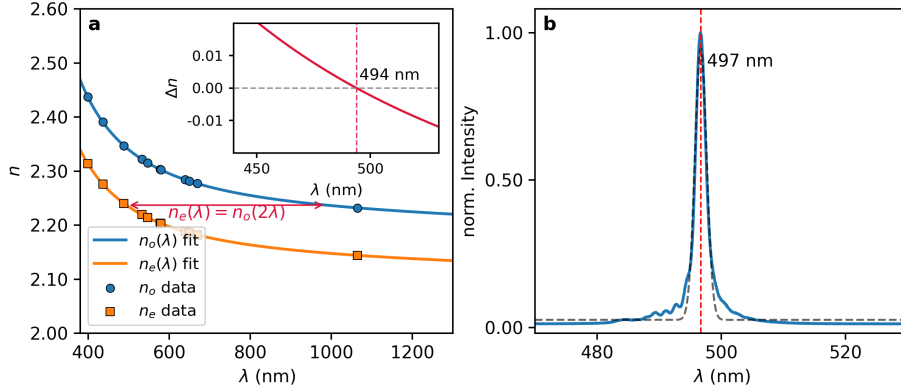


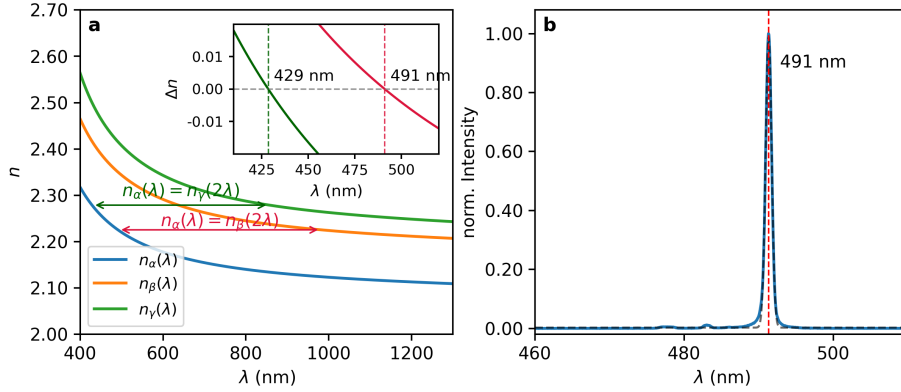
## Supplementary Information

### Self-selected phase-matched second harmonic generation in nonlinear optical materials: from phenomenon to applications

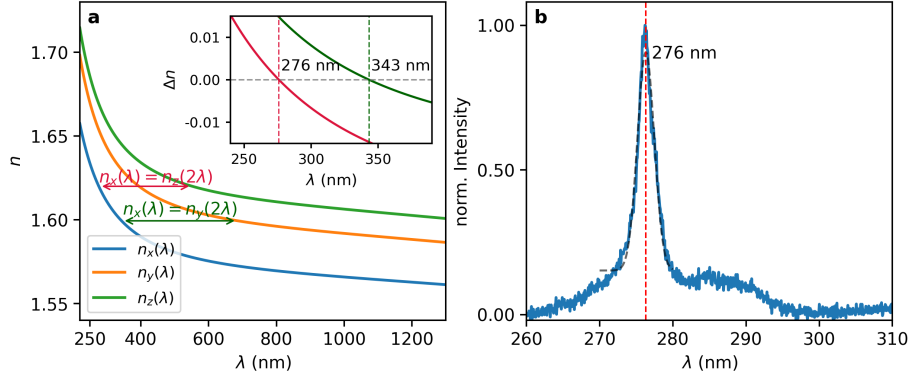
Niklas Dömer, Tobias Hehemann, Felix Sauerwein, Sebastian Inckemann, Steffen Ganschow, and Mirco Imlau



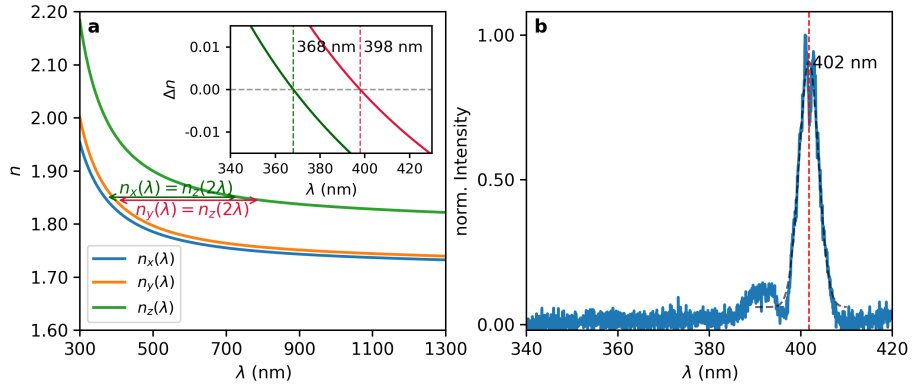
**Fig. S1.** Measurement on 1.3 mol% Mg-doped stoichiometric x-cut LN (Mg:sLN). The sample was supplied by Oxide Corporation and had a thickness of approximately 2 mm. a: Ordinary and extraordinary refractive indices,  $n_o$  and  $n_e$ , of Mg:sLN. Symbols show refractive-index data reported by Shoji *et al.* [1], while solid lines are fitted Sellmeier equations. The inset shows the corresponding noncritical phase-matching condition,  $n_e(\lambda) = n_o(2\lambda)$ , with  $\lambda$  denoting the SHG wavelength. b: Measured SHG spectrum obtained with the OPA centered at 980 nm. The dashed gray line shows a Gaussian fit to the SHG peak, and the red vertical line marks the fitted peak center.



**Fig. S2.** Supplementary measurement on a 2.5 mm-thick potassium niobate ( $\text{KNbO}_3$ ) crystal supplied by SurfaceNet with light propagating along crystallographic  $\gamma$  axis following the axis convention of Zysset *et al.* [2]. a: Principal refractive indices  $n_\alpha$ ,  $n_\beta$ , and  $n_\gamma$  of orthorhombic  $\text{KNbO}_3$  calculated from the 22 °C Sellmeier coefficients reported by Zysset *et al.* [2]. The inset shows the corresponding phase-matching conditions  $n_\alpha(\lambda) = n_\beta(2\lambda)$  (red) and  $n_\alpha(\lambda) = n_\gamma(2\lambda)$  (green), where  $\lambda$  denotes the SHG wavelength. b: Measured SHG spectrum obtained under excitation at 970 nm. The dashed gray line shows a Gaussian fit to the SHG peak, and the red vertical line marks the fitted peak center.



**Fig. S3.** Supplementary measurement on a 3 mm-thick  $y$ -cut lithium triborate (LBO) crystal. a: Principal refractive indices  $n_x$ ,  $n_y$ , and  $n_z$  of LBO calculated from Sellmeier equations [3]. The inset shows the corresponding phase-matching conditions  $n_x(\lambda) = n_z(2\lambda)$  (red) and  $n_x(\lambda) = n_y(2\lambda)$  (green), where  $\lambda$  denotes the SHG wavelength. b: Measured SHG spectrum obtained under excitation at 580 nm. The dashed gray line shows a Gaussian fit to the SHG peak performed only in the range from 270 to 280 nm, and the red vertical line marks the fitted peak center.



**Fig. S4.** Supplementary measurement on a 1 mm-thick  $x$ -cut KTP crystal supplied by Castech Inc. a: Principal refractive indices  $n_x$ ,  $n_y$ , and  $n_z$  of KTP calculated from Sellmeier equations [4]. The inset shows the corresponding phase-matching conditions  $n_x(\lambda) = n_z(2\lambda)$  (green) and  $n_y(\lambda) = n_z(2\lambda)$  (red), where  $\lambda$  denotes the SHG wavelength. b: Measured SHG spectrum obtained under excitation at 780 nm. The dashed gray line shows a Gaussian fit performed only in the range from 390 to 410 nm, and the red vertical line marks the fitted peak center. A double-peak structure was reproducibly observed for several KTP crystals.

## References

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- [3] Velsko, S., Webb, M., Davis, L. & Huang, C. Phase-matched harmonic generation in lithium triborate (LBO). *IEEE Journal of Quantum Electronics* **27**, 2182–2192 (1991).
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