Bright squeezed light via second harmonic generation in a whispering-gallery mode resonator

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ABSTRACT
We investigate the conditions necessary for bright squeezed light generation through second harmonic generation inside a crystalline whispering-gallery mode resonator. We show that the variance of a coherent mode can be reduced by a factor of 9 due to low loss through a nonlinear medium. This results in a one-step process that can generate efficient bright squeeze light at a desired wavelength.

Keywords: whispering gallery mode resonators, nonlinear crystal, second harmonic generation, squeezed light

1. INTRODUCTION
The accuracy of any measurement is ultimately limited by the uncertainty principle, which is usually referred to as the “standard quantum limit”. However, it is possible to beat this limit by using non-classical optical probes with modified quantum noise or/and by using new measurement protocols based on quantum mechanical operators. So far, the most common and reliable technology for generation of such states is based on nonlinear conversion of one or more optical fields of one frequency into optical field(s) of a different frequency. Since in such nonlinear processes photons are created or absorbed in pairs, this process can significantly modify the photon statistics of all involved optical fields, and lead, for example, to generation of correlated photon pairs, amplitude or phase squeezed light or squeezed vacuum. However, traditional realizations of such sources, based on bulk nonlinear crystals, are rather complex and bulky, and usually require powerful lasers, high quality optical cavities, and exceptional temperature and mechanical stability.

There are many advantages of using high-quality whispering gallery mode resonators, made out of nonlinear crystals. Crystalline whispering-gallery mode resonators have already dramatically improved the efficiency for various nonlinear processes, including Raman scattering, parametric oscillations and second, third and fourth harmonic generation. Such crystalline WGMRs have demonstrated a low threshold for nonlinear processes (down to tens of µW of pump power) that significantly reduces the power requirements. At the same time, the small volume of all optical elements that makes it less sensitive to environmental mechanical and thermal noises, and easy to integrate with any other optical system.

Here, we discuss a prospects of a source for bright squeezed light using second harmonic generation (SHG) in a crystalline WGMR. This squeezing mechanism in principle requires only one nonlinear cavity under SHG conditions to produce quadrature squeezed light in both fundamental frequency and second harmonics, significantly simplifying the experimental arrangements. The proof-of-principle demonstrations of SHG squeezing have been performed, but our calculations predict that using WGMR can significantly improve these results.

The manuscript is analyzed as follows. First, we derive analytical expression for quantum noise quadratures of optical fields at fundamental (pump) and second-harmonic frequencies. Then, we investigate the squeezing dependence on WGMR parameters, such as its quality factor and coupling efficiency. Finally, we report on experimental progress toward SHG squeezing generation.

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2. QUANTUM NOISE IN SECOND HARMONICS GENERATION PROCESS

During the second harmonic generation a pump beam of fundamental frequency $\omega$ propagates through a crystal, a second harmonic field is produced at frequency $\omega$. Physically, such conversion is possible due to nonlinear dependence of induced electric polarization $P(E)$ on the laser field amplitude $E$ inside the crystal:

$$P(E) = \chi^{(1)} E + \chi^{(2)} E^2 + \cdot$$

(1)

where $\chi^{(1)}$ and $\chi^{(2)}$ are linear and second-order nonlinear susceptibilities, correspondingly. In most nonlinear crystals, $\chi^{(2)}$ is a tensor that depends on the polarizations of the participating optical fields.

The nonlinear interaction between the optical fields inside a resonator is described by the following Hamiltonian:

$$H_{sys} = \hbar \omega_a a^\dagger a + \hbar \omega_b b^\dagger b + \frac{i}{2} \hbar \epsilon (a^\dagger b^\dagger - a b^\dagger).$$

(2)

Here, the intracavity fundamental and second-harmonic fields are $a$ and $b$, respectively, and $\epsilon$ is the normalized nonlinearity, proportional to the nonlinear susceptibility $\chi$, defined as:

$$\epsilon = \chi^{(2)} \frac{2 \pi \omega}{n} \sqrt{\frac{\hbar \omega}{2 \pi c V}}$$

(3)

where $n = n(\omega)$ is the refractive index at the fundamental frequency $\omega$ and $V$ is the mode volume.

To properly calculate the quantum noise of, for example, a fundamental pump field after intracavity second harmonic generation, we used a input-output formalism$^{26}$ to model a WGMR as a cavity with one input/output mirror with effective reflectivity proportional to the coupling efficiency, and one high reflector, corresponding to the optical losses inside the WGMR. The equations of motion for the quantized optical fields, obtained from the Hamiltonian (2), are given below:

$$\dot{a} = -i \omega_a a - \frac{1}{2} \gamma_a^{tot} a + \epsilon a^\dagger b + \sqrt{\gamma_a^{in}} a_{in} + \sqrt{\gamma_a^{out}} a_{a}
$$

$$\dot{b} = -i \omega_b b - \frac{1}{2} \gamma_b^{tot} b - \frac{1}{2} \epsilon a a + \sqrt{\gamma_b^{in}} b_{in} + \sqrt{\gamma_b^{out}} b_{b}$$

(4)

Here, the subscripts $a$ and $b$ indicate parameters for the fundamental and the second harmonics, correspondingly, and factors $\gamma$ describe the decay rate associated with internal losses of a WGMR ($\gamma^{in}$) and with coupling losses ($\gamma^{out}$), and ($\gamma^{tot} = \gamma^{in} + \gamma^{out}$). The first two terms of each equation correspond to propagation and linear absorption of each optical field inside a WGMR; the third and fourth source terms are the contribution of the nonlinear frequency conversion and of the coupled external optical field. Finally, the last terms take into account coupling of coherent vacuum modes $a_{a}, b_{b}$ into the system through optical losses.

We then solve Eqs.(4) decomposing each quantum field into a steady-state mean value (c-number) and the quantum fluctuation operator with zero mean values: $a = \langle a \rangle + \delta a, b = \langle b \rangle + \delta b$ ($\delta a = \langle a \rangle$, etc.) Then the time-independent average values for both fundamental ($\langle a \rangle$) and second-harmonics ($\langle b \rangle$) intracavity fields are governed by the following equations:

$$\frac{1}{2} \gamma_a^{tot} \delta a = \epsilon \delta b + \sqrt{\gamma_a^{in}} a_{in}$$

$$\frac{1}{2} \gamma_b^{tot} \delta b = -\frac{1}{2} \epsilon \delta a + \sqrt{\gamma_b^{in}} b_{in}$$

(5)

as $\langle a \rangle = \langle b \rangle = 0$.

The equations for quantum fluctuation operators are then become:

$$\delta a = -\frac{1}{2} \gamma_a^{tot} \delta a + \epsilon \delta b^\dagger + \epsilon \delta a^\dagger + \sqrt{\gamma_a^{in}} (\delta a_{in} + \sqrt{\gamma_a^{out}} \delta u_a)
$$

$$\delta b = -\frac{1}{2} \gamma_b^{tot} \delta b - \epsilon \delta a + \sqrt{\gamma_b^{in}} (\delta b_{in} + \sqrt{\gamma_b^{out}} \delta u_b)$$

(6)
These linear equations can be solved analytically in frequency domain by taking Fourier transform of corresponding operators, for example:

$$\delta \tilde{a}(\Omega) = \int_{-\infty}^{\infty} \delta a(t)e^{-i\Omega t} dt$$

Then it is convenient to write all operators compactly in the following vector notation:

$$\tilde{x}_c = \begin{pmatrix} \delta \tilde{a} \\ \delta \tilde{a}^\dagger \\ \delta \tilde{b} \\ \delta \tilde{b}^\dagger \end{pmatrix}, \quad \tilde{x}_\text{in} = \begin{pmatrix} \delta \tilde{a}_\text{in} \\ \delta \tilde{a}_\text{in}^\dagger \\ \delta \tilde{b}_\text{in} \\ \delta \tilde{b}_\text{in}^\dagger \end{pmatrix}, \quad \tilde{x}_u = \begin{pmatrix} \delta \tilde{u}_a \\ \delta \tilde{u}_a^\dagger \\ \delta \tilde{u}_b \\ \delta \tilde{u}_b^\dagger \end{pmatrix},$$

such that the fluctuation equations can be expressed in matrix form:

$$i\Omega \tilde{x}_c = M_c \tilde{x}_c + M_{\text{in}} \tilde{x}_\text{in} + M_u \tilde{x}_u$$

where the matrices are defined as:

$$M_c = \begin{pmatrix} -1/2\gamma_{\text{tot}}^a & \epsilon \tilde{b} & c\tilde{a}^* & 0 \\
\epsilon \tilde{b}^* & -1/2\gamma_{\text{tot}}^b & 0 & c\tilde{a} \\
-c\tilde{a} & 0 & -1/2\gamma_{\text{tot}}^b & 0 \\
0 & -c\tilde{a}^* & 0 & -1/2\gamma_{\text{tot}}^b \end{pmatrix}$$

$$M_{\text{in}} = \text{diag} \left( \sqrt{\gamma_{\text{in}}^a}, \sqrt{\gamma_{\text{in}}^b}, \sqrt{\gamma_{\text{in}}^u}, \sqrt{\gamma_{\text{in}}^u} \right)$$

$$M_u = \text{diag} \left( \sqrt{\gamma_{u}^a}, \sqrt{\gamma_{u}^b}, \sqrt{\gamma_{u}^u} \right)$$

Then intracavity fluctuations can be simply calculated:

$$\tilde{x}_c = (i\Omega I - M_c)^{-1} (M_{\text{in}} \tilde{x}_\text{in} + M_u \tilde{x}_u)$$

Since we are interested in quantum state of output fundamental field, we must take into account contributions for both partially reflected input field and outcoupled intracavity field:

$$\tilde{x}_o = \begin{pmatrix} \delta A_o \\ \delta A_o^\dagger \\ \delta B_o \\ \delta B_o^\dagger \end{pmatrix} = M_{\text{in}} \tilde{x}_c - \tilde{x}_\text{in}.$$ 

Thus, by combining Eqs.(12) and (13) the final expression for the output fields’ fluctuations are:

$$\tilde{x}_o = [M_{\text{in}} (i\Omega I - M_c)^{-1} M_{\text{in}} - I] \tilde{x}_\text{in} + M_{\text{in}} (i\Omega I - M_c)^{-1} M_u \tilde{x}_u$$

In practice the noise characterized by the variances (second momentum) of the quantum fluctuations of each optical field. The two orthogonal noise quadratures for, for example, fundamental field are defined as:

$$\delta A_1 = \delta A_o e^{i\phi} + \delta A_o^\dagger e^{-i\phi}, \quad \delta A_2 = -i \left( \delta A_o e^{i\phi} - \delta A_o^\dagger e^{-i\phi} \right),$$
where $\phi$ is the phase difference defined with respect to a local oscillator.\textsuperscript{1} The intensity fluctuations of an optical field are described by the $\delta A$ quadrature with $\phi = 0$, and the measured noise power is proportional to the quadrature variance:

$$\text{Var} (\delta A) = \langle |(\delta A_+ + \delta A_-^\dagger)|^2 \rangle$$ (17)

The variance of the coherent optical field is unity, and is independent of the quadrature phase $\phi$. If the variance of the output field becomes less than unity (for some quadrature phase), this optical field is said to be intensity squeezed. In second-harmonic generation squeezing is expected in intensity quadratures for both fundamental and SHG output optical fields, which is also confirmed by our numerical calculations.

3. MODELING THE QUANTUM NOISE IN A CRYSTALLINE WHISPERING GALLERY MODE RESONATOR

We now connect the general model, developed above, to the parameters of a realistic WGMR. For now we will assume that only one spatial mode is excited for each fundamental and second harmonic optical fields. Also, we consider the input pump optical fields $a_{in}$ to be in a coherent state, and the input second harmonic field $b_{in}$ to be a coherent vacuum ($\tilde{b}_{in} = 0$). As mentioned above, in this model the coupling efficiency of input laser field into WGMR is described by the value of $\gamma^{(i)}_{a,b}$ (in general, the coupling can be different for the two optical fields). All losses inside the WGMR are modelled by the high-reflector loss rate $\gamma^u$, which is connected to the $Q$-factor of a WGMR as $\gamma^u = \frac{\omega}{Q}$. In our theoretical calculations we assume the $Q$-factors in the $10^6 \sim 10^8$ range, that has been experimentally demonstrated\textsuperscript{13} for lithium niobate WGMRs.

The nonlinearity parameter $\epsilon$ is related to the nonlinear susceptibility $\chi^{(2)}$ by Eq.(3). Our experimental design uses type 1 ooe phase matching, thus $\epsilon_{ijk} \rightarrow \epsilon_{311} = \epsilon$. The corresponding susceptibility of lithium niobate is $\chi_{311} = -4.35 \text{ pm/V.}^{27}$

The important parameters governing the resulting variance are the ratio between the fundamental input loss rate and the total rate ($\frac{\Gamma^a}{\Gamma^e}$), the ratio between the total fundamental and second harmonic loss rate ($\frac{\Gamma^a}{\Gamma^b}$), and the input power. For example, Fig. 1 shows the calculated spectral dependence of the output fundamental field variance $V_{out}$ for a LiNbO$_3$ disk with various values of the fundamental frequency quality factor. Here we plot only the minimum noise quadrature variance (corresponding to intensity quadrature $\phi = 0$); the variance of the other is increased to preserve the Heisenberg uncertainty relation. The calculated variances clearly demonstrate the dramatic improvements in the expected value of squeezing at a higher-$Q$ resonator: while for $Q = 10^6$ maximum squeezing does not exceed 1 dB, up to 10 dB of squeezing is predicted for $Q = 10^8$. At the moment,
our model does not take into account any excess noise of a pump laser, which is known to overwhelm squeezing at low frequencies.\textsuperscript{1,23} however, this problem can be alleviated by using an ultra-stable solid-state laser with a linewidth $< 1 \text{ kHz}$.

\begin{figure}[h]
\centering
\includegraphics[width=0.7\textwidth]{figure2.png}
\caption{Squeezing in the fundamental intensity quadrature as a function of pump power. The other parameters are $Q = 10^8$, $\frac{\gamma_{ia}}{\Gamma_a} = 0.991$, $\frac{\Gamma_a}{\Gamma_b} = 0.5$.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=0.7\textwidth]{figure3.png}
\caption{Squeezing in the second harmonic intensity quadrature as a function of pump power. The other parameters are $Q = 10^8$, $\frac{\gamma_{ia}}{\Gamma_a} = 0.91$, $\frac{\Gamma_a}{\Gamma_b} = 0.5$.}
\end{figure}

Similar quantum noise reduction is expected in the output generated second harmonic field. It is important to note that bright squeezing (i.e., squeezed light with non-zero average power) is expected for both optical fields, since the quantum noise modification becomes possible only in the regime of significant nonlinear conversion (in which the changes in both fundamental and second harmonics amplitude become comparable). We model squeezing in the second harmonic using Eq. 14 to get the variance in Eq. 18. The variance of the second harmonic field has a dependence on $Q$-factor similar to the fundamental variance. We predict a maximum level of squeezing of $\sim 7 \text{ dB}$ with a $Q = 10^8$.

$$ Var(\delta B_1) = \langle |(\delta B_o + \delta B_o^\dagger)|^2 \rangle $$

(18)
Increase in quality factor also reduces the power requirements for the pump laser. Fig. 2 shows variance of the fundamental frequency output optical field as a function of input power for a WGMR with a $10^8$ $Q$-factor. It shows that squeezing is expected at low input power down to tens of $\mu$Ws, and optimal squeezing ($> 10$ dB) is predicted for laser power around 500 $\mu$W. This compares favorably to previous work, where $< 1$ dB of fundamental squeezing was obtained from 9 mW of pump power.\textsuperscript{23}

The noise reduction in the second harmonic field displays similar dependence on the input pump power, as shown in Fig. 3. The only difference is that maximum squeezing is predicted at a slightly higher input power, with similar $Q$-factor parameters.

4. PRELIMINARY EXPERIMENTAL RESULTS

Figure 4. Schematics of the experimental setup. The laser beam is focussed at the minimum separation point between a WGRM and a coupling diamond prism. The reflected laser beam and the generated second harmonic are separated using a dichroic mirror (DM) and measured using two separate photodiodes (PDs). Additional polarizer can be inserted to the SHG beam to further reduce the leakage of the main laser into this channel.

In experiments we have successfully produced WGMR disks out of 3-4% MgO:LiNbO$_3$, with non-critical phase matched second harmonic generation at a temperature around 89°C. The diameter of the disk used in the experiment is 7 mm, and the thickness is 1.0 mm. The experimental setup is shown in Fig. 4. The output of the 1064 nm laser is coupled into a WGMR disk using a diamond prism with anti-reflection coating for both 1064 nm and 532 nm. The separation between the disk and the prism (and thus the strings of the coupling) is controlled by a spring-loaded microscrew. Fig. 5 shows an example of narrow resonance feature in the intensity of the reflected laser beam as its frequency scans across the WGMR mode. From the width of this resonance we can estimate the quality factor of the disk at the fundamental mode to be $Q = 5 \cdot 10^7$. This value has been measured at relatively large separation between the prism and the disk; for most experimental conditions this gap is smaller, leading to stronger coupling and correspondingly lower $Q$-factor.

The temperature of the disk is actively stabilized at wide range around 89°C for best phase-matching conditions. The laser frequency can either be scanned or locked to the desired frequency to match the frequency of a particular mode in the disk. The output fundamental and second harmonic have orthogonal polarizations, and exit the prism at slightly different angles due to the optical dispersion of diamond; using a combination of spatial, spectral and polarization filters they can be well separated. Fig. 6 shows an example of observed second harmonic generation in WGMR.

It is clear that the second harmonic output is detected only when the laser is in resonance with some of the fundamental modes, and the amplitude of the second harmonic is not necessarily proportional to the coupling efficiency of the fundamental mode. This is because the SHG efficiency depends not only on the quality factors of the two optical fields, but also on the geometrical overlap of their modes inside the resonator. The analytical expression for the efficiency has been derived in Ref.:\textsuperscript{12}

$$\frac{P_{\text{out}}^{(2\omega)}}{P_{\text{in}}^{(\omega)}} = \frac{6}{S} \left[ \sqrt[3]{1 + S + \sqrt{S(1 + S)}} + \sqrt[3]{1 + S - \sqrt{S(1 + S)}} - 2 \right]^2, \quad (19)$$

where $P_{\text{in}}^{(\omega)}$ and $P_{\text{out}}^{(2\omega)}$ are the powers of the input fundamental and output second harmonics optical fields, correspondingly, and $S = 54 P_{\text{in}}^{(\omega)}/P_0$ is the saturation parameter. The saturation power $P_0$ is determined by the
Figure 5. Output intensity (in arb. units) of the pump laser. Narrow resonance is due to destructive interference between the directly reflected light and the light coupled into and out of the disk.

Figure 6. An example of fundamental (i) and second-harmonic (ii) optical fields outputs (the arbitrary units are different for the two fields). The temperature of the disk is \( T = 89^\circ C \), and the laser power before the disk is 63 mW.

Refractive indices of the disk material at different optical frequencies \( n(\omega) \) and \( n(2\omega) \), nonlinear susceptibility \( \chi^{(2)} \), mode volume for each field \( (V(\omega), 2\omega)) \) and their overlap \( (V^{\text{over}}) \), as well as their quality factors:

\[
P_0 = \frac{(n(\omega))^4(n(2\omega))^2}{64\pi^3 \left[\chi^{(2)}\right]^2} \left( \frac{V(\omega)}{Q(\omega)V^{\text{over}}} \right)^2 \omega V^{(2\omega)} \left( \frac{Q^{(2\omega)}}{Q(\omega)} \right),
\]

(20)

Fig. 7 shows measured SHG conversion efficiency as a function of the pump field power for two different separations between the disk and the prism. Varying the disk-prism separation increased the conversion efficiency, as the \( Q \) increased by a factor of 1.55 from \( Q = 4.6 \cdot 10^6 \) to \( Q = 7.0 \cdot 10^6 \). Since we do not have sufficient information about individual parameters in Eq.(20), we extract the value of the saturation parameters by fitting the SHG efficiency measured experimentally using Eq.(19). The obtained values are \( P_0 = 900 \) W for the measured \( Q = 7.0 \cdot 10^6 \), and \( P_0 = 2200 \) W for the measured \( Q = 4.6 \cdot 10^6 \). The obtained ratio between the two saturation powers is consistent with \( P_0 \sim \frac{1}{Q^2} \) predicted by Eq.(20). This shows that our conversion efficiency is currently...
Figure 7. Measured SHG conversion efficiency for two different loaded Q-factors. The values of Q are extracted from the WGMR mode linewidth. The lines show fits for the saturation power. A higher Q reduces the saturation power, and improves the conversion efficiency.

limited by the low Q-factor. The reduction of Q by an order of magnitude from originally measured $5 \times 10^7$ may be due to some deterioration of the surface of the disk during the measurements, or due to photorefractive damage.

5. CONCLUSIONS

Here we theoretically analyze the perspective of bright squeezing generation via second harmonic nonlinear conversion in LiNbO$_3$ WGMR disks. The main advantage of WGMRs compared to conventional mirror cavities is significant reduction in required pump power and high conversion efficiency. Our analysis predicts significant squeezing (up to $8-10$ dB) in intensity fluctuations of both fundamental and second-harmonic optical fields. However, high quality factor ($Q \geq 10^8$) is required in order to realize significant squeezing in either field. Our preliminary experimental results show that such high values may be within our reach (as $Q = 5 \times 10^7$ has been observed in a newly-polished disk). In addition, we should be able to improve it further by annealing after polishing. We have tested that annealing a similar disk at 600°F for 24 hours improved its Q by a factor of about 2 (from $Q = 3 \times 10^6$ to $7 \times 10^6$). We expect to use this method to improve our Q-factor from $5 \times 10^7$ to over $10^8$. We can then use the separation to control the coupling rate ($\gamma_i$) to tune to the optimal parameters for squeezing.

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