Metasurface holograms reaching 80% efficiency

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Supplementary Section 1. Physical model of plasmonic nanorod

Here we use a simple model to show that the high efficiency conversion between the two circular polarizations arises from the interplay of the antenna resonance and the Fabry Pérot effect of the multilayer structure. We first derive the complex transmission and reflection coefficients through the antenna monolayer residing at the interface between two media of index $n_1$ and $n_3$. The sheet of ultrathin antennas is treated as a homogeneous layer with refractive index $n_2$, and thickness $d$, which will later be taken to the limit of zero thickness. For the three layer medium as shown in Supplementary Fig. 1, for the configuration the Fresnel equations can be expressed as

$$t^{-1} = \frac{n_1 + n_3}{2n_1} \cos(n_2 k_0 d) - i \left( \frac{n_3}{n_2} + \frac{n_2}{n_1} \right) \sin(n_2 k_0 d)$$  \hspace{1cm} (1)$$

$$rt^{-1} = \frac{n_1 - n_3}{2n_1} \cos(n_2 k_0 d) - i \left( \frac{n_3}{n_2} - \frac{n_2}{n_1} \right) \sin(n_2 k_0 d)$$  \hspace{1cm} (2)$$

$$r't^{-1} = \frac{n_3 - n_1}{2n_3} \cos(n_2 k_0 d) - i \left( \frac{n_3}{n_2} - \frac{n_2}{n_3} \right) \sin(n_2 k_0 d)$$  \hspace{1cm} (3)$$

$$t'^{-1} = \frac{n_3 + n_1}{2n_3} \cos(n_2 k_0 d) - i \left( \frac{n_3}{n_2} + \frac{n_2}{n_3} \right) \sin(n_2 k_0 d)$$  \hspace{1cm} (4)$$

where $t$ and $r$ are the complex transmission and reflection coefficients for light incident from medium $n_1$, whereas $t'$ and $r'$ are the coefficients for light incident from medium $n_3$. We next

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relate the susceptibility of medium $n_2$ to the polarizability of individual antenna as,

$$\chi_e = \frac{\alpha_e}{a^2 d}$$

(5)

where $\alpha_e$ is the antenna polarizability, $a$ is the in-plane lattice constant, and $d$ is the thickness. Due to the localized Plasmon resonance of the antennas, $\alpha_e$ can be assumed to have a Lorentzian form $\alpha_e \propto \frac{1}{\omega - \omega_0 + i\gamma}$. The permittivity is given by $\varepsilon = 1 + \chi_e \approx \chi_e = \frac{\alpha_e}{a^2 d_m}$

for sufficiently small $d$. The refractive index of medium 2 can therefore be expressed as,

$$n_2 \approx \frac{1}{a} \sqrt{\frac{\alpha_e}{d_m}}$$

(6)

Thus, for sufficiently small $d_m$, $n_1 = 1$ (air), $n_2 = n$ and $n_3 = n_3$ (substrate), the transmission coefficient is approximated as,

$$t^{-1} = \frac{1 + n^2_n}{2} \cos(n_k d) - \frac{i}{2} \left(\frac{n_n}{n} + n \sin(n_k d) \right) \approx \frac{1 + n^2_n}{2} - \frac{i}{2} \frac{k_0 \alpha_e}{a^2}$$

$$\Rightarrow t = 1/(\frac{1 + n^2_n}{2} - i\omega P)$$

(7)

whereas $P = \frac{\alpha_e}{2ca^2} = \frac{g}{\omega - \omega_0 + i\gamma}$ has the same resonance form as the antenna polarizability, and $g$ is a parameter indicating how strong the antenna couples with the incident light. Following the same procedure for the reflection coefficient and for light incident from the side of medium $n_3$ (substrate), we can write

$$r = (\frac{1 - n^2_1}{2} + i\omega P)/(\frac{1 + n^2_n}{2} - i\omega P)$$

(8)

$$t' = n_3/(\frac{1 + n^2_n}{2} - i\omega P)$$

(9)

$$r' = (\frac{n_3 - 1}{2} + i\omega P)/(\frac{1 + n^2_n}{2} - i\omega P)$$

(10)

To retrieve the unknown parameters $g$, $\gamma$ and $\omega_0$, we simulated the transmission spectrum for light incident from the air side onto a monolayer of antennas with uniform orientation sitting on top of a MgF$_2$ substrate using Comsol software. Figure Supplementary 2 shows the fitting of equation (7) to the simulated transmission coefficients. In the simulation, the polarization of light is along the long axis of the antennas. As a result, we obtain for the retrieved parameters $g=0.19620$, $\gamma=2\pi\times6.35\times10^{12}$ rad/s and $\omega_0=2\pi\times3.53\times10^{14}$ rad/s.

In the next step, we consider the configuration of the entire metasurface – the sheet of antennas on top of a finite thick MgF$_2$ layer with a thick metal ground plane underneath, as shown in Figure 1a of the main text. The reflection coefficient of this three layer system is given by

$$R = r + \frac{tt' r_m e^{i2n_k d_1}}{1 - r'r_m e^{i2n_k d_1}} \approx r + \frac{tt' e^{i\alpha}}{1 - r'e^{i\alpha}}$$

(11)
Supplementary Fig. 2: Numerical simulations of the transmission amplitude (upper) and phase (bottom) coefficients for light incident from air onto a monolayer of antennas, and the corresponding fitted data with eq. 7. These curves show the results for light with polarization along the long axis of the nanoantennas.

where \( r_m \) is the complex reflection coefficient at the MgF\(_2\)/gold interface, and 
\[
\alpha = 2n_k d_3 + \varphi(r_m)
\]
is the round trip phase of the dielectric layer plus that of the reflection at the ground plane (here \( d_3 \) is the thickness of MgF\(_2\) and \( \varphi(r_m) \) the phase of the complex reflection coefficient \( r_m \)). By substitution the expressions for \( t \), \( t' \), \( r \), \( r' \) and \( P \) into Eq. 11, we obtain

\[
R = \frac{n_k - 1}{2} (\omega - \omega_0 + i\gamma) + i\omega g - \frac{n_k + 1}{2} (\omega - \omega_0 + i\gamma) - i\omega g e^{i\alpha}
\]

In the above equation, if \( \alpha = 2\pi \) for \( \omega = \omega_0 \), the reflection is simply given by

\[
R = -\frac{i\gamma + i2\omega g}{i\gamma + i2\omega g} = -\frac{2\omega g - \gamma}{2\omega g + \gamma}
\]

Thus, under this condition, the reflection coefficient along the long axis of the antenna becomes negative which corresponds to a phase shift of \( \pi \) compared to the incident light. To this end, we assume the antenna is only interacting with the incident electric field in \( x \) direction (along the long axis of the antenna), and does not interact with light at all along \( y \) direction (short axis). Hence, the parameter \( g \) becomes \( g = 0 \) for incident light along \( y \) direction and the phase of the reflection coefficient is approximately \( 2\pi \), which is the sum of the round trip propagation phase (\( \pi \)) and the reflection phase at the metal ground plane (\( \pi \)).

Thus, the phase difference between the two orthogonal polarization directions is \( \pi \). In addition, the coupling of the antenna to the radiation field is much stronger than the ohmic loss, therefore, the amplitude of \( R \) is close to 1. Consequently, the rotation direction of the electric field of a circularly polarized incident beam is almost completely reversed upon reflection on such a three layer system.
We next show that the phase difference between the two reflection coefficients with orthogonal polarizations is dispersionless around the resonance frequency, leading to a broadband operation. Around the resonance frequency, $\alpha$ can be expressed as

$$\alpha \approx f\Delta \omega$$  

(14)

Where $f$ represents the frequency dispersion of $\alpha$, which consists of the dispersion of the round trip phase in the dielectrics and the dispersion of the reflection phase at the ground metal surface. If we substitute Eq. 14 into the expression for $R$ and expand the expression in $\Delta \omega$, we obtain

$$R = \frac{n_2 - 1}{2} (\Delta \omega + i\gamma) + iog - \frac{n_2 - 1}{2} (\Delta \omega + i\gamma) - iog (1 + if\Delta \omega)$$  

(15)

By neglecting $\gamma$ since it is much smaller than $\omega_g$, we obtain

$$R \approx \frac{2og + i\Delta \omega (1 + f\omega_g)}{2og - i\Delta \omega (1 - f\omega_g)}$$  

(16)

Thus, the phase of $R$ can be approximated as

$$\phi(R) \approx \pi + \frac{\Delta \omega (1 + f\omega_g) - \Delta \omega (1 - f\omega_g)}{2og} = \pi + \frac{2\Delta \omega f\omega_g}{2og} \approx \pi + f\Delta \omega = \pi + \alpha$$  

(17)

Note that along the orthogonal direction the reflection phase is close to $\alpha$, with a slight deviation caused by the weak Fabry Pérot effect due to the small mismatch between the refraction indices of air and MgF$_2$. Thus, the phase difference between the reflections of orthogonal polarizations is dispersionless around the resonance frequency of the antenna, leading to broadband operation.

Supplementary Fig. 3: Phase delay vs wavelength for normal incident light reflection on a gold substrate.

For a more realistic case the metal (gold) is modeled by a Drude model with $\omega_p = 1.37 \times 10^{16}$ rad/s and $\gamma_p = 1.215 \times 10^{14}$ rad/s. At the resonance wavelength for the long axis (849 nm), the reflection phase is $\phi(r_m)$ rad (Supplementary Fig. 3). Substituting this value into the round trip phase $\alpha = 2n_r k_r d + \phi(r_m)$ leads to a dielectric thickness of 133 nm, which slightly less
than a quarter of the wavelength in the dielectric medium (155 nm). Supplementary Fig. 4 shows the modeled reflectivity curves $|R_{LR}^2|$ and $|R_{LL}^2|$ for the dielectric thickness of 133 nm. It is shown that the efficiency of circular polarization conversion reaches 90% while the reflectivity with opposite circular polarization is zero over a broad frequency range. This broad band operation is due to the dispersionless phase difference between the reflection coefficients of the two orthogonal linear polarizations.

**Supplementary Fig. 4:** The modeled reflectivity curves $|R|^2$ for both cross polarization and co-polarization light by taking into account the effect of finite metal conductance. Only the response along the long axis of the antenna has been considered in this curve.

A modification of the optimized dielectric thickness is due to the antenna response along the short axis. Despite that the resonance frequency for the polarization along the short axis is very high and falls out of the frequency range we are interested, it nonetheless contributes to a phase term which is not negligible. Again, we use equation (7) to fit the transmission curve for polarization along the short axis, which shows very good agreement with the numerical simulations (Supplementary Fig. 5). For the fitting parameters we obtain $g=0.15772$, $\gamma=2\pi\times3.5714\times10^{12}$ rad/s and $\omega_0=2\pi\times7.1554\times10^{14}$ rad/s.

**Supplementary Fig. 5:** Numerical simulations of the transmission amplitude (upper) and phase (bottom) coefficients for light incident from air onto a monolayer of antennas, and the corresponding fitted data with eq. 7. These curves show the results for light with polarization along the short axis of the nanoantennas.
Thus, by considering the response of the antenna along the short axis, the dielectric thickness is reduced to 105 nm. The modeled reflectance spectra by using the fitted parameters for the polarization along both long and short axis are plotted in Supplementary Fig. 6, which are in good agreement with the full wave simulation results. The reflectivity shows a broad bandwidth from 600 to 950 nm with over 80% conversion between the two circular polarizations, meanwhile maintaining close to zero reflectance with the same circular polarization.

**Supplementary Fig. 6:** a The modeled and, b simulated reflectivity curves $|R|^2$ for both cross polarization and co-polarization light. The thickness of the MgF$_2$ film is 105 nm.

Our fabricated metasurface has a thickness of 90 nm, which is close but not exactly the optimized value. Despite this deviation, we nonetheless obtain a broadband performance with very high conversion efficiency. The modeled and simulated results with the parameter of our realized metasurface are shown in Supplementary Fig. 7. The slight deviation of the modeling from the full wave simulation result is due to the near field coupling of the antenna array with the metal ground plane, which is not taken into account in our modeling.

**Supplementary Fig. 7:** a The modeled and, b simulated reflectivity curves $|R|^2$ for both cross polarization and co-polarization conversion. The thickness of the MgF$_2$ film is 90 nm as in the actual realized metasurface.
Supplementary Section 2. Simulation of ohmic loss of nanorod-based antenna

The key to the high efficiency and low ohmic loss is the interplay between the antenna resonance and the Fabry Perot resonance. It has been shown previously that a similar structure but with much more reduced dielectric spacer thickness can be used as perfect absorber (almost 100% loss) at the resonance frequency$^1$. In these experiments the near field coupling between the antennas and the ground metal plane plays an important role. As shown in equation 13, under the Fabry Perot condition in our configuration, the ohmic loss at the resonance frequency of antenna is around $2\gamma /\omega_0 g$, which leads to very small loss (14% in simulation and less than 20% in experiment) for a considerably large $g$ (radiative coupling). Indeed, the ohmic loss in our configuration is very close to that of light transmitting through a single metasurface layer (without the ground metal plane) around the resonance wavelength (800 - 850 nm) of the antenna, as shown in Supplementary Fig. 8.

![Ohmic Loss vs Wavelength](image)

**Supplementary Fig. 8:** Numerical simulations of ohmic loss vs wavelength. The loss for a single-layer structure is calculated by $1 - |r_x|^2 - n_s |t_x|^2$, where $r_x$ and $t_x$ are the reflection and refraction coefficients along the long axis direction, respectively, $n_s$ is the refractive index of the MgF$_2$ film. The loss for a three-layer structure is calculated by $1 - |R_s|^2$, where $R_s$ is the reflection coefficient along the long axis direction.

Supplementary Section 3. Hologram design using the concept of Dammann gratings

In our work, we use a 2x2 pattern hologram design based on concept of Dammann gratings$^{4,5}$, which is helpful to avoid the appearance of laser speckles$^6$ and thus improves the image fidelity. We simulated the intensity in far field for both 2x2 periodic and single holograms with the same phase distribution (shown in Supplementary Fig. 9). As can be seen from the image the 2x2 periodic hologram generates an image consisting of discrete spots. In comparison, a single hologram produces a continuous image with lower image fidelity, i.e. more laser speckles. Our current design can be further optimized by a $N \times N$ ($N$ is an integer) Dammann grating design, which would increase the image quality further. However, this will in turn require the need for longer fabrication time.
Supplementary Fig. 9: a and b, Simulated holographic image of Einstein’s portrait and a close up of the character 'M' using 2x2 periodic hologram. c and d, Identical simulated holographic image but with a single hologram.

Supplementary Section 4. Simulation of the diffraction efficiency of the metasurfaces
Each antenna of the metasurface is excited not only by the incident field, but also by the retarded fields from all other antennas. Therefore it might not be appropriate to simply look at the angular distribution of the radiation from an isolated antenna. However, full wave numerical simulation of our hologram is beyond our simulation capability due to the large array of antennas contained in the hologram. On the other hand, a hologram can be considered as diffraction of light into the desired diffraction orders, and the phase profiles of realistic holograms are far from being random. This can be seen from the zoom-in view of the nanoantennas in Fig. 3a, where the orientations of the antennas vary only gradually in both x and y directions in most part of the hologram. To address the issue of antenna response in a non-uniform array, here we studied the efficiency of diffraction for metasurfaces with different linear phase gradients such that for a normal incident light a range of diffraction angles can be achieved. While this configuration does not represent a typical hologram that diffracts light into many diffraction orders, it at least contains antennas with different orientations.

In the simulation, we construct metasurfaces with the number of antennas $m=3, 4, 5, 6$ and $10$, with the linear phase gradient varying between $2\pi/(10a)$ to $2\pi/(3a)$, where $a$ is the pixel size of the nanorod. The angular dependence of the diffraction efficiency at different wavelengths is shown in Supplementary Fig. 10. Our holographic image has a projection angle of 60° (full angle) in the x direction and 70° (full angle, $30°+5.2°*2=70°$) in the y direction. From Supplementary Fig. 10, one can see that the efficiency at a diffraction angle of 35° drops by only 4-10 % (depending on the wavelength) compared to that of normal
incidence/reflection (corresponding to 0°) over a broad wavelength band from 650 – 1050 nm. For a smaller wavelength band the efficiencies are still reasonably high for angles up to 60°. Hence from the scattering characteristics of the nanorod antennas we conclude that our hologram works also with high efficiency at larger diffraction angles.

Supplementary Fig. 10: Scattering efficiency of the nanorods vs diffraction angle for four different wavelengths.

The numerically simulated diffraction efficiency spectra are shown in Supplementary Fig. 11. At small phase gradients (4, 5, 6, 10 antennas in the unit cell) it is only slightly different from that of uniform metasurface shown in Fig. 1f of main text. On the other hand, a large phase gradient (3 antennas in a unit cell) would reduce both the efficiency and the bandwidth dramatically.

Supplementary Fig. 11: Numerical simulations of the diffraction efficiency (upper) and zero-order efficiency (bottom) for light incident on linear phase gradient gratings which contain 3, 4, 5, 6 and 10 unit cells in one period respectively.

Supplementary Section 5. Phase deviations from coupling effects of neighboring nanorods
In this section, the effect of the near field coupling between neighboring nanorods is investigated. The coupling will be strongest if the corners of the two nearest rods are closest to each other and weakest when they are most apart from each other.
Here we have investigated the phase deviations from the designed value (2\(\phi\), \(\phi\) is the orientation angle) for a uniform array of nanorods but with different orientations (Supplementary Fig. 12a). Our analysis shows (see Supplementary Fig. 12b) that deviations of the reflected phase (LCP to RCP or vice versa) from that of the designed value (twice the orientation angle) is very small (2°-4°) for several wavelengths, which is much less than the phase step (22.5°/step) used in our hologram design.

**Supplementary Fig. 12:** a, Schematic illustration of the arrangement of periodic nanorods with various orientation angles. b, Simulated phase deviation caused by near field coupling of adjacent nanorods at different working wavelengths. The phase shows over the entire range a phase deviation that is smaller than 9°.

**Supplementary Section 6. Experiments**

**Holographic imaging:** For the experiments we used several laser sources including a red laser (He-Ne laser, wavelength: 632.8 nm) and a near infrared fiber-coupled diode laser (New Focus, wavelength 780 nm). The collimated laser beam is normally incident on the hologram after passing through a linear polarizer and a quarter waveplate. The diameter of the laser beam is around 1.5 mm, which fully covers the hologram. The reflected holographic image is projected onto a white screen 300 mm away from the sample. We captured the red and near infrared holographic images (Figure 3 in the main text) by using a commercial digital camera (Nikon D3200) and an ELOP-Contour CMOS Infrared Digital Camera, respectively.

**Optical efficiency measurement:** The setup of the optical efficiency measurement is shown in Supplementary Fig. 13. The polarization state of the laser source is converted to circularly polarization by passing through a linear polarizer and a quarter waveplate. A lens with focal length of 300 mm is used to focus the incident beam onto the hologram. Color filters are used to remove the unwanted light generated by the fiber laser. In addition, an iris is used to block the scattered beam from multi-reflections between the optical interfaces. Two identical condenser lenses with high numerical aperture (N.A.=0.6) are used to collect and focus the diffracted light for the measurement by using a power meter in the wavelength range: 400 nm-1100 nm. The beam focused onto the sample with a spot size of \(~300 \mu m\) in
diameter, which is less than the size of the hologram (666.6 x 666.6 μm²). The intensity of the light is measured at two points in the beam path, Point 1 and Point 2, as shown in Supplementary Fig. 13. The window efficiency \( \eta_W \) can be evaluated by:

\[
\eta_W = \frac{P_z - P_2}{P_1} \cdot \frac{1}{T_c},
\]

where \( P_z \) is optical power of zero-order beam, \( P_1 \) and \( P_2 \) are optical power measured at point 1 and 2, separately. \( T_c \) is the transmission efficiency of the two condenser lenses, which is obtained by ray tracing simulations and experimental measurement. For our off-axis hologram design, the zero-order beam is separated from the signal beam and can be captured and measured separately.

In Supplementary Fig. 14 it is shown that the numerical aperture is slightly larger than that of the projected image. However, the background noise, except the zero order signal captured within the imaging field, is extremely low. By analyzing the captured holographic image, the background noise is only around 1.1% of the total energy we collected.

Supplementary Fig. 13: Illustration of the optical efficiency measurement setup. The incident circularly polarized beam is generated by the linear polarizer (LP) and the quarter waveplate (QWP). The incident circularly polarized beam is focused on the sample and the diffracted light in reflection with opposite circular polarization is collected by two condenser lenses and an optical power detector. For the efficiency measurement, the power was measured at point 1 and 2.

Supplementary Fig. 14: Illustration of geometric parameters of the hologram image at the collection window of the first condenser lens.
We also measured the angle dependent efficiency by fixing the working wavelength at 825 nm (peak efficiency position). The measured results are shown in Supplementary Fig. 15. It is observed that the optical efficiency of hologram drops when the incident angle is increased from 9° to 45°.

Supplementary Fig. 15: Measured optical efficiency as a function of the incident angle.