SUPPLEMENTARY INFORMATION

Efficient unidirectional polarization-controlled excitation of surface plasmon polaritons

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Details on modelling, fabrication and characterization

Modelling. All modelling results are performed using the commercial available finite element software Comsol Multiphysics, ver. 4.3b. For 1D-periodic SPP couplers, the configuration and the incident Gaussian beam are assumed to be invariant along the \( y \)-direction (Figure 1a and 1c), hereby reducing the problem to a 2D calculation. The coupling efficiency and directivity are obtained by integrating the \( x \)-component of Poynting’s vector on vertical 3-\( \mu \)m-long lines positioned 15 \( \mu \)m to the left and right of the SPP coupler. The coupling efficiency corresponds to the power flowing in the \( +x \)-direction relative to the incident power, whereas the directivity is the ratio of power flowing in the \( +x \)- and \( -x \)-direction. Note that the coupling efficiency is corrected for the exponential decay of the power of the SPP as it propagates from the coupler to the evaluation point. For 2D-periodic SPP couplers and metascatterers, we resort to full 3D calculations. However, in the attempt to quantify the performance of the SPP coupler consisting of 6\( \times \)6 super cells and to reduce the size of the computational domain, we treat each row of the super cell (Figure 2c) as its own super cell, assuming the new couplers to be infinitely extended (i.e., periodic) in the \( y \)-direction and six super cells wide in the \( x \)-direction. Accordingly, the \( x \)-polarized Gaussian input beam is taken to be invariant along the \( y \)-direction. The SPP power flowing in \( \pm x \)-direction is evaluated 10 \( \mu \)m away from the SPP coupler. Regarding 1D- and 2D-periodic homogeneous GSP-based metasurfaces (Figure 1a and Figure 2a, respectively), we model one unit cell by applying periodic boundary conditions on the vertical sides of the cell. The complex reflection coefficients (Figures 1b and 2b) are determined on the top surfaces of the nanostrips/nanobricks when the impinging light is linearly polarized and normal to the surface. In our gold-glass-gold configurations, the permittivity of gold is described by interpolated experimental values\(^1\) while glass, assuming to be silicon dioxide, takes

on the constant refractive index $n = 1.45$. The medium above the configurations is chosen to be air. The air domain is truncated using perfectly matched layers to minimize reflections, while a perfect electric conductor boundary condition is applied on the bottom side of the optically thick gold film.

**Fabrication.** The investigated samples are composed of a 170-µm-thick glass slide covered with a 3-nm-thin titanium adhesion layer and an 80-nm-thick gold film. Onto the gold film we deposit a 50-nm-thick phosphosilicate polymer (Honeywell ACCUGLASS P-5S spin-on-glass) with refractive index 1.45 in the near-infra red as obtained by ellipsometry measurements (Jobin Yvon Horiba UVISEL). The SPP couplers are defined onto the spin-on-glass polymer by electron-beam lithography. A 200-nm-thick PMMA film is exposed, using a JEOL JSM-6500F field emission scanning electron microscope (SEM) equipped with a Raith Elphy Quantum lithography system, at 20 kV acceleration voltage followed by deposition of a 3-nm-thick titanium adhesion layer and 50-nm-thick gold film. In the case of glass spacers located only below the strips/nanobricks (Figure 5a), we do not use a spin-on-glass approach, but instead we deposit 3 nm of titanium and 50 nm of silica by thermal evaporation prior to electron-beam evaporation of 3 nm of titanium and 50 nm of gold.

**Optical characterization.** In the setup used for leakage radiation microscopy (LRM), a collimated and polarized incident beam with Gaussian intensity distribution from a fiber-coupled tunable laser source (Anritsu OSICS, wavelength range 1500-1620 nm) is focused by an objective O1 onto the SPP coupler in order to excite the SPPs (Figure 3c). The SPP propagation is then visualized by collecting the leakage radiation from the sample plane with a high N.A. oil-immersion objective O2 which is thereafter projected onto a cooled IR CCD-camera (Xenics xeva-651) by the microscope tube lens, thereby enabling the assessment of propagation length $L_p$, coupling efficiency $C$, and directivity $D$. As stated in the main text, the intensity distribution of the SPP in the transverse direction of propagation can be approximated with a Gaussian function in which the product of the
maximum intensity $I_x$, and the beam waist $w_x$, approximately satisfies the relation

$$I_x w_x = A \exp(-u / L_p),$$

where $A$ is a constant, $u = |x - x_0|$, and $x_0$ is the $x$-coordinate of the excitation spot; $A$ and $L_p$ are found by a linear fit to $\ln(I_x w_x)$ (Figure 4b).

In order to evaluate the coupling efficiency, one must estimate both the direct transmitted power of the incident Gaussian beam, $P_T$, and the leakage radiation power, $P_{LR}$, transmitted by the propagating SPP, respectively. The power transmitted by the incident Gaussian beam is evaluated by positioning the beam on the sample away from the metasurface and is given by

$$P_T = I_0 \pi w_0^2,$$

where $I_0$ and $w_0$ are the peak intensity and waist and are estimated by fitting a Gaussian function to the transmitted beam cross section. On the other hand, the leakage radiation power in the $+x$-direction can be expressed as:

$$P_{LR} = \int \int I_x(u) \exp\left(-\frac{u^2}{w_x^2(u)}\right) du dy = \sqrt{\pi} \int_0^\infty I_x w_x du = \sqrt{\pi} A \int_0^\infty \exp(-u / L_p) du = \sqrt{\pi} AL_p.$$

Furthermore, one must account for the loss ratio between absorption in the gold film and leakage radiation:

$$\Gamma = \frac{\text{Im} \left[ n_{\text{app}}^{(1)} \right]}{\text{Im} \left[ n_{\text{app}}^{(2)} \right] - \text{Im} \left[ n_{\text{app}}^{(1)} \right]},$$

where $n_{\text{app}}^{(1)}$ and $n_{\text{app}}^{(2)}$ are the effective mode indexes of the SPP without and with leakage radiation losses, respectively (i.e., gold film of infinite and finite thickness, respectively). For a 80-nm-thick gold film with a 50-nm glass layer at $\lambda = 1500$ nm, the loss ratio is $\Gamma \approx 36$. Since the leakage radiation power is related to the power carried by the SPP, $P_{SPP}$, by $P_{LR} = P_{SPP} / (1+\Gamma)$, with $P_{SPP} = C*P_{in}$ and $P_{in} = P_T / T$, where $T$ relates the incident and transmitted beam powers, we can finally define the coupling efficiency as:

$$C = \frac{T(1+\Gamma)P_{LR}}{P_T}.$$
Wavelength sensitivity of 1D- and 2D-periodic SPP couplers

Despite the fact that surface plasmon polariton (SPP) couplers in this work are designed for efficient and unidirectional excitation of SPPs at wavelength $\lambda=1500$ nm, optical characterization of the fabricated couplers demonstrates a rather broadband performance. This is illustrated in Fig. S1 for the 1D-periodic SPP coupler (see Figure 3a in the main text), where it is evident from the LRM-images that SPPs are dominantly excited in the $+x$-direction for the wavelength range 1500-1600 nm. As expected, the coupling efficiency, $C$, and directivity, $D$, decreases for increasing wavelength, but the deterioration of the performance is not dramatic, showing $C \approx 19\%$ and $D \approx 43$ at $\lambda=1600$ nm. The same kind of moderate wavelength sensitivity is seen for polarization-controlled unidirectional excitation of SPPs using 2D-periodic coupler (Fig. S2). LRM-images clearly demonstrate how orthogonal polarized light excites SPPs in orthogonal directions, with a slight decrease in coupling efficiency and directivity for increasing wavelength, resulting in $C \approx 16$ and $D \approx 44$ for $\lambda=1600$ nm.

Fig. S1. Optical characterization of 1D-periodic SPP coupler. (a,c) Recorded leakage radiation microscopy (LRM) images for a free-space wavelength of 1550 nm and 1600 nm, respectively. For each image, the coupling efficiency, $C$, propagation length, $L_p$, and directivity, $D$, are indicated. The Gaussian input beam with spot radius $w_0=2.8\pm0.3$ $\mu$m is in each case positioned to optimize the coupling efficiency and directivity. (b,d) Variation of the product of SPP intensity and beam waist, $I_w w_0$, on a logarithmic scale as a function of the distance $u = |x - x_0|$ where $x_0$ is the excitation spot $x$-coordinate. The first order polynomial fitted to the data points (blue curves) is used to evaluate the coupling efficiency and the propagation length (see the Methods section in the main text).
Fig. S2. Optical characterization of 2D-periodic polarization-controlled SPP coupler. Recorded LRM-images for x- and y-polarized incident light at a free-space wavelength of (a,b) \( \lambda = 1550 \text{ nm} \) and (c,d) \( \lambda = 1600 \text{ nm} \). For each image, the coupling efficiency, \( C \), propagation length, \( L_p \), and directivity, \( D \), are indicated. The Gaussian input beam with spot radius \( w_0 = 2.8\pm0.3 \mu m \) is in each case positioned to optimize the coupling efficiency and directivity. Scanning electron microscopy (SEM) image of the SPP coupler can be seen in Figure 3b in the main text.

Pillar-shaped gap-plasmon resonators and SPP couplers

As discussed in the main text, gap-surface plasmon (GSP) resonators and the associated SPP couplers can also be made with the dielectric material located only below the nanostructures and not as a continuous layer atop the gold film. In the following, we are going to discuss the design, fabrication, and optical characterization of these pillar-shaped SPP couplers, beginning with the 1D-periodic case. The 1D-periodic SPP coupler is designed by considering the gold-glass-gold metasurface in Fig. S3a for which the unit cell size is \( \Lambda = 498 \text{ nm} \), corresponding to one-third of the SPP wavelength at free-space wavelength \( \lambda = 1500 \text{ nm} \). Next, we fix the glass and gold strip thicknesses to 50 nm and calculate the complex reflection coefficient of the metasurface for a normal incident TM-polarized plane wave (Fig. S3b). Clearly, we can construct a super cell of size \( \lambda_{spp} = 3\Lambda \) with a linear reflection-phase gradient by chosen the three strip widths marked with dotted
Fig. S3. Design of 1D couplers for efficient and unidirectional SPP excitation. (a) Sketch of 1D-periodic GSP-based metasurface. The incident field is TM-polarized (i.e., x-polarized) and propagates normal to the surface. (b) Amplitude and phase of reflected light from metasurface in a as a function of strip width \( w \) when \( \lambda = 1500 \) nm, \( \Lambda = 498 \) nm, and \( d = t = 50 \) nm. Vertical dotted lines indicate the three chosen strip widths (with phase difference of \( 2\pi/3 \)) constituting a super cell in the SPP coupler. The time convention is \( \exp(-i\omega t) \). (c) Electric field of the incident x-polarized Gaussian beam overlaid by a sketch of the SPP coupler. The SPP coupler consists of 6 super cells, corresponding to an overall size of \( L_c = 18\Lambda = 9.0 \mu m \), and the Gaussian beam is centered at \( x_0 = 0.67L_c \) with a beam radius of \( w_0 = 3 \mu m \). (d) \( z \)-component of the electric field, corresponding to the transverse electric field component of SPPs.

vertical lines. The three strip widths, corresponding to \( w_1=60 \) nm, \( w_2=290 \) nm, and \( w_3=342 \) nm, are characterized by an equidistant three-step discretization of the \( 2\pi \) phase space. We now numerically study a realistic 1D-periodic SPP coupler consisting of six super cells being illuminated by a normal incident Gaussian beam of beam radius 3 \( \mu m \) and positioned in such a way to maximize the excitation efficiency of SPPs in the +x-direction (Fig. S3c and S3d). The SPP coupler is characterized by a coupling efficiency \( C \approx 36\% \) and directivity \( D \approx 14 \), which is somewhat lower than obtained for the non-optimized 1D-periodic SPP coupler (with a continuous glass layer) in the main text (see Figure 1). We believe that the difference in performance is related to the weakly bound SPP for an air-gold interface at \( \lambda = 1500 \) nm, allowing the mode to easily scatter-off as it propagates.
within the SPP coupler. In order to improve the performance of the 1D-periodic SPP coupler, we have iteratively changed the unit cell period and the strip widths, reaching $C \approx 47\%$ and $D \approx 186$ for $\Lambda = 468$ nm, $w_2 = 284$ nm, and $w_3 = 350$ nm.

We now consider the design of polarization-controlled 2D-periodic SPP couplers, following the procedure outlined in the main text. In the quest to design a birefringent GSP-based metasurface with orthogonal phase gradients for orthogonal polarizations and super cell periodicity equal to the SPP wavelength, we calculate the complex reflection coefficient of a (homogeneous) metasurface with the unit cell in Fig. S4a for both $x$- and $y$-polarized incident light, using the nanobrick widths,
Fig. S5. Fabricated pillar-shaped plasmonic couplers. (a,b) SEM-images of a fabricated 10 µm long 1D-periodic SPP coupler composed of 6 super cells with design period and strip widths taken from the optimized theoretical design, and b 2D-periodic polarization-sensitive SPP coupler composed of 4×4 super cells (see Fig. S4c).

$L_x$ and $L_y$, as geometrical parameters to control the reflection phase (Fig. S4b). The appropriate super cell, shown in Fig. S4c, is found by realizing that with the reflection phase being discretized in steps of $2\pi/3$, the nanobricks possessing the proper reflection phases for both polarizations correspond to the intersections marked with circles in Fig. S4b. The polarization-controlled excitation of SPPs is verified in Fig. S4d, displaying the $z$-component of the E-field 400 nm above the nanobricks when a Gaussian beam, being ether $x$- or $y$-polarized, impinges on a single super cell. In order to estimate the coupling efficiency and directivity of the 2D-periodic SPP coupler consisting of 6×6 super cells, we follow the approach described in the main text for $x$-polarized light in which each row of the super cell (Fig. S4c) is treated as its own super cell with the purpose of reducing the computational demand. Such a series of calculations for a Gaussian input beam with beam radius $w_0=3$ µm result in coupling efficiencies and directivities in the range $C \approx 38-39\%$ and $D \approx 22-23$, respectively, which, due to symmetry, also applies to $y$-polarized incident light.

We now move on to the proof-of-principle experimental verification of the above designed SPP couplers. The structures are fabricated using a single step of electron beam lithography (Fig. S5); however, unlike the case of continuous glass spacer, the GSP resonators are created by depositing both glass and gold in the resist profile. The outcome is GSP-resonators with slightly inclined side walls, making the actual dimensions of the fabricated strips and nanobricks smaller than design values. That said, the 1D-periodic SPP coupler in Fig. S5a has been characterized with
Fig. S6. Efficient unidirectional polarization-controlled SPP excitation. Optical characterization of (a,b) 1D-periodic SPP coupler (Fig. S5a) and (c,d) 2D-periodic polarization-controlled SPP coupler (Fig. S5b). a,c,d Recorded LRM-images for a free-space wavelength of 1500 nm. The input beam is in each case positioned to optimize the coupling efficiency and directivity, and in a \( w_0 = 2.8 \) µm whereas in c,d \( w_0 = 1.7 \) µm. b Variation of the product of SPP intensity and beam waist, \( I_w \), on a logarithmic scale as a function of the distance \( x' = |x-x_0| \) where \( x_0 \) is the excitation spot x-coordinate. The first order polynomial fitted to the data points (blue curve) is used to evaluate the coupling efficiency and the propagation length.

LRM, demonstrating a reasonably high coupling efficiency \( C \sim 22\% \) and directivity \( D \sim 10 \) (Fig. S6a). The slightly reduced performance compared to coupler with continuous layer of glass (Figure 4a in the main text) is related to the difficulty in reaching the design dimensions and cross-sections (vertical sides). Note how the propagation of SPPs along the bare gold-air interface allows for propagation lengths of \( L_p \sim 210 \) µm which is in reasonable agreement with the theoretical value of 260 µm.

As a way of experimentally verifying the unidirectional polarization-controlled excitation of SPPs with the 2D-periodic coupler, whose super cell is depicted in Fig. S4c, we apply the LRM procedure for a coupler consisting of 4×4 super cells (Fig. S5b) when the incident Gaussian beam (beam spot is \( w_0 = 1.7 \) µm) is either x- or y-polarized (Fig. S6c and S6d). One notices the orthogonal excitation of SPPs for orthogonal polarizations, but it is clear that the usage of only 4×4 super cells together with too small nanobrick dimensions degrades the performance considerably compared to numerical estimations, demonstrating a coupling efficiency of \( C \sim 6-8\% \) and directivity of \( D \sim 4-9 \).
Regular grating couplers

In order to fully appreciate the advantages of employing SPP couplers based on phase-gradient metasurfaces, one should compare their performance with regular grating-based SPP couplers in the same configuration. As an example, we numerically study the regular grating depicted in Fig. S7a, which consists of six nanostrips on top of a glass spacer and gold substrate separated by $\Lambda=1467$ nm, corresponding to the SPP wavelength at the excitation wavelength $\lambda=1500$ nm. The other geometrical parameters are chosen to $t = d = 50$ nm in order to correlate with the gradient 1D-periodic SPP couplers in the main text, and the strip width $w$ is the free variable. Using a TM-polarized Gaussian incident field with beam radius $w_0 = 3$ $\mu$m, the maximum SPP coupling efficiency and associated directivity as a function of strip width were calculated (Fig. S7b). The curves are obtained by scanning, for each strip width, the input beam across the grating while noting when the excitation of SPPs propagating in $+x$-direction is maximum, i.e., maximum coupling efficiency. It is clear that the coupling efficiency is influenced by the strip width, with the maximum efficiency reaching $C \approx 21\%$ with a directivity of $D \approx 2$ at $w = 925$ nm, implying that $\sim 10\%$ of the incident power is converted into SPPs propagating in the “wrong” direction. Compared with the non-optimized 1D-periodic SPP coupler in the main text, showing $C \approx 40\%$ and $D \approx 53$, the best regular grating coupler demonstrates a significantly reduced amount of power being converted into SPPs propagating in the desirable direction and a rather poor directivity.

Fig. S7. Excitation of SPPs using regular grating coupler. (a) Sketch of regular SPP grating coupler consisting of six equally spaced gold nanostrips on top of a glass spacer and gold substrate. (b) Maximum SPP coupling efficiency and associated directivity as a function of strip width $w$ for fixed parameters $\Lambda=1467$ nm, $t = d = 50$ nm and incident wavelength $\lambda=1500$ nm. The incident field is a TM-polarized (i.e., $x$-polarized) Gaussian beam with beam radius $w_0 = 3$ $\mu$m propagating normal to the surface.