Doped metasurfaces: Etched structure-free films based on regular spatially doped semiconductor and compatible with general optical ones

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Highlights
The metasurfaces obtained by the spatial regular doping of semiconductor thin films
The metasurfaces without etching micro-nano structure can be attached with other optical films
The infrared transmittance of the metasurfaces with anti-reflection film is maintained at 90%, and the LUE is as high as 82.2

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Doped metasurfaces: Etched structure-free films based on regular spatially doped semiconductor and compatible with general optical ones

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SUMMARY
The metasurfaces through the reasonable design and arrangement of subwavelength nanostructures to control the spatial light field are expected to replace the traditional lens elements. However, the low light use efficiency (LUE) and difficulty in preparation caused by the etching process restrict the development of its application. Here, an idea of “doped metasurfaces” based on a spatial and regular doping of semiconductor thin films is proposed for the first time. Since the metasurfaces has no etched micro-nano structure, other optical functional films are allowed to be added, which greatly improves and enriches its optical performance. The effectiveness of the design is verified by simulating a suitable meta-surface lens. The simulation results show that this designed MIR metalens possesses wide operating range, high transmittance, and high LUE. The method proposed here provides a new idea or perspective for constructing metasurfaces devices compatible with traditional optical thin films.

INTRODUCTION
Metasurfaces, an innovative concept in the fields of optics, have attracted extensive attention (Fan and Xu, 2017; Zou et al., 2020; Ma and Cui, 2020). Metasurfaces refer to a two-dimensional functional planar structure composed of numerous subwavelength nanostructure units. Through an interaction between the subwavelength structure and incident light field, an efficient modulation on amplitude, phase, and polarization of electromagnetic (EM) wave can be achieved, which is expected to subvert conventional optical components theoretically (Yu et al., 2011; Zhao et al., 2020). A practical application of metasurfaces is namely metalens, which is known for its characteristics of being ultrathin and flat, as well as the ability to focus light just like ordinary lens (Aieta et al., 2012; Dunkelberger et al., 2017). These performance makes the metalens expected to be a revolutionary technology in optics field by replacing the complex and bulky lens group, so as to change mobile phones, cameras and other products smaller and thinner in the future, which explains why this concept could lead a research boom in optics field.

When it comes to the design of metasurfaces, there are three main categories including metallic antenna array metasurfaces (Yu et al., 2011; Aieta et al., 2012; Ni et al., 2012), dielectric metasurfaces (Lin et al., 2014), and metal-dielectric hybrid metasurfaces (Pors and Bozhevolnyi, 2013; Shi et al., 2021). However, behind all the types listed above lie some specific scientific or technical obstacles which hinder the practical implementation. For example, a strong loss would be resulted from the interaction between EM wave and free electrons of metal, which generally causes low transmission and LUE of metal-based metasurfaces (Sun et al., 2012; Pors et al., 2013). Situation is improved to some extent for dielectric-based metasurfaces whose LUE is much higher, but it is still limited by a scattering of unit structure and a mismatch between the equivalent admittance of the metasurfaces’ surface layer and the incident medium or substrate (Arbabi et al., 2015). In addition, dielectric metasurfaces are usually made up of high refractive index dielectric columns with a length comparable to the wavelength of EM wave as shown in Figure 1A, which will lead to a quite difficult preparation and processing process when they are used in mid- or far-infrared range due to the large depth-width radio of dielectric column (Fan and Xu, 2017; Zhao et al., 2021).

Facing those problems, we propose a new design scheme here based on spatial regular doping of semiconductor films: through the technical means like plasma implantation, specific region of the semiconductor thin films can be doped to change the dielectric function of that doped region, after which an
array structure similar to that of traditional metasurfaces can be further designed to realize an efficient modulation of EM wave, as shown in Figures 1B and 1C. The doped region unit here is in fact equivalent to the etched nano-structure of conventional metasurfaces, which means a new type of metasurfaces, namely Doped metasurfaces, can be constructed by reasonably designing and arranging the doped units. Compared with traditional etched-structure metasurfaces, doped metasurfaces have unique merits: without etched structure, they have a smooth and flat surface which makes some functional films on it like optical anti-reflection film accessible, as shown in Figure 1E. In other words, doped metasurfaces can both realize the focusing function owned by normal metasurfaces and stay compatible with functional optical thin films as what traditional lens does, so as to further improve and expand their optical properties.

RESULTS AND DISCUSSION

Based on the design scheme introduced above, we apply Generalized Snell’s Law to the doped metasurfaces. Figure 1D shows the schematic of structure and phase changes of doped metasurfaces, where there are two interfaces with one between incident medium and doped layer and the other between doped layer and substrate. The generalized Snell’s Law is applied to these two interfaces, respectively, as follows (Yu et al., 2011):

\[
\sin(\theta_d)n_d - \sin(\theta_i)n_i = \frac{\lambda_0}{2\pi} \varphi_0
\]  

(Equation 1)

\[
\sin(\theta_i)n_i - \sin(\theta_d)n_d = \frac{\lambda_0}{2\pi} \varphi_1
\]  

(Equation 2)
Figure 2. Design and phase change of doped metasurfaces

(A) Doped unit of metasurfaces, which is a cuboid spatially and regularly arranged in 1 \( \mu m \times 1 \mu m \) with length, width and thickness as 0.8 \( \mu m \), 0.2 \( \mu m \) and 0.1 \( \mu m \), respectively.

(B) Phase change as a function of azimuthal angle of the doped unit at a wavelength of 4.5 \( \mu m \), changing from 0 to 2\( \pi \).

(C) Dielectric function of the doped region.

(D and E) The amplitude distribution of linearly polarized light with vibration directions parallel to and perpendicular to the long axis of the doped region on the surface of the unit structure.

where \( \theta_i \) is incident angle; \( \theta_d \) and \( \theta_p \) are the refraction angle of doped layer and substrate, respectively; \( n_u, n_v, n_i \) and \( n_t \) are the refractive index of incident medium, doped layer and the substrate, respectively; and \( \nabla \phi_2 \) and \( \nabla \phi_1 \) are the phase gradient generated by the two interfaces of metastructures.

In the metasurface layer, the equivalent refractive index \( n_{\text{equiv}} \) of doped region can be expressed as a function between angular frequency \( \omega \) and optical properties of doping metasurfaces obtained by simulation as shown in Figure 2, in which Figure 2A shows the phase change can cover a range from 0 to 2\( \pi \), and the refractive index \( n \) of incident medium, doped layer, and the substrate, respectively; and \( \sin \theta_2 n_i - \sin \theta_1 n_t + 2h((\cos \theta_2 n_{\perp} + \sin \theta_2 n_i) - (\cos \theta_1 n_{\perp} + \sin \theta_1 n_i)) = \frac{\lambda_0}{2\pi} \nabla \phi_2 \) (Equation 3)

where \( h \) is the thickness of the metasurfaces layer, and \( \theta \) is the counterclockwise rotation angle of the doped area.

By adding Equations (1)–(3) together, a equation for calculating the phase gradient of doped metasurfaces is obtained:

\[
\sin (\theta_2) n_i - \sin (\theta_1) n_t + 2h((\cos (\theta_2) n_{\perp} + \sin (\theta_2) n_i) - (\cos (\theta_1) n_{\perp} + \sin (\theta_1) n_i)) = \frac{\lambda_0}{2\pi} \nabla \phi_2 + \frac{\lambda_0}{2\pi} \nabla \phi_1 + \frac{\lambda_0}{2\pi} \nabla \phi_2
\]

(Equation 4)

It can be seen from Equation (4) that the phase change introduced by doping metasurfaces is contributed by the upper and lower interfaces and the middle doped layer. Because the thickness of the doped layer is far less than the wavelength of the light, the phase gradient caused by the refractive index anisotropy of the light passing through the doped layer is small and can be ignored. Therefore, the phase change from 0 to 2\( \pi \) can be realized by reasonably designing the phase gradient of the upper and lower interfaces.

In order to verify the correctness of this idea of doped metasurfaces, a metalens based on a Sn-doped In\(_2\)O\(_3\) thin film doped metasurfaces for mid-infrared range is designed and FDTD method is used to simulate the metals’ optical properties. According to Drude model, the dielectric function \( \epsilon_m \) of doped region can be expressed as a function between angular frequency \( \omega \) of incident light and free-electron concentration \( N_{\text{e}} \) (Brewer and Franzen, 2002):

\[
\epsilon_m = \epsilon_{\text{eo}} + i \epsilon_{\text{em}} = \epsilon_{\text{eo}} - \frac{N_{\text{e}} q^2}{m_e (\omega^2 + \gamma_e^2) \epsilon_0} + i \frac{N_{\text{e}} q^2 \gamma_e}{m_e \omega (\omega^2 + \gamma_e^2) \epsilon_0}
\]

(Equation 5)

where \( q \gamma_e \), \( m_e \), \( \epsilon_0 \), and \( \omega \) are electron charge, electron damping rate, high frequency dielectric constant \( (\epsilon_{\text{eo}} = 3.8) \), electron effective mass \( (m_e = 1.4 m_0) \), and vacuum dielectric constant, respectively. The unit structure and optical properties of doping metasurfaces obtained by simulation are shown in Figure 2, in which Figure 2A shows the doped metasurfaces and one unit structure. The diameter of the whole metasurfaces lens is 30 \( \mu m \), and the unit structures with the same azimuth angle \( \phi \) form a concentric ring. The azimuth angle \( \phi \) is the angle between the Y axis and the long axis of the metasurfaces, the substrate is Si, the free electron concentration of the doped region is \( 8 \times 10^{20} \text{cm}^{-3} \), and the length, width, and thickness of the single doped region are 0.8 \( \mu m \), 0.2 \( \mu m \), and 0.1 \( \mu m \), respectively and oriented in a fixed direction. The spatial regular arrangement of these doped area units constitutes a doped metasurface with a period of 1 \( \mu m \times 1 \mu m \), and the incident beam is a left-handed circularly polarized plane light with a diameter of 30 \( \mu m \). Figure 2B shows that when left-handed circularly polarized light with a wavelength of 4.5 \( \mu m \) is incident, the phase change can cover a range from 0 to 2\( \pi \) with the change of angle \( \phi \); Figure 2C shows the dielectric function of that doped region in 3–6 \( \mu m \) range. Figures 2D and 2E show the amplitude distribution of the linearly polarized light with the vibration direction parallel to and perpendicular to the long axis of the doped region on the surface of the unit structure, respectively. It is obvious from the amplitude distribution in the figure that the scattering of light with different polarization directions varies greatly in the...
doped region. A spherical wavefront is constructed by hyperbolic phase distribution. For normal incident beam with wavelength of $\lambda$, the phase field can be expressed as $\phi(x,y) = \frac{2\pi}{\lambda} (f - \sqrt{x^2 + y^2 + f^2})$, where $f$ is the focal length of lens and $(x, y)$ is the position coordinates of doped unit. Figure 3 shows the light field distribution of the doped metalens with different thickness (0.1 μm, 0.2 μm) metasurfaces layer at wavelengths of 3.5 μm, 4.5 μm, and 3.5 μm. It can be seen from the figure that the focal spot size and the intensity of metasurfaces with two thickness are basically the same, indicating that the increase in the thickness of the metasurface layer does not significantly change the focal length of the metasurface lens. This situation can be explained by Equation (4), which points out that the phase change introduced by doped metasurfaces is mainly related to the interface and the refractive index on both sides. The thickness of the metasurface layer is much smaller than the wavelength, so the contribution to the phase gradient can be ignored. In Figure 3, the light intensity of the focus on the 0.2 μm -thick doped metasurfaces is higher than that on the 0.1 μm-thick doped metasurfaces, because the doped layer itself has antireflection effect on the substrate. The antireflection effect of the 0.2-μm-thick doped layer at the wavelength of 4.5 μm is greater than that of the 0.1-μm-thick doped layer, so the intensity of the focus on the 0.2-μm-thick doped metasurfaces increases. In general, the simulation calculation shown above confirms the reliability of doped metasurfaces which are expected to be applied to practical fabrication.

**Figure 3. Light field distribution of focusing lens with different thickness of doped metasurfaces**

(A and B) Thickness of the doped layer is 0.1 μm and 0.2 μm, respectively.
As mentioned before, due to the absence of etched structure, the performance of some functional films such as antireflective films can be further improved by being deposited on doped metasurfaces. In order to verify how optical film affects the performance of doped metasurface, we design an anti-reflection film for the substrate in combination with a metasurface layer. The dielectric function of the metasurface is calculated using equivalent medium theory, then the vertical and parallel dielectric functions can be expressed as (Kannegulla and Cheng, 2016):

$$
\varepsilon_v = \frac{(1 + f')\varepsilon_m \varepsilon_{\text{ZnS}} + (1 - f')\varepsilon_{\text{In}_2\text{O}_3}^2}{(1 - f')\varepsilon_m + (1 + f')\varepsilon_{\text{ZnS}}} \\
\varepsilon_h = \frac{f'\varepsilon_m + (1 - f')\varepsilon_{\text{ZnS}}}{(1 - f')\varepsilon_m + (1 + f')\varepsilon_{\text{In}_2\text{O}_3}}
$$

(Equation 6)

(Equation 7)

where $f'$ is the doping duty cycle. ZnS and SiO$_2$ which are transparent in mid-infrared range and firmly bonded with In$_2$O$_3$ thin film as well as the substrate are chosen as the antireflection film materials. Multilayer characteristic matrix is used to calculate the antireflection film:

![Figure 4. Comparison of the light field intensity distribution effect achieved by a metasurface compatible with optical films](image-url)

(A) The light field intensity distribution diagram of the metasurface lens without anti-reflection film at the wavelengths of 3.5 µm, 4.5 µm and 5.5 µm. (B) The light field intensity distribution diagram of the corresponding metasurface lens under the anti-reflection film.
where $\delta_i$ and $\eta_i$ are the phase thickness and material admittance of the $i$th layer, respectively, and $\eta_{N+1}$ is the substrate admittance. During the calculation, let $Y = \frac{B_C}{B_B} = n_0$ be the effective admittance (where $n_0$ is air refractive index). Simulation result shows that the antireflection film should be composed of 0.4-μm-thick ZnS and 0.1-μm-thick SiO$_2$. Figure 4 compares the light field distributions of doped metasurfaces at different wavelengths (3.5 μm, 4.5 μm, 5.5 μm) without and with antireflection film. It can be seen that the focal length decreases with the increase of wavelength, this is due to the stronger diffraction ability of light with larger wavelengths. Under the condition of antireflection film, the light intensity at the focus is much higher than that without anti-reflection film, but the focus position is not affected. This shows that the antireflection film only increases the transmittance of the metasurfaces and has little effect on its focusing performance. Without antireflection film, the full width at half maxima (FWHM) of different wavelengths (3.5 μm, 4.5 μm, 5.5 μm) is 4.45 μm, 4.60 μm, and 4.51 μm; With antireflection film, the FWHM is 4.43 μm, 4.57 μm, and 4.49 μm. It can be seen that the FWHM of the same wavelength is basically the same, indicating that the antireflective film does not affect the size of FWHM. Figure 5 is the transmittance curve of 3.0–6.0 μm range and the LUE is at 3.5 μm, 4 μm, 4.5 μm, 5 μm, and 5.5 μm wavelengths, from which it can be seen that the transmittance of metalens with antireflection film is about 90% in 3.5–5.5 μm range, which is much higher with an average increase of more than 35% than that without coating. In midwave infrared, the currently known highest-efficiency metasurface transmittance can reach about 75% (Zhang et al., 2018), which is lower than our design, indicating that our design of doped metasurface combined with the antireflection film is a very efficient method. We calculated the LUE by comparing the optical energy of focal spot with total incident light energy. For LUE, metalenses with antireflection film has a much higher efficiency with an average increase of over 27% than that without coating, and the highest LUE can reach 82.2%, which shows that antireflection film is a key role in improving the efficiency of metasurfaces.

Conclusions

Generally speaking, the concept of doped metasurfaces realized by changing dielectric function via doping is put forward for the first time in this paper. The characteristics of doped metasurfaces in midinfrared range are studied. Besides, based on the property that doped metasurfaces can be added to functional optical film, the influence of antireflection film on transmission and LUE of doped metasurfaces is also studied. Simulation results show that the transmittance and LUE of doped metasurfaces with the antireflection film is much higher than those without this film. The structure of doped metasurfaces combined with antireflection film proposed in this paper is expected to solve the problems including the low infrared efficiency of metal-based metasurfaces and the difficult preparation of dielectric metasurfaces. This technology can be applied to the fields of planar lens, vortex phase plate, holographic phase plate, polarization converter, and wavelength selector, which brings new perspectives and ideas to the design of metasurfaces devices.
Limitations of the study
The metasurface is limited to the regulation of infrared light, and applications in other wavelengths are yet to be developed.

STAR+ METHODS
Detailed methods are provided in the online version of this paper and include the following:

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AUTHOR CONTRIBUTIONS
KJ and WZ proposed the idea, KJ completed the theoretical calculation, YD, WZ and HQ analyzed the data and discussed the results, All the authors wrote the paper and approved the final manuscript.

DECLARATION OF INTERESTS
The authors declare no competing financial interests.

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STAR METHODS

KEY RESOURCES TABLE

| REAGENT or RESOURCE | SOURCE | IDENTIFIER |
|---------------------|--------|------------|
| Software            |        |            |
| FDTD Solutions      | https://www.lumerical.com/products/fdtd/ | version 8.6.0 |

RESOURCE AVAILABILITY

Lead contact
Further information and requests for resources should be directed to and will be fulfilled by the lead contact, Wei Zheng (zhengw37@mail.sysu.edu.cn)

Materials availability
This study did not generate new unique reagents.

Data and code availability
This study did not generate any unique dataset.

METHOD DETAILS
The theoretical results of the paper were verified by FDTD Solutions simulation, which simulated the phase change, amplitude distribution, focused light field distribution, and light use efficiency. The transmittance is calculated using TFCalc according to the equivalent medium theory.