Toward broadband, dynamic structuring of a complex plasmonic field

Shibiao Wei,1,2,3 Guangyuan Si,3 Michael Malek,2 Stuart K. Earl,2,3 Luping Du,1 Shan Shan Kou,2 Xiaocong Yuan,1* Jiao Lin1,3,4*

The ability to tailor a coherent surface plasmon polariton (SPP) field is an important step toward many new opportunities for a broad range of nanophotonic applications. Previously, both scanning a converging SPP spot and designing SPP profiles using an ensemble of spots have been demonstrated. SPPs, however, are normally excited by intense, coherent light sources, that is, lasers. Hence, interference between adjacent spots is inevitable and will affect the overall SPP field distributions. We report a reconfigurable and wavelength-independent platform for generating a tailored two-dimensional (2D) SPP field distribution by considering the coherent field as a whole rather than as individual spots. With this new approach, the inherent constraints in a 2D coherent field distribution are revealed. Our design approach works not only for SPP waves but also for other 2D wave systems such as surface acoustic waves.

INTRODUCTION

Surface plasmon polaritons (SPPs) are surface electromagnetic waves coupled with collective oscillations of electrons at metal-dielectric interfaces. SPPs continue to inspire research interest due to their intriguing properties such as electromagnetic field enhancement and their short effective wavelength (1, 2). The ability to tailor a coherent SPP field plays a key role in numerous applications such as sensing (3, 4), nanocircuitry (2, 5), optical data storage (6, 7), superresolution imaging (8, 9), plasmonic tweezers (10, 11), in-plane communications (12), and in-plane data processing (13).

For in-plane SPP waves to exist, they have to satisfy the Helmholtz equation, as do all electromagnetic waves, regardless of their dimensionality (14). Because of their tight confinement at a metal-dielectric interface, the propagation of SPPs can be modeled using a two-dimensional (2D) wave equation by "losing" one of the dimensions relative to electromagnetic waves propagating in free space. At a metal-dielectric interface in the x-y plane, the out-of-plane electric field component $E_{zd}$ (in the dielectric half-space) is the most common component used to characterize the 2D field distribution. For a monochromatic SPP, the field distribution of $E_{zd}$ is governed by the 2D wave equation

$$\frac{\partial^2 E_{zd}}{\partial x^2} + \frac{\partial^2 E_{zd}}{\partial y^2} + k_{spp}^2 E_{zd} = 0 \tag{1}$$

where $k_{spp} = k_0 \sqrt{\varepsilon_m \varepsilon_d / (\varepsilon_m + \varepsilon_d)}$ is the vector describing the SPP waves at the interface, $k_0$ is the wave vector in vacuum, and $\varepsilon_m$ and $\varepsilon_d$ are the permittivities of the metal and dielectric, respectively. A number of SPP field distributions that satisfy the above equation have been investigated as the 2D counterparts of free-space solutions to the 3D wave equation. These include in-plane self-accelerating beams (15, 16), diffraction-free beams (17, 18), and 2D plasmonic vortex (PV) beams (19, 20). Dynamically scanning an SPP focal spot (a point-like solution) and modulating interference patterns have previously been demonstrated (21–23). Intricate intensity patterns have also been generated by carefully arranging the in-plane converging spots of SPPs (24, 25). It is possible to generate these SPP spot arrays because a converging spot is a solution to Eq. 1, with its amplitude profile described by a zeroth-order Bessel function of the first kind (26). However, the Bessel function amplitude profile has many side lobes, and therefore, the entire SPP field must be assembled using discrete points with a reasonable spatial separation. In contrast, here, we demonstrate the complete tailoring of complex SPP fields as a single inseparable entity. By considering the SPP field as a whole, the complex fields, including their amplitude, phase, and subsequently the in-plane energy flow of the entire 2D field distribution, can be manipulated. Furthermore, by using a wavelength-independent subwavelength structure to excite the SPP waves, we demonstrate the broadband nature of this approach. Experimentally, an electronically addressed spatial light modulator (SLM) was used to imprint information onto the incident beam before the excitation of the structured SPP field. Hence, the resultant SPP fields become digitally controllable, which will facilitate many novel inter-disciplinary applications that require a dynamically reconfigurable SPP field.

Our aim was to launch tailored monolithic SPP fields onto a metal surface by projecting beams with a specifically designed launching condition onto the ring coupler (depicted in Fig. 1), a subwavelength annular ring groove that functions as an in-plane coupler to convert light into SPP waves. Here, we used an iterative algorithm that involves a 2D in-plane Fourier transform (FT) operator inside the loop to retrieve the initial launching condition at the annular ring source (figs. S1 and S2). To simplify the design procedure, we set the amplitude distribution of the initial launching condition at the ring coupler to be uniform during each iteration. The treatment is similar to the widely used Gerchberg-Saxton algorithm for phase retrieval (27). The resultant phase distribution along the ring is subsequently transformed into a 2D radial pattern to be loaded onto the SLM. A free-space, collimated laser beam was reflected from the SLM and imprinted with the designed phase distribution (also known as the initial launching condition) before it was projected onto the ring coupler to launch the desired SPP field. This was achieved by a 4-f system, as shown in Fig. 1. As SPP waves can only be excited by transverse magnetic electromagnetic radiation, a radially polarized beam would, ideally, be used to launch the SPP waves from the ring. In the experiment, a circularly polarized beam was used instead, as the generation of a radially polarized beam would have added
with the target patterns (more discussion in section S4).

Wei et al., Sci. Adv. 2018;4 eaao0533 1 June 2018

SPP field modulation is dependent on the phase distribution associated to the application of the paraxial approximation in the derivation of the full-wave simulation, and the measurement results, primarily due to the initial conditions. They are both degenerate solutions of Eq. 1. This is the case when the matrices of the initial conditions are the same. However, due to the nature of the iterative process of the algorithm, one of the solutions can be identified. The dynamic control of a 2D phase distribution

The dynamic control of a 2D phase distribution
As shown in Fig. 3, it is possible to change the direction of the phase gradient from a "forward" to a "reverse" direction (the naming of these directions was defined arbitrarily) without altering the intensity distribution. The algorithm discussed earlier provides only a single output for the intensity target (which has two degenerate phase distributions that differ by the direction of their phase gradient), both solutions have the same probability of being found by the iterative process of the algorithm, and the initial conditions determine which one is identified. It is possible to obtain the other solution for the target field by changing the initial conditions. They are both degenerate solutions of Eq. 1. This allows one to manipulate the in-plane energy flows in either direction. A near-field experiment was designed to demonstrate the phase control by placing a straight groove into the generated plasmonic field as an in-plane reflector that helps reveal the direction of SPP energy flow (more details in section S6). In the time domain, the phase gradient and the propagating SPP waves could be directly detected using time-resolved two-photo photoemission electron microscopy (29, 30).

RESULTS
Tailoring SPP intensity distributions
An aperture-type NSOM (NTegra Solaris, NT-MDT) with an aluminum-coated fiber tip (tip aperture approximately 100 nm in diameter) was used in collection mode to measure the near-field intensity profile of the structured SPPs. Figure 2 shows the results of the various SPP field intensity distributions designed for this experiment (detailed configurations of the experiment and more results in figs. S4 and S5). There are some deviations when comparing the algorithm calculation, the full-wave simulation, and the measurement results, primarily due to the application of the paraxial approximation in the derivation of the in-plane FTs. On the metal surface, the “axis” is the center of the ring groove. This deviation of plasmonic field will become stronger when the field moves farther out from the center, while the full-wave simulation calculates the strict solution of the SPP field without any approximations. More significant energy distributions outside the desired patterns were detected in the measurement results. These arise because of the roughness of the metal film. The roughness of our metal film measured by the NSOM is about 4 nm (root mean square). Imperfections like this scatter and perturb the converging plasmonic waves. This significantly increases the loss of the plasmonic wave, resulting in the observed lower intensity of the desired patterns at the center during NSOM measurement. In addition, because the central target field results from the interference of all the constituent plasmonic waves traveling to the center from the ring coupler, the cumulative perturbations experienced by the waves during propagation eventually converge to the center and contribute to the inhomogeneity and distortions observed in the experimental data.

The dynamic control of a 2D phase distribution
As shown in Fig. 3, it is possible to change the direction of the phase gradient from a “forward” to a “reverse” direction (the naming of these directions was defined arbitrarily) without altering the intensity distribution. The algorithm discussed earlier provides only a single output for the intensity target (which has two degenerate phase distributions that differ by the direction of their phase gradient), both solutions have the same probability of being found by the iterative process of the algorithm, and the initial conditions determine which one is identified. It is possible to obtain the other solution for the target field by changing the initial conditions. They are both degenerate solutions of Eq. 1. This allows one to manipulate the in-plane energy flows in either direction. A near-field experiment was designed to demonstrate the phase control by placing a straight groove into the generated plasmonic field as an in-plane reflector that helps reveal the direction of SPP energy flow (more details in section S6). In the time domain, the phase gradient and the propagating SPP waves could be directly detected using time-resolved two-photo photoemission electron microscopy (29, 30).
Broadband property of the approach

Frequently, structured SPPs are launched via complex nanostructured arrays (24, 25, 31). The nanostructured units are placed over a specific interval designed for a predetermined wavelength. Nanostructures are normally unable to generate SPPs in a broadband manner owing to their wavelength-specific configurations, which are defined by their constituent materials and dimensions. In contrast, the structure proposed here is a wavelength-independent ring groove, albeit one whose coupling efficiency varies depending on the wavelength; its only function is to couple the incident light (which itself carries all the information that defines the structured SPP field) from free space into the in-plane SPPs. Figure 4 shows the numerical results of a variety of structured SPP fields using several incident wavelengths to demonstrate the broadband behavior of our approach.

DISCUSSION

The 2D field profile of a coherent monochromatic SPP consists of two parts, namely, its phase and amplitude. A question that may arise is whether we can structure the in-plane phase and amplitude of an SPP field concurrently, the answer to which is dependent on whether the desired field distribution satisfies Eq. 1. When considering one of the parts (normally, the amplitude) by relaxing the distribution of the other part (for example, the phase), it is easier to find a resultant field that both satisfies the wave equation and approximates the target field distribution (for example, the amplitude distribution). Unfortunately, an arbitrary complex amplitude (both amplitude and phase) distribution of an in-plane electromagnetic wave does not necessarily satisfy Eq. 1. Take, for example, a PV beam. The optical vortex (OV) field carries optical angular momentum, which comprises concentric rings and has an associated azimuthal phase distribution (32). In free space, the radii of the intensity profile for an OV beam can be varied (33). For a 2D surface wave such as an SPP, the size of a PV becomes unchangeable, as their sizes are correlated to topological charge (34). The eigenmodes of PVs on a gold film are \( E_{l \phi}(\phi, r) \sim \psi_l(k_{spp} r) \exp(il\phi) \), where \( l \) is an integer representing the topological charge of the vortex and \( J_l \) stands for the \( l \)th-order Bessel function of the first kind (20). The radius of the main intensity profile of a PV field is described by the first peak of the Bessel function. The peak indicates an SPP wave circulating along this ring with a phase change of \( l \times 2\pi \); therefore, intuitively, the circumference of the ring is fixed to \( l \times \lambda_{spp} \). The results obtained by our method, presented in Fig. 5, also confirm that it is impossible to find an appropriate output field whose radius does not match the correct value. For

![Fig. 3. In-plane phase controlling.](image3.png)

**Fig. 3. In-plane phase controlling.** (A) Ellipse intensity target intensity. There are two degenerate solutions that obey the in-plane wave equation found by our algorithm that approximate the targets. (B and F) The intensities of these two solutions are the same. (C and G) The phase gradients of the solutions are reversed from counterclockwise to clockwise. (D and H) The zoomed-in areas of (C) and (G) (close-ups of the areas indicated by the yellow dashed squares) indicate that the propagation directions of the SPP waves are reversed. Here, the phases of the low-intensity areas are set to zero to highlight the phase gradient. (E and I) Poynting vectors with intensity distributions in the background (the same areas as (D) and (H)) show that the energy flows are reversed. Scale bar, 5 \( \mu \)m.

![Fig. 4. Broadband structuring of SPP fields.](image4.png)

**Fig. 4. Broadband structuring of SPP fields.** (A to C) Rows: Different SPP field targets excited by incident beams with different wavelengths (532, 633, and 780 nm). (A1 to C1) Target intensity distributions. (A2 to C2) Intensity distributions of the 2D field found by our algorithm that approximate the targets. (A3 to D3) Full-wave simulation results of resultant SPP waves excited by the corresponding initial ring phase distributions.
instance, the PV field, pictured in Fig. 5 (E to H), with a target ringshape intensity profile intentionally set to be larger than that of Fig. 5 (A to D), could not satisfy Eq. 1. Therefore, the output SPP field generated using our algorithm no longer resembles the target—in other words, an unwrapped target SPP field could not be generated at a metal-dielectric interface.

In summary, we have reported a new and versatile platform to dynamically control in-plane SPP fields. We have demonstrated the modulation and generation of a number of intensity patterns by treating the coherent SPP field as a single entity. In addition, this technology could dynamically control the direction of the phase gradient of monolithic SPP fields, which corresponds to being able to control the direction of the (in-plane) energy flow. Because the phase information required to launch the SPP field is carried by the incident laser beam without requiring wavelength-dependent nanostructures, the launching structure can be used for a wide range of wavelengths without alteration. Finally, the limits of 2D modulation of SPP fields discussed here could act as a guide for the manipulation of other surface-confined waves. The combination of a well-designed intensity profile and in-plane energy flow has the potential to enable the dynamic control of metallic nanoparticles using plasmonic tweezers. The broadband capability of the method used to launch the SPPs here offers potential applications for generating SPP fields with different colors, for high-speed in-plane communications, and for use in large-capacity data storage. The method reported in this paper was a general method for calculating the initial launching condition for any in-plane 2D beam shaping, such as surface acoustic waves (35, 36), Bloch surface waves (37, 38), and so on.

METHODS

Numerical simulation
All numerical simulation work was performed with the commercial software FDTD Solutions from Lumerical Solutions Inc. In the simulation, the dielectric constant of the gold film was pulled from the built-in materials database within the software, in which the raw data were taken from the study of Palik (39), and the refractive index of glass was set to be 1.51. Perfectly matched layer boundaries were used in the $x$, $y$, and $z$ directions. A global mesh size of 20 nm was used in the calculations.

Sample preparation
A 100-nm-thick gold film was deposited onto a quartz substrate by electron-beam evaporation with a 5-nm germanium adhesion layer. The ring-groove structures were fabricated using focused ion beam milling (FEI Helios NanoLab600 DualBeam FIB-SEM system). During focused ion beam milling, a 28-pA beam current was selected with an accelerating voltage of 30 kV.

SUPPLEMENTARY MATERIALS
Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/4/6/eaao0533/DC1

section S1. In-plane FT for converging SPP waves
section S2. Algorithm for obtaining the initial launching distribution of the SPP fields
section S3. Compensating spiral phase carried by the circularly polarized beam
section S4. Discussion on the spatial resolution of the generated SPP fields
section S5. Experimental setup
section S6. Experimental demonstration of the phase control
section S7. Additional results for dynamically structuring SPP fields
fig. S1. Derivation of the in-plane FT of surface waves.
fig. S2. Algorithm process flow to obtain the initial launching distribution of the SPP fields.
fig. S3. Schematic diagram of the spiral phase compensation by an inverse spiral phase distribution.
fig. S4. Single focused spot of plasmonic field.
fig. S5. Two equally bright coherent spots separated by one full width at half maximum with the phase difference between the two spots as 0, π, and 0.5π.
fig. S6. Schematic diagram of the experimental setup.
fig. S7. Experimental demonstration of dynamic switching of the phase gradient.
fig. S8. Results for dynamically structuring SPP fields.
movie S1. Clockwise energy flow for the elliptical plasmonic field.
movie S2. Counterclockwise energy flow for the elliptical plasmonic field.

REFERENCES AND NOTES
1. W. L. Barnes, A. Dereux, T. W. Ebbesen, Surface plasmon subwavelength optics. Nature 424, 824–830 (2003).
2. E. Ozbay, Plasmonics: Merging photonics and electronics at nanoscale dimensions. Science 311, 189–193 (2006).
Toward broadband, dynamic structuring of a complex plasmonic field
Shibiao Wei, Guangyuan Si, Michael Malek, Stuart K. Earl, Luping Du, Shan Shan Kou, Xiaocong Yuan and Jiao Lin

Sci Adv 4 (6), eaao0533.
DOI: 10.1126/sciadv.aao0533