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Blue-shifted dispersive wave generation by the diffraction-arrested solitons for coherent anti-Stokes Raman scattering microscopy in a photonic crystal fiber



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ABSTRACT

The broadband blue-shifted dispersive waves (DWs) are efficiently generated by the diffraction-arrested solitons in a photonic crystal fiber (PCF) designed and fabricated in our laboratory. By optimizing the pump parameters and the fiber length, the DWs can be used as the pump pulses for the high resolution coherent anti-Stokes Raman scattering (CARS) microscopy. The CARS microscopy based on the broadband DWs can be an attractive tool for simultaneously measuring the vibrational dephasing times of multiple Raman modes of the biological and chemical samples with the C–H and O–H stretch vibration resonances of 2700–3000 cm⁻¹ and 3000–3750 cm⁻¹.

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1. Introduction

Coherent anti-Stokes Raman scattering (CARS) microscopy as a powerful tool for the chemical imaging of biological and material systems is a high resolution nonlinear imaging technique with a fast data acquisition time, where a co-propagating pump beam and a Stokes beam with frequencies ω_P and ω_S interact with a sample through the third order susceptibility. The CARS signal can be generated and enhanced when the frequency difference between the pump wave and the Stokes wave matches with the Raman resonance frequency of the sample. Since its first demonstration in 1982 [1], the CARS microscopy has been successfully applied to the live cell imaging and chemical sample detecting. However, the set-ups for generating the pump and Stokes beams in the conventional CARS microscopy are complex and very expensive.

Because of the high nonlinearity in conjunction with the tailored dispersion profile of photonic crystal fibers (PCFs) [2–4], it is possible to tailor the output spectral distributions in PCFs for

concentrating the powers at the desired spectral ranges relevant for the CARS microscopy. The CARS microscopy with PCFs has attracted considerable attention [5–8]. Soliton self-frequency shift (SSFS) [9–11] as an effective frequency conversion technique experiences the continuous frequency down-shifting induced by the Raman effect, and the nonlinear optical process can be substantially enhanced due to a strong field confinement in a small core. Because the wavelength-dependent effective mode area can give rise to observable effects in the waveguide spectral broadening of ultrashort laser pulses, the diffraction-arrested solitons can be formed as the balance between the diffraction and index-step guiding is broken and the SSFS is slowed down [12]. At the same time, the dispersive waves (DWs), also known as Cherenkov radiations or nonsolitonic radiations, will be generated when the resonance conditions are satisfied [13–17].

In this paper, the blue-shifted DWs are efficiently generated by the diffraction-arrested solitons in a PCF, and are demonstrated to be suitable for the CARS microscopy, which can be an attractive tool for simultaneously measuring the vibrational dephasing times of multiple Raman modes of the biological and chemical samples with the C-H and O-H stretch vibration resonances from 2700 to 3000 cm⁻¹ and 3000 to 3750 cm⁻¹.

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2. Theory model

In order to analyze the process of SSFS in PCFs, the generalized nonlinear Schrödinger equation (GNLSE) is numerically solved by the split-step Fourier method, where the nonlinear polarization term including the retarded nonlinearity of the fiber material and the wavelength-dependent effective mode area is considered [12,18,19]:

$$\frac{\partial A(\xi,\tau)}{\partial \xi} = i \sum_{k=2}^{6} \frac{(i)^{k}}{k!} \beta^{(k)} \frac{\partial^{k} A(\xi,\tau)}{\partial \tau} + P_{nl}(\xi,\tau)$$
 (1)

where $A(\xi, \tau)$ is the field envelope, ξ is the propagation coordinate, τ is the retarded time, and $\beta^{(k)} = \partial^k \beta / \partial \omega^k$ are the coefficients in the Taylor-series expansion of the propagation constant β . The nonlinear polarization term $P_{nl}(\xi, \tau)$ is defined as follows:

$$P_{nl}(\xi,\tau) = i\hat{F}^{-1} \left(\frac{n_2 \omega}{cS_{eff}(\omega)} \hat{F}(A(\xi,\tau) \int_{-\infty}^{\infty} R(t) |A(\xi,\tau-t)|^2 dt) \right)$$
 (2)

where n_2 is the nonlinear refractive index of the fiber material, ω is the current frequency, c is the speed of light in the vacuum, $S_{eff}(\omega)$ is the frequency-dependent effective mode area, and the operators $\hat{F}^{-1}(\cdot)$ and $\hat{F}(\cdot)$ denote the inverse and no-inverse Fourier transforms. $P_{nl}(\xi,\tau)$ includes both the instantaneous, Kerr nonlinearity, and the retarded Raman contribution via the response function:

$$R(t) = (1 - f_R)\delta(t) + f_R\Theta(t) \frac{\tau_1^2 + \tau_2^2}{\tau_1 \tau_2^2} e^{-t/\tau_2} \sin\left(\frac{t}{\tau_i}\right)$$
 (3)

where f_R is the fractional contribution of the Raman response, $\delta(t)$ and $\Theta(t)$ are the delta and Heaviside step function, and τ_1 and τ_2 are the characteristic times of the Raman response of the fiber material. For the fused silica, f_R =0.18, τ_1 =12.5 fs, and τ_2 =32 fs.

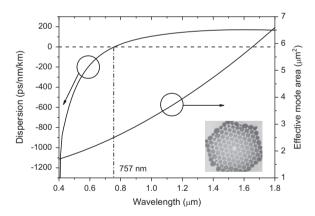


Fig. 1. The group velocity dispersion and effective mode area curves calculated for the fundamental mode of PCF, the inset showing the cross-section of PCF used in the experiment; the vertical dash-line corresponds to the zero dispersion wavelength of 757 nm.

3. PCF properties and experiment

Fig. 1 shows the group velocity dispersion and effective mode area curves calculated by the Multi-pole method (MPM) [20,21] for the fundamental mode of PCF designed and fabricated by the improved stack-and-draw technique [22,23] in our laboratory, and the cross-section of PCF with the core diameter of 2.05 μ m and the relative hole size of 0.89 is shown in the inset. As seen from Fig. 1, the zero dispersion wavelength is at 757 nm, and the effective mode area increases from 2.63 to 4.18 μ m² as the radiation wavelength increases from 0.8 to 1.255 μ m, by 59%.

As demonstrated in Fig. 2, a mode-locked Ti:sapphire ultrafast laser with the FWHM of 120 fs at a pulse repetition rate of 76 MHz is used as the pump source. The near transform-limited pulses are obtained by compensating the group velocity dispersion with a pair of fused silica prisms. The input pump power is adjusted by a variable attenuator, and an isolator is used to prevent the back-scattering light at the input end of the fiber from entering into the laser system. The pump pulses are coupled by a $40 \times$ objective into the PCFs, and the coupling efficiency can be up to 65%. The fundamental mode can be selectively excited by changing the distance between the input end of the fiber and the objective to exactly adjust the angle between the input beam and the fiber axis (the offset pumping technique). CCD1 and CCD2 are used to observe the coupling state of input light and the distribution of output mode field. The output spectra from the PCF are monitored by two optical spectrum analyzers (OSA, Avaspec-256 and Avaspec-NIR-256) with the measurement scopes of 200-1100 nm and 900-2500 nm and the resolutions of 0.025 nm and 15 nm.

4. Results and discussion

In Fig. 3(a), when the pump with the input average power of 300 mW (the peak power of 36 kW) works at 800 nm and the fiber length L increases from 40 to 50 cm, the diffraction-arrested fundamental solitons are formed due to the interplay between the negative dispersion and the self-phase modulation (SPM) along with the distinct increase of the effective mode area, and shifted from 1200 to 1255 nm induced by the intrapulse Raman scattering (IRS) as it propagates through the PCF. As a result of the perturbation induced by the higher-order dispersions and the resonance condition, the blue-shifted DWs are effectively generated at the central wavelengths λ_{DW} of 648 nm, 644 nm, and 637 nm, corresponding to the Raman shifts of 2932 cm⁻¹, 3028 cm⁻¹, and 3199 cm⁻¹, respectively, as shown in Fig. 3(b) and (c). The simulation parameters are chosen as follows: the peak power of Gauss-shaped pump pulse $P_p=36$ kW, the FWHM of pulse T_{FWHM} = 120 fs, and the nonlinear-index coefficient of fused silica n_2 = 2.6 × 10⁻²⁰ m²/W. The nonlinear polarization term is considered from Eq. (2) to include the effect of the wavelengthdependent effective mode area. The simulation result agrees well with the experimental one for L of 50 cm. The inset of Fig. 3(b)

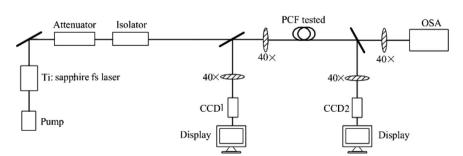


Fig. 2. The experimental set-up.

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