



Focusing light with a metal film coated patchy particle

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Abstract: Microsphere-assisted super-resolution imaging is a promising technique that can significantly enhance the resolution of conventional optical microscopes. The focus of a classical microsphere is called photonic nanojet, which is a symmetric high-intensity electromagnetic field. Recently, patchy microspheres have been reported to have superior imaging performance than pristine microspheres, and coating microspheres with metal films leads to the formation of photonic hooks, which can enhance the imaging contrast of microspheres. Understanding the influence of metal patches on the near-field focusing of patchy particles is important for the rational design of a nanostructured microlens. In this work, we theoretically and experimentally showed that the light waves can be focused and engineered using patchy particles. When coating dielectric particles with Ag films, light beams with a hook-like structure or S-shaped structure can be generated. Simulation results show that the waveguide ability of metal films and the geometric asymmetry of patchy particles cause the formation of S-shaped light beams. Compared with classical photonic hooks, S-shaped photonic hooks have a longer effective length and a smaller beam waist at far-field region. Experiments were also carried out to demonstrate the generation of classical and S-shaped photonic hooks from patchy microspheres.

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1. Introduction

The Abbe diffraction limit is a fundamental problem in optical science and engineering, as it limits the resolution of optical imaging systems. Microlenses have proven to be a powerful tool for overcoming this diffraction limit. In 2009, Lee et al. first found that spherical microlenses made of organic molecules can resolve features beyond diffraction limit [1]. Later in 2011, Wang et al. reported that silica microspheres with a diameter between 2 - 9 μm have super-resolution ability and can resolve 70 nm feature size under white light illumination [2]. Since then, many discoveries and improvement have been made to this technology, such as liquid-immersion microscopy [3,4], scanning microsphere imaging [5,6], development of biomaterial microlenses [7,8], etc. Nowadays, microsphere-assisted imaging has become one of the most promising nanoscopy technique with the merits of enabling label-free and real-time super-resolution imaging.

When a microsphere is illuminated by light waves at visible spectrum, a highly concentrated, non-evanescent energy flux with subwavelength beam waist will be generated at the shadow side of the microsphere [9]. This optical phenomenon is called photonic nanojet (PNJ). Although the mechanism of microsphere imaging is still not clear [10], it has been experimentally demonstrated

that the imaging performance of a microlens can be significantly influenced by the position, effective length and full-width-half-maximum (FWHM) of its PNJ [11–13]. Various methods have been proposed to engineer the PNJ and to improve the imaging performance of microspheres. In 2016, Wu et al. reported the super-focusing phenomenon of center-covered microsphere [14]. The patterned microsphere can adjust the transverse component of the incident beam and achieve a sharp PNJ with a FWHM of 0.387λ . In 2021, microspheres with segmented region of diffractive patterns were successfully used to generate a bottle-like PNJ [15]. Later in 2022, Wu et al. coated microspheres with bilayer films to suppress interface reflection and enhance the quality of imaging [16].

In addition to this jet-like light beam, microparticles also can generate energy flow with curved trajectory, which is called photonic hook (PH) due to its hook-like structure [17]. Compared with PNJ, the PH has a smaller FWHM and a better tunability [18–20]. Their unique subwavelength bending may have potential applications in particle manipulation and laser micromachining. An important practical application of PH is in super-resolution microscopic imaging. In 2021, Shang et al. found that patchy microspheres have a better imaging performance than pristine microspheres. Coating metal films on microspheres significantly enhances their imaging contrast to $6.5 \times$ higher [20]. The anisotropic optical property of patchy particles breaks the symmetry of imaging systems, and leads to the formation of PHs [21]. This near-field asymmetric illumination significantly improves the contrast of the sample, which in turn leads to better imaging performance of the microscope. Asymmetric illumination is a commonly used technique in computational microscopic imaging to enhance the contrast of objects [22]. The concept of the PH-assisted contrast enhancement technique was also extended to the microscopic imaging in the terahertz spectrum [23]. However, the light focusing of patchy microparticles still needs further investigation to understand the physical mechanism behind this phenomenon.

In this work, we investigated the light focusing properties of patchy particles under plane wave illumination. 2D and 3D simulations based on the Finite-Difference Time-Domain (FDTD) method have been performed to theoretically study the light fields generated by anisotropic microcylinders and microspheres, respectively. Experiments were also carried out using patchy microspheres to observe the classical and S-shaped PH phenomenon. To the best of our knowledge, it is the first time that the generation of PHs from patchy particles is experimentally demonstrated. The findings of this work have potential applications in super-resolution imaging, micromachining, optical trapping, etc.

2. Simulation methods

First, we theoretically investigated the focusing performance of patchy microcylinders. Dielectric microcylinders partially covered with Ag films are illuminated with plane waves. The light field at the shadow side of the microcylinder was obtained using the FDTD method with a commercial software (Lumerical FDTD Solutions). Figure 1 are the schematic drawing of the 2D sectional view of the investigated model. As shown in the drawing, an intense focusing of light will occur on the shadow side of the cylinder when a S-polarized monochromatic plane wave ($\lambda = 550$ nm) propagating parallelly to the X axis passes through the cylinder. In this study, the microcylinder has a constant refractive index (RI) of 1.90, the same as the RI of BaTiO₃, which is a high-index dielectric material widely used in microsphere-assisted super-resolution imaging [5,20,24]. The diameter of the cylinder changes between 1 - 5 μm and the RI of the background changes between 1.00 - 1.52. For the entire computational domain, non-uniform meshes with RI-dependent element size were used and all of them are smaller than $\lambda/30$. The size and position of Ag films can be defined by their opening angles (α, η) and position angles (θ, δ).

A typical focusing of a patchy microsphere starts out with an approximately straight energy flow. The intensity distribution has a curved structure and eventually straightens out after a certain distance. The structure of a S-shaped PH can be approximately defined by the start point

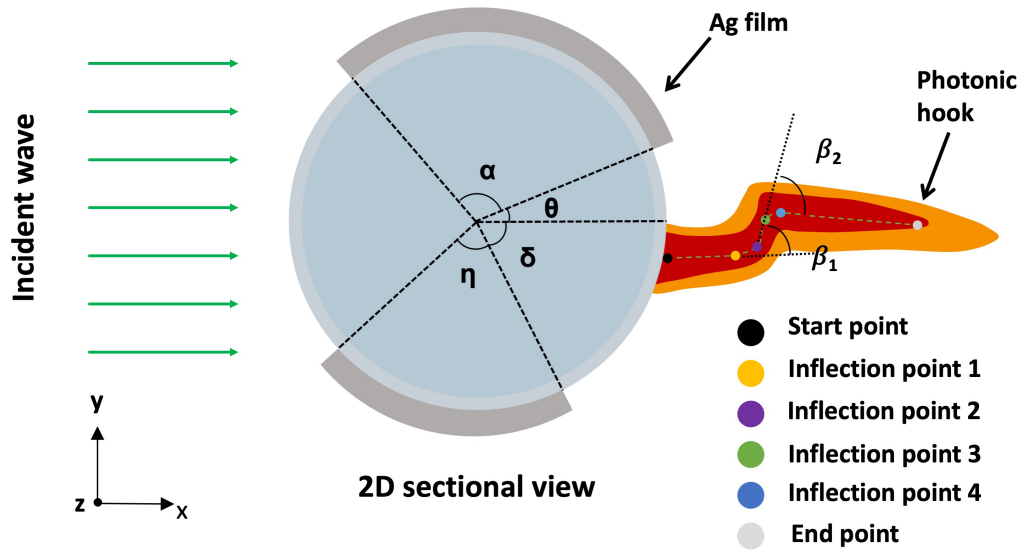


Fig. 1. Schematic drawing of a patchy particle illuminated with plane waves.

(SP), the four inflection points (IP₁, IP₂, IP₃ and IP₄) and the end point (EP) of the PH. The effective length (L_{eff}) is defined as the distance between the SP and EP. To find these points, we sliced the PHs along the propagation direction of incident light at a step resolution of 100 nm, and find the maximum intensity positions in each slice. After fitting the maximum intensity points, we can obtain a smooth line representing the structure of PHs. We call this line the midline because this line is approximately in the middle of the PHs.

The inflection points are the points at which the curvature state of the midline changes [18]. The end point in this study is defined as the point on the midline of the PH with an intensity enhancement factor of I_{max}/e [25,26]. The I_{max} is the largest $|E|^2$ enhancement formed at the shadow side of the patchy microcylinder. Its value is the enhancement factor of $|E|^2$ with respect to that of the incident light. Based on these points, the curvature of the S-shaped PHs can be defined by the bending angle β_1 and β_2 . β_1 is the angle between the two lines respectively connecting the SP with the IP₁, and the IP₂ with the IP₃. β_2 is the angle between the two lines respectively connecting the IP₂ with the IP₃, and the IP₄ with the EP.

3. Results and discussion

3.1. Comparison between the PNJ, PH and S-shaped PH

Then, we compared the light fields formed by a pristine microcylinder, a patchy cylinder with one piece of Ag film and a patchy cylinder with two pieces of Ag films. The microcylinders have a constant RI of 1.90 and a diameter of 2 μm . The background RI is 1.33, same as the RI of water. As shown in Fig. 2(a), plane waves propagating through a pristine cylinder become a PNJ on the shadow side of the cylinder, which has a symmetric spatial distribution of the intensity along the propagation direction of light. A PH with a curved structure can be generated from patchy microcylinders with one piece of Ag film [Fig. 2(b)]. By using two pieces of metal films, PHs with a S-shaped structure can be effectively generated, as shown in Fig. 2(c). We also performed simulations using the ray tracing method based on geometrical optics, and we found that the geometrical optics cannot predict the formation of S-shaped PHs, but it makes a good prediction about the formation of PNJ and PH [the insets in Figs. 2(a)-(c)].

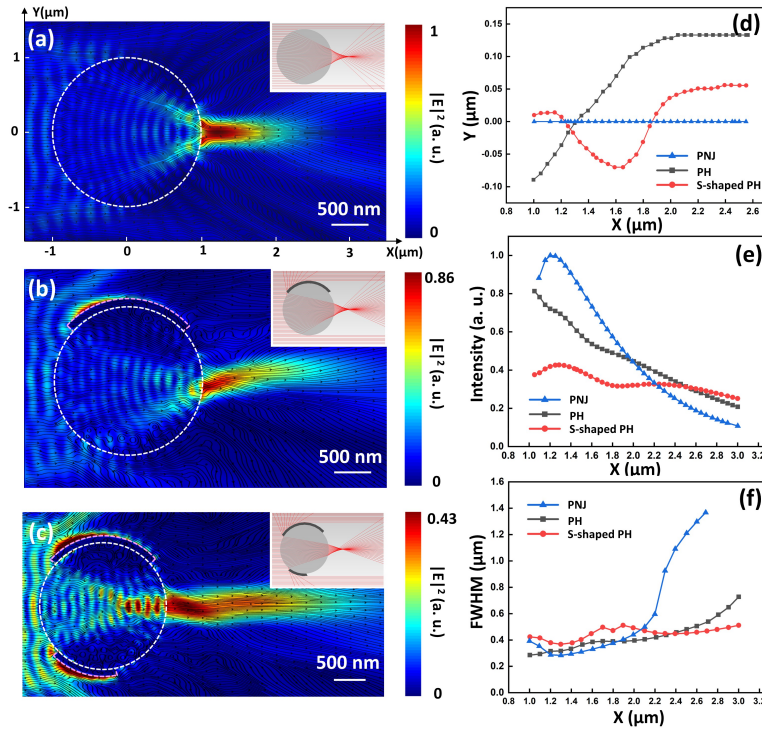


Fig. 2. (a)-(c) Simulated light fields corresponding to the normalized $|E|^2$ distribution of (a) a photonic nanojet, (b) a classical photonic hook and (c) a S-shaped photonic hook. (d)-(f) The properties of the three types of energy flow; (d) The change of the maximum intensity position along the X axis; (e) The change of the value of the maximum intensity along the X axis; (f) The change of the FWHM of the intensity profiles along the X axis.

To show the properties of the light fields, we sliced them along the X axis and recorded the position and value of the maximum intensity as well as the FWHM of the intensity profiles in each slice. The variation of the maximum intensity position along the X-axis confirms the straight, bended and S-shaped structure of PNJ, PH and S-shaped PH, respectively [Fig. 2(d)]. As shown in Fig. 2(e), the S-shaped PH maintains in a concentrated light wave after the propagation of 3.7λ outside the right edge of the patchy microcylinder. The intensity of the PNJ, PH and S-shaped PH decreased to 11%, 25% and 60% of their corresponding maximum values, respectively. The S-shaped PH has the smallest rate of decline compared to the other two types of energy flow. The S-shaped PH also has the longest effective length. The effective length of a PNJ, a classical PH and a S-shaped PH is 2.05λ , 2.84λ and 5.02λ , respectively.

In this study, we also compared the waist of the three types of concentrated light waves. As shown in Fig. 2(f), we found that the S-shaped PH can maintain a subwavelength waist of FWHM = 0.9λ even after a propagation distance of 3.7λ outside of the right edge of particle, while the FWHM of the PNJ increases to 2.5λ after 3.1λ propagation. The PH has a FWHM of 1.4λ at a propagation distance of 3.7λ . The S-shaped PHs have the narrowest beam waist after existing the particle and propagating another distance of 2.55λ .

3.2. Formation mechanism of S-shaped PHs

As reported in the previous work [17,26], the formation mechanism of the curved light field formed by patchy particles can be analyzed with the time-averaged Poynting vectors. In this

work, the field lines of the Poynting vector are shown as black lines in Figs. 2(a)-(c). We can see that the energy flow of the light field of pristine cylinders is focused into a classical photonic nanojet at the shadow side of the microcylinder [Fig. 2(a)]. As for patchy cylinders, the incident electromagnetic (EM) waves are partially blocked by the Ag film and then reflected backwards to the free space. This breaks the symmetry of illumination and makes the spatial distribution of the energy flow inside the microcylinder asymmetric. This asymmetric flow of energy is then focused into a curved beam after leaving the patchy cylinder, as shown in Figs. 2(b),(c).

However, the unbalanced energy flow alone cannot explain the formation of S-shaped spatial distribution of light field shown in Fig. 2(c). Therefore, we used the Movie Monitor in the Lumerical FDTD Solutions to record the evolution of the $|E|^2$ field component for the duration of the simulation. The result is shown in Visualization 1. As shown in Visualization 1, we found that the incident light excites the surface plasmon polariton (SPP) on the surface of Ag films, and its propagation direction can be significantly changed. The superposition of the SPP waves and the conventional PHs leads to the formation of such S-shaped intensity distribution. SPPs are EM modes propagating along metal–dielectric interfaces, in which surface collective excitations of free electrons in the metal are coupled to evanescent EM fields in the dielectric layer. Metal films have been reported to be able to transport EM energy as SPP waveguides [27]. Minin et al. also theoretically and experimentally demonstrated the generation of plasmonic hooks based on SPP excitation [28,29].

3.3. Influence of Ag film positions and background RI

To investigate the effect of silver film position on the light focusing properties of patchy particles, we first study the situation when the microcylinder is covered with only one piece of Ag film. As shown in Fig. 3, the cylinder has a diameter of $2\ \mu\text{m}$. The opening angle of the silver film is kept constant at 98° . The Ag film was rotated counterclockwise by increasing θ from 10° to 50° . As shown in Fig. 3(a), we find that the silver film will obstruct the formation of PHs when it is close to the SP of the PH. We found that the PHs can be effectively generated when the silver film moves away from the SP [Figs. 3(b)-(d)]. At a rotation angle of 20° , we can observe the formation of S-shaped PHs generated by the superposition of SPP waves and conventional PHs [Fig. 3(b)]. As the silver film continued to rotate counterclockwise, the influence of SPP waves gradually weakened, and conventional PHs become dominant in the light field.

Then, we keep the position of the Ag film on top unchanged and rotate the Ag film at bottom clockwise by increasing δ from 20° to 90° [Fig. 4(a)]. Patchy microcylinders cannot generate PHs when δ is less than 20° . As shown in the red line in Fig. 4(b), the focal length (FL) is defined as the distance between the $I_{\max}$ point and the center of the microcylinder. We found that the FL can be effectively adjusted by changing the position of Ag films. The FL obtained at $\delta = 90^\circ$ is $\sim 25\%$ longer than that obtained at $\delta = 30^\circ$. The furthest focal point (FL $\sim 1.3\ \mu\text{m}$) is obtained at $\delta = 80^\circ$. The FL is constant when δ is between 20° and 30° because the Ag film is not in the propagation path of incident light, and it has an approximately linear increase when increasing δ from 30° to 80° . We also measured the $I_{\max}$ of the PHs at different δ angles, and found that the $I_{\max}$ decreases to less than 50% of its original value when increasing δ from 20° to 90° due to the blocking effect of Ag films [blue line in Fig. 4(b)]. We can obtain the following formulas: $\text{FL} = 0.0059\ \delta + 0.8497$ and $I_{\max} = -0.055\ \delta + 8.615$ by performing curve fitting within the range of $\delta = 30^\circ$ to 80° , and 30° to 90° , respectively.

The bending of the light intensity distribution can also be effectively adjusted by changing the position of Ag films. Figure 4(c) shows the evolution of the first bending (β_1) and second bending (β_2) of the generated light field as a function of δ angle. When the Ag film is rotated from $\delta = 30^\circ$ to $\delta = 50^\circ$, the opening at the left edge of patchy microcylinder has a higher degree of geometrical symmetry, and the incident light consequently has a smaller phase velocity difference. This makes β_1 decrease from 52.1° ($\delta = 20^\circ$) to 39.2° ($\delta = 50^\circ$). Further rotating

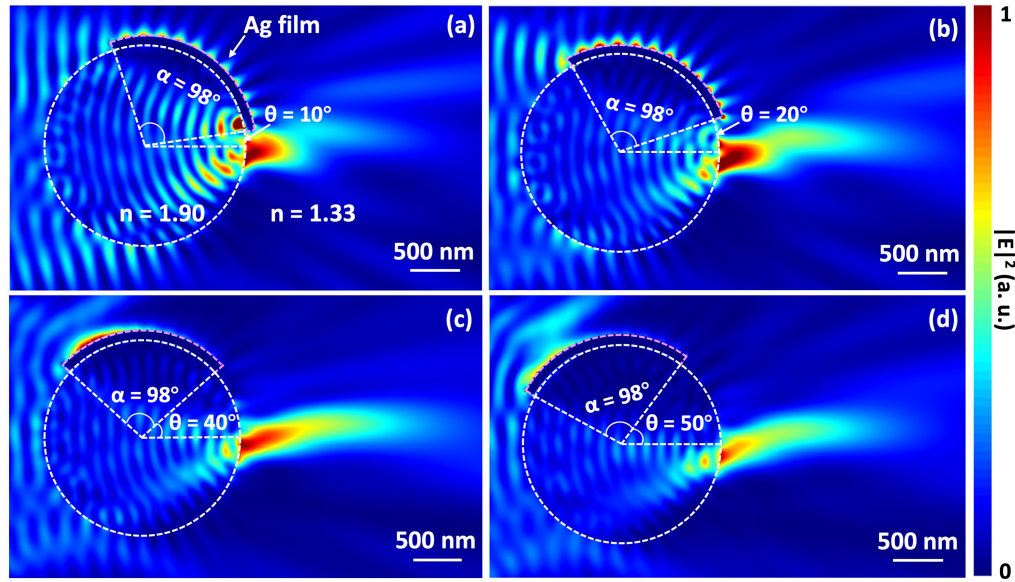


Fig. 3. Light fields generated by 2 μm -diameter patchy microcylinders coated with one piece of Ag film. The rotation angle (θ) of the Ag film is (a) 10° , (b) 20° , (c) 40° and (d) 50° , respectively.

the Ag film from $\delta = 50^\circ$ to $\delta = 90^\circ$ increases the geometrical asymmetry of the left opening, making the light waves obtain a higher phase velocity difference. β_1 thus increases from 39.2° ($\delta = 50^\circ$) to 60.1° ($\delta = 90^\circ$). The bending angle (β_2) corresponding to the second inflection point slightly increases from 35.7° to 43.9° when increasing δ from 30° to 90° . The bending angles obtained at $\delta = 20^\circ$ are same as those obtained at $\delta = 30^\circ$. In this study, the light field with the highest curvature ($\beta_1 = 60.1^\circ$, $\beta_2 = 43.9^\circ$) is obtained at $\delta = 90^\circ$.

By controlling the refractive index of the immersion medium, we can adjust the effective length (L_{eff}) of the energy flux. We found that the microcylinder in air ($n_{bg} = 1.00$) has a short PH with an effective length of $0.6 \mu\text{m}$ [Fig. 5(a)]. As shown in Fig. 5(b), increasing the n_{bg} to 1.52 leads to the formation of a longer photonic flux ($L_{\text{eff}} = 4.0 \mu\text{m}$). The PH clearly shows a S-shaped structure. We also set the refractive index of the immersion medium to that of water ($n_{bg} = 1.33$). The effective length of the PH is found to be $2.2 \mu\text{m}$ in water [Fig. 5(a)]. Considering that water is one of the most widely used biocompatible liquids in both research and industry, we will use water as the immersion medium in the following studies.

3.4. 3D simulations with patchy microspheres

In addition to patchy microcylinders, we also performed 3D simulations to study the light fields generated by Ag film-coated microspheres. As shown in Fig. 6, patchy microspheres ($n_{\text{sphere}} = 1.90$) partially covered with different ratios of 100 nm-thick Ag films were illuminated by 550 nm plane waves. The microspheres have a diameter of $2 \mu\text{m}$ in Figs. 6(a1), (b1), (c1), and a diameter of $10 \mu\text{m}$ in Figs. 6(a2), (b2), (c2). The coverage ratio (CR) of Ag films, which is defined as the area of Ag films divided by the area of the microspheres, is 30% [Fig. 6(a)], 43% [Fig. 6(b)] and 54% [Fig. 6(c)], respectively. The background medium is assumed to be water ($n_{bg} = 1.33$). The center of the microspheres is at the origin of coordinates. The incident light propagates in the Z direction (black arrows) and is polarized along the Y axis (red arrows).

As shown in Fig. 6, we found that similar to patchy microcylinders, coating microspheres with Ag films also leads to a curved focus under plane wave illumination. The microsphere with the

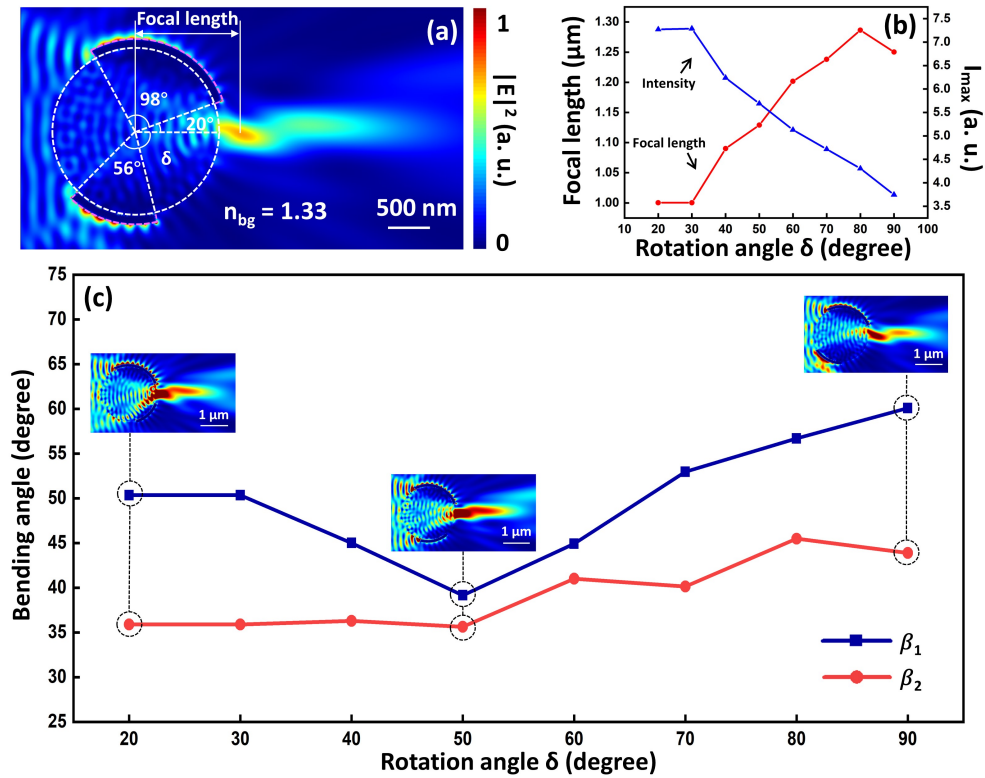


Fig. 4. (a) Normalized light fields generated by $2\ \mu\text{m}$ -diameter patchy particles when the background RI (n_{bg}) is 1.33; (b), (c) Properties of the light field generated by the patchy microcylinder in water as a function of rotation angle δ : (b) The focal length of the patchy microcylinder and (c) the bending angle of the photonic hooks.

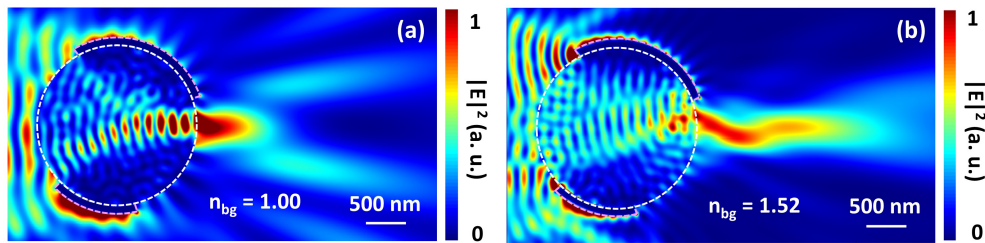


Fig. 5. (a)-(b) Normalized light fields generated by $2\ \mu\text{m}$ -diameter patchy particles when the background RI is (a) $n_{bg} = 1.00$, (b) $n_{bg} = 1.52$.

largest CR of Ag films produce PHs with the highest curvature [Figs. 6(c1), (c2)]. This optical phenomenon can be explained as follows. In this work, the formation of curved focus is caused by the structural asymmetry of patchy particles. We can see that the structure of the microspheres for simulation is symmetric to the X-Z plane and asymmetric to the Y-Z plane. Then, we can decompose the energy at focus into two parts. The first part is the light energy from the center of patchy microspheres, which obtains a curved structure after being modulated by the metal films. The second part is the light energy focused by the naked edges of patchy microspheres, which generates a symmetrical intensity distribution. Therefore, by removing the second part of light

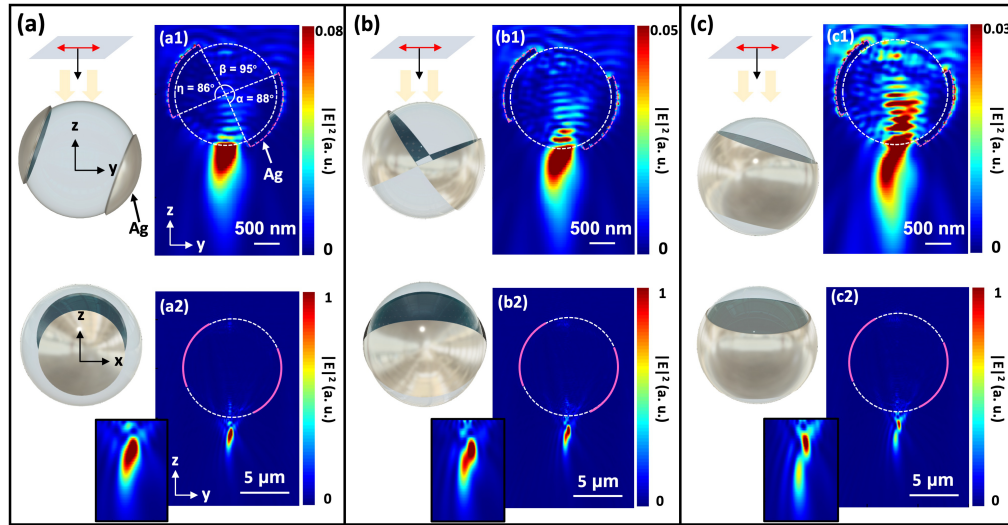


Fig. 6. 3D simulations of the photonic hooks generated by patchy microspheres. (a1)-(c1) The diameter of the microsphere is $2\ \mu\text{m}$; (a2)-(c2) The diameter of the microsphere is $10\ \mu\text{m}$.

energy from the concentrated light field, the curved structure of PHs can be more prominent, and the features such as subwavelength bending can thus become more visible.

The effective length of PHs is another important feature, which determines the working distance of these energy flows. People usually prefer PHs with long effective lengths because they can be used to process samples remotely. As shown in Fig. 6, we can effectively obtain PHs with different effective lengths by properly designing the coverage ratio of the Ag films on the two sides of patchy microspheres. The corresponding effective length increases from $1.04\ \lambda$ [Fig. 6(a1)] to $1.64\ \lambda$ [Fig. 6(c1)] for $2\ \mu\text{m}$ -diameter microspheres, and from $5.07\ \lambda$ [Fig. 6(a2)] to $5.82\ \lambda$ [Fig. 6(c2)] for $10\ \mu\text{m}$ -diameter microspheres. The detailed physical parameters of the generated PHs are shown in Table 1.

Table 1. Focusing properties of patchy microspheres

Microsphere Diameter	$2\ \mu\text{m}$			$10\ \mu\text{m}$		
CR (%)	30	43	54	30	43	54
I_{max} (a. u.)	33.5	19.2	11.1	269.9	241.0	215.8
L_{eff} (μm)	0.57	0.73	0.90	2.79	2.92	3.20
FWHM (λ)	0.62	0.62	0.62	0.73	0.62	0.56

3.5. Experimental observation of PHs

In this work, we experimentally demonstrated that PHs with S-shaped spatial distribution can be generated from patchy microspheres. To fabricate patchy particles, pristine barium titanate glass (BTG) microspheres (Microspheres-Nanospheres, USA) with a diameter of $10\ \mu\text{m}$ were deposited with 30 nm-thick Ag films by the glancing angle deposition method [30]. The inset in Fig. 7(a) is the SEM image of a patchy microsphere, in which we can observe the presence of Ag films on both sides of the microsphere. Then, we illuminated the patchy microsphere using a halogen white light source with a green interference filter (central wavelength $\lambda = 550\ \text{nm}$, 45 nm bandwidth) from below. This created a light field above the patchy microsphere. A

microscopic objective lens (100 $\times$, 0.9 NA, EC EPIPLAN, Carl Zeiss) was used to observe the evolution of the light field during the Z axis scanning with a step resolution of 100 nm [Fig. 7(b)]. The experiment was carried out under an optical microscope (Axio AX10, Carl Zeiss). During scanning, serial transverse cross sections corresponding to serial focal planes along the Z axis were recorded with a digital camera (DFC295, Leica).

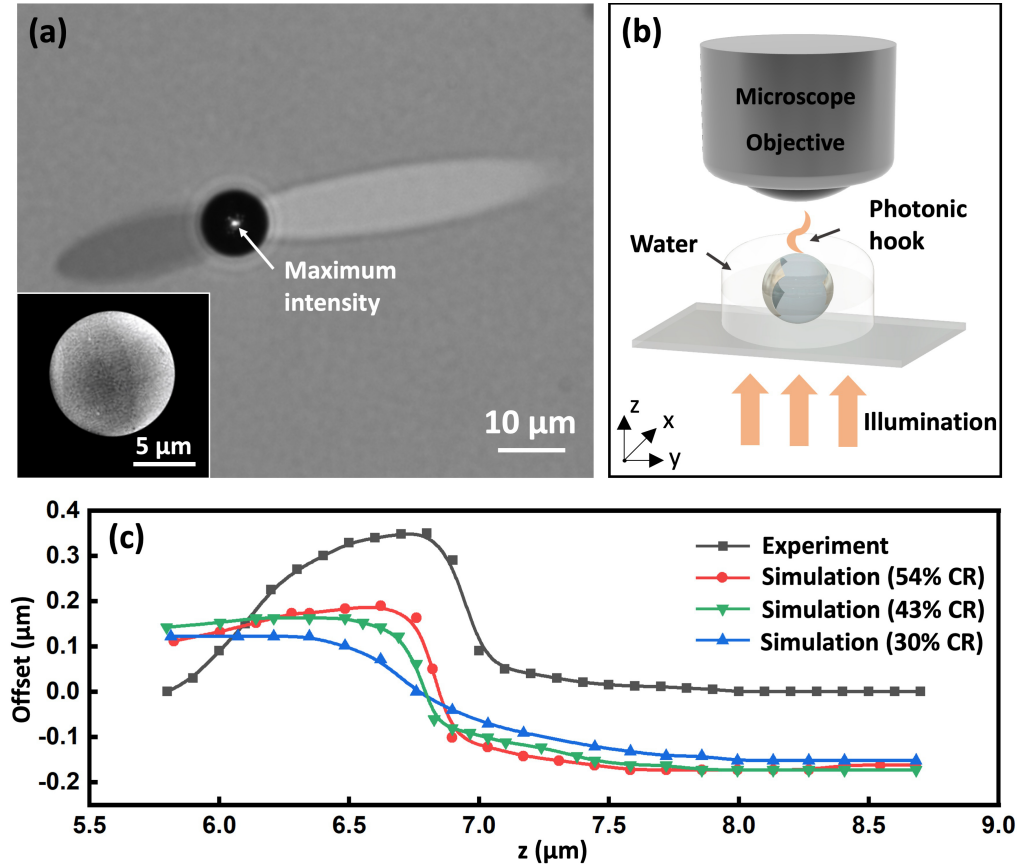


Fig. 7. (a) Optical microscopic images and scanning electron microscopic (SEM) image of a patchy microsphere after Ag deposition; (b) Schematic drawing of the experimental setup for the observation of the curved focus; (c) Experimental and simulated maximum intensity profiles of the PHs obtained by scanning the Z axis.

We found that the position of the maximum intensity area, i.e. the bright spot indicated by the white arrow in Fig. 7(a), moves during scanning. The change of the maximum intensity position as a function of the Z-axis offset distance were shown by the black line in Fig. 7(c). The experimental results show that the PH first moves away from the center of the microsphere, and then it moves back to the centre. The PH finally disappears in the center of the microsphere at a propagation distance of 9 μm . The corresponding video is shown in the [Visualization 2](#) in the Supplementary Materials. We also fabricated patchy microspheres for the generation of classical PHs. The patchy microsphere was covered with one piece of Ag film and observed with the same setup. We found that during the scanning process, the maximum intensity position of the light field gradually moved from the off-axis position to the center of the microsphere ([Visualization 3](#)). The trajectory of the bright spot movement during scanning is consistent with the shape of a classical PH [21]. To compare the experimental and simulation results, we retrieved the

trajectories of the maximum intensity points from the simulation results in Figs. 6(a2)-(c2). We found that the shape of the measured PHs is close to the shape of the simulated PHs. It is interesting to see that the experimentally obtained S-shaped PH has a one-sided shape and a larger curvature than the simulation results. We believe this discrepancy can be attributed to the differences between the experimental and simulation parameters. For example, the illumination source in simulations is monochromatic plane waves while that in experiments has a bandwidth. Additionally, the Ag film has a uniform 100 nm thickness in modeling, but in experiments, the Ag film is 30 nm thick and its thickness varies on different areas of microspheres.

4. Conclusions

In this work, we theoretically and experimentally investigated the light focusing properties of patchy particles. By properly designing the desired Ag films, PHs and S-shaped PHs can be generated from patchy microcylinders or patchy microspheres. Compared with classical PHs, the S-shaped PHs have a longer effective length and a narrower beam waist in the far-field region. The FDTD simulation shows that in this work both the excitation of SPP waves and the asymmetry nature of patchy particles contribute to the formation of curved light beams. Experiments were also carried out with patchy microspheres to prove the presence of PHs and S-shaped PHs. This new type of curved photonic flow may be beneficial to various applications, such as super-resolution imaging, micromachining, optical trapping, etc.

Funding. National Natural Science Foundation of China (61905115, 62105151, 62105156, 62175109, U21B2033); National Key Scientific Instrument and Equipment Development Projects of China (62227818); Leading Technology of Jiangsu Basic Research Plan (BK20192003); Outstanding Youth Foundation of Jiangsu Province of China (BK20190445, BK20210338); Biomedical Competition Foundation of Jiangsu Province (BE2022847); Key National Industrial Technology Cooperation Foundation of Jiangsu Province (BZ2022039); Fundamental Research Funds for the Central Universities (30920032101); Open Research Fund of Jiangsu Key Laboratory of Spectral Imaging and Intelligence Sense (JSGP202105, JSGP202201); European Regional Development Fund (SPARCII c81133); Royal Society (IEC\NSFC\181378, IEC\R2\202178); Fonds De La Recherche Scientifique - FNRS (T.0126.22, U.N027.18).

Disclosures. The authors declare no conflicts of interest.

Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

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