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Investigation of fiber Bragg grating as a spectral notch shaper for single-pulse coherent anti-Stokes Raman spectroscopy

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ABSTRACT

We experimentally demonstrate compact and efficient single-pulse coherent anti-Stokes Raman spectroscopy (CARS) via spectral notch shaping implemented with a fiber Bragg grating. We show that a fiber Bragg grating can serve as a narrowband notch filtering component on a 90 nm broadband femtosecond pulsed laser without spectral distortion. Finally, we obtain CARS spectra of various samples in the fingerprint region of molecular vibrations. This scheme has potential for compact implementations of all-fiber single-pulse multiplex CARS due to its compatibility with fiber optics.

1. Introduction

Coherent anti-Stokes Raman scattering (CARS) has received a considerable amount of attention in the fields of spectroscopy and microscopy based on molecular vibrations [1,2]. CARS microscopy has enabled label-free measurements [1,2], and CARS spectroscopy has found applications for the stand-off detection of hazardous agents due to its excellent sensitivity [3–6]. The signal levels in CARS can be more than 5 orders of magnitude greater than those in spontaneous Raman scattering [2].

Despite the growing demands for using a CARS system in various fields, its commercialization is hampered by the complexity involved in CARS instrumentation. A typical CARS system requires spatial overlap and temporal synchronization of two pulsed lasers to create a beat frequency which makes the overall systems bulky in size, complex, and sensitive to alignment. To overcome these disadvantages, several research groups have developed novel methods for CARS systems [6–17].

Fiber-based light sources for CARS microscopy were recently developed to reduce size and alleviate the alignment issues [12–14]. The method used a narrow bandwidth light source, optimized to measure the C–H stretching vibrations, where it may show limitations when applied to measure the fingerprint spectral region [12–14]. Another study demonstrated fiber-based supercontinuum sources for the fingerprint region of CARS, in which it still remains alignment issues similar to those for a typical CARS system [16]. Single-pulse CARS is a novel method that uses only one pulsed laser to obtain the fingerprint region of molecular vibrations [6–9]. These methods can obtain a full spectrum of the sample simultaneously that it can be used in both microscopy and spectroscopy applications. The most recent system used a resonant photonic crystal slab as a pulse shaper [17] which requires careful selection of materials and suitable nanotechnology [18].

In this work, we demonstrate a new single-pulse CARS approach with a fiber Bragg grating, which is a widely used passive pulse-shaping component [10,19–21]. An important issue in the development of this approach is to propagate the broad bandwidth of a femtosecond pulsed laser in a fiber Bragg grating without significant spectral distortion. We found a suitable condition for using a fiber Bragg grating as a notch shaper in a femtosecond pulsed laser. Finally, we obtained CARS spectra of various samples in the fingerprint spectral region. Fiber Bragg gratings can also be easily manufactured with a narrowband and a high rejection rate such that the spectral resolution and sensitivity can be easily enhanced. Another advantage is that fiber Bragg gratings can be used as rapid pulse shaping modulators that allow for the use of lock-in detection [22,31]. This new scheme has potential for compact implementations of single-pulse CARS because of its natural compatibility with fiber optics [14,20].

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2. Principle and concept of single-pulse CARS

In general, CARS is a four-wave-mixing (FWM) process, and it can be described as [8,9]

$$P(\omega_p - \omega_s + \omega_{pr}) = P_R^{(3)} + P_{NR}^{(3)} = (\chi_R^{(3)}(\Omega) + \chi_{NR}^{(3)}(\Omega))E_p(\omega_p)E_s(\omega_s)E_{pr}(\omega_{pr}), \quad (1)$$

where P_R and P_{NR} are the resonance and non-resonance polarizations, respectively, from a third-order nonlinear process. The E_p, E_s, and E_{pr} are the electric fields of the pump, Stokes, and probe, respectively. The $\chi_{NR}{}^{(3)}$ is non-resonant third-order nonlinear susceptibility, which is not related to the Raman signal and generally has a constant value. The $\chi_R{}^{(3)}$ is a resonant third-order nonlinear susceptibility that it is related to the Raman spectrum of the sample, and it is defined as [8,9]

$$\chi_R = G/(\Omega_R - \Omega) + i\Gamma, \quad (2)$$

where $\hbar\Omega_R$ is the molecular vibrational energy with bandwidth Γ and G is a coefficient of the Raman strength. In single-pulse CARS, one pulsed laser simultaneously acts on the three electric fields in Eq. (1). Therefore, the resonant third-order nonlinear polarization is described as [8,9]

$$P_R^{(3)}(\omega) = G\int_0^\infty (E(\omega - \Omega)/(\Omega_R - \Omega) + i\Gamma)A(\Omega)d\Omega, \quad (3)$$

where $A(\Omega) = \int E^*(\omega - \Omega)E(\omega)d\omega$ is the vibrational excitation amplitude [8]. As shown in Eq. (3), when Ω is equal to ΩR, the equation has a dominant value because the denominator has the smallest number. At that point, if $E(\omega - \Omega R)$ has a value of almost zero because of the notch-filtered location in the pulsed laser, then the total integral equation should have a relatively small value compared to the left- and right-hand-side values. Therefore, the narrowband notch-filtered pulsed laser causes a dip in the third-order nonlinear signal. The energy difference between the notch-filtered location and the dip in the third-order nonlinear signal corresponds to a Raman energy level in a sample.

3. Investigation of the spectral distortion of pulsed laser with respect to group-velocity-dispersion pre-compensation

To obtain desirable CARS signals in the single-pulse CARS scheme, transform-limited laser pulses with a broad spectral bandwidth accompanying a narrowband notch shape are required. The detectable spectral range of the CARS signal is proportional to the bandwidth of a laser pulse used, whereas the CARS spectral resolution and the contrast of resonant CARS dips, on the other hand, are primarily determined by the bandwidth and rejection rate of the notch shape in the pulsed laser, respectively.

In our experiment, we generate a narrow notch-filtered pulsed laser by passing through a fiber Bragg grating (FBG) written in a single-mode fiber (SMF, 780-HP, Thorlabs). Therefore, we have to consider nonlinear effects in the optical fiber when the intense pulsed laser delivered with a SMF is subject to spectral distortion and temporal pulse broadening related to self-phase modulation (SPM) and positive group-velocity dispersion (GVD) in the SMF [23–27].

Since the optical fiber we used has a positive dispersion at 800 nm, we first examine whether the negative pre-chirped pulse can maintain its spectral shape with a short pulse duration after propagation in the optical fiber. For stable guiding of the laser pulse in the FBG, we investigated the proper conditions for the incident pulsed laser.

To evaluate the possibility of the pre-compensation scheme, the experimental setup was constructed as shown in Fig. 1(a). We started from a commercial Ti: sapphire femtosecond oscillator (Micra-10, Coherent) with an average power of 800 mW and central wavelength tunable in the range of 780 nm to 820 nm with a spectral bandwidth from 20 nm to 90 nm. The system consisted of a variable neutral density filter to change the input pulse energy and two chirped mirror sets (reflectivity $>99\%$, -175 fs^2 GDD per reflection, DCMP175, Thorlabs) to control the large number of GDD with high energetic efficiency. We used a quasi-parallel chirped mirror pair to control the number of reflections with respect to angle θ, as shown in Fig. 1(c). Then, the pulsed laser was launched into a 100 mm 780-HP fiber by coupling the objective lens (LU Plan, 20X/0.45, Nikon) with a coupling efficiency of 60%. Finally, the spectral shape was measured using an optical spectrum analyzer (MS9710C, Anritsu).

The total positive GDD value in our experiment was almost 14,000 fs^2. The GDD value of each optical component was estimated using a phase and intensity from correlation and spectrum only (PICASO) algorithm [28,29]. We used a Ti:S oscillator (Chameleon Vision-S, Coherent) equipped with an autocorrelator (AA-M autocorrelator, Avesta) and spectrometer (HR2000+, Ocean Optics). The automated dispersion pre-compensation system allows precise control over the GDD value in the range from 0 to $>22{,}000$ fs^2. The following presents the estimated GDD values of each of the optical components that we used: the summed value of the Faraday isolator, fiber coupling objective lens and several mirrors for guiding the laser was 8000 fs^2; the 100 mm SMF fiber was 4000 fs^2; and the sample objective lens was 2000 fs^2 (Fig. 1(b)). Therefore, before guiding in the SMF, a negatively pre-chirped pulse with a GDD of -6000 fs^2 is needed for the transform-limited pulse at the sample position.

Spectral narrowing is a common phenomenon accompanied by the laser pulse propagation in optical fibers with negative chirping [23]. We give particular attention to such spectral narrowing. Since the spectral extent of the CARS signal is proportional to the bandwidth of the pulsed laser, it is crucial to avoid the spectral narrowing of the laser. In contrast, the oscillatory and/or asymmetric spectral shape only leads to a nonuniform spectral efficiency of CARS signals for which the original CARS spectrum can be recovered by a proper spectrum normalization [15], meaning that the resolution of a resultant CARS spectrum is not degraded by the multi-peak spectral feature.

There are no actual experimental studies on the spectral distortion in the broad bandwidth of a short pulsed laser. Fig. 2 shows the changes in the spectral shape in the pulsed laser with a bandwidth of 40 nm and central wavelength of 800 nm with respect to input GDD from -6450 fs^2 to 4100 fs^2 for the same output power (i.e., 50 mW). In Fig. 2(a), the gradual spectral narrowing alleviation phenomenon is shown from negatively chirped to positively chirped pulses.

Previous numerical studies investigated the formation of triangular and parabolic spectral shapes in the pulsed laser after a short piece of 780-HP fiber delivery [26,27]. According to the simulation results, the condition of the initial lase pulse for triangular pulse shaping is generally shorter than the dispersion length with a large negative chirp parameter and higher soliton order [27]. Fig. 2(b) shows the triangular spectral shape in our experiment with a GDD value of -6450 fs^2. The fiber was shorter than the dispersion length with a chirp parameter of $-7.5 \le C \le -6.5$, and the soliton order was 8. Moreover, in the case of a small amount of negative chirp, the parabolic pulse shape was observed with a large undulation, as shown in Fig. 2(b).

All the experimental results were in good agreement with the numerical results obtained in previous studies [26,27]. We confirmed that there is strong spectral distortion due to nonlinearity and dispersion effects in the case of a negatively chirped pulsed laser. Therefore, in the case of an optical fiber with a small core