nature portfolio

Peer Review File

High-order diffraction for optical superfocusing

Editorial note: Parts of this Peer Review File have been redacted as indicated to remove third-party material where no permission to publish could be obtained.

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The manuscript presents the design and characterization of a diffractive zone plate. The authors claim that the zone plate achieves "superfocusing" because the full-width half maximum of the observed focal spot is less than. In short, the manuscript presents the design of a flat lens with very low efficiency and no particular new features, and its claims seem to stem from misunderstandings about how flat lenses operate, as a result, I do not recommend publishing its publication.

Zone plates have been known for a very long time and using their higher order diffraction is not new. The main issue with using them is their low efficiency. Unfortunately, the manuscript presents the topic as if it is a new idea/concept and discusses it in such a way that can be considered misleading. The goal of designing a flat lens (either diffractive, metalens, etc) is to direct a significant amount of the power into the specific diffraction orders and achieve high efficiency. This has been done by high NA metalenses which achieve high efficiency. Metalenses do not use first-order diffraction as the authors suggest, but multiple meta-atoms form larger supercells that efficiently deflect light into high orders. If efficiency was not an issue, there was no need for using small structures and meta-atoms, one could just use a zone plate with a few percent efficiency (as is done by the authors).

The claim of "superfocusing" is also a result of an incorrect assumption that the focal spot shape of high NA metalenses is an Airy disk with an FWHM of ~0.515λ/NA, and thus anything smaller is "superfocusing". This is an incorrect assumption. The focal spot shapes of high NA flat lenses deviate significantly from an Airy disk and its FWHM is also smaller than 0.5λ (see for example "A. McClung, M. Mansouree, S. Samudrala, and A. Arbabi, "Properties of Ideal Flat Metalenses," CLEO 2020, paper FM1R.1.")

In addition, the confocal microscopy and ablation experiments do not show any higher resolution than other flat lenses (e.g., metalenses). Confocal microscopy already has a higher resolution even for lenses with Airy spot (see for example https://www.olympus-lifescience.com/en/microscope-resource/primer/techniques/confocal/resolutionintro/), and ablation is a nonlinear process and much higher resolutions are already demonstrated without “superfocusing” (see Joglekar AP, Liu HH, Meyhöfer E, Mourou G, Hunt AJ. Optics at critical intensity: applications to nanomorphing. PNAS, 101(16), pp.5856-5861).

Reviewer #2 (Remarks to the Author):

Planar lenses, based on micro-/nano-structures, are usually influenced by the diffraction phenomenon, mostly the size of focal spot that is concerned. This paper takes the interest in the high-order diffraction (HOD), not only giving its analytical formula which correlates the diffraction order and the width of each transmissive zone, but also proposing two new lens samples by manipulating HOD independently and showing their potentials in the imaging beyond diffraction limit and laser direct writing lithography. Besides, the zones consisting the HOD lens are of non-subwavelength widths, ensuring the low-cost fabrication. This work is interesting and focuses a new research theme of micro-/nano-structure-based planar lenses. However, the following comments still need to be addressed.

1. As the authors mentioned, the higher order of the diffraction, the less efficiency it behaves. How to consider this restriction, especially for the engineering applications?
2. How to obtain the Fig.1b? More explanations should be given.
3. Eq.(1) analytically correlates the dimension of each zone with the diffraction order. Does it consider the polarization state of the incident light? Will it influence the ultimate optical field achieved? As shown in the experimental sections, only the circularly polarized illumination was employed.
4. Comparing Fig.1e with Fig.1d, the HOD FZP gives a much more diverging optical field than the standard FZP. It causes lots of sidelobes around the central focal spot, as shown in Fig.2c with both simulation and experimental testing. Is it possible to completely eliminate these sidelobes? If not, how they will affect the realistic applications?
5. In Fig.2e, how to define the longitudinal distance of 10.86 μm? The intensity value of 0.5 should be also labelled.
6. “The measured optical needle has a lateral FWHM of approximatly 180 nm, …”, in this sentence, “approximatly” has an “e” missing. In the same sentence, “Rayleigh criterion”

with a short name of RC and “super-oscillation criterion” with a short name of SOC should be given.

7. “Theoretically, the longer distance is also possible if more rings are used in the phase mask…”, is this point for the same focal length? In that case, more rings will result in a larger size of the lens, which increases the NA. Normally, as the NA increases, DOF will decrease accordingly. So, more details or examples are needed to support this conclusion.

8. “More complex nano-objects can also be imaged with high resolution by using our SCM.” This conclusion needs to be clarified more clearly. Are these nano-objects planar or three-dimensional, transparent or semi-transparent or opaque, etc?

9. “sub-diffraction-limit focusing”, “subdiffraction-limited focusing”, it should be consistent in the whole paper. Also in the supplementary material, “1st-order lens” and “1-order lens” should be consistent.

10. Both developed lens samples have the NA of 0.9. How will other values of NA influence the HOD performance?

11. In the “METHODS” section, “when the diffraction order M is small or the focal length f is large, which means that the second term in the right part of Eq. (1) can be ignored due to relatively small value (compared with the first item)”, more details are needed to provide the evidence to support this conclusion, for example, how small of M and how large of f? It is important to use the proposed method for some practical applications.

12. Considering the laser-ablation lithography, more details to discuss the influences of the uniformity of the achieved long focusing needle on the fabrication results are needed.

13. In “Supplementary Figure 4”, “The heights and widths of the slits are 2 μm and 50 nm, respectively.” should be “The lengths and widths of the slits are 2 μm and 50 nm, respectively.”

14. In “Supplementary Section 6”, “…indicates that the experimental longitudinal FWHM can reach 20 μm” should be “… indicates that the experimental longitudinal DOF can reach 20 μm”.

Reviewer #3 (Remarks to the Author):

The authors proposed independent manipulation of unexploited high-order diffraction through an analytical formula that correlates the diffraction order and the width of each zone. The large spatial frequencies enabled the proposed lenses to reduce the lateral focal size down to 0.44λ (NA=0.9). They experimentally demonstrated high-order lens-based confocal imaging with a center-to-center dry resolution of 190 nm, and laser-ablation lithography with achieved direct-writing resolution of 400 nm (0.385λ). The authors demonstrated the applications of the proposed diffractive lenses, and the data in the manuscript are convincing. However, there are some concerns that must be carefully addressed before the paper is fit for publication.

1. On page 3 of the main text, the authors claimed that "We note that the additional phase also modulates other undersigned orders but does not influence optical focusing at the designed order M. Thus, by using the phase delay of each zone, we can realize the independent manipulation of the phase at the expected order." How do the authors modulate the optical focusing at the designed order M by designing additional phase, and what effect does this have on other orders?
2. As can be seen from Figs 1d and 1e, such a design will bring obvious interference to the diffraction characteristics of the device, greatly reducing the focusing efficiency of FZP. Are there other ways to achieve optical superfocusing? The authors should discuss them.
3. In applying the designed device to laser-ablation-based lithography, do the authors utilize only the higher-order part? How do they filter out the rest of the orders?
4. The efficiency of the proposed device appeared to be much lower than those listed in Supplementary Table 1, which is detrimental to its practical application.
5. The focal length designed by the author is 1 mm. Can the focal length of such diffractive elements be set relatively small? Like tens or hundreds of microns, how does that affect their performance?

Comments from the reviewers and our point-by-point replies

Contents

Reviewers' comments:

Comments from Reviewer #1

1. The manuscript presents the design and characterization of a diffractive zone plate. The authors claim that the zone plate achieves "superfocusing" because the full-width half maximum of the observed focal spot is less than. In short, the manuscript presents the design of a flat lens with very low efficiency and no particular new features, and its claims seem to stem from misunderstandings about how flat lenses operate, as a result, I do not recommend publishing its publication.

Reply: We thank the referee for the critical comments on our work, which allow us to re-check and reaffirm the novelty of our manuscript carefully. We hope that the reviewer can re-evaluate our work again.

For the "super-focusing" issue, we find that the suggested CLEO conference paper might demonstrate *incorrect* conclusion about the simulated focal spot of an ideal metalens. For your better reference, we show the proofs and discussions in the following reply section 4.

According to our careful review of literatures, we reaffirm that our work is the first to demonstrate subdiffraction-limit focusing with high-order diffraction (the evidence is provided in the following reply section 5).

The "low efficiency" is an intrinsic property for high-order diffraction, which has been discussed in our original manuscript. The low-efficiency zone plate are still useful in X-ray optics and other application scenarios (please refer to the following reply section 2).

Our flat lens has new features of subdiffraction-limit focusing with large zone width and millimeter-scale focal length, which enable a scanning imaging resolution of 190 nm, the highest one for reported confocal microscopies in the visible(see the reply section 5). Note that, the imaging resolution 0.4λ/NA in the suggested Olympus website is just a theoretical value, which, to the best of our knowledge, have never been demonstrated experimentally.

Our team have been working on zone-plate-based flat lenses for subdiffraction-limit focusing since 2014 and published a series of papers on the topic, *e.g.*, Nat. Commun. 6, 7059(2015), Nat. Commun. 14, 5838(2023), Adv. Mater. 29, 201602721(2017), Adv. Mater. 30, 1704556(2018), Laser Photon. Rev., 8, 152(2014), Light 10, 22(2021), Nano Lett. 20, 7972(2020), Sci. Rep. 5, 09977(2015). In an invited review paper (Adv. Mater. 30, 1704556(2018)), we have provided a comprehensive introduction about working principles and applications of planar diffractive lenses. If there are any "misunderstandings about how flat lenses operate" in either our previous works or current manuscript, we are very happy to discuss in details at any time.

Finally, we appreciate your instructive comments again, which helped us to polish our manuscript and doubly confirm the novelty of our work. Hope that our careful revision and detailed replies can help us to win your support.

2. Zone plates have been known for a very long time and using their higher order diffraction is not new. The main issue with using them is their low efficiency. Unfortunately, the manuscript presents the topic as if it is a new idea/concept and discusses it in such a way that can be considered misleading.

Reply: Thank you for the comment. It is right that zone plates have been known for a very long time. However, our work is the first to realize subdiffraction-limit focusing with high-order diffraction. Although high order diffraction in zone plates can be used to enhance imaging resolution in X-ray (Phys. Rev. Lett. 103, 110801(2009)), the demonstrated imaging results are still *diffraction-limited*. So, it is novel to demonstrate super-focusing with high-order diffraction.

Even though the zone plates have low efficiency, they are the only choice for focusing or imaging at short-wavelength spectra, such as soft X-ray, where the transmission elements are not available. So, the zone plates are still useful, besides in other areas where they can play unique roles.

To avoid any misleading, in the abstract we re-organize the claim as "Here we report independent manipulation of HODs, *which are unexploited for subdiffraction-limit focusing in diffractive lenses*, through…".

Furthermore, in our revised manuscript, we have cited the relative references that reported high-order diffraction zone plates for diffraction-limited focusing or imaging, and give a detailed discussion about high-order diffraction in diffractive lenses, as shown below:

"*For soft X-ray, the HODs of zone plates have been used to enhance imaging resolution*[22]*, which, however, is still diffraction-limited due to the lack of independent manipulation of HODs.* "

3. The goal of designing a flat lens (either diffractive, metalens, etc) is to direct a significant amount of the power into the specific diffraction orders and achieve high efficiency. This has been done by high NA metalenses which achieve high efficiency. Metalenses do not use first-order diffraction as the authors suggest, but multiple meta-atoms form larger supercells that efficiently deflect light into high orders. If efficiency was not an issue, there was no need for using small structures and meta-atoms, one could just use a zone plate with a few percent efficiency (as is done by the authors).

Reply: Thank the reviewer for the comments on diffractive lens and metals. We agree that metalenses with multiple meta-atoms in supercells can efficiently direct light into specific diffraction orders.

Efficiency is indeed important but it could be compromised at some situations. For example, in soft X-ray, almost all materials are absorptive so that transparent elements or high-efficiency meta-atoms are not available, which makes the low-efficiency zone plates the choices in various X-ray applications. In this case, realizing the focusing purpose is more important than the low efficiency for X-ray optics. Furthermore, in other spectra such as visible frequencies, if one wants to realize a high-NA (>0.9) lens with a ultralong focal length of centimeter scale, our proposed high-order lenses with non-subwavelength feature will be a good candidate due to its easy fabrication by using laser direct-writing lithography. In contrast, to realize the same purpose, both the traditional objectives and metalenses are facing extremely challenging fabrication issues.

The current status of zone-plate in X-ray is that, the focusing and imaging resolution is nearly equal to the smallest feature size of the zone plates. Constrained by the fabrication technology, the smallest feature is usually 10 nm ~20 nm, which leads to the limited imaging and focusing resolution in X-ray. A natural question arises: *whether the imaging and focusing resolution can be increased further when the smallest feature of zone plates is fixed*? The significance of this work is that it confirms one still can focus light into a subdiffraction-limit spot with the help of high-order diffraction even if the smallest feature size of zone plate is fixed.

4. The claim of "superfocusing" is also a result of an incorrect assumption that the focal spot shape of high NA metalenses is an Airy disk with an FWHM of ~0.515λ/NA, and thus anything smaller is "superfocusing". This is an incorrect assumption. The focal spot shapes of high NA flat lenses deviate significantly from an Airy disk and its FWHM is also smaller than 0.5λ (see for example "A. McClung, M. Mansouree, S. Samudrala, and A. Arbabi, "Properties of Ideal Flat Metalenses," CLEO 2020, paper FM1R.1.")

Reply: It has been widely accepted that focusing light into a spot with an FWHM smaller than $0.515\lambda/NA$ (the size of an Airy spot) is taken as "subdiffraction-limit focusing" or "super-focusing"[R1,2]. In this manuscript, we use "super-focusing" for the shorter title and "subdiffraction-limit focusing" in the main text.

The referee might be misled by the incorrect conclusion in the CLEO paper[R3]. In fact, an ideal high-NA metalens CANNOT realize a focal spot smaller than an Airy spot. The mistakes in the CLEO paper [R3] are threefold: 1) Wrong apodization factor $a(\theta)$; 2) Unsuitable diffraction theory used in calculating the focal field of a focused spherical wave; 3) Inconsistences between their reported results (in the CLEO paper [R3]) and other experimental results. The detailed proofs are shown below:

1) Wrong apodization factor $a(\theta)$

[figure redacted]

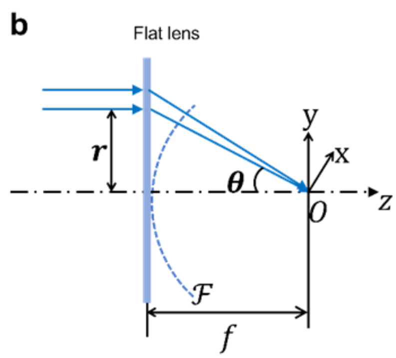

Fig. R1 (a) Unreliable schematic ray diagram of a flat lens in Ref.[R3]. (b) The correct schematic ray diagram of a flat lens.

In Ref. [R3], a collimated narrow beam is incorrectly shaped into another collimated beam after the ideal flat lens, see Fig. R1(a), which yields the relationship $|E_2|=|E_1|\cos\theta^{-1/2}$ (the refractive indices $n_1=n_2$). The ideal metalens with the phase profile of $\exp[-2\pi if/(n_2\lambda_0\cos\theta)]$ will yield a convergent spherical wave instead of a plane wave as sketched in Fig. 1a in Ref. [R3]. In fact, after the flat lens, the transmitted light should be shaped into a convergent spherical wave, see Fig. R1(b). For a diffractive flat lens, the radial position of flat lens r and the focal length of f meet the Helmholtz condition [R4-6]:

$$r/f=\tan\theta, \tag{R1}$$

where θ is the convergence angle in Fig. R1(b). Eq. (1) denotes the Helmholtz condition[R4-6], which is valid for all flat metalenses. According to law of energy conservation, we can obtain the relationship between the amplitude $|E_1|$ of incident light and $|E_2|$ of converged spherical wave[R4-6]:

$$|E_1|^2 2\pi r dr=|E_2|^2 2\pi f\sin\theta f d\theta, \tag{R2}$$

From Eq. (R1) and (R2), one can know that $|E_1|$ and $|E_2|$ should meet: $|E_2|\propto|E_1|\cos\theta^{-3/2}$. Therefore, for an ideal flat lens, the apodization factor $a(\theta)$ is $\cos\theta^{-3/2}$, which is different from $\cos\theta^{-1/2}$ in Ref. [R3].

2) Unsuitable diffraction theory for simulation

Rayleigh-Sommerfeld diffraction[R7,8] or Richard-Wolf theory[R4-6] can be used frequently to simulate the focal electric field of a planar diffractive metalens. But, each method is used under different condition.

For the Rayleigh-Sommerfeld diffraction (equivalent to angular spectrum method), the initial input is the product of the incident electric field and the phase profile of the metalens, without any need of the pre-processed apodization factor a(θ) in Ref. [R3]. But, the authors in Ref. [R3] misused the a(θ) item in their "full-wave propagation", which leads to the incorrect result that "*the simulated focal spots for NA=0.8 and NA=0.95 are much smaller than the Airy spot (see Fig. 1c in Ref.* [R3])". Because the authors have not pointed out the detailed diffraction integral, we take the "full-wave propagation" as the Rayleigh-Sommerfeld diffraction or angular spectrum method.

For Richard-Wolf theory, its initial input must be the wavefront of a convergent spherical wave so that one has to do some mathematics to change the incident plane wave into the convergent wavefront by following the steps of energy conservation in Ref. [R4-6]. In this process, it leads to an apodization factor a(θ), which will be used in the calculation of Richard-Wolf integral. Note that, in Richard-Wolf theory, the spherical phase profile of a metalens or an objective has been taken as the mathematical process of changing the incident plane wave into the convergent wavefront. As a result, there is no need to involve the phase profile of the metalens or objective in the calculation by using Richard-Wolf theory.

If both methods of Rayleigh-Sommerfeld diffraction and Richard-Wolf theory are used correctly, one can find that both simulated focal spots agree with each other and are also highly consistent with the Airy spot. To show the proofs, we implement optical focusing of an incident circular-polarization (*i.e.*, $E_1 = e_x + ie_y$, which is used to keep the consistence with Ref. [R3]) light via an ideal metalens with uniform amplitude by using Rayleigh-Sommerfeld diffraction and Richard-Wolf theory, respectively.

By using Rayleigh-Sommerfeld diffraction, we have the phase profile $U_0 = e^{ik(f-\sqrt{r_0^2+f^2})}$ for the metalens, where f is the focal length, k is the wave number and $r_0=(x_0^2+y_0^2)^{1/2}$ is the radial coordinate. If the circular-polarization light is illuminated, the vector electric fields of transmitted light can be expressed as

$$E_{2x}(x,y,z) = \frac{-1}{2\pi}\int\int E_{1x}(x_0,y_0)\frac{\partial}{\partial z}\left(\frac{e^{ikR}}{R}\right)dx_0dy_0$$

$$E_{2y}(x,y,z) = \frac{-1}{2\pi}\int\int E_{1y}(x_0,y_0)\frac{\partial}{\partial z}\left(\frac{e^{ikR}}{R}\right)dx_0dy_0 \qquad \text{(R3)}$$

$$E_{2z}(x,y,z) = \frac{1}{2\pi}\int\int\left[E_{1x}(x_0,y_0)\frac{\partial}{\partial x}\left(\frac{e^{ikR}}{R}\right)+E_{1y}(x_0,y_0)\frac{\partial}{\partial y}\left(\frac{e^{ikR}}{R}\right)\right]dx_0dy_0$$

where $E_{1x}= U_0$ and $E_{1y}=i*U_0$. By using Eq. (R3) with the numerical integral (without any pixelated process as the practical metalenses), we can simulate the vector electric fields at the focal plane, as shown by the red curves in Fig. R2(a). The intensity profiles of the transverse electric field have good agreement with the Airy spot for each NA. However, the predicted large Ez component for high NA=0.95 leads to that the total intensity has slightly larger FWHM than the Airy spot. Basically, the simulated results by Rayleigh-Sommerfeld diffraction is highly consistent with the Airy spot.

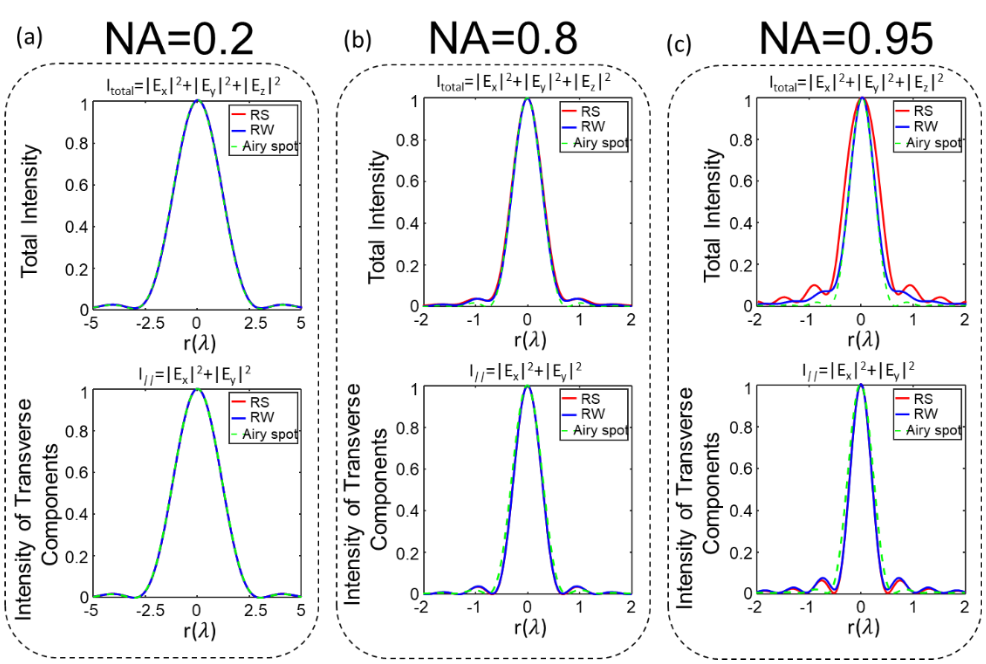

Fig. R2. Simulated intensity profiles of a metalens with different NAs (NA=0.2 in (a), NA=0.8 in (b) and NA=0.5 in (c)) by using Rayleigh-Sommerfeld (RS, labelled in red curves) diffraction and Richard-Wolf (RW, labelled in blue curves) theory. For a better comparison, the intensity profiles are also shown in green dashed curves. The total intensity profiles including all vector components are shown in the upper panel, while the intensity profiles of the transverse components are provided in the lower panel for each case of *NA*. In our simulations, the metalens has a fixed diameter of D=1000λ and a changeable focal length for each NA.

By using the Richard-Wolf diffraction, we only consider the apodization factor $a(\theta)= \cos\theta^{-3/2}$ because the electric field of incident circular-polarization light is uniform. Thus, we have the electric field near the focal plane

$$E = -\frac{\mathrm{i}f\pi}{\lambda}\int_0^{\theta_{max}} cos\theta^{-3/2}[\begin{pmatrix} [(1+cos\theta)\cdot J_0(krsin\theta)+cos2\phi(1-cos\theta)J_2(krsin\theta)]e_x \\ sin2\phi(1-cos\theta)J_2(krsin\theta)e_y \\ 2isin\theta cos\phi J_1(krsin\theta)e_z \end{pmatrix} + i\cdot\begin{pmatrix} sin2\phi(1-cos\theta)J_2(krsin\theta)e_x \\ [(1+cos\theta)J_0(krsin\theta)+cos2\phi(cos\theta-1)J_2(krsin\theta)]e_y \\ 2isin\theta sin\phi J_1(krsin\theta)e_z \end{pmatrix}]\mathrm{e}^{ikzcos\theta}sin\theta d\theta, \quad (R4)$$

where θ_{max} is the maximum convergent angle of the metalens and $\tan\theta_{max}$=D/2*f*. For the metalenses with different NAs, the simulated total and transverse intensity profiles are presented in the blue curves in Fig. R2, and also has good agreement with the Airy spot. More importantly, both simulated spots by Rayleigh-Sommerfeld diffraction and Richard-Wolf integral are nearly the same for different NAs, which verifies that we have implemented the corrected simulations.

Thus, we have to emphasize that, from the above simulations, we have NOT observed the obvious decrease in the spot size of an ideal metalens with NA=0.95. All the simulated spots have the sizes of ~0.5λ/NA. Based on our simulated results, we think that the simulated spots for high-NA cases in Ref. [R3] might be incorrect.

So, the CLOE paper (*i.e.*, Ref. [R3]) cannot be taken as a convincing proof to deny the well-accepted definition of "subdiffraction-limit focusing" in terms of 0.5λ/NA.

3) Inconsistences between their reported results (in the CLEO paper [R3]) and other experimental results

For a metalens with the phase profile of $U_0=e^{ik(f-\sqrt{r_0{}^2+f^2})}$, their experimental results have also been demonstrated in different groups. We list their parameters in the following Table R1. One can find that all the experimentally achieved spots at different NAs are diffraction-limited by 0.515λ/NA (*i.e.*, the FWHM of an Airy spot). Thus, the experimental results in the reported literatures also don't support the conclusion "An ideal metalens can realize the sub-diffraction-limit focusing" in the CLEO paper.

Table R1. Experimental focal spots of the reported metalenses with different NAs.

Reference	Wavelength (nm)	Pixel pitches （λ）	NA	Experimental spot size (FWHM, λ)	The size of an Airy spot: 0.515λ/NA (λ)
Liang et al.[R9]	532	0.4135	0.98, 1.48	0.52, 0.40	0.5255, 0.3480
Chen et al.[R10]	532	0.4511	1.1	0.57	0.4682
Khorsaninejad et al[R11]	405, 532, 660	0.4938,0.6109,0.6516	0.8	0.67, 0.63, 0.73	0.6438
Paniagua-Dominguez et al.[R12]	715	0.3636	0.99	0.54	0.5202
Lin et al.[R13]	500	Not reported	0.43	1.27	1.1977
Wang et al.[R14]	400-600	0.2~0.3	0.106	5.3	4.8585
Arbabi et al.[R15]	1550	0.516	0.97	0.57	0.5309

5. In addition, the confocal microscopy and ablation experiments do not show any higher resolution than other flat lenses (e.g., metalenses). Confocal microscopy already has a higher resolution even for lenses with Airy spot (see for example https://www.olympus-lifescience.com/en/microscope-resource/primer/techniques/confocal/resolutionintro/).

Reply: From our response in section 4, we have confirmed that the metalenses have the diffraction-limited focusing, see the experimental results in Table R1. In contrast, our achieved focal spot in this work has the lateral size of 0.44λ, which is smaller than 0.515λ/NA=0.57λ at NA=0.9. So, our work did show higher resolution than metalenses.

For confocal microscopy, the theoretical resolution is indeed close to $0.515\lambda/\mathrm{NA}/\sqrt{2} \approx 0.4\lambda/\mathrm{NA}$[R16], which, however, is derived from the assumption that the object is infinitesimal (*i.e.*, a point object) and a pinhole in the confocal microscopy is also infinitesimal (*i.e.*, the pinhole has a transmission function of $\delta(x)$). In fact, these assumptions cannot be realized in practice, so that the realistic confocal microscopies by using traditional diffraction-limited objectives and lenses cannot approach the theoretical resolution, which is also pointed out by Stefan Hell in 2007[R1].

More importantly, in our original manuscript we have already demonstrated the imaging resolution (Fig. 3j) of confocal microscopy based on the diffraction-limit objectives, where other optical systems such as pinhole are the same with our flat-lens-based confocal microscopy. The results (Fig. 3k) of our high-order-lens-based confocal microscopy show higher resolution than those in Fig. 3j. The referee might have overlooked our experimental results or misunderstood the detailed status of confocal microscopy.

To show the advantages of imaging resolution for our confocal microscopy, we have summarized all the recently reported confocal microscopies by using flat lenses, including metalenses, super-oscillation lenses (SOLs), and super-critical lenses (SCLs). The comparison is given in Table R2. One can find that our confocal microscopy is the only one to break the 200 nm-limit among all these microscopies at the visible wavelengths.

Table R2. A comparison among far-field label-free confocal microscopies based on flat lenses

Reference	Material	Lens type	Modulation type	Smallest feature size (λ) of lens	λ	Ambient Medium	Working Mode	Working Distance	Center To Center Distance
Rogers et al.[R17]	Al	SOL	Amplitude	0.31	640 nm	oil	Transmission	10.3 μm	315 nm(0.49λ)
Qin et al.[R18]	Cr	SCL	Amplitude	2.96	405 nm	air	Transmission	55 μm	228 nm(0.56λ)
Chen et al.[R19]	TiO_2	Meta lens	Phase	0.45	532 nm	oil	Transmission	125 μm	400 nm(0.75λ)
Yuan et al.[R20]	Gold	SOL	Amplitude	<1	800 nm	air	Transmission	10 μm	320 nm(0.4λ)
Wang et al.[R21]	MoS_2	SCL	Amplitude	0.55	450 nm	air	Transmission	20 μm	200 nm(0.44λ)
He et al. [R22]	SiO_2	SCL	Phase	0.56	405 nm	air	Reflection	1000 μm	200 nm(0.49λ)
Li et al. [R23]	Si_xN_y	SOL	Phase	5.12	488 nm	air	Transmission	428 μm	258 nm (0.53λ)
This work	SiO_2	SCL	Phase	2.72	405 nm	air	Transmission	1000 μm	190 nm(0.46λ)

6. Ablation is a nonlinear process and much higher resolutions are already demonstrated without "superfocusing" (see Joglekar AP, Liu HH, Meyhöfer E, Mourou G, Hunt AJ. Optics at critical intensity: applications to nanomorphing. PNAS, 101(16), pp.5856-5861).

Reply: We appreciate the reviewer for the valuable comment. As demonstrated in this PNAS paper, the deep-subwavelength structures can indeed be fabricated with optical ablation by using ultrafast pulses. However, this phenomenon happens only when deterministic optical breakdown is achieved by carefully tuning the pulse intensity near the threshold [R24], which is not easy for a commonly used laser without any customized functionality. In our case, the pulse energy and duration is fixed without any tuning, so that we cannot observe the deep-subwavelength structures, which, however, will be one of our future works.

Comments from Reviewer #2

Planar lenses, based on micro-/nano-structures, are usually influenced by the diffraction phenomenon, mostly the size of focal spot that is concerned. This paper takes the interest in the high-order diffraction (HOD), not only giving its analytical formula which correlates the diffraction order and the width of each transmissive zone, but also proposing two new lens samples by manipulating HOD independently and showing their potentials in the imaging beyond diffraction limit and laser direct writing lithography. Besides, the zones consisting the HOD lens are of non-subwavelength widths, ensuring the low-cost fabrication. This work is interesting and focuses a new research theme of micro-/nano-structure-based planar lenses. However, the following comments still need to be addressed.

Reply: We thank the reviewer for the insightful comments and strong support to our work.

1. As the authors mentioned, the higher order of the diffraction, the less efficiency it behaves. How to consider this restriction, especially for the engineering applications?

Reply: We thank the reviewer for the valuable comment. The theoretical efficiency is given in Eq. (7), which is a good estimation for the efficiency of a high-order lens. In practical applications, one should consider this restriction from the viewpoint of the entire optical systems, including laser source, optical devices (*i.e.*, high-order lens), and detection. Although the efficiency of the high-order lens is low, high power source or high-sensitivity detector is helpful to compensate the efficiency issue in real applications, such as, our demonstrated confocal microscopy (with high-sensitivity photomultiplier) and laser ablation (using pulse laser with high transient power) can be taken as the proof.

In addition, the thicker zone structures in the high-order lens can be also used to enhance optical efficiency, which has also been proposed and verified (Schneider, Appl. Phys. Lett., 71, 2242(1997)).

As suggested, we have provided the detailed discussion in our revised manuscript and show it in the following for your reference:

"*Low efficiency from HODs is obtained under the assumption of ultra-thin zone structures. In fact, when the height of the zone structures increases, the waveguide effect in the thick zone can be used to enhance the efficiency with the optimized thickness and line-to-space ratio*[57]*. Moreover, the shrunken efficiency from HODs can be compensated by using higher-power laser sources or high-sensitivity detector in practical applications. For example, although the high-order lens in our confocal microscopy has low efficiency, optical signal is detected by using a photomultiplier tube that can record weak optical signals . In the laser-ablation lithography, high transient power of the incident ultrafast pulses can also compensate low efficiency of the high-order lens so that the focused light is sufficiently strong to pattern various structures on the materials.*"

2. How to obtain the Fig.1b? More explanations should be given.

Reply: Figure 1b shows the simulated radial intensity as a function of the ring width Δr (=r_n-r_{n-1}), where the ring has a fixed central radius of 75λ (*i.e.*, (r_n+r_{n-1})/2=75λ). To obtain Fig. 1b, we need to do the following steps:

1) changing Δr to get a new ring;

2) simulating the diffraction pattern of the new ring by using Rayleigh-Sommerfeld diffraction;

3) choosing the simulated radial intensity within the radial range of $0 \leq \rho \leq 75\lambda$ and storing it as one volume of a two-dimensional matrix. The volume refers to the radial intensity and the row refers to the ring width Δr. In Fig. 1b, the ring width Δr is valued from 0.1λ to 5λ with an interval of 0.1λ.

4) repeating steps 1-3 so that the matrix is completely filled.

5) drawing the matrix with a 3-dimensionl view by using a "surf" function in MATLAB.

6) drawing the dashed white lines by using the relationship $M = \lfloor \Delta R/\lambda \rfloor$ (where $\lfloor a \rfloor$ denotes the integer part of a.

3. Eq.(1) analytically correlates the dimension of each zone with the diffraction order. Does it consider the polarization state of the incident light? Will it influence the ultimate optical field achieved? As shown in the experimental sections, only the circularly polarized illumination was employed.

Reply: Thank the reviewer for the valuable comment. Eq. (1) is derived without considering the polarization of incident light. Hence polarization has no influence on Eq. (1), which means that it does not affect the diffraction order of ring.

According to vector Rayleigh Sommerfeld diffraction integral[R25,26], the transverse components of electric field at the focal plane is the same as that of scalar Rayleigh Sommerfeld integral[R27]. This means that the transverse component is independent on the polarization of incident light. For example, the intensity profile of 2nd-order lens (main text) is the same under the illumination of x-polarized light and circularly polarized light (as shown in Fig. R3(a) and R3(d)).

However, the longitudinal component is dependent on the polarization of incident light[R25,26]. Therefore, under the illumination of x-polarized light and circularly polarized light, the 2nd-order lens has the different longitudinal components, as shown in Fig. R3(b) and R3(e).

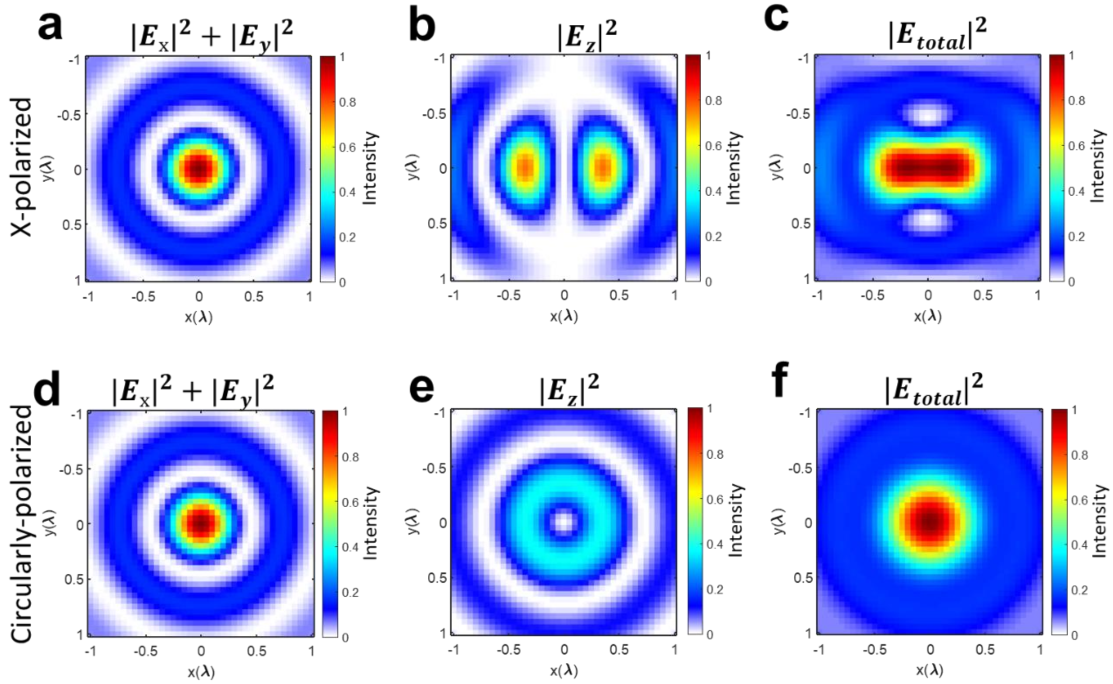

Fig. R3 The focus intensity of transverse and longitudinal component of 2nd-order lens(main text). (a-c)Under the illumination of x-polarized light, the transverse(a), longitudinal(b) component and total(c) intensity of focus. (d-e)Under the illumination of circularly-polarized light, the transverse(ad), longitudinal(e) component and total(f) intensity of focus. The intensity of longitudinal component is normalized by intensity of transverse component.

Although the longitudinal component influences the total focal intensity for a polarized incident light (see Figs. R3c and R3f), it will be filtered by the imaging system with a high magnification objective[R28]. Hence, the measured focal intensity (Fig. 2c in the main text) of the 2nd-order lens is the same as the simulated intensity of the transverse component (Fig. 3a, d). Similarly, in the confocal microscopy (Fig. 3a in the main text), only the transverse component of the focal spot can be detected by the detector PMT. Hence, the resolution of SCM is not influenced by the longitudinal component.

In the laser-ablation-based lithography, this longitudinal component will widen the patterned lines. Under the illumination of circularly polarized light, the focal spot broadens (Fig. R3f), and the line width of the fabricated slit also increases. Consequently, as depicted in Fig. 4c in the main text, the measured widths of the fabricated slits are larger than the theoretical values (obtained by the measured intensity of transverse component shown in Supplementary Figure 5b). In this work, circularly polarized illumination is employed to obtain a focal spot with good circular symmetry (Fig. R3f), facilitating the fabrication of uniform structures.

4. Comparing Fig.1e with Fig.1d, the HOD FZP gives a much more diverging optical field than the standard FZP. It causes lots of sidelobes around the central focal spot, as shown in Fig.2c with both simulation and experimental testing. Is it possible to completely eliminate these sidelobes? If not, how they will affect the realistic applications?

Reply: We thank the reviewer for the comments. With the reduction in the size of the main focal spot, the energy of the main spot will decrease. According to energy conservation, the other energy at the focal plane will be transferred to the sidelobe inevitably[R27]. Therefore, it is hard to eliminate these sidelobes completely for sub-diffraction limit focal spot.

If the peak intensity of sidelobe is so strong that it can be comparable or even larger than the peak intensity of main spot, double image will appear in the images of HOL-based SCM, thus destroying the imaging resolution. In lithography, the strong sidelobes will yield a bad patterning resolution because the materials at the larger area will also be etched. Therefore, the focal spot must be designed carefully for practical applications.

For example, in our design, the peak intensity of the sidelobe does not exceed 20% of the peak intensity of the main spot (see Fig. 2d in main text and Supplementary Figure 5c). So, the sidelobe issue is still acceptable in realistic applications. This can be observed by the clear curves in our demonstrated confocal imaging and the patterned lines in laser-ablation lithography. Hence, after careful design, the sidelobe is not a big issue in real applications.

5. In Fig.2e, how to define the longitudinal distance of 10.86 μm? The intensity value of 0.5 should be also labelled.

Reply: Thank the reviewer for the suggestion. The longitudinal distance of 10.86 μm is defined by the full width at half maximum (FWHM) of the normalized peak intensity. In our revised manuscript, the label 0.5 has been shown in Fig.2e in the main text.

6. "The measured optical needle has a lateral FWHM of approximatly 180 nm, …", in this sentence, "approximatly" has an "e" missing. In the same sentence, "Rayleigh criterion" with a short name of RC and "super-oscillation criterion" with a short name of SOC should be given.

Reply: We thank the reviewer for pointing out the typo and the valuable suggestions. In our revised manuscript, the sentence has been revised as "*The measured optical needle has a lateral FWHM of approximately 180 nm (Fig. 2f), which is smaller than the Rayleigh criterion (RC, calculated by 0.515λ/NA=0.57λ) but larger than the super-oscillation criterion (SOC, calculated by 0.358λ/NA=0.4λ)*".

7. "Theoretically, the longer distance is also possible if more rings are used in the phase mask…", is this point for the same focal length? In that case, more rings will result in a larger size of the lens, which increases the NA. Normally, as the NA increases, DOF will decrease accordingly. So, more details or examples are needed to support this conclusion.

Reply: Thank the reviewer for the valuable suggestion. In our design, the HOL is composed by a HOD FZPs and a phase mask, as shown in Fig. 2a in main text. The radius of HOD FZPs is labelled in r_n and the radius of the phase mask is shown by ρ_j (j=1, 2, …, J). The maximum radius ρ_J of the phase mask is always smaller than the maximum radius r_N of the HOD FZPs. Here, "*more rings are used in the phase mask*" means that the phase mask (not the HOD FZPs) has more rings, *i.e.*, larger J, and the radius number of the HOD FZPs is fixed (*i.e.*, the

NA of the HOL is fixed, too.). More rings in phase mask will increase more degree of freedom to achieve longer optical needle with sub-diffraction-limit lateral size.

To avoid the confusion, in our revised manuscript we update the sentence as

"*Theoretically, without changing the high-order zone plates (i.e., its focal length and NA are fixed), the longer distance is also possible if more rings (i.e., having a larger J in Fig. 2a) are used in the phase mask due to the greater spatial degrees of freedom available for light shaping*".

8. "More complex nano-objects can also be imaged with high resolution by using our SCM." This conclusion needs to be clarified more clearly. Are these nano-objects planar or three-dimensional, transparent or semi-transparent or opaque, etc?

Reply: We thank the reviewer for the valuable advice. Here, the "*More complex nano-objects*" are planar and transparent samples, such as the bird as shown in Fig.3h in main text. In our revised manuscript, the sentence has been updated as

"*More complex, planar and transparent nano-objects can also be imaged with high resolution by using our SCM.*"

9. "sub-diffraction-limit focusing", "subdiffraction-limited focusing", it should be consistent in the whole paper. Also in the supplementary material, "1st-order lens" and "1-order lens" should be consistent.

Reply: As suggested, all the items in our revised manuscript have been updated by using "subdiffraction-limit focusing" and "1st-order lens".

10. Both developed lens samples have the NA of 0.9. How will other values of NA influence the HOD performance?

Reply: The high NA of 0.9 is chosen to obtain the small focal spot for the application in nano-imaging and nano-lithography. Exactly, the NA can decrease. In real applications, once the focal length is fixed, the decrement of NA is realized by removing the outermost of the HOD zone plates. But the focal spot for a small NA will enlarge meanwhile optical efficiency will increase slightly (as shown in Fig. 1f in the main text).

11. In the "METHODS" section, "when the diffraction order M is small or the focal length f is large, which means that the second term in the right part of Eq. (1) can be ignored due to relatively small value (compared with the first item)", more details are needed to provide the evidence to support this conclusion, for example, how small of M and how large of f? It is important to use the proposed method for some practical applications.

Reply: Thank the reviewer for the valuable suggestion. We have added a brief discussion in the Methods, as shown below:

"*We can rewrite Eq.(1) as:*

$$r_n = \sqrt{n(2M+1)\lambda f}\sqrt{1+\frac{n\lambda(2M+1)}{4f}}. \quad (2)$$

According to Eq. (2), r_n *can be approximately as* $\sqrt{n(2M+1)\lambda f}$ *when* $n \ll 4f/[\lambda(2M+1)]$*. Because the zone index n is valued between 0 to N, it requires* $N \ll 4f/[\lambda(2M+1)]$*, which yields the diffraction order* $M \ll 2f/(N\lambda)-0.5$ *and* $f \gg \sqrt{2}r_N/4$*. This approximation indicates that the radius* r_N *is small compared with the focal length f, thus leading to small NA of the lens. Therefore, optical efficiency derived under the approximation matches the realistic efficiency only for a small-NA lens. By using the approximation, we have* $r_n^2 = n(2M+1)\lambda f$.”

12. Considering the laser-ablation lithography, more details to discuss the influences of the uniformity of the achieved long focusing needle on the fabrication results are needed.

Reply: The fabricated results are influenced by the longitudinal intensity distribution, the lateral beam size and mechanical vibration, which has been discussed in our revised Supplementary Section 7. We list them in the following for your reference:

“*In laser-ablation-based lithography, the uniformity of optical needle is important to obtain well-behaved patterning. To evaluate the uniformity of the needle, two parameters, including longitudinal intensity distribution and the lateral beam size, are used here. For the longitudinal intensity distribution, the measured results in Supplementary Fig. 5b show slowly varied intensity with a peak at Δz=0. Because the longitudinal intensity is not uniform, only the above-threshold parts of needle can be used to fabricate the samples in lithography. On the other hand, the patterning resolution is also related with the second parameter, i.e., the lateral beam size, which is measured and shown in Supplementary Fig. 5d. The measured beam size shows a slowly decreasing tendency at the above-threshold region from Δz=-8 μm to Δz=14 μm. After comparing the line widths (Fig. 4c in the main text) of the fabricated lines, we find that the widths of the fabricated lines have the similar tendency to that of the longitudinal intensity distribution. It means that the longitudinal intensity above the threshold dominates in the fabrication when the lateral beam size has no significant variation. Beyond these issues, the mechanical vibration of optical system will influence the shape of the fabricated lines. To show it, we provide the transmissive optical microscope images of these slits in Supplementary Fig. 6b, which signifies that some of the slits are not straight due to mechanical vibration.* ”

13. In “Supplementary Figure 4”, “The heights and widths of the slits are 2 μm and 50 nm, respectively.” should be “The lengths and widths of the slits are 2 μm and 50 nm, respectively.”

Reply: We thank the reviewer for careful reading of our manuscript. This typo has been corrected as “*The lengths and widths of the slits are 2 μm and 50 nm, respectively.*”.

14. In "Supplementary Section 6", "…indicates that the experimental longitudinal FWHM can reach 20 μm" should be "… indicates that the experimental longitudinal DOF can reach 20 μm".

Reply: We thank the reviewer for spotting this error and the suggestion. In our revised manuscript, the sentence has been updated as "*···indicates that the longitudinal depth-of-focus (DOF) of the on-axis intensity is 20 μm*"

Comments from Reviewer #3

The authors proposed independent manipulation of unexploited high-order diffraction through an analytical formula that correlates the diffraction order and the width of each zone. The large spatial frequencies enabled the proposed lenses to reduce the lateral focal size down to 0.44λ (NA=0.9). They experimentally demonstrated high-order lens-based confocal imaging with a center-to-center dry resolution of 190 nm, and laser-ablation lithography with achieved direct-writing resolution of 400 nm (0.385λ). The authors demonstrated the applications of the proposed diffractive lenses, and the data in the manuscript are convincing. However, there are some concerns that must be carefully addressed before the paper is fit for publication.

Reply: We greatly appreciate the reviewer for confirming the quality of our experiment results.

1. On page 3 of the main text, the authors claimed that "We note that the additional phase also modulates other undersigned orders but does not influence optical focusing at the designed order M. Thus, by using the phase delay of each zone, we can realize the independent manipulation of the phase at the expected order." How do the authors modulate the optical focusing at the designed order M by designing additional phase, and what effect does this have on other orders?

Reply: 1) To realize optical focusing at the designed order M, we have implemented three steps, which are shown below.

Step 1: Selecting the designed order M by using the structural parameters, which is described in Eq. (1) of main text. With the help of Eq. (1), we can make sure that the M-order diffraction light from all the zones plates (defined by Eq. (1)) is used to form the expected focal spot.

Step 2: Modulating the M-order diffraction light by adding a binary phase of 0 or π into each zone. Because each zone diffracts the M^{th}-order light into the focal spot, the M^{th}-order diffraction is also modulated by the binary phase that is encoded in the phase mask (see Fig. 2a).

Step 3: Optimization of the phase mask for subdiffraction-limit focusing. To decrease the focal spot, we need to optimize the structures of the phase mask so that the spot size is below the diffraction limit (*i.e.*, 0.515λ/NA) of light. The optimization is implemented by using particle swarm algorithms that can change the structures of the phase mask. For an updated phase mask during the optimization, we evaluate its optical performance (*i.e.*, the focal spot size) by simulating the focal electric field with Rayleigh-Sommerfeld diffraction. The optimization process will stop when the focal spot satisfies the expected subdiffraction-limit focusing.

Using these three steps, we can tune optical focusing with the M^{th}-order diffraction from the designed lens.

2) Light at other orders will also have the same binary phase that is encoded in the phase mask. Because the structures of the phase mask are designed at the diffraction order M, the

focal spots at other orders are usually not optimal. Considering that the subdiffraction-limit focusing needs careful control of interference, the light from other orders is not designed so that the size of its focal spot usually cannot be below the diffraction limit of light. It means that our proposed lens is valid for only the designed order.

If one wants to control other orders, we suggest that *M* should be changed to the desired order by repeating our proposed method. Thus, she/he can obtain the subdiffraction-limit focusing at the desired order.

2. As can be seen from Figs 1d and 1e, such a design will bring obvious interference to the diffraction characteristics of the device, greatly reducing the focusing efficiency of FZP. Are there other ways to achieve optical superfocusing? The authors should discuss them.

Reply: We thank the reviewer for the valuable comments.

1) Efficiency issue.

Almost all the reported diffraction lenses need the subwavelength structures to diffract light into the focal spot, but the fabrication of subwavelength structures at shorter-wavelength spectrum is quite challenging. So, the motivation of this work is to answer the question of whether a diffractive lens without any subwavelength structure can focus light into a subdiffraction-limit spot. We propose the solution for subdiffraction-limit focusing with high-order diffraction of light without subwavelength structure. Our manuscript has proved its feasibility in both theory and experiment with demonstrations in nano-imaging and nono-lithography. Certainly, the accompanying issue of using high-order diffraction is the shrunken efficiency, which can be solved by increasing the height of the zone structures. In our revised manuscript, we add some brief discussions about increasing optical efficiency of high-order diffraction, as shown below:

"*Low efficiency from HODs is obtained under the assumption of ultra-thin zone structures. In fact, when the height of the zone structures increases, the waveguide effect in the thick zone can be used to enhance the efficiency with the optimized thickness and line-to-space ratio*[57]".

2) Other methods for superfocusing.

There are two ways to realize optical superfocusing beyond the evanescent region are: i) diffractive lenses with subwavelength structures that are challenging for a large-scale device; ii) traditional objectives with additional phase modulation. The bulky nature of the traditional objectives is a barrier in an integrated optical system.

3. In applying the designed device to laser-ablation-based lithography, do the authors utilize only the higher-order part? How do they filter out the rest of the orders?

Reply: We thank the reviewer for the valuable comment. In laser-ablation-based lithography, only 1^{st}-order diffraction of lens is utilized. As shown in Supplementary Figure 1, the focuses for other diffraction orders are located at different *z*. In our experiment, the sample is firstly placed at the designed focal plane of z=1 mm and then etched by 1^{st}-order focus. It is important to point out that the intensity from other diffraction orders is quite weak (below the intensity threshold for ablation) at our designed focal plane of z=1 mm so that light from other orders has no contribution to the ablation process. So, in our experiment we don't block or filter out the rest orders of diffraction.

4. The efficiency of the proposed device appeared to be much lower than those listed in Supplementary Table 1, which is detrimental to its practical application.

Reply: Thank the reviewer for the comment. We agree with the fact that high efficiency is desired in practical applications. We consider the efficiency issue from two aspects:

1) To increase the efficiency of high-order diffraction by using thicker zone structures. It has been discussed in the section 2 of this reply.

2) To compensate low efficiency by using high-power laser sources or high-sensitivity detector. This point has been shown in the Discussion section of our revised manuscript, as shown below:

"*Moreover, the shrunken efficiency from HODs can be compensated by using higher-power laser sources or high-sensitivity detector in practical applications. For example, although the high-order lens in our confocal microscopy has low efficiency, transmitted light is detected by using a photomultiplier tube that can record weak optical signals. In the laser-ablation lithography, high transient power of the incident ultrafast pulses can also compensate low efficiency of the high-order lens so that the focused light is sufficiently strong to pattern various structures on the materials*."

5. The focal length designed by the author is 1 mm. Can the focal length of such diffractive elements be set relatively small? Like tens or hundreds of microns, how does that affect their performance?

Reply: Thank the reviewer for the comment. Without destroying other optical performances, the proposed diffractive elements can be smoothly fabricated with smaller focal lengths of tens and hundreds of microns, where their fabrications are much easier than those for our proposed lenses.

In fact, for a high-*NA* diffractive lens, it is more challenging to achieve a long focal length at the scale of millimeter because one have to fabricate a larger-size lens. It can be seen by the definition of $NA=\sin(\arctan(R/f))$ for a diffractive lens, where R is the radius of the lens and f is the focal length. If the focal length f is large, the radius R of the lens should also be large for a fixed *NA*. Because the diffractive lenses contain fine details, it is more difficult to obtain high-uniform microstructures in a larger-size device, which is caused by the limited capability of the current fabrication tools. Due to this difficulty, almost all the other lenses in Supplementary Table 1 are reported with small focal lengths from several to hundreds of microns.

The reason why we demonstrate the long focal length of 1 mm is to facilitate practical usage of the proposed lens. For example, in our confocal microscopy, the objects must be located at the focal plane of our lens. If the focal length is small, the substrate of the objects will touch the substrate of the lens once they are not parallel to each other, thus increasing the experimental difficulty. Therefore, the long focal length is more pursued in many applications.

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REVIEWERS' COMMENTS

Reviewer #2 (Remarks to the Author):

All my questions have been addressed and this revised vesion seems much better. It can be accepted for publication.

Reviewer #3 (Remarks to the Author):

The manuscript titled "High-order diffraction for optical superfocusing" presents a remarkable study on the manipulation of high-order diffraction (HOD) for optical superfocusing applications. After carefully reviewing the revised manuscript, I would like to provide my remarks on several key aspects.

The noteworthy results of this work include the development of an analytical formula that correlates the diffraction order with the width of each zone, enabling independent manipulation of HODs. This innovation is significant as it opens up new possibilities for sub diffraction-limit focusing in diffractive lenses without relying on sub wavelength features. Furthermore, the authors demonstrate the practical applications of their approach through high-order lens-based confocal imaging and laser-ablation lithography, achieving impressive resolutions.

This work holds significant importance for the field of optics and related areas such as microscopy and lithography. The ability to reduce the lateral focal size down to 0.44λ without sub wavelength features facilitates large-scale manufacture and could potentially revolutionize optical imaging and fabrication techniques.

In comparison to the established literature, this manuscript introduces a novel approach to managing HODs that has not been previously explored. While suppressing HODs using subwavelength structures has been studied, the independent manipulation of HODs for superfocusing purposes is a unique contribution.

The work presented in this manuscript is original and, to the best of my knowledge, has not been reported elsewhere. The authors have responded appropriately to previous reviewer comments, addressing all concerns related to content, structure, argumentation, and data sufficiency.

The data analysis, interpretation, and conclusions presented in the manuscript are robust and support the claims made by the authors. The experimental demonstrations, including confocal imaging and laser-ablation lithography, provide solid evidence of the practical utility and effectiveness of the proposed method.

The methodology employed in this study is sound and meets the expected standards in the field of optics. The authors provide sufficient detail in the methods section, enabling the work to be reproduced by other researchers.

In conclusion, the revised manuscript "High-order diffraction for optical superfocusing" represents a significant advancement in the field of optics. The authors have addressed all previous concerns raised by reviewers, and the manuscript is now well-structured, logically coherent, and linguistically polished. The innovative approach, rigorous methodology, and impressive experimental results make this work a valuable contribution to the scientific community. I strongly recommend the acceptance and publication of this manuscript

Reviewer #4 (Remarks to the Author):

Since the authors have addressed all my concerns, I therefore recommend the publication of this manuscript.

Reviewer #5 (Remarks to the Author):

The manuscript presents an innovative approach to creating flat lenses for subdiffraction-limit focusing. The authors discuss the design and characterisation of flat lenses based on high-order zone plates. While the use of zone-plate-based lenses for subdiffraction-limit focusing is not new, the authors explore and demonstrate the use of high-order diffraction for this purpose in this work. Higher-order diffractive zones typically consist of larger and more widely spaced elements, which reduces the need to create high-density, sub-wavelength-sized feature building blocks in the flat lens.

Two flat lenses are designed to utilise either the 1st or 2nd diffraction orders of their Fresnel zone plates for super-focusing in imaging and laser ablation applications. These lenses have a large numerical aperture (NA=0.9), a relatively long focal distance, and subdiffraction-limit focusing with a lateral focal size of 0.44λ. The design of the flat lenses includes different diffractive zones that direct the input light to the focal point, along with an integrated binary phase mask (0 and π phase shift). The manuscript also provides an analytical formula connecting the geometrical parameters of each zone to the desired diffraction order. The binary phase mask (0 and π phase shift) is adapted within the diffractive zones and optimised to minimise the size of the focal spot.

The designed high-order diffraction lenses have been characterised and experimentally examined in different applications. The authors showed that scanning confocal microscopy with the flat lens had superior resolving power than commercial objectives. Moreover, they demonstrated the patterning of a chromium thin film using laser ablation, benefiting from the relatively large longitudinal depth and subdiffraction-limit focus point size of the 1st-order diffraction flat lens. The flat lens presented in the manuscript outperforms traditional objectives in resolution. Although a high-order diffraction flat lens inherently has low efficiency, as the authors suggest, it may have various niche applications.

The approach of manipulating higher-order diffraction to design flat lenses is highly novel, and the manuscript provides a new path for achieving resolutions beyond the Rayleigh criterion. The revised manuscript contains high-quality data and clear explanations, thanks to the reviewers' comments and the authors' detailed responses.

Comments from the reviewers and our point-by-point replies

Reviewer #2 (Remarks to the Author):

All my questions have been addressed and this revised vesion seems much better. It can be accepted for publication.

Reply: We thank the reviewer for the efforts and recommendation to our work.

Reviewer #3 (Remarks to the Author):

The manuscript titled "High-order diffraction for optical superfocusing" presents a remarkable study on the manipulation of high-order diffraction (HOD) for optical superfocusing applications. After carefully reviewing the revised manuscript, I would like to provide my remarks on several key aspects.

Reply: We thanks the reviewer for his/her strong recommendation to this work.

The noteworthy results of this work include the development of an analytical formula that correlates the diffraction order with the width of each zone, enabling independent manipulation of HODs. This innovation is significant as it opens up new possibilities for sub diffraction-limit focusing in diffractive lenses without relying on sub wavelength features. Furthermore, the authors demonstrate the practical applications of their approach through high-order lens-based confocal imaging and laser-ablation lithography, achieving impressive resolutions.

Reply: We thank the reviewer's confirmation about the novelty of our manuscript.

This work holds significant importance for the field of optics and related areas such as microscopy and lithography. The ability to reduce the lateral focal size down to 0.44λ without sub wavelength features facilitates large-scale manufacture and could potentially revolutionize optical imaging and fabrication techniques.

Reply: We thank the reviewer's conformation about the significance of this work.

In comparison to the established literature, this manuscript introduces a novel approach to managing HODs that has not been previously explored. While suppressing HODs using subwavelength structures has been studied, the independent manipulation of HODs for superfocusing purposes is a unique contribution.

The work presented in this manuscript is original and, to the best of my knowledge, has not been reported elsewhere. The authors have responded appropriately to previous reviewer comments, addressing all concerns related to content, structure, argumentation, and data sufficiency.

Reply: We thank the reviewer's confirmation about our efforts in this work.

The data analysis, interpretation, and conclusions presented in the manuscript are robust and support the claims made by the authors. The experimental demonstrations, including confocal imaging and laser-ablation lithography, provide solid evidence of the practical utility and effectiveness of the proposed method.

The methodology employed in this study is sound and meets the expected standards in the field of optics. The authors provide sufficient detail in the methods section, enabling the work to be reproduced by other researchers.

Reply: We thank the reviewer's strong recommendation to the technical parts of the work.

In conclusion, the revised manuscript "High-order diffraction for optical superfocusing" represents a significant advancement in the field of optics. The authors have addressed all previous concerns raised by reviewers, and the manuscript is now well-structured, logically coherent, and linguistically polished. The innovative approach, rigorous methodology, and impressive experimental results make this work a valuable contribution to the scientific community. I strongly recommend the acceptance and publication of this manuscript

Reply: The strong recommendation from the reviewer is a confirmation of our persistent efforts in this area over the past decade. It encourages a lot to keep more energy and time in this area. Hope we can make more interesting results in the near future.

Reviewer #4 (Remarks to the Author):

Since the authors have addressed all my concerns, I therefore recommend the publication of this manuscript.

Reply: We thank the reviewer for the great efforts and time to our work. We also appreciate it for the valuable suggestions to improve our work.

Reviewer #5 (Remarks to the Author):

The manuscript presents an innovative approach to creating flat lenses for subdiffraction-limit focusing. The authors discuss the design and characterisation of flat lenses based on high-order zone plates. While the use of zone-plate-based lenses for subdiffraction-limit focusing is not new, the authors explore and demonstrate the use of high-order diffraction for this purpose in this work. Higher-order diffractive zones typically consist of larger and more widely spaced elements, which reduces the need to create high-density, sub-wavelength-sized feature building blocks in the flat lens.

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Reply: Thanks a lot for the reviewer's careful reading of this work and the confirmation of the technical achievements in this work.

The approach of manipulating higher-order diffraction to design flat lenses is highly novel, and the manuscript provides a new path for achieving resolutions beyond the Rayleigh criterion. The revised manuscript contains high-quality data and clear explanations, thanks to the reviewers' comments and the authors' detailed responses.

Reply: We thank the reviewer's confirmation about the novelty and technique quality of our work. Appreciate it again for the great recommendation to this work.