Supporting Information:
Linear electro-optic effect in silicon nitride waveguides enabled by electric-field poling

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Supplementary Note 1: Device design and properties

The silicon nitride (Si₃N₄) optical waveguides used in thermally assisted electric-field poling and third-order susceptibility measurements were fabricated by LIGENTEC using its AN800 technology platform. Waveguides with two different cross-sections were used in this study - 1.31 × 0.81 µm² (wide) and 0.57 × 0.81 µm² (narrow) as shown in Fig. S1(a)). In the chip design, electrodes are located in a plane 10 µm apart from each other and 1.7 µm above a waveguide. The latter offset is due to fabrication limitations. In future designs, the electrodes could be placed in the same plane as the waveguide hence providing means for increasing the electric field intensity in the core in comparison to the current design. The waveguides are folded in meanders on 5 × 10 mm² chips by using curved sections with a radius of 75 µm, as schematically shown in Fig. S1(b). The electric and optical field interaction lengths L are 4.5 cm and 8.3 cm for the waveguides having 1.31 µm and 0.57 µm
widths, respectively. The material constants used in this study are summarized in Table S1. Here, it is important to note that in the literature, the ratio of the third-order susceptibility tensor elements $\chi_{1111}^{(3)}:\chi_{1122}^{(3)}$ of silica (SiO$_2$) was found to vary from 1.6:1 to 7:1.$^{31}$ Therefore, as an approximation of $\chi_{1122}^{(3)}$, we considered the reported value of $\chi_{1111}^{(3)}$ and divided it by 3 so that Kleinman’s symmetry rule is met.

![Figure S1: (a) Schematic cross-section of devices used in the study. Here, Si$_3$N$_4$ waveguides are buried in silica (SiO$_2$) and have widths $w$ of 1.31 µm and 0.57 µm. In each device, aluminum (Al) electrodes were introduced to enable application of an electric field. The electric and optical field interaction lengths $L$ are 4.5 cm and 8.3 cm, respectively. (b) Schematic layout of the devices used in the study.](image)

![Table S1: Material constants used in the numerical simulations](table)

| Description                                         | Value                  |
|-----------------------------------------------------|------------------------|
| Si$_3$N$_4$ refractive index $n_{Si3N4}$ at $\lambda = 1560$ nm | 1.986                  |
| SiO$_2$ refractive index $n_{SiO2}$ at $\lambda = 1560$ nm    | 1.444                  |
| SiO$_2$ third-order susceptibility $\chi_{1111}^{(3)}$     | $2.0 \times 10^{-22}$ m$^2$/V$^2$ S2 |
| SiO$_2$ third-order susceptibility $\chi_{1122}^{(3)}$     | $0.67 \times 10^{-22}$ m$^2$/V$^2$ S2 S3 |
| Si$_3$N$_4$ dielectric constant $\epsilon_{Si3N4}$         | 7.5$^{34}$             |
| SiO$_2$ dielectric constant $\epsilon_{SiO2}$             | 4.2$^{34}$             |

**Supplementary Note 2: Two-photon imaging**

We used an upright Leica LSM 710 NLO two-photon microscope (TPM) at Cellular Imaging Facility of the University of Lausanne for measuring inscribed electric field in optical waveguides after thermally assisted electric-field poling. Initially, a He-Ne laser operating at 633 nm was used for optical alignment, while a Coherent Chameleon Ultra II IR Ti:Sapphire laser giving 1010 nm horizontally (relative to the image plane) polarized light was employed for
excitation of second-harmonic (SH) in the poled regions of the device. During the TPM measurement, the focal point is raster-scanned across a sample, and the SH signal is collected. The optical resolution of a retrieved image is estimated to be around 310 nm.

Fig. S2 shows an optical image (a) and two TPM images ((b) and (c)) before and after electric-field poling, respectively, of the device comprising a waveguide with dimensions $0.57 \times 0.81 \, \mu m^2$. As evident in both TPM images, a signal along the sides of the electrode boundaries is detected in both cases. The latter could be due to symmetry breaking at the metal-dielectric interface.\textsuperscript{S5} Importantly, as evident from Fig. S2(b), there is no response along the waveguide located between the electrodes meaning that there is no intrinsic nonlinearity present in the waveguide core. Notably, after thermally assisted electric-field poling, we found that optical nonlinearity can be induced in the waveguide in case it would have only one electrode (anode) in its vicinity. The latter case is displayed in the TPM image in Fig. S2(c). As seen here, two electrodes are placed along the waveguide, the latter being in the middle of the image, while the electrode on the left side ends earlier than that on the right. Despite this, there is some SH signal generated at the waveguide location throughout its entire length, as shown. It is also worth noting that bright spots in the TPM image, e.g., Fig. S2(b), are due to accidental deposition of a thermal compound on the chip surface. The thermal compound is consistently used for establishing a thermal contact between the chip and the thermal stage.

**Supplementary Note 3: Electric and optical field distribution and overlap simulations**

We use COMSOL Multiphysics for the calculation of third-order susceptibility from the experimentally measured refractive index modulation in the proposed devices. The connection between these two parameters is established through Eq. (6) from the main text, which requires electric and optic field simulations, as well as geometrical parameters of the device.
Figure S2: (a) An optical image of the device comprising a waveguide with dimensions \(0.57 \times 0.81 \, \mu\text{m}^2\). In the same device, (b) and (c) show TPM images of the same region before and after thermally assisted electric-field poling, respectively. Note that in (c), the waveguide is poled also in the region where it transitions from being between two electrodes to having only a single electrode (anode) in its vicinity.

These are showcased in Fig. S3(a) to (d), considering the case of the narrow waveguide. Here, we initially define the geometry of the waveguide as shown in Fig. S3(a). Two regions having different optical properties are evident in the figure corresponding to the Si\(_3\)N\(_4\) core and SiO\(_2\) cladding. It is important to note that in the model, the corners of elements were rounded. The latter is introduced in order to avoid very high electric fields near the edges during poling dynamics simulations. The known refractive indices, dielectric constants, and third-order susceptibilities of the employed materials are assigned according to values in Table S1. Fig. S3(b) and (c) show the simulated optical and applied electric field profiles before poling. The \(\chi^{(3)}\), the optical mode \(E(x, y)\), and the electric field \(E_m(x, y)\) profiles are multiplied according to the nonlinear interaction defined in Table 1 in the main text. For the case of refractive index change at twice the applied modulation frequency (or \(2\omega_m\)), the profile of \(\chi^{(3)}|E(x, y)|^2|E_m(x, y)|^2\) is as shown in Fig. S3(d). The latter is integrated across device cross-section following equation:

\[
\Delta n_{\text{eff}} = 2c\varepsilon_0 \int \int \frac{3\chi^{(3)}(x, y)}{4} |E(x, y)|^2 |E_m(x, y)|^2 \, dx \, dy,
\]

where \(c\) is the speed of light in vacuum, \(\varepsilon_0\) is the permittivity of free space. Importantly, \(E(x, y)\) is such that \(\int \int (E(x, y)^* \times H(x, y) + E(x, y) \times H(x, y)^*) \hat{z} \, dx \, dy = 1\), where \(H(x, y)\) is the magnetic field profile of an optical mode. The value obtained using Eq .1 is matched
to the experimentally measured through variation of third-order susceptibility of Si$_3$N$_4$. The calculated third-order susceptibility of Si$_3$N$_4$ is then used for fitting the thermally assisted electric-field poling curves. Here, charge carrier concentration $C$ and diffusion coefficient $D$ will determine the electric field profile in the waveguide, which in turn can be used for estimation of induced nonlinearity following steps as described above. It is important to note that, during simulations, only the $x$-component of the applied electric field is considered. The latter is justified by the fact that the $x$-component is at least one order of magnitude larger than the $y$-component, according to the numerical simulation. It is displayed in Fig. S3(e) and (f), where both applied field components are shown across waveguides having cross-sections of $1.31 \times 0.81 \ \mu m^2$ and $0.57 \times 0.81 \ \mu m^2$, respectively.

Figure S3: (a) Device cross-section ($0.57 \times 0.81 \ \mu m^2$) showing two regions having different optical properties. Here, only third-order susceptibilities of Si$_3$N$_4$ and SiO$_2$ are indicated. (b) and (c) the simulated TE polarized optical and applied electric field profiles, respectively, before electric-field poling. (d) Normalized profile of $\chi^{(3)}|E(x, y)|^2|E_m(x, y)|^2$ according to Eq. 1. (e) and (f) externally applied electric field $x$- ($E_x$) and $y$- ($E_y$) components across waveguides having cross-sections of $1.31 \times 0.81 \ \mu m^2$ and $0.57 \times 0.81 \ \mu m^2$, respectively. Displayed values correspond to the case when voltage of 400 V is applied between the electrodes.
Supplementary Note 4: Linear and quadratic electro-optic effect with TM polarized light

The studied devices were also characterized for operation with TM polarized light following the same workflow as discussed in the main text where TE operation is presented. Below in Fig. S4(a) and (b), we show the TM polarized optical mode field profile in the studied waveguides. It is important to note that in the case of the narrow waveguide, as shown in Fig. S4(b), the TM mode has a higher effective refractive index than the TE mode, hence it is more confined in the waveguide core. Fig. S4(c) and (d) show the effective refractive index change $\Delta n_{\text{eff}}$ measured via quadratic and linear electro-optic (EO) as a function of modulation voltage amplitude $V_m$. In the case of the latter, constant DC voltage $V_{DC} = 100$ V was applied between the electrodes. Quadratic and linear trends of $\Delta n_{\text{eff}}$ growth with $V_m$ for quadratic and linear EO effect, respectively, are evident similarly as displayed in the main text for the case when the devices are tested in the TE operation regime. There are two main differences when comparing the performance of the same devices operating with TM and TE polarized light. Firstly, in the TM regime, the achieved $\Delta n_{\text{eff}}$ is lower due to smaller $\chi^{(3)}$ experienced by the light. Secondly, the difference between induced $\Delta n_{\text{eff}}$ measured in the narrow and wide waveguide is smaller because of the higher confinement of light for TM mode in the narrow waveguide core, as mentioned above. Fig. S4(e) displays the $\Delta n_{\text{eff}}$ per $V_m$ measured using TM polarized light in the wide and narrow waveguides at $\omega_m$ after each thermally assisted electric-field poling interval. These values are marked with points in the graph. The solid lines show the simulated dynamics of the refractive index change in the studied waveguides using parameters derived from the evolution of $\Delta n_{\text{eff}}$ with the optical mode polarized in TE polarization (see main text). Finally, in Fig. S4(e), the dashed lines mark the $\Delta n_{\text{eff}}$ per $V_m$ via linear EO effect with DC field applied. As evident, there is a good agreement between the experimentally measured and numerically simulated refractive index change values.
Figure S4: (a) and (b) TM-mode profile in waveguides with $1.31 \times 0.81 \, \mu m^2$ and $0.57 \times 0.81 \, \mu m^2$ cross-sections. (c) and (d) Effective refractive index change $\Delta n_{\text{eff}}$ due to quadratic and linear EO effect (under $V_{DC} = 100 \, V$) as a function of the modulation voltage $V_m$ using TM polarized light. (e) Effective refractive index change $\Delta n_{\text{eff}}$ per $V_m$ (points) measured in both waveguides at $\omega_m$ after each electric-field poling interval using TM polarized light. Solid lines show the simulated dynamics of the refractive index change and dashed lines correspond to $\Delta n_{\text{eff}}$ per $V_m$ via linear EO effect with DC field applied.

Supplementary Note 5: Poling dynamics simulations

During numerical simulation of dynamics of the refractive index change in the thermally assisted electric-field poling process, it was found that the saturation values and growth rates are very sensitive to the charge carrier diffusion coefficient $D$ and number concentration $C$, particularly in the device comprising the narrow waveguide. This is showcased in Fig. S5, in which experimentally measured and numerically simulated poling dynamics in both narrow and wide waveguides are considered. As mentioned in the main text of the paper, the
charge carrier diffusion coefficient $D$ mainly determines the induced nonlinearity growth time constant (see Fig. S5(a) and (c)), while the concentration $C$ determines both the saturation value and growth time (see Fig. S5(b) and (d)). The induced nonlinearity at the end of electric-field poling is particularly sensitive to the concentration $C$ of charge carriers. This is because the $C$ value determines the depletion zone size, which within the considered charge carrier concentration range takes a considerable amount of waveguide core in which the optical mode is strongly confined. Also, from the measurement and simulation results in Fig. S5, it is clear that, because of the lack of measurement points in the initial part of electric-field poling, the fitting error for the diffusion coefficient $D$ is expected to be large.

![Graphs](image)

Figure S5: Experimentally measured $\Delta n_{\text{eff}}$ per $V_m$ during thermally assisted electric-field poling along with simulated dynamics using different charge carrier diffusion coefficient $D$ and number concentration $C$ values included in the legends. Measurements are done with TE polarized light. (a) and (b) correspond to the case of the wide waveguide, while (c)-(d) to the narrow waveguide.
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