

Supplementary Information for

Photoelastic plasmonic metasurfaces with ultra-large near infrared spectral tuning

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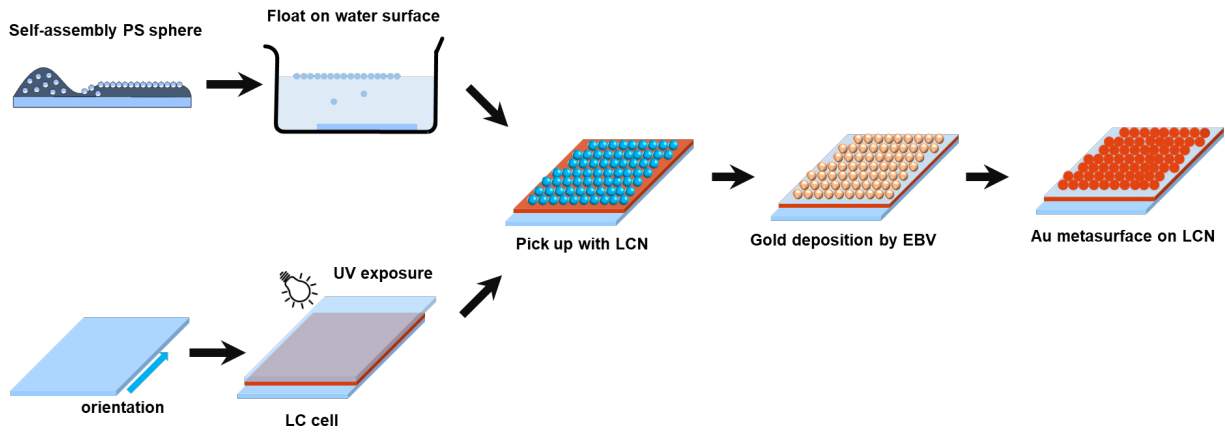
Supplementary Figures 1 to 16

Supplementary Table 1

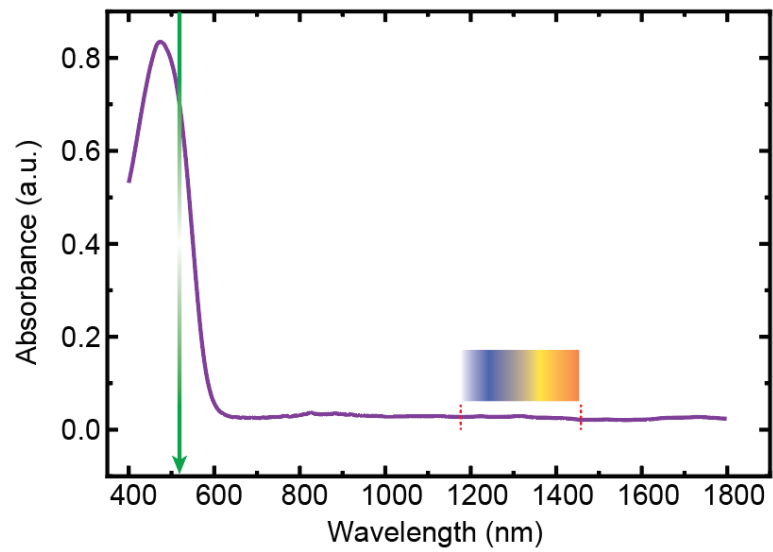
Materials and Methods.

1. Finite-difference time-domain (FDTD) method.
2. Experiment setup for the transmission spectrum.
3. The optical setup for measurements of the temporal response.

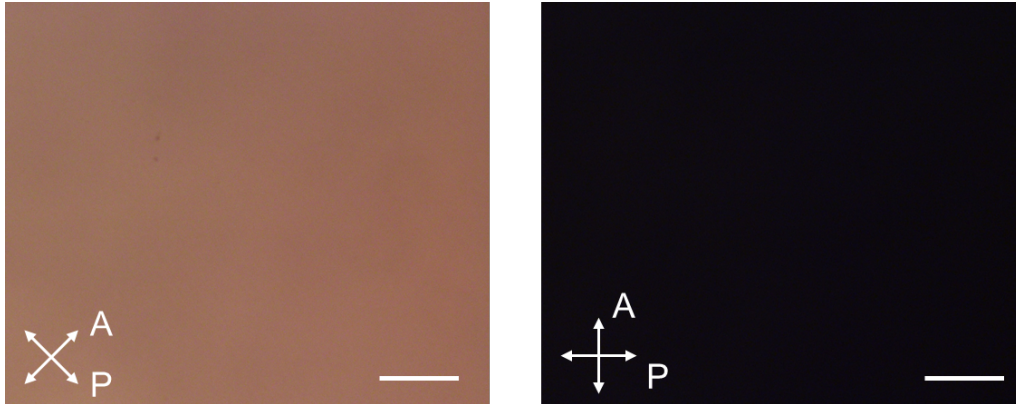
Supplementary Figures.



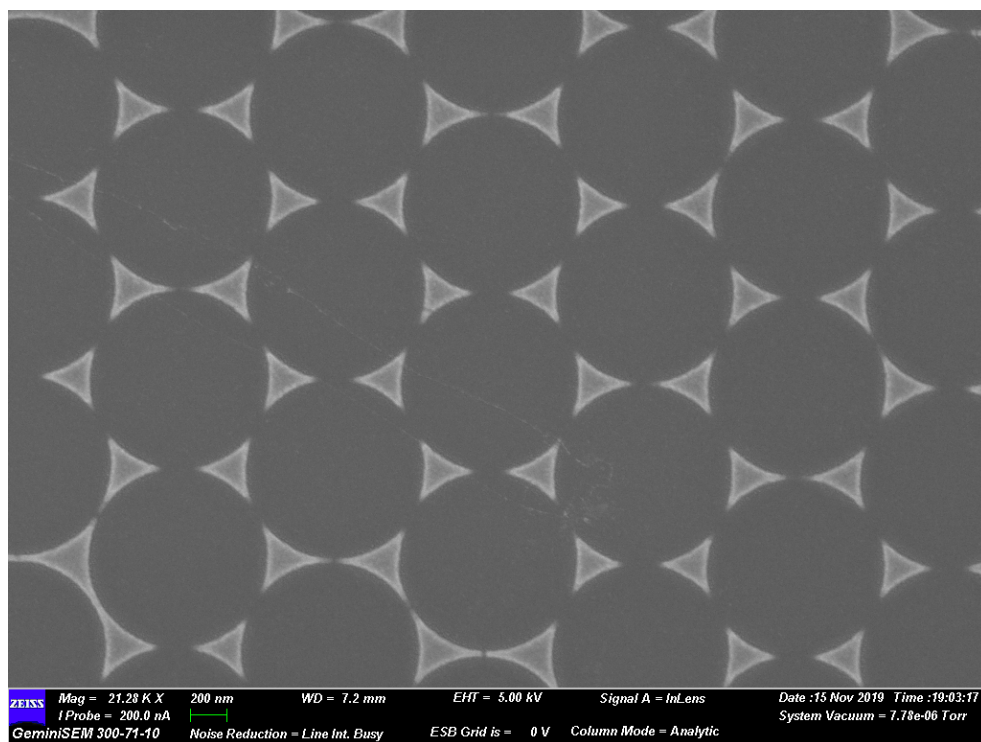
Supplementary Figure 1. Schematic drawing of the fabrication process. Polystyrene (PS) nanosphere monolayer was formed via the air-liquid interface method; LCN was prepared by cell infiltration followed by UV polymerization. The LCN substrate was used to lift-up the self-assembled PS sphere layer, followed by gold deposition and cleaning all the spheres, leaving the gold metasurface on the LCN surface.



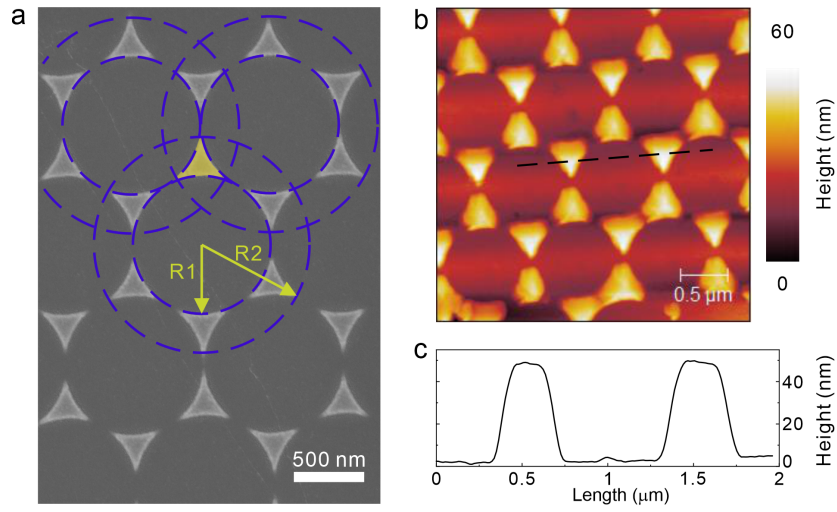
Supplementary Figure 2. Absorption spectrum of the LCN. The green arrow represents the excitation wavelength (532 nm).



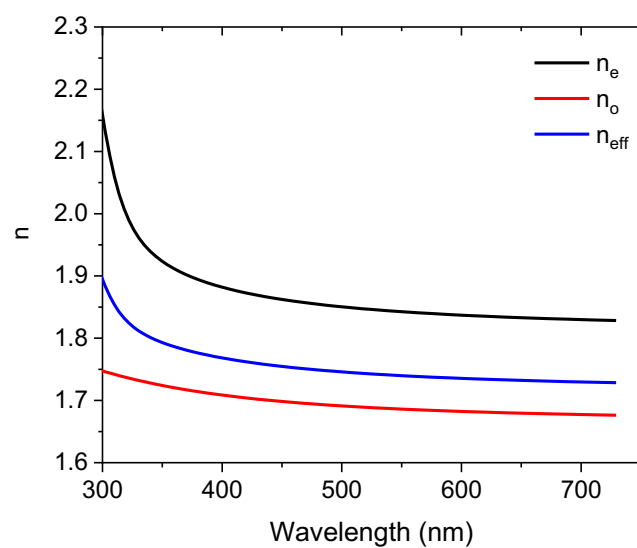
Supplementary Figure 3. Cross polarized optical microscope images of the LCN with the LC director set at $\pm 45^\circ$ (left) and $0/90^\circ$ with respect to the polarizer/analyzer. Scale bar: $20\ \mu\text{m}$.



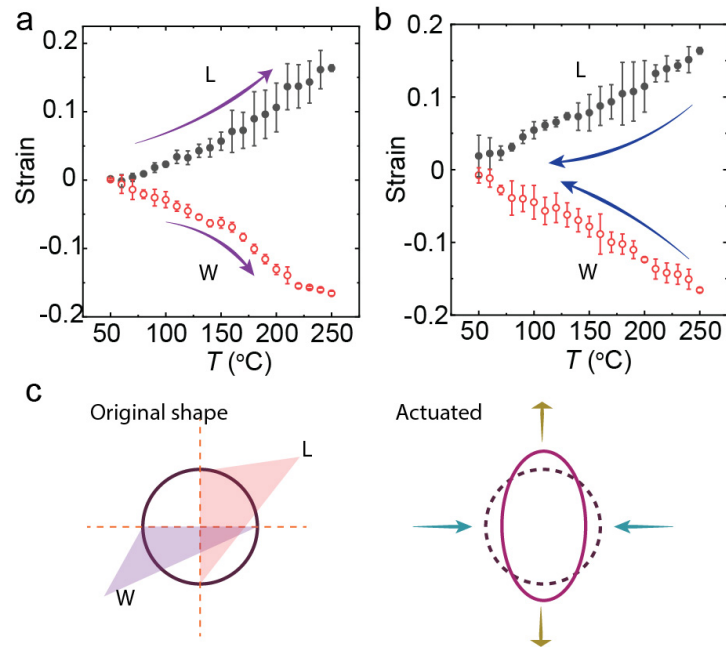
Supplementary Figure 4. Scanning electron microscope (SEM) image of the gold nanoarray on glass substrate.



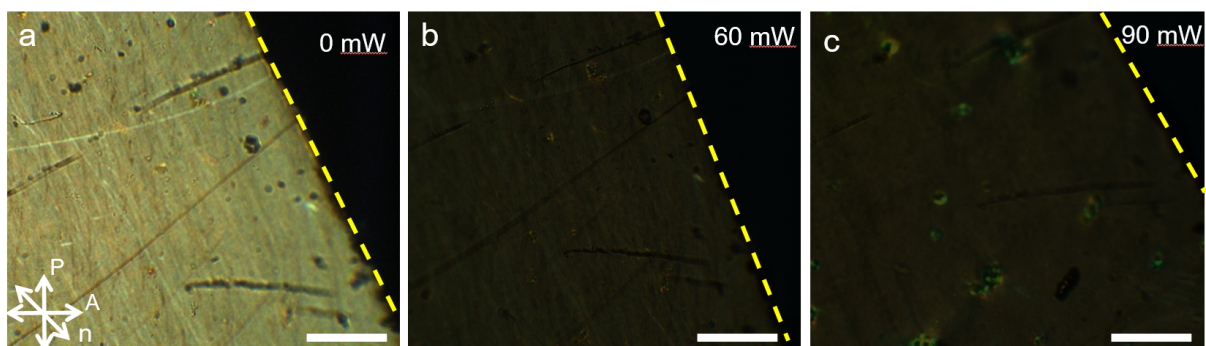
Supplementary Figure 5. Geometry characterization of the Au nanoarray. (a) Detailed SEM image of the Au nanoarray. The radius of R1 and R2 are measured to be 489 nm and 600 nm, respectively. The distance between two neighboring nanotriangles is 577 nm. (b) Atomic force microscope image of the Au metasurface. (c) The height profile along the black dotted line in (b). The thickness of the Au metasurface is about 50 nm.



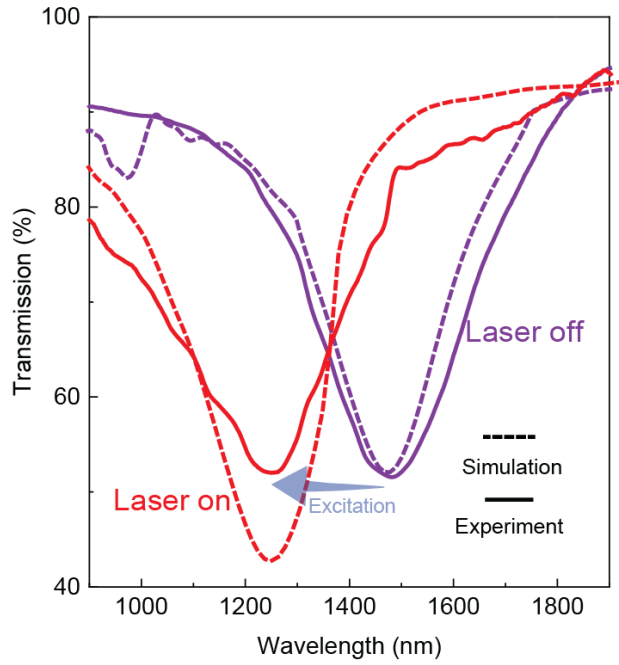
Supplementary Figure 6. Refractive index of the LCN substate. The measurement is conducted with a spectroscopic ellipsometer (UVISEL, HORIBA) at room temperature.



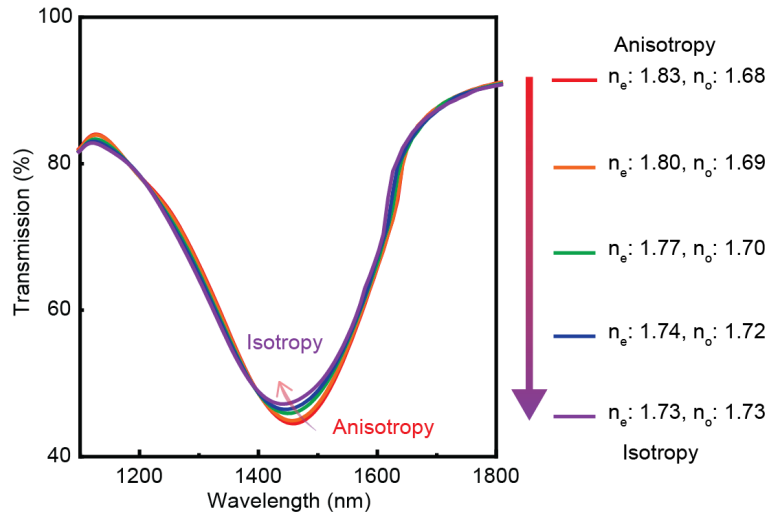
Supplementary Figure 7. Heat induced deformation of the LCN substrate. The shape changes of the LCN substrate during heating (a) and cooling (b). (c) Schematic drawing of the deformation, indicating the anisotropic shape change.



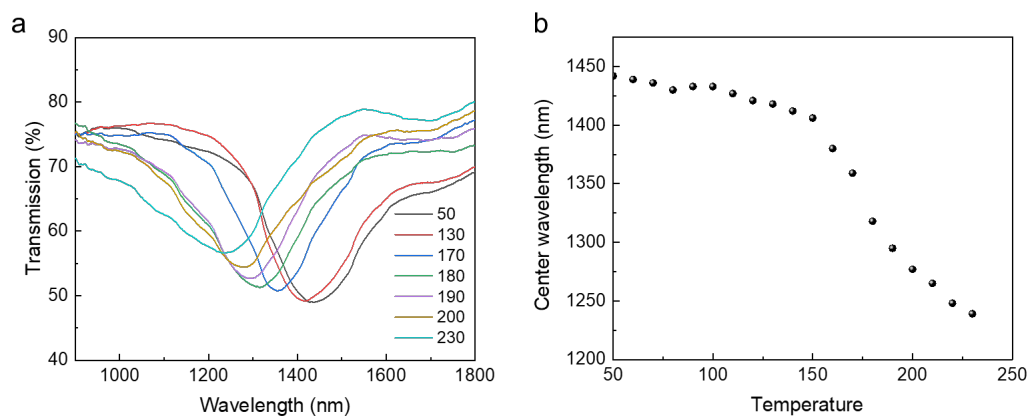
Supplementary Figure 8. Cross polarized microscope images of an LCN substrate without laser excitation (a), and when exposed to a 60 mW (b) and 90 mW (c) excitation (532 nm, spot size 700 μm). The dashed lines indicate the edge of the LCN substrate. Scale bars: 50 μm .



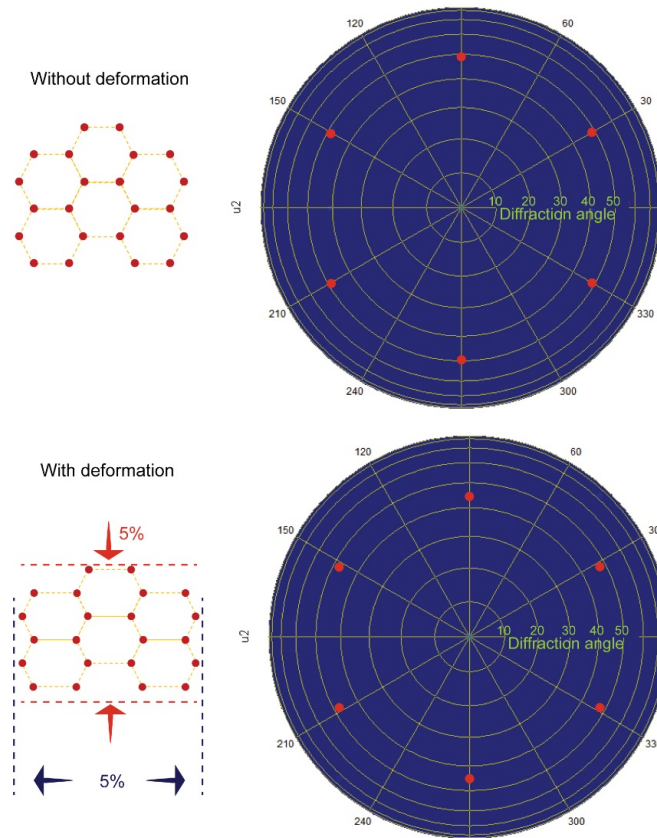
Supplementary Figure 9. Simulated and measured spectral shifting upon laser excitation. In the simulations, due to negligible dispersion in the near-IR region, extraordinary refractive index n_e of 1.828 and the ordinary refractive index n_o of 1.676, measured at 725 nm, are chosen to approximate the LCN birefringence at 1000-1800 nm^[1,2]. An effective refractive index $n_{\text{eff}}^2 = (n_o^2 + 2n_e^2)/3$ is used for the isotropic LCN under light irradiation. The simulated spectrum is obtained using fixed lattice deformation of 20% in both orthogonal directions. In the experiment, the laser excitation is induced by a focused 532 nm laser with 90 mW power and 700 μm focal spot.



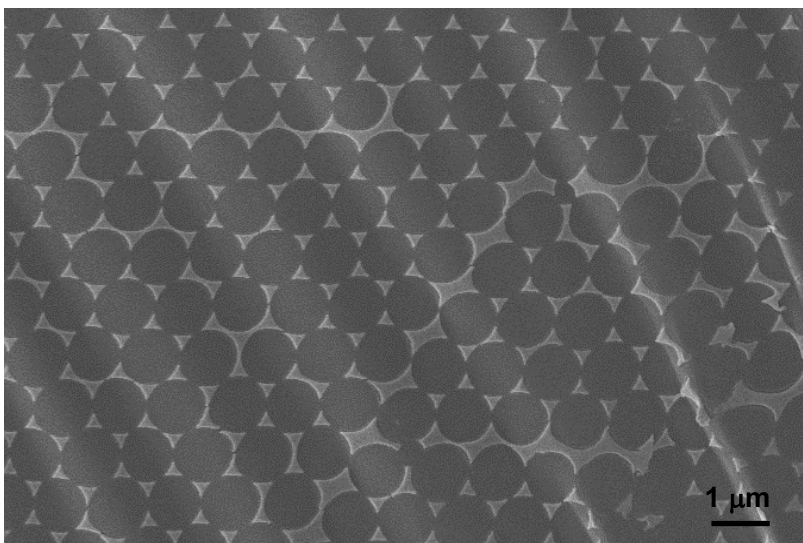
Supplementary Figure 10. Simulation of the spectral shifting by change of refractive index. The LCN substrate refractive index is tuned from anisotropic to isotropic conditions, while the metasurface lattice remains unchanged.



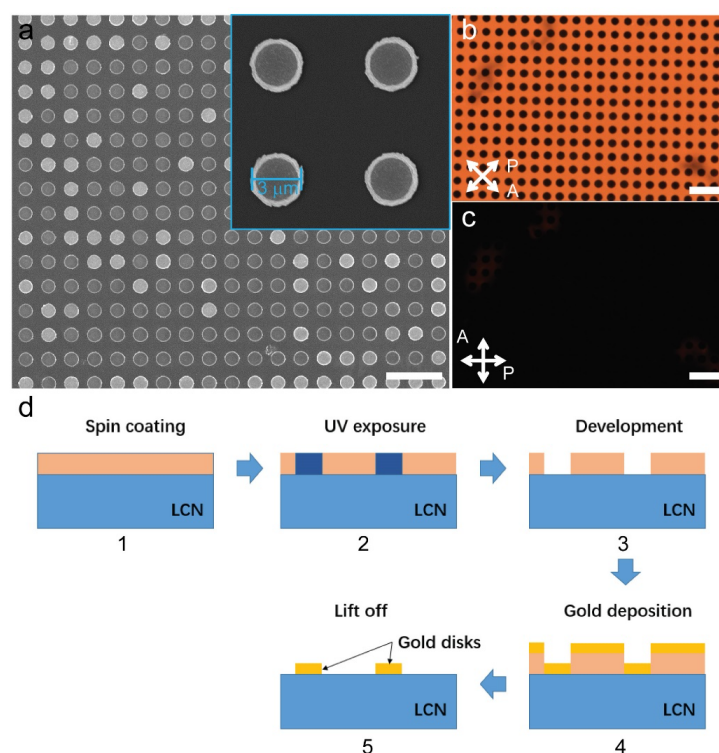
Supplementary Figure 11. Heat-induced spectral change. (a) Transmission spectra at different temperatures. **(b)** Center wavelengths of plasmon resonance extracted from (a) as a function of temperature.



Supplementary Figure 12. Simulation of far-field diffraction pattern. The results are based on FDTD simulation of undeformed metasurface lattice (top) and a deformed lattice (bottom) with 5% anisotropic strains.



Supplementary Figure 13. SEM image of a wrinkling LCN surface. The metasurface has been irradiated for about 30 min (532 nm, 90 mW, spot size 700 μm).



Supplementary Figure 14. Fabrication of microarray on a LCN substrate with standard UV lithography. (a) SEM image of a microdisk array on an LCN substrate. (b, c) Cross polarized optical microscopy images confirming the uniaxial alignment of the LCN substrate. Scale bars: 10 μm . (d) Schematic illustration of the fabrication process. Positive photoresist (RZJ-304) is spin-coated on the LCN substrate (1). The hole pattern is inscribed using masked UV exposure (2). The exposed areas are removed using RZX-3038 developer (3), after which 50 nm gold layer is evaporated onto the sample (4). After the lift off process using acetone, gold disk array fabricated on the LCN substrate was obtained (5).

Supplementary Table 1

Year	Mechanism	Functional material	Functionality	Wavelength	Refs.
2016	Electrically-tunable carrier doping	ITO	+1 order beam diffraction, ~ 17 nm resonance shift	Near IR, ~1550 nm laser	Nano Lett. 2016 ^[3]
2017	Electrically-tunable carrier doping	ITO	Transmittance decrease from 18% to 10%	Visible, ~670 nm	Adv. Mater. 2017 ^[4]
2017	Mechanical stretch	PDMS	~150 nm scattering shifting	Visible	Nano Lett. 2017 ^[5]
2018	MEMS Electromechanical	Dielectric medium	~8 mm focus length shifting,	Near IR, 915 laser	Nat. Commun. 2018 ^[6]
2018	Pump-probe	Ga-ZnO	15 nm shift of a Fabry–Pérot resonance	Near IR, ~1160 nm	Nano Lett. 2018 ^[7]
2018	Heating phase transition	VO ₂	~200 nm reflectance shifting	Near IR, 1100-1300 nm	ACS Photonics 2018 ^[8]
2019	Heating phase transition	GST	10% reflectance modulation	375-450 nm	Nano Lett. 2019 ^[9]
2019	Electrically-tunable	ITO	~ 17 nm resonance shift, Dynamic beam steering	Near IR, ~917 nm	Nat. Commun. 2019 ^[10]
2021	Heating phase transition	VO ₂	~40 nm absorption shifting	Near IR, 1300-1400 nm	ACS Photonics 2021 ^[11]
This work	All-optically photoelastic deformation	LCN	~250 nm resonance shifting Dynamic polarization	Near IR, 1200-1450 nm	

Materials and Methods

1. Simulation for transmission spectra, near-field electric field, and far-field diffraction

For all simulations, we used the finite-difference time-domain (FDTD) method. According to the geometric parameters from Supplementary Figure 5, we constructed the Au metasurface on the LCN substrate, with refractive index of gold taken from Ref. [12]. The refractive index of the anisotropic LCN was measured by spectroscopic ellipsometry, as shown in Supplementary Figure 7. The average index was used to simulate the isotropic LCN under the laser excitation^[1,2]. The transmission is calculated from the total power passing through the sample and divided by the incident power before arriving at the sample.

To simulate an unpolarized plane wave source, two simulations with orthogonal plane waves with 0° and 90° polarizations were taken. The transmission spectrum in Figure 2c in main text is the average from these two simulations. For the calculated electric field intensity in Figure 2b in the main text, the quantity $\langle |E| \rangle$ refers to the time averaged electric field intensity of an unpolarized beam source

$$\langle |E| \rangle = \sqrt{\frac{|E_0|^2 + |E_{90}|^2}{2}},$$

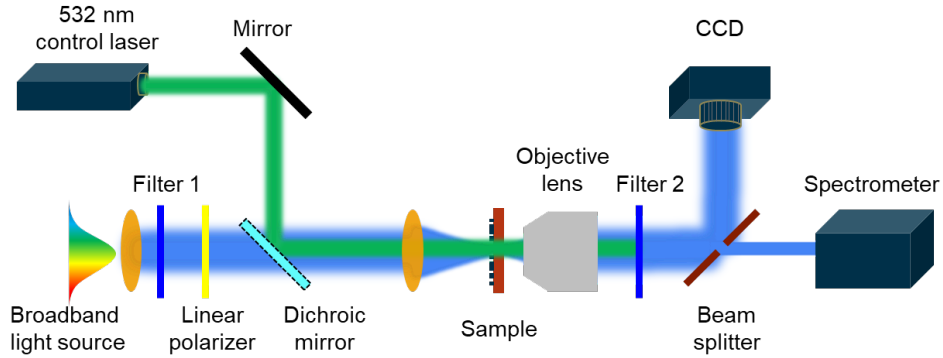
where E_0 and E_{90} are the field intensities of the Au nanoarray with polarization angle of incident source at 0° and 90°, respectively.

To simulate the far-field diffraction pattern, a profile and power monitor is used to record the transmitted data, which allow the electromagnetic fields to be calculated anywhere in the far field as a post-processing step. We assume that there are 100×100 periodic areas, and the light source is set as a plane wave with wavelength of 633 nm.

2. Measurement for the transmission spectrum

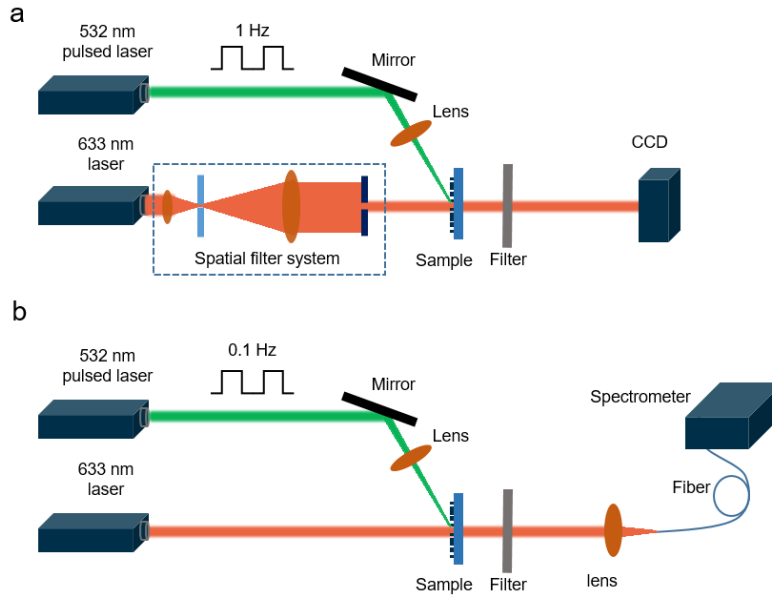
The transmission spectra were measured by an optimized microspectrometer (CRAIC Technologies), as shown in Supplementary Figure 15. A 70W xenon lamp (Olympus) was used as the broadband source. Filter 1 is a longpass filter with cut-off wavelength of 550 nm, which was used to avoid the photothermal effect of the light below 550 nm from xenon lamp onto LCN. Dichroic mirror with cut-off wavelength of 650 nm is used to reflect the control laser (continuous-wave, 532 nm) to the sample. Filter 2 is used to eliminate the control laser. A special beam splitter

took one part of the transmission to the spectrometer analysis while the other part to camera imaging.



Supplementary Figure 15. Schematic diagram of the experimental setup of the spectral measurements.

3. Measurements of the temporal response



Supplementary Figure 16. The optical setup for measurements of the temporal response. The optical setup for measurements of the far field diffraction pattern (a) and intensity modulation (b).

The setup for measuring diffraction and temporal response consists of a continuous-wave laser (633 nm), spatial filter, tunable pulsed laser (532 nm), focus lens, longpass filter of 550 nm, fiber coupled spectrometer, and CMOS camera. Spatial filter system in Supplementary Figure 16a

was used to obtain a high quality collimated probe beam for creating diffraction pattern from the metasurface.

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