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Optical gap-surface plasmon metasurfaces for spin-controlled surface plasmon excitation and anomalous beam steering

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ABSTRACT: Multifunctional metasurfaces featuring diversified functionalities offer unprecedented possibilities for developing versatile ultracompact micro/nanophotonic devices and systems. Until recently, most multifunctional metasurfaces were designed for light incidence with orthogonal linear polarizations, being unable to function with orthogonal circularly polarized (CP) light, which is of vital importance in spin photonics, chiroptical spectroscopy/imaging, and optical sensing. Here we consider the design of efficient spin-decoupled multifunctional gap-surface plasmon (GSP) gradient metasurfaces, and experimentally demonstrate simultaneous spin-controlled unidirectional surface plasmon polariton (SPP) excitation and anomalous beam steering in the optical regime under orthogonal right and left CP light incidence, respectively. The spin-
decoupled GSP gradient metasurface, consisting of rotated GSP-based nanoscale half-wave plates, combines both propagation and geometric phases to produce two different spin-dependent linear phase gradients enabling SPP excitation and anomalous reflection. The proof-of-concept fabricated metasurface exhibits broadband (850-950 nm) operation featuring efficient (> 22%) unidirectional SPP excitation and high-efficiency (48% on average) anomalous beam steering for right and left CP incident light, respectively. Our demonstration of metasurface-enabled spin-controlled unidirectional SPP excitation along with free-propagating beam steering opens new possibilities for spin photonics and plasmonics with potential applications ranging from biomedical diagnosis, chiroptical spectroscopy and imaging to optical sensing.

**KEYWORDS:** gap-surface plasmon metasurface, spin-decoupled, multifunctional, phase gradient, surface plasmon excitation, beam steering
Metasurfaces offer unique possibilities for controlling electromagnetic (EM) fields at subwavelength scales and provide a highly flexible and versatile platform for future high-density micro-/nanophotonic devices and circuits.\textsuperscript{1-4} Numerous applications have been demonstrated in recent years, including metalenses,\textsuperscript{5,6} optical holograms,\textsuperscript{7,8} waveplates,\textsuperscript{9,10} polarimeters,\textsuperscript{11-13} waveguide couplers\textsuperscript{14,15} and mode converters,\textsuperscript{16} etc. However, to date, most of the metasurfaces were designed to exhibit a single functionality, which does not meet the current trend in photonics towards larger capacities and more functionalities within single compact-form-factor components. Thus, the development of multifunctional metasurfaces has become an increasingly important direction in nanophotonic researches, with many fascinating demonstrations being reported.\textsuperscript{17-27} Most of the aforementioned multifunctional metasurfaces operate with linearly polarized (LP) light, utilizing polarization-resolved phase responses in birefringent meta-atoms with precisely varied dimensions to impose independent phase modulations to orthogonal LP incident light.\textsuperscript{17,21-24,27} However, adjusting the dimensions of each meta-atom in two orthogonal directions is rather demanding for nanofabrication as $N$-level phases require $N$ different meta-atoms. Besides, these phase gradients are typically wavelength-dependent, resulting in limited operation bandwidths. In contrast, metasurfaces operating with circularly polarized (CP) light rely on the wavelength-independent geometric phases and can be designed to realize multiple functionalities by proper arrangement of differently oriented identical meta-atoms, thereby releasing fabrication challenges and broadening operation bandwidths.\textsuperscript{19,20,28} At the same time, the geometric phase has intrinsically opposite signs for orthogonal circular polarizations, resulting in locked and mirrored functionalities for right CP (RCP) and left CP (LCP) beams, such as free-space beam steering into symmetrical directions,\textsuperscript{29,30} surface plasmon polariton (SPP) excitation to opposite directions\textsuperscript{15,31} and vortex beam generation with opposite topological charges.\textsuperscript{19}
Very recently, the spin-locked limitations were released by combining both the propagation (size-dependent) and geometric (orientation-dependent) phases in each meta-atom, providing a general and practical guideline towards realization of spin-decoupled functionalities with a single metasurface for orthogonal CP states.\textsuperscript{18,32} Based on this approach, various spin-decoupled multifunctional metasurfaces have been realized, such as spin-multiplexing holograms,\textsuperscript{32,33} arbitrary spin-to-orbital momentum converters,\textsuperscript{34} spin-decoupled multifoci metalenses\textsuperscript{35,36} and spin-decoupled wavefront shaping and polarization conversion.\textsuperscript{37-40} However, the aforementioned spin-decoupled metasurfaces are mainly focused on molding free-space propagating waves. High-efficiency spin-decoupled manipulation of both propagating waves and surface waves remains so far largely unexplored, especially in the optical regime.

In this work, we consider the design of high-efficiency spin-decoupled multifunctional gap-surface plasmon (GSP) gradient metasurfaces, and experimentally demonstrate simultaneous spin-controlled unidirectional SPP excitation and anomalous beam steering in the optical regime under RCP and LCP light incidence, respectively, dictated by the corresponding spin-multiplexed linear phase gradients. The spin-decoupled GSP metasurface, consisting of rotated GSP-based nanoscale half-wave plates (nano-HWPs), combines both propagation and geometric phases to produce two different spin-dependent linear phase gradients enabling SPP excitation and anomalous reflection (Fig. 1). The proof of concept fabricated metasurface exhibits efficient (~ 28\% at 900-nm-wavelength) and broadband (1dB bandwidth ~ 100 nm) unidirectional SPP excitation under normal RCP light incidence, whereas for the LCP light, high-efficiency broadband (~ 48\% within 800-950 nm) beam steering is realized.
Figure 1. Schematic of the spin-decoupled multifunctional GSP gradient metasurface composed of rotated GSP-based meta-atoms, manifesting distinct linear phase gradients under normally incident orthogonal CP light beams. The red- and cyan-colored beams illustrate the spin-decoupled functionalities: unidirectional SPP coupling and anomalous beam steering under normally incident RCP and LCP light, respectively.

RESULTS AND DISCUSSION

Let us first introduce the underlying physical principles of combining both the propagation and geometric phases to realize independent control of arbitrary orthogonal polarization states\textsuperscript{32} and subsequently apply this approach to implement our design. Generally, the rotated meta-atom can be described by the Jones matrix under orthogonal CP polarization basis:\textsuperscript{32,34}

\[
R(\theta)^{tr} = \begin{bmatrix}
\frac{1}{2}(r_{xx} + r_{yy}) & \frac{1}{2}(r_{xx} - r_{yy})e^{-j2\theta}
\end{bmatrix}
\]

(1)

where \(r_{xx}\) and \(r_{yy}\) represent the complex reflection coefficients of the meta-atom under orthogonal \(x\) and \(y\) LP incidence, and \(\theta\) is the meta-atom’s orientation. To achieve high-efficiency geometric phase modulation, the reflection coefficients \(r_{xx}\) and \(r_{yy}\) should satisfy the relationship of \(r_{xx} = \)
- \( r_{yy} \),\(^{30}\) implying that the meta-atom should be designed as a nano-HWP. After introducing the LP reflection amplitude and phase explicitly \( r_{xx} = A e^{i\varphi} \), the Jones matrix \( R(\theta)^{lr} \) becomes simplified:

\[
R(\theta)^{lr} = \begin{bmatrix}
0 & A e^{i(\varphi + 2\theta)} \\
A e^{i(\varphi - 2\theta)} & 0
\end{bmatrix}
\]

(2)

where \( A \) and \( \varphi \) represent the magnitude and phase of the complex reflection coefficient under the \( x \)-polarized light incidence, respectively.

It turns thus out that, by independently adjusting the propagation phases with shape/dimension-varied meta-atoms and the geometric phases with spatially-varied orientations \( \theta(x,y) \), the spin-decoupled arbitrary phase modulation of \( \varphi(x,y) \pm 2\theta(x,y) \) can be added to the reflected LCP and RCP light, respectively. Specifically, here we implement two independent linear phase gradients, \( \frac{d\varphi_{\text{LCP}}}{dx} = \frac{\theta(x,y) + 2\theta(x,y)}{\Lambda} = \frac{\Delta \varphi + 2\Delta \theta}{\Lambda} \) and \( \frac{d\varphi_{\text{RCP}}}{dx} = \frac{\theta(x,y) - 2\theta(x,y)}{\Lambda} = \frac{\Delta \varphi - 2\Delta \theta}{\Lambda} \), along the \( x \)-axis for the LCP and RCP incident light, respectively, thus effecting the possibility to simultaneously realize, under orthogonal CP light incidence, the spin-controlled unidirectional SPP excitation and anomalous beam steering.

To achieve high-efficiency operation of both SPP excitation and beam steering, we use the GSP resonators as meta-atoms of the spin-decoupled multifunctional metasurface, which consist of gold (Au) nanoparticles arranged periodically in the \( x-y \) plane, and a continuous Au film, separated by a silicon dioxide (SiO\(_2\)) dielectric spacer (see insets in Fig. 2). The GSP resonators offer large reflection magnitudes and \( 2\pi \) phase coverage upon reflection for free-propagating optical fields, as was demonstrated in the experiments on high-efficiency beam focusing and steering in the optical regime.\(^{23,24,41,42}\) Besides, the dielectric spacer and metal bottom layer in our GSP configuration are continuous, making the GSP configuration naturally compatible with SPP
excitation and propagation.\textsuperscript{24,43} One should note that here we use both nanobrick (Fig. 2a) and nanocylinder (Fig. 2b) GSP resonators as meta-atoms to improve the performance of our spin-decoupled GSP gradient metasurface by selecting nano-HWPs with desired reflection phases and large reflection magnitudes.

**Figure 2.** Calculated complex reflection coefficients $r$ as a function of (a) nanobrick and (b) nanocylinder dimensions ($L_x, L_y$) for $A = 240$ nm, $t_m = 35$ nm, and $d = t_s = 50$ nm at 850-nm-wavelength. Color maps show the reflection coefficient magnitudes for $x$-polarization, while the blue and green solid lines are contours of the reflection coefficient phases for $x$- and $y$-polarization. Black dashed lines indicate the nano-HWPs with $|\phi_x - \phi_y|$ equal to 180°. Note that the reflection
magnitude map for \( y \)-polarization can be obtained by mirroring the map for \( x \)-polarization along the line \( L_x = L_y \). Insets are the sketches of the nanobrick and nanocylinder meta-atoms.

We implement three-dimensional (3D) full-wave simulations with the commercially available software Comsol Multiphysics (ver. 5.5) to calculate the complex reflection coefficients of the meta-atoms under both \( x \)- and \( y \)-polarized light incidence at the design wavelength of 850 nm, with \( L_x \) and \( L_y \) being the nanobrick/nanocylinder dimensions varying to optimize our nano-HWPs and other geometrical dimensions being kept constant: \( A = 240 \) nm, \( d = t_s = 50 \) nm, and \( t_m = 35 \) nm. Besides, the permittivity of Au is described by interpolated experimental values,\(^{44}\) and the SiO\(_2\) spacer is taken as a lossless dielectric with a constant refractive index of 1.45. Periodic boundary conditions are used in both \( x \)- and \( y \)-directions. After acquisition of all the complex reflection coefficients under \( x \)- and \( y \)-polarized light incidence, the available nano-HWPs with \( |\varphi_x - \varphi_y| \) matched to 180° can be extracted from the complex reflection coefficient maps (black dashed lines in Fig. 2). Two nano-HWPs with large reflection magnitudes and desired reflection phases are selected as the design nano-HWPs (red circles in Fig. 2) from the nanobrick and nanocylinder libraries, respectively, with a propagation phase difference of \( |\Delta \varphi = 90^\circ| \) under \( x \)-polarized incidence. Here we set the propagation phase differences of adjacent nano-HWPs to \(-90^\circ\), and then arrange them in a periodic manner (Fig. 3a). Note that the third and fourth meta-atoms are generated by simply rotating the first two meta-atoms by \( 90^\circ \), resulting in the supercell arrangement with a period of \( 4A \) along \( x \)-direction (Fig. 3a) that imposes the propagation phase gradient of \(-0.8854k_0\).
Figure 3. Schematic of the spin-decoupled multifunctional GSP gradient metasurface (a) before and (b) after rotating each meta-atom for introducing geometric phases. (c) Calculated reflection phase profiles of the spin-decoupled GSP gradient metasurface under RCP (red dots) and LCP (black squares) incidence at 850-nm-wavelength. (d) SEM images of the fabricated spin-decoupled GSP gradient metasurface.

In order to realize spin-decoupled functionalities of simultaneous unidirectional SPP coupling and beam steering under RCP and LCP incident light, we further introduce the geometric phases by properly rotating each nano-HWP. Here, for RCP light, the phase gradient should meet the
phase-matching condition of \(k_{RCP} = (\Delta \varphi - 2\Delta \theta)/\Lambda = k_{SPP} = k_0 N_{eff}\) for high-efficiency SPP coupling at normal incidence,\(^{15,24,43}\) where \(k_{RCP}\) is the linear phase gradient added to the reflected RCP light by the metasurface, \(k_{SPP}\) is the propagation constant of the SPP wave supported by the air/50-nm-SiO\(_2\)/50-nm-Au/SiO\(_2\) interface, \(k_0\) is free-space wavevector and \(N_{eff}\) is the effective mode index of the SPP wave (\(N_{eff} = 1.0859\)). According to the above phase-matching condition, the orientation difference between adjacent nano-HWPs \(\Delta \theta\) is set to 10.28°. Whereas for LCP light, the phase gradient encoded upon reflection is \((\Delta \varphi + 2\Delta \theta)/\Lambda = -0.6832k_0\), corresponding to a deflection angle of 43.09° for the reflected light in free space. The final arrangement of meta-atoms along \(x\)-direction in our spin-decoupled gradient metasurface (Fig. 3b) is obtained by rotating each nano-HWP from the original position (Fig. 3a) by \(N\Delta \theta\), where \(N\) indicates the \(N^{th}\) nano-HWP and \(\Delta \theta\) is the orientation difference between adjacent nano-HWPs (\(\Delta \theta = 10.28^{\circ}\)). The calculated reflection phase profiles of thus chosen and oriented meta-atoms, under normally incident RCP and LCP light at the design wavelength of 850 nm, manifest clearly the distinctly different spin-dependent linear phase gradients for orthogonal CP light incidence (Fig. 3c).

To experimentally validate the spin-dependent functionalities of the spin-decoupled GSP gradient metasurface, we fabricated the metasurface with overall dimensions of 30 \(\mu\)m \(\times\) 30 \(\mu\)m (Fig. 3d) by standard thin-film deposition, electron-beam lithography (EBL) and liftoff process (see more details about fabrication in Methods). The zoom-in SEM image (Fig. 3d) indicates that the fabrication quality is quite good, and that the resulting metasurface is in overall accordance with our design requirements despite some rounded corners of the meta-atoms. The expected performance of our spin-decoupled GSP gradient metasurface as a SPP coupler for the RCP incidence is evaluated by simulating the wavelength-dependent SPP coupling efficiency of the proposed metasurface (Fig. 4a) and considering the electric field distributions (Figs. 4b and 4c).
The pronounced excitation of the left-propagating SPPs is expected under the normally incident RCP Gaussian beam (beam radius $w_0 = 1\mu m$), reaching up to $\sim 23\%$ with $1$ dB bandwidth $\sim 170$ nm. The wavelength ensuring the maximum SPP coupling efficiency is $\sim 850$ nm, which is consistent with the nano-HWP’s design wavelength, proving that each nano-HWP maintains their respective reflection properties after constructing the metasurface.\textsuperscript{41} Note that the laser beam center is positioned away from the metasurface center to optimize the SPP coupling efficiency, owing to the tradeoff between the beam-metasurface overlap and the SPP scattering loss introduced by the metasurface.\textsuperscript{24,43} Therefore, the distance between the laser beam center and the left edge of the metasurface should be roughly equal to the beam radius, as shown in Fig. S1. After optimizing the incident beam spot position, efficient unidirectional SPP coupling by the metasurface under normally incident RCP light is realized (Fig. 4c and Fig. S5).
**Figure 4.** Simulation and experimental results of the spin-decoupled GSP gradient metasurface for unidirectional SPP coupling under normally incident RCP light. (a) Calculated and measured wavelength-dependent SPP coupling efficiencies of the excited left-propagating SPPs with an optimally positioned RCP Gaussian beam ($w_0 = 1\mu m$) at normal incidence. (b) Electric field ($x$-component) of the normally incident RCP Gaussian beam ($w_0 = 1\mu m$). The offset of the focused beam center from the left edge S1 of the metasurface is $\sim 1\mu m$. (c) Electric field ($z$-component) of the excited SPPs. (d, e) Measured LRM images at (d) the Fourier plane and (e) the real plane under RCP incidence at 900-nm-wavelength. Inset in (e) is an optical microscopic image of the sample, S1 and S2 indicate the left and right edges of metasurface, respectively, scale bar 10 $\mu m$.

With the above numerical simulations illustrating the possibility of efficient unidirectional SPP excitation with RCP light, we conducted experimental characterization of the fabricated metasurface (Fig. 3d) with a home-made leakage radiation microscopy (LRM, schematic of setup in Fig. S2a), which is widely applied for visualizing and characterizing SPP excitation and propagation.\(^{45,46}\) A wavelength-tunable laser (Spectra Physics 3900S Ti:Sapphire laser) is used as a light source in the LRM, with its output being first converted into the RCP state using a HWP, polarizer and quarter-wave plate (QWP), and then focused onto the sample into a $\sim 1$-$\mu m$-radius spot. Subsequently, we optimized the SPP coupling efficiency of the left-propagating SPPs by carefully moving the sample with a multi-axis translation stage. To quantitatively estimate the SPP coupling efficiency, we first evaluated the total SPP power by measuring the SPP leakage radiation power under LRM, and then compared it with the incident optical power:\(^{43}\)

\[
E_C = \frac{P_{SPP}}{P_{in}} = \frac{T(1 + \Gamma)P_{LR}}{P_T}
\]

(3)

\[
\Gamma = \frac{Im[n_{SPP2}]}{Im[n_{SPP2}]-Im[n_{SPP1}]}
\]

(4)
Here $E_C$ is the SPP coupling efficiency, $P_{SPP} = (1 + \Gamma)P_{LR}$, and $P_{in} = P_T/T$, where $P_{SPP}$ is the total SPP power, $P_{in}$ is the incident optical power, $T$ is the transmittance of the substrate without top-layer Au nanoparticles, $\Gamma$ relates the loss ratio between absorption in the gold film and leakage radiation for SPP waves, $P_{LR}$ is the SPP leakage radiation power, $P_T$ is the optical power after transmitting through the substrate, and $n_{SPP1}$ and $n_{SPP2}$ are the effective mode indices of SPP waves with finite and infinite bottom metal layers (Figs. S4a,b), respectively.

Following the above procedure, we experimentally characterized the SPP excitation and estimated the corresponding coupling efficiencies. First, we evaluated $\Gamma$ by measuring the wavelength dependent SPP propagation lengths that are determined by both absorption and leakage losses of the SPP waves and comparing the experimental results with the calculations employing different weighted imaginary part of the gold permittivity (Fig. S3). In Fig. S3c, the experimental SPP propagation lengths match perfectly with the calculation results obtained by increasing the imaginary part of gold permittivity by a factor of 2. After extracting the gold permittivity, we could calculate $\Gamma$ by comparing the imaginary parts of effective mode indices of two SPP modes with finite (50-nm-thick, SPP1, with leakage loss) and infinite bottom Au layer (SPP2, without leakage loss), respectively (Fig. S4). After estimating $\Gamma$, we measured $P_{LR}$ and $P_T$ directly from two successive Fourier images obtained by exposing the sample with/without metasurface to the RCP incident light, respectively. In Fourier images (Fig. 4d, Figs. S6a-d), the direct-transmitted light and SPP waves are located apart due to their distinct transverse wavevectors, rendering convenient extraction of SPP leakage power ($P_{LR}$) as well as the transmitted light power ($P_T$) for estimating SPP coupling efficiencies. For each wavelength, we optimized the focused beam position and measured 4 times. The measured wavelength-dependent SPP coupling efficiency of the spin-decoupled GSP gradient metasurface is shown in Fig. 4a, with
center wavelength around ~900 nm, exhibiting some redshift compared to the simulation results, which may be related to the fabrication imperfections of the meta-atoms as well as the uncertainty of the SiO$_2$ spacer thickness. Besides, the LRM images clearly manifest the efficient and unidirectional coupling of the RCP incident light into left-propagating SPPs, as shown in Fig. 4e (also in Figs. S6e-h). To further validate the spin-decoupled phase gradient of our metasurface, we also experimentally focused LCP and RCP light around the left and right edges (S1 and S2) of our metasurface with optimized positions, respectively. As expected, no evident SPP excitation and propagation occurs (Fig. S7), confirming the ability of the spin-decoupled GSP gradient metasurface to act as an efficient SPP meta-coupler only for RCP light incidence.

For LCP light incidence, the spin-decoupled GSP gradient metasurface works as a broadband beam deflector. To verify the broadband beam steering performance, we first performed 3D full-wave numerical simulations to calculate the far-field reflection patterns from the metasurface under LCP incident light and estimated corresponding diffraction efficiencies, increasing for consistency also in this case the imaginary part of gold permittivity by a factor of 2. Fig. 5a shows the wavelength-dependent efficiencies of the diffracted and specular-reflected light, as well as the contrast between them within 780–950 nm wavelength range. Here the contrast is defined as:

$$C = 10 \log_{10} \left( \frac{R_D}{R_S} \right)$$

where $R_D$ is the diffraction efficiency related to the amount of light reflected to the designated direction, and $R_S$ is the specular reflection efficiency. The operating wavelength with maximum diffraction efficiency $R_D$ and contrast $C$ in simulations is around 850 nm, coinciding well with both the nano-HWP’s design wavelength and the wavelength of the calculated maximum SPP coupling efficiency under RCP incidence. Fig. 5b and 5c illustrate the electric fields ($y$-components) of the incident LCP Gaussian beam ($w_0 = 1 \mu m$) and the diffraction light reflected
from the metasurface at 850-nm-wavelength, respectively. The reflection field in Fig. 5c displays a smooth wavefront with fewer distortions, manifesting that the reflected light is efficiently diffracted to one direction. When the operating wavelength deviates further from the designed value, the reflected wavefront becomes more and more inhomogeneous (Figs. S8a-d), resulting in more pronounced specular reflection which could be visualized in the farfield reflection patterns (Figs. S8f-j). Despite slight disturbances, the high-efficiency broadband beam steering is still sustained over a wide spectral range, centered around 850 nm with 1dB bandwidth of 140 nm.

**Figure 5.** Simulation and experimental results of the spin-decoupled GSP gradient metasurface for anomalous beam steering under normally incident LCP light. (a) Calculated wavelength-dependent diffraction efficiencies (black solid and dotted lines) and contrast (red dashed line). (b) Electric field (y-component) of the incident LCP Gaussian beam ($w_0 = 1 \mu m$) at 850-nm-
wavelength. (c) Electric field (y-component) of the reflection fields from the metasurface at 850-nm-wavelength. (d) Measured wavelength-dependent diffraction efficiencies (black solid and hollow rectangles) and contrast (red solid circles). (e) Measured farfield reflection pattern from the metasurface under normally incident LCP light at 900-nm-wavelength.

To experimentally characterize the broadband anomalous beam steering functionality of the spin-decoupled GSP gradient metasurface for LCP incident light, we use the same metasurface sample (Fig. 3d) which has been demonstrated as an efficient SPP coupler under RCP incident light. Fig. 5d shows the wavelength-dependent diffraction efficiencies of the metasurface, which was measured by a home-made optical setup capable of recording angle-resolved reflected optical power from the metasurface (schematic of the setup in Fig. S2b). In general, reasonable agreement is observed between the measured and calculated diffraction efficiencies, verifying the high-efficiency broadband beam steering functionality under LCP incident light. The measured average diffraction efficiency is up to ~ 48% within 800-950 nm wavelength range, and the wavelength of the maximum contrast is around 900 nm, which shows some slight redshift compared to the calculated result, but coincides well with the experimentally demonstrated wavelength of maximum SPP coupling efficiency under RCP light incidence. Furthermore, we measured the farfield patterns of the reflection field from the metasurface under LCP incident light at 900-nm-wavelength (Fig. 5e), indicating the excellent directionality of the reflection field and thus high diffraction efficiencies of the spin-decoupled GSP gradient metasurface under LCP incident light.

CONCLUSIONS

In this work, we have experimentally demonstrated a spin-decoupled multifunctional GSP gradient metasurface for simultaneous high-efficiency unidirectional SPP coupling and beam steering under normally incident orthogonal CP light in the optical regime. The metasurface is
constituted of spatially rotated GSP-based nano-HWPs encoded with propagation and geometric phases, providing two spin-dependent distinct linear phase gradients along the same direction. The proof-of-concept fabricated metasurface exhibits unidirectional SPP excitation with measured coupling efficiency > 22% within 850–950-nm wavelength range under RCP incident light. Besides, high-efficiency (48% on average) and highly-directional broadband beam steering is experimentally demonstrated for the LCP incident light. As an additional comment, we emphasize that the efficiencies of the dual-functional spin-decoupled metasurface could be further improved by employing more complex GSP meta-atoms that could provide better HWPs with finer phase sampling steps (Fig. S9-10), or using single-crystalline gold materials in the fabrication process to reduce the Ohmic loss (Fig. S11). Finally, it should be noted that, our proposed spin-decoupled GSP gradient metasurface can be scaled to more sophisticated multifunctionalities in the optical regime, such as spin-decoupled SPP excitation and beam steering to arbitrary direction, spin-controlled unidirectional SPP excitation to orthogonal directions and spin-controlled multi-foci reflecting mirror, by suitable design and configuration of the spin-decoupled reflection phase distributions. Owing to the compactness and excellent performance, we believe that, the proposed spin-decoupled multifunctional gradient metasurface provides a flexible and versatile way to manipulate spin photonics and spin plasmonics with potential applications ranging from chiroptical spectroscopy, chiral imaging to optical sensing.

METHODS

**Sample fabrication.** The spin-decoupled gap-surface plasmon (GSP) gradient metasurface was fabricated by standard thin-film deposition, electron-beam lithography (EBL) and lift-off technique. First, successive layers of 3 nm titanium (Ti), 50 nm gold (Au), 1 nm Ti and 50 nm silicon dioxide (SiO$_2$) were deposited onto a 170-$\mu$m-thick glass coverslip. Then, the metasurface
was defined using electron beam lithography (EBL, JEOL JSM-6500F field emission scanning electron microscope equipped with a Raith Elphy Quantum lithography system) employing a 100-nm-thick poly(methyl methacrylate) (PMMA, 2% in anisole, Micro Chem) layer at an accelerating voltage of 30 keV. After development in 1:3 solution of methyl isobutyl ketone (MIBK) and isopropyl alcohol (IPA), a 1 nm Ti adhesion layer and a 35 nm Au layer were deposited subsequently using ohmic evaporation. The Au nanoantennas were finally formed on the top of the SiO$_2$ film after a lift-off process.

**Leakage Radiation Microscopy.** The leakage radiation microscopy (LRM, Fig. S2a) was implemented for visualizing the surface plasmon polariton (SPP) excitation and propagation$^{45,46}$, as well as the quantitative estimation of corresponding SPP coupling efficiency$^{43}$. A fiber-coupled wavelength-tunable continuous-wave laser source (Spectra Physics 3900S Ti:Sapphire laser, wavelength range: 700 nm ~ 1000 nm) with Gaussian intensity distribution passed through first half-wave plate (HWP), a Glan-Thomson polarizer, a second HWP, and a quarter-wave plate (QWP) successively, and then focused by an objective O1 (magnification 60×, numerical aperture $NA = 0.85$) onto the metasurface. The input intensity can be adjusted by rotating the first HWP, and the spin of the input CP light can be switched by rotating the QWP. The SPP propagation was visualized by collecting the leakage radiation from the sample plane with a high NA oil-immersion objective O2 (63×, $NA = 1.25$), which thereafter split into two optical paths and projected to respective CCD cameras for obtaining both real plane and Fourier plane LRM images simultaneously. The SPP propagation length $L_p$ can be derived from the real plane LRM image,$^{43}$ where the intensity distribution of the SPP waves in the transverse direction of propagation is first fitted using a Gaussian function $I = I_x \exp \left( -2x^2/w_x^2 \right)$ with maximum intensity $I_x$ and the beam waist $w_x$, and subsequently the product of the $I_xw_x$ approximately satisfies the relation $I_xw_x = \ldots$
\( A \exp(-u/L_p), \) where \( A \) is a constant, \( u = |x - x_0| \), and \( x_0 \) is the \( x \)-coordinate of the excitation spot. Finally \( L_p \) could be extracted by a linear fit to \( \ln(I_{xw_x}) \) (Figs. S3a,b).
ASSOCIATED CONTENT

Supporting Information.

The Supporting Information is available free of charge on the ACS Publication website.

Optimization of the beam position for SPP coupling, experimental setup, estimation of SPP propagation lengths, effective SPP mode indices, calculated Ex and Ey components under RCP incident light, LRM characterization of SPP coupling under RCP incident light, experimental verification of the spin-dependent phase gradient, calculated reflection fields and farfield patterns under LCP incident light, optical GSP metasurfaces for spin-controlled beam steering and SPP coupling with other propagation/geometric-phase-step pairs ($\Delta \varphi$, $2\Delta \theta$), calculated efficiencies with different imaginary part of gold permittivity.

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Author Contributions

F. D. and S.W.T conceived the idea. C.M. and S.W.T. performed the numerical calculations. C. M. fabricated the samples and performed the optical characterization. F. D. and S.I.B. supervised the project and provided feedback on the experiments. All authors contributed to the interpretation of results and participated in the preparation of manuscript.

Notes
The authors declare no competing financial interest.

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Title: Optical gap-surface plasmon metasurfaces for spin-controlled surface plasmon excitation and anomalous beam steering

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Description to TOC: The spin-decoupled GSP gradient metasurface, consisting of rotated GSP-based nanoscale half-wave plates, combines both propagation and geometric phases to produce two different spin-dependent linear phase gradients enabling SPP excitation and anomalous reflection.