Second-Harmonic Enhancement from a Nonlinear Plasmonic Metasurface Coupled to an Optical Waveguide

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ABSTRACT: Metasurfaces are commonly constructed from two-dimensional arrangements of nanoresonators. Coherent coupling of the nanoresonators through extended photonic modes of the metasurface results in a modified collective optical response, and enhances light–matter interactions. Here we experimentally demonstrate that strong collective resonances can arise also from coupling the metasurface to an optical waveguide. We explore the effect this waveguide-assisted collective interaction has on second-harmonic generation from the hybrid system. Our measurements indicate an enhancement factor of 8 for the transmitted second harmonic in comparison to incoherent collective scattering. In addition, complementary simulations predict about a 100-fold enhancement for the second harmonic that remains confined inside the waveguide. The ability to control the hybrid modes by the waveguide’s design provides broader control over the formation of the collective interaction and new tools to tailor the nonlinear interactions. Our findings pave a promising direction to realize nonlinear photonic circuits with metasurfaces.

KEYWORDS: metasurface, waveguide, nonlinear, collective scattering, guided-mode resonance, guided lattice resonance

In recent years much effort has been devoted to the research of nonlinear optical metamaterials and nonlinear metasurfaces. Various frequency conversion processes utilizing nonlinear metasurfaces have been reported, including second- to high-harmonic generation. Also, difference frequency generation processes were shown to yield terahertz radiation, for example, and even to generate entangled photon pairs. Moreover, the ability to control the optical response on a microscopic scale through a precise design of the metasurface provides means to modulate the nonlinear wavefront.

Although metasurfaces were found to be compelling compact and versatile platforms for tailored nonlinear optical interactions, low total conversion efficiencies hinder their adoption for technological applications.

In general, the optical response of a metasurface is determined by both the single nanoresonator’s properties and the collective interactions between different lattice sites. When the lattice spacing is approaching the effective wavelength in the host medium, resonant collective scattering between lattice sites dramatically alters the optical response of the array. This is known in the literature as a Rayleigh–Wood anomaly (RA), which is when a diffraction order is on the edge between a radiating to an evanescent mode. This type of optical anomaly is associated with sharp spectral features, as the collective scattered fields coherently build into a surface wave. In a metasurface made out of resonant nanoantennas, the localized modes and the distributed surface mode at the RA condition can hybridize, to form a surface lattice resonance (SLR). These hybrid modes were found to significantly enhance nonlinear wave mixing. Yet, to fully harness the potential of these modes, the metasurface needs to be placed in a homogeneous dielectric background. When the integration of metasurfaces in compact optical systems is considered, scattered fields from inhomogeneities reduce the quality of the RAs. This imposes some limitations and restrictions for incorporating collective scattering effects in metasurface integrated systems, such as photonic circuits.

When a diffractive periodic array is in optical contact with a waveguiding structure, diffraction orders may couple to guided modes. This coupling results in sharp spectral features associated with the leaky modes known as guided mode resonances (GMRs). For arrays of subwavelength scatterers, this means coherent scattering that is mediated by the guided modes. While gratings have been extensively used to couple light in and out of waveguides, the interactions between localized modes in metasurface and propagating GMRs have been left relatively unexplored. Such interactions lead to mode hybridization similar to the SLRs found when localized
modes are coupled to RAs. In the literature these hybrid modes are sometimes referred to as waveguide-plasmon polaritons.\(^\text{23}\) Since this collective phenomenon is not unique to plasmonics, we prefer to use the more general term guided lattice resonance (GLR) instead. GLRs were found to enhance fluorescence\(^\text{24,25}\) and even stimulate lasing.\(^\text{26}\) GMRs by themselves were reported to enhance nonlinear optical interactions, such as second\(^\text{27,28}\) and third-harmonic\(^\text{29}\) generation, or to achieve ultrahigh-quality linear and nonlinear metasurfaces.\(^\text{30,31}\) However, in these works the nonlinearity originates from the susceptibility of the waveguide’s bulk media and from surface effects, but not from the metasurface’s nonlinearity. Here, we study experimentally and numerically the effect of nonlinear GLRs on the enhancement of second-harmonic generation (SHG) emitted to free space and how these modes, in contrast to SLRs, are insensitive to the interparticle spacings but not from the metasurface.

When a plane wave is incident upon the metasurface the diffraction orders are determined by the conservation of parallel momentum

\[
k_{m_1,m_2}^\parallel = k_{\text{inc}}^\parallel + m_1 b_1 + m_2 b_2
\]

where \(b_{1,2}\) are the primitive reciprocal lattice vectors, \(m_{1,2}\) are integers, \(k_{\text{inc}}^\parallel\) and \(k_{m_1,m_2}^\parallel\) are the incident and the diffracted wave vectors, respectively, and the superscript \(\parallel\) stands for the vector’s projection on the metasurface plane. If the metasurface is in optical contact with a planar waveguide, a GMR is formed when

\[
k_{m_1,m_2}^\parallel = \beta_M
\]

where \(\beta_M\) is the \(M\)th-order guided mode’s propagation constant. This condition is schematically described by the arrows in Figure 1a. For a waveguiding slab, finding \(\beta_M\) requires solving transcendental equations for both the transverse electric (TE) and transverse magnetic (TM) polarizations

\[
\text{TE: } \tan(\kappa_M h) = \frac{k_M(\gamma_M + \delta_M)}{k_M^2 - \delta_M \gamma_M} \quad (3)
\]

\[
\text{TM: } \tan(\kappa_M h) = \frac{\varepsilon_{\text{core}} k_M^2 (\gamma_M \varepsilon_{\text{sub}} + \delta_M \varepsilon_{\text{sup}})}{k_M^2 \varepsilon_{\text{sub}} \varepsilon_{\text{sup}} - \varepsilon_{\text{core}}^2 \delta_M \gamma_M} \quad (4)
\]

where \(\kappa_M = \sqrt{\varepsilon_{\text{core}} k_M^2 - \beta_M^2}\), \(\gamma_M = \sqrt{\varepsilon_{\text{core}}^2 k_M^2 - \beta_M^2}\), \(\delta_M = \sqrt{\beta_M^2 - \varepsilon_{\text{sub}} k_M^2}\), \(h\) is the slab’s thickness, \(k_0\) is the wavenumber in vacuum, and \(\varepsilon_{\text{core}}\), \(\varepsilon_{\text{sub}}\), and \(\varepsilon_{\text{sup}}\) are the permittivities of the core, substrate, and superstrate, respectively. Each guided mode may couple to multiple diffraction orders, resulting in a large number of supported GMRs. The coherent scattering at the metasurface, together with the near-field enhancement associated with guided modes can be beneficial for nonlinear wave-mixing processes such as SHG. In general, the nonlinearities may originate from each of the hybridized system’s constituents. However, in this work we focus on the case where the quadratic nonlinearity originates from the metasurface.

The studied system of a metasurface—waveguide hybrid is schematically described in Figure 1a. The waveguiding layer is made of a 320 nm thick TiO\(_2\) film, sputtered on a fused silica substrate. A 100 \(\times\) 100 \(\mu\)m\(^2\) metasurface of gold split-ring resonators (SRRs) was fabricated on top of the waveguide by a conventional e-beam lithography technique. Figure 1a,b illustrates the SRRs’ shape, dimensions, and lattice spacing and Figure 1c presents a scanning electron microscope (SEM) image of the metasurface. The TiO\(_2\) surface roughness led to some irregularities of the fabricated SRRs’ shape; thus, the dimensions presented in Figure 1b are the typical mean values measured from multiple scanning electron microscope images. The meta-atoms were fabricated in a rectangular lattice, with interparticle spacings \(a_y = 550 \text{ nm}\) and \(a_x = 260 \text{ nm}\), so as to support diffraction in \(y\) and suppress the diffraction in \(x\). The guided modes’ dispersions were evaluated using eqs 3 and 4 with the substrate’s refractive index taken from the literature.
and the evaluated refractive index of the sputtered TiO$_2$ (see Supporting Information). The light lines and the modes’ dispersion are presented in Figure 1d, where the reciprocal lattice vectors are marked by dashed vertical lines. The intersections of these lines with the guided modes’ dispersion and light lines represent the GMR and RA conditions, respectively. By taking oblique incidence angles, parallel momentum is added/subtracted to shift these intersections and provide the means to spectrally tune the GMR. Noncentrosymmetric SRRs were chosen for the metasurface elements, as they were shown to support strong quadratic nonlinearities and have already been investigated extensively in the context of SHG.$^{3,7,33}$ When the pump is polarized parallel to the base of the SRR, it excites the LSPR at the fundamental

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**Figure 2.** Angle- and polarization-resolved transmission measurements where the reference measurements are made from a region with no metasurface. The left and right panels show the transmission for the TE and TM polarizations, respectively. The black dashed lines show the GMRs of zeroth-order guided modes and the dotted-dashed lines those of the first guided mode. The GMRs were labeled by the guided mode’s order (subscript) and polarization and the diffraction order (superscript). The white dotted lines mark the LSPRs excited in SRRs for each polarization. Values slightly exceeding 100% in the TM polarization are due to some random spectral noise in the illumination source.

**Figure 3.** Transmitted second harmonic. (a) Measured transmitted SH as a function of the pump’s incident angle and wavelength. The measured photon count was normalized by the pump power squared. (b) Simulated transmitted SH normalized by the total SH from the same metasurface without the waveguide, to give an evaluation of the SHG enhancement. The white dashed lines represent the dispersion of the TE GMRs. The dotted-dashed lines represent the dispersion of the TM GMR modes corresponding to the SH wavelength $\lambda_{pump}/2$. 

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The azimuthal angle measurements (see the Supporting Information). The enhancement to promote the nonlinear wave-mixing.

The expected, the high refractive-index contrast between the manifested by the splitting of the transmission dips. As the GMR spectrally overlaps with a LSPR, which are sensibly polarized, parallel to the SRR arms. This cross-polarization behavior is an indication that the SH originates from the metasurface and not from the dielectric interfaces of the waveguiding stratified structure. The resonators’ dimensions can be adjusted to tune the LSPRs to the frequencies of interest and benefit from near-field enhancement to promote the nonlinear wave-mixing.

The first step in characterizing the hybrid system was to perform angle- and polarization-resolved linear transmission measurements (see the Supporting Information). The azimuthal angle was kept constant at 90°, while was varied to sweep over the parallel momentum . The measured transmission spectra are presented in Figure 2, along with the calculated, angle-dependent GMR dispersion. Each mode is labeled by a subscript marking the guided modes order and a superscript labeling the different GMR-related spectral features. Additionally, we may notice how TE or TM GMRs interact with different LSPR modes. GLRs are formed when the GMR spectrally overlaps with a LSPR, which are manifested by the splitting of the transmission dips. As expected, the high refractive-index contrast between the waveguide’s core and the superstrate diminish RA-related features in the transmission spectra.

To characterize the way the GLRs affect the SHG, the sample was pumped in TE polarization by a tunable femtosecond laser with an average power of 200–300 mW and pulse length of 140 fs (see Supporting Information for additional details). The measured average SH photon counts were corrected by the quantum efficiency of the detector and were normalized by the square of the pump’s power. Figure 3a presents the TM-polarized SH measured at the zeroth-order transmission. The dispersions of the GMRs were added with the TM modes corresponding to SH wavelengths at while the TE mode corresponds to . The region of maximum SHG follows the dispersion of the TE- GMR, which means that it is predominantly enhanced by resonances at the FF. When the angle increases, the resonance for the FF red-shifts to better overlap with the LSPR of the SH. This overlap leads to an estimated enhancement factor of 8, relative to the results obtained at normal incidence, where the GMRs are distant and barely contribute. These findings are comparable to the reports for RA SLRs at the pump frequencies. Additionally, GMR features related to SH frequency, i.e. the TM modes, appear as dips that indicate the coupling of the generated SH to the waveguide.

To gain a better understanding of the nonlinear dynamics, we performed full-wave simulations using a commercial solver with the hydrodynamic model as the source of the nonlinear generation. From the simulation at the frequency of the pump () the linear polarization (P) can be found. Using the hydrodynamic model, the relation between the induced nonlinear surface currents (K) in the plasmonic nanoresonators to the linear polarization is approximated by

\[
K_NL = \frac{i \omega c}{\eta_0} \left[ \mathbf{n} (p_{1}^* p_{2}) \right] + \mathbf{n} \frac{1}{2} \left[ \frac{3 \omega + i \gamma}{2 \omega + i \gamma} (p_{1})^2 \right]
\]

where \( \eta_0 = 5.7 \times 10^{28} \text{ m}^{-3} \) is the electron density, \( \gamma = 1.07 \times 10^{14} \text{ s}^{-1} \) is the phenomenological damping rate, and \( \mathbf{n} = \mathbf{t} \) are the unit vectors pointing perpendicular and parallel to the metallic surface, respectively. Similarly, superscripts of \( P \) indicate the polarization component perpendicular and parallel to the metallic surface. These nonlinear currents serve as the radiative source for the simulation at the SH frequency. To
evaluate the enhancement factor, the transmitted SH in the
simulation was normalized by the results obtained for the same
metasurface on a semi-infinite TiO₂ substrate. In the simula-
tions, periodic boundary conditions defined a unit cell
that follows the lattice spacing mentioned in Figure 1a. The
SRR, with the dimensions mentioned in Figure 1b, was
positioned in the center of the unit cell. The resulting LSNRs
were red-shifted by about 30 nm in comparison to those in the
measurements; these led to some deviations of the SHG
features presented in Figure 3b. The enhancement seen near
λ_{pump} = 1.2 μm at small angles is related to the red-shift of the
GLR at λ_{pump}'. The steep spectral feature starting at ~5° is the
free space RA at the superstrate. It is not captured in the
experiment due to the imperfections of the fabricated
waveguide and SRRs. Other RA-related features do not appear
at all, as expected due to the high refractive index contrast at
the metasurface’s plane. The overall resemblance to the
experimental results validated the simulations, which provided
us with the means to probe the near fields.

We used the simulations to qualitatively study how the
collective interaction affects the SHG coupled to the
waveguide. Figure 4a shows the normalized SHG power
carried by the guided waves as a function of incidence angle and
wavelength. Since the SH is coupled through the diffraction
orders, it can couple to counterpropagating TM modes.
Therefore, the sign and color in Figure 4a indicate the
direction of the power flow. It can be seen how the
enhancement factor may reach up to 2 orders of magnitude.

The upper panel shows a cross-section at of this enhancement
at λ_{pump} = 1.29 μm. Figure 4b reveals the normalized TM field
(H_y) of the generated SH in the unit cell for λ_{pump} = 1.32 μm
and at x = \frac{a}{2} for two different angles (stated at the panels’
upper right corners). The field profiles match the familiar
mode profiles from guided-mode theory and validate how
SHG feeds the TM modes. Overall they demonstrate how
coupling of the SHG to the GMRs leads to an enhancement
inside the waveguide by 2 orders of magnitude, which is 1
order of magnitude larger than the SH emitted to free space by
the same system and also exceeds those in reports for the
enhancement obtained from RA-based SLRs.13,15 Additionally,
the guided mode profiles, described by the white lines in
Figure 4b, reveal how the coupling of nonlinear metasurface
was obtained by placing it at the evanescent tail of the guided
modes. A thoughtful design, in which the metasurface is better
positioned relative to the guided modes’ profile, may lead to an
even stronger enhancement of the nonlinear emission into the
waveguide.

To conclude, we have demonstrated how a metasurface in
optical contact with a planar waveguide has additional channels
available to achieve coherent scattering between lattice sites.
This occurs through the coupling of the metasurface’s
diffraction orders to the guided modes. When these GMRS
spectrally overlap with LSNRs, it results in polarization-
dependent GLRs. These, in contrast to the RA-based SLRs,
is insensitive to index matching and do not require a
homogeneous dielectric environment. The GLRs provide
similar enhancements of the SH emitted to free space, in
comparison to SLRs. Moreover, simulations predict an
additional order of magnitude increase in the enhancement of
the SH confined to the waveguide. This enhancement may
be attributed to two mechanisms. The first is the increase in
effective polarizability of the nonlinear SRRs due to the
collective resonances. The second is the near-field enhance-
ment in the vicinity of the waveguide upon excitation of a
guided mode20 that can occur for both the FF and the SH. The
increase in effective polarizability and the stronger near fields
enhance the light–matter interactions and the nonlinear
conversion process. On the basis of these results, together
with the large number of design degrees of freedom in the
hybrid system, even higher enhancements in the wave-mixing
conversion process may be achieved. Eventually, combining
nonlinear metasurfaces with optical waveguides provides new
means to infuse future photonic devices with nonlinear
interactions.

ASSOCIATED CONTENT
Supporting Information
The Supporting Information is available free of charge at
https://pubs.acs.org/10.1021/acs.nanolett.1c04584.
Refractive indices used for both the calculation of the
guided modes’ dispersion and simulations and details
and description of the experimental setups and methods
used for the linear and second-harmonic measurements
(PDF)

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Notes
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