Optical sum-frequency generation in a whispering-gallery-mode resonator

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Abstract
We demonstrate sum-frequency generation between a telecom wavelength and the Rb D2 line, achieved through natural phase matching in a nonlinear whispering gallery mode resonator. Due to the strong optical field confinement and ultra high $Q$ of the cavity, the process saturates already at sub-mW pump peak power, at least two orders of magnitude lower than in existing waveguide-based devices. The experimental data are in agreement with the nonlinear dynamics and phase matching theory based on spherical geometry. Our experimental and theoretical results point toward a new platform for manipulating the color and quantum states of light waves for applications such as atomic memory based quantum networking and logic operations with optical signals.

Keywords: nonlinear optics, sum-frequency generation, whispering-gallery-mode resonators

1. Introduction

Strong optical nonlinearities have been the foundation of many applications in classical and quantum optics. Recently, the burgeoning field of high-$Q$ nonlinear micro- and nano-cavities [1] has emerged as a new chip-scalable platform for photonic information processing, which
requires very low (below 1 fJ) energies [2–4] and can be nearly lossless. In addition, by utilizing the quantum Zeno effect, interaction-free operations can be implemented which eliminate the otherwise inevitable energy dissipation and background scattering processes. A pursuit of low light level optical interactions, hence, can simultaneously address fundamental and practical problems faced by both classical and quantum information processing [5]. In fact, by analyzing a $\chi^{(2)}$-nonlinear lithium niobate microresonator, it has been predicted that strong, noise-free interaction can be realized among single photons, thereby uncovering pathways to unprecedented applications such as single-photon optical transistors and deterministic quantum logic gates [6]. Such a realization has an inherent advantage over resonant optical interactions with matter systems due to its compact experimental setup and room-temperature operation.

All of these proposals place exacting criteria on optical resonators, requiring a high quality factor across a broad optical range, small mode volume, and strong overlap between the interacting modes. While a small mode volume is available in photonic-crystal microcavities, multiply-resonant, high-$Q$ cavities are difficult to fabricate [7]. In this paper we present, for the first time, naturally phase-matched sum-frequency generation (SFG) in a triply-resonant high-$Q$ lithium niobate whispering gallery mode (WGM) microresonator with strongly non-degenerate frequencies, which could serve as a significant step towards achieving these goals simultaneously.

Optical WGM resonators with $\chi^{(2)}$ nonlinearity thus far have been successfully used for either single frequency multiplication (e.g. doubling [8–10], tripling [11–13], and quadrupling [14]), or parametric down conversion [15–18]. In contrast, in this experiment we demonstrate SFG between a 1560 nm pump and a 780 nm signal reaching saturation already at sub-mW pump power levels, which is at least two orders of magnitude lower than in best known to us waveguide-based devices [19]. Such efficient cross-band coupling opens avenues to several narrow-band frequency conversion experiments that have been hitherto challenging. Indeed, SFG can be employed for efficient room-temperature detection of far-infrared, and even sub-THz, photons [19, 20]. Since SFG does not disturb the quantum state [21], it also can lead to efficient manipulation of the color and shape of single-photon signals [22] for interfacing optical flying qubits with narrowband atomic quantum memories [23]. Furthermore, the discrete and sharp nature of the resonance lines in such devices can greatly suppress incoupling of Raman noise, and potentially lead to new optical tools for mode discrimination and reshaping of narrowband quantum signals [24]. Our experiment is an important first step towards all of these applications in both the classical and quantum domains.

2. Experiment

2.1. Experimental set-up and measurement technique

We observed SFG in a MgO-doped lithium niobate z-cut microdisk ($R \sim 0.6$ mm) evanescently coupled to a diamond prism. Diamond-polishing was used to obtain absorption-limited $Q \gtrsim 2 \times 10^7$ and $Q \gtrsim 4 \times 10^7$ for the signal and pump waves, respectively. A 780 nm narrow (below 300 kHz) linewidth tunable diode laser was used as the signal, and a distributed feedback laser provided a 1560 nm pump. The pump linewidth of approximately 2 MHz was not insignificant compared to the WGM linewidth (approximately 5 MHz) which should limit the coupling efficiency to some 70%. However, as we will see in the following, the coupling
efficiency was more strongly affected by imperfect spatial mode matching. Two input waves are ordinarily (in the resonator plane) polarized to achieve the type-I phase-matching with the upconverted wave at 520 nm that has the extraordinarily (perpendicular to the resonator plane) polarization. The two input waves were combined on a dichroic mirror and focused onto the prism-resonator interface by an objective lens. A lateral offset of the beams before the lens allowed for optimizing of the pump and probe in-coupling angles individually. A similar lens was used to collect the output light, with a dichroic mirror separating the pump and the signal while a polarization beam splitter separating the sum-frequency wave. All three optical powers were measured by photodetectors, whose signals were fed into a data acquisition unit.

As the laser frequencies were continually swept at 50 Hz across several linewidths, the signal and pump WGMs were tracked by a computer program continuously adjusting the lasers central wavelengths to follow the chosen WGMs. The program ensured that the pump and signal WGMs were in the center of the sweeps and that they were pumped simultaneously. We prefer this technique to a standard laser locking because it proved more robust against the mode crossing, frequently occurring during the thermal tuning, and also allowed us to record the SFG signal as a function of detuning in every sweep. The peak sum-frequency power recorded during such a fast sweep will be referred to as the SFG signal throughout the rest of the paper. The resonator was temperature-controlled to allow for thermal tuning of the SFG phase matching. In addition, it was coated with silver paste and wired to a voltage source to perform this tuning electro-optically (figure 1).

A typical phase matching temperature/voltage tuning curve is shown in figure 2. We see that the SFG is achieved within a rather narrow temperature range of approximately 7 mK. This width is determined by the relative shifts of the pump, signal and sum-frequency WGMs from the energy conservation condition \( \omega_p + \omega_s = \omega_f \) in response to varying temperature or electric field. To measure the phase matching width in temperature units we first calibrated the temperature variation against the electro-optical bias voltage variation by a compensation technique, and then recorded the SFG peak signal versus the bias voltage sweep.

Having the phase matching achieved, we measured the sum-frequency output power for various input pump and signal powers and for various WGMs. Since the temperature
stabilization of the resonator at the required level was deemed difficult and time consuming, we carried out these measurements in transient by slowly varying the electro-optic bias voltage to record the peak SFG efficiencies.

The signal and pump waves were critically coupled and over-coupled, respectively. The longer wavelength pump was coupled stronger than the shorter wavelength signal due to the nature of the evanescent coupling. Due to the spatial-mode mismatch between the input Gaussian beam and the WGM profile, in this measurement we achieved a critically-coupled contrast of 48%. We took this into account by only utilizing the in-coupled powers for our theoretical analysis.

2.2. SFG channels

Even when the energy conservation condition $\omega_p + \omega_s = \omega_f$ is fulfilled, different combinations of WGMs (which we will call channels in the following) may lead to different SFG efficiency. For some combinations this process may even be completely prohibited. In the theoretical section below we will discuss the dependence of the SFG channels efficiency on the orbital ($L, m$) and radial ($q$) numbers of the three interacting WGMs and show that for the most efficient SFG we need to identify the equatorial (i.e. such that $L = m$) fundamental (i.e. $q = 1$) signal and pump modes. While the equatorial modes can be usually identified as those with the best coupling, identifying a WGM’s $q$ may present a considerable experimental challenge. Fortunately, the WGM’s free spectral range (FSR) depends on its $q$ much stronger than on $L$ and $m$, because $q$ affects the effective length of the resonator. Therefore a WGM’s $q$ can be inferred from the FSR measurements. We carried out the FSR measurements for the pump laser by frequency-modulation technique [25]. The results of such measurements carried out with 11 best-coupled modes within one FSR are shown in figure 3. The theoretical FSR values shown in figure 3 were derived from the WGM dispersion equation [26]. We fit the theory value for $q = 1$.
to the smallest measured FSR of 32.362 GHz by varying the resonator radius from the initially measured $0.65 \pm 0.01$ mm to 0.6505 mm. No fitting was done to match other $q$ values.

The theoretical FSR value for $q = 1$ WGMs at the signal wavelength is 31.049 GHz. We found a high-contrast mode with a very close FSR value of $31.04 \pm 0.004$ GHz. The majority of the signal modes had smaller FSRs and did not follow the trend of figure 3. This contradicts the assumption that the selected probe mode has $q = 1$ and is also inconsistent with the resonator radius measurement. We attribute this discrepancy to localized photorefractive damage induced by the 780 nm light [27, 28]; however, to further validate the FSR measurement technique we have performed additional measurements that are discussed in appendix A. Coupling the pump and signal lasers to these WGMs we slowly varied the resonator temperature while monitoring the SFG signal, and acquired the data for figure 4.

Figure 3. Theoretical FSR values for $q = 1...8$ WGMs at 1560 nm (vertical lines), and the measurement results.

Figure 4. The out-coupled SF emission is measured and normalized to the input signal powers. Symbols represent the experimentally measured data, and the solid lines are theoretical fits. Note that the theory predictions for 2.5 $\mu$W and 5 $\mu$W are identical because the nonlinear loss is negligible at such powers.
2.3. Results

Using the selected SFG channel, we observed efficient SFG with a maximum in-coupled pump power of only 1.22 mW. In figure 4 we plot the out-coupled SFG efficiency with respect to the input signal versus the in-coupled pump power. Here each data point represents the average of three consecutive measurements.

As we varied the signal powers, saturation of the peak conversion efficiency was observed in all cases with sub-mW incoupled powers, instead of a cyclic behavior that is observed in the traveling-wave configuration [29]. Similar saturation has been observed in frequency-doubling WGM experiments [8, 9]. At higher pump powers, an additional nonlinear loss for the signal wave is created due to its upconversion, leading to the reduction of its internal Q-factor and consequently, of the coupling contrast. As a result, a smaller portion of the signal wave enters the resonator and the SFG efficiency is reduced. This behavior is a manifestation of the ‘coherent’ quantum Zeno effect for the signal wave, where the ‘potential’ for the upconversion decouples the signal field from the cavity [30].

3. Theory

3.1. Wave mixing in WGM resonators

A theoretical description of the SFG in WGMRs is warranted because manifestly different behavior is observed at high pump powers compared to a traditional traveling-wave geometry. Neglecting Rayleigh backscattering and assuming linearly polarized fields, we find the following scalar equations of motion in the cavity:

\[ \frac{\partial c_p}{\partial t} = -\kappa_p c_p + i \sqrt{\frac{\omega_p}{Q_p}} a_p + i \Omega c_s^* c_f \]  

(1)

\[ \frac{\partial c_s}{\partial t} = -\kappa_s c_s + i \sqrt{\frac{\omega_s}{Q_s}} a_s + i \Omega c_p c_p^* \]  

(2)

\[ \frac{\partial c_f}{\partial t} = -\kappa_f c_f + i \Omega^* c_s c_p \]  

(3)

where \( Q_p \) and \( Q_s \) denote the coupling and intrinsic Q-factors, \( \kappa_\mu = \left( \frac{\alpha_\mu}{2Q_\mu} + \frac{\alpha_\mu^*}{2Q_\mu^*} - i \Delta_\mu \right) \) for \( \mu = s, p, f \) indicating respectively signal, pump and the sum-frequency, \( \Delta_\mu \) are detunings of the optical frequencies from their respective WGM resonances, and \( \Omega = \frac{\omega_\mu}{\hbar} d_31 \int dV E_j^* E_p^* E_s^* \) is the internal conversion efficiency of the SFG process. The input field operators, \( a_\mu \), are related to the output fields by \( b_\mu = \sqrt{\mathbb{T}} a_\mu + i \frac{\alpha_\mu}{\sqrt{Q_\mu}} c_\mu \). Using quasi-static analysis we solve equations (1)–(3) for the out-coupled sum-frequency field, \( |b_f|^2 = \left| i \frac{\alpha_f}{\sqrt{Q_f}} c_s c_p \right|^2 \).

Before we provide the solution, a few salient points are to be noted. In this formulation, \( \Omega \) is the internal conversion efficiency of the SFG process, whereas only the SF out-coupled power is measured experimentally. Since \( Q_p^\prime \) are determined by the distance from the prism to the resonator \( d \), we need only to fit to \( \Omega \) and the intrinsic Q-factor for the SF. Moreover, these
equations are difficult to solve in general for \( c_p \), \( c_s \), and \( c_f \) analytically. To guide us, however, we make the undepleted pump approximation and then use numerical methods to acquire the generic solution. This approximation yields:

\[
c_p = \frac{i \sqrt{\frac{n_p}{\omega_p}} a_p}{\kappa_p}, \quad c_s = \frac{i \sqrt{\frac{\alpha_s}{\omega_s}} a_s}{\kappa_s + \sqrt{\frac{\alpha_p}{\kappa_s} \frac{a_p}{a_s}}},
\]

\[
|b_j|^2 = \frac{\alpha_j \alpha_j \alpha_j}{Q^2_p Q^2_s Q^2_f} \left| \frac{\Omega}{\kappa_s + \sqrt{\frac{\alpha_p}{\kappa_s} \frac{a_p}{a_s}}} \right|^2 |a_p|^2 |a_s|^2. \tag{5}
\]

Note that the expressions for \( c_p \) and \( c_s \) are asymmetric due to the nature of the undepleted pump approximation. These solutions indicate that we do not observe an oscillatory behavior in the frequency-conversion dynamics. For low pump and signal energies, the SFG output behaves linearly, \( \propto b_j \) (see figure 4), whereas at higher energies, the dynamics are different, i.e. the upconversion and subsequent down conversion processes are asymmetric in this geometry [31].

Using the in-coupled pump and signal powers, and measured \( Q_p^\mu, Q_s^\mu \) for \( \mu = s, p \), we can provide a theoretical fit for the measured data in figure 4 with the fitting parameters \( \Omega \) and \( Q_f^\mu \). Fitting our efficiency measurements at 2.5 \( \mu \)W signal inputs (where the undepleted pump approximation is still valid) with equation (5), we can estimate \( \Omega \), which does not vary as the same WGM triplet is studied throughout the experiment. Throughout this procedure, we assumed that the SFG peak occurred when the three waves were exactly on resonance, i.e. \( \Delta = \Delta_p = \Delta_f = 0 \).

We then solved for equations (1)–(3) numerically, which provided the efficiency curves for data where the undepleted pump approximation is invalid. The total \( Q \)-factors for the pump and signal waves were not constant through the experiment, which we account for by using \( Q_f^\mu \) as a fitting parameter in figure 4. We attribute this variation in part to the unaccounted photorefractive effects in the resonator, caused by the SF. These effects were most evident for the highest signal power used, \( P_s = 60 \mu \)W. Indeed, this particular data set in figure 4 also presents the largest data scatter and the worst agreement with theory.

In spite of these theoretically unaccounted background processes, by using just the \( \Omega \)-parameter, which determines the shape of the efficiency curves, we are able to accurately model the signal and pump transmission spectra as well as the SF emission spectrum. Moreover, we were able to calculate the fundamental channels modes overlap to give \( \Omega_{\text{theor}} = 253 \) kHz, whereas the empirical value is \( \Omega_{\text{expt}} = 5 \) kHz. In calculating this latter value, we used \( Q_f^\mu = 3.25 \times 10^7 \), the intrinsic sum-frequency \( Q \)-factor and that it was strongly undercoupled to the resonator, \( \frac{Q_p}{Q_f} = 164 \), as shown in appendix B.
3.2. Phase-matching in WGM resonators

Not every pair of signal and pump WGMs can generate sum-frequency. SFG in a triple-resonant system requires the phase matching between these modes, which can be viewed as conservation of the integrals of motion determined by the system’s symmetry.

Usually WGMs have nearly perfect spherical symmetry, so their eigenfunctions inside the resonator can be well approximated by

$$\Psi_{Lmq} (r, \theta, \varphi) = Y_L^m (\theta, \varphi) j_L \left( k_q r \right),$$

where \( r, \theta, \varphi \) are spherical coordinates, \( L, m, q \) are azimuthal, polar and radial mode numbers, respectively, \( Y_L^m (\theta, \varphi) \) is a spherical harmonic and \( j_L \left( k_q r \right) \) is a spherical Bessel function. The radial wave number \( k_q \) is found from the boundary conditions.

Spherical symmetry may be a good approximation for determining the eigenfunction shape (but not always the eigenfrequencies) in a spheroid WGM resonator whose aspect ratio is not too large, because the variation of the boundary from a true sphere in the region of significant WGM field is very small. Due to the spherical symmetry the phase matching in WGM resonators corresponds to conservation of photons’ orbital momenta [9, 10, 16, 32]. If \( L \gg 1 \) the overlap integral in \( \Omega \) breaks up into a product of the angular and radial part, former depending on \( L - m \) and latter on the \( q \) values for all three modes. Each part also weakly depends on wavelengths and the three values of \( L \). The angular parts give rise to the SFG selection rules corresponding to the Clebsch–Gordan coefficients. In particular, they enforce

$$m_p + m_s = m_j, \quad (7)$$
$$L_p + L_s \geq L_f, \quad (8)$$
$$L_p + L_s + L_f = 2N, \quad (9)$$

where \( N \) is an integer. The radial parts lead to no particular selection rules, however as we will see they strongly favor the cases when \( q_p + q_s \approx q_f \).

For mm-size resonators the orbital numbers \( L, m \) are large, and evaluating the overlap integrals with Legendre polynomials and Bessel functions of such orders is impractical. Appropriate asymptotic approximations need to be made. Let us introduce

$$\Psi (r, \theta, \varphi) \approx \Psi_{\text{ang}} (\theta) \Psi_{\text{rad}} (r) e^{im \varphi}$$

inside the resonator, and \( \Psi (r, \theta, \varphi) \equiv 0 \) outside, thereby neglecting the evanescent field. The angular asymptotic for (10) is given by [33]

$$\Psi_{\text{ang}} (\theta) = \frac{N_s}{\sqrt{2\pi}} H_{L-m} \left( \sqrt{L} \cos (\theta) \right) \exp \left\{ -\frac{1}{2} L \cos^2 (\theta) \right\}, \quad (11)$$

and radial asymptotic is

$$\Psi_{\text{rad}} (r) = \frac{N_q}{\sqrt{\pi}} \text{Ai} \left( \alpha_q \frac{L - k_s \left( L + 1/2, q \right) r}{L - k_s \left( L + 1/2, q \right)} \right), \quad (12)$$
In (11) $H_{L-m}$ is the Hermite polynomial of the order $L - m$, and $N_a$ is the normalization factor,

$$N_a^{-2} = \int_{-1}^{1} H_{L-m}^2(\sqrt{L}x) \exp \{ -Lx^2 \} \, dx. \quad (13)$$

In (12), $R$ is the resonator radius, $Ai$ is the Airy function, $\alpha_q$ is its $q$th root (positive value), dimensionless wave number can be approximated [26] as

$$k_r(L, q) \approx L + \alpha_q(L/2)^{1/3}, \quad (14)$$

and the normalization factor is

$$N_r^{-2} = R^3 \int_{0}^{1} Ai^2 \left( \frac{L - k_r(L + 1/2, q) x}{\sqrt{L - k_r(L + 1/2, q)}} \right) x \, dx. \quad (15)$$

The factors (13) and (15) provide the asymptotic wavefunction (10) normalization: $\int dV |\Psi(r, \theta, \varphi)|^2 = 1$.

Approximation (10) allows us to evaluate the radial and angular overlap integrals separately. These overlap factors are shown in figure 5 in the normalized form. To determine the overlap integral for a particular channel one needs to take the appropriate angular part and multiply it by the appropriate radial part. For the fundamental modes, i.e. such that $q_p = q_s = q_f = 1$, $L_p - m_p = L_s - m_s = L_f - m_f = 0$ (the most efficient SFG channel for our wavelengths and resonator size) $\left| \int dV \Psi_p^* \Psi_s^* \Psi_s \right|^2 \approx 2.66 \times 10^6 \text{ cm}^{-3}$. For comparison, the fundamental WGM mode volume in our resonator ranges from approximately $1 \times 10^{-7} \text{ cm}^3$ for the SF to $4 \times 10^{-7} \text{ cm}^3$ for the pump.

We show, therefore, that many SFG channels are possible albeit with different conversion efficiencies, whereas most efficient channel is the one that couples the fundamental modes.
3.3. Temperature tuning of SFG channels

Once an SFG channel has been selected we need to achieve the energy conservation
\[ \omega_p \left( L_p, m_p, q_s \right) + \omega_s \left( L_s, m_s, q_s \right) = \omega_f \left( L_f, m_f + m_p, q_f \right) \] to within a WGM linewidth. In lithium niobate resonators this can be achieved for ordinary pump, ordinary signal and extraordinary sum-frequency WGMs, due to different temperature dependencies of the ordinary and extraordinary refraction indices. We find the phase matching temperatures by iteratively solving the energy conservation condition using the WGM dispersion equation [26] and temperature-dependent Sellmeier equations [34]. Let us point out that for sub-mm resonators the resulting temperatures may significantly (by tens of degrees) differ from the bulk phase matching temperature, due to the geometrical, or waveguide, part of the WGM dispersion.

Based on the previous section analysis, we can rank various SFG channels by their efficiency. We then numerically find the dependence of the frequency detuning \( \Delta \omega = \omega_f - \omega_s - \omega_p \) on temperature and determine the phase matching (\( \Delta \omega = 0 \)) temperature as well as the WGM frequencies \( \omega_f, \omega_s, \omega_p \) at that temperature, for each channel from the ranked list.

In figure 6 we show the theoretical prediction for phase matching temperatures in the range between 120 and 150 °C. The pump modes in this simulation have been assumed equatorial \( \left( L_p - m_p = L_s - m_s = 0 \right) \) and with \( q_{s,p} < 8 \). These assumptions are justified by the high contrast of the observed WGMs and by the FSR measurements discussed above. No assumptions have been made about the sum-frequency WGMs as they are outside of our control. From figure 6 we see that the most efficient \([1,1,1]\) SFG channels have the phase matching temperature near 129 °C, far from other channels of significant efficiency. However, there are also many ‘minor’ SFG channels at this temperature. In our experiment, we indeed observed several other SFG channels with the efficiency at least an order of magnitude below than the one we have been using.
We have verified the theoretical result shown in figure 6 in experiment by tuning the pump and signal lasers across an integer number of FSRs. The wavelengths and phase matching temperatures for these channels are shown in figure 7 together with the numeric simulation for the fundamental channel. A good agreement was achieved by using the MgO concentration as a fitting parameter. This parameter affects the phase matching temperature by entering lithium niobate dispersion [34]. Unfortunately its value is not precisely known for our congruent wafer, except that it should slightly exceed the threshold value of approximately 5%. The fitting yielded a very plausible MgO concentration of 5.63%. It should be pointed out that no other efficient SFG channels can fit the observations with any reasonable MgO concentration. However, as we have already pointed out, many less efficient equatorial channels exist for the same temperatures. The lower observed Ω suggests that the WGM triplet may not have been fundamental. This suggestion is made plausible by the uncertainty in the sum-frequency WGM numbers.

4. Conclusion

To summarize, we have demonstrated efficient triple-resonant SFG in a WGM resonator. We have extended the theoretical analysis for finding the phase-matched WGMs inside the resonator and understood the nonlinear dynamics of frequency conversion in the strong pump-signal coupling regime. The efficiency of this process in the resonator is in general higher than in a traveling-wave geometry due to cavity enhancement, requiring sub-mW powers for saturation. We expect to find applications of this interaction in the fields of spectroscopy, optical communications and data processing, both at classical and at quantum levels. Other conceivable applications are for fundamental tests of quantum theory, e.g. in cavity optomechanics and quantum non-demolition measurements.
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Appendix A. Validation of a WGM $q$ measurement

Since the FSR-based $q$ measurement technique described in the experimental section provided only a limited number of data points at the pump wavelength for our working resonator, and had even more limited success at the signal wavelength, we feel compelled to demonstrate its robustness in a separate measurement with a different test resonator. The test resonator was made from the same material but had a larger radius, $R \approx 700 \text{ m}$, and a denser WGM spectrum. The denser spectrum has allowed us to measure the FSR for 50 modes within a single FSR frequency interval. The results of this measurement carried out with the pump laser are shown in figure A1. Here again, we have forced the $q = 1$ theoretical value to match the two lowest-FSR modes by correcting the resonator radius value from $700 \text{ m}$ to $699.3 \text{ m}$. This has achieved a remarkable agreement up to the $q = 8$ with no further fitting. At higher $q$’s the WGM coupling contrast is considerably reduced, and the measurements become unreliable.

Appendix B. Acquiring the internal conversion efficiency $\Omega$

For low signal powers, the sum-frequency efficiency can be understood in the undepleted pump regimes (figure B1). Since equation (5) is valid only in the undepleted pump approximation, we can infer $\Omega$ only for datasets where the signal powers are very small. Additionally, we assume that the inherent nonlinearity is so weak that for both pump and signal powers below 5 $\mu$W,
there is no noticeable conversion (which is justified by all of our data sets, wherein the efficiency is nearly zero in this region). Since we use the same WGM triplet for the entire experiment, and $\Omega$ is an overlap measure between the excited WGMs, we operate under the condition that $\Omega$ is the same for arbitrary input powers. However, what we can acquire from the measured data is the ratio $\Omega\kappa$, where $\kappa = \omega_j \left( \frac{1}{2Q_s} + \frac{1}{2Q_i} \right)$. The coupling $Q$-factors, $Q_{\mu}^s$, are given by [35] for a TM WGM in a spherical resonator:

$$Q_{\mu}^s = \frac{\pi \sqrt{n_s (n_s - 1)}}{2 \sqrt{n_r^2 - n_s^2}} \left( Rk^2 \right)^{3/2} \exp \left( 2\gamma d \right)$$

(B.1)

$\mu = s, p$, where $n_s, n_p$ are the indices of refraction in the resonator and prism respectively, $R$ is the resonator radius, $\gamma = \sqrt{k^2 (n_s^2 - 1)}$, and $d$ is the distance between the prism and resonator. We will use (B.1) as an order of magnitude estimate for our spheroidal resonator, and multiply it by $n_s^4$ when computing $Q_f^s$ for the extraordinarily polarized 520 nm sum-frequency wave, which is due to the different boundary conditions. Since we operate under critical coupling conditions, i.e., $Q_f^c = Q_f^i$, for the $\lambda = 780$ nm signal wave, we can acquire an empirical value for $d$. In our experiment, $Q_f^c = Q_f^i = 4.88 \times 10^7$ leads to $d = 70.5$ nm. For this distance, for the 520 nm wave we calculate $Q_f^i = 5.34 \times 10^9$. From data fitting we find $Q_f^i = 3.25 \times 10^7$, which is very close to an earlier reported [9] value of $Q^i = 4 \times 10^7$ in the same material at a close wavelength of 532 nm. Let us point out that in lithium niobate a lower $Q$ is indeed expected at a shorter visible wavelength.

Our estimate leads to the undercoupling factor $Q_f^c/Q_f^i = 164$. Therefore, the internally generated green light was coupled out very inefficiently. This leads us to believe that the photorefractive damage induced by the green light could have been significant. Moreover, from equation (B.1), we see that any changes in the index of refraction due to thermo-optic effects,
light-induced charge transport, or even the electro-optic effect due to charge movement on the periphery of the resonator can significantly affect $Q_c^\mu$. The exponential dependence on $d$ places stringent stability criteria, which may have been breached due to the mechanical hysteresis in the brass oven that was employed to allow for temperature-tuning, leading to a time-varying $Q_c^\mu$.

In fact, by studying the transmission curves over the duration of the experiment, we have seen that the total $Q$ of the resonator does change. Therefore, we conclude that the $\Omega$ we acquire while assuming that $\Delta_f = 0$ and neglecting these concurrent effects, might be only used as an order of magnitude estimate. This may further explain why the empirically measured internal conversion efficiency is lower by a factor of 50 than the theoretical prediction. However let us point out that using the $\Omega$ and $\kappa_f$ parameters estimated from the experimental data, we were able to model the signal and pump transmission spectra, as well as the sum-frequency emission spectra, achieving a good agreement with the measurement, as shown in figure B2. No additional fitting parameters were used for this modeling.

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