Efficient photoinduced second-harmonic generation in silicon nitride photonics

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Silicon photonics lacks a second-order nonlinear optical ($\chi^{(2)}$) response in general, because the typical constituent materials are centrosymmetric and lack inversion symmetry, which prohibits $\chi^{(2)}$ nonlinear processes such as second-harmonic generation (SHG). Here, we realize high SHG efficiency in silicon photonics by combining a photoinduced effective $\chi^{(2)}$ nonlinearity with resonant enhancement and perfect phase matching. We show a conversion efficiency of $(2,500 \pm 100)\% \text{W}^{-1}$ that is two to four orders of magnitude larger than previous field-induced SHG works. In particular, our devices realize milliwatt-level SHG output powers with up to $(22 \pm 1)\%$ power conversion efficiency. This demonstration is a breakthrough in realizing efficient $\chi^{(2)}$ processes in silicon photonics, and paves the way for further integration of self-referenced frequency combs and optical frequency references.

Second-order ($\chi^{(2)}$) nonlinear optics is a cornerstone for many classical and quantum applications. For example, to achieve compact functionalities for optical-frequency synthesis and optical atomic clocks, frequency combs based on third-order ($\chi^{(3)}$) nonlinear processes need to be self-referenced by efficient second-harmonic generation (SHG), ideally on the same silicon chip. However, common materials in silicon photonics, including silicon (Si), silicon nitride ($\text{Si}_3\text{N}_4$) and silicon dioxide ($\text{SiO}_2$), do not support the $\chi^{(2)}$ response in bulk within the electric-dipole approximation. It is therefore particularly challenging to realize efficient SHG on a silicon chip (for example, for $2f-2f$ locking in frequency combs). As a result, systems based on silicon photonics technology have often relied on conventional platforms such as centimetre-scale periodically poled lithium niobate waveguides for $\chi^{(2)}$ functionalities. Alternatively, there has been considerable progress in realizing efficient SHG in non-silicon-based thin-film platforms, including aluminium nitride, gallium arsenide and lithium niobate materials. Such advances generally require heterogeneous integration with a silicon-based platform for optimized performance in the aforementioned frequency comb applications. On the other hand, silicon carbide nanophotonics has recently made major strides, demonstrating $\chi^{(2)}$ processes in both photonic-crystal cavities and microring resonators. However, the fabrication processes that realize high performance in silicon carbide (SiC) suggest that its integration with the rest of the silicon photonics platform may be challenging.

There has also been work aiming to demonstrate effective $\chi^{(2)}$ processes directly in typical silicon photonics materials. One approach uses the weak $\chi^{(2)}$ nonlinearity present in silicon-based systems (for example, due to symmetry breaking) in conjunction with perfect phase matching in high-quality-factor (Q) microwires to boost the normalized SHG efficiencies to 0.1% W$^{-1}$ (ref. 19) and 0.049% W$^{-1}$ (ref. 20). These efficiencies can be improved by optimized input/output waveguide–resonator coupling, but are ultimately limited by the weakness of the $\chi^{(2)}$ nonlinearity. Another approach uses a large effective $\chi^{(2)}$ nonlinearity created through the combination of an electric field and the medium's $\chi^{(3)}$ nonlinearity in photonic waveguides without cavity enhancement. This electric field can be induced directly by external electrodes or optically through the photogalvanic effect and has resulted in $\chi^{(2)}$ nonlinearities up to 41 pm V$^{-1}$ and normalized SHG efficiencies as high as 13% W$^{-1}$ (ref. 21). The induced field not only produces a nonlinearity that substantially exceeds the existing intrinsic nonlinearity, but also supports quasi-phase matching (QPM), with phase either pre-determined or self-organized/photoinduced. However, the above approaches, when used separately, are inefficient compared to devices using traditional $\chi^{(2)}$ materials, and, as a result, are far from generating milliwatt-level continuous-wave SHG output.

To realize efficient SHG, we engineered devices that take advantage of both a strong effective $\chi^{(2)}$ nonlinearity and resonant enhancement. We use the $\text{Si}_3\text{N}_4$ platform, which has been successfully applied to many wide-band nonlinear photonics demonstrations, including octave-spanning frequency combs, classical/quantum frequency conversion and optical parametric oscillation. Here, the physical process in use is photogalvanic field-induced SHG, first discovered in germanium-doped glass fibres decades ago. In contrast to reports of a photoinduced $\chi^{(2)}$ in non-resonant geometries such as $\text{Si}_3\text{N}_4$ waveguides and $\text{SiO}_2$ fibres, we demonstrate an effective photoinduced $\chi^{(2)}$ nonlinearity in a high-Q $\text{Si}_3\text{N}_4$ microresonator. We show that this resonantly enhanced, photoinduced $\chi^{(2)}$ nonlinear process enables high-efficiency SHG with appreciable output power for continuous-wave inputs.

The physical process behind our approach involves three $\chi^{(2)}$ and $\chi^{(3)}$ nonlinear interactions among three modes. The modes are illustrated in Fig. 1a, and consist of a direct-current (d.c.) mode, a pump mode and a second harmonic mode. The three interactions are illustrated in Fig. 1b, labelled I–III:

- **Seedling SHG (I):** this process yields a nanowatt-level SHG signal due to a small intrinsic $\chi^{(2)}$ nonlinearity, for example, from surface symmetry breaking.

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Fig. 1 | Photoinduced SHG in a Si3N4 microring resonator. a, Illustration of the device and modes involved in perfect phase matching. A Si3N4 microring is integrated with two coupling waveguides for pump (ω) and SHG (2ω) light. Within the microring, the mode profiles of three interacting modes at 2ω, ω and d.c. frequencies are shown from top to bottom. Red and blue indicate positive and negative phases of the electric fields, that is, pointing outwards or inwards in the radial direction. The darkness of the colours depicts the local field strength. These modes satisfy perfect phase matching for both χ(2) and χ(3) processes. We note that, while the azimuthal distribution of the d.c. field is uniform (due to the perfect phase matching), the exact radial distribution pattern of the d.c. field is unknown and is assumed to be uniform here as a rough estimate when, for example, extracting the device’s effective χ(2).

b, Photogalvanic SHG with perfect phase matching in a microring. (I) A small seedling SHG (2ω) field is generated with a weak seedling intrinsic χ(2) nonlinearity. (II) This 2ω field, together with the ω field, builds up a d.c. field through the coherent photogalvanic effect, which is distinctively slow (up to minutes) compared with ordinary nonlinear optical processes (<1 ps). (III) The generated d.c. field and the pump field in turn generate SHG light through a field-induced SHG effect, that is, the d.c. Kerr effect. Once seeded by (I), (II) and (III) work together to build up the SHG field. c, Our SHG scheme (red) is superior in power efficiency to other processes in χ(2) silicon photonics, including biased QPM (orange), photogalvanic QPM (blue) and high-Q resonators with weak nonlinearities (purple). This toy model is used to compare the physics of our scheme to related SHG processes in silicon photonics and is qualitative in nature. It only considers QPM and the threshold of the process, and cavity Q enhancement is not considered for illustration purposes. The cavity will provide an additional enhancement related to its finesse. See the Supplementary Information for more details. PG, photogalvanic; PPM, perfect phase matching; WG, waveguide.

- Photogalvanic effect (II)9,40: this process coherently builds up a d.c. electric field through the pump field and the seedling SHG field. The d.c. field can be very large, up to $1 \times 10^7 \text{ V m}^{-1}$ (ref. 41).
- Field-induced SHG (III)40: the electric field generated by the photogalvanic effect combines with the medium’s χ(3) to create an effective χ(2) nonlinearity, which in turn amplifies the SHG field (and hence the photogalvanic effect).

Importantly, the scheme is unique in the context of photoinduced SHG in realizing perfect phase matching for both the intrinsic χ(2) process and the field-induced χ(2) process simultaneously. Such perfect phase matching is sometimes referred to as modal phase matching; however, here, we use perfect phase matching specifically to distinguish from cases where engineered modal dispersion and quasi-phase matching are used together15,43. First, phase and frequency matching of the intrinsic χ(2) process, as shown in Fig. 1b, interaction (I), is supported by recent techniques developed in nanophotonic dispersion engineering22. Once such matching is fulfilled, the χ(2) processes, as shown in Fig. 1b (II,III), are automatically matched given the nature of the d.c. field, which is stationary, with zero angular momentum and a frequency of $\omega = 0$. The χ(2) and χ(3) processes can therefore work together seamlessly; that is, the induced χ(2) can feed on the intrinsic seedling SHG to self-start (Fig. 1b), rather than relying on external electrodes or a SHG laser for initiation21. Second, the field-induced χ(2) in our scheme, through perfect phase matching with a d.c. field, is more efficient than those reported previously, which are achieved through quasi-phase matching with radio-frequency (RF) fields21,22,24,28, as shown in Fig. 1c. Owing to such perfect phase matching, our induced χ(2) from the d.c. field is always at its maximum, instead of having periodic modulations that decrease the effective χ(2) to 2/π and 1/π times the maximum value for square41 and sinusoidal22,24,28 longitudinal profiles of the RF fields, respectively. Moreover, the resonance nature enables the induced χ(2) to remain spatially uniform inside the resonator, instead of forming build-up and decay-down regions as observed in waveguide geometries22,23,28. Through this phase-matched, photoinduced and resonant SHG process, we report a SHG conversion efficiency of $(2.500 \pm 100)\% \text{ W}^{-1}$ in a Si$_3$N$_4$ microring resonator, which is two to four orders of magnitude larger than previous works in typical silicon-based materials19-24. Our absolute SHG efficiency is $(22 \pm 1)\%$ at an output power of $(1.9 \pm 0.1) \text{ mW}$, with an input pump power of $(8.8 \pm 0.1) \text{ mW}$. This performance level is promising for f-2f self-referencing of octave-spanning microresonator frequency combs within a common Si$_3$N$_4$ platform2,4,29,30. Moreover, considering the recent integration of Si$_3$N$_4$ photonics with Rb vapour cells6,41, high-performance stabilization of telecom lasers (which have also recently been integrated with Si$_3$N$_4$ photonics42) through efficient SHG that connects the lasers to narrow-linewidth atomic transitions49 may now be possible within a single integrated photonic platform.

The SHG device is a Si$_3$N$_4$ microring integrated with two coupling waveguides, as shown in Fig. 1a. A pump laser in the telecom band (~1,560 nm) is coupled by a straight waveguide (bottom) into the microring, in which the 1,560-nm light is frequency-doubled to 780 nm, and the 780-nm light is coupled out by a separate waveguide (top). This top waveguide only supports the 780-nm light; it does not support any modes for 1,560 nm (see the Supplementary Information for more details on coupling). Inside the microring, three modes are involved in the process, the d.c. mode, the pump mode (1,560 nm) and the SHG mode (780 nm). The d.c. mode is stationary (resonance frequency of $\omega = 0$) with zero azimuthal angular momentum, and can be characterized by $(n, m) = (0, 0)$ when depicted in the whispering gallery mode terminology, where $n$ represents the radial mode number and $m$ represents the azimuthal mode number. To perfectly match the mode numbers for the pump and SHG, we use the fundamental transverse-electric
Using this method, we can selectively modify radial and azimuthal mode numbers. In particular, the (1, 154) and (3, 308) modes (labelled in red) satisfy perfect phase matching and are used in the experiment as pump and SHG modes. The pump mode number is identified using mode-splitting devices targeting (1, 155) and (1, 154), respectively, and the corresponding cavity modes are clearly identified by a mode splitting of ~0.7 nm. Their intrinsic $Q$-factor values are $6.0 \times 10^6$ and $1.6 \pm 0.1 \times 10^6$, respectively. The errors represent 1 s.d. uncertainties in nonlinear fitting of the resonances.

Although phase matching is quantized and is perfect once the appropriate modes are identified, frequency matching of those modes typically needs thermal/power tuning, as shown in Fig. 3a,b. The device has loaded quality factors of $>0.5 \times 10^6$ and intrinsic $Q$ values of $>1 \times 10^6$ for both pump and SHG modes (Fig. 2d,h). These high $Q$ values necessitate frequency matching to be within ~0.3 GHz.
Fig. 3 | Laser detuning to optimize photoinduced SHG in the microring. a,b. Experimental results (a) and illustrations (b) showing three regions with a distinctive SHG response. In b, the x axis represents the wavelength and SHG can happen when the pump mode wavelength (λm) and twice the SHG mode wavelength (2λm) overlap (this is equivalent to the overlap of ν2m and 2νm, but the schematic is drawn from the experimental perspective in which light is injected into the telecom mode as the wavelength is increased, as in a). In regions (i) and (iii), the device has no observed SHG response, because of the inefficient seedling SHG process, as the cavity modes are frequency-mismatched. In region (ii) (green area), the cavity frequencies match and photoinduced SHG can self-start, either through forward (blue) or backward (purple) laser tuning. As shown in the inset, both tuning methods require forward tuning at first to drop laser power into the cavity, because of thermal bistability indicated by the dashed red line, whose height is proportional to the pump power that is dropped into the cavity. In region (ii), photoinduced SHG can self-start and the response time depends on the laser detuning, which affects the cavity frequency mismatch, as shown in b(ii). Darker green indicates a faster response in region (ii). The dashed black line in a indicates the output SHG power after the laser is tuned for 10 s. The error bars in a represent 1 s.d. uncertainties from the calibration of the on-chip power. c. The response takes a few seconds or less than a second when the pump is below 1,558.11 nm (blue), and takes from 20 s at 1,558.12 nm (green) to 110 s at 1,558.15 nm (red). This slow response is a signature of the photogalvanic process, in contrast to other nonlinear processes that are typically ultrafast (<1 ps). The dashed black line indicates a time of 10 s after laser tuning, corresponding to that in a. The lines are guides to the eye.

The thermo-optic bistability exhibited by the high-Q cavity, indicated by dashed red triangles in Fig. 3a,b, requires the pump laser to be scanned from blue detuning to red detuning to drop power into the cavity. When pump power is first dropped into the cavity, illustrated by region (i) in Fig. 3a,b, the pump and SHG mode are mismatched in frequency, similar to the cold cavity case. Here, the seedling SHG process is only resonantly enhanced by the pump cavity mode but not by the SHG cavity mode, and yields no observable SHG signal (the minimum power our detector can resolve is 0.1 mW). Without such seedling SHG, effective photoinduced SHG cannot self-start. When the pump laser is tuned into region (ii) in Fig. 3a,b, the two cavity modes start to have spectral overlap, which results in an appreciable seedling SHG power to start the photogalvanic effect.

To reach optimal SHG power, both forward and backward tuning of the pump laser are required, with the specifics dependent on the laser–cavity detuning. These two tuning methods are illustrated in the inset of Fig. 3a. For example, the SHG power of 1.15 mW can be directly generated by forward tuning, where the laser is tuned from <1,558.06 nm to 1,558.09 nm, as indicated by the blue arrow. However, the larger SHG power of ~1.2 mW at 1,558.08 nm can only be accessed through backward tuning: that is, the laser is first tuned in the forward direction from <1,558.06 nm to >1,558.09 nm, and then tuned in the backward direction to 1,558.08 nm, as indicated by the purple arrow. In comparison, when the laser is directly forward-tuned from <1,558.06 nm to 1,558.08 nm, no SHG signal is observed. Such hysteresis is probably due to pump depletion, as the cavity frequency matching is different when SHG just starts (without depletion) in comparison to when it has already started (with depletion). We have verified that with a lower input power (~4 mW), such hysteresis is substantially decreased.

As discussed earlier, when the pump laser is set to a wavelength between 1,558.09 nm and 1,558.15 nm, SHG can self-start simply by forward tuning. The response time of the process is determined by the photogalvanic process. This time depends critically on the cavity frequency matching. When the frequency is matched well (top panel (ii), Fig. 3b), that is, λm = 1,558.09–1,558.11 nm, the response time is within a few seconds. By contrast, when the cavity modes are not well frequency-matched (bottom panel (ii), Fig. 3b), that is, λm = 1,558.12–1,558.15 nm, the SHG has a slow build-up time ranging from 20 s to over a minute, as shown in Fig. 3c. Note that our photogalvanic build-up time is much shorter than those reported in previous works. Even the slowest case has a response time of ~110 s, and this response time is in general two to three orders of magnitude faster than previous photogalvanic work in waveguides[12,13,14]. Such a notable change in response time is related to the cavity enhancement (the cavity linewidth). The resonance wavelengths of the pump and SHG modes are 1,557.785 nm and 778.877 nm, respectively, recorded by a wavemeter at room temperature when the optical power is small so that both Kerr and thermo-optic shifts are negligible. The SHG mode thus needs an 7.7-GHz per °C (0.58 pm per °C) (details are provided in the Supplementary Information).
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Our results have superior efficiency compared to previous works due to the use of perfect phase matching and resonant enhancement rather than quasi-phase matching in waveguides (dashed blue line) and silica fibres, while our much larger nonlinearity leads to improved performance relative to all other resonant schemes in silicon photonics (dashed purple line). We note that the efficiency does not show a linear dependence on pump power. Although our normalized SHG efficiency of (2,500 ± 100)% W⁻¹ is only 1% of the recently reported χ⁽²⁾ record in lithium niobate microrings, our absolute SHG efficiency of (22 ± 1)% (Supplementary Information) is comparable to state-of-the-art χ⁽²⁾ nanophotonic devices. Moreover, the maximal SHG power we obtained is ~2.2-mW SHG in the waveguide with ~15-mW input pump power (bottom panel, Fig. 4a). Achieving such milli-watt-level output SHG powers with high conversion efficiency is indicative of how this work demonstrates photoduced SHG that is comparable to state-of-the-art χ⁽²⁾ results. The Supplementary Information presents a more detailed discussion of this comparison, as well as the estimated induced χ⁽²⁾, d.c. field and related discussions.

The maximal induced χ⁽²⁾ value is χ⁽²⁾ = (0.20 ± 0.04) pm V⁻¹ (Supplementary Information), near the lower end of previous Si₃N₄ photogalvanic results, which range from 0.3 pm V⁻¹ to 3.7 pm V⁻¹. The electric field is estimated to be (0.6 ± 0.1) MV cm⁻¹, which is ~15–20% of the electric breakdown voltage of Si₃N₄ (ref. 51), that is, 3–4 MV cm⁻¹. Our induced nonlinearity is currently limited by pump depletion and can probably be improved further. For example, in previous work, the applied field is 0.25 MV cm⁻¹, which is 62.5% of the electric breakdown voltage of Si (0.40 MV cm⁻¹). Operation near the breakdown field of silicon nitride, for example, 4 MV cm⁻¹, suggests an upper-bound estimate of 1.3 pm V⁻¹ as potentially accessible in our system.

Efficient SHG only occurs in our system at sufficiently high (milli-watt-level) injected powers, and an unsolved problem for photogalvanic SHG is the origin and value of such threshold powers. Previously, such threshold behaviour has only been reported empirically in fibre (5-kW peak power) and waveguide (10-W peak power), both using quasi-phase matching and pulse pumping. We believe our system, with perfect phase matching and continuous-wave pumping at milli-watt levels, is a promising platform to probe the origin of this threshold, for example, whether it
comes from the photogalvanic process alone or the overall nonlinear process.

In summary, we have demonstrated efficient photoinduced SHG with perfect phase matching in silicon photonics, achieving record-high conversion efficiencies in comparison to prior silicon-based devices, and absolute efficiencies and output powers on par with the highest values demonstrated in nanophotonic media with much larger intrinsic $\chi^{(2)}$ nonlinearities. Our demonstration opens up promising avenues for $\chi^{(2)}$ nonlinear silicon photonics, including f$\to$2f locking for $\chi^{(2)}$-mediated octave-span frequency combs, SHG-based connections between telecom lasers and atom-based frequency standards, sum/difference-frequency generation and electro-optical modulation. Going forward, the integration of external electrodes to create the d.c. electric field can enable efficient SHG across a wider range of input powers, produce a larger effective $\chi^{(2)}$ response and realize additional levels of tunability and control.

Online content
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Data availability
The data that support the plots within this paper and other findings of this study are available from the corresponding authors upon reasonable request.

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Author contributions
X.L. led the design, fabrication and measurement of the SHG devices. G.M., A.R. and K.S. provided assistance with design and measurement. D.A.W. provided assistance with fabrication. All authors participated in the analysis and discussion of results. X.L. and K.S. wrote the manuscript with assistance from all authors, and K.S. supervised the project.

Competing interests
The authors declare no competing interests.

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