I. INTRODUCTION

The development of optical frequency combs, since their first reporting two decades ago [1, 2], is still being advanced due to their important applications in metrology, spectroscopy, optical communication, and quantum information. A Kerr frequency comb can be generated by coupling a continuous-wave (CW) laser into a high finesse microresonator and exploiting the third-order nonlinearity of the optical material [3]. Silica whispering gallery resonators (WGRs) have been used extensively for this purpose [4–7]. The advantage of using silica for frequency comb generation is related to its abundance of use in photonics for both passive and active devices due to its ultralow loss and broadband transparency [8]. Silica is used as a cladding material for the integration of optical waveguides with high refractive indices, such as silicon nitride [29, 30] and lithium niobate [31].

Conventionally, most studies on silica-based photonics, especially involving fiber optics, are conducted in the infrared (IR) wavelength band. However, there is significant motivation in pushing silica photonics to the near-IR and visible wavelengths. Developing applications in many areas such as biological imaging, underwater communication and detection, atomic clocks, and quantum technologies all rely on wavelengths in this range. To date, near-visible optically pumped parametric oscillation and Kerr frequency combs have been realized in silica WGRs [9, 10, 11, 12]. Green and blue light generation through a third-harmonic process with near-infrared pumping near 1550 nm has also been observed [13]. To our knowledge, there is, as of yet, no report on the effect of a blue laser pump on nonlinear optical processes in silica WGRs.

In this paper, we report on the generation of a Kerr frequency comb and Raman scattering in a silica micro-
sphere:tapered fiber coupled system pumped in the blue wavelength region. Moreover, we show that the nonlinear processes cannot be maintained for long periods of time, since the ability of the tapered fiber to transmit blue light and the optical quality of the whispering gallery modes (WGMs) appear to degrade rapidly. We have studied the dramatic annihilation of the nonlinear optical processes by monitoring the transmission of light through the tapered fiber for different pump wavelengths and powers, while simultaneously observing the Raman signal in the WGR and the WGM spectrum. The suspected cause of the degradation is photodarkening due to the creation of color centers from two-photon absorption in the blue-band [37] that causes excess absorption loss.

II. BLUE-BAND COMB GENERATION AND STIMULATED RAMAN SCATTERING

The experimental setup is schematically illustrated in Fig. 1. A tunable laser (Toptica Photonics, DL Pro HP 461) with a center wavelength of 462 nm was used as pump light to excite the nonlinear optical processes in the silica microsphere resonator. The light was coupled into a single-mode fiber (SMF, 460HP, Thorlabs), then passed through a polarization controller (PC) and a 90/10 inline beam splitter (BS). The 10% port was connected to a power meter to measure the actual power coupled into the tapered fiber through the 90% port. The tapered fiber, the silica microsphere resonator, and the corresponding coupling system were placed in an enclosed chamber to avoid environmental disturbance. Another 50/50 inline BS was connected to the output of the tapered fiber: one port was connected to an optical spectrum analyzer (OSA, AQ6373B, Yokogawa) and the other port was connected to a photodetector (PD). A digital acquisition card (DAQ) connected to a computer was used to monitor the transmission spectra in real-time.

![Experimental setup for measuring the frequency comb generated using a silica microsphere.](image)

For the measurements, a microsphere resonator with a diameter of \( \sim 100 \mu \text{m} \) was fabricated from 460HP SMF using a CO\(_2\) laser and the Q-factor was determined to be \( 1.8 \times 10^7 \) at low pump power. By tuning the wavelength of the pump laser and adjusting the polarization state of the PC, Kerr frequency combs and cascaded stimulated Raman scattering (SRS) were observed separately, see Fig. 2. Using the maximum hold mode of the OSA and scanning the pump laser over multiple WGMs, a comb with 13 teeth was excited with an input power of \( \sim 30 \text{ mW} \) at a pump wavelength of 462 nm, as shown in Fig. 2(a). The measured free spectral range (FSR) was 0.48 nm, which agrees with the calculated FSR of 0.46 nm. It is worth noting that it is usually challenging to achieve blue-band hyperparametric oscillation and a Kerr comb in silica resonators because the group velocity dispersion (GVD) of silica is in the normal regime; however, the local dispersion around the pump can be easily modified to be anomalous with the assistance of mode coupling between different transverse modes, which usually exist in microsphere resonators [38, 39]. In Fig. 2(b), up to five orders of cascaded SRS can be seen. The spacing between the center of the 1st order SRS band and the pump light is \( \sim 10 \text{ nm} \) (\( \sim 13.8 \text{ THz} \)) and the result is consistent with the Raman shift of bulk silica glass [40]. Despite our observations, the optical nonlinear processes were highly unstable. Neither the optical frequency comb nor the SRS could be maintained for an extended period of time. Both the transmission of the tapered fiber and the quality of the resonator decreased dramatically with blue laser pumping and we explored this phenomenon in further detail.

![Graphs showing frequency comb and Raman scattering spectra.](image)

III. ANNIHILATION CHARACTERISTICS OF NONLINEAR PROCESSES

In order to understand the annihilation processes that led to destruction of the nonlinear features, we first characterized the transmission of light through tapered optical fibers as a function of time for different wavelengths. The lasers used were all fixed wavelength and multimode, except for the 462 nm laser which was single-mode, nar-
row linewidth and tunable. We used a different SMF (980HP, Thorlabs) for these studies, since it has a larger core than the 460 nm fiber used in the previous tests. This ensured that it was easier to couple light over a wider range of wavelengths. To reduce the number of modes in the tapered fiber waist at shorter wavelengths, the diameter was kept at \(\sim 500 \text{ nm}\). The power coupled into the fiber was 7 mW for each wavelength and it was kept constant, with the transmission being recorded over time.

A typical normalized transmission signal as a function of time is shown in Fig. 3(a). It can be seen that, for this particular fiber sample, the transmission decreased at different rates for the different wavelengths. The reduction in the transmission at a wavelength of 980 nm was slow and it only reduced by 0.04\% over the test time (more than 4 hours). At 633 nm, the light decayed rapidly with an exponential time constant of 6 minutes. After 20 minutes, the normalized transmission remained nearly fixed at 80\%. At shorter wavelengths the behavior was noticeably different; the transmission for 520 nm was similar to that for 633 nm for the first hour, but it decreased to 4\% after four hours with an exponential time constant of \(\sim 50\) minutes. For 460 nm light, the transmission dropped rapidly to 62\% in 4 minutes, then oscillated for 2.5 hours, and finally decayed to 1\% after 4 hour with an exponential time constant of 25 minutes. The above results demonstrate that under the same input power, the power drop at short wavelengths was more significant. This phenomenon follows the same trend as photodarkening in untapered Yb-doped fibers [41] and we assume this is the dominant process behind the observed behavior.

![FIG. 3. (a) The normalized transmission of the tapered fiber at a fixed input pump power of 7 mW but different wavelengths. (b) The normalized transmission of the tapered fiber with a fixed wavelength of 460 nm but different input pump powers (the transmittance of an untapered fiber is given for comparison).](image)

The excess optical loss caused by photodarkening in untapered fibers is also related to the intensity of the pump light [42]. Therefore, we investigated the transmission through a tapered fiber at a wavelength of 462 nm over time for different input powers. The tapered fibers were made from 460HP fiber and the diameter was approximately 500 nm in each case. The results are shown in Fig. 3(b) and, as a reference, the transmission through an untapered 460HP fiber is also presented. We see that the transmission of the untapered fiber was reduced by only 0.9\% over 6 hours for an input power of 7 mW. However, for the tapered fibers, their transmission was unstable when the input power exceeded 1.25 mW. As the power increased from 1.25 mW to 7 mW, the power downtrend was more dramatic; at an input power of 7 mW, the normalized transmission dropped close to zero after 2 hours. Although the transmission seems to remain stable for low powers (dozens of \(\mu\)W) for a reasonable period of time and whispering gallery modes can be excited, the powers are not sufficient to reach the threshold needed for the observation of nonlinear effects.

![FIG. 4. Images of the waist of the tapered fiber over time for an input power of 7 mW.](image)

A CCD camera was used to record the scattered light intensity evolution from the tapered fiber (for the 7 mW case) and images at 20 minute time intervals are shown in Fig. 4. Here, the exposure time was set to the lowest value at the beginning. As the loss of 462 nm light in the tapered fiber increased with time, the brightness of the recorded image also gradually increased. This observation helps us to understand the loss in transmission of the tapered fiber caused by photodarkening. It is worth emphasizing that the observed scattered light was not caused by dust particles, since the tapered fiber was kept in a clean chamber and a similar phenomenon was not observed when pumping at 980 nm. After irradiation, the tapered fiber was examined with a scanning electron microscope (SEM) and no trace of scatterers or damage was found on its surface.

Similar to the degradation of the tapered fiber, the blue comb and Raman scattering could only be sustained for a short period of time. To confirm this, we recorded the optical spectrum for SRS and the transmission spectrum of the WGMs simultaneously, see Fig. 5. A tapered fiber was used to couple 462 nm laser into the microsphere to excite the WGMs and generate the SRS. The tapered fiber and micropshere were all made from 460HP fiber. The laser frequency was scanned over 13.3 GHz at a rate of 10 Hz and the pump power was fixed at 12 mW. A DAQ was connected to a computer to record the real-time resonance data. Since the laser frequency was scanning, the Raman spectra were continuously acquired in five second intervals using the maximum hold mode of the optical spectrum analyzer to ensure most of the Raman peaks were captured.
FIG. 5. (a) Recorded stimulated Raman scattering spectra with max hold mode for 5 seconds at different times. (b) Corresponding transmission spectra through the tapered fiber. The red line represents the piezo voltage of the pump, which increases with the laser’s frequency.

As shown in Fig. 5(a), two orders of SRS were visible during the first 5 s; however, the second order SRS disappeared after 480 s and no SRS could be observed after 600 s. The evolution of the SRS spectra can be understood by monitoring the fiber transmission spectra, which are shown in Fig. 5(b). Note that the thermal broadening of the resonances was weakened and no obvious thermal broadening could be observed after 600 s. This phenomenon should not be mainly attributed to the degradation of the tapered fiber itself. We deduce that it is due to a photodarkening-induced reduction of the microcavity Q-factor that resulted in the annihilation of the SRS signal. Indeed, the transmission of the tapered fiber deceased to 40% within 600 s, although we estimate that there was still 1.33 mW at the tapered fiber waist and this should be more than enough power to generate SRS in newly made microspheres. We verified this by replacing the tapered fiber with a newly fabricated tapered fiber (with the same dimensions) and then pumping the degraded microsphere with the same power (12 mW). We found that we could no longer achieve SRS.

IV. DISCUSSION AND CONCLUSIONS

A blue-band Kerr frequency comb with 13 teeth and 5th order cascaded SRS were successfully excited in a silica microsphere resonator for the first time, a significant result in itself. The nonlinear processes and the supporting WGMs proved to be highly unstable. As a result, neither the optical frequency comb nor the Raman signal could be maintained at reasonable pump powers. These results imply that there is some limiting factor, possibly photodarkening, for the advancement of nonlinear photonics at short wavelengths in silica-based devices.

There are controversies in the literature about the specific generation mechanism of color centers that lead to photodarkening, but it can be generally viewed as being due to oxygen deficiency centers (ODCs) [43], charge transfer band (CT band) transitions [44], or the influence of Tm$^{3+}$ impurities [45] in doped fiber. One common color center in pure silica is a partially bound oxygen atom with one free electron, that is the non-bridging oxygen hole center (NBOHC) [46]. This defect often forms as a result of optically induced breaking of a bond in a stressed multiple member Si-O ring [47]. It is also noted that dangling bonds on the surface of the silica play a role in some laser-induced color centers.

In contrast to the available studies on photodarkening in commercial optical fibers when using high powers (typically a few Watts) [46, 48, 49], the threshold for photodarkening-like effects in the microresonators and nanofibers reported herein is very low (a few mW). The most likely explanation is the increased probability of two-photon absorption due to the very high optical intensities (MW/cm$^2$ - GW/cm$^2$) in these micro- and nanodevices. Such high intensities arise from the small mode volume and high optical quality factors. Our observations indicate that photodarkening is a possible obstacle for directly extending the micro/nano silica photonics from the IR into the visible band, even for ultralow-threshold nonlinear optics applications.

As we have shown, light intensity clearly plays a role in the signal annihilation. The incurred losses in the tapered fiber take hours to manifest, whereas the microsphere WGMs are degraded within minutes for the same optical power. In fact, at higher powers, the WGMs can be eliminated in a matter of seconds. It is not clear at this point how this mechanism can be overcome and it warrants further study to determine the process in detail and to find possible solutions.

The results presented herein also illustrate that the tapered optical fiber can be used as a tool to observe, in situ, the dynamics of color center formation. The one dimensional, ultrathin fiber provides us with an easy technique for imaging the process and could facilitate the study of color centers in different materials using sub mW pump powers.

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