Optimal third-harmonic generation in an optical microcavity with $\chi^{(2)}$ and $\chi^{(3)}$ nonlineairties

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Third-harmonic generation can be realized via both $\chi^{(3)}$ and cascaded $\chi^{(2)}$ nonlinear processes in a triply-resonant microcavity. It is still unknown how these processes interfere with each other and the optimization of the conversion efficiency still remains as a question. In this work, the interplay between the direct third-harmonic generation and the cascaded process combining of the second-harmonic generation and the sum-frequency generation are investigated. It is found that the interference effect between these two processes can be used to improve the conversion efficiency. By optimizing the cavity resonance and the external coupling conditions, the saturation of the nonlinear conversion is mitigated and the third-harmonic conversion efficiency is increased. A design rule is provided for achieving efficient third-harmonic generation in an optical microcavity, which can be generalized further to the high-order harmonic generations.

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INTRODUCTION

Nonlinear photonics based on the third-order nonlinear process ($\chi^{(3)}$) can be used for various applications in classical and quantum fields [1–4]. Generally speaking, the efficiency of the third-order nonlinear process is very low, due to the very weak $\chi^{(3)}$ susceptibilities in most materials. High-Q optical microcavities are often used to enhance the nonlinear interaction $\chi^{(3)}$, because the optical mode is confined tightly and the density of state is greatly enhanced. During the last decades, $\chi^{(3)}$-assisted third-harmonic generation were studied theoretically [9] and realized experimentally in various types of microcavities, including microdroplets [10,11], silica toroid/microsphere/microbottle [12–15], SiN microring [16], silicon photonic-crystal micro/nano-cavities [17] and microcavities with hybrid materials [18].

To enhance the efficiency of the third-harmonic generation (THG), one should increase the nonlinear susceptibility of the nonlinear processes, reduce the mode volume, optimize the modal overlap as well as increase the quality factor of the microcavity [6]. For a given material system, the size and geometry of an optimal microcavity should be designed to satisfy the phase-matching and maximize the mode overlap factor [17,22]. By using the intermodal dispersion of higher-order modes for phase matching and optimizing these parameters, third-harmonic generation with a efficiency of 180% $W^{-2}$ has been observed on an integrated photonic chip [18]. On the other hand, one should notice that the quality factor of an microcavity is usually limited due to the fabrication imperfection and the material absorption, which builds a barrier to increase the efficiency of THG.

Fortunately, the mechanism of cascading different nonlinear processes provides a promising approach for engineering the nonlinear susceptibility of the material. For example, it has been investigated that effective $\chi^{(3)}$ nonlinear process can be constructed with low-order $\chi^{(2)}$ process [23]. In a high-Q microcavity, multiple and cascaded nonlinear processes with high efficiency can be observed simultaneously. People have demonstrated the second-harmonic generation (SHG), sum-frequency generation (SFG), as well as THG could be observed simultaneously under the multiple-resonant condition in a cavity by introducing materials with both $\chi^{(2)}$ and $\chi^{(3)}$ nonlinearity, e.g., Lithium Niobate and Aluminum Nitride (AIN). The cascading of SHG and SFG (SHG-SFG) can also convert photons from the fundamental to the third-harmonic frequency, which is equivalent to the direct $\chi^{(3)}$ assisted THG (DTHG). Recently, the cascading of $\chi^{(2)}$ processes for achieving THG was observed experimentally in periodically-poled MgO : LiNbO$_3$ cavity [23] and LiNbO$_3$ microdisk [24]. The authors demonstrated that the cascaded effect SHG-SFG in a high $\chi^{(2)}$ cavity can exceed the essential DTHG based on the $\chi^{(3)}$ nonlinearity. Besides, people have also observed DTHG accompanied by Raman scattering and four-wave mixing [25,26], which increases the potential for multiple applications. In addition to THG, some other cascaded nonlinear processes in cavities have also been applied for the realization of frequency combs, broadband wavelength conversion, quantum entanglement, etc [27,28].

Since the TH light can be generated by both processes of $\chi^{(2)}$ and cascaded $\chi^{(2)}$, it is interesting to understand how these processes influence each other and which one is better to maximize the THG efficiency. In our previous work, we have demonstrated that different nonlinear processes will interfere with each other [23]. Here, we study a system based on a microring resonator (MRR) in which both the processes of direct $\chi^{(3)}$ and cascaded $\chi^{(2)}$ are...
involved. The behavior of these processes and the interplay between them are investigated quantitatively. Both regimes with a low pump power and a high pump power are investigated. It is shown that the interference happens when the quality factor of the MRR approaches a critical value. The interference can be utilized to improve the conversion efficiency. In particular, when operating with a high pump power, one should optimize the resonant frequencies and the external coupling ratio of the MRR to compensate the saturation effect for achieving a maximal conversion efficiency.

**SYSTEM AND MODEL**

Fig. 1 schematically illustrates a triply-resonant microcavity coupled with a bus waveguide. The three resonant modes are the fundamental (FM) mode $a$, the second-harmonic (SH) mode $b$ and the third-harmonic (TH) mode $c$ with frequency $\omega_{i,0}$ ($i \in \{a, b, c\}$). This cavity is formed by or filled with nonlinear materials that have both non-zero $\chi^{(2)}$ and $\chi^{(3)}$ nonlinear susceptibilities. The inset of Fig. 1 illustrates three possible nonlinear interactions in this cavity: (i) DTHG between $a$ and $c$ via the intrinsic $\chi^{(3)}$ nonlinearity. (ii) SHG between $a$ and $b$ via the $\chi^{(2)}$ nonlinearity. (iii) SFG between $a$, $b$ and $c$ via the $\chi^{(2)}$ nonlinearity. According to the phase-matching condition, the conservation of angular momentum quantum number should be satisfied, i.e., $m_a = m_b/2 = m_c/3$. The cavity is driven by a continuous-wave laser with the frequency $\omega_a$ around the resonance $\omega_{a,0}$ of the fundamental mode. The system Hamiltonian is given by

$$H = H_0 + H^{(2)} + H^{(3)} + H_d,$$

where $H_0$ is the free Hamiltonian of the three modes, $H^{(2)}$ is the nonlinear interactions due to $\chi^{(2)}$, $H^{(3)}$ is the nonlinear interaction due to $\chi^{(3)}$, and $H_d$ is the external coherent driving to the fundamental mode. One has

$$H_0 = \omega_{a,0}a^\dagger a + \omega_{b,0}b^\dagger b + \omega_{c,0}c^\dagger c,$$

$$H^{(2)} = g_{22}(a^2b + a^2b^\dagger) + g_{21}(a^\dagger b^\dagger c + abc^\dagger),$$

$$H^{(3)} = g_3(a^3c + a^3c^\dagger),$$

$$H_d = \epsilon_a(e^{i\omega_a t} + a^\dagger e^{-i\omega_a t}),$$

where $g_3$ is the single photon coupling strength of DTHG, $g_{22}$ is the single photon coupling rate of SHG, $g_{21}$ is the single photon coupling rate of SFG, $\epsilon_a = \sqrt{2\kappa_{a,1}P_p/\hbar \omega_a}$ is the pump parameter with $\kappa_{a,1}$ and $P_p$ being the external coupling rate and pump power, respectively.

For a rotational symmetric whispering gallery microcavity, the single photon coupling rate of the $\chi^{(2)}$ and $\chi^{(3)}$ nonlinear processes can be calculated using the method in Ref. [31]. The typical values for a AlN MRR are $g_{22,21}/2\pi \sim 10^3 \text{Hz}$, $g_{3}/2\pi \sim 10 \text{Hz}$ [19, 35, 36]. Specially, the values can be improved greatly in a Niobate Lithium microcavity with optimized fabrication and structures [37]. In this model, the self-phase modulation and cross-phase modulation effects are neglected, since these effects only shift the resonance frequencies at very high pump powers. It is convenient to solve this problem in the rotating frame of $\omega_a a^\dagger a + 2\omega_a b^\dagger b + 3\omega_a c^\dagger c$ to eliminate the high frequencies, and then the free Hamiltonian

$$H_0 = \delta_a a^\dagger a + \delta_b b^\dagger b + \delta_c c^\dagger c,$$

where $\delta_i = \omega_i,0 - \omega_i$ ($i \in \{a, b, c\}$) represents the detuning of the resonant mode to the photon frequency. Following the Heisenberg-Langevin equation and the mean field approximation, both the operators and mean field obey the following form,

$$\frac{da}{dt} = \alpha_a a - i3g_3a^2c - i2g_{22}a^\dagger b - ig_{21}b^\dagger c - ic_a,$$

$$\frac{db}{dt} = \alpha_b b - ig_{22}a^2 - ig_{21}a^\dagger c,$$

$$\frac{dc}{dt} = \alpha_c c - ig_3a^3 - ig_{21}ab,$$

where $\alpha_i = -i\delta_i - \kappa_i$, $\kappa_i = \kappa_{i,0} + \kappa_{i,1}$ with $\kappa_{i,0}$ and $\kappa_{i,1}$ being the intrinsic decay rate of the cavity and the external coupling rate between the cavity and waveguide. The intracavity photon amplitude of each mode at steady state can be solved by setting $\frac{da}{dt} = \frac{db}{dt} = \frac{dc}{dt} = 0$. Using the input-output relationship for the microcavity $\epsilon_{in} = -i\sqrt{2\kappa_{c,1}c_s}$ [38], the output power of the TH light in the waveguide and the absolute conversion efficiency can be calculated as

$$P_{TH} = 2\hbar \omega_c |c_s|^2,$$

$$\eta = P_{TH}/P_p,$$

respectively. According to Eqs. (7-10), both the internal nonlinear processes and the external coupling rates should be optimized to achieve the maximal conversion efficiency.
Weak-pump regime

To reveal the basic physical insight of this system, we first investigate the behavior of the system under a weak pump. In this regime, the conversion efficiency from the fundamental mode $a$ to the harmonic modes $b$, $c$ are very weak. As a result, it is reasonable to apply the non-depletion approximation on mode $a$, under which the intracavity photon number can be treated as a constant since the backaction from the harmonic modes are negligible. The Hamiltonian of the simplified model reduces to

$$H = \delta_b b^\dagger b + \delta_c c^\dagger c + g_3 |a_s|^3 (c + c^\dagger) + g_{22} |a_s|^2 (b + b^\dagger) + g_{21} |a_s| (b^\dagger c + bc^\dagger),$$

(12)

where $a_s = i\epsilon_a / (-i\delta_a - \kappa_a)$ is the intracavity photon number amplitude of the mode $a$ by neglecting the backaction from modes $b$ and $c$. Since the phases of $a_s$ and the coupling strengths can be absorbed into the operators $b$ and $c$, we replace $a_s$ by $|a_s|$ in the Hamiltonian. The dynamics of the SH and TH modes is described as,

$$\frac{db}{dt} = (-i\delta_b - \kappa_b) b - ig_{22} |a_s|^2 - ig_{21} |a_s| c$$  

(13)

$$\frac{dc}{dt} = (-i\delta_c - \kappa_c) c - ig_3 |a_s|^3 - ig_{21} |a_s| b.$$  

(14)

The intracavity photon amplitude of the TH mode can be solved as

$$c_s = \frac{ig_3 - g_{21}g_{22}}{\alpha_c + \frac{g_{21}g_{22}}{\alpha_b}} |a_s|^2 |a_s|^3 \approx \left( \frac{ig_3 - g_{21}g_{22}}{\alpha_c + \frac{g_{21}g_{22}}{\alpha_b}} \right) |a_s|^3.$$  

(15)

Here the first term in the bracket represents the contribution of DTHG and the other term is due to the cascaded SHG-SFG process. It can be seen that both the direct $\chi^{(3)}$ and the cascaded $\chi^{(2)}$ processes contribute to the generation the TH light. As an analog to $g_3$, the effective third-order nonlinear coupling strength of the cascaded SHG-SFG process can be defined as $-i \frac{g_{21}g_{22}}{\alpha_b}$. Now, we can judge which process dominates the generation of the TH light by directly comparing $g_3$ and $\left| \frac{g_{21}g_{22}}{\alpha_b} \right|$. The coupling strength $g_3$ and $g_{21}$ depend on the mode volume and the mode overlap, which is relevant to the size and geometry of the optical cavity. For a given optical cavity, the effective coupling strength $\left| \frac{g_{21}g_{22}}{\alpha_b} \right|$ scales inversely with the decay rate and detuning of the SH mode $b$. The higher the quality factor of mode $b$, the stronger the effective coupling strength. For zero detuning of mode $b$, $\left| \frac{g_{21}g_{22}}{\alpha_b} \right|$ reduces to $\frac{g_{21}g_{22}}{\alpha_b}$. The critical quality factor $Q_{cr}$, at which $g_3$ equals $\frac{g_{21}g_{22}}{\alpha_b}$, is given as

$$Q_{cr} = \frac{\omega}{\gamma} \frac{g_{21}g_{22}}{g_3}.$$  

(16)

When $Q_b \gg Q_{cr}$, the cascaded SHG-SFG process is more efficient than the DTHG process. In contrast, for low quality factor $Q_b \ll Q_{cr}$, the DTHG process is dominant. Therefore, it is necessary to engineer the SH mode $b$ if an high-Q optical cavity is available.

When the quality factor is close to $Q_{cr}$, the interplay between DTHG and SHG-SFG should be taken into account. Using the nonlinear coupling strengths demonstrated previously [18, 19], the $Q_{cr}$ for an AlN MRR is calculated around $2.0 \times 10^5$ when operating at the wavelength around 775 nm. This $Q$ is achievable for the MRRs fabricated with the current fabrication technologies. In Fig. 2(a), we plot the relationship between the intensity of the TH light and the pump power for DTHG, SHG-SFG and the combined process. The parameters used in the calculations are $g_3/2\pi = 10$ Hz, $g_{21}/2\pi = 0.1 MHZ$, $\kappa_b/2\pi = 1 GHZ$, $\delta_b = 0.5 \kappa_b$, $\kappa_{a,b}/2\pi = 0.4 \times 10^9$ Hz, $\kappa_{a,b} = 4 \kappa_{a,b}$, $\kappa_{a,b} = 10 \kappa_{a,b}$, $\delta_b = 0.5 \kappa_b$. All the three modes are critically coupled to the waveguide.

![FIG. 2. Interference between DTHG and SHG-SFG process.](image)

(a) The intensity of the TH mode generated from three different processes. (b) The relation between the normalized intensity of the TH mode and the detuning of the SH mode. The intensities are normalized to that of the DTHG process with $P^{TH}_d / P^{TH}_D$ (it represents the intensity of the TH mode from DTHG, SHG-SFG and the combined process). The parameters used in the calculations are $g_3/2\pi = 10$ Hz, $g_{21}/2\pi = 0.1 MHZ$, $\kappa_b/2\pi = 1 GHZ$, $\delta_b = 0.5 \kappa_b$. All the three modes are critically coupled to the waveguide.

The crucial factor that accounts for the interference is the phase difference $\Delta \theta$ between $ig_3$ and $-\frac{g_{21}g_{22}}{\alpha_b}$. For
\(-\pi/2 < \Delta \theta < \pi/2\), DTHG and cascaded SHG-SFG process interfere constructively and the efficiency can be enhanced. Oppositely, the destructive interference suppresses the generation of the TH light. We can define the effective third-order coupling strength \(g_{\text{eff}} = ig_3 - \frac{g_{21}g_{22}}{\kappa_0 - \kappa}\) to describe the whole process. Fig. 2(b) shows the relationship between the intensity of the TH light (normalized to the DTHG process) and the detuning of the SH mode. Extraordinarily, THG is not the most efficient for zero detuning of mode \(b\), at which the cascaded \(\chi^{(2)} - \chi^{(3)}\) is the most efficient (Dashed Red line in Fig 2(b)). In the case shown in Fig. 2(b), the THG efficiency can be even suppressed up to an order of magnitude. Therefore, one must carefully control the detuning of the intermediate mode \(b\) to optimize the conversion efficiency.

**Strong-pump regime**

When the intracavity photon number of the pump increases, the cooperativity of SHG \(C_{\text{SHG}} = \frac{g_{22}^2 |a|^4}{|\alpha_c|^2}\) or DTHG \(C_{\text{DTHG}} = \frac{g_{22}^2 |a|^4}{|\alpha_c|^2}\) approaches to unity. The nonlinear coupling induced changes of the decay rates of the cavity modes become significant. The down-conversion from the harmonic modes to the fundamental mode will suppress the up-conversion. Non-depletion approximation is not valid for large pump powers and the conversion efficiency saturates. The saturation effect has been theoretically observed for SHG [39] and experimentally predicted for THG [39]. In this situation, Eqs. (7)-(9) must be solved without approximation to analyze the behavior of the system. Then, the intracavity photon amplitude of mode \(a\) can be derived as

\[
\begin{align*}
- \left( \frac{g_{21}}{\alpha_b^*} - ig_3 - \frac{g_{21}g_{22}}{\alpha_b} \right) \left[ \frac{g_{21}}{\alpha_c^*} + \frac{g_{21}g_{22}}{\alpha_b} \right] \left[ \frac{g_{21}}{\alpha_c} + \frac{g_{21}g_{22}}{\alpha_b} \right] |a|^6 + \frac{2g_{22}^2 |a|^2}{\alpha_b} \\
\left( i3g_3 - \frac{2g_{21}g_{22}}{\alpha_b} + \frac{g_{21}g_{22}}{\alpha_b^*} \right) \left[ \frac{g_{21}}{\alpha_c} + \frac{g_{21}g_{22}}{\alpha_b} \right] |a|^4 + \frac{2g_{22}^2 |a|^2}{\alpha_b} \right] = i_0 c_a
\end{align*}
\]

and the amplitude of the harmonic modes are

\[
c = \frac{ig_3 - g_{21}g_{22}}{\alpha_c} |a|^3 \tag{17}
\]

\[
b = \frac{i}{\alpha_b} \left( g_{22} + g_{21} \right) \frac{ig_3 - g_{21}g_{22}}{\alpha_c} |a|^2 \tag{18}
\]

Equation (18) has the similar form as Eq. (15) with an additional intensity dependent \(\frac{g_{21}^2}{\alpha_b} |a|^2\) term in the denominator, which was neglected for weak pump powers. Also, the intracavity photon number in the fundamental mode no longer scales linearly with the pump power with quite a portion of energy being converted to the harmonic modes. By numerically solving Eqs. (17)-(18), the relationship between the conversion efficiency and the pump power \(P_p\) is plotted in Fig. 3(a). The blue, red and black lines represent the conversion efficiency of the DTHG, SHG-SFG and the combined process, respectively. It can be seen that the efficiency of THG from neither DTHG nor cascaded SHG-SFG no longer scale cubically with the pump power when the conversion efficiency is high. Due to the saturation, not only the SHG is suppressed, but also the SFG efficiency from mode \(a\) to \(c\) decreases. Because \(g_3 << g_2\), \(C_{\text{THG}}\) is much smaller than \(C_{\text{SHG}}\), the saturation of the cascaded SHG-SFG appears ahead of the DTHG along with the increase of the pump power. The efficiency of cascaded SHG-SFG increases much slower than DTHG, which can be found by comparing the blue and red lines. At very high pump powers, the efficiency of DTHG can even exceed that of the SH-SFG.

Since the saturation behaviors of the direct \(\chi^{(2)}\) and the cascaded \(\chi^{(2)}\) nonlinear processes are different, their contributions to the whole process scale differently with the pump power. The interference will be different for high pump powers. Fig. 3(b) shows the conversion efficiency against the detuning \(\delta_0\) of the SH mode for different pump powers. As a consequence of the unsynchronized saturation effect of the cascaded SHG-SFG and DTHG processes, the interference fringe becomes less pronounced. The maximal enhancement factor decreases as the pump increases and the optimal detuning \(\delta_{0,m}\) for maximum THG efficiency shifts to larger values when the pump increases (Fig. 3(c)). For very high values of the pump power, the individual DTHG is efficient enough. The presence of the SH mode and their coupling add nonlinear losses to the TH mode, thus reducing the conversion efficiency. In this case, tuning the SH mode off the resonance is more favorable. Simultaneously, the TH mode also add nonlinear loss to the SH mode and the SHG efficiency is also reduced.

To summarize briefly, the efficiency of THG is determined in two ways in the high-pump regime. On the one hand, the generation of the TH photons via the cascaded SHG-SFG process and the DTHG interfere with each other. On the other hand, the cascaded SHG-SFG and DTHG process change the decay rates of the fundamental and TH modes. The cavity modes experience an additional nonlinear decay channel, with the total decay rate \(\kappa = \kappa_0 + \kappa_1 + \kappa_{nd}\), which suppresses the nonlinear frequency conversion. Since the saturation of these two process are unsynchronized, the constructive interference is also destroyed.

**OPTIMAL THIRD-HARMONIC GENERATION**

In this section, we come to a more realistic question for practical applications that what is the upper limit of the THG efficiency and how to achieve that efficiency if the pump power and the intrinsic quality factor of the microcavity are limited. In the above sections, we have explicitly pointed out that the interference between DTHG
Conversion efficiency %

Optimal detuning $d_b,m/k_b$

Maximum intensity

Pump power /mW

Conversion efficiency

Pump power /mW

Normalized intensity

Optimal detuning $\delta_{b,m}/k_b$

Optimal detuning $\delta_{a,nl}/k_a$

Maximum intensity

Pump power /mW

FIG. 3. Saturation effects in TH light generation. (a) Relation between the conversion efficiency from the fundamental frequency to the TH frequency for three different processes. The cascaded SHG-SFG saturates first when increasing the pump power. (b) Interference fringes for different pump powers. The saturation of the SHG-SFG process destroys the constructive interference significantly. (c) Optimal detuning (Blue line) of the SH mode for TH light generation and the corresponding maximum enhancement factor (Orange line) over the DTHG process. When the pump increases, the optimal detuning shifts to larger values and the constructive interference is destroyed. In the high-pump regime, THG is dominated by the DTHG process.

and cascaded SHG-SFG process can be used to improve the THG efficiency with low pump powers. While operating with high pump powers, the enhancement provided by the interference is weakened. Also, the decay rates or quality factors of the cavity mode decreases due to the nonlinear coupling [19,10].

For experiments, several parameters are fixed due to the limitations of material and fabrication, therefore the nonlinear coupling strengths $g_{21,22,3}$ and the intrinsic quality factor $Q_{i,0} = \frac{2\pi}{\kappa_{i,0}}$ of the three resonant modes (i.e. $a, b, c$) are limited. So, we can engineer the mode frequency detuning $\delta_b$ and the external coupling strengths $\kappa_{i,1}$ to optimize the THG. The role of $\delta_b$ is to control the interference between the DTHG and the SHG-SFG processes, while the $\kappa_{i,1}$ balances the extraction of the TH light from the cavity $(\kappa_{e,1}/\kappa_{c})$, the cooperativity of the nonlinear frequency conversion and also the saturation effect.

Fig. 4(a) shows the absolute conversion efficiency $\eta$ for different external coupling rates with pump power $P_p = 10$ mW (weak pump) and $P_p = 1000$ mW (strong pump). The detuning $\delta_b$ is chosen to be the optimal detuning as studied in Fig.3(b). It is found that, the critical coupling for both the fundamental mode and the TH mode is required to achieve maximum conversion efficiency for weak coupling regime. The reason is that the cooperativities $C_{THG} \ll 1$ and $C_{SHG} \ll 1$ in this regime, the backaction induced decay rate $\kappa_{i,nl}$ (saturation effect) can be neglected compared to $\kappa_{i,0}$. In this case, the optimal external coupling rate that balances the extraction and cooperativity is $\kappa_{c,1} \approx \kappa_{c,0}$ and $\kappa_{a,1} \approx \kappa_{a,0}$. Since there is no input or output required for the mode $b$, thus we want $\kappa_{b,1} \approx 0$.

When operating with a high pump power $P_p = 1000$ mW, the maximum conversion efficiency 51.7 % is obtained at $\kappa_{a,1} = 2.95 \kappa_{a,0}$ and $\kappa_{c,1} = 5.01 \kappa_{c,0}$, which lies in the over-coupled region. If the external coupling rates are set as $\kappa_{a,1} = \kappa_{a,0}$ and $\kappa_{c,1} = \kappa_{c,0}$, the conversion efficiency is only 25.0 %. This indicates that the cooperativities approaches to 1, leading to relatively large changes of the effective decay rates of the modes, $\kappa_{i,nl} \sim \kappa_{i,0}$. Therefore, the loaded quality factors of mode $a$ and $c$ will decrease and the waveguide-cavity coupling rates must be optimized to a higher value to ensure the critical coupling condition $\kappa_{a,1} = \kappa_{a,0} + \kappa_{a,nl}$ and $\kappa_{c,1} = \kappa_{c,0} + \kappa_{c,nl}$.

Taking both the internal and external coupling parameters into consideration, we calculate the achievable conversion efficiency of THG using a microcavity with given pump power. By optimizing the detuning $\delta_b$ of the SH mode $b$ and the external coupling rates of the fundamental and TH modes, we obtain the relationship between the pump power and the maximum conversion efficiency, as shown in Fig. 4(b). The dashed red line shows the efficiency of DTHG without optimal condition for the external coupling. By comparing the optimal and DTHG process, it is beneficial to utilize the cascaded SHG-SFG and the constructive interference effect when the pump power is low. As the pump increases, the advantage of the cascaded process and the interference is weakened by the saturation and the two lines get close to each other. Therefore, there is no need to design the cascaded SHG-SFG process if the pump is too high since DTHG dominates the TH light generation. In the top right of the Fig. 4(b), the separation of the two lines becomes large again. In this area, the higher conversion efficiency mainly attributes from the optimized external coupling conditions.
FIG. 4. Optimization of the conversion efficiency. (a) Relationship between the conversion efficiency and the external coupling rates $\kappa_{a,1}$ and $\kappa_{c,1}$. Left: $P_p = 10$ mW. Right: $P_p = 1000$ mW. In the high-pump regime, the cavity modes should be designed over-coupled to the waveguide to achieve optimal efficiency. (b) Relationship between the optimal THG efficiency and the pump power. Black line: the detuning $\delta$, external coupling rates $\kappa_{a,1}$ and $\kappa_{c,1}$ are optimized; Red line: efficiency of DTHG with all the modes critical-coupled to the waveguide. Orange: optimal external coupling rate of the fundamental mode $a$. Blue: optimal external coupling rate of the TH mode $c$. The external coupling rates shown in this figure are normalized to the corresponding intrinsic decay rates $\kappa_{i,0}$.

CONCLUSION

In conclusion, we have studied the third-harmonic generation in an optical microcavity, which allows both the $\chi^{(3)}$-assisted direct THG and $\chi^{(2)}$-assisted cascaded SHG and SFG for the generation of the third-harmonic light. Under the non-depletion approximation for weak pump, we explicitly show the contributions of direct THG and the cascading of SHG and SFG processes and predict the interference between them. According to our analysis, the detuning of the intermediate second-harmonic mode plays a significant role on the conversion efficiency. For high pump powers, we analyze the different saturation effects of these processes. The interference fringe becomes less pronounced and the external coupling condition should be optimized to improve the THG efficiency. At last, the optimal conversion efficiency for a microcavity with given parameters and pump power is studied. Our results clearly explain the mechanisms of THG in presence of both $\chi^{(2)}$ and $\chi^{(3)}$ nonlinearity and give guidelines to optimize the nonlinear conversion efficiency. Since our study is based on the parameters of microcavities with current fabrication and experimental technologies, we believe the new phenomenon predicted in our work will be experimentally demonstrated and find their applications in the near future. The analyses can be also generalized to other nonlinear processes, such as SHG, Raman, and high-order harmonic generations [41], which would stimulate more new physics with cascaded nonlinear optics effects.

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[1] R. W. Boyd, Nonlinear optics (Academic press 2003).
[2] G. P. Agrawal, Nonlinear fiber optics (Academic press 2007).
[3] C. Reimer, M. Kues, P. Roztocki, B. Wetzel, F. Grazioso, B. E. Little, S. T. Chu, T. Johnston, Y. Bromberg, L. Caspani D. J. Moss and R. Morandotti, Generation of multiphoton entangled quantum states by means of integrated frequency combs, Science 351, 1176 (2016).
[4] J. Leuthold, C. Koos, and W. Freude, Nonlinear silicon photonics, Nat. Photonics 4, 535 (2010).
[5] K. J. Vahala, Optical microcavities, Nature 424, 839 (2003).
[6] D. V. Strekalov, C. Marquardt, A. B. Matsko, H. G. Schwefel, and G. Leuchs, Nonlinear and quantum optics with whispering gallery resonators, J. Opt. 18, 123002 (2016).
[7] G. Liu, A. Coillet, and Y. K. Chembo, Nonlinear photonics with high-Q whispering-gallery-mode resonators, Adv. Opt. Photon. 9, 828 (2017).
[8] H. Rong, A. Liu, R. Jones, O. Cohen, D. Hak, R. Nicolaescu, A. Fang, and M. Paniccia, An all-silicon raman laser, Nature 433, 292 (2005).
[9] H. Hashemi, A. W. Rodriguez, J. D. Joannopoulos, M. Solja´ ci´ c, and S. G. Johnson, Nonlinear harmonic generation and devices in doubly resonant kerr cavities, Phys. Rev. A 79, 013812 (2009).
[10] D. H. Leach, R. K. Chang, W. P. Acker, and S. C. Hill, Third-order sum-frequency generation in droplets: experimental results, J. Opt. Soc. Am. B 10, 34 (1993).
[11] I. Kasparian, B. Krämer, J. P. Dewitz, S. Vajda, P. Rairoux, B. Vezin, V. Boutou, T. Leisner, W. Hübner, J. P. Wolf, L. Wöste, and K. H. Bennemann, Angular dependences of third harmonic generation from microdroplets, Phys. Rev. Lett. 78, 2952 (1997).
[12] T. Carmon and K. J. Vahala, Visible continuous emission from a silica microphotonic device by third-harmonic
generation, Nat. Phys. 3, 430 (2007).

[13] R. Ismaeel, T. Lee, M. Ding, N. G. R. Broderick, and G. Brambilla, Nonlinear microfiber loop resonators for resonantly enhanced third harmonic generation, Opt. Lett. 37, 5121 (2012).

[14] D. Farnesi, A. Barucci, G. C. Righini, S. Berneschi, S. Soria, and G. Nunzi Conti, Optical frequency conversion in silica-whispering-gallery-mode microspherical resonators, Phys. Rev. Lett. 112, 093901 (2014).

[15] M. Asano, S. Komori, R. Ikuta, N. Imoto, Ş. K. Özdemin, and T. Yamamoto, Visible light emission from a silica microbottle resonator by second- and third-harmonic generation, Opt. Lett. 41, 5793 (2016).

[16] J. S. Levy, M. A. Foster, A. L. Gaeta, and M. Lipson, Harmonic generation in silicon nitride ring resonators, Opt. Express 19, 11415 (2011).

[17] M. Galli, D. Gerace, K. Weim, T. F. Krauss, L. O’Faolain, G. Guizzetti, and L. C. Andreani, Low-power continuous-wave generation of visible harmonics in silicon photon crystal nanocavities, Opt. Express 18, 26613 (2010).

[18] J. B. Surya, X. Guo, C.-L. Zou, and H. X. Tang, Efficient third-harmonic generation in composite aluminum nitride/silicon nitride microrings, Optica 5, 103 (2018).

[19] X. Guo, C. Zou, and H. Tang, Second-harmonic generation in aluminum nitride microrings with 2500 %/ W conversion efficiency, Optica 3, 1126 (2016).

[20] Z. Lin, X. Liang, M. Lončar, S. G. Johnson, and A. W. Rodriguez, Cavity-enhanced second-harmonic generation via nonlinear-overlap optimization, Optica 3, 253 (2016).

[21] K.-H. Kim, M.-S. Hwang, H.-R. Kim, J.-H. Choi, Y.-S. No, and H.-G. Park, Direct observation of exceptional points in coupled photonic-crystal lasers with asymmetric optical gains, Nat. Commun. 7, 13893 (2016).

[22] Y. Zeng, I. Roland, X. Checoury, Z. Han, M. El Kurdi, S. Sauvage, B. Gayral, C. Brimont, T. Guillet, F. Semon and P. Boucaud, Imaging of photonic crystal localized modes through third-harmonic generation, ACS Photonics 3, 1240 (2016).

[23] K. Sasagawa and M. Tsuchiya, Highly efficient third harmonic generation in a periodically poled MgO:LiNbO3 disk resonator, Appl. Phys. Express 2 (2009).

[24] S. Liu, Y. Zheng, and X. Chen, Cascading second-order nonlinear processes in a lithium niobate-on-insulator microdisk, Opt. Lett. 42, 3626 (2017).

[25] A. Chen-Jinnai, T. Kato, S. Fujii, T. Nagano, T. Kobatake, and T. Tanabe, Broad bandwidth third-harmonic generation via four-wave mixing and stimulated raman scattering in a microcavity, Opt. Express 24, 26322 (2016).

[26] W. Liang, A. A. Savchenkov, Z. Xie, J. F. McMillan, J. Burkhart, V. S. Ilchenko, C. W. Wong, A. B. Matsko, and L. Maleki, Miniature multioctave light source based on a monolithic microcavity, Optica 2, 40 (2015).

[27] L. Wang, L. Chang, N. Volet, M. H. Pfeiffer, M. Zervas, H. Guo, T. J. Kimpenberg, and J. E. Bowers, Frequency comb generation in the green using silicon nitride microresonators, Laser Photonics Rev. 10, 631 (2016).

[28] S. Fujii, T. Kato, R. Suzuki, and T. Tanabe, Third-harmonic blue light generation from kerr clustered combs and dispersive waves, Opt. Lett. 42, 2010 (2017).

[29] H.-T. Tan and H. Huang, Bright quadripartite entanglement from competing $\chi^{(2)}$ nonlinearities, Phys. Rev. A 83, 015802 (2011).

[30] V. Ulvila, C. Phillips, L. Halonen, and M. Vainio, Frequency comb generation by a continuous-wave-pumped optical parametric oscillator based on cascading quadratic nonlinearities, Opt. Lett. 38, 4281 (2013).

[31] H. Jung, R. Stoll, X. Guo, D. Fischer, and H. X. Tang, Green, red, and IR frequency comb line generation from single ir pump in aln microring resonator, Optica 1, 396 (2014).

[32] R. Wolf, I. Breunig, H. Zappe, and K. Buse, Cascaded second-order optical nonlinearities in on-chip micro rings, Opt. Express 25, 29927 (2017).

[33] M. Li, C. L. Zou, C. H. Dong, X. F. Ren, and D. X. Dai, Enhancement of the second-harmonic generation on the cascaded second- and third-order nonlinear processes in a multimode optical microcavity, arXiv preprint arXiv:1805.08388 (2018).

[34] X. Guo, C.-L. Zou, H. Jung, Z. Gong, A. Bruch, L. Jiang, and H. X. Tang, Efficient visible frequency comb generation via cherenkov radiation from a kerr microcomb, arXiv preprint arXiv:1704.04264 (2017).

[35] X. Guo, C. L. Zou, H. Jung, and H. X. Tang, On-Chip Strong Coupling and Efficient Frequency Conversion between Telecom and Visible Optical Modes, Phys. Rev. Lett. 117, 123902 (2016).

[36] X. Guo, C.-L. Zou, C. Schuck, H. Jung, R. Cheng, and H. X. Tang, Parametric down-conversion photon-pair source on a nanophotonic chip, Light Sci. Appl. 6, e16249 (2017).

[37] M. Zhang, C. Wang, R. Cheng, A. Shams-Ansari, and M. Lončar, Monolithic ultra-high-Q lithium niobate microring resonator, Optica 4, 1536 (2017).

[38] D. F. Walls and G. J. Milburn, Quantum optics (Springer Science & Business Media 2007).

[39] R. Yu, C. Ding, J. Wang, and D. Zhang, Enhanced visible light generation in an active microcavity via third-harmonic conversion beyond the non-depletion approximation, J. Appl. Phys. 122, 244303 (2017).

[40] J. Li, H. Lee, T. Chen, and K. J. Vahala, Characterization of a high coherence, brillouin microcavity laser on silicon, Opt. Express 20, 20170 (2012).

[41] J. Moore, M. Tomes, T. Carmon, and M. Jarrahi, Continuous-wave ultraviolet emission through fourth-harmonic generation in a whispering-gallery resonator, Opt. Express 19, 24139 (2011).