Miniaturization of optical structures makes it possible to control light at the nanoscale, but on the other hand it imposes a challenge of accurately handling numerous unit elements in a miniaturized device with aperiodic and random arrangements. Here, we report both the new analytical model and experimental demonstration of the photon sieves with ultrahigh-capacity of subwavelength holes (over 34 thousands) arranged in two different structural orders of randomness and aperiodicity. The random photon sieve produces a uniform optical hologram with high diffraction efficiency and free from twin images that are usually seen in conventional holography, while the aperiodic photon sieve manifests sub-diffraction-limit focusing in air. A hybrid approach is developed to make the design of random and aperiodic photon sieve viable for high-accuracy control of the amplitude, phase and polarization of visible light. The polarization independence of the photon sieve will also greatly benefit its applications in optical imaging and spectroscopy.
Nanotechnology has provided a strong support for realizing the goal of controlling light at the nanoscale by using various nanostructures. This enabled the emergence of new physical phenomena, such as surface-enhanced Raman scattering, extraordinary optical transmission, and surface plasmons optics. However, miniaturization of structures gives rise to an intractable problem particularly for controlling visible light with devices composed of enormous unit elements, that is, realization of superfocusing in air by beating the diffraction limit beyond the evanescent range. Creation of a small focusing spot necessitates high spatial frequency to be carried by the transmitted beam, which imposes inseparable conditions for a large array size and a huge number of unit elements. Recently, devices with aperiodic or randomly distributed nanostructures have drawn great attention owing to their novel properties, such as absorption-enhancing proximity effect and near-field manipulation of light. Nanophotonic devices can acquire rich degrees of freedom (that is, spatial position and geometric size of the nanostructure) from structural aperiodicity and randomness to realize complex functionalities, which are not achievable through periodic or quasi-periodic features with limited control in geometry. However, the physics of designing such a kind of aperiodic or random device possessing a large capacity of both position-size-varying unit structures are yet to be explicitly addressed upon technical challenges.

The first challenge comes from the lack of an effective strategy to determine the geometry positions and sizes of individual holes, in particular for the aperiodic and random cases. The second challenge lies in the unavailability of an appropriate method for direct modelling of the overall device performance oriented by specific objectives such as holography-based beam shaping with high uniformity and superfocusing, which require highly accurate information about the optical field diffracted from an individual nanostructure. One approach reported recently was to approximate the optical performance (that is, amplitude and phase of diffracting light) of a single nanostructure by using that of its diffracting optical field in the whole space by FDTD. This hybrid approach greatly improves the efficiency in optimizing the design in terms of arbitrary distribution, size variation and boundless number.

To overcome these challenges, we propose a hybrid approach that strategically combines the coupled-mode theory and the multipole expansion method to describe the diffracted field from a huge number of subwavelength holes with high accuracy. It gains physical insight into the diffraction issue in terms of elementary mathematical functions, and it makes the optimization algorithms feasible in the structural design of a photon sieve aiming for high-quality holograms and superfocusing.

Results
Hybrid approach. Although the vectorial Rayleigh–Sommerfeld diffraction and angular spectrum theory have been proposed to carry out the simulation of diffraction field at a targeted plane of photon sieves, lack of essential capacity in describing the polarization and intensity profiles of electric field immediately after a single nanohole makes them behave inaccurately for far-field diffraction. Succeeding in it as a powerful tool, finite-difference time domain (FDTD) demands tremendous computing resources to create ultrafine grids so as to accurately calculate the vector field in overall space, not to mention the huge degree of uncertainties during the optimization of such a complex photon sieve.

To alleviate this issue, we propose a twofold method: (1) a hybrid approach combining the coupled-mode theory and multipole expansion method is proposed to accurately model the optical field diffracted from a single subwavelength hole, as shown in Fig. 1a, which is formulated by the superimposing elementary mathematical functions (see equation (3) in the Methods section); (2) the far-field diffracted field of the entire photon sieve sketched in Fig. 1b is obtained by coherently superimposing optical fields diffracted from each subwavelength hole (see equation (4)). A comparison between our method and FDTD for calculating a relatively small photon sieve has been carried out with good agreement, as detailed in the Supplementary Figs 1–4 and Supplementary Notes 1–4. It verifies the effectiveness and reliability of this hybrid approach. In our method, the size parameter (that is, radius $a$) and position information of the subwavelength hole are taken into account to get its diffraction characteristics by calculating the electric field of interest only, which is free from the requirement of simulating the electric field in the whole space by FDTD. This hybrid approach greatly improves the efficiency in optimizing the design in terms of arbitrary distribution, size variation and boundless number.

Design principle. Genetic algorithm inspired by biological evolution is used to implement the design, because it can be easily combined with other algorithms such as particle swarm optimization, cuckoo search and ant colony optimization owing to their common inheritance on old generation. The basic configuration containing key operations of pair, cross-over and mutation are schematically shown in Fig. 1c. After the standard genetic algorithm, we encode the photon sieve information into one chromosome containing $N$ genes, each of which is labelled by the hole size $(d_n)$ and position $(p_n)$, as shown in Fig. 1d. The evolution operation of every chromosome can thus be carried out on the basis of the hybrid approach. In practice, the hole size is constrained to a certain range owing to fabrication issues, which implies that we only need to calculate the electric field of light diffracted from some limited-geometry holes before the evolution operation. Next, we superimpose the known electric field of every hole by addressing its size. It significantly expedites an evolution operation by four orders of magnitude (see Supplementary Fig. 5 and Supplementary Notes 5–6), which makes it more feasible in carrying out the design of a large-scale photon sieve.

Optical hologram. A randomly distributed photon sieve to realize high-uniformity optical holography is schematically shown in Fig. 2a. The design of the photon sieve is implemented by a modified genetic algorithm with mutation and evolution operations only (see Supplementary Fig. 5 and Supplementary Note 6). The photon sieve was fabricated with 34,034 holes of 150 nm radius randomly distributed in a 100-nm-thick chromium film via electron-beam lithography (ELS-7000, Elionix). The scanning
electron microscope images, as depicted in the inset, clearly show randomness with the minimum center-to-center distance of 450 nm between two neighbouring holes, which demonstrates an improvement by nearly 200 times in spatial resolution compared to the similar halftone-based hologram with the smallest pixel pitch of 80 μm.\(^{29–31}\)

We experimentally verified the performance of this photon sieve by normally projecting a linearly polarized 532-nm-wavelength light on it and collecting the transmitted light with a 10 magnification objective lens. The intensity was directly recorded by a charge-coupled device camera, as shown in Fig. 2b. A bird pattern was identically reconstructed by the simulated and measured intensity profiles, as shown in Fig. 2c with excellent agreement. The intensity uniformity is defined as

\[
\sigma_{\text{r.m.s.}} = \left[ \frac{\langle (I(x,y) - I_{\text{avg}})^2 \rangle}{I_{\text{avg}}} \right]^{1/2},
\]

where \(I(x,y)\) denotes the intensity inside the bird pattern area, \(I_{\text{avg}} = \langle I(x,y) \rangle\) and \(\langle \cdot \rangle\) stands for the average intensity and the average operator, respectively. The simulated \(\sigma_{\text{r.m.s.}}\) equals 7%, which is so small that it even behaves better than traditional holography for beam shaping (for example, \(\sim 11%\) in ref. 32). As shown in Fig. 2d, the measured \(\sigma_{\text{r.m.s.}}\) shows a uniformity of \(\sim 17%\), which might attribute to the imperfection (for example, irregularly circular shape, the hole’s in-wall not perpendicular to the exit surface) of fabricated holes. The dependence of the uniformity on the incident wavelength (ranging from 460 to 580 nm) has been simulated. Our results (not shown here) demonstrate that the amplitude uniformity varies within less than 1% in the aforementioned wavelength range.

We have also investigated the diffraction efficiency formulated by

\[
\eta = \frac{I_t}{I_0},
\]

where \(I_t\) is the total intensity of the bird pattern and \(I_0\) is the transmitted intensity. Both of the simulated and measured results have an approximately equal value of \(\sim 47%\), as shown in Fig. 2d. This means that nearly half of the transmitted intensity contributes to the reconstruction of the image. It is a significant advantage over metasurface holograms with polarization dependence\(^{12,13}\), because half of their transmitted light is filtered out by a polarizer. That, in fact, leads to much lower diffraction efficiency (below 20% in refs 12–13,32–35). Owing to the rotational symmetry of the circular hole, our photon sieve hologram exhibits overall polarization independence to account for high diffraction efficiency. Plotted in Fig. 2e are the experimentally captured images of nearly identical intensity profiles when varying the polarization orientation of incident light at 0°, 45° and 90° (angle between the x axis and polarization), respectively.

Beyond these, our photon sieve hologram is free from problematic twin image and high-order diffraction that are present in nanostructured\(^{33,34}\), nanotube\(^{35}\) and nanoparticle-based\(^{36}\) holograms. Traditionally, the twin image also appears for spatial light modulators, and it has been the subject of a long-standing debate\(^{37–40}\) since holography was invented in 1948 ref. 41. Our results suggest that the twin image could be eliminated when the size of the elementary diffraction unit (DU) of the hologram is chosen to be smaller than the wavelength, as demonstrated in refs 12,13. In our case, this DU is a 150-nm-radius hole purposely made at the subwavelength scale, whereas...
the elementary DU (consisting of several pixel pitches) of those nanostructured holograms$^{33,34}$ is larger than the wavelength. For nanotube$^{35}$ and nanoparticle-based$^{36}$ holograms with twin images, their effective DUs with a wavelength-scale size can be taken as a combination of two horizontally or vertically pixel pitches, because two neighbouring pixel pitches share the same phase or amplitude in the hologram design. In their design processes, the retrieval of phase or amplitude in a hologram is based on the feedback of optical inverse fast Fourier transform of target intensity, which has no capacity to distinguish the high spatial frequency embedded in two subwavelength pixels (in connection with evanescent wave), resulting in an effective DU larger than the wavelength. To avoid this in designing the photon sieve hologram, we used the genetic algorithm based on our hybrid approach and made an exemplary attempt by randomly addressing the isolated nanoholes in hologram without referring to the amplitude or phase feedback$^{35,36}$, which has followed the proposal by Gerchberg and Saxton$^{42}$.

**Superfocusing.** Far-field super-resolution focusing under visible light has been demonstrated with periodically or quasi-periodically structured lenses$^{9,10}$. The requirements on a large amount of nanostructures and accurate manipulation of light beyond the evanescent region are challenging in simulation and design optimization. As a result, most approaches adopt the lens with its nanostructures arranged in period or quasi-period distribution having limited manipulation of light.

To obtain a circular focusing spot, we propose an optical nanosieve with circular symmetry as sketched in Fig. 3a, composed of 7,240 subwavelength holes located at 22 concentric rings with a radius of $R_n$ ($n$ is the index of rings with an ascending order from inner to outer). These rings are non-periodically distributed in radial position, while the holes in each ring are uniformly allocated and equally sized with a radius of $a_n$. When $n$ increases, $a_n$ reduces from 150 nm ($n = 1$) to 50 nm ($n = 22$), while $R_n$ rises from 10.85 to 19.65 μm. Hence, the overall structure exhibits aperiodic behaviour in $a_n$ and $R_n$ which allows it to sieve light with appropriate amplitude and phase for achieving a small spot$^{43}$. The central region blocks the direct transmission of light carrying low spatial frequencies while only the component of high spatial frequencies passes through to constructively interfere, accounting for a sharp focusing$^7$. The correlation between $a_n$ and $R_n$ can be encoded by the hole diameter $d_n$ and the interval $t_n$ between adjacent rings. The parameters $d_n$ and $t_n$ are used to construct the chromosome information, which could be systematically optimized (see Supplementary Fig. 7 and Supplementary Note 7) through the standard genetic algorithm to reach a sub-diffraction-limit focusing spot.

Figure 3b depicts the measurement setup with an illumination wavelength of 632.8 nm, and the inset shows the scanning electron microscope image of a part of the fabricated aperiodic photon sieve (the structural dimensions and fabrication process are provided in detail in Supplementary Table 1 and Supplementary Notes 8 and 9). Figure 3c shows the
experimentally scanned intensity pattern at the focal plane in an area of $1.2 \times 1.2 \mu m$ with a step size of $6 nm$. A sub-diffraction-limit focal spot with about $200$-nm FWHM ($\approx 0.32\lambda$) is formed at a distance of $z = 13.3 \mu m$ ($\approx 21\lambda$ at $\lambda = 632.8 nm$) together with an annular side lobe. Figure 3d correspondingly plots the theoretical intensity profile of transversely polarized field $E_{||}$. The agreement between the experimental spot and theoretical $E_{||}$ indicates that the recorded focal spot is transversely polarized, while the longitudinal component $E_z$ predicted in Fig. 3e becomes experimentally undetectable. Furthermore, we plot the normalized cross-sectional profiles of experimental and theoretical results in Fig. 3f without any fitting, and both match with each other with a small discrepancy of amplitude and lateral position, which can be attributed to the cumulative error of the three-dimensional stage and the imperfect alignment between photon sieve and objective lens.

The experimental absence of a longitudinal component $E_z$ is indeed an interesting issue. The simulated $E_z$ has a two-lobe transverse intensity profile, which is normalized to the on-axis intensity of $E_{||}$. From their scale bars, the maximum intensity of two bright spots under $E_z$ is twice that of the spot under $E_{||}$. Upon collection of an objective lens with a magnification of $M$, the intensity of optical field $E_z$ undergoes an $M$-fold reduction in amplitude to become $E_z/M$ at the imaging plane, as shown in Fig. 3g. As a consequence, the amplitude of $E_z$ at the image plane has the order of $1/M^2$ (for example, $10^{-4}$ in our experiment with $M = 100$) of the actual $E_z$ at the objective plane. However, the transverse component of the electric field has no significant attenuation by the objective lens. That is why $E_z$ is usually negligible at the image plane of conventional optical imaging systems. In fact, it is difficult to measure the $E_z$ component, which requires special techniques, such as utilizing tip-enhanced second-harmonic generation in scanning near-field optical microscopy or using surface-enhanced Raman scattering.

It is important to emphasize that our FWHM is normalized to the wavelength in the same ambience (that is, air) throughout, instead of normalizing the FWHM observed in dielectric ambience (for example, oil) by the wavelength in air. In fact, the experimentally achieved $0.32\lambda$ spot is much smaller than the super-oscillatory criterion ($r_s = 0.38\lambda$/numerical aperture (NA))\textsuperscript{43}, validating the physics of design to be in line with the super-oscillation phenomenon. This super-oscillatory spot requires the precise control of coherent interference of light from photon sieves, which means that our lens is more sensitive...
to chromatic aberration than traditional spherical lenses or other nanostructured lenses\textsuperscript{14–17} that display larger spots. Although our photon sieve lens also suffers from low efficiency as other nanostructured lenses\textsuperscript{14–17}, this technique is able to offer a direct and non-invasive way to realize super-resolution imaging without evanescent wave by incorporating it with scanning confocal microscopy\textsuperscript{37}. It is worth commenting here that although in this reference the E-field at the focusing spot has an important longitudinal component, as in our case, the good imaging properties of the lens are preserved. Notice also that our approach has the advantage of sieving a single mode, as holes are used as DUs and not rings, as used in ref. 47. Our approach could also be designed to use various sophisticated optical sieving devices with randomness, and the currently reported lens is just one example showing the superfocusing performance. In addition, the proposed high-NA photon sieve with purely subwavelength-size units gets rid of the broken symmetry of the focusing spot, which usually occurs in subwavelength zone plates\textsuperscript{16}. With its direct sub-diffraction-limited focusing beyond the evanescent region in air ambient, ultrathin planar characteristics\textsuperscript{48} and polarization insensitivity, the photon sieve high-NA lens is promising to integrate with conventional imaging and focusing systems for optical micro-nanoscopy and nanolithography application.

Discussion

In summary, we have experimentally demonstrated the accurate manipulation of light beyond the evanescent region by using a random photon sieve to realize a uniform, twin-image free and high diffraction-efficiency hologram and a non-periodic photon sieve to achieve high-NA lens is promising to integrate with conventional methods in integrated photonics and beam steering with arbitrary distribution.

Experimental setup. The experimental measurement of the photon sieve hologram and superfocusing are carried out in a photon scanning tunneling microscope (Alpha 300S, WITec GmbH) with conical configuration, as shown in Figs 2b and 3b. A laser light with linear polarization is coupled to a polarization-scope (Alpha 300S, WITec GmbH) with confocal configuration, as shown in Figs 2b and 3b. A laser light with linear polarization is coupled to a polarization-scope (Alpha 300S, WITec GmbH) with confocal configuration. The objective lens can image the bird pattern onto a charge-coupled device (CCD) after the two-dimensional stage moved and controlled by a computer. The correlation between the three-dimensional stage position and the detected light signal allows us to reconstruct two-dimensional image of the diffraction field of the photon sieve. Far-field diffraction of nanosieve. Subsequently, by using the coherent superposition of the diffracting field from every hole, we can get the far-field diffraction of the nanosieve

$$E_{\text{far}}(r') = \sum_{j=1}^{N} E_{\text{hil}}(r-j r_0),$$

where $r'$ is the position of the $j$-th hole in a nanohole array and $N$ is the total number of holes. We have to emphasize that the hole number $N$ can be infinite large according to practical requirement. Equation (4) is valid for the hole array with arbitrary distribution.

References

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