Spectra of spontaneous Raman scattering in taper-drawn micro/nano-fibers*

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(Received 5 July 2016; revised manuscript received 29 August 2016; published online 25 October 2016)

We study the spontaneous Raman scattering (RS) in taper-drawn micro/nano-fibers (MNFs) by employing the photon counting technique. The spectra of RS in five MNFs, which are fabricated by using different heating flames (hydrogen flame or butane flame) and with different diameters, are measured within a frequency shift range of 1435 cm^{-1}–3200 cm^{-1}. From the measured spectra, we observe the RS peaks originated from silica and a unique RS peak with a frequency shift of ∼2905 cm^{-1} (∼87.2 THz). Unlike the former ones, the latter one is not observable in conventional optical fibers. Furthermore, the unique peak becomes obvious and starts to rapidly increase with the decrease of the diameter of MNFs when the diameter is smaller than 2 μm, and the intensity of the unique peak significantly depends on the heating flame used in the fabricating process. Our investigation is useful for the entanglement generation or optical sensing using taper-drawn MNFs.

Keywords: nonlinear optics, micro/nano-fibers, Raman scattering

PACS: 42.65.–k, 42.81.Qb, 78.30.–j **DOI:** 10.1088/1674-1056/25/12/124205

1. Introduction

Raman scattering (RS) is the inelastic scattering of photons by a medium. The incident photons interact with molecular vibrations or other excitations in the medium, resulting in their energy being shifted up or down. The frequency down-shifted photons are referred to as Stokes photons, and the frequency up-shifted photons are referred to as anti-Stokes photons. Many applications take advantage of RS, such as Raman spectroscopy, Raman laser, and Raman amplifier.[1–3] While in some other applications, RS is considered as a crucial noise factor, such as optical parametric amplification and photon pair generation.[4–6] Therefore, it is important to characterize the RS spectrum of a particular medium, for the purpose of either enhancing or suppressing RS.

Micro/nano-fibers (MNFs) have the merits of a miniaturized modal area, engineerable dispersion, large evanescent field and strong near-field interaction, which make it promising for a variety of applications.[7] Among the various kinds of fabrication methods of MNF, taper-drawing of conventional optical fibers is a general and simple one, which fulfills the requirements of many applications, such as nonlinear optics, optical sensing, and manipulating atoms or quantum dots, etc.[8–11] In particular, the taper-drawn MNFs have a relatively high grade of geometric uniformity and surface smoothness, and their ends usually connect to conventional fibers through adiabatic tapers, which greatly facilitates the optical launching and mechanical handling.

So far, a number of nonlinear optical effects in taper-drawn MNFs have been investigated, such as supercontinuum generation, second/third harmonic generation, and four wave mixing.[12–15] However, there are a few works that focus on the RS in MNFs. In Ref. [16], the stimulated RS in a liquid probed by the evanescent field of MNFs, which can be used for sensing, has been reported. In Ref. [17], correlated photon pairs are generated via four wave mixing in MNFs, but the purity of the photon pairs is affected by RS. So it is important to study the RS in MNF in details. In this paper, we measure the spectra of spontaneous RS in taper-drawn MNFs by employing the photon counting technique,[5] identify the RS peaks with different origins, and characterize the influences of various factors on the RS spectra including the diameter of MNF and the heating flame used in the taper-drawing process.

The rest of this paper is organized as follows. The fabrication process of the MNFs and the experimental setup for the spectral measurement of RS are illustrated in Sections 2 and 3, respectively. We devote Section 4 to the experimental results and discussions. Our results show that, in addition to the RS peaks originated from silica, there exists a unique RS peak at

*Project supported by the National Natural Science Foundation of China (Grant Nos. 11304222 and 11527808) and the State Key Development Program for Basic Research of China (Grant No. 2014CB340103).

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$\sim$2905 cm^{-1} in the measured spectra, and the unique RS peak, in accordance with the carbon–hydrogen stretching vibration, is influenced by the heating flame and the field intensity at the surface of MNFs. Finally, we conclude in Section 5.

2. Fabrication of taper-drawn MNFs

The MNFs used in our experiment are fabricated by taper-drawing conventional single mode fibers (SMFs, model: SMF-28). Before the drawing process, a few millimeters of acrylate jacket is stripped from the middle part of an SMF, and a lens cleaning tissue soaked in ethanol is repeatedly wiped along the stripped section of fiber to remove the remaining acrylate powder. The SMF is then carefully clamped on the motor stages of the taper-drawing system.[15] During the drawing process, a small section of $\sim$1 millimeter of the fiber is heated by a heating flame. The heating flame employed in the taper-drawing system can be either a butane flame or a hydrogen flame. The fiber is first tapered down to approximately 50 microns in diameter, with the two stages moving in the opposite directions. Then the stages move equal-directionally, with the relative displacement increasing gradually. The pulling parameters are tuned to optimize the transmittance.

The typical uniform length of the taper drawn MNF, with diameters ranging from 1 μm to 3 μm, can reach more than 9 cm. The MNFs are inspected by using an optical microscope and no obvious fluctuation of the diameter within the uniform region is observed. Therefore, we deduce that the fluctuation of the diameter of the MNFs is less than 0.2 μm. The prepared MNF with two tapers is finally sealed into a plastic housing to prevent dust contamination and to ease the handling. At each end of the MNF, about one meter long SMF is retained. Table 1 lists the diameters, lengths, and heating flames of the five MNFs (MNF1-MNF5) used in our experiment. The microscope pictures of MNFs with different diameters are shown in Fig. 1.

Table 1. Parameters of the MNFs used in experiment

	MNF1	MNF2	MNF3	MNF4	MNF5
Diameter/μm	$\sim$1	$\sim$2	$\sim$3	$\sim$1	$\sim$1.5
Length/cm	$\sim$9	$\sim$9	$\sim$9	$\sim$15	$\sim$15
Heating flame	butane	butane	butane	hydrogen	hydrogen

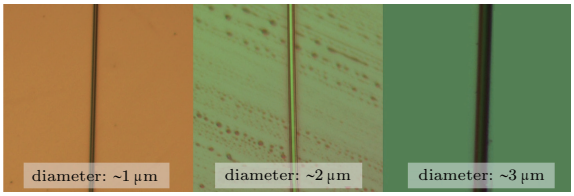


Fig. 1. (color online) Microscope pictures of MNFs with diameters ranging from $\sim$1 μm to $\sim$3 μm.

We then measure the transmission efficiency of each MNF. The results shown in Fig. 2 are obtained by recording the output and input powers at the two SMF ends of each sealed MNF (see Fig. 3) and calculating the ratio between the two powers when the wavelength of the incident light is changed. One sees that comparing with the efficiency at 1310 nm or 1550 nm, the efficiency at 1390 nm is rather low, particularly for the MNFs (MNF4 and MNF5) fabricated by using the hydrogen flame, because the OH^- absorbtion around 1390 nm is enhanced due to the flame heating process.[18]

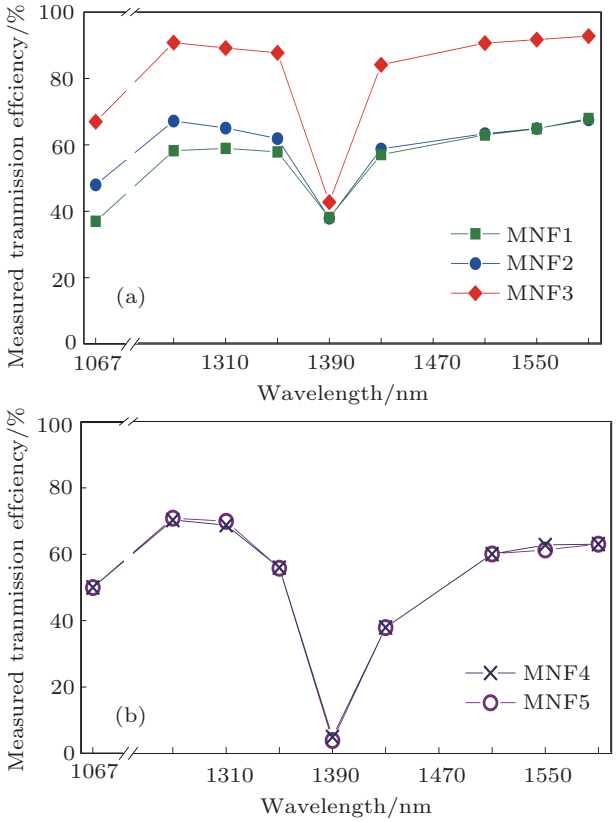


Fig. 2. (color online) Measured transmission efficiencies of the five MNFs (MNF1-MNF5) as a function of wavelength. The sizes of error bars are smaller than those of the data points.

3. Experimental setup for spectral measurement of RS

The experimental setup is shown in Fig. 3. In the experiment, we launch a pulsed pump centering at 1067 nm into each MNF respectively, and measure the intensity spectra of the Stokes photons generated in each MNF within a wavelength range of 1260 nm–1620 nm (corresponding to a Raman shift range of 1435 cm^{-1}–3200 cm^{-1}). For the five MNFs, the phase matching condition of spontaneous four wave mixing (SFWM) is not satisfied in the range of 1260 nm–1620 nm,[17] therefore, all the Stokes photons recorded in this range are originated from spontaneous RS.

The pump laser employed in the experiment is a home-made mode-locked fiber laser based on Yb-doped photonic crystal fiber.[19] The repetition rate and pulse duration of the pulse trains are 62.56 MHz and 250 fs, respectively, while the central wavelength, full width at half maximum (FWHM), and

the power of the laser output are 1041 nm, 8 nm, and 1 W, respectively. In order to obtain the specified pump, we first send the laser output into a 0.4-m long highly nonlinear fiber (HNF) to expand the spectrum to 100 nm via the self phase modulation effect, and then carve out the desired pump by using transmission grating G. By doing so, we obtain the pulsed pump, with the central wavelength, FWHM and average power of 1067 nm, 1.3 nm, and 2 mW, respectively.

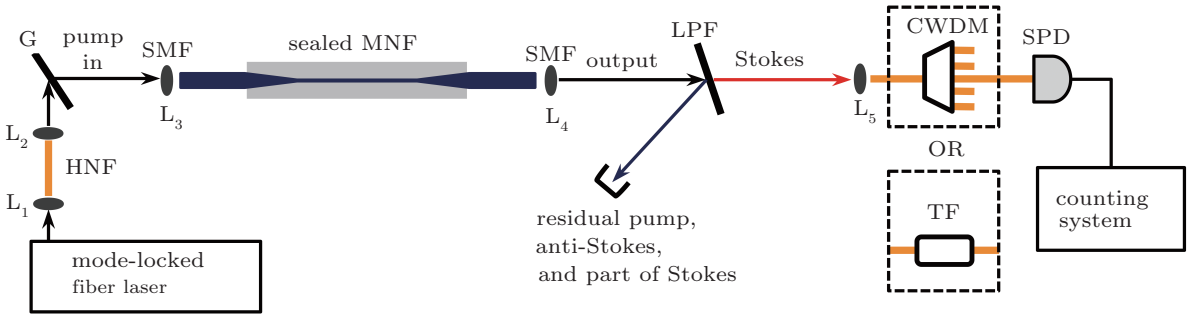


Fig. 3. (color online) Experimental setup. HNF: highly nonlinear fiber; SMF: single mode fiber; G: grating; MNF: micro/nano-fiber; LPF: long pass filter; CWDM: coarse wavelength division multiplexer; TF: tunable filter; SPD: single photon detector; L: lens.

Since the probability of the Raman photons being scattered by a typical 1-ps-duration pump pulse that contains approximately 1.7×10^8 photons is about 0.01 photon per pulse, to reliably detect the Stokes photons produced by RS in MNFs, one must effectively prevent the pump photons from reaching the detector. We achieve this by first passing the output of the MNF through an interferometric long pass filter (LPF, cut-off wavelength 1200 nm). Stokes photons with wavelength longer than 1200 nm can pass through the LPF, while photons with wavelength shorter than 1200 nm, including the residual pump, anti-Stokes, and part of Stokes photons, are reflected.

We then let the output of LPF propagate through a coarse wavelength division multiplexer (CWDM) or a tunable filter (TF). The CWDM is used to realize a coarse measurement with a large wavelength range, and the TF is used to realize a fine spectral measurement within a small range. The CWDM has 18 channels, whose central wavelengths range from 1270 nm to 1610 nm. For each channel, the transmission spectrum is super-Gaussian shaped and the corresponding 0.5-dB bandwidth is about 18 nm. While for the TF, the tuning range is 1530 to 1565 nm and the transmission spectrum is Gaussian shaped with an FWHM of about 1 nm.

The Stokes photons are detected by a single photon detector (SPD, model: SPD4, Langyan), which is based on an InGaAs/InP avalanche photon diode. The SPD is working in the gated Geiger mode, and its trigger originates from the repetition signal of the laser. The detection efficiency of the SPD is $\sim$14%.

4. Results and discussions

We first coarsely measure the spectra of the Stokes photons from RS in each MNF. During the measurement, the wavelength of the photons is selected by the CWDM and the photon counting rate of each CWDM channel is recorded. Figure 4 is plotted after correcting the measured raw data by normalizing the counting rate to one channel of CWDM with transmission efficiency of 24% and by subtracting the counting rate contributed by the SMF ends of the corresponding MNF. From the measured spectra of the five MNFs, we find that the peak locations of counting rates of each MNF are the same. The significant peak at the 1290-nm channel is identified to be the fourth-order RS peak of silica. Although the fifth-order RS peak of silica is supposed to appear in the 1390-nm channel, a dip instead of a peak is observed owing to the OH^- absorbtion induced transmission loss. In addition to the RS peaks of silica, for each MNF, there is a unique RS peak locating at the 1550-nm channel, which is not observable in conventional optical fibers or photonic crystal fibers.

In optical fiber, the intensity of Raman scattering is proportional to $g_R P L_{eff}/A_{eff}$, where g_R, determined by the imaginary part of the third-order nonlinear susceptibility, is the Raman-gain coefficient, P is pump power, L_{eff} is the effective length of the fiber, and A_{eff}, proportional to the square of the fiber radius, is the effective area of the fiber.[20] Comparing the spectra in Fig. 4(a), it is clear that the fourth-order RS peak of silica at 1290 nm increases gradually with the decrease of the diameter of MNF due to the accordingly decreased effective area. In contrast, the growing trend of the unique RS peak around 1550 nm is much more significant, especially when the diameter of the MNF is less than 2 μm. For MNF3 with $\sim$3-μm diameter, the unique RS peak is hard to observe, while for MNF1 with $\sim$1-μm diameter, the unique RS peak becomes even higher than the fourth-order RS peaks at 1290 nm. The similar variation trend of the RS peaks can be observed from the spectra of MNFs in Fig. 4(b). Moreover, comparing the RS spectra of MNF1 and MNF4, having the same diameter of $\sim$1 μm and being fabricated by the heating flame of butane and hydrogen, respectively, we find that when the length and transmission efficiency of the two MNFs are considered (see Fig. 2), the height of the fourth-order RS peak per unit length for the two MNFs are about the same, but the height of the

unique RS peak per unit length for MNF4 is obviously lower since the fourth-order RS peak is the highest among all the observed RS peaks of MNF4. Thus, we infer the unique RS peak around 1550 nm is not only influenced by the diameter of the MNF.

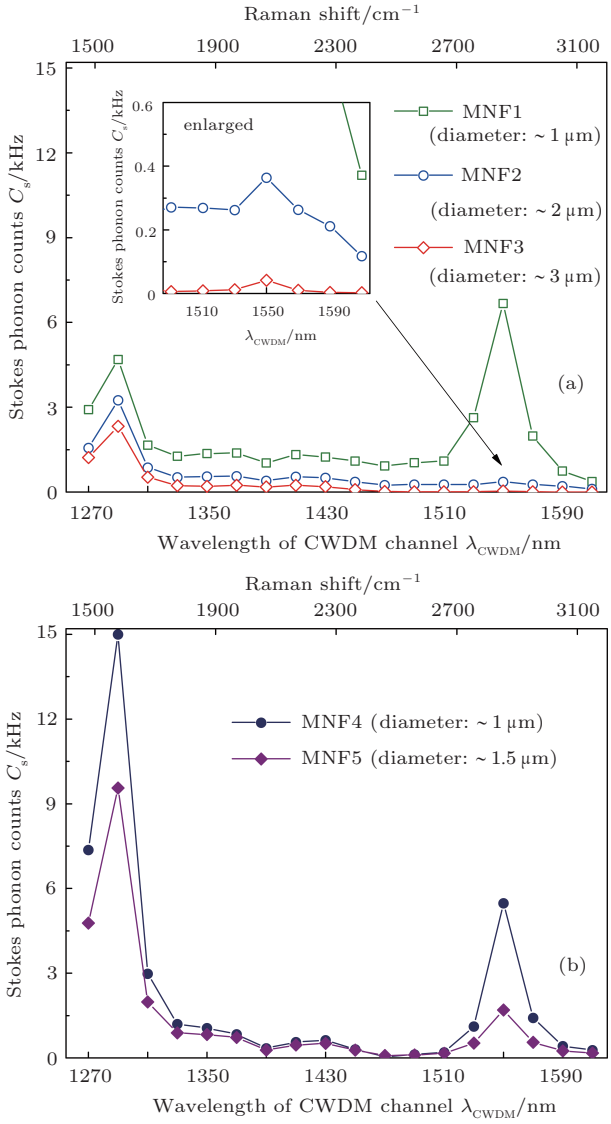


Fig. 4. (color online) Measured Stokes photon counts versus the central wavelength of the CWDM channel for (a) MNFs fabricated by using the flame of butane (MNF1-MNF3) and (b) MNFs fabricated by using the flame of hydrogen (MNF4 and MNF5). The sizes of error bars are smaller than those of the data points. See Table 1 for detailed parameters and features of each MNF.

To further study the unique RS peak, we then precisely scan the spectrum of the Stokes photons around 1550 nm by replacing the CWDM with the TF. Figure 5 is the results obtained in MNF1. From Fig. 5, we find the unique RS peak with an FWHM of about 14 nm is centering at 1546.4 nm, which is ~87.2 THz from the pump (corresponding to a frequency shift of ~2905 cm^{-1}) and is in accordance with the carbon-hydrogen stretching vibration.[21] Therefore, we think that the molecules of carbon compounds are responsible for the appearance of this unique RS peak.

The carbon compounds on the surface of MNFs may be induced by the two steps during fabricating: one is the fuel gas used in the heating process, the other is the very small amount of residual acrylate coating adhering to the fiber after the coating stripping process. Since the density of the carbon compounds originated from the acrylate coating of SMF decreases with the diameter of MNFs, the former step is generally responsible for the difference between the RS spectra of MNF1 and MNF4, since the two MNFs have the same diameter but are fabricated by different heating flames.

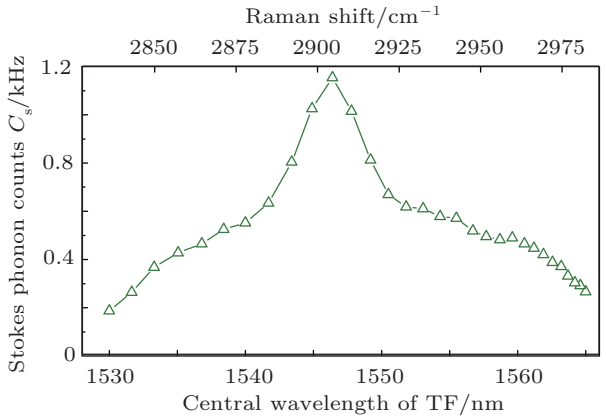


Fig. 5. (color online) Measured Stokes photon counts versus central wavelength of TF. The measurement is done by using MNF1, and the sizes of error bars are smaller than those of the data points.

To further understand why the height of the unique RS peak increases so rapidly with the decrease of diameter, particularly for MNFs with a diameter less than 2 μm, we calculate the field intensity on the surface of MNFs with a diameter of 1 μm, 2 μm, and 3 μm, respectively, by following the method in Ref. [22]. The calculation is conducted under the following assumption: in each MNF, the pump light propagating in the fundamental mode with rotating polarization has a fixed power and wavelength (1067 nm). The calculated results indicate that the field intensity on the surface of the MNF with a diameter of 1 μm (2 μm) is about 67 (4.8) times that with a diameter of 3 μm. Therefore, it is reasonable to deduce that, for the RS originated from the carbon compounds on the surface on MNFs, the higher field intensity at the surface will result in a higher RS peak.

5. Conclusion

In conclusion, we have studied the spectra of spontaneous RS in taper-drawn MNFs having different diameters and being fabricated by different kinds of heating flames. For the measured spectra in the frequency shift range of 1435 cm^{-1}–3200 cm^{-1}, RS peaks originating from silica can be identified and their intensities grow gradually with the diameter decrease due to the accordingly decreased effective area, which are the same as in conventional optical fibers and in photonic crystal fibers. However, a unique RS peak with a frequency shift of ~2905 cm^{-1} (~87.2 THz) is observed as well. Further study shows the unique RS peak, which is in accordance

with the carbon-hydrogen stretching vibration, might originate from the molecules of carbon compounds on the surface of MNFs. Compared with the butane flame, using hydrogen flame in the fabricating process helps to reduce the intensity of RS at $\sim$2905 cm^{-1}, but the trade off is the reduced transmission efficiency around 1390 nm due to the enhanced OH^- absorption. Our investigation indicates the substances on or around the surface of MNF can have a significant contribution to the RS spectra, and the scattering intensity is highly related to the diameter of the MNF. Therefore, for the applications utilizing MNFs, such as the entanglement generation, for which RS is considered as an origin of background noise,[17] and the optical sensing, which relies on measuring the particular peaks in RS spectra,[16] the fabrication of MNFs, including the fiber preparation and the heating technique, should be carefully considered.

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