecular gases. Our numerical results indicate that in hydrogen (not shown), where the rotational gain spectrum consists of well-separated narrow lines, a smooth supercontinuum can still be formed.

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The similar broadening of the vibrational Raman comb into a supercontinuum by either the Kerr or rotational Raman response is not due to any similarity of those processes, but because both effects phase-modulate the ultrafast pulse structures underlying the vibrational comb (Figure 4) to varying degrees based on their peak power and temporal location. It is this non-constant phase modulation which leads to broadening and merging of the vibrational comb lines, and this is independent of the specific physical mechanisms involved.

Figure 1f shows the full supercontinuum simulations, including all contributions to the nonlinear polarization. Importantly it is the vibrational comb that establishes the full continuum extent,
covering three octaves, and then the Kerr and rotational Raman responses that broaden the vibrational lines to form a true continuum.

Ionization plays a negligible role in the dynamics—the low peak intensity (a maximum of $3 \times 10^{13}$ W cm$^{-2}$ is reached) leads to a negligible ionization fraction of 0.002%. This is in contrast to supercontinuum generation in gas-filled hollow fibers pumped with shorter pulses based on modulational instability,

The supercontinuum spectra shown in Figure 1 are an average of 100 simulations to account for shot-to-shot fluctuations. Like other noise seeded supercontinuum mechanisms, such as modulational instability, each laser shot produces slightly different spectrum. To illustrate these fluctuations the top panel of Figure 1f shows each individual shot as a thin grey line, with the average in blue. These fluctuations are not detrimental to most applications, which acquire time-averaged spectra.

In the numerical simulations described so far we used a gas pressure gradient from 1 to 40 bar, corresponding to our experiments. To check whether the pressure gradient is critical to the dynamics shown here, Figure 5 shows a simulation for a static gas pressure of 15 bar in the fiber, instead of a pressure gradient. All other parameters are identical to Figure 1f. The overall dynamics are virtually identical, confirming that the supercontinuum mechanism we have identified does not depend on a specific gas pressure or gradient. The difference in spectral extent at high frequencies is due to variations in phase-matching.

### 3. Experimental Section

These dynamics were experimentally verified using a single-ring nodeless hollow-core anti-resonant fiber consisting of six untouched thin tubes with diameter of 12 μm and membrane thickness of 210 nm forming a negative-curvature core shape with inscribed diameter of 26 μm. The fiber exhibited a minimum attenuation of 80 dB km$^{-1}$ at the pump laser wavelength of 532 nm and its first-order transmission band spans from 440 nm to $>1200$ nm with single-mode operation.

The ultraviolet transmission band was not measured, but results from finite element modeling indicated that it spanned 250 to 360 nm. A 10 m length of fiber was used, and for experimental convenience the input end was left open to atmosphere, while keeping the output end sealed inside a gas cell filled with 40 bar of nitrogen. The 532 nm pump laser emitted 20 ps duration pulses at 1 kHz, and was coupled to the fiber through a telescope and plano-convex lens. The output spectrum was collected by an integrating sphere and two optical spectrometers.

Figure 6 shows the output spectral evolution as the input pulse energy increased from 0.5 to 18.4 μJ coupled into the fiber. A wide supercontinuum spectrum was generated, in agreement with the numerical simulations, spanning more than 775 nm from 440 to 1200 nm (the long-wavelength limit of our measurements). Furthermore, observe deep ultraviolet (DUV) generation was observed from 260 to 360 nm. The presence of the DUV component was further confirmed in the inset. The spectral gap from 375 to 440 nm was due to the high resonance loss in the hollow-core fiber,

Figure 6b shows the output spectral evolution as the input pulse energy is gradually increased from 0.5 to 18.4 μJ as indicated. The pump line is labeled (p) as are the discrete vibrational Stokes lines at 607 nm (s1), 707 nm (s2), 847 nm (s3), and 1056 nm (s4).
the near infrared, ultrafast coherent pulse shaping of a dispersive wave in the ultraviolet, and asymmetrical spectral broadening for pulse post-compression.

With much shorter pump pulses, Raman effects combine with a strong Kerr contribution, allowing for enhanced supercontinuum generation. In the impulsive regime, with a pump pulse duration close to the Raman oscillation period, Raman dynamics combined with Kerr-driven soliton self-compression have enabled supercontinuum generation from the vacuum ultraviolet to the near infrared, ultrafast coherent pulse shaping of a dispersive wave in the ultraviolet, and asymmetrical spectral broadening for pulse post-compression.

The use of intermediate pulse durations of a few picoseconds leads to completely different nonlinear dynamics characterized by efficient noise-seeded generation of vibrational lines and a frequency comb followed by spectral broadening and supercontinuum generation. Recently a single frequency-shifted vibrational line was generated by pumping an atmospheric air-filled HC-PCF with 6 ps pulses and this line broadened, along with the pump spectrum, into a high-power sub-octave continuum. Here we have shown that even a broad frequency comb, consisting of a multitude of vibrational lines (we experimentally demonstrate ten vibrational lines, numerically more than twelve) can be broadened into a flat continuum. This dramatically increases the width of the continuum to cover multiple octaves. For the fiber parameters, gas species and pressure that we use, the pump pulses cannot drive Kerr-based broadening mechanisms, such as self-phase modulation, soliton dynamics, or modulational instability, by themselves. Instead, it is only through the sequential effects of vibrational comb formation and consequent breakup of the long pump pulse into few-femtosecond pulses, which then proceed to drive both rotational Raman scattering and self-phase modulation, that the continuum is created.

We expect our observations to be a general result for other molecular gases too, albeit with different characteristics. This scheme could be expanded for VUV and mid-infrared supercontinuum generation by altering the pump wavelength and using a fiber with suitably tuned resonances. Furthermore, this technique may be scalable to high average power by increasing the repetition rate. Research in that direction must carefully consider possible heating of the gas due to excitation through Raman scattering.

4. Discussion

Experiments in molecular gases in HC-PCF to date have mostly concentrated on pumping with pulses of much longer duration than the Raman oscillation period. Apart from comb generation, this system has also been used for extremely efficient Raman conversion and generation, solitary pulse generation, self-similar evolution, and supercontinuum up-conversion. With much shorter pump pulses, Raman effects combine with a strong Kerr contribution, allowing for enhanced supercontinuum generation. In the impulsive regime, with a pump pulse duration close to the Raman oscillation period, Raman dynamics combined with Kerr-driven soliton self-compression have enabled supercontinuum generation from the vacuum ultraviolet to the near infrared, ultrafast coherent pulse shaping of a dispersive wave in the ultraviolet, and asymmetrical spectral broadening for pulse post-compression.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

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