Giant nonlinear response at a plasmonic nanofocus drives efficient four-wave mixing

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Efficient optical frequency mixing typically must accumulate over large interaction lengths because nonlinear responses in natural materials are inherently weak. This limits the efficiency of mixing processes owing to the requirement of phase matching. Here, we report efficient four-wave mixing (FWM) over micrometer-scale interaction lengths at telecommunications wavelengths on silicon. We used 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.

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 telecommunication systems 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 nanoplasmic 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. Although metals can be highly nonlinear and afford extreme optical localization, at telecommunication 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 means of solution processing (21), and their low refractive index minimizes propagation loss.

Illustrated in Fig. 1 is the silicon hybrid gap plasmon waveguide (HGPW) (11, 17, 22) that we have used to mediate pump degenerate FWM (DFWM) in the nonlinear polymer poly[2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene] (MEH-PPV) (18). 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). In our approach, phase-matching is irrelevant because the propagation distances are considerably shorter than the DFWM coherence lengths under investigation. Near a wavelength of λ = 1500 nm over a pump-to-signal bandwidth of Δλ = 30 nm, the W = 25 nm waveguide has a coherence length of hundreds of micrometers, which is far longer than its 2-μm propagation length. Even a bandwidth of Δλ = 300 nm near 1500 nm would have a coherence length greater than the propagation length. More details on phase-matching and dispersion in the HGPWs are available in (23).

The nanofocusing mechanism is illustrated in Fig. 1 (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 µm because its modal overlap with metal is minimal. For W < 50 nm, the mode becomes concentrated in the gap region. Although the mode only propagates for a few micrometers in this state, the gap's field enhancement is dramatic (17). The taper angle to access this confined mode is selected to minimize propagation loss and reflections or scattering that would reduce the nanofocusing efficiency. More details on the taper/grating coupling efficiencies and the waveguide propagation losses are provided in (23).

In order to investigate DFWM in this plasmonic device, two spectrally distinct pulses centered at λ_1 = 1450 nm (signal pulse) and λ_p = 1480 nm (pump pulse) were generated by filtering a femtosecond pulse centred at λ = 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 λ_i = (2λ_1^2 - λ_p^2)^1/2 = 1510 nm was measured on a spectrometer from the out-coupling grating after spectrally filtering out the pump and signal (23). The normalized input and filtered output spectra are compared in Fig. 2A for a peak pump power of 30 W and a HGPW with W = 25 nm and L = 2 µm. The input and output spectral counts represent the power spectral density (PSD), P(λ, 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). Shown in Fig. 2B is the simulated conversion spectrum overlaid with the 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_i(0, L)] to the peak signal PSD at the start [P_s(0, 0)] directly from Fig. 2A (23). For this HGPW, CE = 33.3 dB. This was the highest conversion efficiency measured in this study and is comparable with ultrastrip 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: P_i(0), to both the peak signal power, P_s(0), and the peak pump power, P_p(0), at the start of the waveguide, P_i(0) / [P_s(0)]/P_p(0)]^2]. For the most efficient device considered here, η = 0.025% 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. In Fig. 3B, the measured conversion efficiency is compared with that simulated by 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 curedd by a critical power at which nonlinear absorption of the pump beam dominates. For P_p(0) > 30 W in the narrowest waveguides (W = 25 nm), the MEH-PPV degraded, setting the upper power limit for our data set. 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, owing to the short device lengths.

The observed conversion efficiencies were ~20 dB less than those expected from NLSE simulations by using the measured nonlinear parameters of MEH-PPV films. Because the discrepancy was systematic across all measured devices (Fig. 4), we can identify a number of reasons. First, poor infiltration of MEH-PPV into the gap would not only affect the waveguide's nonlinear coefficient but also the mode confinement. Second, 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. Last, 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 to 3 times less than that inferred from Z-scan measurements.

The plasmonic waveguide width and length clearly influence the DFWM conversion efficiency; whereas 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? The conversion efficiency of HGPWs with W = 25 nm and P_p(0) = 30 W for L = 1 to 5 µm is shown in Fig. 4A. The experiment is broadly consistent with the theory that conversion efficiency increases with device length until a maximum is reached because of 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 > 25 nm despite the increase in peak interaction length (Fig. 4B). Complementary data are shown in Fig. 4, C and D, on how the conversion efficiency varies with gap width for two fixed HGPW lengths of L = 3 µm and L = 5 µm, at P_p(0) = 30 W. Although the gap width affects both propagation loss and nonlinear coefficient, broad agreement between NLSE simulations and experiment is observed.
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 with the vacuum wavelength of light. Remarkably, at the minimum gap width of 25 nm in this study, we are still operating far from where nonlocal and quantum effects arise at the subnanometer scale (26), which suggests 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 nonresonant 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.

REFERENCES AND NOTES

Fig. 4. DFWM conversion efficiencies for a variety of different HGPW devices. (A and B) Conversion efficiency versus waveguide length for HGPWs of (A) W = 25 nm at $\beta_p(0) = 30$ W and (B) $W = 50$ nm at $\beta_p(0) = 40$ W. (C and D) Conversion efficiency versus waveguide width for HGPWs of (C) $L = 3$ μm and (D) $L = 5$ μm at $\beta_p(0) = 30$ W. Solid lines show theoretical conversion efficiencies calculated with the NLSE using $\gamma/2.5$.
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A plasmonic route for mixing waves
Nonlinear optics typically requires photons to interact over distances spanning hundreds or thousands of wavelengths. Nonlinear optical devices therefore tend to be bulk components. Nielsen et al. used a polymer material with a high nonlinear coefficient that they embedded within a plasmonic cavity to show that the interaction length scale could be reduced dramatically. The plasmonic cavity focused the light down to the nanoscale, providing an intense electromagnetic field that induced the nonlinear process of four-wave mixing in the polymer. The technique provides a versatile platform for compact nonlinear optical devices. Science, this issue p. 1179