Coherent Visible-Light-Generation Enhancement in Silicon-Based Nanoplasmonic Waveguides via Third-Harmonic Conversion

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We report visible third-harmonic conversion at \( \lambda = 517 \) nm in subwavelength silicon-based nanoplasmonic waveguides at an unprecedented conversion efficiency of \( 2.3 \times 10^{-5} \). This marks both the highest third-harmonic conversion efficiency in a silicon-based or nanoplasmonic structure and the smallest silicon waveguide structure demonstrated to date. The high conversion efficiency is attributed to tight electric field confinement and strong light-matter coupling arising from surface plasmon modes in the nanoplasmonic waveguide, enabling efficient nonlinear optical mixing over micrometer length scales. The nonresonant geometry of the waveguide enables the entire \( \lambda = 1550 \) nm femtosecond pulse spectrum to be converted to its third harmonic, which may be easily extended to the entire visible spectrum. We envisage that third-harmonic generation in silicon-based nanoplasmonic waveguides could provide a platform for integrated, broadband visible light sources and entangled triplet photons on future hybrid electronic-silicon photonic chips.

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Understanding the potential for efficient frequency conversion in nano-optical circuits is of key importance for future chip-scale information processing. Optical harmonic generation marked the beginning of nonlinear optics and it remains a powerful process for converting coherent optical radiation to higher frequencies [1]. In the particle interpretation of light, harmonic generation is understood as the union of \( N \) low-energy photons into a single photon with \( N \times \) the energy. For example, third-harmonic generation (THG) occurs when three photons of the fundamental frequency \( \omega \) interact in a nonlinear medium to generate one photon at the \( 3\omega \) frequency. In the wave description of THG, a driving electric field \( E_\omega \) induces a third-order electric polarization in a medium that acts as a \( 3\omega \) radiation source as follows [2]:

\[
P^{(3)}(3\omega) = \frac{E_\omega^3}{4} \chi^{(3)}(3\omega; \omega, \omega, \omega)\]

where \( \chi^{(3)}(3\omega; \omega, \omega, \omega) \) is the third-order electric susceptibility and \( E_\omega \) is the permittivity of free space. Although second-harmonic generation (SHG) is only possible in materials without inversion symmetry, THG occurs in any nonlinear medium for sufficient electric field strength.

Typically, high-efficiency harmonic generation is achieved in exotic crystals that are incompatible with modern semiconductor processing, making implementation in planar photonic circuitry a challenge. As a critical material for electronics, Si has also become the foundation for integrated optical systems, and it merges electronics, photonics, and nanoplasmonics on a single platform. The potential of Si as a functional nonlinear optical material has incited many nonlinear frequency conversion studies, including stimulated Raman scattering, self- and cross-phase modulation, and four-wave mixing [3]. Although a variety of optical functionalities have been demonstrated in Si, efficient light generation remains challenging due to its indirect band gap. Rather than endeavor light emission through phonon-assisted transitions from the valence band to the conduction band, it is more feasible to generate coherent light through nonlinear optical interactions such as THG [2]. Temporal pulse compression in dispersion engineered Si photonic waveguides offers a potential means to improve the THG conversion efficiency to \( \sim 10^{-7} \) [4]. However, photonic waveguides are bulky compared to modern electronic circuitry and it is desirable to reduce them to the nanoscale, permitting unidimensional interfacing to electronic circuits.

Despite decades of development, efficient SHG and THG in nanoscale structures remains a formidable challenge. Nanoplasmonic structures incorporate metallic features, allowing propagating optical signals to couple strongly to the conduction electrons of the metal, enabling subwavelength optical confinement [5]. Confinement of input radiation to a nanoscale volume not only facilitates compact frequency conversion structures suitable for planar optical circuitry, but also enhances the optical intensity, increasing nonlinear frequency conversion efficiencies. Although “plasmonic” metals are characterized by large \( \chi^{(3)} \) coefficients (e.g., \( \chi^{(3)} \sim 0.2 \) nm² V⁻² for gold [6]), high losses restrict nonlinear interactions to the metallic surfaces where few nonlinear dipoles contribute to the \( 3\omega \) signal, resulting in low conversion efficiencies. Furthermore, previous investigations of THG in plasmonic structures, such as aperture arrays and gratings, have not been conducive to integration in planar photonic circuits [7–14]. For example, rough silver films on a Si substrate have been shown to enhance
THG conversion efficiencies compared to a thick silver film [7]. However, such films are not nanoscale devices and no absolute measurement of the THG conversion efficiency was reported. Nanoplasmonic waveguides can be easily loaded with a high-nonlinearity material, enabling plasmonically enhanced nonlinear interactions in the volume (rather than the surface) of a nonlinear subwavelength structure. Specifically, a Si-loaded nanoplasmonic waveguide \( \chi^{(3)} = 0.19 \text{ nm}^2 \text{ V}^{-2} \) for Si [15]) would enable high-efficiency THG in waveguides with sub-100-nm dimensions, delivering a compact visible light source for all-optical signal processing and quantum information encoding [16–18].

In this Letter, we present the first demonstration of THG in a nanoplasmonic waveguide. Broadband THG in the green spectral region is generated in a Si-based nanoplasmonic waveguide that is simple to fabricate, compatible with modern electronics and photonics technologies, and occupies an ultracompact footprint of 0.40 \( \mu \text{m}^2 \). Femtosecond laser pulses at \( \lambda = 1550 \text{ nm} \) with input powers in the range of 85 \( \mu \text{W–1.52 mW} \) are used to generate strong THG with conversion efficiencies up to 2.3 \( \times \) 10^{-5}, the highest reported to date in any Si-based device or nanoplasmonic structure. The nonresonant waveguide enables 3\( \omega \) up-conversion of the entire pulse spectrum, which can be extended to a much wider spectral range [19]. These results demonstrate a unique, enabling platform for integrated frequency conversion sources spanning visible to near-IR wavelengths.

We investigate a nanoplasmonic waveguide that consists of a Si core with width \( w = 95 \text{ nm} \), height \( h = 340 \text{ nm} \), length \( L = 5 \mu \text{m} \), and a \( t = 60 \text{ nm} \) thick gold cap, as shown in Fig. 1(a). Because of the subdiffraction dimensions of the nanoplasmonic waveguide, only a single quasi-TM waveguide mode at \( \lambda = 1550 \text{ nm} \) exists. The effective refractive index \( n_{\text{eff}} \) of the waveguide mode is plotted versus wavelength between 1500 and 1600 nm in Fig. 1(b) and decreases from 2.71 to 2.63 over this range. The third harmonic of \( \lambda = 1550 \text{ nm} \) lies in the green region of the visible spectrum where the photon energy, \( E_{3\omega} = 2.40 \text{ eV} \), exceeds the indirect band gap energy of Si, \( E_g = 1.1 \text{ eV} \). This observation comes with two important consequences: the 3\( \omega \) signal will be absorbed strongly and high dispersion in Si will hinder phase matching. Four quasi-TM and three quasi-TE modes exist for the 3\( \omega \) signal, and \( n_{\text{eff}} \) for each of these modes is also plotted in Fig. 1(b). Strong dispersion of the Si core produces a higher \( n_{\text{eff}} \) for the low-order 3\( \omega \) modes than the fundamental mode. For higher-order 3\( \omega \) modes, more energy propagates outside of the high-index Si core, resulting in a lower \( n_{\text{eff}} \). Although the dispersion relations of the TE\(_{22}\) and TM\(_{00}\) modes intersect at \( \lambda = 1552 \text{ nm} \), we will demonstrate through simulations that phase matching between these two modes is not possible because the TM\(_{00}\) cannot excite the TE\(_{22}\) mode.

It is also important to consider the relevant length scales of each waveguide mode. At \( \lambda = 1550 \text{ nm} \), the TM\(_{00}\) mode has an effective wavelength, \( \lambda_{\text{eff}} = 581 \text{ nm} \), and decays by a factor of \( e^{-1} \) as it propagates, \( L_{\text{prop}}^{\text{TM}_{00}} = 3.1 \mu \text{m} \) (loss = 1.4 dB/\( \mu \text{m} \)). The tight mode confinement and short propagation length signify strong light-matter coupling and are ideal for efficient nonlinear interactions in an ultracompact structure. Similarly, the TM\(_{02}\) mode has \( L_{\text{prop}}^{\text{TM}_{02}} = 140 \text{ nm} \) and propagates a characteristic length, \( L_{\text{prop}}^{\text{TM}_{02}} = 634 \text{ nm} \) (1.1\( \omega_{\text{TM}_{02}} \)), demonstrating that phase matching over multiple wavelength scales is not feasible. For comparison, the TM\(_{20}\) mode has \( \lambda_{\text{eff}} = 235 \text{ nm} \) and propagates a characteristic length \( L_{\text{prop}}^{\text{TM}_{20}} = 260 \text{ nm} \) (0.45\( \omega_{\text{TM}_{20}} \)). For similar reasons, previous observation of THG in bulk Si has been measured from surface reflection, where the 3\( \omega \) signal is generated within the absorption depth of both \( \omega \) and 3\( \omega \) [20–26]. Although this nanoplasmonic structure does not provide the benefits of phase matching, it enables 3\( \omega \) up-conversion of broadband femtosecond pulses with minimal spectral amplitude modulation.

To gain fundamental insight into the generation and propagation dynamics of the 3\( \omega \) radiation, we incorporate a third-order polarization into finite-difference time-domain simulations, via Eq. (1). Linear losses and \( \chi^{(3)} \) values for Si, Au, and SiO\(_2\) are taken from prior experimental measurements [6,15,27–29]. We propagate pulses of \( \tau_p = 84 \text{ fs} \) duration centered at \( \lambda = 1550 \text{ nm} \) through the Si-based nanoplasmonic waveguide depicted in Fig. 1(a) and characterize the resulting THG signal. A snapshot of the \( E_z \) electric field of the 3\( \omega \) signal taken at the arrival time of the pulse peak is shown in Fig. 2(a). As expected, the most efficient THG occurs where the fundamental radiation is the most intense, near the Si-Au interface and close to the waveguide input facet (\( z = 0.31 \mu \text{m} \)) [30]. Based on the intrinsic dielectric losses at 3\( \omega \), the skin depth of Si is \( \delta_{\text{Si}} = 723 \text{ nm} \), whereas the skin depth of Au is \( \delta_{\text{Au}} = 18.9 \text{ nm} \). Therefore, any 3\( \omega \) that is radiated in the direction of the Au film (\( t = 60 \text{ nm} \)) will either be reflected into the Si or absorbed in the Au. Any 3\( \omega \) that is radiated into the Si core will contribute to the propagating 3\( \omega \).

**FIG. 1** (color online). (a) Schematic depiction of the silicon-based nanoplasmonic waveguide. (b) Mode dispersion curves for the fundamental and 3\( \omega \) wavelengths. Time-averaged intensity distributions (\( \langle I \rangle \)) for three waveguide modes are shown on the right-hand side.
Evidently, phase matching is not sufficient to compensate for the high ηeff efficiency through each plane of the waveguide, the THG conversion efficiency
\[ \eta \] of the Si core is measured to be w × h = 95 nm × 340 nm and the gold cap has a thickness of t = 60 nm.

Free-space radiation from a laser emitting τp = 84 fs pulses centered at λ = 1550 nm is collinearly coupled into the nanoplasmonic waveguides using a microscope objective (N.A. = 0.85). For peak input powers above \[ P_{\text{peak}} = 9.9 \text{ W} \], bright visible light emission is observed from the waveguides under normal room lighting. Visible emission collected from above the sample by a 20× microscope objective and delivered to a charge-coupled device camera is shown in Fig. 3(d). Bright white light emission with a strong blue component is visible at the input facet of the nanoplasmonic waveguide and distinct THG (green light) is observed at the output facet. Depending on input excitation alignment and power, strong white light emission could be observed along the entire length of the waveguide. The white light emission arises practically challenging and a special scheme must be realized. As such, the nanoplasmonic waveguides are fabricated on a narrow ridge that is etched to a depth \[ d_{\text{etch}} = 75 \mu m \], allowing for collinear access with standard microscope objectives and lensed SMFs [30]. An artistic rendering of the sample along with the excitation and detection scheme are shown in Fig. 3(a). Figure 3(b) presents a SEM of a single Si-based nanoplasmonic waveguide. A narrow ridge including several nanoplasmonic waveguides embedded in silica cladding is shown in Fig. 3(c), and the inset depicts a cross-sectional SEM of a nanoplasmonic waveguide end facet. Cross-sectional dimensions of the Si core are measured to be w × h = 95 nm × 340 nm and the gold cap has a thickness of t = 60 nm.

FIG. 2 (color online). (a) Time snapshot of the 3ω \[ E_z \] electric field at the fundamental pulse peak arrival. (b) Time-averaged intensity distribution of the 3ω radiation at z = 0.5 μm. (c) Third-harmonic conversion efficiency along the waveguide z axis for peak electric fields of \[ E_{\text{peak}} = 1, 2, 3, 4, \] and 5 V/μm.

A time-averaged intensity distribution in the z = 0.5 μm plane is shown in Fig. 2(b), depicting the mode transformation from a field localized to the Au-Si interface into a distribution that resembles the TM03 waveguide mode. A time-averaged intensity distribution in the z = 0.5 μm plane is shown in Fig. 2(b), depicting the mode transformation from a field localized to the Au-Si interface into a distribution that resembles the TM03 waveguide mode. By recording the fundamental and 3ω power transmission through each plane of the waveguide, the THG conversion efficiency \[ \eta^{\text{THG}} \] is calculated for several peak electric fields, as shown in Fig. 2(c). The maximum \[ \eta^{\text{THG}} \] is observed at z = 310 nm from the input facet and decreases along the z axis of the waveguide. For \[ E_{\text{peak}} = 5 \text{ V/μm} \], conversion efficiencies up to \[ \eta = 1.40 \times 10^{-4} \] are calculated. Oscillations in \[ \eta^{\text{THG}} \] with a period of \[ L_\text{c} = 487 \text{ nm} \] occur along the length of the waveguide, which closely matches the coherence length \[ L_\text{c} = 2\pi/\Delta k = 505 \text{ nm} \] predicted from the \[ n_{\text{eff}} \] data in Fig. 1(b) for a pure TM03 mode. Evidently, phase matching is not sufficient to compensate for the high 3ω losses. Setting \[ \chi^{(3)} = 0 \] for Au and SiO2 reveals that 98.9% of the THG occurs in the Si, demonstrating that loading nanoplasmonic waveguides with a highly nonlinear medium drastically increases \[ \eta^{\text{THG}} \] over a bare nanoplasmonic structure.

To accurately measure THG in a Si-based nanoplasmonic waveguide, it is crucial to directly excite nonlinearities in the waveguide so that the measurements are not intertwined with nonlinear excitations in an on-chip coupling scheme. By direct collinear coupling of free-space \( \lambda = 1550 \text{ nm} \) radiation into the nanoplasmonic waveguides using a microscope objective and outcoupling the generated visible emission with a lensed single-mode optical fiber (SMF), detailed spectral analysis and direct extraction of the THG conversion efficiency become possible. When considering a very short nanoplasmonic waveguide, the substrate must be reduced to ≤10 μm dimensions, which is
from collisions between hot electrons and valence electrons close to the input facet of the nanoplasmonic waveguide, where a two-photon absorption-generated free-carrier population interacts with the tightly confined nanoplasmonic mode. These free-carriers can be ponderomotively accelerated to energies exceeding the threshold for impact ionization in Si, leading to avalanche multiplication of the free-carrier population [31]. Although interesting, white light emission from hot electrons is not a coherent light source and is not investigated in these measurements. We optimize the input alignment conditions for the strong-source and is not investigated in these measurements. Movie S1 in the Supplemental Material [30] presents THG from several individual nanoplasmonic waveguides as the beam is scanned across, demonstrating that emission was only observed from waveguides and not from other areas of the sample. The THG signal was present in every nanoplasmonic waveguide that was tested regardless of the waveguide length.

The coherent light emission spectrum is measured by coupling both the \( \omega \) and \( 3\omega \) light emitted from the output facet of the nanoplasmonic waveguide to a lensed SMF. Figure 4(a) depicts the outcoupled laser spectrum together with the \( 3\omega \) spectrum, which spans a wavelength range of \( 500 \leq \lambda \leq 530 \) nm. The broad \( 3\omega \) spectrum and absence of sharp peaks confirm that phase matching does not play a role in the THG. The \( 3\omega \) spectrum as calculated from the measured fundamental spectrum is also shown for reference. The excellent agreement between the calculated and the measured \( 3\omega \) spectra further confirms the absence of phase matching. Similar measurements on a bare Si photonic waveguide produce a similar THG signal strength, confirming that the dominant nonlinear response arises from the Si features. However, the THG signal from the Si-based nanoplasmonic waveguide is approximately 27\% stronger in a footprint area that is reduced by over a factor of 3 [30].

A surface plot depicting the \( 3\omega \) spectral power \( P_{\text{out}}^{3\omega} \) as a function of the average input laser power \( P_{\text{in}}^{\omega} \) is shown in Fig. 4(b). Regardless of \( P_{\text{in}}^{\omega} \), the \( 3\omega \) spectrum spans \( 500 \leq \lambda \leq 530 \) nm, and the peak wavelength is at \( \lambda = 517 \) nm. The overlaid plot conveys a measured \( 3\omega \) spectrum for \( P_{\text{in}}^{\omega} = 1.36 \) mW (dashed line in the figure). As shown in Fig. 4(c), a log-log plot of the \( P_{\text{in}}^{\omega} \) versus the total \( 3\omega \) power \( P_{\text{out}}^{3\omega} \) (obtained by integrating the \( 3\omega \) spectrum) scales with a slope of 2.99 ± 0.02. Similarly, the slopes for specific wavelengths in the \( 3\omega \) spectrum, \( \lambda = (512, 517, 520) \) nm, are found to be 3.04 ± 0.01, 2.93 ± 0.01, and 3.16 ± 0.04, respectively. The \( P_{\text{out}}^{3\omega} \propto (P_{\text{in}}^{\omega})^3 \) dependence confirms that the green light originates from the third-order \( \chi^{(3)}(-3\omega; \omega, \omega, \omega) \) nonlinear optical interaction [2] and demonstrates that the entire \( 3\omega \) spectrum grows uniformly with increasing input power.

Using a calibrated spectrometer detector, an absolute measure of the \( 3\omega \) power and conversion efficiency is obtained. The THG conversion efficiency from the nanoplasmonic structure is plotted as a function of the input power on a log-log scale in Fig. 4(d). A linear fit to this data produces a line with a slope of 3.84, which is very close to the expected value of 4. This quartic power scaling arises from the expected \( 3\omega \) conversion efficiency dependence, i.e., \( P_{\text{out}}^{3\omega} / P_{\text{in}}^{\omega} \propto (P_{\text{in}}^{\omega})^4 \). For a peak electric field of \( E_{\text{peak}}^{\omega} = 4.8 \) V/\( \mu \)m, the conversion efficiency is measured to be \( \eta_{\text{THG}} = 2.3 \times 10^{-5} \) and green light powers up to \( P_{\text{out}}^{3\omega} = 35 \) nW are generated. For reference, the calculated \( \eta_{\text{THG}} \) is plotted on the same graph, for two physical scenarios: perfectly smooth Au films and Au films with nanoscale roughness that would enhance the average electric fields by an additional factor of 1.09. The scaling trend for an electric field enhancement of 1.09 matches the experimental measurements very closely and, therefore, the roughness of the Au film cannot be neglected. Finally, we also plot an estimate of the maximum internal conversion efficiency in the nanoplasmonic waveguides. From this, we extract the maximum internal conversion efficiency in the experiments to be \( \eta_{\text{THG}} = 4.71 \times 10^{-4} \).

The measured \( \eta_{\text{THG}} = 2.3 \times 10^{-5} \) marks a 400× increase in \( \eta_{\text{THG}} \) over previous plasmonic arrays [10], a 230× increase in \( \eta_{\text{THG}} \) over bulky silicon photonic structures [4], and a ~10^6× increase over SHG efficiencies in a single plasmonic structure [32].

FIG. 4 (color online). (a) Spectra of the fundamental (red), the measured third harmonic (green), and the third harmonic calculated from the fundamental spectrum (blue). (b) Surface depicting third-harmonic power versus wavelength and input power. The black overlaid \( 3\omega \) spectrum is measured for \( P_{\text{in}}^{\omega} = 1.36 \) mW. (c) Log-log plots of detected \( 3\omega \) power versus input power for the entire spectrum (green) and individual wavelengths, \( \lambda = (512, 517, 520) \) nm. (d) Third-harmonic conversion efficiency as a function of peak electric field.
0.40 μm², with potential for further reduction. The absence of saturation in the \( P_{\text{out}} \propto (P_{\text{in}})^{3} \) trend or damage to the sample demonstrate that \( n_{\text{THG}} \) can be increased further simply by increasing the input power. We note that further increases in the input power will eventually lead to depletion of the fundamental pulses due to two-photon absorption. For example, the critical free-carrier density in Si of \( n_c = 1.44 \times 10^{21} \text{ cm}^{-3} \) is reached for an electric field of \( E = 2.2 \text{ V/nm} \). The fact that we do not yet observe saturation in \( n_{\text{THG}} \) is due to the short interaction length in the nanoplasmonic waveguide and the near-instantaneous nature of the THG process. Furthermore, the nonresonant characteristics of the Si-based nanoplasmonic waveguide enable the visible light color to be tuned by simply changing the excitation wavelength, where an excitation wavelength of \( \lambda^{\text{in}} = 1200 \text{ nm} \) would generate violet light and \( \lambda^{\text{in}} = 2100 \text{ nm} \) would generate red light. The compatibility of these structures with electronics and photonics processing techniques make them ideal for monolithic integration with existing technologies. We note that military specification electronic circuits use Au interconnects, and it is anticipated that integration of Au features into CMOS electronics would not affect electronic performance. Alternatively, CMOS-compatible plasmonic materials, such as Al or TiN, could replace the Au features.

In summary, we have demonstrated dramatic increases in the third-harmonic conversion efficiency in Si-based nanoplasmonic waveguides. Absorption of the third-harmonic signal in the Si waveguide core enables uniform conversion of broadband femtosecond pulses. These measurements are performed in a nanoscale, Si-based structure, making it attractive for use in Si photonic or electronic circuitry. The conceptual simplicity of this design leaves room for tailoring the waveguide geometry to further enhance the conversion efficiency, with the possibility of achieving integrated coherent visible light sources with microwatt powers. These results demonstrate the feasibility for conversion of near-IR radiation to visible radiation with technologically significant efficiencies, enabling nanoscale Si-based visible light sources on a chip.

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