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Wide-range cyclic phase matching and second harmonic generation in whispering gallery resonators

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We report on a wide-range efficient method for optical second harmonic generation based on a whispering gallery mode resonator made from crystalline beta barium borate. In this single resonator, we were able to generate second harmonic fields for four different pump wavelengths ranging from the infrared (1557 nm) to the visible (634 nm) regime. The highest conversion efficiencies achieved in this whispering gallery mode resonator are as high as $4.6\% (\text{mW})^{-1}$. This conversion process is based on type-I phase matching with continuously varying optical axis orientation in an xy-cut configuration of the resonator. In such a geometry, the second harmonic whispering gallery mode experiences an oscillatory modulation of the refractive index. This enables wide-range cyclic phase matching along the circumference of the disk resonator. © 2013 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4827538>]

In nonlinear optical processes, birefringent phase matching offers great flexibility in wavelength selection.^{1–3} Type-I second harmonic (SH) generation in beta barium borate (BBO), for instance, is possible for SH wavelengths ranging from $0.2 \mu\text{m}$ in the ultraviolet up to around $1.5 \mu\text{m}$ in the infrared.⁴ The general limits of this type of phase-matching range are given by the birefringence and the transparency window of the nonlinear material used. In a bulk crystal, the phase matching condition for a given wavelength conversion can be achieved via tuning the angle of incidence of the pump beam. For uniaxial crystals, this conversion range can be addressed within a single crystal plane that includes the optic axis.

Whispering gallery mode (WGM) resonators demonstrated their strength in nonlinear optics in a variety of processes.^{5–8} This strength arises from the high quality factor along with the small mode volume of WGMs.⁹ Phase matching in the WGM resonator has been achieved through natural geometric dispersion and quasi-phase-matching.^{7,10–12} Experiments in uniaxial crystalline WGM resonators were performed exclusively in a z-cut geometry, where the optic axis and the resonator symmetry axis coincided. So the optic axis was not included in the plane of propagation, and the broad birefringence phase matching was not possible. On the other hand, in the xy-cut geometry where the optic axis is in the plane of the resonator disk, the circularly propagating light has a continuously varying angle relative to the optic axis in the plane of the crystal, offering the possibility of efficient conversion in the entire birefringence phase matching range. However, it was not clear whether these resonators are still able to exhibit high Q whispering gallery modes. Recent experiments indeed showed that high Q whispering gallery modes exist in a non-z-cut configuration,¹³ which opens up the possibility of wide-range phase-matching.

In this Letter, we demonstrate a WGM resonator made from highly birefringent crystalline BBO that generated

second harmonic for pump wavelengths between 634 nm and 1557 nm. This was achieved by aligning the optic axis of BBO perpendicular to the symmetry axis of the WGM resonator as shown in Fig. 1(a). Conversion efficiencies for second harmonic generation of nearly 5% per mW pump power were observed. A theoretical model was developed for describing this *cyclic phase matching* (CPM) of light fields in the WGM resonator based on the type-I phase-matching approach.

A whispering gallery mode resonator made from a homogeneous dielectric supports two orthogonally polarized mode families, the transverse electric (TE) and magnetic (TM) modes as illustrated in Fig. 1(a). To a good approximation, the spatial distribution of the electric field $u(r, \vartheta, \varphi)$ for a TM mode and analogously the distribution for the magnetic field for a TE mode are described in spherical coordinates by

$$\vec{r} \cdot \vec{u}(r, \vartheta, \varphi) = C_m^{q,\ell}(r, \vartheta) e^{im\varphi}, \quad (1)$$

with $C_m^{q,\ell}(r, \vartheta)$ describing the radial and polar fields distribution and the radial and polar modes numbers q and ℓ . Azimuthally, the field is described by a complex exponential,

FIG. 1. (a) Whispering gallery mode resonator in xy-cut geometry. The optic axis is in the plane of disk resonator. The two polarisation mode families TE and TM are depicted in the side view. (b) Refractive index for both mode families along the circumference for 1557 nm and 779 nm. Collinear birefringent phase matching is achieved, when the refractive indices are equal.

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which reflects the rotational symmetry of the resonator. The three mode numbers are thereby related to the wavelength of the mode via the dispersion relation.¹⁴

For phase-matching, momentum conservation in bulk material transforms to the conservations of angular momentum in WGM resonators, expressed via selection rules on the mode numbers of the modes involved.¹⁵ In particular, they require that the azimuthal mode number m has to be conserved.

In a WGM resonator made from a uniaxial crystal in the z -cut geometry, the rotational symmetry is preserved and both mode families exist. The TM and TE mode families experience the ordinary and extraordinary refractive index n_o and n_e , respectively. This geometry was used in sub- and second harmonic generation.^{7,10–12,16–21}

A more interesting but complex situation arises when the WGM resonator axis lies in the (xy) crystallographic plane (thus the xy -cut geometry), as illustrated in Fig. 1(a). In this case, the resonator supports two mode families in general, with TE modes now experiencing the ordinary refractive index. The TM modes, however, experience an oscillation in the refractive index $n(\varphi)$ along the circumference between the ordinary and extraordinary values of the crystal. This situation is depicted in Fig. 1(b) for BBO at 1557 nm. Considering possible type-I second harmonic generation in BBO, the index variation of a TM mode at 779 nm is also presented. We find four regions where the indices of a TE polarized pump mode at 1557 nm and its harmonic TM mode are equal. These are four discrete regions where phase matching occurs. Since the circulating fields in the resonator repeat these phase matching in every round trip, we refer to this birefringent phase matching in a resonator as CPM.

From a theoretical point of view, as the xy -cut geometry still exhibits a periodicity in the azimuthal or φ direction, hence the electric field of both mode families can still be expressed with complex valued exponential functions with adapted polar and radial functions. To account for the oscillatory behaviour of the refractive index of TM modes, the electric field of TM modes has to be described by a superposition of complex exponential functions with different azimuthal mode numbers. Thus, there is a variety of azimuthal modes available for phase matching, increasing the bandwidth of wavelength for phase matching.

We can model Type I second harmonic generation in an xy -cut BBO resonator numerically, by evaluating the SH field evolution for a single round trip because of the cyclic resonant wave propagation. For further simplicity, we unfolded the circular geometry to a linear propagation along z of both fields along a 7 mm long nonlinear medium, which corresponds to the WGM resonator circumference. The second harmonic field E_2 is described in general by

$$\frac{dE_2}{dz} = \frac{2ik_2}{n_2^2} d_{\text{eff}}(z) E_1^2 e^{i \int_0^z \Delta k dz}, \quad (2)$$

where $\Delta k = 2k_1 - k_2$ represents the difference between the fundamental wave vector $k_1 = 2\pi n_1/\lambda_1$ and that of the second harmonic field $k_2 = 2\pi n_2/\lambda_2$, with their corresponding wavelengths $\lambda_1 = 2\lambda_2$. For this simulation, the refractive indices for both fields were determined from the WGM

resonator geometry: a constant one n_1 for the TE pump, and an oscillating function n_2 such as shown in Fig. 1(b) for the TM SH field. We further assumed an undepleted pump. The variation of the effective nonlinear coefficient $d_{\text{eff}}(z)$ of BBO in this geometry along the circumference is included in our model. The effect of the oscillation of the nonlinear coefficient in WGM resonators for quasi-phase matching was already studied theoretically and experimentally in GaAs.^{12,22–24} So does the similar use of orientation-dependent third-order nonlinearity in silicon nanowires.^{25,26} In our case, however, the birefringence dominates the phase matching and mismatching.

The calculated conversion efficiency normalized to a perfect phase matching situation is plotted for different wavelengths in Fig. 2(a). As predicted, there exists a broad range of phase matching. The overall efficiency of CPM is smaller than that for perfect phase matching. This however can be compensated by the high quality factors available in WGM resonators. The conversion efficiency shows an oscillatory behaviour with two distinct periods. These arise from interference between four phase-matched locations along the circumference, one pair closer together than the other (see Fig. 1(b)), similarly to that reported in Ref. 27. The trajectory of the complex harmonic field along the propagation direction explains this interference. This field evolution is plotted versus the index mismatch $\Delta n(\varphi) = n_o - n(\theta)$ for constructive and destructive interference situations in Figs. 2(b) and 2(c). For large Δn , the field is locally spiralling in the complex plane. When $\Delta n = 0$, there is a large gain for the field. In the case of constructive interference in Fig. 2(b), the individual increments of the field amplitude add in phase, while in the case of destructive interference in Fig. 2(c), they tend to compensate one another.

The conversion profile in Fig. 2(a) is determined by the refractive indices and the resonator size. The tuning of the conversion maxima and minima by the resonator size can be seen in Fig. 2(a) (blue) for a radius variation of 0.2%. The refractive index can also be changed by the thermo-refractive and electro-optic effect. It can be changed by choosing different resonator modes thus using the modal dispersion at the same time. It should also be noted that continuous tuning of SHG over a wide frequency range can be achieved by a combination of temperature tuning and mechanical tuning (e.g., by a small deformation of the resonator) for maintaining the doubly resonant condition.

For the experimental demonstration of CPM SHG, we fabricated an xy -cut WGM resonator from a crystalline BBO substrate. The resonator rim was polished into a spheroidal shape of optical grade. The disk has a diameter of about 1.82 mm and a few hundred microns thickness. The BBO resonator is mounted on a post with active temperature control close to a movable sapphire prism, which is used as an evanescent coupler. This setup is illustrated in Fig. 3(a). Four tunable laser sources at 634, 870, 974, and 1557 nm are used as fundamental pump sources. The TE polarized pump beam is focused onto the backside of the prism for evanescently coupling to the WGM resonators. The output beam from the prism is collimated again with the pump and harmonic fields spectrally separated using a dichroic mirror. A bandpass filter is placed in front of the second harmonic

FIG. 2. (a) Conversion efficiency of the second harmonic field for different wavelengths from the visible to the infra-red regime. (b) and (c) Harmonic field gain in the complex plane, plotted against the refractive index mismatch between fundamental and harmonic fields: (b) constructive interference. (c) destructive interference (both situations are indicated in Figure (a)).

detector to further reduce the pump light. Both fields are monitored simultaneously with two detectors.

The linewidths of the resonances of the BBO WGM resonator were measured at all wavelengths by scanning the laser frequency and observing resonances in the pump intensity. From these linewidth measurements, we inferred the corresponding Q factors at the pump wavelengths. We found the Q factors in the order of 10^7 for both TE and TM modes, which confirm the existence of whispering gallery modes also in the xy-cut geometry. In addition, we measured carefully the free spectral range of the TE and TM WGMs at 1557 nm.²⁸ We find the average refractive indices of $(1.59) \pm 0.01$ and (1.65 ± 0.01) , respectively. These values match with the

FIG. 4. Conversion efficiency of the harmonic field versus pump power for a pump wavelength of 634 nm, showing a linear dependence. Inset: Frequency sweep of the pump field, showing several whispering gallery modes. One of the modes is phase matched with a second harmonic mode.

average value of 1.587 for the TE whispering gallery mode and the ordinary index of refraction of 1.647 for the TM mode in Fig. 1. Experimentally, we also observed that the required TM excitation angle at the prism changed dramatically when changing the disk coupling position along its circumference, consistent with a spatially varying refractive index of TM modes.²⁹

For SHG, we pumped the WGM resonator with a few milliwatts of power with the four mentioned laser sources while over-coupled the resonator with a fixed gap distance between the WGM resonator and prism. The pump laser frequency was scanned over several modes to find phase matching. The inset of Fig. 4 shows an example of such a frequency scan at 634 nm. When CPM was satisfied at the doubly resonant condition, a harmonic signal was observed. We confirm the SHG linearity of the conversion efficiency with the in-coupled pump power as shown in Fig. 4. Here, the pump power was corrected for the in-coupling contrast, giving the in-coupled pump power. We observed second harmonic generation for all four pump laser sources in the same BBO resonator. Figure 3(b) shows the images of the second harmonic output beams observed on a paper card inserted behind the dichroic mirror or band pass filter. The violet and blue-green correspond to the second harmonic signals at 435 nm and 487 nm. The blue and red spots are related to

FIG. 3. (a) Sketch and photograph of the experimental setup. The sketch illustrates the polarization of the pump and harmonic field. The wavelength separation is implemented with a dichroic mirror. (b) True color images of the second harmonic beam profiles generated from different pump wavelengths: 974 nm, 870 nm, and 634 nm, from left to right, respectively.

TABLE I. Summary of harmonic generation for four different pump wavelengths: incoupled pump power, second harmonic power, loaded Q-factor of the pump mode, and conversion efficiency per mW incoupled pump power.

Pump wavelength (nm)	Pump power (mW)	Harmonic power (μ W)	Pump Q-factor (10^6)	Conversion efficiency (%/mW)
1557	1.1	0.76	3	0.063
974	0.25	2.9	7	4.6
870	0.87	13.5	10	1.8
634	0.42	1.3	4	0.74

luminescence from 317 nm on the paper card and the residual pump laser at 634 nm, respectively. The spatial separation between blue and red is due to the dispersions in the WGMs in the resonator and the coupling prism. The results of the SHG measurements are summarized in Table I. Thermal tuning was used to optimize the conversion efficiency at each wavelength. The large variation of the conversion efficiency achieved at the different wavelengths in the single resonator corresponds to the efficiency variation shown in Fig. 2(a). Compared with previously reported cavity-enhanced SHG systems at ultraviolet (UV) wavelengths,^{30,31} the demonstrated overall efficiencies of CPM are comparable and higher. If one compares the compact and wide wavelength range WGM resonator device to a comparably small single-pass doubling setup,³² the improvement factor is of several orders of magnitude.

The polarization of the outgoing second harmonic beam is confirmed to be along the expected TM direction, i.e., in the plane of the disk resonator. In fact, by rotating the WGM resonator and therefore changing the local optical axis angle at the coupling point, we observed a periodic angular variation of the SHG output beam spot due to the modulation of the refractive index of a TM mode.²⁹

For further confirmation of the cyclic type-I phase matching, we also set the pump polarization in the TM direction, and we could not observe any second harmonic signal. In addition, we investigated WGM resonators made from z-cut BBO and x-cut quartz. In both cases, there are no theoretical phase matching regions found between TE pump and TM second harmonic. We were unable to observe any second harmonic signals under similar conditions in the experiments.

In summary, we have theoretically predicted and experimentally confirmed birefringent Type-I phase matching in a crystalline WGM resonator with its optic axis parallel to the resonator plane. With a single resonator made from BBO, this cyclic phase matching efficiently generated second harmonic light from 779 nm down to 317 nm in UV, i.e., over more than an octave from the ultraviolet through the whole visible range. This also demonstrates a practical way for continuous wave (CW) second harmonic generation into the UV regime in a WGM resonator. In principle, this phase matching technique covers the entire birefringent phase matching range of the nonlinear crystal used. This

wide-range cyclic phase matching technique may open many possibilities for nonlinear frequency conversion in WGM resonators including micro CW laser sources into deep UV by SHG and mid-IR by parametric down conversion.

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