DEVELOPMENT OF NEAR INFRARED COHERENT ANTI-STOKES RAMAN SCATTERING MICROSCOPY USING SUPERCONTINUUM GENERATED FROM A PHOTONIC CRYSTAL FIBER

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Keywords: *coherent anti-Stokes Raman scattering, microscopy, photonic crystal fiber, supercontinuum*

Abstract: Coherent supercontinuum generated from a photonic crystal fiber is applied to a near infrared coherent anti-Stokes Raman (NIR-CARS) microscopy. A clear CARS image of polystyrene microsphere has been obtained using the CH_2-stretching band.

Coherent anti-Stokes Raman scattering (CARS) microscopy has been attracting much attention as a new technique for vibrational spectroscopic imaging [1-3]. Since CARS is based on the third-order nonlinear optical process, it has an inherent three-dimensional sectioning capability without a pinhole as a confocal microscope. In addition, owing to the intense, uni-directional CARS signal, it does not require long data-acquisition time. In order to generate the CARS signal, the vibrational coherence must be created by a difference frequency of two lasers, namely pump and Stokes lasers. However, it requires complicated laser systems such as two synchronized Ti:sapphire oscillators [1,3] or an optical parametric amplifier with a Ti:sapphire regenerative amplifier system [2]. Although several reports have been made of single pulse CARS [4], the frequency bandwidth is so narrow that it does not cover the fingerprint region. In the present study, we use a supercontinuum generated from a photonic crystal fiber (PCF) for the Stokes pulse. The PCFs consist of the microstructured cladding region with a hexagonal array of air holes to guide light into the pure silica core. By injecting readily available low-power ultrashort pulses into the PCF, an ultrabroadband phase-coherent supercontinuum spanning more than one octave has been achieved [5]. Owing to the easily handling and compactness, the supercontinuum has been used in various applications including spectroscopy [6-8].

Figure 1 shows a schematic overview of the experimental setup. The light source is an unamplified Ti:sapphire laser (Coherent, Vitesse-800). Typical duration, pulse energy, repetition rate, and peak wavelength are 100fs, 12nJ, 80MHz, and 801nm, respectively. The laser pulses are divided into the pump and seed pulses for the supercontinuum generation. The seed pulse is coupled into the PCF (Crystal Fibre, NL-1.7-690 or Mitsubishi cable, LFR-141) with a microscope objective. For the CARS signal generation, the fundamental of the Ti:sapphire laser (ω_1) and the

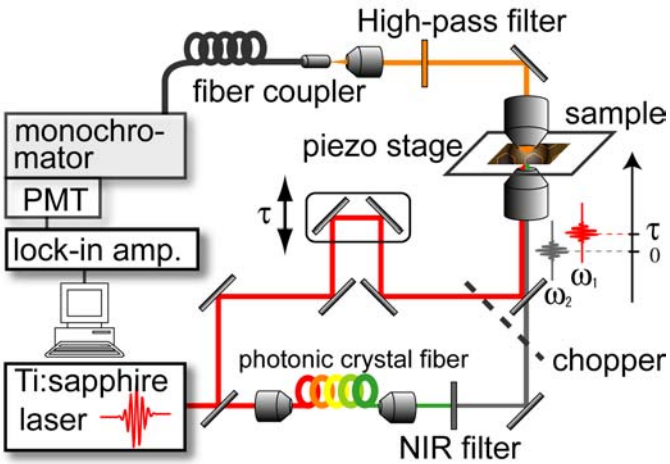

Fig. 1. Experimental setup for NIR-CARS microscopy.

supercontinuum (ω_2) are employed as the pump and Stokes pulses, respectively. In the present study, only the near-infrared (NIR) frequency part of the supercontinuum, namely 850-1100nm, is used for the Stokes pulse. Since all laser pulses are in the NIR region, we can expect low photo damage, suppression of the nonresonant background signal, and a deep penetration depth for the opaque sample. In these aspects, our setup, NIR-CARS microscopy, is contrasted with that of the previous report [7]. All three laser pulses are superimposed collinearly and then tightly focused using a microscope objective. Because of the large numerical aperture, the phase-matching conditions are relaxed due to the large angular dispersion and the short interaction length.

Figure 2 shows the CARS imaging of polystyrene microspheres with diameter of 3 μm in water on a coverslip. The detected vibrational frequency is 3055 cm^{-1}, which corresponds to the CH_2-stretching mode. As shown in Fig. 2, four spheres are clearly detected. A very weak nonresonant CARS signal due to water is also observed as an offset. In order to confirm that the signal originates from the CARS process, the excitation power dependence is examined. The CARS signal intensity, I_{CARS}, is reproduced well by $I_{\mathrm{CARS}} \propto I_{\omega_1}^{1.8\pm0.1} I_{\omega_2}^{1.1\pm0.1}$, respectively. Here I_{ω_1} and I_{ω_2} represent the intensities of the ω_1- and ω_2-lasers, respectively. These results are in good agreement with the theoretical expectation ($I_{\mathrm{CARS}} \propto I_{\omega_1}^2 I_{\omega_2}$). A further proof of our detection of CARS signal is the delay time dependence. The CARS signal is observed only when the ω_1- and ω_2-laser pulses are temporally overlapped.

In conclusion, NIR-CARS microscopy has been developed using supercontinuum generated from the PCF. It is simple, robust, and compact system. The vibrational imaging of the polystyrene microsphere is demonstrated at the CH_2-stretching region.

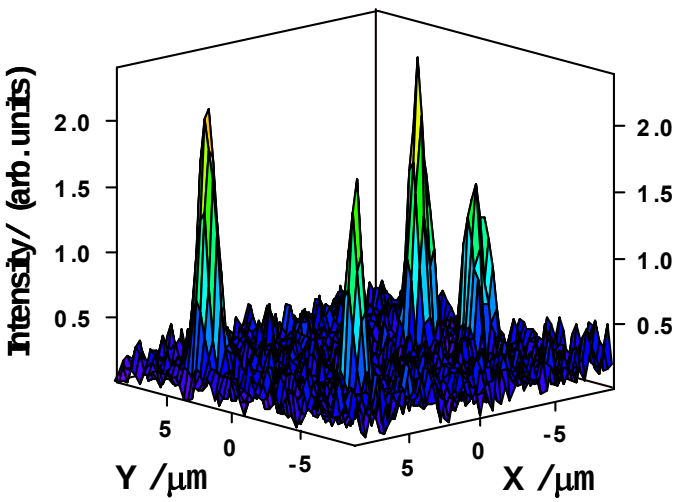

Fig. 2. CARS imaging of polystyrene microspheres.

Acknowledgements:
The authors would like to gratefully acknowledge Crystal Fibre A/S and Rikei co. for kindly providing the PCF. This research is partially supported by Grant-in-Aid for Young Scientists (B) (15750005) to HK.

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