Plasmonic Mirrorless Optical Parametric Oscillator

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Supporting Information

ABSTRACT: We analyze a nondegenerate four-wave mixing process on a metal–dielectric interface in which the signal and idler waves are counter-propagating surface plasmons and the process is pumped by a free wave propagating in air. We show that, similarly to mirrorless parametric oscillations, above a certain threshold intensity of the pump, the system exhibits instability, which results in the spontaneous generation of plasmons propagating on the metal surface. Interestingly, we find that, in such a plasmonic nonlinear interaction, phase-matching is naturally fulfilled by the geometry of the process, allowing a broadband tunability of the output frequencies by varying the angle of incidence of the pump.

KEYWORDS: Surface plasmons, four-wave mixing, parametric oscillations, surface nonlinearities, counter-propagating waves

Nonlinear interaction of counter-propagating beams can give rise to a multitude of fascinating phenomena due to the inherent feedback mechanism imposed by the unique boundary conditions. In particular, in frequency conversion processes, such feedback can give rise to parametric instability, which can result in parametric oscillations in a nonlinear medium even without a cavity.4−6 In such a mirrorless optical parametric oscillator (MOPO), the pump photons are converted into pairs of counter-propagating signal and idler photons at different energies. Above a certain threshold intensity of the pump, the instability of this backward interaction leads to the spontaneous buildup of both the signal and the idler. This makes the MOPO a highly attractive source of coherent radiation, which is simple, robust, and does not require any cavity alignment or seeding. Moreover, because such a process is essentially a nondegenerate down-conversion process, the generated signal and idler plasmons could be quantum-correlated.4−6 Although the basic concept behind the MOPO was already introduced in the 1960s,1 it is only recently that such a device has been demonstrated experimentally.7−10 This is because the counter-propagation geometry results in a significant momentum mismatch. Although quasi-phase-matching can be obtained by periodically modulating the interaction medium, such a processes requires that the grating period would be on a subwavelength scale,1 which is extremely challenging. This was finally achieved by Canalias et al.,7 who demonstrated for the first time a MOPO operating in the near-infrared region using a periodically poled nonlinear crystal. In these experiments, it was also shown that the output wavelengths can be varied over a range of several tens of nanometers by changing the pump wavelength. Alternatively, it was also shown that the phase-matching problem for such backward nonlinear processes can be eliminated using negative-index metal materials,11 zero-index materials,12 or hyperbolic materials.13

Here, we propose and analyze a plasmonic mirrorless parametric oscillator in which the nonlinear interaction between counter-propagating surface plasmons is pumped by a free-propagating plane wave. We show that, in such a plasmonic MOPO, phase-matching conditions can be readily obtained on a smooth metal film without any periodic structuring. Unlike quasi-phased-matched MOPOs, in which the periodicity is typically optimized for a specific wavelength combination, the plasmonic MOPO can be conveniently tuned over a broad spectral band from the visible range to the far-infrared.

Nonlinear frequency conversion on metal surfaces has been a very active research topic for several decades,14 covering, for example, second15 and third16 harmonic generation, four-wave mixing,17−19 negative refraction,20 and nonlinear super-resolution imaging.21 Here, we consider the nonlinear interaction sketched in Figure 1a. An intense pump beam, taken as a plane-wave, is launched onto the metal film from the air side at an angle of incidence θp. The signal and idler plasmon waves propagate in the positive and negative directions along the metal–air interface, taken as the x-axis, and z corresponds to the direction normal to the interface. Instead of the three-wave mixing process, which is usually considered in MOPOs, here we consider partially degenerate four-wave mixing (FWM) in which the single pump beam enters the interaction twice as two identical fields. Essentially, such an interaction will convert a pair of identical pump photons at a frequency ωp into a pair of correlated, counter-...
proceeding photons with frequencies $\omega_s$ and $\omega_i$ occupying the signal and idler plasmon modes, as illustrated in Figure 1a.

For frequencies below the metal plasma frequency, all the waves decay exponentially along the $z$ direction and in particular into the metal film, where the nonlinear interaction takes place. Therefore, for such an interaction, momentum should only be conserved in the direction parallel to the metallic surface.\(^{18,21}\) In that sense, the present configuration is different from the previously studied transversely pumped MOPO\(^2\) in which phase-matching needs to be achieved in both the parallel and perpendicular directions. The energy and momentum conservation (phase-matching condition) can thus be written as

$$2\omega_p = \omega_s + \omega_i$$

and

$$2k_p \sin \theta_p = k_s - k_i$$

where $k_p = \omega_p/c$ is the pump wavenumber, with $c$ being the speed of light in vacuum, and $k_s$ and $k_i$ are the (in-plane) wavenumbers of the signal and idler, respectively. With the signal and idler being surface-plasmon modes propagating on a homogeneous metal film, their wavenumbers can be derived from the surface-plasmon dispersion given by

$$k_{si} = \frac{\omega_{si}}{c} \sqrt{\frac{\varepsilon_m}{1 + \varepsilon_m}}$$

where $\varepsilon_m$ is the dielectric function of the metal evaluated at the frequency of the signal or idler waves. As illustrated in Figure 1b, the phase-matching condition, as formulated by eqs 1–3, can be represented graphically\(^2\) by using a simple interpretation of the energy/momentum conservation. Choosing particular values for the signal and idler frequencies (blue and red circles in Figure 1a), the pump frequency is simply given by the average of these two values. Furthermore, when we plot the dispersion curves of the forward-propagating signal and the backward-propagating idler (with negative $k$-values), the in-plane momentum of the pump is given by the midpoint between $k_s$ and $k_i$. As shown in Figure 1b, the solution to the frequency and in-plane momentum of the pump (which is given by $k_p\sin \theta_p$) defines a point on the dispersion diagram that may reside within the air light-line. This illustrates how the FWM configuration can indeed fulfill the phase-matching condition with the pump beam taken as a free plane-wave, which uniquely defines the angle of incidence of the pump. The existence region for such a solution is given by the condition that $0 \leq \sin \theta_p \leq 1$. In that respect, our configuration resembles previous schemes that relied on nonlinear interactions to directly couple a free-space plane-wave to surface plasmons.\(^{18,21}\) Note also that, because we restrict the angle of incidence to positive values, the phase-matching condition imposes that $\omega_s \geq \omega_i$. Using eqs 1–3, the pump incident angle required for obtaining phase-matching is given by

$$\sin \theta_p = \frac{\omega_s}{\omega_s + \omega_i} \sqrt{\frac{\varepsilon_m(\omega_s)}{1 + \varepsilon_m(\omega_s)}} + \frac{\omega_i}{\omega_s + \omega_i} \sqrt{\frac{\varepsilon_m(\omega_i)}{1 + \varepsilon_m(\omega_i)}}$$

The solution for the pump angle as a function of the energies of the signal and idler plasmons within this existence region is presented in Figure 2a as calculated for an air–silver interface. As can be seen, close to the line defined by $E_s = E_i$, the incident angle is close to $0^\circ$ as expected for this symmetric situation. However, as the signal energy increases (while the idler decreases in energy), the pump needs to be launched at a large angle in order to conserve momentum in the FWM process. As can also be seen in Figure 2a, when the energy of the signal wave is too large, there is no solution for which $\sin \theta_p \leq 1$, implying that the pump beam should also be a surface wave to satisfy the phase-matching condition.

It is important to note that, because the momentum values of the plasmon wave are not purely real, the solution of the phase-matching equations results in a small imaginary component for the pump in-plane momentum. In Figure 2b, we plot this imaginary component normalized to the free-space wavenumber of the pump field given by $\text{Im}(k_p)/k_p = \text{Im}(\sin \theta_p)$ (in logarithmic scale). As can be seen, for most of the existence region, the imaginary part is smaller than 1%, ensuring that this momentum mismatch is negligible and becomes important only when the signal energy approaches $\sim 3.5$ eV.

A different manner to present the phase-matching condition is to fix the energy and incident angle of the pump beam and

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then to plot the solution for the energies of the signal and idler plasmons. This is shown in Figure 3a and b, which also shows the allowed parameter space for $E_p = h\omega_p$ and $\theta_p$. Once again, the data in Figure 3 demonstrate that, for a given pump energy, the resulting signal and idler can be tuned by varying the pump angle. For example, with $h\omega_p = 1.55$ eV (corresponding to 800 nm wavelength) as the incident angle is varied between 0° and 85°, the signal can be tuned throughout the whole visible range. At the same time, the idler wave will move into the mid-infrared region.

With the phase-matching condition satisfied, we now proceed to analyze the nonlinear interaction between the waves. The FWM process described above is mediated by two propagating plasmons, which depends on the pulse duration time. Moreover, the interaction length is also limited by the propagation losses of the plasmons, which weaken the feedback between the counter-propagating plasmons. As a result, when the absorption in the metal is taken into account, the threshold intensity increases above the value predicted by eq 9 (see Supporting Information). Nevertheless, when the interaction length is comparable to the plasmon decay distance or smaller, the increase in the critical intensity is rather moderate (see Supporting Information).

Taking the experimentally measured value for the third-order nonlinear susceptibility of Ag as $\chi^{(3)} = 0.28$ nm$^2$/V$^2$ and an interaction length of $L = 50$ μm (corresponding to a 166 fs pulse duration), the critical intensity for inducing spontaneous oscillations in the system can be evaluated numerically. Figure 4 shows the critical intensity as a function of the signal and idler energies (assuming that the pump incident angle is tuned to comply with the phase-matching condition for the $z$-dependent profiles of the interacting fields and their overlap (see Supporting Information). For the boundary conditions, we consider an interaction length of $L$ where the signal and idler waves are injected from opposite sides, as illustrated in Figure 1a, with the input amplitudes fixed as $a_{i}(0)$ and $a_{i}(L)$. These equation have the same structure as the usual MOPO system, and likewise, they exhibit an instability at a critical coupling strength. When $GL = \pi/2$, the output amplitudes for both waves [i.e., $a_{s}(L)$ and $a_{i}(0)$] diverge for a finite value of the input amplitudes. Similar to the instability occurring in normal lasers and optical parametric oscillators, this divergence means that, even for zero input amplitudes, the MOPO will spontaneously generate the signal and idler plasmons with finite output amplitude due to a positive feedback acting on random fluctuations and quantum noise. However, here as in other types of MOPOs, the feedback originates from the counter-propagation of the plasmons and no mirrors are required.

Using eq 8, we obtain the critical pump intensity as

$$I_c = \frac{\lambda_s\lambda_i\epsilon_0c}{2\pi\chi^{(3)}L} \sqrt{\frac{\beta\beta_i}{k_k\alpha\alpha_i}}$$

(9)

which scales inversely with the interaction length. However, one should note that the interaction length cannot be arbitrarily chosen, and essentially, it is set by the extent of the pump beam and the walk-off distance of the counter-propagating plasmons, which depends on the pulse duration time. Moreover, the interaction length is also limited by the propagation losses of the plasmons, which weaken the feedback between the counter-propagating plasmons. As a result, when the absorption in the metal is taken into account, the threshold intensity increases above the value predicted by eq 9 (see Supporting Information). Nevertheless, when the interaction length is comparable to the plasmon decay distance or smaller, the increase in the critical intensity is rather moderate (see Supporting Information).

![Figure 3](image-url) **Figure 3.** Tunability range for the plasmonic parametric oscillator output based on the phase-matching condition and showing the signal (a) and idler (b) energies as a function of the pump incident angle and energy.

![Figure 4](image-url) **Figure 4.** Calculated pump threshold intensity for the onset of spontaneous oscillations as a function of the signal and idler energies. Contour lines correspond to $\times10$ increments.
condition) and including the metal losses, as detailed in the Supporting Information). As can be seen, the critical intensity for relatively short signal and idler waves is on the order of 100 GW/cm², which is comparable to the typical intensities used in experiments involving nonlinear plasmon interactions. However, for wavelengths toward the near-infrared region the critical intensity increases, quickly reaching values that are above the damage threshold of metals, which limits the practical implementation of the current scheme in experiments. The main reason for the high threshold intensities is the short penetration depth of the waves into the metal and the long exponential tail of the plasmons at the air side, which lead to a very weak nonlinear interaction among them. This limitation may be overcome by using a nonlinear dielectric medium on top of the metal (instead of air), which will make the interaction more efficient. Furthermore, it is worthwhile noting that, in our calculations, we assumed that the nonlinear susceptibility is wavelength-independent and used a value extracted from four wave mixing experiments performed with wavelengths below 1 μm. However, it is most probable that, similarly to the second-order susceptibility in metals, also increases with the wavelength, which may compensate the decrease in the effective interaction volume inside the metal to some extent. At the same time, at longer wavelengths the reduced absorption and longer propagation distances (which scale as the square of the wavelength) permit longer interaction length compared to the 50 μm taken above, which will also assist in lowering the threshold intensity for the oscillations.

In conclusion, we proposed a new type of mirrorless OPO in which the emerging fields are counter-propagating surface plasmons. As we have shown, such plasmonic mirrorless parametric oscillations can be excited on a plane metal film pumped by a free-propagating beam, and the wavelengths of the resulting plasmons can be conveniently tuned by adjusting the incident angle of the pump beam. Furthermore, we showed that such a system does not require any periodic spatial modulation for obtaining phase-matching conditions. In previously studied MOPOs, which rely on quasi-phase-matching, negative-index metamaterials or waveguides, the phase-matching always depends on the typical length-scale of the underlying structure, which limits the possible spectral tuning range of the MOPO. In sharp contrast, because the plasmonic MOPO does not require any structuring, it can operate over a broad spectral band from the visible range to the far-infrared. One may also readily recognize that, for a similar counter-propagating plasmonic OPO, but based on second-order nonlinearity, the same value of $\theta_p$ (being the propagation angle of the pump at the air side) satisfies the phase-matching condition when the pump frequency is doubled. However, because in that case the nonlinearity originates from the surface alone, an analysis along similar lines as for the FWM process presented here shows that the threshold intensities are higher by a few orders of magnitude. Nevertheless, as for the FWM-based mirrorless OPO, the nonlinear interaction between the waves might be strengthened by incorporating dielectric materials with inherent nonlinearity or by using metamaterials with enhanced nonlinearities. Finally, the proposed configuration is not limited to plasmonic modes but can also be implemented using other types of surface waves for which the propagation and phase accumulation of the counter-propagating signal and idler is restricted to a plane. One example would be Bloch surface waves on 1D photonic crystals in which third harmonic generation was recently demonstrated.

## SUPPORTING INFORMATION

### References


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