

Structureless Excitation and Manipulation of Dynamic Holographic Plasmonic Slides

Corresponding Author: Professor Changjun Min

Parts of this Peer Review File have been redacted as indicated to remove third-party material.

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The manuscript demonstrates a novel and fully reconfigurable method to excite arbitrary surface-plasmon patterns on a flat metal film by encoding both amplitude and phase at the pupil plane of a focusing objective. An inverse-design algorithm computes the required ring-shaped holograms. The authors validate their approach using four target patterns (oval, arc, helix, and cycloid), quantify efficiency improvements, and demonstrate dynamic particle transport along complex trajectories using orbital angular momentum.

We recommend this paper for publication, after the authors address the following concerns:

1. The discussion beginning with "In contrast, Figs. 2 (D and E) illustrate ... high modulation freedom and flexibility." appears verbatim in both lines 116–126 and 133–142. Please remove the redundant copy so that the explanation appears only once.
2. The colorbars in Figs. 2(B), 2(E), and 4(A1) are not specified properly. What do the values 30 in Fig. 2(B), 44 in Fig. 2(E), and -60 in Fig. 4(A1) represent? Are these normalized quantities?
3. The authors report correlation indices larger than 0.8 between the designed and measured plasmonic fields, but it is not clear how sensitive this metric is to the choice of region of interest (ROI) based on equations 6 and 7. Could you please clarify how the spatial ROI used for the correlation calculation was defined? Was it the entire scanned area or only the high-intensity belt? Have you examined how the correlation index changes with different ROI bounds?
4. The Inverse-Design Algorithm section provides a high-level description of the plane-wave decomposition of the target field. Please add: a) a step-by-step outline or flowchart detailing the computation of the inner product over the ring aperture b) the number of sampling points/angles used, and c) an estimate of the computational cost of the algorithm.
5. Did the inverse design procedure involve an iterative solver or optimization loop? A brief paragraph comparing your inverse-design approach with other numerical techniques would help the reader to understand its novelty, performance advantages, and limitations.
6. The manuscript reports improved "efficiency" compared to etched rings, but the metric is not explicitly defined. If efficiency refers to the fraction of input optical power coupled into the target plasmonic mode, please specify how it is measured and include the formula used in the calculation.

Reviewer #2

(Remarks to the Author)

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Reviewer #3

(Remarks to the Author)

This manuscript developed a method to generate plasmonic fields on a metal film without using pre-fabricated nanostructures. It also demonstrated an energy flow control to control particles along the plasmonic intensity profiles. This work is interesting and novel by providing a versatile and flexible platform for controlling nanoscale particles, which might be interesting for many single particle based quantum applications. I would recommend this paper to be accepted by Nature Communications.

Here are some more detailed questions:

1. In figure2, the comparison between the structureless method and the conventional methods is clearly shown. However, it would be better to have a quantitative comparison. For example, the author can define a FoM of the generated pattern. How much do they differ? Or for some other particle trapping profile, to achieve similar trapping depth, how much input energy differs? A quantitative comparison here might be interesting to see.
2. In figure3, it was not clear to me why the simulation looks worse than the theory, especially for the high-spatial-frequency patterns. Also, why does the experimental results look worse than the simulation result? Is it due to misalignment of the lenses?
3. In the experiment, a gold particle was used. May I ask why using gold particles? Is it because gold has a very strong plasmonic response? How does this technique apply for other particles with weaker plasmonic responses?

Reviewer #4

(Remarks to the Author)

The authors of the manuscript "Structureless Excitation and Manipulation of Dynamic Holographic Plasmonic Slides" present a way to manipulate the phase, intensity and properties of plasmon excited on a flat metal-dielectric interface, to produce a holographic image. I found the paper interesting and engaging, with the experimental results showing improvement on current methods used. However, I'm unsure if the progress on the method is significant enough for publication in Nature Communications. I believe that this work will be more suited in a more specialised journal, such as Nanophotonics.

Nevertheless, I have a couple of points that the authors can improve on, before submitting the paper in a more specialized journal:

- 1) I'm a bit puzzled why the authors chose to do their experiments by optically trapping nanoparticles. Would a probe-tip not be a better choice, since it won't suffer from Brownian motion which alters the scattered fields measured during the experiment? Also, if the authors want to stick with optically trapped nanoparticles, then why not use dielectric ones that will induce less scattering that affects their results?
- 2) I'm not sure how the authors match the momentum of the incident beam with propagating SPP modes. From Fig.2, it seems that they always excite with normal incidence. I understand in Fig2A that there is a nanoring slit that allows for this momentum matching for certain part of the incident normal beam. But how do the authors manage to achieve a better momentum match in fig.2D (and future configurations in the paper)?
- 3) The first sentence of the introduction is not technically correct. The propagating SPPs on metal-dielectric interfaces are eigenmodes, which means that they are always there. One does not need to illuminate the interface with light for the evanescent modes to emerge. But one needs external illumination to couple light into these modes, which is what I believe the authors were trying to communicate.
- 4) Terms like "stray signals" or "stray light" are not proper scientific terminology. This light/signal comes from diffraction, and one should say this.
- 5) Line 53: "Some other properties, such as energy flow, are challenging to manipulate according to desired patterns, ..." Why is this the case? What is stopping current methods to do this?
- 6) I'm not sure what is a toroidal orbital angular momentum. It would be convenient for the reader if the authors defined what they mean by it.
- 7) What is the asterisk on I_1^* in table 1? It is never explained, or if it is I missed it completely.
- 8) There are a 3-4 places that the language and grammar can be improved. For example:
line 121 "Fig.2E is significantly clean with higher excitation..." should probably be: "Fig.2E is significantly more clean with higher excitation..."
line 175 "This imperfect is mainly...", should probably be : "These imperfections are mainly ..."

Overall, although the manuscript shows interesting results, they are primarily incremental so I believe that they will be of more interest to a more specialized audience than Nature Communications.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The authors have satisfactorily addressed all of our comments. We recommend accepting the revised manuscript in its current form.

Reviewer #2

(Remarks to the Author)

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Reviewer #3

(Remarks to the Author)

I appreciate the authors' responses and revisions. All of my previous questions and comments have been addressed, and the manuscript is now complete and clear. I would recommend the paper for publication.

Reviewer #4

(Remarks to the Author)

The authors improved the manuscript significantly and answered my questions well. I'm happy to accept now the paper for publication in Nature Communications.

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Response to the Decision Letter

Response to reviewer #1

Comment: The manuscript demonstrates a novel and fully reconfigurable method to excite arbitrary surface-plasmon patterns on a flat metal film by encoding both amplitude and phase at the pupil plane of a focusing objective. An inverse-design algorithm computes the required ring-shaped holograms. The authors validate their approach using four target patterns (oval, arc, helix, and cycloid), quantify efficiency improvements, and demonstrate dynamic particle transport along complex trajectories using orbital angular momentum. We recommend this paper for publication, after the authors address the following concerns.

Comment 1: The discussion beginning with “In contrast, Figs. 2 (D and E) illustrate ... high modulation freedom and flexibility.” appears verbatim in both lines 116–126 and 133–142. Please remove the redundant copy so that the explanation appears only once.

Reply 1: We thank the reviewer for the careful reading and for pointing out this duplication. The repeated paragraph (lines 133-142 in the original manuscript) has now been removed, and the explanation is now included only once in the revised manuscript.

Comment 2: The colorbars in Figs. 2(B), 2(E), and 4(A1) are not specified properly. What do the values 30 in Fig. 2(B), 44 in Fig. 2(E), and -60 in Fig. 4(A1) represent? Are these normalized quantities?

Reply 2: We appreciate the reviewer’s insightful question. In Fig. 2, the colorbar values represent the magnitude of the electric field, $|E|$, obtained from the FDTD simulations. The incident field is normalized to a unit amplitude of $1 \text{ V} \cdot \text{m}^{-1}$, providing a consistent reference for comparing field enhancements between our tightly focused excitation and conventional structured illumination. For context, achieving a stable optical trap requires an optical potential depth exceeding $1k_B T$ (k_B is the Boltzmann constant and T is the temperature, which is fixed at $T=300 \text{ K}$ for normalization [28]) to ensure that the particle can overcome thermal fluctuations. In Fig. 4(A1), the colorbar indicates the dimensionless potential depth by $k_B T$, which is a normalized quantity. Additional clarifications regarding colorbar normalization and labeling have been added in the revised manuscript to improve clarity.

Correspondingly,

Line 124-126, Page 4: “For a quantitative comparison, the amplitude distributions in Fig. 2B and Fig. 2E are calculated from electric fields that are normalized to an incident beam with an electric-field amplitude of $1 \text{ V} \cdot \text{m}^{-1}$. In the absence of structure-induced stray light, the pattern in Fig. 2E exhibits a significantly cleaner background and a higher excitation efficiency than that in Fig. 2B.”

The caption of Fig. 2, Page 5: “Fig 2 | Comparison of a holographic plasmonic helix field excited by structural and structureless methods. (A) Schematic of a nanoring slit assisted plasmonic field excitation. (B) Distribution of the structure assisted plasmonic helix field, with the field amplitude normalized to that of an incident beam whose electric-field amplitude is set to $1 \text{ V} \cdot \text{m}^{-1}$. The yellow dashed line indicates the slit’s position. (C) The generated chaotic plasmonic field when the nanoring slit is cleared off. (D and E). The schematic and final excited plasmonic helix of our method. F. The intensity and phase of the incident beam, intensity is normalized along the radius.”

Fig.4, Page 8:

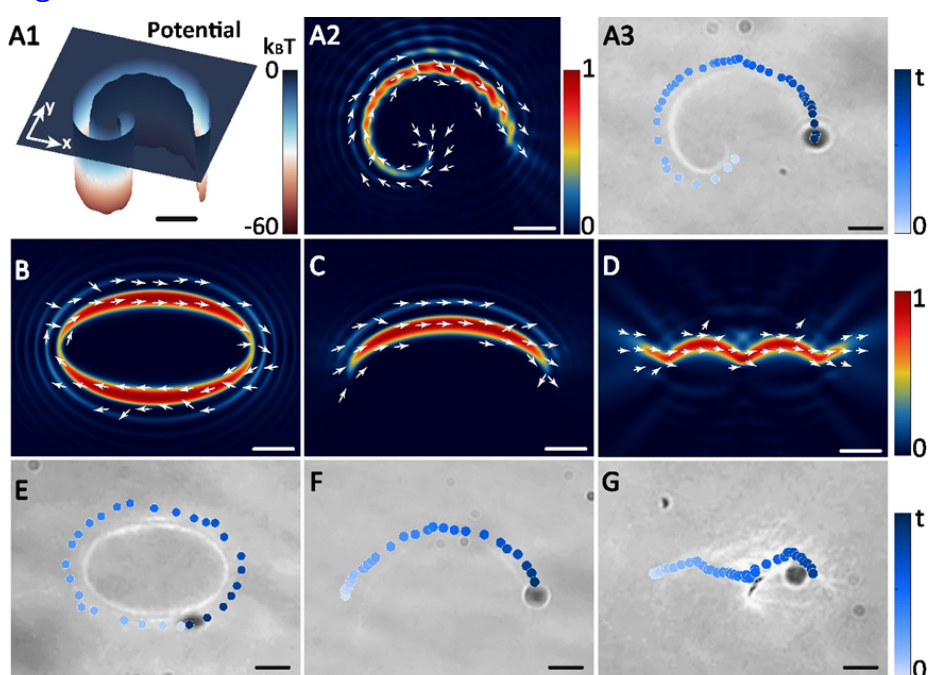


Fig. 4 | The energy flux and particle transportation results of excited plasmonic slide fields. (A) The potential well (in units of $k_B T$), energy flux, and experimental trajectory of an Au particle in the excited Helix plasmonic field. Here k_B is the Boltzmann constant and T is the temperature, which is fixed at $T=300 \text{ K}$ for normalization. (B to D) The energy flux arrows (background is intensity) and (E to G) the corresponding experimental trajectory of the Au particle in the Oval, Helix, and Cycloid shaped plasmonic fields, respectively. Different colors of the trajectories represent positions of the particle at different times, specifics see Supplementary Videos. Scalebars are $1 \mu\text{m}$.

Comment 3: The authors report correlation indices larger than 0.8 between the designed and measured plasmonic fields, but it is not clear how sensitive this metric is to the choice of region of interest (ROI) based on equations 6 and 7. Could you please clarify how the spatial ROI used for the correlation calculation was defined? Was it the entire scanned area or only the high-intensity belt? Have you examined how the correlation index changes with different ROI bounds?

Reply 3: Thanks for raising this important point. For the designed plasmonic field described by Eq. 1, the cross-intensity profile of the focal curve follows a Gaussian envelope. In this work, without loss of generality, the threshold boundary for selecting the region of interest (ROI) in the correlation calculation is defined as the area where the Gaussian intensity drops to 50% of its peak value. To evaluate the robustness of this choice, we further calculate the correlation coefficient across a series of ROI boundaries corresponding to Gaussian attenuation levels ranging from 20% to 80%. As shown in Fig. R1, the correlation values remain essentially unchanged over this range, indicating that the metric is insensitive to the precise ROI selection. An expanded analysis of the ROI robustness has been added to the supplementary of the revised manuscript.

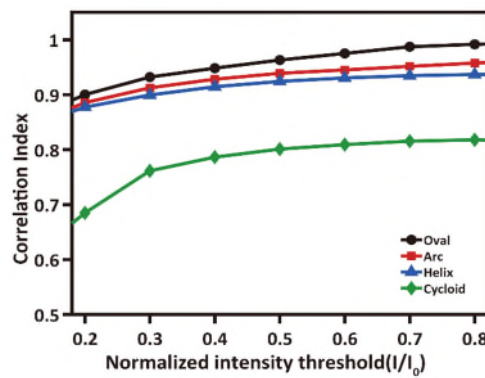


Fig. R1 | Correlation Index of the plasmonic patterns under different Region-of-Interest (ROI) boundaries

Correspondingly,

Line 168, 169, Page 6: “... correlation indices of both the intensity and complex amplitude of the plasmonic fields are calculated (see Methods section and Supplementary Note S4).”

Add Supplementary Note S4: Robustness of the Correlation Index against Region-of-Interest (ROI) Boundary Selection.

For the designed plasmonic field described by Eq. 1, the cross-sectional intensity profile along the focal curve follows a Gaussian envelope. In the main text, the Region-of-Interest (ROI) for the correlation calculation is defined as the area where the Gaussian intensity drops to 50% of its peak value.

To assess the robustness of the correlation index used to compare the designed and experimental plasmonic fields, we evaluate its stability with respect to the choice of ROI boundary for four representative plasmonic patterns (oval, arc, helix, and cycloid), as indicated by the different colored curves. The ROI boundary is specified by the normalized Gaussian intensity threshold I/I_0 , which is varied from 0.2 to 0.8 to examine the sensitivity of the correlation metric, as shown in Fig. S3. Since the primary energy remains predominantly confined within the high-intensity band, the correlation coefficient exhibits almost no variation once the threshold exceeds 0.3 and remains essentially stable across this range, confirming the robustness of the metric and its insensitivity to the precise ROI

selection.

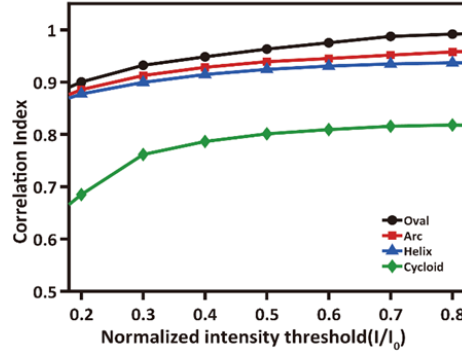


Fig. S3 | Correlation Index of the plasmonic patterns under different Region-of-Interest (ROI) boundaries.

Comment 4: The Inverse-Design Algorithm section provides a high-level description of the plane-wave decomposition of the target field. Please add: a) a step-by-step outline or flowchart detailing the computation of the inner product over the ring aperture b) the number of sampling points/angles used, and c) an estimate of the computational cost of the algorithm.

Reply 4: We thank the reviewer for this constructive suggestion. The detailed computational procedure for evaluating the inner product over the ring aperture is summarized in the flowchart as shown in Fig. R2. Briefly, we begin by importing the normalized target plasmonic field. Next, we define the sampling variable θ (sampling points/angles), using a discretized angle with a step size of $\pi/500$. For each θ , a corresponding plasmonic plane wave $\Phi_\theta = \exp[ik_x \sin(\theta)] \cdot \exp[ik_y \cos(\theta)]$ is generated and subsequently normalized. The inner product shown in Eq.(3) between Φ_θ and $\mathbf{E}_0$ is then computed to obtain the complex coefficient $\mathbf{u}$. This process is repeated for all θ values until the complete function $\mathbf{u}(\theta)$ is obtained.

It is important to emphasize that our method does not require iterative optimization; the only loop calculation involves traversing the sampled θ angles. Consequently, our approach offers a significant advantage in computational efficiency compared with traditional optimization algorithms. Implemented in MATLAB and executed on a machine equipped with an Intel i7-12700 CPU, the total computation time is 30.95 seconds.

In the revised manuscript, we have added a detailed, step-by-step description of the inner-product computation in the supplementary.

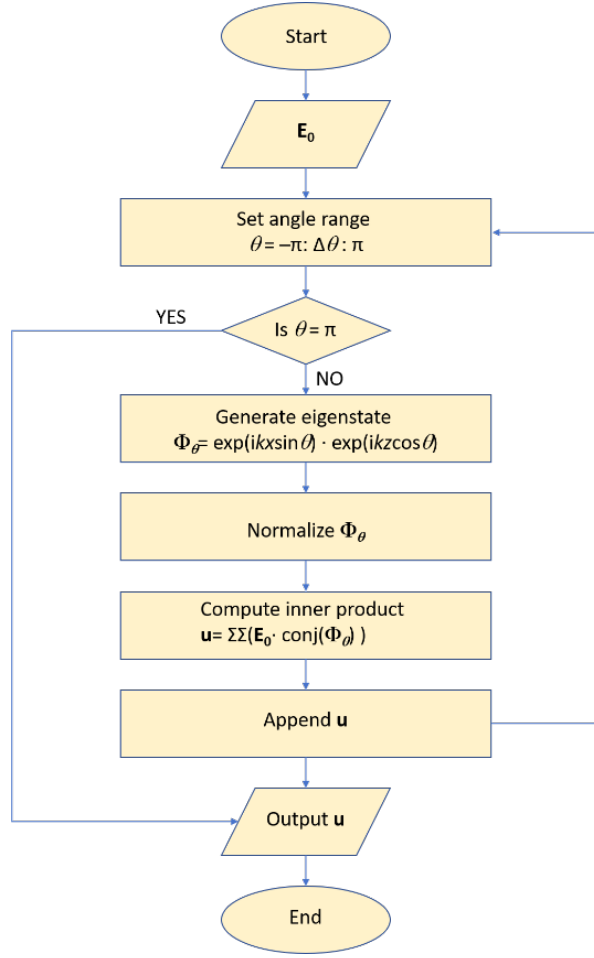


Fig. R2 | Flowchart for computing the inner product over the ring aperture

Correspondingly,

Line 336-337, Page 13: “where the symbol $\langle \cdot | \cdot \rangle$ represents the inner product of two fields, and the detailed simulation procedure is provided in Supplementary Note S7. From Eqs. (1 to 3), the amplitude and phase distribution of initial excited plasmonic plane waves at different angle θ can be calculated, as shown in Fig. 6B. ”

Add supplementary Note S7. Numerical Calculation of the Inner Product.

To explicitly present the numerical procedure used in the inverse-design process, Fig. S5 details the full computational workflow for evaluating the angular coefficient function. Compared to the inverse Fourier transform in free space, the inverse design algorithm represents the target plasmonic field as a linear combination of directionally separable plasmonic plane waves. Initially, the algorithm introduces the normalized target field distribution $\mathbf{E}_0$. Subsequently, a sampling variable θ is defined, representing discrete sampling points or angles, with a specified angular step size of $\pi/500$. For each discrete θ , a corresponding plasmonic plane wave Φ_θ is generated and subsequently normalized. This procedure effectively discretizes the continuous plasmonic modes within the angular spectrum space. The inner product between Φ_θ and $\mathbf{E}_0$ is then computed to obtain the complex coefficient $\mathbf{u}$. By repeating this procedure for all sampled angles, the complete angular coefficient function $\mathbf{u}(\theta)$ is constructed. This inner-product operation directly yields the

projection complex amplitude of the target field onto the plasmonic eigenmode at each angle, representing the complex weight of its corresponding component in the angular spectrum. Therefore, the algorithm enables an end-to-end mapping through complete complex-amplitude control, without the need for any additional iterative optimization. Implemented in MATLAB and executed on a machine equipped with an Intel i7-12700 CPU, the total computation time is 30.95 seconds.

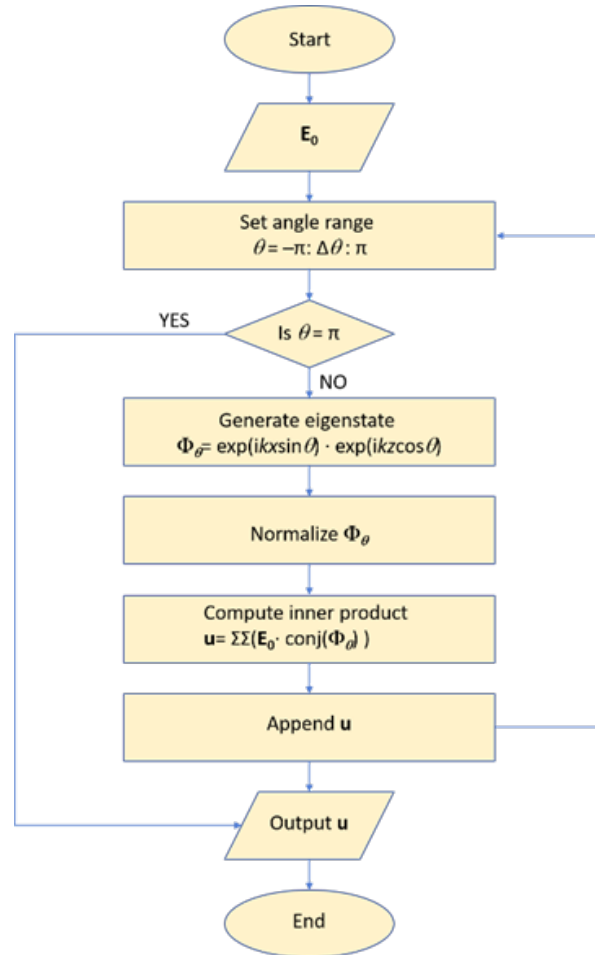


Fig. S5 | Flowchart for computing the inner product over the ring aperture

Comment 5: Did the inverse design procedure involve an iterative solver or optimization loop? A brief paragraph comparing your inverse-design approach with other numerical techniques would help the reader to understand its novelty, performance advantages, and limitations.

Reply 5: Thanks for this insightful suggestion, this is indeed a key point. A major advantage of our approach is that the inverse-design procedure does not rely on iterative solvers or optimization loops. Instead, it is formulated directly from the principle of quantum-state superposition: the target field is expressed as a linear combination of basis plasmonic modes, and the complex weighting coefficients are obtained analytically through inner-product evaluation. This fundamentally distinguishes our

method from conventional inverse-design techniques, such as Gerchberg–Saxton–type phase retrieval or adjoint-based gradient optimization, which typically require numerous iterations, involve convergence checks, and may be susceptible to local minima. In contrast, our method avoids these issues, offers higher computational efficiency, and retains a clear physical interpretation. A comparative discussion highlighting these differences has been added to the revised manuscript (lines 348-352).

Correspondingly,

Line 348-352, Page 13: “Distinct from conventional inverse-design techniques, such as Gerchberg–Saxton–type phase retrieval or adjoint-based gradient optimization, which typically require numerous iterations, rely on convergence checks, and are prone to local minima, our method avoids these issues, offers significantly higher computational efficiency, and retains a clear physical interpretation.”

Comment 6: The manuscript reports improved “efficiency” compared to etched rings, but the metric is not explicitly defined. If efficiency refers to the fraction of input optical power coupled into the target plasmonic mode, please specify how it is measured and include the formula used in the calculation.

Reply 6: Thank you for this helpful suggestion. To facilitate a clear and quantitative comparison between the structured and unstructured configurations, we define the coupling efficiency, β , as the ratio of the power coupled into the excited plasmonic field to the total input power, calculated as:

$$\beta = \frac{\iint |\mathbf{E}_o(x, y)|^2 dx dy}{\iint |\mathbf{E}_{in}(x, y)|^2 dx dy} \quad (\text{R1})$$

Our results show that the structured configuration achieves a coupling efficiency of approximately 12.3%, whereas our inverse-designed configuration reaches 18.5%. This efficiency can be further improved by generating a perfect radially polarized beam, which can enhance the trapping stiffness by compressing the energy into a thin ring that satisfies the plasmonic coupling conditions [39]. In the revised manuscript, we have added the definition of β , its corresponding formula, and relevant references to improve clarity.

Correspondingly,

Line 129-133, Page 5: “In the absence of structure-induced diffraction background, the pattern in Fig. 2E exhibits a significantly cleaner background and a higher excitation efficiency than that in Fig. 2B. Quantitatively, we define the coupling efficiency β as the ratio of the power coupled into the excited plasmonic field to the total input power (see Supplementary Note S2 for the formal definition). With this definition, the coupling efficiency of our inverse-designed configuration reaches 18.5%, exceeding that of the ring-shaped structural approach (12.3%) by 6 percentage points, corresponding to a 50% relative improvement.”

Add supplementary Note S2.

Comparison of coupling efficiency.

To facilitate a clear and quantitative comparison between the ring-shaped slit and tightly focused reverse design configurations, we define the coupling efficiency, β , as the ratio of the power coupled into the excited plasmonic field to the total input power, calculated as:

$$\beta = \frac{\iint |\mathbf{E}_0(x, y)|^2 dx dy}{\iint |\mathbf{E}_{in}(x, y)|^2 dx dy} \quad (\text{S1})$$

where $\mathbf{E}_0(x, y)$ represents the electric field distribution within the target plasmonic field profile and $\mathbf{E}_{in}(x, y)$ represents incident electric Field on the front surface of the gold film, both obtained from the FDTD simulations.

To ensure a fair comparison between the two excitation schemes, the incident field in both cases is normalized to a unit amplitude, and the colorbars in Fig. S1 represent the corresponding relative electric-field magnitudes. This normalization allows the field distributions shown in Fig. S1A and S1B to directly reflect the efficiency with which each excitation configuration couples energy into the plasmonic mode. As a result, the visible enhancement in the ring-shaped slit and inverse-designed configurations provides an intuitive comparison. The ring-shaped slit configuration achieves a coupling efficiency of approximately 12.3%, whereas our inverse-designed configuration achieves an efficiency of 18.5%. This enhancement is also evident in the increased intensity of the excited plasmonic field. The efficiency can be further improved by generating a perfect radially polarized beam, which can enhance the trapping stiffness by compressing the energy into a thin ring that satisfies the plasmonic coupling conditions ^[39].

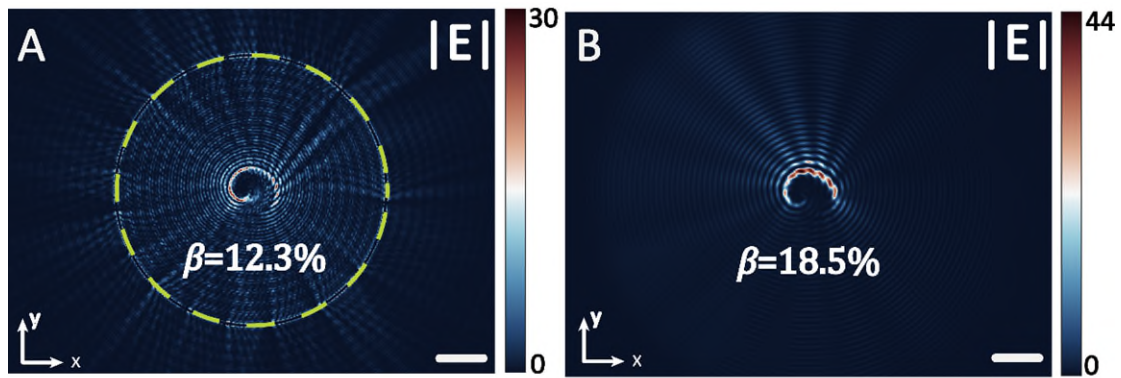


Fig. S1 | Efficiency Comparison of a holographic plasmonic helix field excited by (A) structural and (B) structureless methods.

Add Reference:

39. X. Wang, Y. Zhang, Y. Dai, et al. Enhancing plasmonic trapping with a perfect radially polarized beam. *Photonics Research*. **6**, 847 (2018).

Response to reviewer #2

Comment: I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

We sincerely thank the reviewer and the co-reviewer for their time and efforts in providing valuable feedback.

Response to reviewer #3

Comment: This manuscript developed a method to generate plasmonic files on a metal film without using pre-fabricated nanostructures. It also demonstrated an energy flow control to control particles along the plasmonic intensity profiles. This work is interesting and novel by providing a versatile and flexible platform for controlling nanoscale particles, which might be interesting for the many single particle based quantum applications. I would recommend this paper to be accepted by Nature Communications.

Comment 1: In figure2, the comparison between the structureless method and the conventional methods is clearly shown. However, it would be better to have a quantitative comparison. For example, the author can define a FoM of the generated

pattern. How much do they differ? Or for some other particle trapping profile, to achieve similar trapping depth, how much input energy differs? A quantitative comparison here might be interesting to see.

Reply 1: Thank you for this valuable suggestion. To provide a quantitative comparison between the structured and unstructured configurations, we define the coupling efficiency, β , as the ratio of the optical power delivered into the target plasmonic mode to the total input laser power. The calculation is as follows:

$$\beta = \frac{\iint |\mathbf{E}_0(x, y)|^2 dx dy}{\iint |\mathbf{E}_{in}(x, y)|^2 dx dy} \quad (\text{R1})$$

Our results show that the structured configuration achieves a focusing efficiency of approximately 12.3%, while the inverse-designed configuration attains 18.5%. The efficiency can be further improved by generating a near-ideal radially polarized beam, which enhances the longitudinal field at the focus and increases the plasmonic trapping stiffness [39]. In the revised manuscript, we have added the definition of β , its corresponding formula, and relevant references for clarity.

Correspondingly,

Line 129-133, Page 5: “In the absence of structure-induced diffraction background, the pattern in Fig. 2E exhibits a significantly cleaner background and a higher excitation efficiency than that in Fig. 2B. Quantitatively, we define the coupling efficiency β as the ratio of the power coupled into the excited plasmonic field to the total input power (see Supplementary Note S2 for the formal definition). With this definition, the coupling efficiency of our inverse-designed configuration reaches 18.5%, exceeding that of the ring-shaped structural approach (12.3%) by 6 percentage points, corresponding to a 50% relative improvement.”

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Where $\mathbf{E}_0(x, y)$ represents the electric field distribution within the target plasmonic field profile

and $E_{in}(x,y)$ represents incident electric Field on the front surface of the gold film, both obtained from the FDTD simulations. To ensure a fair comparison between the two excitation schemes, the incident field in both cases is normalized to a unit amplitude, and the colorbars in Fig. S1 represent the corresponding relative electric-field magnitudes. This normalization allows the field distributions shown in Fig. S1A and S1B to directly reflect the efficiency with which each excitation configuration couples energy into the plasmonic mode. As a result, the visible enhancement in the ring-shaped slit and inverse-designed configurations provides an intuitive comparison. The ring-shaped slit configuration achieves a coupling efficiency of approximately 12.3%, whereas our inverse-designed configuration reaches 18.5%. This enhancement is also evident in the increased intensity of the excited plasmonic field. The efficiency can be further improved by generating a perfect radially polarized beam, which can enhance the trapping stiffness by compressing the energy into a thin ring that satisfies the plasmonic coupling conditions ^[39].

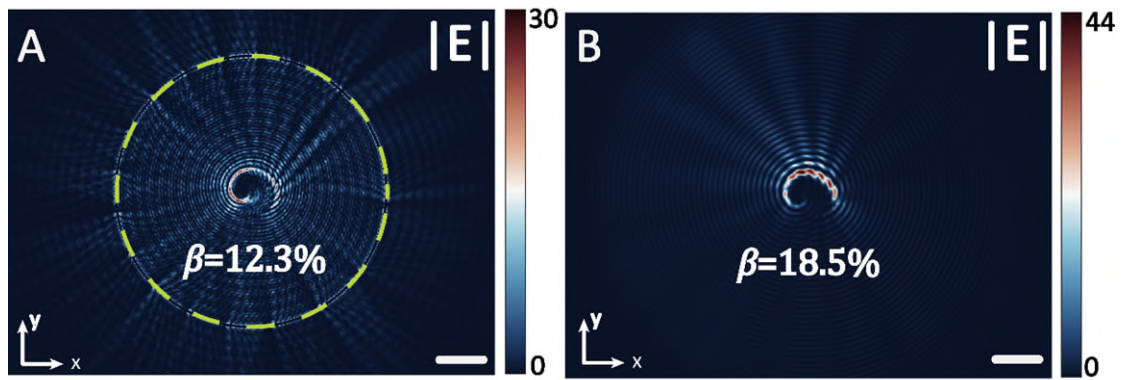


Fig. S1 | Efficiency Comparison of a holographic plasmonic helix field excited by (A) structural and (B) structureless methods.

Comment 2: In figure3, it was not clear to me why the simulation looks worse then the theory, especially for the high-spatial-frequency patterns. Also, why does the experimental results look worse than the simulation result? Is it due to misalignment of the lenses?

Reply 2: Thank you for your thoughtful comment. In our method, the theoretical target plasmonic field is constructed as a superposition of plane waves, with the corresponding complex amplitude weights determined through an inverse-design procedure. During the actual propagation of the plasmonic field, however, the Joule losses and other dissipative mechanisms inevitably distort these complex weights, resulting in deviations between the simulated fields and the ideal theoretical patterns, especially for high-spatial-frequency components. To quantify these differences, we performed FDTD simulations of the designed fields and evaluate their consistency with the theoretical targets using correlation coefficients.

For the experimental results, as you mentioned, misalignment of the optical elements is indeed one factor that affects the accuracy of the generated field. Although such errors can be reduced by improving the positional accuracy of the optical components, they remain a practical limitation. Additionally, the experimental optical system inherently behaves as a low-pass filter, suppressing high-frequency components and making it challenging to reproduce sharp features in the target field. This filtering effect further contributes to the discrepancies observed between the experimentally measured fields and simulated patterns.

We have added these discussions in the revised manuscript, correspondingly,

Line 184-190, Page 7: “Furthermore, Table 1 shows that the FDTD results are slightly degraded compared with the ideal theoretical values. This discrepancy mainly arises because the plasmonic field in theory is constructed as a superposition of plane waves, whose complex amplitude weights are prescribed by the inverse-design procedure. During the actual propagation of the plasmonic field, however, Joule losses and other dissipative mechanisms inevitably reshape these complex weights, leading to deviations between the simulated fields and the ideal target pattern, particularly in the high-spatial-frequency components.”

Comment 3: In the experiment, a gold particle was used. May I ask why using gold particles? Is it because gold has a very strong plasmonic response? How does this technique apply for other particles with weaker plasmonic responses?

Reply 3: Thanks for this question. In this work, gold nanoparticles are chosen primarily due to their strong absorption and scattering properties, which pose significant challenges for trapping in conventional optical tweezers. Consequently, our results underscore the advantages of the plasmonic approach we have developed. Importantly, our approach is not limited to metallic particles; it also holds for dielectric particles exhibiting considerably weaker plasmonic responses.

As illustrated in Figure R3, we add validation experiments using SiO₂ particles, which demonstrate stable trapping and guided transport along the designed holographic plasmonic trajectory. These results confirm the generality and robustness of our technique across different particle types. These new experimental data have been incorporated into the revised manuscript and Supplementary Materials.

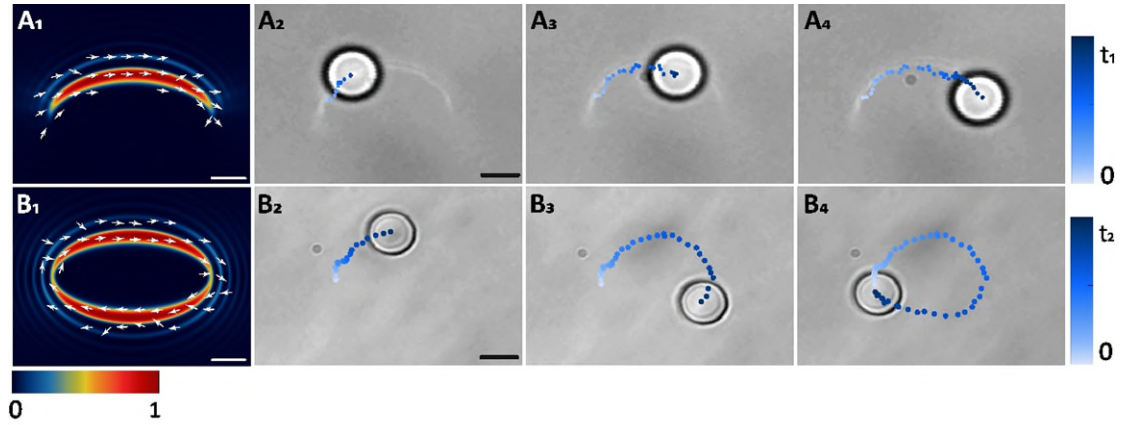


Fig. R3 | The corresponding experimental trajectory of the SiO₂ particles in the (A) Arc, and (B)Oval shaped plasmonic fields. Scalebars are 3 μ m.

Correspondingly,

Line 277-282, Page 11: “In addition to trapping metallic nanoparticles, our method is also applicable to the micromanipulation of dielectric particles (see Supplementary Note S6). To demonstrate its generality, we further perform trapping experiments with SiO₂ microspheres, showing that these dielectric particles can likewise be guided along trajectories prescribed by the holographic plasmonic fields. These results confirm the universality of our approach for microparticle trapping and transport.”

Add supplementary Note S6.

Experimental Transport of Dielectric particles (SiO₂)

Benefiting from the fine control of plasmonic energy flow, the holographic plasmonic tweezer system is not limited to metallic particles but is equally effective for dielectric particles. We additionally performed trapping experiments on dielectric particles, which exhibit relatively weak plasmonic responses. In experiment, a diluted suspension of SiO₂ microspheres was dropped onto the gold film. SiO₂ particles located near the potential wells were stably captured at the deepest valley under the axial gradient forces, and then continuously transported along the designed trajectories under the drive of the plasmonic energy flow, as shown in Fig. S4. It should be emphasized that the trapping power is also crucial for ensuring stable transportation. Considering different trajectory lengths lead to variations in the locally distributed energy, the incident power was set to 5 mW to balance trapping stability and transportation efficiency. Compared with metallic particles, dielectric particles exhibit weaker scattering, which contributes to their higher stability during transportation. These results confirm the generality and robustness of our technique across different particle types.

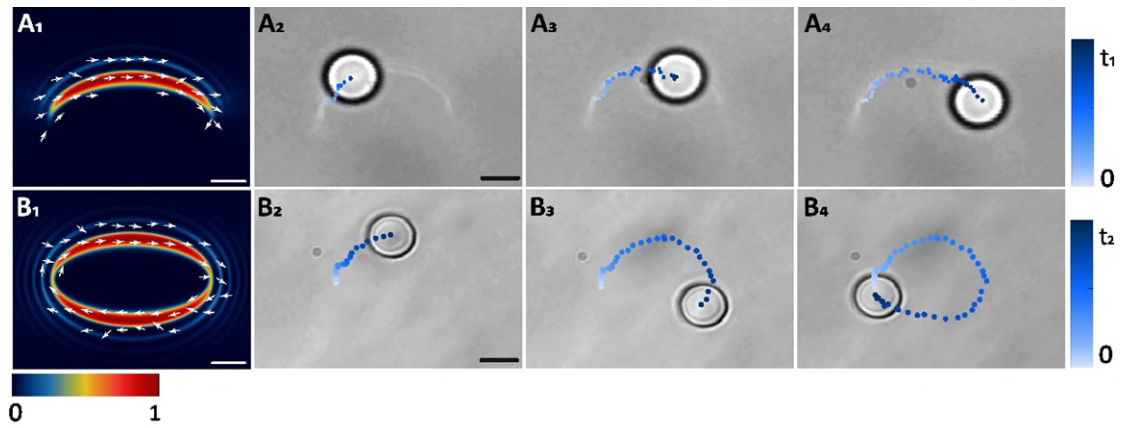


Fig. S4 | The experimental trajectory of the SiO₂ particles in the (A) Arc, and (B) Oval shaped plasmonic fields. Scalebars are 3 μm .

Response to reviewer #4

Comment: The authors of the manuscript "Structureless Excitation and Manipulation of Dynamic Holographic Plasmonic Slides" present a way to manipulate the phase, intensity and properties of plasmon excited on a flat metal-dielectric interface, to produce a holographic image. I found the paper interesting and engaging, with the experimental results showing improvement on current methods used. However, I'm unsure if the progress on the method is significant enough for publication in Nature Communications. I believe that this work will be more suited in a more specialised journal, such as Nanophotonics.

Reply: We sincerely thank the reviewer for the time and effort dedicated to evaluating our manuscript and for acknowledging that the work is interesting, engaging, and experimentally improves upon existing approaches. We appreciate the opportunity to clarify the broader impact and fundamental importance of our contribution.

The conventional plasmonic manipulation approaches rely on nanostructures, which inherently suffer from structural scattering, limited reconfigurability, and the inability to control key properties such as energy flow. These limitations have long restricted the fidelity and adaptability of plasmonic field engineering. In contrast, the present work fundamentally addresses these challenges by introducing an unstructured, reverse-engineered holographic excitation framework that enables an all-optical methodology.

This approach enables:

1. the generation of high-fidelity plasmonic fields devoid of structural scattering and mode distortion;

2.real-time reconfigurability of plasmonic profiles integrated with optomechanical functionality, representing a significant advancement toward scalable, and adaptive plasmonic systems;

3.controllable energy flow driven by orbital angular momentum (OAM), capable of transporting microparticles, thereby effectively realizing a structureless plasmonic optical tweezers.

This nanofabrication-free and reprogrammable platform transcends the limitations of static plasmonic configurations. To the best of our knowledge, this is the first demonstration of dynamically reconfigurable, structure-free holographic plasmonic fields that support functional energy-flow manipulation. We believe that these developments hold substantial potential for applications in precision manipulation, on-chip sensing, and reconfigurable photonics.

Nevertheless, I have a couple of points that the authors can improve on, before submitting the paper in a more specialized journal:

Comment 1: I'm a bit puzzled why the authors chose to do their experiments by optically trapping nanoparticles. Would a probe-tip not be a better choice, since it won't suffer from Brownian motion which alters the scattered fields measured during the experiment? Also, if the authors want to stick with optically trapped nanoparticles, then why not use dielectric ones that will induce less scattering that affects their results?

Reply 1: Thank you for your thoughtful and constructive comments. We address your questions point by point below.

(1) Why conduct experiments using optically trapped nanoparticles, instead of using a probe tip for field mapping?

Optical manipulation at the nanoscale has become increasingly important as advanced manufacturing, chip-level processing, and life sciences continue to progress toward smaller length scales. Plasmonic optical tweezers are particularly attractive in this context because the near-field confinement of surface plasmon polaritons enables significantly stronger gradient forces and weaker scattering forces compared with conventional free-space tweezers. However, this very advantage of plasmonic near fields also imposes a fundamental limitation: their confinement to the metal surface makes flexible modulation extremely challenging. Consequently, previous plasmonic tweezers have only demonstrated relatively simple trapping and translation operations.

The central contribution of our work lies not only in the creation of a structureless holographic plasmonic field, but also in demonstrating that such engineered fields can substantially expand the manipulation capabilities of nanoparticles, enabling functions that were previously inaccessible in plasmonic-field-based optical manipulation. The use of gold nanoparticles highlights the capability of our method to achieve complex and dynamic trapping in a highly challenging regime.

It is important to clarify that the field-mapping experiment is conceptually distinct from the optical-tweezing experiment, and is specifically designed to verify that we indeed generate the desired holographic plasmonic field. In the field-mapping measurements, the gold nanoparticle is immobilized on the substrate and therefore does not undergo Brownian motion. By analyzing the scattered field from this stationary nanoparticle, we reconstruct the spatial distribution of the holographic plasmonic field, following the procedure described in Ref. [25]. In contrast, in the optical-tweezing experiments, the nanoparticles are freely trapped and transported by the engineered plasmonic field in a liquid environment.

We fully agree that a probe-tip-based near-field method could also be used to measure holographic plasmonic fields. Here, we chose not to adopt this approach primarily for reasons of system compatibility and experimental simplicity. The stationary-nanoparticle field-mapping method can be implemented within the same optical platform as the plasmonic tweezers, without adding a separate near-field probe system. This integration allows us to characterize and manipulate the holographic plasmonic field in a unified setup, which is more practical and robust for the purposes of this work.

(2) Why not use dielectric particles for trapping?

Gold nanoparticles exhibit strong absorption and scattering, resulting in significant scattering forces that make them difficult to trap using traditional free-space optical tweezers. Therefore, demonstrating stable trapping of these challenging particles provides clear validation of the enhanced capabilities offered by our holographic plasmonic platform.

That said, our method is equally applicable to dielectric particles. To demonstrate this generality, we have included experimental results using SiO₂ microspheres, which exhibit stable trapping and guided transport along the designed holographic plasmonic trajectory (see Fig. R4 and Supplementary Video). These results confirm that the technique is robust for both metallic and dielectric particles.

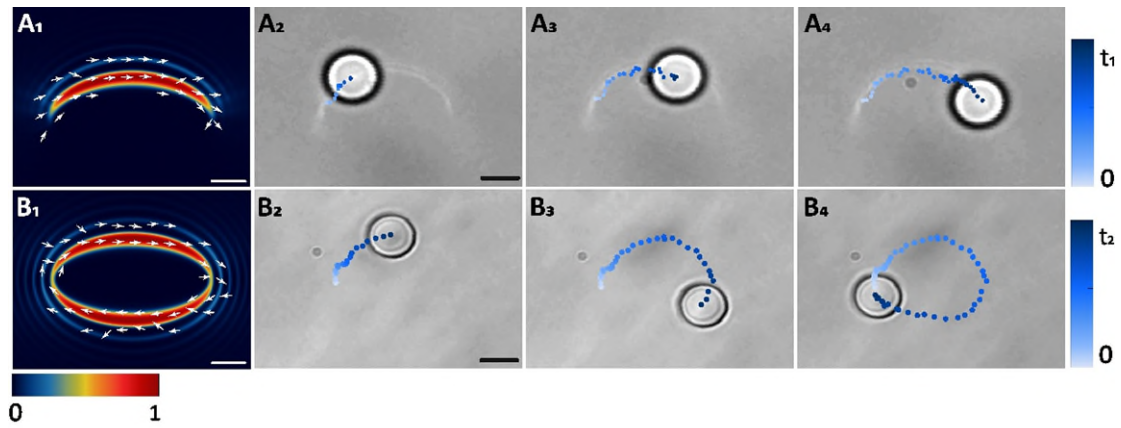


Fig. S4 | The corresponding experimental trajectory of the SiO₂ particles in the (A) Arc, and (B) Oval shaped plasmonic fields. Scalebars are 3 μ m.

We appreciate this comment for highlighting aspects that were not sufficiently emphasized in the original manuscript, particularly the significance of optical trapping. In the revised version, we have updated the title, abstract, and introduction to clarify the role and importance of plasmonic-field-based optical manipulation. Additionally, we have included the SiO₂ trapping experiments in the supplementary materials.

Correspondingly,

Line 277-282, Page 4: “In addition to trapping metallic nanoparticles, our method is also applicable to the micromanipulation of dielectric particles (see Supplementary Note S6). To demonstrate its generality, we further perform trapping experiments with SiO₂ microspheres, showing that these dielectric particles can likewise be guided along trajectories prescribed by the holographic plasmonic fields. These results confirm the universality of our approach for microparticle trapping and transport.”

Line 407-414, Page 16: “In this work, we perform two types of experiments: a field-mapping measurement to verify the generation of the holographic plasmonic field, and an optical-tweezing experiment to demonstrate its applicability to nanoparticle manipulation. Both experiments share the same optical setup; the main difference lies in the role of the nanoparticle. In the field-mapping experiment, the particle is immobilized on the metal film and used as a nanoscale probe that scatters the holographic plasmonic field for reconstructing its spatial distribution²⁵. In the optical-tweezing experiment, the particle initially undergoes unrestricted motion within a liquid medium before being confined and manipulated by the holographic plasmonic field.”

Add supplementary Note S6.

Experimental Transport of Dielectric (SiO₂) Microparticles in Holographic Plasmonic Fields

Benefiting from the fine control of plasmonic energy flow, the holographic plasmonic tweezer system is not limited to metallic particles but is equally effective for dielectric particles. We

additionally performed trapping experiments on dielectric particles, which exhibit relatively weak plasmonic responses. In experiment, a diluted suspension of SiO₂ microspheres was dropped onto the gold film. SiO₂ particles located near the potential wells were stably captured at the deepest valley under the axial gradient forces, and then continuously transported along the designed trajectories under the drive of the plasmonic energy flow, as shown in Fig. S4. It should be emphasized that the trapping power is also crucial for ensuring stable transportation. Considering different trajectory lengths lead to variations in the locally distributed energy, the incident power was set to 5 mW to balance trapping stability and transportation efficiency. Compared with metallic particles, dielectric particles exhibit weaker scattering, which contributes to their higher stability during transportation. These results confirm the generality and robustness of our technique across different particle types.

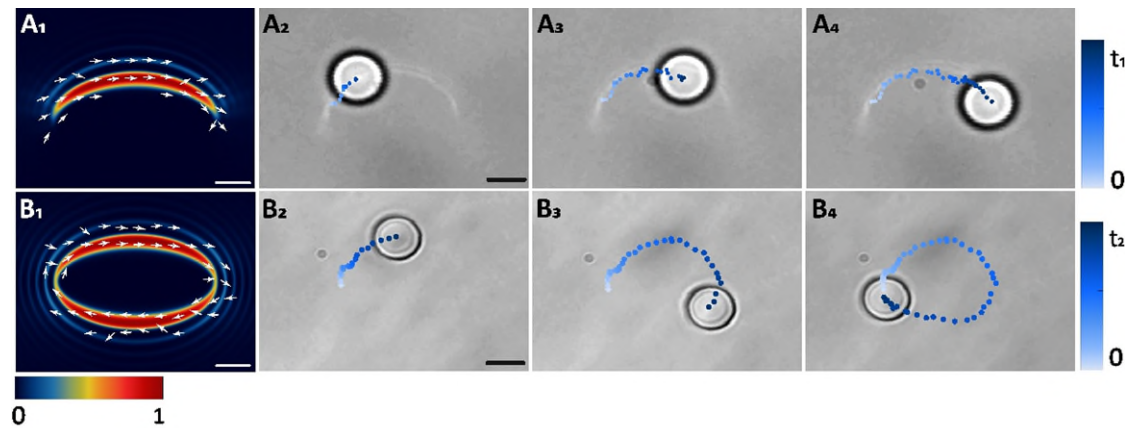


Fig. S4 | The corresponding experimental trajectory of the SiO₂ particles in the (A) Arc, and (B) Oval shaped plasmonic fields. Scalebars are 3 μ m.

Comment 2: I'm not sure how the authors match the momentum of the incident beam with propagating SPP modes. From Fig.2, it seems that they always excite with normal incidence. I understand in Fig2A that there is a nanoring slit that allows for this momentum matching for certain part of the incident normal beam. But how do the authors manage to achieve a better momentum match in fig.2D (and future configurations in the paper)?

Reply 2: Thank you for raising this important question. It is well known that the in-plane momentum of an SPP exceeds that of light in free space, and therefore additional momentum compensation is required for efficient excitation. Several common wavevector-matching mechanism as follows.

The first configurations is prism-based coupling. The classical Otto and Kretschmann configurations, introduced in 1968, compensate the in-plane momentum by tuning the incidence angle(Fig. R5a-b) [*Zeitschrift für Naturforschung A*. 23, 2135-2136 (1968); *Zeitschrift für Physik A Hadrons and nuclei*. 216, 398-410 (1968)]. This scheme is simple and efficient, However, the propagating SPP diverge along the metal surface

and do not generate sufficiently strong optical gradient force, this approach has long been considered unsuitable for optical trapping.

Second, surface plasmons can also be excited by near field scattering by a tip near a metal surface (Fig. R5d), such as by particles or nanometre-sized scatters on a surface (Fig. R5e). Since near-field scatterers have a wide spatial frequency spectrum, there are components that will satisfy the matching conditions required to excite the SPP. This approach is simple but often inefficient because only a small fraction of the scattered light contributes to excitation. To enhance efficiency, periodic gratings are often introduced. They supply defined spatial frequency components that mix with the incident light and create strong momentum components matching the SPP wavevector (Fig. R5f). The fixed nature of nanostructures greatly limits the dynamic modulation of the excited plasmonic field. In addition, imperfections in the structures can introduce extra scattering, leading to a reduction in the intensity of the desired optical field.

In our system, a high-NA objective produces a continuum of incident wavevectors around the nominal optical axis (Fig. R5c). When the metal film is positioned before the focal plane, only the annular portion of the illumination that meets the surface-plasmon resonance (SPR) angle contributes to coupling. This annular band functions similarly to the ring illumination used in prism-based schemes: it selectively concentrates optical power near the required in-plane momentum while suppressing direct transmission through the metal. The excited SPP then act as secondary circular wave sources that propagate inward and coherently interfere to form a strong central convergence point Ref. [5] & [*Optics letters*. **37,4627-4629 (2012)].**

This coupling mechanism is directly responsible for the improved momentum matching observed in Fig. 2D and subsequent configurations. The designed holographic structures reshape the spatial phase of the incident field so that the high-NA objective preferentially redistributes energy into the angular components that most efficiently satisfy the SPP dispersion relation. As a result, this configuration inherently provides the in-plane momentum required for efficient excitation of surface plasmons, while eliminating fabrication-related constraints and maintaining high excitation efficiency. [*Plasmonics*. **13**, 991-996 (2018)]. Furthermore, this efficiency can be improved by employing perfect radially polarized beams, by compressing the energy into a thin ring that satisfies the plasmonic coupling conditions.[*Photonics Research*. **6**, 847-852 (2018)].

[editorial note: figure redacted]

Correspondingly,

Line 75-77, Page 3: “Principles. To generate a holographic plasmonic field, as illustrated in Fig. 1A, a specifically designed incident light field is utilized in a tight focusing system to illuminate a smooth metallic thin film. The high-NA focusing system provides large incident angles, so that the in-plane wave-vector components satisfy the required wave-vector matching condition for plasmon excitation⁵. In our approach, the incident light field is produced with a reversal design algorithm, which enables the precise control of the formed plasmonic field as desired. ...”

Comment 3: The first sentence of the introduction is not technically correct. The propagating SPPs on metal-dielectric interfaces are eigenmodes, which means that they are always there. One does not need to illuminate the interface with light for the evanescent modes to emerge. But one needs external illumination to couple light into these modes, which is what I believe the authors were trying to communicate?

Reply 3: Thank you for the clarification. We agree with the reviewer’s statement that SPP at a metal–dielectric interface are eigenmodes that inherently exist without external illumination. In the revised manuscript, we have corrected the introductory sentence accordingly to accurately reflect this physical concept.

Correspondingly,

Line 27-31, Page 1: “Surface plasmons are electromagnetic eigenmodes bound to metal – dielectric interfaces, with evanescent fields that decay away from the surface¹. By coupling incident light into these surface modes under appropriate wave-vector matching conditions, one obtains a propagating plasmonic wave that runs along the interface and remains tightly confined near it, effectively forming a near-field optical “belt”²⁻⁴. ”

Comment 4: Terms like "stray signals" or "stray light" are not proper scientific terminology. This light/signal comes from diffraction, and one should say this.

Reply 4: Thank you for your comment. To improve scientific accuracy, we have used the “diffraction components” in the revised manuscript.

Correspondingly,

Line 14-16, Page 2: “However, holographic plasmonic fields are usually realized with complex nanostructures, introducing [diffraction background](#) and limiting flexibility and dynamic reconfigurability.”

Line 48-50, Page 2: “Primarily, the metallic nanostructures introduce unavoidable [diffraction components](#) and significant energy losses, which detract from the fidelity of the generated plasmonic field²⁰.”

Line 116-118, Page 4: “Furthermore, the [diffraction background](#) induced by the structure significantly degrades the signal-to-noise ratio and accuracy of the generated plasmonic field.”

Line 127-128, Page 4: “In the absence of structure-induced [diffraction background](#), the pattern in Fig. 2E exhibits a significantly cleaner background and a higher excitation efficiency than that in Fig. 2B.”

Comment 5: Line 53: "Some other properties, such as energy flow, are challenging to manipulate according to desired patterns, ..." Why is this the case? What is stopping current methods to do this?

Reply 5: Thank you for your comment. Controlling the energy flow distribution is inherently more challenging because it requires precise manipulation of both amplitude and phase. Current limitations in micro- and nano-fabrications introduce unavoidable structural deviations that generate diffraction and background crosstalk, making it difficult to accurately reproduce the target phase distribution. Moreover, most current SPP modulation schemes rely solely on phase control [*Nano Letters*, **17**, 7920(2017); *Sci. Adv.* **4**, eaao0533 (2018); *Nat. Commun.* **6**, 8360 (2015)], whereas our inverse-design approach enables full complex-amplitude modulation. To the best of our knowledge, this work is the first experimental demonstration of controlled trajectory transport of small structures using plasmonic tweezers.

Correspondingly,

Line 53-56, Page 2: “Some other properties, such as energy flow, are challenging to manipulate according to desired patterns [because the diffraction background perturbs the phase of the plasmonic field](#)^{12,19-21}. These [properties](#) are particularly important for the modulation of on-chip optical energy and particle transmission.”

Add Reference:

21. P. R. Huft, J. D. Kolbow, J. T. Thweatt, N.C. Lindquist, Holographic plasmonic nanotweezers for dynamic trapping and manipulation. Nano Letter. 17, 7920(2017).

Comment 6: I'm not sure what is a toroidal orbital angular momentum. It would be convenient for the reader if the authors defined what they mean by it.?

Reply 6: Thank you for your insightful comment. We apologize for any possible confusion caused by the use of the term “toroidal orbital angular momentum (OAM)” in the original manuscript, as it may be mistaken for toroidal vortices or toroidal dipoles in other contexts. In our work, the inverse-design approach reshapes the OAM distribution into a ring-shaped pattern, generating a circular in-plane energy flow on the plasmonic surface. To avoid confusion, we have replaced “toroidal OAM” with “ring-shaped OAM” throughout the revised manuscript.

Correspondingly,

Line 62-63, Page 2: “ Furthermore, it is demonstrated that by loading the ring-shaped orbital angular momentum (OAM) onto the incident light,...”

Comment 7: What is the asterix on I1* in table 1? It is never explained, or if it is I missed it completely.?

Reply 7: Thank you for pointing out this issue. The asterisk (“*”) appearing after I₁ in Table 1 was an inadvertent typographical error and does not indicate any special condition or footnote. There is no corresponding explanation in the text. In the revised manuscript, we have removed the asterisk to avoid confusion and ensure consistency.

Correspondingly,

Line 191, Page 7:

Table 1 | Quantitative analysis of the holographic plasmonic OAM belts

	<i>Correlation index</i>	I_1/I_0	I_2/I_1	I_3/I_1
<i>Oval</i>	γ	0.9390	0.9874	0.8445
	η	0.8255	0.8276	/
<i>Arc</i>	γ	0.9243	0.9438	0.8873
	η	0.8493	0.8108	/
<i>Helix</i>	γ	0.9335	0.9406	0.7747
	η	0.8191	0.7573	/
<i>Cycloid</i>	γ	0.8011	0.7804	0.5831

	η	0.6578	0.5649	/
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Comment 8: There are a 3-4 places that the language and grammar can be improved.

For example:

line 121 "Fig.2E is significantly clean with higher excitation..." should probably be:

"Fig.2E is significantly more clean with higher excitation..."

line 175 "This imperfect is mainly...", should probably be : "These imperfections are mainly ..."

.?

Reply 8: Thank you for pointing out these language issues. The highlighted phrases have been corrected in the revised manuscript. In addition, we have also carefully reviewed the manuscript for comparable grammatical and stylistic concerns, and the wording has been refined in multiple sections to enhance clarity and readability. All modifications are highlighted in the revised version.

Response to the Decision Letter

REVIEWERS' COMMENTS

Reviewer #1 (Remarks to the Author):

The authors have satisfactorily addressed all of our comments. We recommend accepting the revised manuscript in its current form.

Response: We thank the reviewer for the positive assessment and recommendation. We appreciate your careful review and are pleased that our revisions have fully addressed your comments.

Reviewer #2 (Remarks to the Author):

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Response: We sincerely thank the reviewer for their contribution and for participating in this peer-review training initiative. We appreciate the effort and thoughtful evaluation of our work, and we are grateful for the support of this initiative.

Reviewer #3 (Remarks to the Author):

I appreciate the authors' responses and revisions. All of my previous questions and comments have been addressed, and the manuscript is now complete and clear. I would recommend the paper for publication.

Response: We sincerely thank the reviewer for the encouraging feedback. We are glad that the revisions addressed all of your previous questions and comments, and we appreciate your recommendation for publication.

Reviewer #4 (Remarks to the Author):

The authors improved the manuscript significantly and answered my questions well. I'm happy to accept now the paper for publication in Nature Communications.

Response: We thank the reviewer for the positive evaluation. We appreciate your recognition of the improvements and are grateful for your support of publication.