ADVANCED OPTICAL MATERIALS

# Supporting Information

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# Plasmonic Metasurfaces with High UV–Vis Transmittance for Photopatterning of Designer Molecular Orientations

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# Plasmonic Metasurfaces with High UV-Vis Transmittance for Photopatterning of Designer Molecular Orientations

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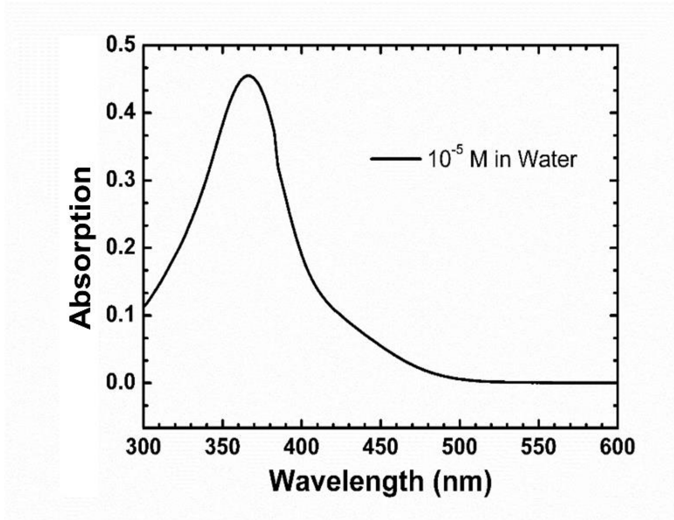


**Figure S1** Absorption spectrum of SD-1 in water.

## 1. Absorption Spectrum of SD-1

The sulfuric dye 1 (SD-1) is a popular photoalignment material developed by Chigrinov et al.[1,2] To measure the absorption spectrum of SD-1, we dissolve it in DI water at $10^{-5}$ M concentration. Fig. S1 shows the absorption spectrum measured by using a PerkinElmer UV-VIS photospectrometer.

## 2. Local Field Distribution for Parallelepiped Orientation $\theta = 30°$

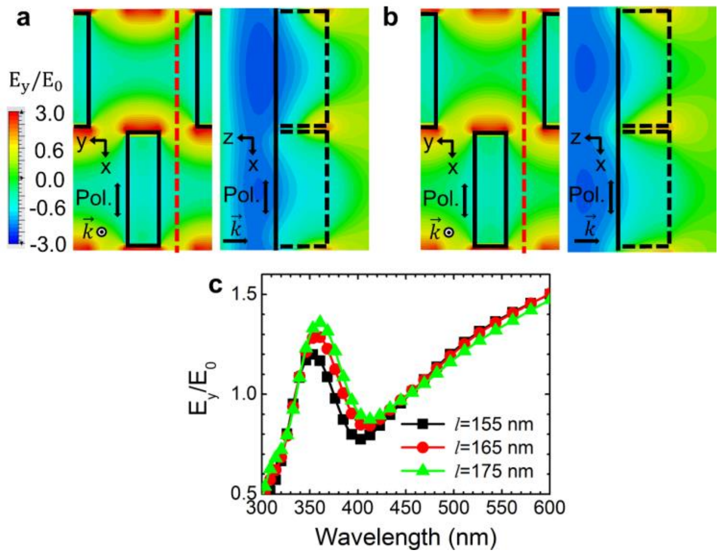


**Figure S2** (a, b) Snapshots of the local electrical field distributions, at two resonant wavelengths, a) 375nm and b) 480 nm respectively. The incident polarization is along the y-axis, and *l*=180 nm, *h*=80 nm, *w*=50 nm, *p*=220 nm, $\theta$=30°. (c) Calculated local electric field enhancement as a function of wavelength at the top center of the gap between two neighboring parallelepipeds. Here *p*, *h* and $\theta$ are fixed at 220 nm, 80 nm and 30° respectively.

## 3. Plasmonic Lattice Resonance

With the couple dipole (CD) approach, the parallelepipeds can be treated as electrical dipoles. For the i-th parallelepiped at position $r_i$, the dipole moment can be expressed as: $\boldsymbol{P}_i = \alpha_0 \cdot \boldsymbol{E}_i$, where $\alpha_0$ is the polarizability.[3,4] The electric field acting on the dipole $P_i$ equals the sum of the incident electrical field and the electrical fields generated by the other parallelpipeds:

$$E_i = E_0 + \sum_{j=1} \frac{e^{ik\boldsymbol{r}_{ij}}}{\boldsymbol{r}_{ij}{}^3} \{k^2 \boldsymbol{r}_{ij} \times (\boldsymbol{r}_{ij} \times \mathbf{P}_j) + \frac{1 - ik\boldsymbol{r}_{ij}}{\boldsymbol{r}_{ij}{}^2} \times [\boldsymbol{r}_{ij}{}^2 \mathbf{P}_j - 3\boldsymbol{r}_{ij}(\boldsymbol{r}_{ij} \cdot \mathbf{P}_j)]\}$$

$E_0$ is the incident field. When the incident wave vector is perpendicular to the plane of the diple arrays, the electrical fields on different dipoles are the same, say $E_i = E_{total}$. Then all are equal, i.e. $\boldsymbol{P}_i = \alpha_0 \cdot \boldsymbol{E}_{total}$.

$$E_{total} = E_0 + \sum_{i \neq j} [\frac{(1 - ikr_{ij})(3cos^2\theta_{ij}e^{ikr_{ij}})}{{r_{ij}}^3} + \frac{k^2sin^2\theta_{ij}e^{ikr_{ij}}}{r_{ij}}] \, (\alpha_0 \cdot E_{total})$$

In this formula, $\theta_{ij}$ is the angle between the vector $\boldsymbol{r}_{ij}$ and the electrical direction. By defining a retarded dipole sum S(k):[3–5]

$$S(k) = \sum_{i \neq j} [\frac{(1 - ikr_{ij})(3cos^2\theta_{ij}e^{ikr_{ij}})}{{r_{ij}}^3} + \frac{k^2sin^2\theta_{ij}e^{ikr_{ij}}}{r_{ij}}]$$

the dipole moment $\boldsymbol{P}_i$ can be expressed as $\boldsymbol{P}_i = \alpha_{cluster} \cdot \boldsymbol{E}_0$, where

$$\alpha_{cluster} = \frac{\alpha_s}{1 - \alpha_s S}$$

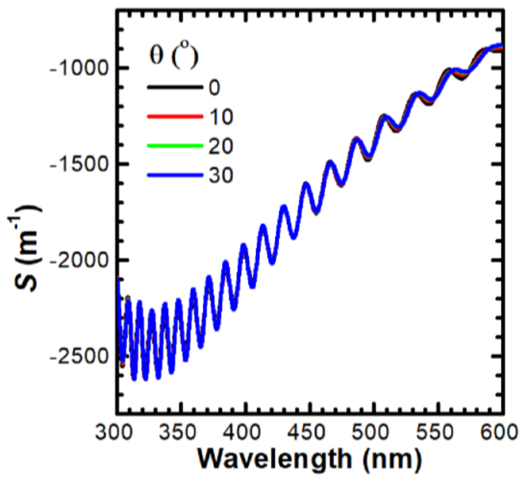


**Figure S3** Calculated S parameter as a function of wavelength and orientational angles of parallelepipeds.

A resonance emerges when $Re(1 - \alpha_s S) = 0$, and the resonant frequency is determined by the real part of the S parameter.[3–5] Since the parallelepipes are positioned in a triangular lattice, we only need to study the cases with $\theta$ varied between 0° and 30°. We calculated the S-parameter by summing over $r_{ij}$ within a distance of 50$p$. The numerical results shown in Figure S2 indicate that the variations of $\theta$ impacts little on S. As a result, the frequency of the lattice resonance is insensitive to $\theta$.

## 4. Projection Photopatterning System

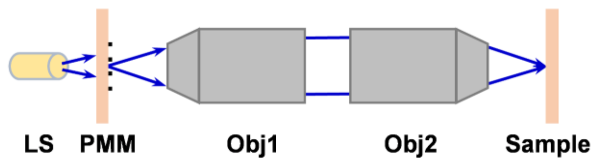


**Figure S4** Schematic setup for the projection photopatterning based on the PMM. LS: white non-polarized light source; Obj1: imaging objective; Obj2: projection objective. The sample substrate is coated with a thin layer of photoalignment material such as SD-1.

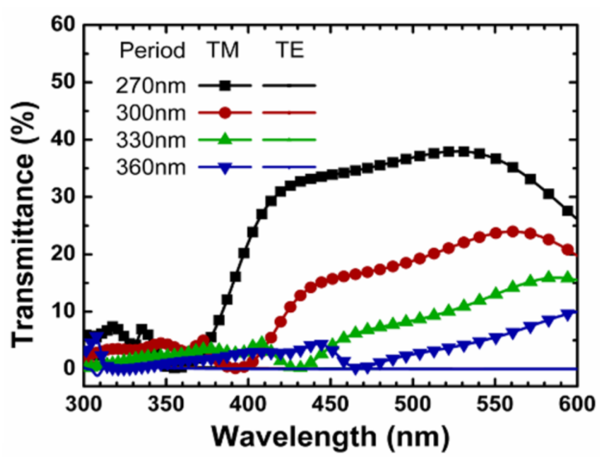


**Figure S5** Simulated transmission spectra for plasmonic metamasks based on rectangular nanoapertures. The length and width of the rectangular nano-apertures are fixed at 220 and 100 nm.

## 5. Transmission Spectra of the Nanoaperture-Based Plasmonic Metamasks

Here we present simulation results for the optical transmission spectra of the plasmonic metamasks based on rectangular nanoapertures developed in our prior work.[6] The optical efficiency is almost negligible in the UV wavelength range where the absorption bands of the SD-1 and PAAD-22 are located.

## Reference


[1] H.-S. Kwok, V. G. Chigrinov, H. Takada, H. Takatsu, *J. Disp. Technol.* **2005**, *1*, 41.

[2] V. Chigrinov, H. S. Kwok, H. Takada, H. Takatsu, *Liq. Cryst. Today* **2005**, *14*, 1.

[3] L. Zhao, K. L. Kelly, G. C. Schatz, *J. Phys. Chem. B* **2003**, *107*, 7343.

[4] S. Zou, N. Janel, G. C. Schatz, *J. Chem. Phys.* **2004**, *120*, 10871.

[5] F. J. García de Abajo, *Rev. Mod. Phys.* **2007**, *79*, 1267.

[6] Y. Guo, M. Jiang, C. Peng, K. Sun, O. Yaroshchuk, O. Lavrentovich, Q.-H. Wei, *Adv. Mater.* **2016**, *28*, 2353.