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# Quasi-phase-matched nonlinear optical frequency conversion in on-chip whispering galleries: supplementary material

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## 1. FABRICATION

The fabrication of pp-LNoQ substrates is done basically in four steps: domain inversion of a bulk LN chip by field-assisted poling; surface cleaning and functionalizing of the LN chip and of an $\alpha$-quartz substrate; thermal bonding of the LN on quartz; and lapping and polishing of LN to the final thin film.

We write calligraphically a linear grating with 23 $\mu$m period length and approximately 5 $\mu$m domain width into a 300-$\mu$m-thick 5-mol.%-MgO-doped optically-polished $z$-cut-LN chip (chip size: $17 \times 18\,\mathrm{mm}^2$). A grating with 50 % duty cycle and a period length of 10 $\mu$m would be even better, however due to our fabrication setup this is hard to achieve. That's why we used a higher order grating structure, which allows us to use larger period lengths [1]. A 150-nm-thick chromium layer is sputtered on the +$z$-side of the crystal and serves as the backelectrode. We write the domains with a tungsten carbide tip moving along the $y$-crystal axis. While writing the domains, the applied high voltage is controlled in a way that a constant poling current of 30 nA flows, and the chip temperature is set to 150 °C. After poling the chromium layer is removed by selective wet-chemical etching.

Next we clean the periodically-poled sample and also a z-cut-$\alpha$-quartz chip with acetone and isopropanol, followed by a further cleaning and surface functionalization procedure. It basically consists of three steps: ultra-sonic-assisted rinsing of the samples for 10 minutes in a $H_2SO_4/H_2O_2$ (1:1) solution, then in a $HCl/H_2O_2/H_2O$ (1:1:5) solution and finally in a $H_2O_2/NH_4OH/H_2O$ (1:1:5) solution. Each step is followed by rinsing the samples with deionized water for 5 minutes, and we dry the sample with nitrogen in the end.

After bonding of the LN chip on the quartz substrate, we enhance the bonding force by tempering. We heat the sample on a hot plate with 4 K/min to 325 °C and temper the sample for 5 hours.

Subsequently, we reduce the thickness of the periodically-poled LN to 2 $\mu$m by lapping and polishing with a standard wafer-polishing machine (*PM5, Logitech*). This is done in three steps. First we use a solution with 9-$\mu$m-$Al_2O_3$ particles and a lapping pressure of 1 N/cm$^2$ to lap the LN down to a thickness of 12 $\mu$m. Next, we reduce the LN-film thickness to approximately 6 $\mu$m, deploying a solution with 1-$\mu$m-ceroxide particles and a lapping pressure of 2.5 N/cm$^2$. Last, to achieve substrates with optical grade surface, we use SF1 Syton (*Logitech*) and a polishing pressure of 2.5 N/cm$^2$ to polish the LN-film down to the final thickness of 2 $\mu$m. The film-thickness measurements are done with a profilometer. The WGRs are structured in the middle area ($6 \times 8\,\mathrm{mm}^2$) where we have a film thickness homogeneity of $\pm$250 nm.

## 2. CALCULATION OF EFFECTIVE NONLINEAR-OPTICAL COEFFICIENT

The effective nonlinear optical coefficient is given by $d_{eff} = |S_M|d_{\max}$ with the unitless coefficient $S_M$. In this section we explain how $S_M$ can be calculated according to [2] for three different cases: First, a single domain resonator for birefringent-phase matching (eoo), second, a radially poled resonator for quasi-phase matching (eee) and last, a linearly poled resonator for quasi-phase matching (ooo) (Fig. S1). For one roundtrip in the WGR the spatial variation of the nonlinear-optical coefficient can be described by $d(\varphi) = S(\varphi)d_{\max}$ with the unitless function $S(\varphi)$ varying between $\pm 1$. $S(\varphi)$ can be influenced by two components: A variation $S_{\mathrm{QPM}}$ due to the domain pattern and a variation $S_{\mathrm{crystal}}$ due to the crystallographic orientation. $S(\varphi)$ is given by $S(\varphi) = S_{\mathrm{QPM}}(\varphi) \times S_{\mathrm{crystal}}(\varphi)$.

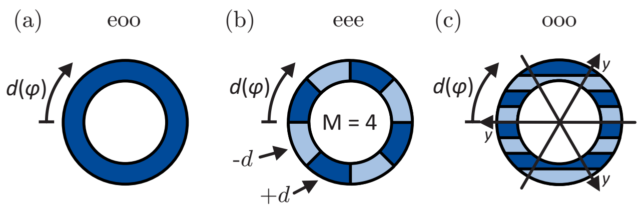


**Fig. S1.** Sketches of the three different cases, discussed in this section. (a) Single-domain WGR with birefringent phase matching. (b) Radially poled FGR with (eee)-quasi-phase matching. (c) Linear poled FGR with (ooo)-quasi-phase matching.

The first case with the birefringent-phase-matched single-domain WGR is simple. No domain patten is applied ($S_{\mathrm{QPM}} = 1$) and the nonlinear optical coefficients $d_{311} = d_{322}$ are relevant so that there is no $\varphi$-dependence of $d$ ($S_{\mathrm{crystal}} = 1$, Fig. S2).

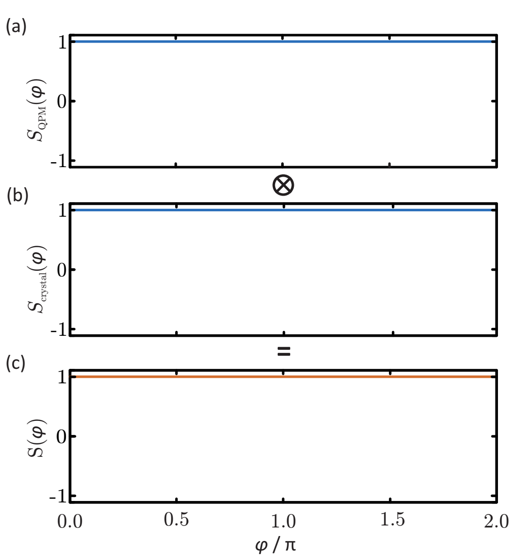


**Fig. S2.** Case one: Single-domain WGR with birefringent-phase matching. (a) No variation of the nonlinear optical coefficient along the WGR perimeter $d_{\mathrm{eff}}(\varphi)$ by $S_{\mathrm{QPM}}$ since there is no domain pattern applied. (b) No variation of $d_{\mathrm{eff}}(\varphi)$ by $S_{\mathrm{crystal}}$ since only $d_{311} = d_{322}$ is employed in birefringent-phase matching. (c) Finally variation of $d_{\mathrm{eff}}(\varphi)$ described by $S(\varphi) = S_{\mathrm{QPM}}(\varphi) \times S_{\mathrm{crystal}}(\varphi)$ which is constant in this case.

In the second example $d$ is inverted every $\pi/4$, which is described by $S_{\mathrm{QPM}}$ like it is shown in Fig. S3 (a). Since only e-polarized light is involved the nonlinear coefficient $d_{333}$ is deployed, which means that $S_{\mathrm{crystal}} = 1$ and thus $S(\varphi) = S_{\mathrm{QPM}}$ Fig. S3.

The third case is the most complex one. Here, $d(\varphi)$ is effected by the linear domain pattern which is described by $S_{\mathrm{QPM}}$ in Fig. S4 (a). We neglect the mode extension in radial direction and thus that different radial parts of the modes cross the domain walls at slightly different angles $\varphi$. Furthermore, only o-polarized light is involved, which means that the nonlinear coefficients $d_{111} = 0$ and $d_{222} = 2.1\,\mathrm{pm/V}$ are of relevance. The nonlinear coefficient varies as $S_{\mathrm{crystal}}(\varphi) = \cos(3\varphi)$ [3] (Fig. S4 (b)) which results in a final variation $S(\varphi)$ shown in Fig. S4 (c).

The coefficients $S_M$ are calculated by a Fourier-transformation of the respective $S(\varphi)$ [2]

$$d_{\mathrm{eff}}(M) = \frac{d_{max}}{2\pi} \left| \int_0^{2\pi} e^{iM\varphi} S(\varphi) d\varphi \right| = |S_M| d_{\max}. \quad \text{(S1)}$$

The corresponding $|S_M|$ to the three cases are shown in Fig. S5. In the first case there is only one coefficient different from zero with $S_{M=0} = 1$. The radially poled domain pattern provides a coefficient different from zero with $S_{M=4} = 2/\pi$ and at higher orders $M = (2n-1) \times 4$ with $n \in \mathbb{N}$. This means that we can achieve with a radially poled domain pattern a $(2\,d_{333}/(\pi/d_{311}))^2 \approx 14$-times higher efficiency with $d_{311} = 3.4\,\mathrm{pm/V}$ and $d_{333} = 20.3\,\mathrm{pm/V}$. In the case with the linear domain pattern there is a $S_M \neq 0$ for all $M$. This means that there are many possibilities to compensate for different phase-mismatch $M$ to achieve quasi-phase matching. However since the coefficients $|S_M|$ are relatively small this leads to a smaller $d_{\mathrm{eff}}$. According to this procedure the coefficients $S_M$ in Fig. (5c) of the main part are calculated for a domain pattern with 23 $\mu$m period length and 5 $\mu$m domain width for (ooo)-polarization.

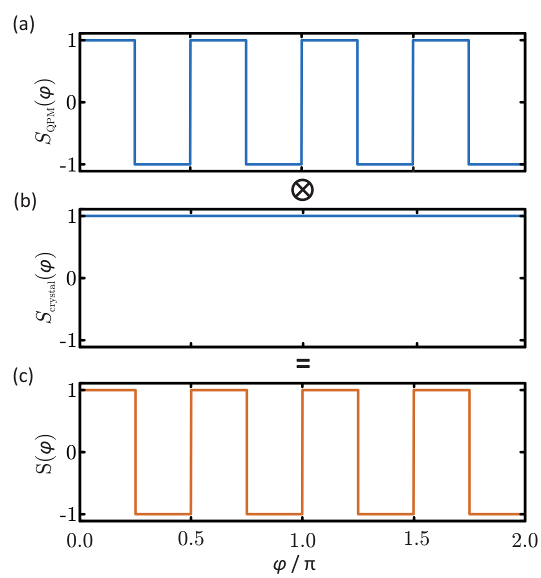


**Fig. S3.** Case two: Radially poled WGR with quasi-phase matching (eee). (a) Variation of the nonlinear optical coefficient along the WGR perimeter $d_{\mathrm{eff}}(\varphi)$ described by $S_{\mathrm{QPM}}$. (b) No variation of $d_{\mathrm{eff}}(\varphi)$ by $S_{\mathrm{crystal}}$ since only $d_{333}$ is employed (c) Finally variation of $d_{\mathrm{eff}}(\varphi)$ described by $S(\varphi) = S_{\mathrm{QPM}}(\varphi) \times S_{\mathrm{crystal}}(\varphi)$.

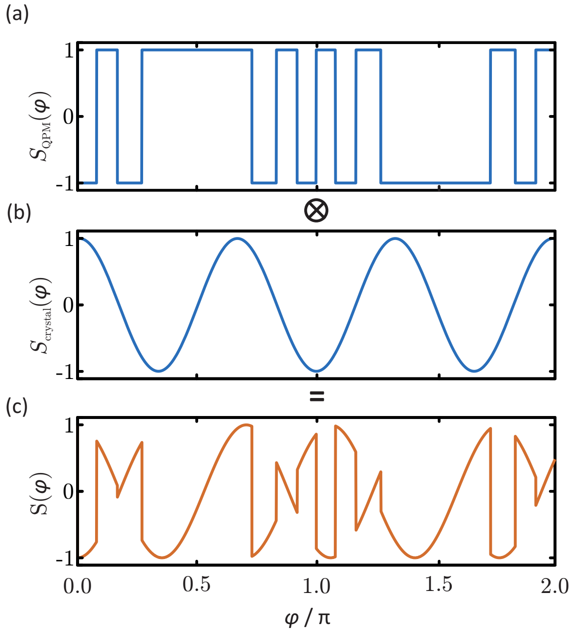


**Fig. S4.** Case three: Linearly poled WGR with quasi-phase matching (ooo). (a) Variation of the nonlinear optical coefficient along the WGR perimeter $d_{\mathrm{eff}}(\varphi)$ described by $S_{\mathrm{QPM}}$. (b) Variation of $d_{\mathrm{eff}}(\varphi)$ in case of o-polarized light due to the fact, that $d_{111} = 0$ and $d_{222} \neq 0$. (c) Finally variation of $d_{\mathrm{eff}}(\varphi)$ described by $S(\varphi) = S_{\mathrm{QPM}}(\varphi) \times S_{\mathrm{crystal}}(\varphi)$.

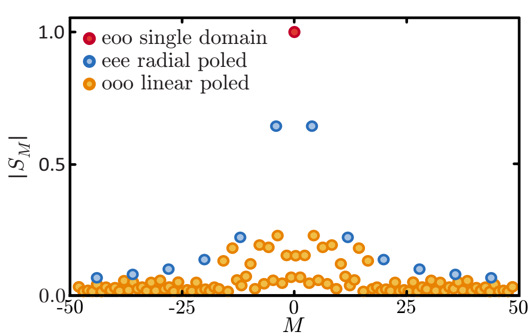


**Fig. S5.** Coefficients $|S_M|$ for the three different cases discussed above.

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