Ultrahigh Efficiency Laser Wavelength Conversion in a Gas-Filled Hollow Core Photonic Crystal Fiber by Pure Stimulated Rotational Raman Scattering in Molecular Hydrogen

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We report on the generation of pure rotational stimulated Raman scattering in a hydrogen gas hollow-core photonic crystal fiber. Using the special properties of this low-loss fiber, the normally dominant vibrational stimulated Raman scattering is suppressed, permitting pure conversion to the rotational Stokes frequency in a single-pass configuration pumped by a microchip laser. We report 92% quantum conversion efficiency (40 nJ pulses in 2.9 m fiber) and threshold energies (3 nJ in 35 m) more than 1×10^6 times lower than previously reported. The control of the output spectral components by varying only the pump polarization is also shown. The results point to a new generation of highly engineerable and compact laser sources.

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Though the past decade has seen a dramatic growth in the number of compact laser sources, some wavelength bands are still unavailable, hampering technological development. One simple way of filling the remaining gaps is through wavelength conversion by nonlinear optical processes such as stimulated Raman scattering (SRS). Historically, SRS has been plagued by high pump power requirements, inefficient conversion to the desired frequency, and incidental conversion to other, unwanted frequencies. Previous experiments on SRS using single-pass gas cells required pulsed lasers with high peak power (a few MW) to reach threshold. We have previously demonstrated that this reliance on high power laser sources can be avoided by using a hollow-core photonic crystal fiber (HC-PCF) filled with a Raman active gas, resulting in lowering the threshold energy for vibrational SRS generation in hydrogen to the sub-micro-Joule level [1]. This value was more than 100 times lower than previously reported in a single-pass configuration.

Here we report the use of the latest ultralow loss HC-PCF to construct hydrogen gas cells that provide tight confinement of laser light, in a single transverse mode, over extremely long (tens of m) interaction lengths. This makes possible even larger reductions in threshold power P_{th}:

$$P_{\text{th}} = \frac{A_{\text{eff}}}{g} \frac{\alpha_{\text{p}}(G + \alpha_{\text{s}} L)}{1 - \exp(-\alpha_{\text{p}} L)}, \tag{1}$$

where A_{eff} is the effective area of the fiber mode, L is the fiber length, g is the Raman gain coefficient of the gas (a few cm per GW), α_{p} and α_{s} are the intensity loss coefficients (m^{-1}) at the pump and Stokes frequencies, respectively, and $G \equiv gLP/A_{\text{eff}}$ is the net gain factor. Defining P_{th} as the power at which the conversion efficiency to the Stokes sideband reaches 1% (i.e., $G = 25$ [2]), single-pass SRS generation at pump powers well below 1 W is within reach using the latest HC-PCF with transmission losses less than 2 dB/km [3].

An associated long-standing problem, limiting the use of SRS in laser wavelength conversion, is conversion of light to unwanted frequencies via other Raman resonances, higher order Raman shifts, or wave mixing between generated Raman lines. Previously, these competing effects unavoidably limited the conversion efficiency to the desired frequency. For example, pure rotational SRS generation has always been difficult because the vibrational transition $Q_{01}(1)$ (frequency shift $\sim$125 THz) has much higher Raman gain than the rotational transitions [e.g., the $S_{00}(1)$ with a shift of $\sim$18 THz] (see Ref. [4] for the notation). Consequently, most reported experiments on SRS in hydrogen have been for vibrational SRS. Indeed, pure rotational SRS has never been demonstrated in a single-pass configuration and, in fact, was only recently observed in a high finesse Fabry-Perot cavity tightly locked to the laser frequency, with conversion efficiencies not exceeding 5% [4].

In our previous work [1] we used a HC-PCF that guided via an incomplete band gap over a broad wavelength range [5,6] with losses of $\sim$1 dB/m. Here we use a PCF that guides via a full photonic band gap but with a narrower bandwidth and much lower transmission loss. The narrower guidance band allows us to suppress the usually dominant $Q_{01}(1)$, which would produce a Stokes signal lying outside the low-loss guidance band. The experimental setup is essentially identical to the one we reported in [1], except that the pump signal is generated by a much less powerful miniature laser source—a passively Q-switched frequency-doubled Nd-doped yttrium aluminum garnet microchip laser delivering 0.8 ns pulses at wavelength 1064 nm and repetition rate 6.3 kHz, with a maximum energy of 1 μJ. The laser power was controlled using a half-wave plate and polarizing beam splitter, the polarization state of the light being controlled using a quarter-wave plate before being launched into the PCF. The signal transmitted through the PCF was then monitored using photodetectors and an optical spectrum analyzer.

The fiber [Fig. 1(a)] was fabricated by the stack-draw technique [7]. It had a core diameter of 7.2 μm and a triangular photonic crystal cladding with an interhole spacing of $\sim$3.1 μm and an air-filling fraction of $\sim$90%. The HC-PCF transmitted over the 1000–1150 nm range with a loss $<$100 dB/km, the minimum loss being 67 dB/km at 1060 nm. The second Stokes frequency (two steps of 18 THz away from the pump and located at 1215 nm) was guided with a loss of 0.6 dB/m.

The fiber was filled with hydrogen to a pressure of 7 bars using the technique described in [1]. In order to efficiently and selectively generate rotational SRS, a circularly polarized laser beam is preferable, because the gain is 1.5 times higher than if the pump light is linearly polarized [8–10]. The pump and Stokes waves, if both circularly polarized in opposite senses, interfere to yield a linear polarization that rotates at the same rate as the molecules. Under these conditions rotational energy is transferred efficiently to the molecules and SRS ensues. Moreover, since the vibrational Raman lines lie outside the transmission bandwidth of the HC-PCF, their overall gain is substantially reduced, giving us an ideal means of achieving full quantum conversion to the rotational Stokes frequency.

The transmitted signal was measured as a function of fiber length by repeatedly cutting back a fiber with a starting length of 35 m. Figures 2(a) and 2(b) show, for two extreme fiber lengths (35 and 2.9 m, respectively), the evolution of the ratio of the transmitted to launched power for the pump (1064 nm) and the first Stokes (1135 nm), respectively. For the longer fiber length, the threshold energy was $\sim 3 \pm 2$ nJ ($\sim 3.75 \pm 2.5$ W peak power). This value is more than 1×10^6 times lower than the lowest value reported in conventional experiments for rotational SRS generation [11–13].

For the shorter fiber length [Fig. 2(b)], although the threshold increased to $\sim$20 nJ, the conversion efficiency was much higher [Fig. 2(a) and 2(b)], reaching a maximum of 86% for the 2.9 m length, compared to only 35% in the longer length. This corresponds to a photon conversion efficiency of 92%, which is to our knowledge the highest ever reported. In Fig. 2(b) it can also be seen that the Stokes signal slowly decreases (over a relatively large pump energy range) after leveling off. The start of this decrease, at around 60 nJ, coincides with the appearance a second Stokes signal (1216 nm). All these observed features agree with numerical solutions of the coupled wave equations where four-wave mixing of the first anti-Stokes (i.e., the high-frequency sideband), the pump, the first Stokes, and the second Stokes are taken into account [14]. Also, in the numerical model, we have assumed a slight ellipticity of the pump in order to take into account the distortion in the laser polarization introduced by the imperfections in the different optical components and by the residual birefringence of the fiber.

Figure 3 shows the dependence of the threshold power for different fiber lengths. The threshold was taken to correspond to an energy conversion fraction of around 2%. The measured threshold level increased as the fiber length shortened and shows good agreement with the theoretical prediction represented by the full and dashed curves, representing the two most extreme values of the Raman gain coefficient found in the literature [4,11,12]. These values were corrected to the pump wavelength used in our experiment using the dependence of the gain coefficient with the pump wavelength given in [15].

The numerical calculations also showed that, for fiber lengths shorter than $\sim$5 m, there is a range of pump

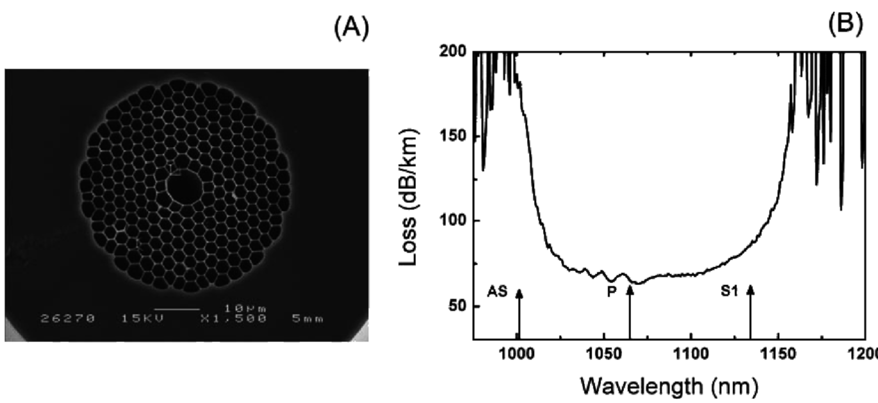

FIG. 1. (a) Scanning electronic micrograph of the HC-PCF. (b) Loss spectrum of the fiber. The arrows pointing upward indicate the wavelength of the pump (P), the first Stokes (S1), and anti-Stokes (AS).

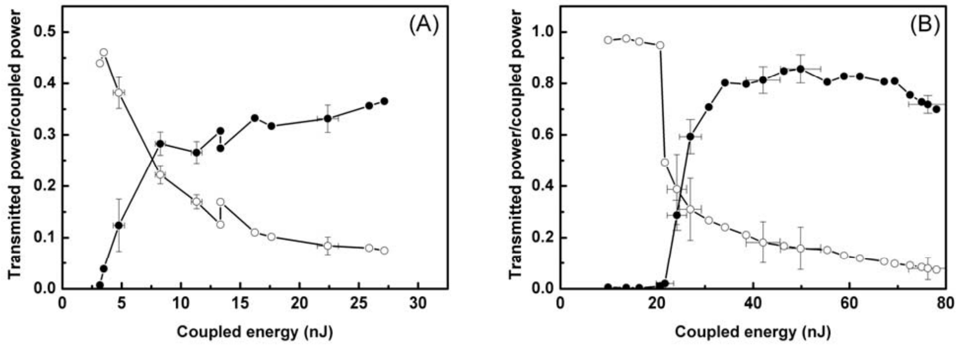

FIG. 2. Evolution of the ratio of transmitted average power over that of the coupled average power as the coupled pump energy is varied for the pump (open circles) and the Stokes (solid circles) in the case of a fiber length of (a) 35 m and (b) 2.9 m.

energies where the conversion efficiency plateaus around 98%—in a good qualitative agreement with the experimental growth at 2.9 m seen in Fig. 2(b). This suggests that efficient and flexible Raman-based frequency converters can be engineered simply by choosing an appropriate fiber length for a given dynamic range of the pump power.

Motivated by the lack of published experimental data, we also investigated the influence of the pump polarization on the coupling between the Stokes and the anti-Stokes signals in rotational SRS. This coupling is maximized when the Raman gain (m^{-1}) is much greater than the phase mismatch rate (m^{-1}) of the coupled waves, i.e.,

$$gP/A_{\mathrm{eff}} \gg -(\lambda_P^2/2\pi c)D(\lambda_P)\Omega_R^2 + 2\gamma P, \qquad (2)$$

where P is the peak power in the pump signal, Ω_R is the angular Raman frequency, c is the speed of light in vacuum, λ_P is the pump wavelength, γ ($\mathrm{m}^{-1}\mathrm{W}^{-1}$) is the Kerr nonlinear coefficient for the hydrogen filled HC-PCF, and $D(\lambda_P)$ ($\mathrm{ps\,nm^{-1}\,km^{-1}}$) is the group velocity dispersion at the pump wavelength [9]. Under our experimental conditions where the dispersion is anomalous at 1064 nm ($D(1064) \sim 50$ ps/km/nm), Eq. (2) is fulfilled for P greater than $\sim 50W$, when the Stokes to anti-Stokes intensity ratio grows as

$$\left|\frac{E_S}{E_{AS}}\right|^2 = \cot^2(\psi - \pi/4), \qquad (3)$$

where ψ is the polarization angle. The experimental measurements are plotted in Fig. 4(a) along with the theoretical predictions and show that for a circularly polarized pump beam the ratio increases dramatically in good agreement with theory. Furthermore, by simply changing the polarization state and power of the pump laser, we were able to control, over a wide range, the amount of power at the pump, first Stokes, second Stokes, and first anti-Stokes frequencies.

To illustrate this, Fig. 4(b) shows four different spectra generated by changing the pump beam polarization or the pump power or both. The magnitude of each spectral component is normalized to that of the transmitted pump. The presence or absence of the anti-Stokes or the second Stokes components in each spectrum illustrates how easily the various processes can be suppressed or enhanced.

In conclusion, HC-PCF enables pure rotational SRS in hydrogen at power levels some 6 orders of magnitude lower than previously reported in a single-pass configuration, and quantum efficiencies close to 100% [11–13]. The well-controlled single guided mode, and absence of beam diffraction, makes it straightforward to maintain phase matching without the need to change the focusing configurations as in more conventional experiments [11,12]. This allowed, for the first time, a convincing exploration of the influence of the pump beam polarization on the Stokes to anti-Stokes coupling close to phase matching. By an appropriate choice of HC-PCF and launch conditions (power and polarization state), it was also possible to control the strength of the different wavelength bands in the output spectrum.

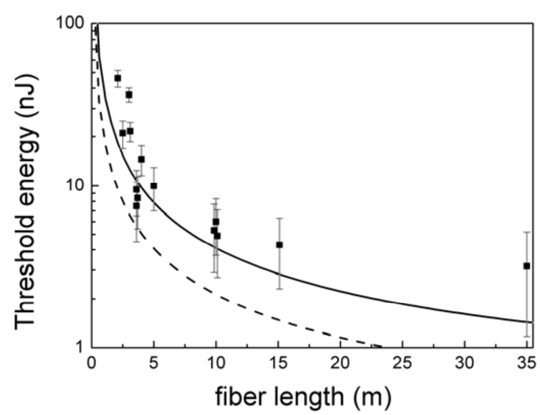

FIG. 3. The evolution of the threshold energy with the fiber length. The points are experimental measurements. The lines are from theory for $g = 1.5$ cm/GW (solid line) and $g = 2.68$ cm/GW (dashed line), taken from Refs. [4,11,12], respectively.

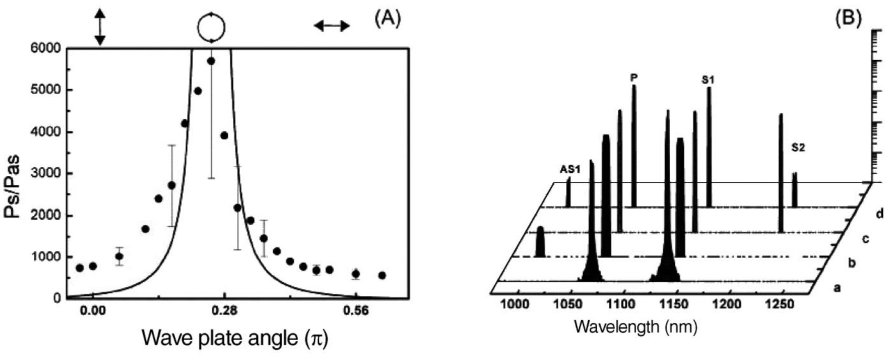

FIG. 4. (a) Stokes to anti-Stokes power ratio as a function of the angle of the quarter-wave plate. The points are the experimental data and the solid line is the theoretical prediction. (b) Four different spectra illustrating the control of the output spectra by changing only the polarization and the input pump power. The coupled energy, the polarization, and the fiber length for each spectrum are (a) circular, 48 nJ, 5 m (b) linear, 30 nJ, 3.6 m (c) elliptical, 20 nJ, 3.6 m (d) elliptical, 87 nJ, 3 m, respectively. The vertical axis represents the transmitted power normalized to that of the transmitted pump.

Gas-filled HC-PCF is an excellent vehicle for generating laser light at wavelengths where semiconductor lasers do not operate even though need is high (e.g., in biomedical applications). The possibility of generating multicomponent spectra at very low threshold power suggests the possibility of practical SRS-based mode-locked lasers [16].

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