Enhancing four-wave-mixing processes by nanowire arrays coupled to a gold film

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Abstract: We consider the process of four-wave mixing in an array of gold nanowires strongly coupled to a gold film. Using full-wave simulations, we perform a quantitative comparison of the four-wave mixing efficiency associated with a bare film and films with nanowire arrays. We find that the strongly localized surface plasmon resonances of the coupled nanowires provide an additional local field enhancement that, along with the delocalized surface plasmon of the film, produces an overall four-wave mixing efficiency enhancement of up to six orders of magnitude over that of the bare film. The enhancement occurs over a wide range of excitation angles. The film-coupled nanowire array is easily amenable to nanofabrication, and could find application as an ultra-compact component for integrated photonic and quantum optic systems.

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References and links

38. Note that Eqs. (2) have been corrected from those published in [17].
1. Introduction

Four wave mixing (FWM) is a nonlinear optical process that has found numerous practical applications, including: optical processing [1–3]; nonlinear imaging [4, 5]; real-time holography and phase-conjugate optics [6–8]; phase-sensitive amplification [9, 10]; and entangled photon pair production [11, 12]. Because material nonlinear susceptibilities are relatively small, significant conversion efficiency in FWM processes requires that light travels over long interaction lengths – available, for example, in silica fibers [7, 10, 12] – with an effort to achieve proper phase-matching conditions. Alternatively, the conversion efficiency can be increased by concentrating optical fields within high-refractive-index materials, such as in silicon waveguides [2, 3, 11, 13].

Here, we propose a method for increasing the efficiency of FWM processes by exploiting metals, which exhibit some of the largest known values for $\chi^{(3)}$ [14, 15]. The third-order nonlinear susceptibility in metals arises from both the nonlinear polarization induced by the motion of bulk valence electrons, as well as from the surface polarization [15–18]. Leveraging the intrinsic nonlinearities of metals has proven difficult, because light is mostly reflected from metal surfaces and hence does not interact significantly with the bulk. However, at optical wavelengths, metals support a variety of surface modes characterized by confined fields whose amplitudes can greatly exceed those of the exciting fields, such that even the small interaction volumes associated with thin metal films can substantially increase the overall efficiency of the nonlinear processes. Additionally, the enhanced fields associated with surface plasmons penetrate into the metal, increasing the volume of the bulk nonlinear interaction. Combined, these effects can produce extremely large enhancements in the effective values of $\chi^{(3)}$, which would manifest itself in a strong increase in the power flow of the of the nonlinearly generated field. In particular, thin metal films support relatively delocalized surface plasmon polaritons (SPPs) that travel along the film surface and decay exponentially away from the interface, while metal nanoparticles support tightly confined localized surface plasmon resonances (LSPRs). The field distributions and scattering characteristics associated with LSPRs depend critically on the geometry of the nanoparticle [19–21]. With either type of the resonance, the nanoscopic nature of the interaction regions suggests that nanopatterned metal structures can operate as an extremely compact device for integrated photonic and quantum optic applications.

Both SPPs and LSPRs have been shown separately to enhance nonlinearities in metal geometries [17, 18, 22–30]. It has been also shown recently that the inherent nonlinearity associated with a gold film can be used to couple light from free space into SPPs via a FWM process [25, 27]. The FWM efficiency of metal films in which SPPs are excited in such a manner can be enhanced [25–30] by up to four orders of magnitude compared to the propagating excitation regime. Likewise, the field enhancement associated with the LSPR suggests that metal nanocomposites should offer a route toward efficient nonlinear media. As an example, isolated gold nanoparticles dispersed in a glass host have been predicted to provide a three orders of magnitude increase in the intensity of phase-conjugated light produced via a degenerate FWM process [17, 22]. The enhancement occurs at wavelengths where the LSPRs of the nanoparticles are present.

Given the sensitivity of nonlinear processes to local field enhancements, it might be expected that a structure that supports both SPPs and LSPRs, leading to the interaction of the two processes, in combination with the free-space SPPs excitation by FWM process, could lead to the
FWM efficiencies significantly larger than with either mechanism acting individually. One system that naturally supports both mechanisms even as the linear response to the applied fields and is easily amenable to nanofabrication is that of a collection of metallic nanoelements with nanometer spacing from a metal film surface [31–34]. Extremely large local fields exist between a nanoelement coupled to its electromagnetic image in a metal film, with field enhancements of two or more orders of magnitude possible. Additionally, by arranging the nanoelements into a periodic array on a film surface, it is possible to achieve efficient grating coupling of the incident beams to the delocalized SPPs, while simultaneously exciting the LSPRs of the individual elements [35–37]. A specific example of nanowires (NW) coupled to a thin film is illustrated in Fig. 1. In this work, we show that the collective effect of the periodic NW arrangement, leading to the interplay between the LSPR and SPP modes, results in many orders of magnitude enhancement of both the peak intensity and the radiated power of the generated FWM light. Additionally, the presence of the interacting SPP and LSPR modes in the pump field, while strongly enhancing both the evanescent and the propagating excitation regimes of the FWM field, can alter on demand the relative efficiency of the FWM generation in these two regimes, e.g., leading to a many orders of magnitude enhancement of the propagating modes of the generated FWM light.

2. Analytical description and numerical approach

Motivated by the geometry studied in [27], we assume two $p$-polarized pump waves at frequencies $\omega_1$ and $\omega_2$ incident at angles $\theta_1$ and $\theta_2$ on the film surface, respectively, as shown in Fig. 1. We consider the FWM process involving the frequencies $\omega_3 \equiv 2\omega_1 - \omega_2$ and $\omega_4 \equiv 2\omega_2 - \omega_1$, where the dominant contributions to the nonlinear polarization arise from the terms:

$$P^{\text{NL}}(\omega_3) = \varepsilon_0 \chi^{(3)}(-\omega_3; \omega_1, \omega_1, -\omega_2) \hat{E}_1 \hat{E}_1 \hat{E}_2^*,$$

$$P^{\text{NL}}(\omega_4) = \varepsilon_0 \chi^{(3)}(-\omega_4; \omega_2, \omega_2, -\omega_1) \hat{E}_2 \hat{E}_2 \hat{E}_1^*. \quad (1)$$

Here, $\hat{E}_i$ ($i = 1, 2$) are the electric field vectors of the incident beams located in the $xy$ plane and $\chi^{(3)}$ is the third-order nonlinear susceptibility tensor with the triple-dot sign denoting the summation over the relevant field and frequency components. Because we do not aim at analyzing the respective contributions of surface versus volume processes, we treat the to-
of nonlinear response as coming from the bulk with an effective third-order susceptibility of $\chi_{1111}^{(3)} = 0.2 \text{nm}^2/\text{V}^2$ [27], where the subscript represents any of the Cartesian coordinates $x$, $y$, or $z$ as shown in Fig. 1, and we assume an isotropic response for the gold medium with $\chi_{1111}^{(3)} = \chi_{1212}^{(3)} = \chi_{3131}^{(3)}$.

We begin by recounting the analytical description of FWM for waves scattering from a bare metal film, which we use to confirm the accuracy of our simulation approach. From momenta conservation and assuming air above a planar metal surface, the $x$-component of the propagation constant above the film for the field generated at $\omega_1$ can be found as $k_{1,x} = \sqrt{\omega_1^2/c^2 - k_{0,y}^2}$, where $k_{1,y} = 2\omega_1/c \sin(\theta_1) - \omega_2/c \sin(\theta_2)$. In the case of the bare film, $k_{1,y}$ has purely imaginary values over a range of pump wave angles $\theta_1$ and $\theta_2$ (we term this the evanescent regime); for these pump wave configurations, the generated FWM light is directed along the surface, with a momentum that coincides with that of the SPP. The expressions for the $x$- and $y$-components of the generated FWM field in the region above the metal surface are given by [14]

$$
E_x^R = -\frac{k^2}{\varepsilon_0} \left[ k_x^2 P_{NL} - \varepsilon k_x^{NL} \right] - k_0^2 \left( k_{x} P_{NL} + k_y^{NL} \right) - k_T^2 \left( k_y P_{NL}^{x_T} - k_y P_{NL}^{y_T} \right);
$$

$$
E_y^R = -\frac{k^2}{\varepsilon_0} \left[ k_y^2 P_{NL} - \varepsilon k_y^{NL} \right] - k_0^2 \left( k_{y} P_{NL} + k_x^{NL} \right) - k_T^2 \left( k_x P_{NL}^{x_T} - k_x P_{NL}^{y_T} \right),
$$

where the indices $R$ and $T$ refer to the waves for the generated FWM field, in air and inside the metal, respectively, and $S$ to the nonlinear source field inside the medium [14, 38]. A contour map showing the FWM efficiency at $\omega_1$ calculated according to Eq. (2) is shown in Fig. 2(a). An increase in the intensity of the generated FWM light is seen in the evanescent regions of the FWM process, consistent with the results described in [27].

To show the dramatic enhancements to $\chi^{(3)}$ that can be obtained with the NW-film system, which is not amenable to analytic approaches, we make use of full-wave, finite-element based simulations using Comsol Multiphysics [39]. The simulations are performed in two dimensions in the frequency domain, solving four coupled wave equations for the pump fields at $\omega_1$ and $\omega_2$ and for the generated fields at $\omega_1$ and $\omega_2$ coupled through the nonlinear polarization term [40].

To confirm our numerical model, we simulate the FWM for a bare gold film (in the absence of NWs) as a function of incident pump wave angle, with the angle of the second pump $\theta_2$ fixed at $46^\circ$, thus moving along the white line in Fig. 2(a). The enhanced FWM field is shown in Fig. 2(b) to be in excellent agreement with the analytical result (solid black). With the thickness of the gold film taken as 200 nm—much larger than the ~50 nm skin depth of gold over the frequency range considered—the film approximates an infinite half-space such that the bare film should be described by Eq. (2). To calculate the FWM efficiency, the maximum of the norm of the electric field is evaluated over the computational domain for each of the excitation angles. We additionally reproduce in Fig. 2(b) the result for the pump configuration employed in [27].

As seen in the solid (dash-dot) lines in Fig. 2(b), the efficiency of the FWM process is enhanced around $-23^\circ$ and $69^\circ$ ($-16^\circ$) (the transition regions between the propagating and evanescent regimes), in agreement with [27]. However, while there is some difference in the enhancement factors of the two curves, the peak efficiency is on the order of $10^6 \text{ V}^2/\text{m}^2$ with either pump wave configuration, indicating the efficiencies achievable by exploiting SPP enhancement alone. The difference in the values for the two curves is due to the dispersion of gold and the different pump wave arrangements. The particular choice of the wavelength and the angles in calculating Fig. 2 is dictated by the linear scattering spectrum of the NW-film system, as explained below.
3. Nanowire-film system

We now consider periodically arranged NWs coupled to the metal film. The linear scattering spectra for NWs spaced 0.6 nm from the film surface, with periods of either 880 nm or 70 nm NW, are shown in Fig. 3 for several excitation angles. Two spacings are chosen, one comparable and one much smaller than the excitation wavelengths. The choice of the particular lattice constants is arbitrary in each case and is taken for convenience in the simulations. With either period, an LSPR resonance is observed around 600 nm, characterized by a strong localized field enhancement of roughly two orders of magnitude (confined between the NWs and film). The position of this resonance is nearly angle-independent, being defined by the separation of the NW from the film, the material parameters, the NW diameter [34, 36, 37], and partially by the lattice period. A smaller period leads to larger NW concentration, increasing the effective refractive index above the film, hence blueshifting the resonance [37] for the case of the 70 nm periodicity. When the lattice constant of the NWs is on the order of the incident wavelength, Wood’s anomalies are produced [42, 43], characterized by narrow, asymmetrically-shaped spectral peaks, with the peak field similarly localized in the gap. The spectral positions of the Wood’s anomalies are strongly angle-dependent, in contrast with that of the LSPR whose peak remains fixed at 628 nm for all excitation angles.

The spectral characteristics of the periodic NW-film system are quite complex, and suggest numerous possibilities for enhancing the nonlinear processes. As a first example, we use the LSPR resonance (628 nm) for the wavelength of the first pump, sweeping its angle between $-90^\circ$ and $90^\circ$, while the second pump is placed at the first Wood’s anomaly peak (780 nm) at an angle of $46^\circ$, thus again following the white line in Fig. 2(a). The angular dependence of the intensity of the FWM field at $\theta_3$ ($\lambda_3 = 525$ nm) is shown in Fig. 4(a). The presence of the NW array and the optimized pump arrangement that exploits the resonances in the linear response of
the NW-film system leads to an enhancement of the intensity of the FWM field by six to eleven orders of magnitude over the angular range (solid green curve), in addition to the enhancements produced by the bare film in either pump configuration (dash-dot and dotted black lines).

For the dense NW array, both pump waves are positioned within the LSPR spectrum, at 588 nm and 600 nm, producing a FWM wave at 577 nm with a similar angle arrangement having $\theta_2$ fixed at 25°. The overall linear enhancement is smaller for the dense NW array, as seen in Fig. 3(b), leading to a somewhat reduced efficiency of the FWM process as compared with the sparse NW array. The overall angle dependence of the FWM process is very uniform in this case, also showing about a six to eight orders of magnitude increase compared to the plain film. The greater peak enhancements of the pump field seen in Fig. 3 for the sparse array can be attributed to the presence of a standing wave in the linear SPP spectrum that occurs for certain wavelengths relative to the array periodicity, leading to the constructive interference at the LSPR resonance. An example of such a standing-wave that appears as a linear response of the NW-film system when excited at the first Wood’s anomaly peak (780 nm) at 46°, is shown in the inset in Fig. 3(a). The coupling to this standing wave mode decreases to zero as the excitation angle approaches zero, however, because the component of the field normal to the surface vanishes [32] and the NW-film LSPR cannot be excited. By contrast, the broadband propagating mode formed between NWs in the dense array (the inset in Fig. 3(b)) can be excited by waves incident from any angle, leading to a FWM efficiency that is much more uniform over excitation angles.

While a strong enhancement of the FWM process for the NW-film geometry is observed in Fig. 4(a), the enhanced field is highly localized to the nanoscale regions between the NWs and the film. It is therefore of practical interest to estimate the available radiated power of the produced FWM field for each of the configurations considered. To estimate the power available for extraction, we calculate the power flow of the generated FWM field across the boundaries of the simulation domain. Equal domains with the size of 1,760 nm along the film and 950 nm normal to the film are used in all cases, with the integration performed over the boundary...
parallel to the film and one of the boundaries normal to the film (the second boundary normal to the film provides the same power flow with an opposite sign due to the periodicity).

The available power flow of the generated FWM field is shown in Fig. 4(b). For the NW-film array, the propagating plasmon at frequency $\omega_3$ can be excited in either the forward or backward direction along the film, depending on the excitation angle and the NWs period: the grating-like effect of the periodic NW arrangement adds an additional momentum along the metal surface. Thus, to estimate the total radiated power, we integrate the absolute values of the power flow over each of the boundaries, independent of the direction along the film. As seen from the figure, the radiated power is enhanced by up to six orders of magnitude in all cases. In addition, the dense NW array provides a similar angular distribution of the FWM efficiency as that for the bare film. However, as seen from Fig. 3(b), in this case stronger enhancement of the radiative power flow is produced in the propagating rather than the evanescent excitation regime (the region between -20° and 50° on the dashed blue curve), in contrast with the plain film case, while both regimes are enhanced by up to four orders of magnitude compared to the plain film.

4. Conclusions

In conclusion, we have shown that it is possible to enhance - by many orders of magnitude - FWM processes using an array of nanowires strongly coupled to a metal film. Both an increase of up to eleven orders of magnitude in the localized fields and of up to six orders of magnitude...
in the radiative power of the generated light is observed. The strongest enhancement is observed when the local enhancement associated with an LSPR intermingles with the enhancement from the delocalized SPP; both modes can interact through the arrangement of NWs periodically on the surface of a metal film. The enhancement can be angle-uniform over the broad range of angles, while the engineering of the collective effect of the periodical arrangement can alter on demand the relative efficiency of the FWM generation in the propagating versus the evanescent regimes. Given the numerous resonances occurring in the spectrum of the linear response of the structure to the applied fields, tuning of the angles of the incident pump beams can lead to even greater enhanced nonlinear processes, especially where the SPP or Wood’s anomalies interfere constructively with the LSRP of the NWs, forging a path towards nonlinear media with vastly enhanced properties.

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