Design of supercontinuum source for coherent anti-Stokes Raman scattering microscopy*

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A new method to obtain supercontinuum(SC) source for multiplex coherent anti-stokes Raman scattering (CARS) microscopy is proposed. The nonlinear propagation in photonic-crystal fibers (PCF) of femtosecond pulse laser with central wavelength at 800.9 nm is studied with scalar wave theory. Based on the incident laser power and dispersion of PCF, super broadband source for multiplex CARS microscopy is designed.

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Coherent anti-stokes Raman scattering (CARS) is a third-order nonlinear optical process that involves a pump and a Stokes laser beam at frequencies of ω_p and ω_s, respectively. The CARS signal at the anti-Stokes frequency of $2\omega_p$-ω_s is resonantly enhanced when ω_p-ω_s is tuned to a Raman band. CARS microscopy has several advantages. First, in contrast with infrared imaging, CARS imaging based on the inherent vibration properties of molecules doesn't require staining or molecules tagging. Second, the CARS signal is not only much larger than the spontaneous Raman signal but also directional for a sizable sample. Third, it doesn't suffer from fluorescence background because the signal is detected at a wavelength shorter than those of incident beams.

The initial method to obtain stokes beam uses stokes frequencies produced by spontaneous Raman of the samples. But its intensities are too weak as the Stokes beam for CARS. As the developing of laser technology, two laser sources are introduced and succeed in tuning the intensity and frequency of the Stokes beam. Dye laser pumped by Nd:YAG laser[1] and the second harmonic waves in idler beams generated by optical parametric oscillator[2] are the most widely used as the Stokes beam. However, it is difficult to control the pump and Stokes beam synchronous and the shifting of two beams in time will result in imaging distortion. Recently, Nirit Dudovich[3] obtained single pulse CARS by using quantum coherent control technique, which solves the problem of getting the beams synchronous. But the spectra coverage of CARS is restricted by the bandwidth of the Stokes laser and is therefore less suitable for the detection of many molecules with broad vibration energy level difference. In this paper, we develop multiplex CARS microscopy using the supercontinuum(SC) spectra generated in photonic-crystal fibers (PCF) as the Stokes beam. Dispersions in PCFs are designed by modifying the design of the fiber structure, which offers a new solution for the enhancement of nonlinear-optical interactions of ultrashort laser pulse. The nonlinear propagation in PCF of femtosecond laser pulses with central wavelength at 800.9 nm is studied with scalar wave theory under the slowly varying envelope approximation condition. Finally, based on the analysis of SC spectra generated in PCF, super broadband source for CARS microscopy is designed as a convenient, flexible and preferable CARS source.

The dispersion features of index-guide PCF are firstly studied with the effective refractive index model [4]. The zero-dispersion wavelength (ZDWL) decreases with the radius of air hole and increases with the radius of the core, as shown in Fig.1. Two typical kinds of PCF are chosen to be studied in details. The ZDWL, core radius, air hole radius and pitch of the first PCF are 0.751 um, 1.0 um, 0.75 um and 1.8 um, respectively. Those parameters of the second PCF are 0.8 um, 1.1 um, 0.7 um and 1.8 um, respectively.

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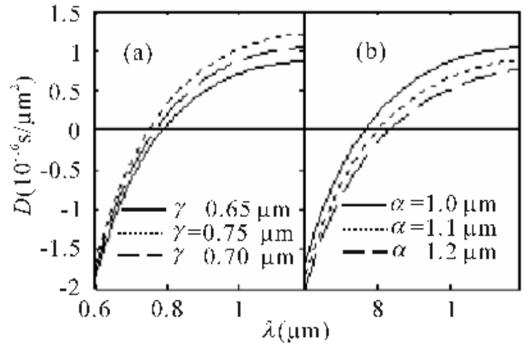

Fig.1 (a) Group-velocity-dispersion (GVD) coefficient of the PCFs with different air radius ; (b)GVD coefficient of the PCFs with different core diameters.

The propagation equation of femtosecond pulses transmitting in the nonlinear fibers is given by [4, 5]

$$\frac{\partial A}{\partial z}+\frac{\alpha}{2}A+\frac{i\beta_2}{2}\frac{\partial^2 A}{\partial T^2}-\frac{i\beta_3}{6}\frac{\partial^3 A}{\partial T^3}+\frac{i\beta_4}{24}\frac{\partial^4 A}{\partial T^4}-\frac{i\beta_5}{120}\frac{\partial^5 A}{\partial T^5}+\frac{i\beta_6}{720}\frac{\partial^6 A}{\partial T^6} = i\gamma[|A|^2+\frac{i}{\omega_0}\frac{\partial}{\partial T}(|A|^2A)-T_R A\frac{\partial |A|^2}{\partial T}] \quad (1)$$

where A, α, β_i and γ are the slowly varying envelope amplitude, attenuation coefficient of fibers, the i^{th} order GVD constant and nonlinear coefficient of the core materials, respectively. T_R is given by

$$T_R = \int_{-\infty}^{\infty} tR(t)dt \quad (2)$$

where $R(t)$ is the response function including both the electronic and vibration contributions.

The mode-locked Ti: sapphire laser is used as laser source. The repetition rate, pulse width and central wavelength are 76 MHz, 123 fs and 800.9 nm, respectively. SC spectra generated in the first kind of PCF with different input powers are plotted in Fig.2, where the second-order GVD constant of PCF is -10.9×10^{-6} fs^2/nm at 800.9 nm. The incident peak powers are 200 W, 500 W, 1 000 W and 4 000 W in Fig.2 (a), Fig.2 (b), Fig.2 (c) and Fig.2 (d) , respectively. The SC spectra indicate higher powers result in stronger self-phase modulation effect. Non-soliton radiation [6] (NSR) peaks shift towards blue and the width of SC spectra becomes wider when the incident power is enhanced.

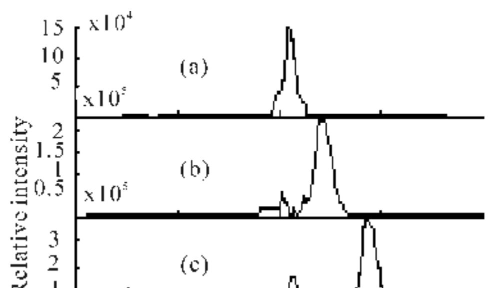

Fig.2 Output spectra dependence on input power

What is the most attractive is that SC spectra generated in the two kinds of PCF with different ZDWL are fully simulated. There is about two order difference between the second-order GVD constants of the two kinds of PCF, as mentioned in Fig.1. Besides, the nonlinear coefficient is different when the core radius of the two kinds of PCF are different, which is referred to

$$\gamma = \frac{n_2 \omega A_{\mathrm{eff}}}{c}, \quad (3)$$

where n_2, ω and A_{eff} are nonlinear reflective index, incident circle frequency and effective area, respectively. However, the difference in γ is slight. Fig.3 (a) and Fig.3 (b) show the output spectra in the first and the second kinds of PCF when the peak power is 1 000 W. The output spectra in the PCF with ZDWL at 0.751 um and 0.8 um are shown in Fig.3 (c) and Fig.3 (d) respectively, when the incident pulse peak power is 4 000 W. Fig.3 indicates that nonlinear transmitting of pulse in PCF with smaller dispersion exhibits stronger self-phase modulation and is more sensitive to the incident power. SC generation influenced by the core radius difference is slightly. Therefore, we come to the conclusion that the PCF with ZDWL closer to laser central wavelength is more suitable for CARS microscopy when the pulse laser transmits in the anomalous dispersion regime of PCF.

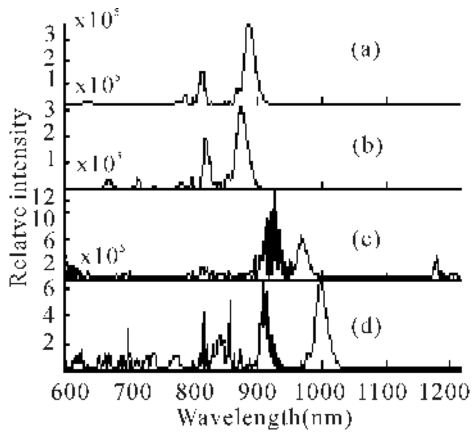

Fig.3 Output spectra in the PCF with different zero-dispersion wavelength.

Based on the above descriptions about SC spectra, a promising light source for CARS microscopy is developed. This method also provides solutions for suppressing non-resonant noises, which is the major drawback for CARS microscopy. Although polarization CARS [7] and heterodyne CARS [8] have been used for background rejection, these techniques also attenuate the resonant contribution and need complicated setups. In our method, the SC spectra generated in the second kind of PCF have demonstrated over a 2 500 cm^{-1} wave number region, which can excite multiplex CARS signals, as shown in Fig.4. The coherent superposition of multiplex CARS signals can efficiently enhance the resonant CARS intensity and significantly improve the signal-to-background ratio. Fig.5 illustrates the scheme of multiplex CARS. An unamplified mode-locked Ti: Sapphire laser is used as the laser source. A large portion of the output from the laser is used for the seed laser to generate the SC spectra in the PCF when the SC coverage associates with incident power. The fundamental of the Ti: Sapphire laser and the SC are used for the pump and Stokes lasers, respectively. Then the two laser pulses are collinearly superimposed and tightly focused onto the sample with a microscope objective. The experiment is carried on in laboratory.

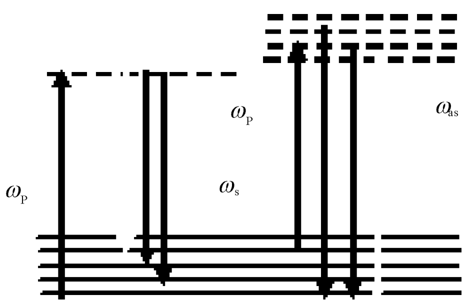

Fig.4 Energy level diagram for multiplex CARS.

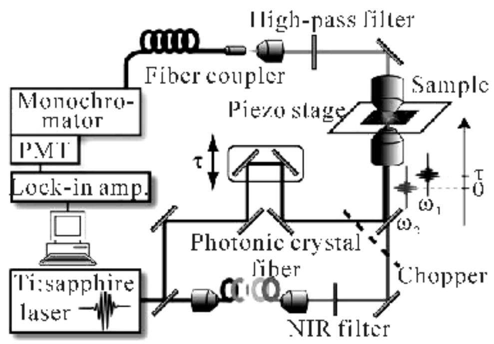

Fig.5 Experimental setup of multiplex CARS.

In this paper, we propose multiplex CARS microscopy using the SC spectra generated in PCF as the Stokes beam. Dispersion in PCFs can be designed by modifying the design of the fiber structure, which offers a new solution for the enhancement of nonlinear-optical interactions of ultrashort laser pulse. The multiplex CARS using SC spectra in PCF as Stokes source is a promising tool for imaging molecules and cells.

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