Title: Giant nonlinear response at a plasmonic nanofocus drives efficient four-wave mixing over micron length scales

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Abstract: Efficient optical frequency mixing typically must accumulate over large interaction lengths as nonlinear responses in natural materials are inherently weak. This limits the efficiency of mixing processes due to the requirement of phase matching. Here we report efficient four-wave mixing (FWM) over micron-scale interaction lengths at telecommunications wavelength on silicon. We use an integrated plasmonic gap waveguide that strongly confines light within a nonlinear organic polymer. The gap waveguide intensifies light by nanofocusing it to a mode cross-section of a few tens of nanometers, thus generating a nonlinear response so strong that efficient FWM accumulates over wavelength-scale distances. This technique opens up nonlinear optics to a regime of relaxed phase matching, with the possibility of compact, broadband, and efficient frequency mixing integrated with silicon photonics.

One Sentence Summary: Efficient wave mixing in plasmonic waveguides on silicon introduces a route to versatile non-resonant nonlinear optical devices with relaxed phase matching limitations.

Main Text:

Nonlinear optics, especially frequency mixing, underpins modern optical technologies and scientific exploration in quantum optics (1, 2), materials and life sciences (3, 4), and optical communications (5, 6). Four-wave mixing (FWM) is an important nonlinear frequency conversion technique used in photonic integrated circuits and telecommunications for signal regeneration (6), switching (7), phase-sensitive amplification (8), metrology (9), and entangled photon-pair generation (10). As a third order nonlinear effect, FWM is extremely sensitive to enhancement by the optical confinement of nanoplasmonic systems (11). For example, FWM has been demonstrated in a variety of metallic nanostructures including nano-antennas (12), rough surfaces (13), and at sharp tips (14). Nonetheless, efficient frequency conversion has remained elusive. While metals can be highly nonlinear and afford extreme optical localization, at telecommunications wavelengths only a small fraction of a plasmonic mode interacts with the metal and increasing this only exacerbates absorption. An alternative strategy is to incorporate low-loss nonlinear materials within nanoplasmonic systems (15, 16). Indeed, recent theoretical studies of FWM in plasmonic waveguides incorporating nonlinear polymers are promising (17). Nonlinear polymers defy Miller's rule by exhibiting large Kerr indices (18, 19) for relatively low

refractive indices, and this has been exploited in recent studies (20). In the context of plasmonics, this brings two advantages: polymers are straightforward to integrate within metallic nanostructures by solution processing (21), and their low refractive index minimizes propagation loss.

In this report we utilize a silicon hybrid gap plasmon waveguide (HGPW) (11, 17, 22) to mediate pump degenerate four-wave mixing (DFWM) in the nonlinear polymer poly[2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene] (MEH-PPV) (18), as illustrated in Fig. 1 (23). The device consists of input and output gratings to launch and collect optical signals, either side of a metallic waveguide of length, L , and width, W , as narrow as $W = 25$ nm, which is accessed via two tapered sections. In recent work (11), we demonstrated this system's capability to enhance more than 100 fold the intensity of light within the narrow gap; a process known as adiabatic nanofocusing (24). In this work, the nonlinear polymer infiltrates the narrow gap section, where the optical field is expected to be maximal.

Unlike conventional DFWM, our approach does not require operation near zero-dispersion wavelengths for phase-matching (23). Here, phase-matching is irrelevant because the propagation distances are considerably shorter than the DFWM coherence lengths under investigation. Near a wavelength of $\lambda = 1500$ nm over a pump to signal bandwidth of $\Delta\lambda = 30$ nm, the $W = 25$ nm waveguide has a coherence length of hundreds of micrometres, which is far longer than its $2\text{ }\mu\text{m}$ propagation length. Even a bandwidth of $\Delta\lambda = 300$ nm near 1500 nm would have a coherence length greater than the propagation length. For more details on phase-matching and dispersion in the HGPWs, see (23).

Figure 1 illustrates the nanofocusing mechanism (11, 22). An input beam polarized parallel to the gratings couples to transverse electric (TE)-like waveguide modes, with dominant electric field component in the plane. For wide gap widths the fundamental TE-like mode propagates primarily in the silicon layer over distances $>100\text{ }\mu\text{m}$ as its modal overlap with metal is minimal. For $W < 50$ nm, the mode becomes concentrated in the gap region. While the mode only propagates for a few microns in this state, the gap's field enhancement is dramatic (11). The taper angle to access this confined mode is selected to minimize propagation loss and reflections or scattering that would reduce the nanofocusing efficiency. For more details on the taper/grating coupling efficiencies and the waveguide propagation losses, the reader is directed to the (23).

In order to investigate DFWM in this plasmonic device, two spectrally distinct pulses centered at $\lambda_s = 1450$ nm (signal pulse) and $\lambda_p = 1480$ nm (pump pulse) were generated by filtering a femtosecond pulse centred at $\lambda = 1480$ nm. The spectral full width at half maxima of these pulses were used to estimate transform limited pulse widths ≥ 1.04 ps. The pump and signal pulses were coupled to HGPWs through the in-coupling grating and the resulting idler pulse centred at $\lambda_i = (2\lambda_p^{-1} - \lambda_s^{-1})^{-1} = 1510$ nm was measured on a spectrometer from the out-coupling grating after spectrally filtering out the pump and signal (23). Figure 2A compares the normalized input and filtered output spectra for a peak pump power of 30 W and a HGPW with $W = 25$ nm and $L = 2\text{ }\mu\text{m}$. The input and output spectral counts represent the power spectral density (PSD, $P(\lambda, z)$) at the start and end of the narrow section of HGPW, respectively, determined from measured grating and tapering efficiencies (23). Our experimental results agree with numerical pulse propagation simulations based on the nonlinear Schrödinger equation (NLSE), using nonlinear material parameters for MEH-PPV, silica and silicon measured with the Z-scan method (23). Figure 2B shows the simulated conversion spectrum overlaid with experimental pump/signal and idler spectra from Fig. 2A.

From the input and output spectra, we can extract the conversion efficiency (CE) of the DFWM process (6), defined as the ratio of the peak idler PSD after the narrow waveguide section ($P(\lambda_i, L)$) to the peak signal PSD at the start ($P(\lambda_s, 0)$) directly from Fig. 2A (23). For this HGPW, $\text{CE} = -13.3$ dB. This was the highest conversion efficiency measured in this study and is comparable to ultrafast DFWM in silicon waveguides over millimeter scale interaction lengths (17). When considering the integrated PSD of each beam, the generated peak power in the idler is 22.7 % of the power in the signal, assuming identical temporal characteristics of the signal and idler. Accounting for the power used in all beams, we also define a DFWM efficiency from the peak idler power after the plasmonic waveguide, $\mathcal{P}_i(L)$, to both the peak signal power, $\mathcal{P}_s(0)$, and the peak pump power, $\mathcal{P}_p(0)$, at the start of the waveguide, $\eta = \mathcal{P}_i(L)/(\mathcal{P}_s(0)\mathcal{P}_p(0)^2)$. For the most efficient device considered here, $\eta = 0.025 \text{ \%W}^{-2}$.

We have confirmed the nature of the conversion process by observing a cubic dependence of idler peak PSD on the combined pump and signal powers (Fig. 3A). The cubic relationship arises from a linear dependence on signal power and a quadratic dependence on pump power. Figure 3B compares the measured conversion efficiency with that simulated using the NLSE as a function of peak pump power for a HGPW of $W = 25$ nm and $L = 2$ μm . The simulated conversion efficiency varies with peak power cubed until a critical power where nonlinear absorption of the pump beam dominates. For $\mathcal{P}_p(0) > 30$ W in the narrowest waveguides ($W = 25$ nm), the MEH-PPV degraded, setting the upper power limit for our dataset. Although the conversion efficiency roll-off was not observed in experiments, it is remarkable that nonlinear absorption should not limit performance until peak powers approaching 100 W, due to the short device lengths.

The observed conversion efficiencies were approximately 20 dB less than those expected from NLSE simulations using the measured nonlinear parameters of MEH-PPV films. Since the discrepancy was systematic across all measured devices (Fig. 4), we can identify a number of reasons. Firstly, poor infiltration of MEH-PPV into the gap would not only affect the waveguide's nonlinear coefficient but also the mode confinement. Secondly, the morphology of the MEH-PPV within the gap could be distinct from that of bulk films, which were used to assess the material's nonlinear parameters. Finally, calculating the waveguide nonlinearity, γ , from the nonlinear responses of the various device materials could require more rigorous theoretical treatment (25). Nevertheless, all data broadly agrees with theory for a waveguide nonlinearity, γ , that is a factor of 2.5 – 3 times less than that inferred from Z-scan measurements.

The plasmonic waveguide width and length clearly influence the DFWM conversion efficiency; while a narrower gap boosts the effective nonlinear coefficient the additional propagation loss limits idler accumulation. This raises the question: what is the optimal interaction length? Figure 4A shows the conversion efficiency of HGPWs with $W = 25$ nm and $\mathcal{P}_p(0) = 30$ W for $L = 1$ to 5 μm . The experiment is broadly consistent with the theory that conversion efficiency increases with device length until a maximum is reached due to growing propagation loss. The fact that the conversion efficiency is maximal near the measured propagation length of 1.9 ± 0.6 μm suggests that DFWM accumulates rapidly and that the optimal gap width is < 25 nm. The dominant role of confinement in these devices is apparent from the much smaller CEs of HGPWs with $W = 50$ nm despite the increase in peak interaction length (Fig. 4B). Figures 4C and 4D show complimentary data on how the conversion efficiency varies with gap width for two fixed HGPW lengths of $L = 3$ μm and $L = 5$ μm , at $\mathcal{P}_p(0) = 30$ W. Although the gap width affects both propagation loss and nonlinear coefficient, broad

agreement between NLSE simulations and experiments remains, demonstrating that this frequency mixing approach is robust and repeatable.

We have shown that the intense light at a nanofocus enables nonlinear optical control over extremely short interaction lengths comparable to the vacuum wavelength of light. Remarkably, at the minimum gap width of 25 nm in this study, we are still operating far from where non-local and quantum effects arise at the sub-nanometre scale (26) suggesting scope for improvement through reducing the gap width, studying wider conversion bandwidth, and exploring alternative nonlinear gap materials. Moreover, our approach mitigates phase matching limitations over large bandwidths in a non-resonant manner (27). With efficient nonlinear processes over distances shorter than a plasmonic mode's propagation length we can also eliminate the key problem of insertion loss that has plagued the application of plasmonics. This shows that plasmonic nanofocusing on a silicon platform can be a powerful tool in nonlinear optics.

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Fig. 1. Nanofocusing devices and modal properties of organic hybrid gap plasmon waveguides on silicon. (A) Schematic representation. Insets show the nanofocusing mechanism with electromagnetic mode distributions with $S = 25$ nm and $M = 40$ nm for a narrow gap, $W = 25$ nm and a wide gap, $W = 500$ nm (the $W = 500$ nm mode has been scaled by x5 for clarity). Lower left inset shows the chemical formula for MEH-PPV. (B) SEM of a $L = 2 \mu\text{m}$ long, $W = 25$ nm wide waveguide without cladding depicting the in-/out-coupling gratings. (C) Close-up of the same waveguide.

Fig. 2. Four-wave mixing spectra. (A) Normalized pump/signal spectrum and out-coupled idler spectrum from a $L = 2 \mu\text{m}$ waveguide of $W = 25 \text{ nm}$ for $\mathcal{P}_p(0) = 30 \text{ W}$. (B) The same spectra (black lines) overlaid by input/output spectra from the NLSE simulations (red lines).

Fig. 3. Degenerate four-wave mixing as a function of combined pump and signal power for a HGPW with $W = 25 \text{ nm}$ and $L = 2 \mu\text{m}$. (A) Peak idler photon counts as a function of average pump power showing the third order dependence on the combined pump and signal power. (B) Signal to idler conversion efficiency versus peak pump power compared solutions of the nonlinear Schrodinger equation for varying waveguide nonlinearity, γ . For this HGPW, the waveguide nonlinearity $\gamma/2.5 = (3.09 + 0.07i) \times 10^4 \text{ W}^{-1}\text{m}^{-1}$.

Fig. 4. Degenerate four-wave mixing conversion efficiencies for a variety of different HGPW devices. Conversion efficiency versus waveguide length for HGPWs of (A) $W = 25$ nm at $\mathcal{P}_p(0) = 30$ W and (B) $W = 50$ nm at $\mathcal{P}_p(0) = 40$ W. Conversion efficiency versus waveguide width for HGPWs of (C) $L = 3$ μm and (D) $L = 5$ μm at $\mathcal{P}_p(0) = 30$ W. Solid lines show theoretical conversion efficiencies calculated with the NLSE using $\gamma/2.5$.

Supplementary Materials:

Materials and Methods

Supplementary Text

Figures S1 to S7

References (28-40)