Lithium-niobate microcavities have not only the ability to resonantly enhance light–matter interaction but also excellent nonlinear optical properties, thereby providing an important platform for nonlinear optical investigations. In this paper, we report the observation of multi-peak spectra in the near infrared range in lithium-niobate microcavities on a chip under the pump of a 1550 nm continuous laser. Such a multi-peak spectrum was attributed to the sum-frequency of the pump laser and its background. The conversion efficiencies of the sum-frequency processes are of the order of 61.5% W⁻¹. The influences of the phenomenon on nonlinear processes were further discussed.

Keywords: lithium niobate; microcavities; nonlinear optics.

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1. Introduction

Whispering gallery mode (WGM) microcavities have attracted much attention in recent years for their broad applications ranging from ultra-sensitive sensing[1], enhanced nonlinear optical effects[2,3] to tunable quantum light sources[4]. These applications benefit from the ability of WGM microcavities to confine light for a long time, which is indicated by quality (Q) factors, in a small mode volume (V) via total internal reflection. Such light trapping mechanisms make WGM microcavities have high Q in very broad spectrum ranges, which is only limited by the transparency window of the material of which the microcavities are made.

It is known that putting a nonlinear optical medium into a high-Q WGM microcavity can improve the efficiencies of the nonlinear optical effects and release the requirement of high intensity pump. Therefore, a pulse laser with high peak power can be replaced by a continuous pump with power even lower than milliwatts. Compared with bulk optical cavities containing nonlinear optical materials, WGM microcavities usually have not only a much smaller size but also a broader resonance window in which high Q can be achieved[5,6]. Therefore, WGM microcavities made of nonlinear optical materials become an excellent testbed for nonlinear optical investigations. Various nonlinear optical effects[7,8] and their cascaded effects[9,10] were observed in high-Q WGM microcavities. Based on cascaded four-wave mixing[11] processes, optical frequency combs[12] that have great important applications in precision measurement are intensively investigated in high-Q WGM microcavities.

Lithium niobate (LN) is a noncentrosymmetrical material with outstanding nonlinear optical properties ($d_{33}=27\ \text{pm/V}$[13]). LN thin film on insulator has aroused wide interest and broad application prospects[14–16]. WGM microcavities made of LN take the advantages of LN and WGM cavities and therefore show great potential in fundamental studies and practical applications of second nonlinear optical effects such as second harmonic generation (SHG)[2,17–22], cascaded harmonic generation[23], sum-frequency generation (SFG)[24,25], and optical parametric oscillation[26–28]. Optical devices including entangled photon pairs[29] and optical frequency combs[30] were recently reported in LN WGM microcavities.

However, all of the nonlinear optical effects reported in LN WGM microcavities are associated with only laser sources. In this paper, we report the observation of a series of nonlinear optical signals, besides second harmonic signals, in an on-chip LN WGM microcavity. These nonlinear optical signals were attributed to the SFGs between the pump laser and its background. This work elucidates the origin of unknown nonlinear optical signals always observed in high-Q WGM resonators that cannot be attributed to high-order harmonic generation.
For a UV lithography, argon ion etching, and hydrogen fluoride etching. by NANOLN by using the microfabrication technique including perpendicular to the plane in which the microdisk lies. to the rotational axis of the microdisk cavity, in other words, less than 200 kHz and a tuning range covering 1520–1550 nm pump is generated by a tunable laser with a linewidth and extract nonlinear optical signals from the same cavity. The was used to couple the 1550 nm pump into the LN microdisk frequency signal in the LN WGM microcavities. A tapered fiber the WGMs related to the sum-frequency processes. the laser controller. Therefore, we can approximately assign the wavelengths measured by the spectrometer and shown by oscilloscope. An optical spectrum analyzer (not shown in Fig. 1) working from 600 nm to 1700 nm that covers the output wavelength of the pump laser and to trigger the oscilloscope. The pump light was monitored by using a photodetector and to an oscilloscope. The tapered fiber that is used to couple the pump collects the nonlinear optical signals as well. The nonlinear optical signals are detected by a spectrometer.

2. Samples and Experimental Setup
LN WGM microcavities with a size of 200 nm in thickness and 40 μm in radius were employed to conduct nonlinear optical experiments. The Q factors of these WGM microcavities are on the order of 10^5, which were fabricated from Z-cut LN film produced by NANOLN by using the microfabrication technique including UV lithography, argon ion etching, and hydrogen fluoride etching. For a Z-cut LN WGM microcavity, the optical axis of LN is parallel to the rotational axis of the microdisk cavity, in other words, perpendicular to the plane in which the microdisk lies.

Figure 1 indicates the experimental setup to observe the sum-frequency signal in the LN WGM microcavities. A tapered fiber was used to couple the 1550 nm pump into the LN microdisk and extract nonlinear optical signals from the same cavity. The 1550 nm pump is generated by a tunable laser with a linewidth less than 200 kHz and a tuning range covering 1520–1570 nm. The nonlinear optical signals were detected by a spectrometer allowing for the detection of weak light with power down to several picowatts (pW). The laser wavelength can be finely tuned by applying a triangular wave voltage on the piezo mirror of the external reference cavity of the laser. The transmission of the pump light was monitored by using a photodetector and an oscilloscope. An optical spectrum analyzer (not shown in Fig. 1) working from 600 nm to 1700 nm that covers the output wavelength of the pump laser was used to measure the transmission spectrum of the laser and its background and to calibrate the wavelengths measured by the spectrometer and shown by the laser controller. Therefore, we can approximately assign the WGMs related to the sum-frequency processes.

3. Experimental Results and Discussions
It is known that second-order nonlinear optical signals can be detected in WGM microcavities made from material lacking of central symmetry with pump down to less than milliwatts[20] when energy, angular momentum conversations, and multiple resonance are fulfilled simultaneously. When one pump was launched into an LN WGM, several kinds of nonlinear optical processes were reported: (1) optical parametric oscillation producing two photons with lower energy[27], (2) SHG giving rise to one photon with energy twice that of the pump[31], (3) sum-frequency between the pump and its second harmonic signal generating a third harmonic signal and other kinds of signals with even higher photon energy[32] and (4) Raman signal with wavelength longer than the pump[33].

We observed nonlinear optical signals with about 10 peaks, as shown in Fig. 2, when only one pump laser at 1521.36 nm was launched into an LN WGM microcavity. Peak 1, which is highlighted in red, has a wavelength of 760.68 nm. It is considered the second harmonic signal of the pump according to the principle of energy conservation, which indicated that the wavelength of the second harmonic signal was half of the pump wavelength. Besides the second harmonic signal, a series of signals ranging from 770 nm to 780 nm, marked in blue in Fig. 2, were unexpectedly observed as well. The nonlinear optical signals in blue have wavelengths between the pump at 1521.36 nm and its second harmonic signal of 760.68 nm, which is different from the aforementioned four kinds of nonlinear optical signals in the wavelength. Such nonlinear signals have not been discussed in detail in LN WGM cavities.

To find the origin of the unexpected nonlinear optical signals, we first suppose they are the sum-frequency signals of the pump and unknown light sources. In this situation, the wavelengths of the undiscovered light sources can be calculated according to energy conversation for sum-frequency processes, i.e., \( \lambda_{\text{SFG}} \). The calculated wavelengths are shown in the third column of Table 1. On the other hand, the transmission spectrum of the pump laser was measured by an optical spectrum analyzer, which is shown in Fig. 3(a). Figure 3(b) is the enlarged version showing the detailed

![Figure 3](image_url)
information of the yellow-background highlighted part of Fig. 3(a). We found that the wavelengths $\lambda_{\text{Mean}}$ of the blue peaks in Fig. 3(b) coincide with $\lambda_{\text{Cal}}$ with deviations from 0 nm to 0.220 nm. Accordingly, we attribute the unexpected nonlinear optical signals to the sum-frequency of the pump laser and its background.

Initially, we suspected these nonlinear optical signals may due to the sum-frequency of the pump laser and its third-order nonlinear optical outputs, such as stimulated Raman or four-wave mixing signal. We measured the transmission spectrum around the pump, and neither the Raman nor four-wave mixing signal was observed in the 1550 nm band, especially at the wavelengths that may generate observed nonlinear signals via the sum-frequency process under a 1521.36 nm pump. The absence of the Raman and four-wave mixing signals further verify that the nonlinear optical signals extending from 770 nm to 780 nm are due to the sum-frequency of the laser and its background that was coupled into the LN WGM microcavity. The high power of the background from 1560 nm to 1600 nm makes it easier to detect optical signals to the sum-frequency of the pump laser and its background.

The normalized conversion efficiencies of the strongest sum-frequency signal (Peak 7 in Fig. 2) and the associated second harmonic signal were derived by measuring the slope of the curve of the conversion efficiency ($P_{\text{signal}}/P_{\text{pump}}$) with respect to the pump power; see Fig. 4. The measured conversion efficiencies of the sum-frequency and SHGs are $6.15 \times 10^{-4}$ mW$^{-1}$ and $4.7 \times 10^{-4}$ mW$^{-1}$, respectively. The efficiency of the SHG is lower than the highest efficiency of second-order nonlinear optical processes related to birefringent phase matching conditions in LN WGM microcavities, which is the order of 0.015 mW$^{-1}$ [27]. The main reason can be described as follows.

(1) According to the reference, phase matching in Z-cut single crystal LN microdisk resonators with less than 200 nm thickness and less than 30 μm radius can be achieved in low-order WGMs [34]. In our experiment, the thickness of the disk is not thin enough to achieve frequency conversion between fundamental modes; therefore, we believe high-order modes get involved in the SHG process. The involvement of high-order WGMs in nonlinear optical processes is one of the main reasons that reduce the nonlinear conversion efficiency due to the poor spatial overlap between the WGMs acting in the nonlinear processes. (2) The absence of a broad-band coupling of the tapered fiber with the LN WGM microcavities, which means the lack of efficient coupling in both the 1550 nm and 775 nm bands, results in low nonlinear conversion efficiency as well due to the decreased collecting efficiency of the second-order signals.

It is noted that the SFG process related to the phenomenon itself has meaningful application and has been reported in both millimeter-sized and micrometer-sized LN WGM microcavities [27,35]. High-efficiency SFG can be utilized in a series of applications, such as single photon detection and bioimaging, by converting relatively weak signals in the infrared band to the visible band and taking advantage of the high efficiency and low cost of photodetectors in the visible band. On the other hand, the phenomenon observed in our experiment may negatively influence other nonlinear optical effects. (1) The multi-peak signals cause pump energy consumption, reducing other nonlinear optical processes. (2) The sum-frequency between the pump laser and the background leads to nonlinear noise. The higher the pump power is, the more the significant noise is. We could take some methods to avoid these negative influences on nonlinear frequency conversion, such as employing a laser with a high side-mode suppression ratio or applying a band-pass filter to the pump laser.
4. Conclusions

To conclude, we observed a series of multi-peak signals in an LN WGM microcavity under the pump of only a continuous laser in the 1550 nm band. These signals were ascribed to the wave mixing of the pump laser and its background, which is usually considered natural light. The influences of SFG between the laser and the background on the nonlinear process were analyzed. The method to suppress the phenomenon was proposed. This work provides a reasonable interpretation for the unknown nonlinear optical signal in WGM microcavities and the suggestions to use and avoid similar phenomena.

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