

Supplementary Information

Electrically Modulated Plasmonic Metasurfaces for Light Communication

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51 **Summarized device performance of electrically modulated metasurfaces**

52 **Table S1. Summarized device performance of electrically modulated metasurfaces**

53

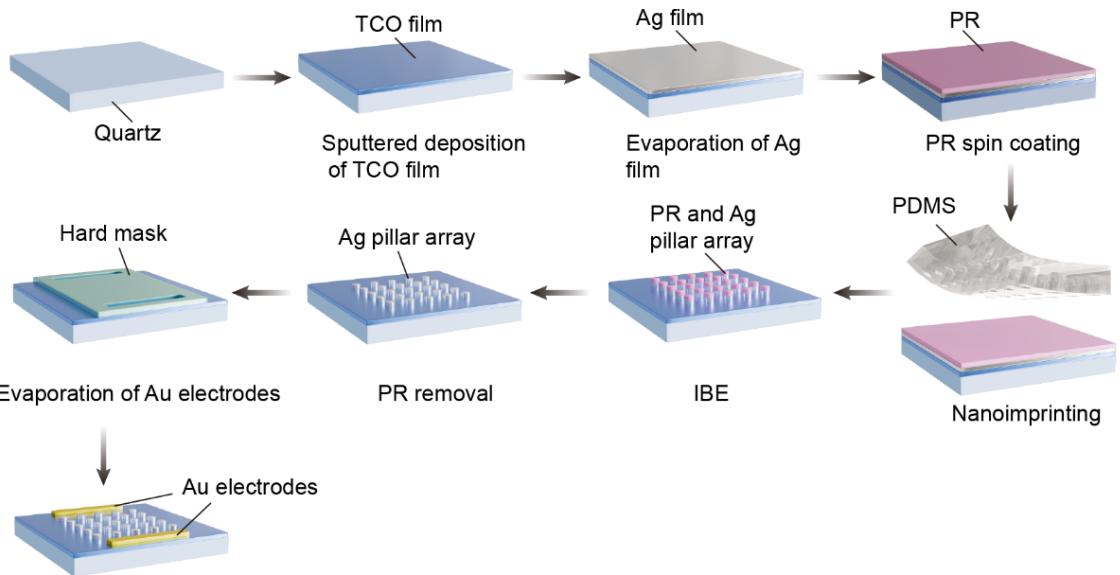
Materials	Wavelength (nm)	Linewidths (nm)	Operation Voltages (V)	Tuning Sensitivity (nm/V)	Modulation Depth	Modulation Speed	Ref.
Phase change material	1320	~200	15.7	-	-	-	[28]
Phase change materials	1430	266	20	8.75	400%	-	[30]
Organic materials	1480	~ 22	80	0.06875	37%	50MHz	[34]
Organic materials	1550	~ 50	60	0.1	67%	GHz	[35]
Organic materials	1500	~ 33	100	0.015	11%	GHz	[36]
Organic materials	1495	~ 5	17	0.16	-	MHz	[37]
Silicon	780	20	4	3.5	90%	100Hz	[38]
Lithium Niobate	968.5	~2	10	0.019	4.2%	kHz	[39]
Liquid crystals	1600	76	70	0.71	700%	-	[41]
Lithium Niobate	1550	26	20	0.07	40%	500 Hz	[42]
Graphene	6500	2000	10	10	100%	20 GHz	[43]
Organic materials	1630	32	10	0.05	0.8%	5 MHz	[44]
Lithium Niobate	1560	1	24	0.011	-	30 GHz	[45]
Liquid crystals	669	30	20	0.3	53%	-	[46]
Phase-change materials	1340	~ 0.1	7.5	0.05	-	-	[47]
DMSO and ITO	550-800	10	Below 5	~1	43%	Hz	This work

54

55

56 **Fabrication of electrically modulated metasurfaces**

57 Ag NP lattices and Al NP lattices on TCO-quartz substrates were fabricated using a
58 nanofabrication process that combines sputter deposition, electron-beam deposition, modified
59 solvent-assisted nanoimprint embossing (SANE), and etching¹. First, a 150 nm TCO layer is
60 deposited by sputtering, and then the Al film or Ag film is deposited by electron beam
61 evaporation, followed by photoresist (PR) S1805 spin-coated on the metal surface at 4000 rpm
62 with an acceleration of 500 rpm/s for 30s. Nanoimprinting is performed on the photoresist to
63 generate periodic PR columns with controlled size and period. The polydimethylsiloxane
64 (PDMS) mask was rinsed with alcohol and then immersed in dimethylformamide (DMF)
65 solution for 90 seconds. After drying with a nitrogen gun, it was pressed onto the sample coated
66 with S1805 and left to stand for 1.5 hours before being removed. Metal NP lattices are formed
67 by ion beam etching (IBE) and removing the PR with acetone. We performed IBE etching using
68 the HAASRODE-I200 etcher from Leuven Instruments. The etching gas Ar flow rate is 24
69 sccm, with an etching angle of 0 degree, a temperature of 7 °C, and a voltage of 400V. The
70 etching times for Al and Ag are 300 seconds and 96 seconds, respectively. Finally, the Au
71 electrodes were deposited via electron beam evaporation with a hard mask to protect the metal
72 NP array.

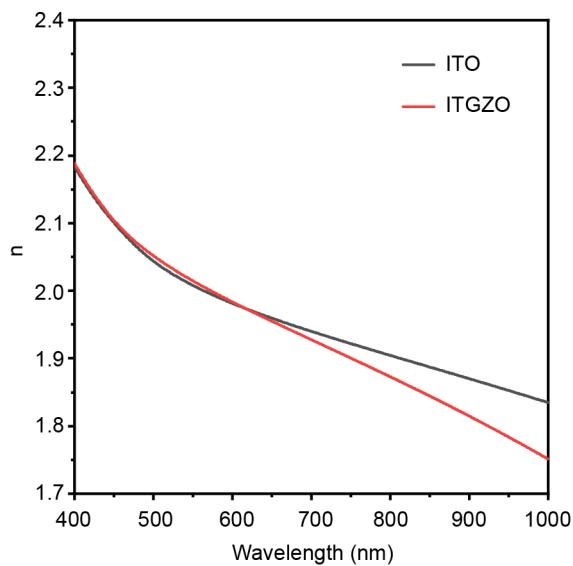


73

74 **Figure S1. Fabrication of electrically modulated metasurface.**

75

76 **Refractive indices of ITO and ITGZO**



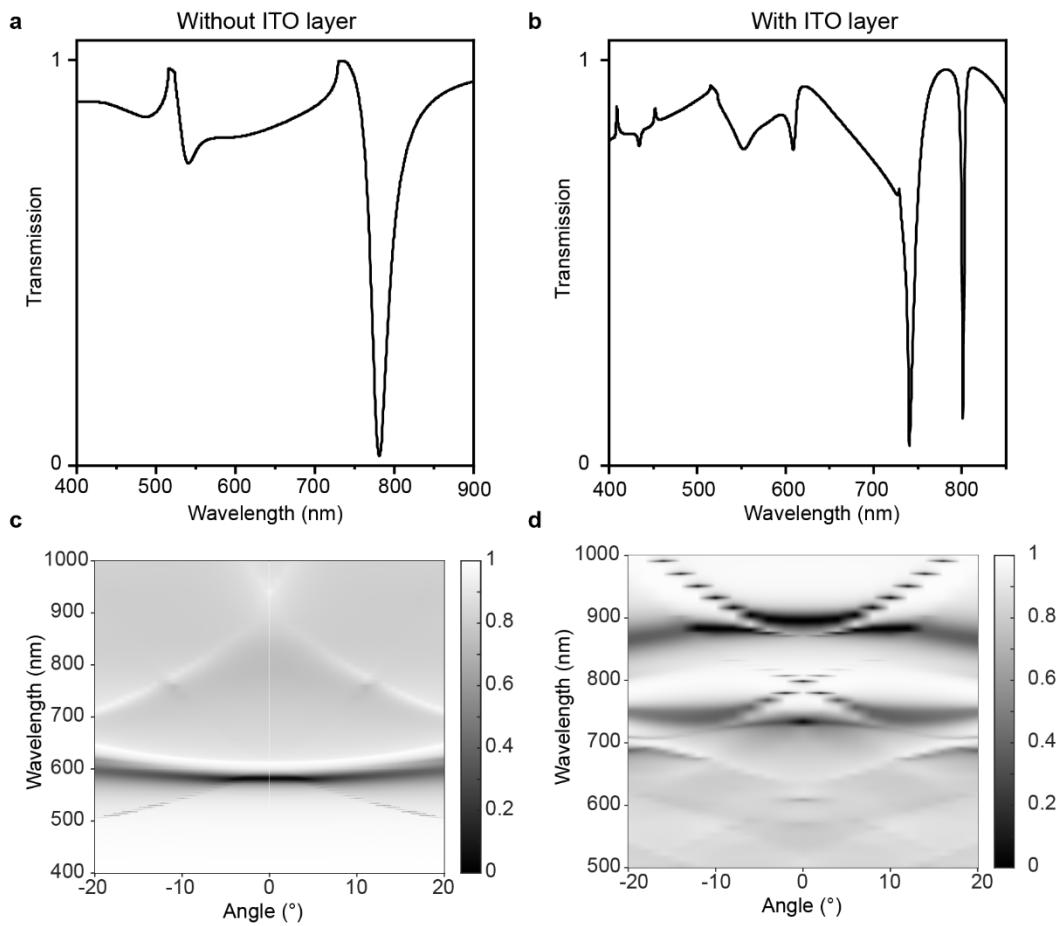
77

78 **Figure S2. Refractive indices of ITO and ITGZO from 400 nm to 1000 nm measured by**
79 **spectroscopic ellipsometry.**

80

81 **Resonance mode analysis of Ag and Al metasurfaces by the FDTD simulation**

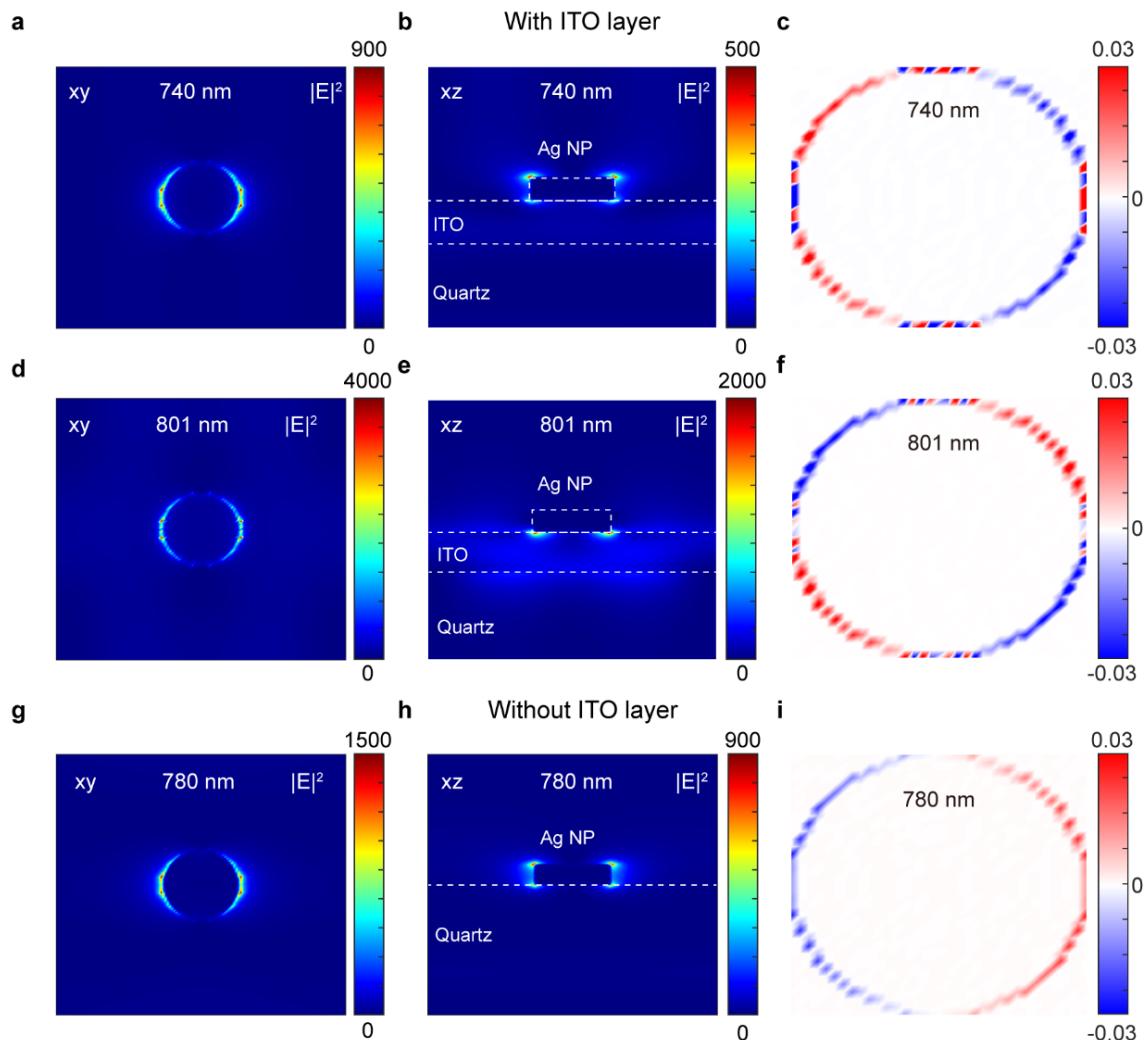
82 Finite-Difference-Time-Domain (FDTD) calculations were performed to simulate the
83 optical responses of Ag NPs lattices and Al NPs lattices on the TCO layer. Optical constants
84 for Ag and Al were taken from Palik measurements (400-1000 nm). These calculations used a
85 uniform lattice size of 4 nm (x, y, and z).



86
87 **Figure S3. Simulated transmission spectra and dispersion of the Ag metasurface with and**
88 **without the ITO layer.** Transmission spectra of Ag metasurface (a) without ITO layer and (b)
89 with ITO layer. Dispersion of Ag metasurface (c) without ITO layer and (d) with ITO layer.

90 We simulated the transmission spectra and dispersion of Ag NP lattices, both with and
91 without the ITO layer, using the FDTD method to illustrate the function of the ITO layer. The
92 period of the Ag NP lattices is 500 nm, the diameter is 130 nm, the height is 80 nm, and the
93 thickness of the ITO layer is 150 nm. Ag metasurface without ITO layer exhibits a single SLR

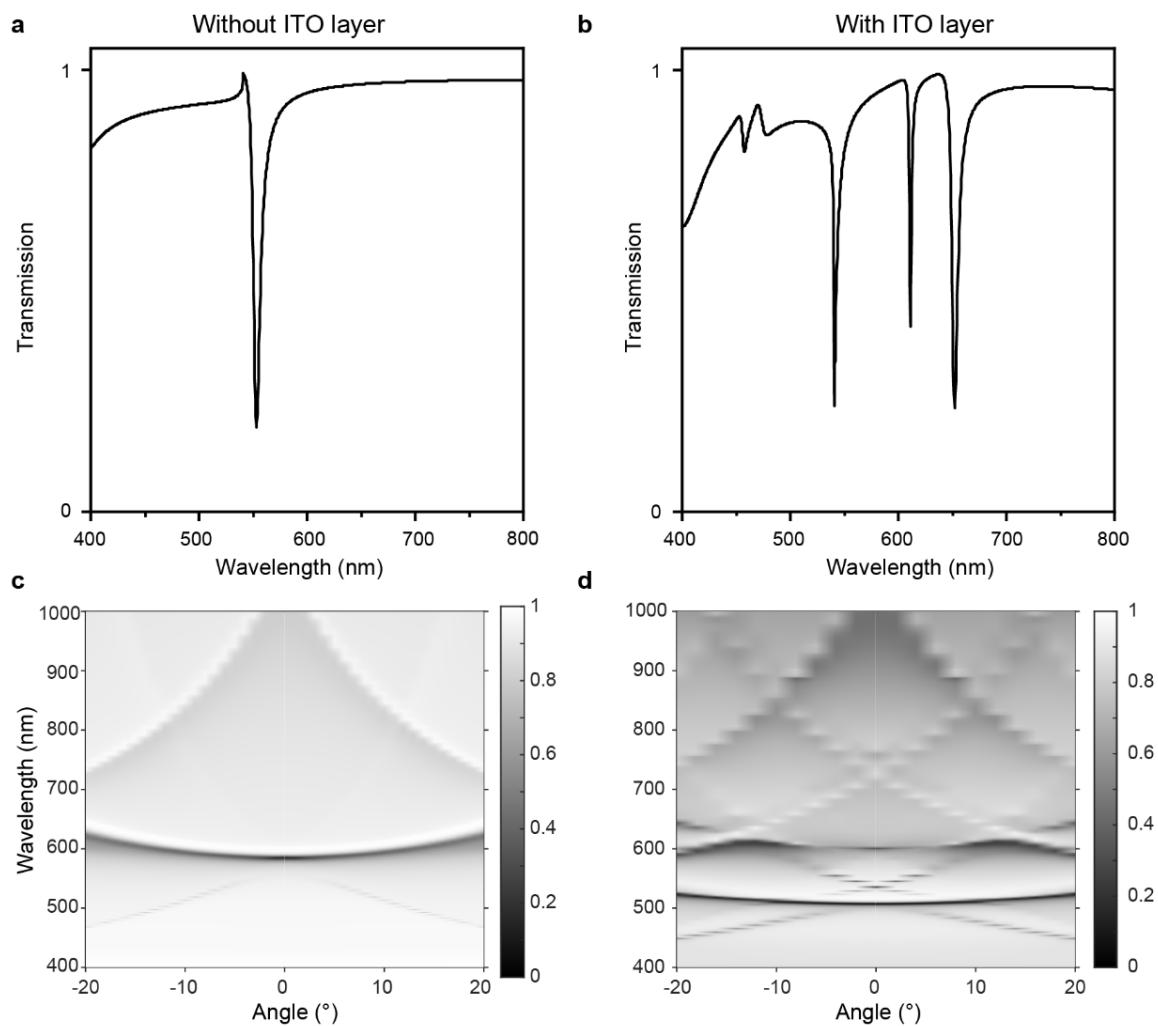
94 mode at 780 nm, while Ag metasurface with ITO layer exhibits multimodes at 740 nm and 801
 95 nm due to the waveguide modes generated by the high refractive index ITO layer coupled with
 96 SLRs².



97
 98 **Figure S4. Simulated near fields and charge distributions of Ag NP lattices at different**
 99 **modes with and without the ITO layer. (a-f)** Nearfield and charge distribution of two modes
 100 **of Ag metasurface with the ITO layer. (g-i)** Nearfield and charge distribution of Ag metasurface
 101 **without the ITO layer.**

102 The hot spots of the Ag metasurface without ITO layers are mainly concentrated on the
 103 upper and lower surfaces of the Ag NP, with a near-field enhancement of ~1500. We focus on
 104 two modes of Ag metasurface with the ITO layer: Mode 1 at 740 nm and Mode 2 at 801 nm.
 105 The hotspot of Mode 1 is mainly located on the upper surface of Ag, exhibiting a near-field

106 enhancement of ~ 900 , whereas the hotspot of Mode 2 is primarily situated at the interface
 107 between Ag NPs and the ITO layer, showing a near-field enhancement of ~ 4000 . Single mode
 108 without ITO layer exhibits dipole charge distributions, while mode 2 displays an in-plane
 109 quadrupole charge distribution with the ITO layer.



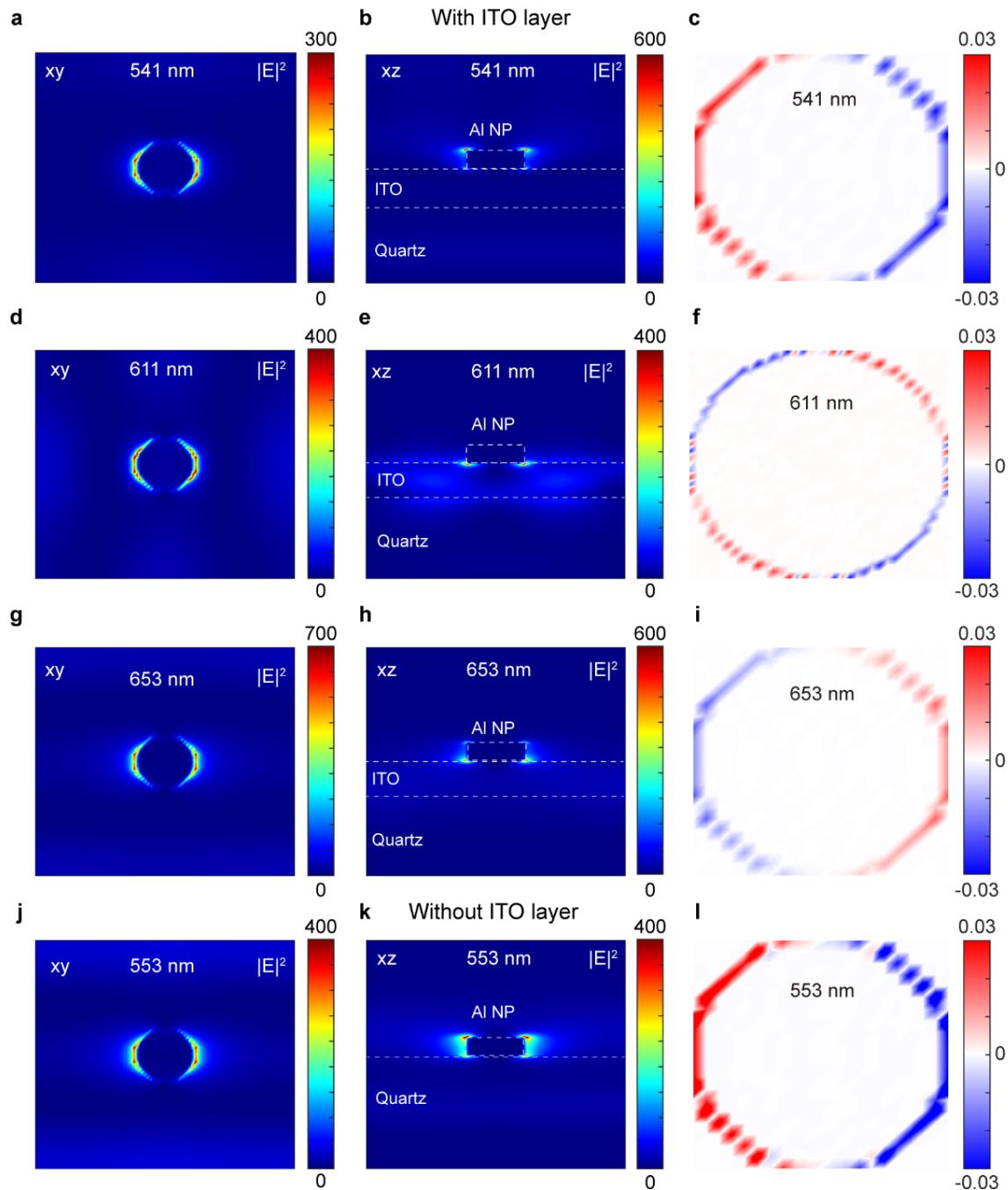
110
 111 **Figure S5. Simulated transmission spectra and dispersion of the Al metasurface with and**
 112 **without the ITO layer.** Transmission spectra of Al NP lattices (a) without ITO layer, and (b)

113 with ITO layer. Dispersion of Al NP lattices (c) without ITO layer, and (d) with ITO layer.

114

115 The period of the Al NP lattices is 370 nm, the diameter is 80 nm, the height is 80 nm, and
 116 the thickness of the ITO layer is 150 nm. An Al metasurface without an ITO layer exhibits a
 117 single SLR mode at 553 nm, while an Ag metasurface with an ITO layer exhibits multimodes

118 at 541 nm, 611 nm, and 653 nm due to the waveguide modes generated by the high refractive
 119 index ITO layer coupled with SLR.



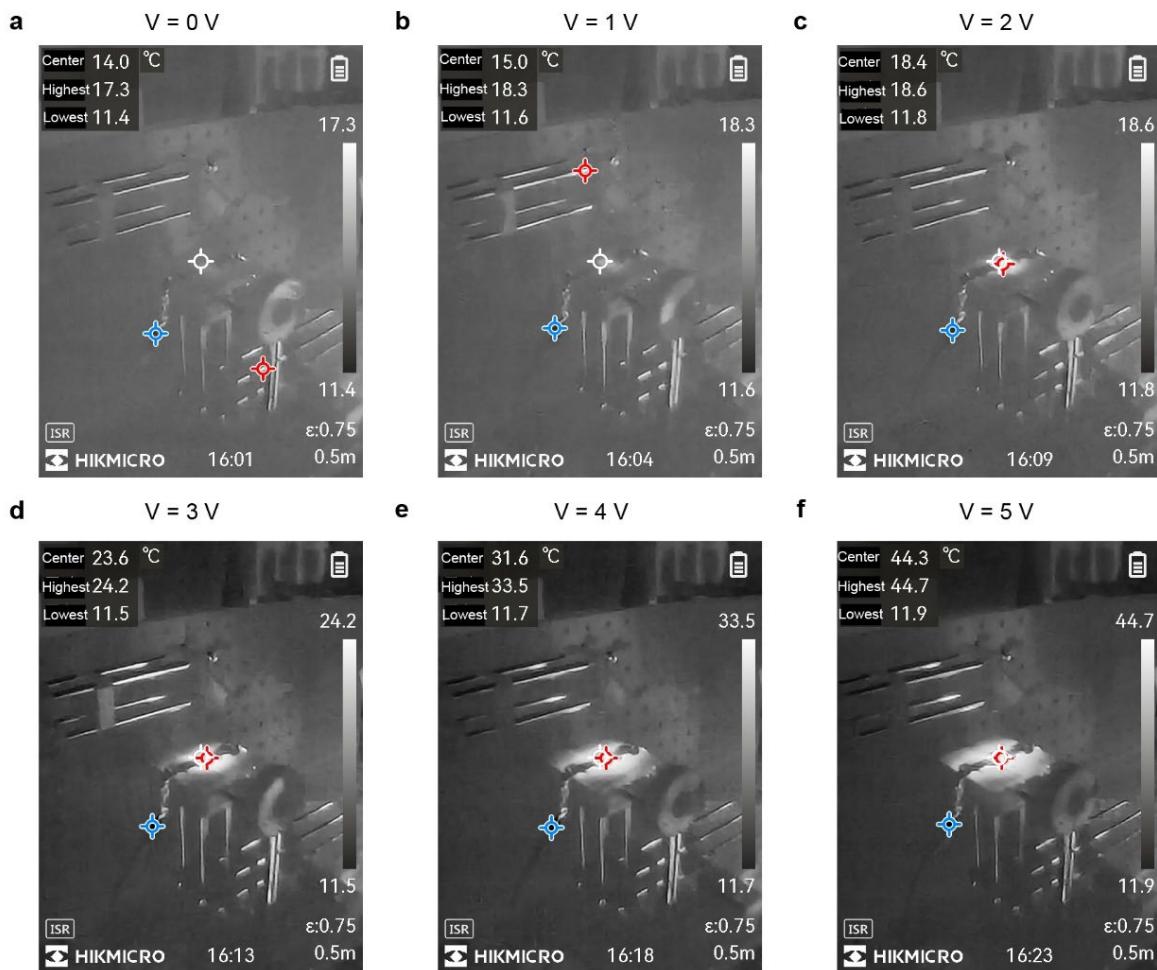
120
 121 **Figure S6. Simulated nearfield and charge distribution of Al NP lattices at different modes**
 122 **with and without the ITO layer.** (a-i) Nearfield and charge distribution of Al NP lattices at 3
 123 modes with the ITO layer. (j-l) Nearfield and charge distribution of Al NP lattices without the
 124 ITO layer.

125 The hot spots of the Al metasurface without ITO layers are mainly concentrated on the
 126 upper and lower surfaces of the Al NP, with a near-field enhancement of ~400. We concentrate

127 on three modes of Al metasurface with the ITO layer: Mode 1 at 541 nm, Mode 2 at 611 nm,
128 and Mode 3 at 653 nm. The hotspot of Mode 1 is mainly located on the upper surface of Al NP,
129 with a near-field enhancement of ~ 600 , while the hotspots of Mode 2 and mode 3 are primarily
130 positioned at the interface between Ag NPs and the ITO layer, with a near-field enhancement
131 of ~ 400 and ~ 700 , respectively. Mode 2 exhibits an in-plane quadrupole, while mode 1, mode
132 3, and the single mode without the ITO layer show in-plane dipole charge distribution.

133

134 **Temperature distribution of the Ag metasurface at different voltages measured by the**
135 **infrared thermal imaging camera**



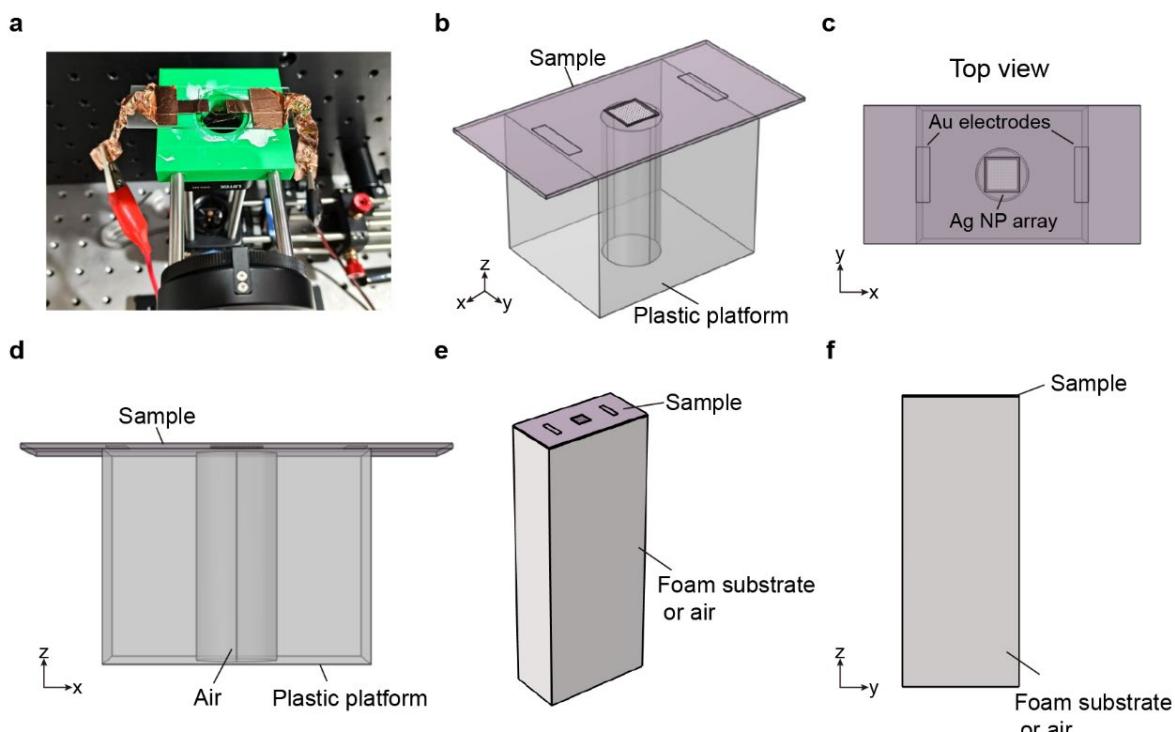
136
137 **Figure S7. Temperature distribution of the Ag metasurface at different voltages.** The white
138 circle indicates the location of the sample, the blue circle indicates the area with the lowest
139 temperature, and the red circle indicates the area with the highest temperature.
140

141 **Simulated temperature distribution of the sample on different substrates**

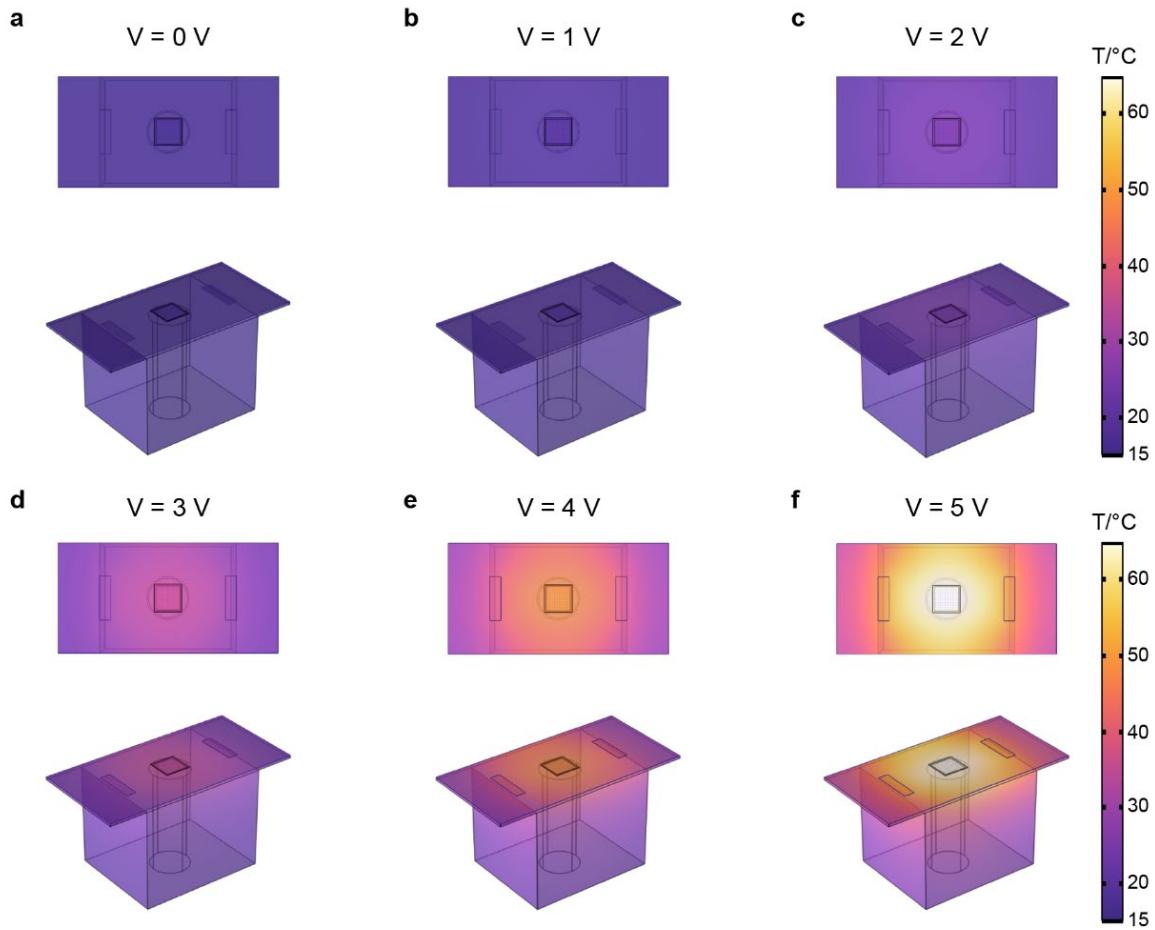
142 We simulate the temperature distribution of the device at different voltages by using the
143 current module with the solid-state heat transfer module in COMSOL. The model is founded
144 on the experimental setup employed for characterizing the modulation performance, with the
145 dimensions of the metasurface in the model aligned to those of the experimental samples, as
146 illustrated in **Figure S6a**. The samples are positioned on a plastic plate featuring a central
147 aperture. It is worth noting that since the size of Al NPs and ITO relative to the overall size is
148 very small, it makes it impossible to establish precise gridding. Consequently, these layers are
149 simplified to a thermally thin layer for the heat transfer analysis. In the current study section,
150 the conductivity of the Al is set as the contact impedance, while the thickness and the
151 conductivity of the ITO are determined by the electrical shielding. This approach overlooks
152 mesh accuracy concerns and yields a highly precise model. We simulated the temperature
153 distribution of the samples positioned on the experimental characterization platform, on foam,
154 and in air at varying voltages.

155 In the sample on the plastic plate, the material within the plastic holes of the test platform
156 is designed as air, thereby establishing heat conduction in the plastic and the holes. The
157 boundary conditions at the bottom of the air column and the bottom of the plastic are set as
158 natural convection boundary conditions. For the sample on the foam, periodic boundary
159 conditions are used around the foam, and natural convective boundary conditions are applied
160 to the bottom of the foam substrate. Natural convective boundary conditions of $T_{e0} = 273 +$
161 $15K$ are used around and above the quartz. The electrical boundary conditions for all three
162 models are electrically insulating due to the substrate approximation of the insulator. The area

163 of the gold electrode without thickness was set as the input of the current, and the temperature
164 distribution of the sample at different voltages was obtained by parameter scanning for voltages
165 from 0 V to 5 V.



166
167 **Figure S8. Modeling of samples placed on different substrates.** (a) Photo of the
168 experimental setup. (b) 3D model and (c) top view, and (d) XZ view of the sample on the
169 plastic platform. (e) 3D model and (f) YZ view of the sample on the foam substrate or air.
170

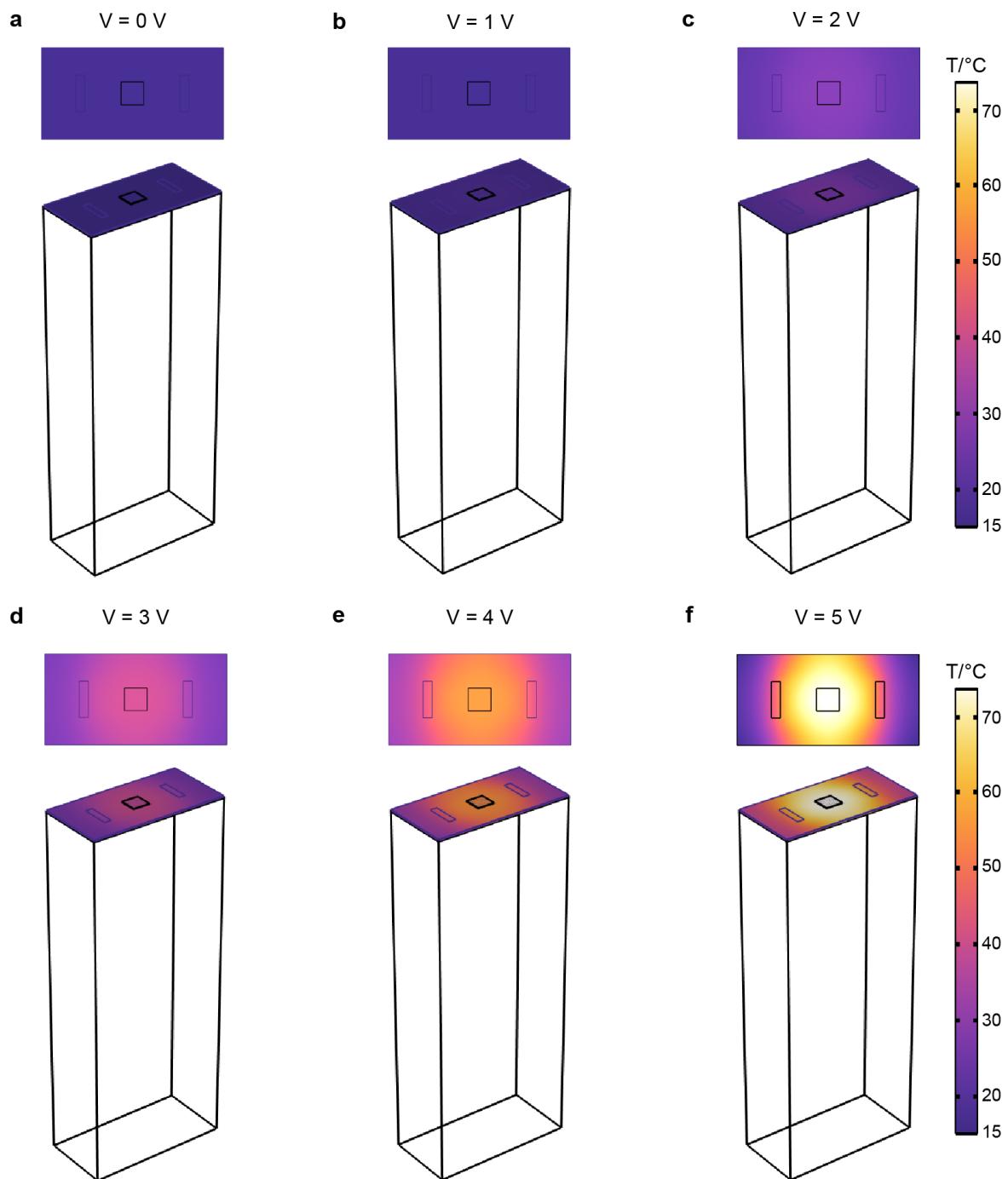


171

172 **Figure S9 Simulated temperature distribution of the sample on the plastic platform with**
 173 **different voltages by COMSOL.** Temperature distribution of the sample at (a) 0 V, (b)

174 (c) 2 V, (d) 3 V, (e) 4 V, and (f) 5 V.

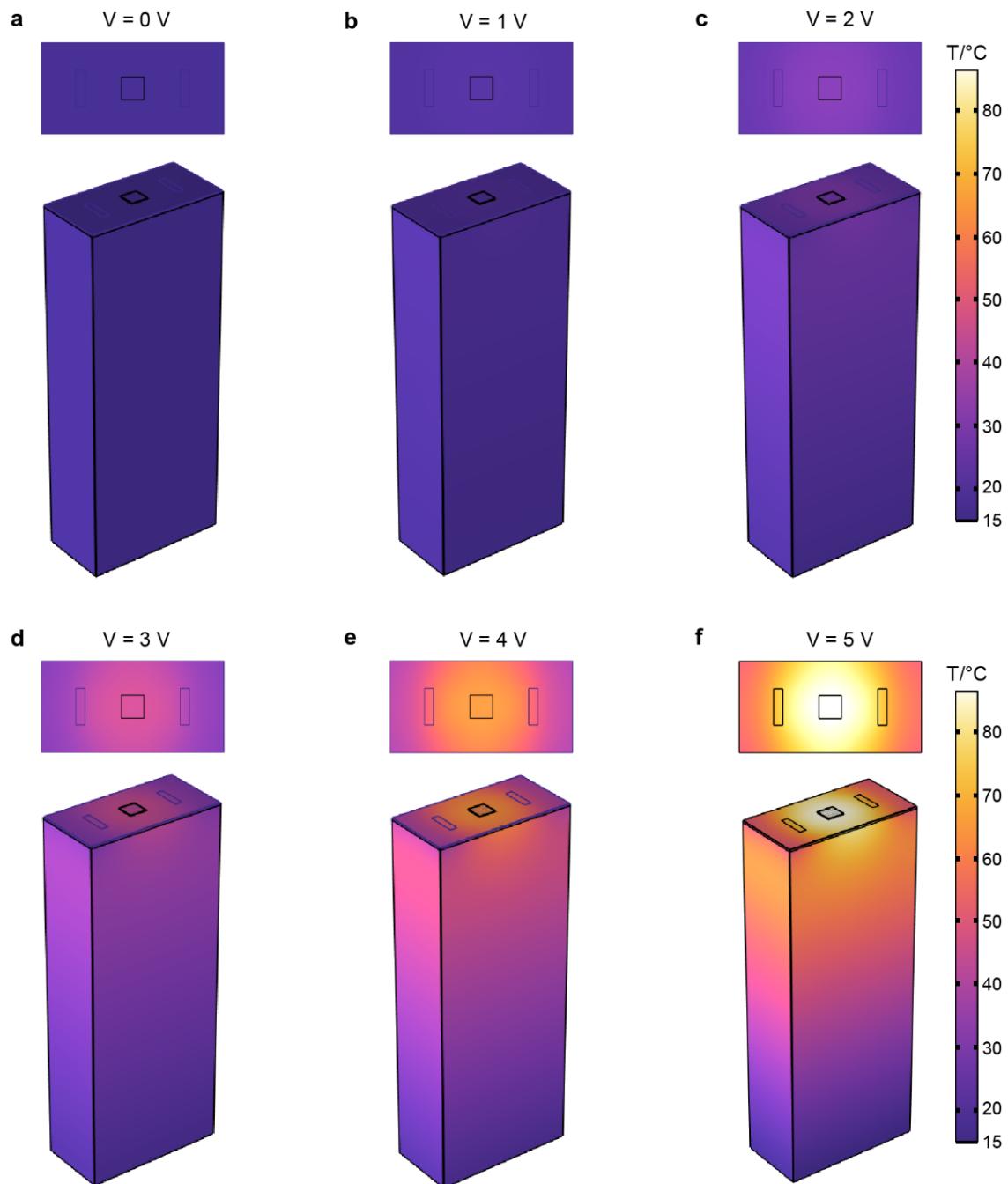
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176

177 **Figure S10. Simulated temperature distribution in air with different voltages by**
 178 **COMSOL.** Temperature distribution of the sample at **(a)** 0 V, **(b)** 1 V, **(c)** 2 V, **(d)** 3 V, **(e)**
 179 and **(f)** 5 V.

180



181
182
183
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185

Figure S11. Simulated temperature distribution of the sample placed on the foam substrate at different voltages. Temperature distribution of the sample at (a) 0 V, (b) 1 V, (c) 2 V, (d) 3 V, (e) 4 V, and (f) 5 V.

186 **The refractive index of DMSO at different temperatures measured by the Abbe
187 refractometer**

188 **Table S2. RI of DMSO at different temperatures**

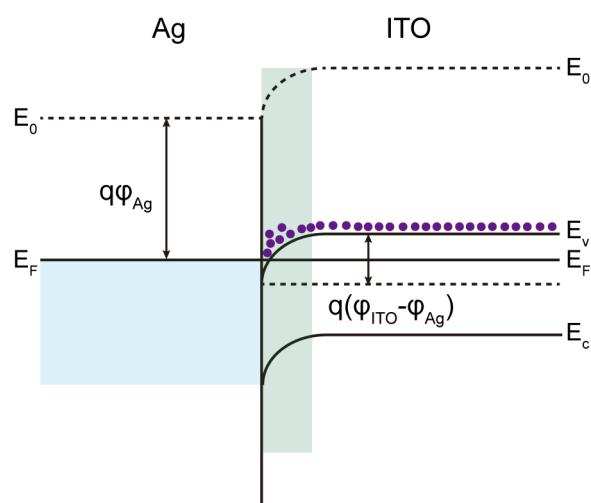
Temperature (°C)	RI of the DMSO
10	1.4789
20	1.4755
30	1.4715
40	1.4681
50	1.4642
60	1.4598

189 We measured the refractive index of dimethyl sulfoxide (DMSO) at various temperatures
190 using an Abbe refractometer. The maximum measurement temperature was limited to 60 °C,
191 as the optical components of the Abbe refractometer may experience thermal expansion, which
192 could result in inaccurate refractive index measurements.

193

194 **The energy band and electric field distribution of the Ag metasurface**

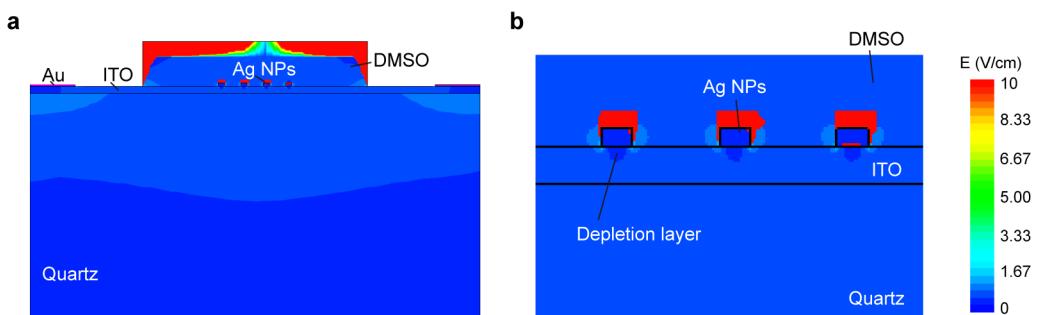
195 We present the energy band diagram for the Ag/ITO interface without the voltage. Here,
 196 E_0 and E_F denote the vacuum energy level and the unified Fermi level of Ag/ITO, respectively.
 197 E_c and E_v represent the conduction band edge and valence band edge energies of ITO,
 198 respectively. $q\phi_{ITO}$ and $q\phi_{Ag}$ denote the work function of the ITO and Ag, respectively.
 199



200

201 **Figure S12. Energy band diagram of the Ag NP array and ITO layer without bias.**

202



203

204 **Figure S13. Simulated electric field distribution of the Ag metasurface. (a)** Electric field
 205 of the Ag NP lattices on the ITGZO layer in the xz plane with a voltage of 5 V. **(b)** Magnified
 206 view of the interface between the Ag NPs and the ITO layer. A depletion layer forms between
 207 the Ag array and the ITO layer. Upon applying voltage, there is virtually no electric field along
 208 the z-direction between Ag and ITO, indicating minimal electron exchange between ITO and
 209 Ag.

210

211

212 We conducted a simulation to analyze the electric field distribution of the Ag metasurface

213 in the xz plane after applying a 5V voltage. The results indicate that voltage application alone,
214 without considering temperature, does not affect electron concentration in either the Ag (Al) or
215 ITO (ITGZO) layers. Therefore, we propose that the electric field generated by the Seebeck
216 effect results in localized variations in electron concentration within ITO. This phenomenon
217 accounts for the greater experimental shift observed, which exceeds the predictions made by
218 the simulation at the same temperature variation.

219

220 **Simulated effect of Ag plasma frequency on transmission spectra**

221 The optical resonance of Ag can be modeled by the Drude model, given by

222

$$\varepsilon_{Al} = \varepsilon_{inf} - \frac{\omega_p^2}{(\omega^2 + i\gamma\omega)}$$

223 Where ε_{inf} denotes the dielectric constant of metals at infinite frequency, ω_p and γ
224 represent the plasma frequency and scattering frequency, respectively. The plasmonic
225 resonance frequency of the metal can be expressed as:

226

$$\omega_p = \sqrt{\frac{Ne^2}{\varepsilon_0 m^*}}$$

227 The instinct plasmonic resonance frequency of Ag material is 1.37×10^{16} rad/s without the bias
228 voltage, with $\varepsilon_{inf} = 3.7$, $\gamma = 0.24 \times 10^{14}$ rad/s^{3,4}. To demonstrate the effect of increasing
229 the electron concentration of Ag NPs surface on transmission spectral shifts, we simulated the
230 transmission spectra at different plasmonic resonance frequencies ω_p of Ag NP ranging from
231 1.37×10^{16} rad/s to 1.4×10^{16} rad/s. A $\Delta \omega_p$ of 0.03×10^{16} rad/s results in a blue shift of ~ 2.3
232 nm for mode 1 and ~ 1.2 nm for mode 2, respectively.

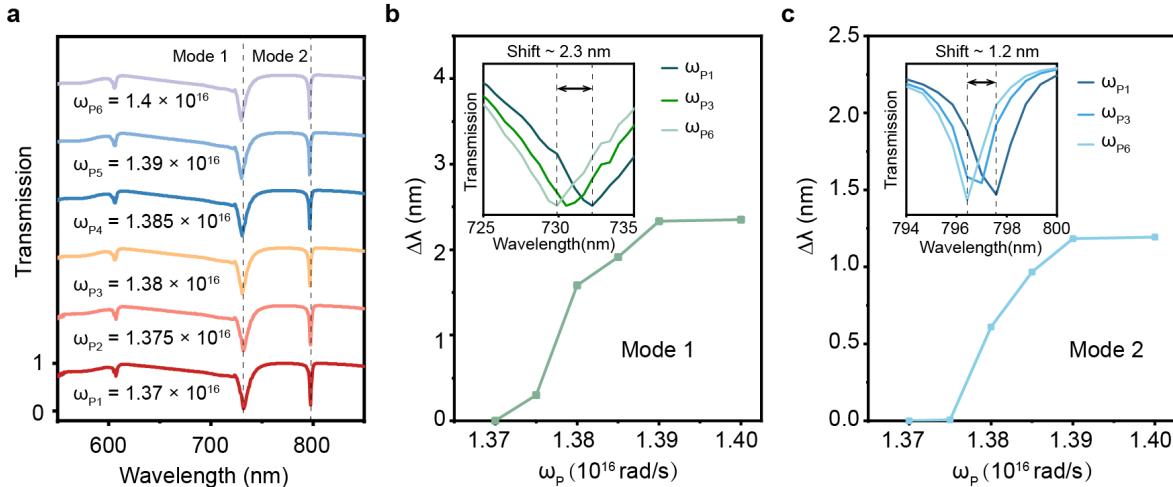


Figure S14. Simulated transmission spectra at different Ag plasma resonance frequencies.

(a) Transmission spectra with Ag plasma resonance frequency from ω_{p1} to ω_{p6} . Wavelength shifts with the increase of ω_p in (b) mode 1. (c) mode 2. The insets in (b-c) show the shifts of transmission spectra in mode 1 and mode 2, with ω_{p1} , ω_{p3} , and ω_{p6} .

Simulated transmission spectra of the metasurface within the communication band by FDTD simulation

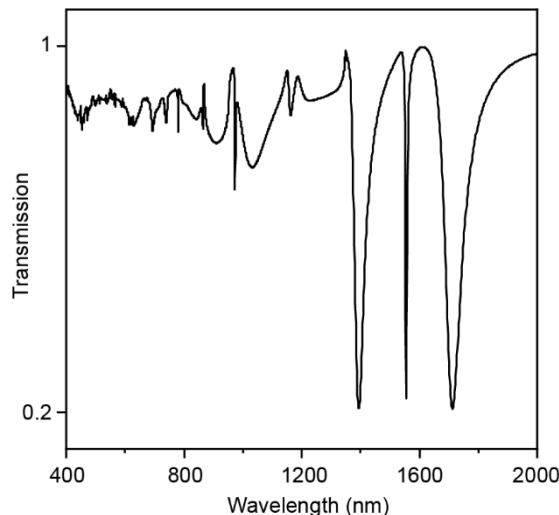


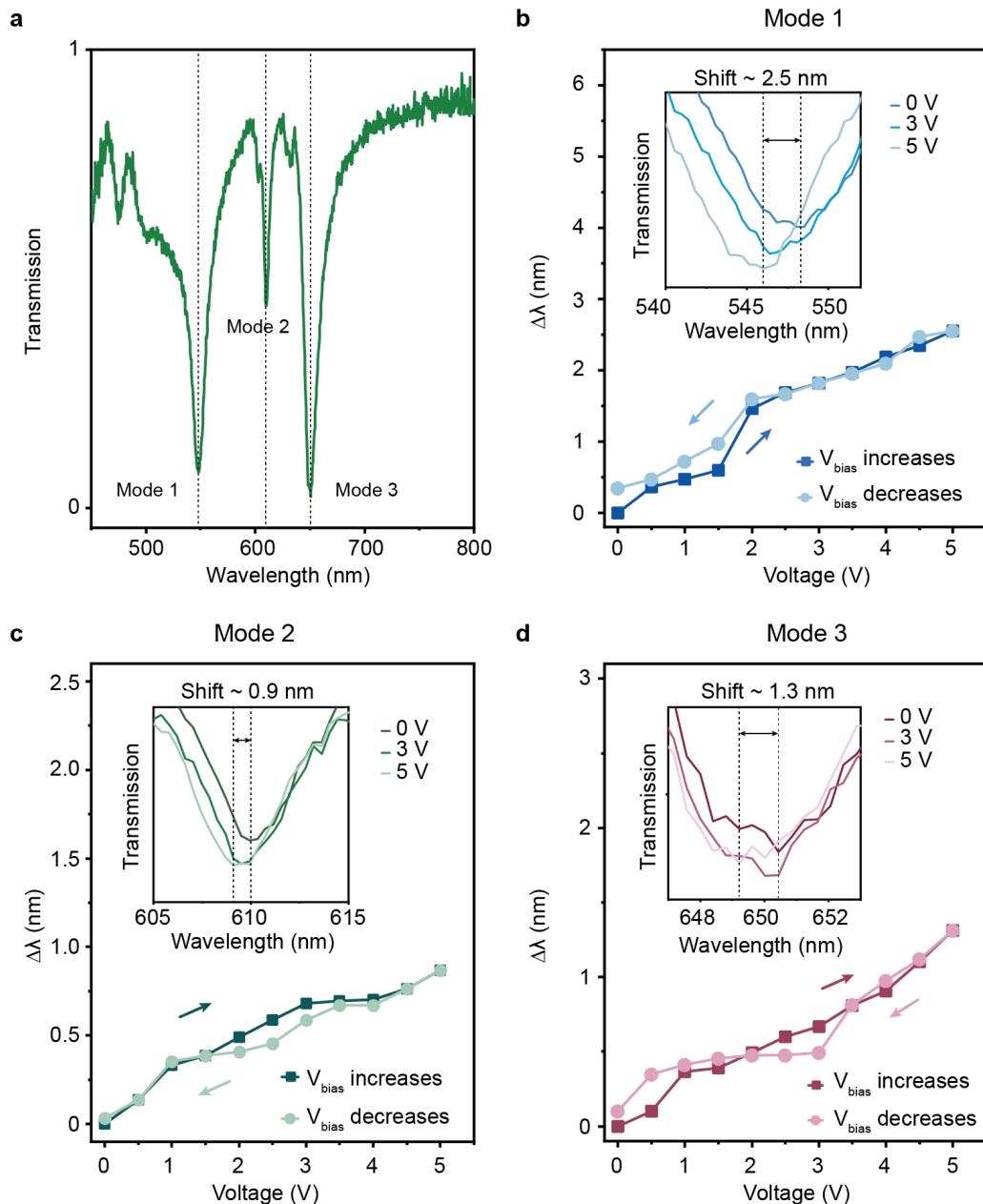
Figure S15. Simulated transmission spectra of the Ag metasurface within the communication band.

This electrically tunable metasurface can be equally suitable for the communication band.

The metasurface is engineered with a transmission spectrum wavelength peak of 1550 nm, an Ag metasurface period of 950 nm, Ag NPs diameter of 300 nm, heights of 100 nm, and an ITO thickness of 150 nm.

249 **Electrical modulation performance of the Al metasurface with the ITGZO layer**

250 The period of the Al metasurface is 370 nm, the diameter of the Al NP is 80 nm, the height
 251 is 80 nm, and the thickness of the ITGZO is 150 nm, with Mode 1 at 548 nm, Mode 2 at 613
 252 nm, and Mode 3 at 650 nm.



253
 254 **Figure S16. DC modulation performance of Al NP lattices with the ITGZO layer.** (a) The
 255 experimental transmission spectra of Al metasurface. The variation of shifts with voltages from
 256 0 V to 5 V of (b) mode 1, (c) mode 2, and (d) mode 3. The insets show the transmission spectra
 257 of mode 1, mode 2, and mode 3 at 0V, 3V, and 5V in (b-d). (e) Electrically modulated
 258 transmission spectra of the Al metasurface with bias voltages ranging from 0 V to 5 V. The
 259 variation of shifts with temperatures from 11 °C to 44 °C of (f) mode 1, (g) mode 2, and (h)
 260 mode 3.

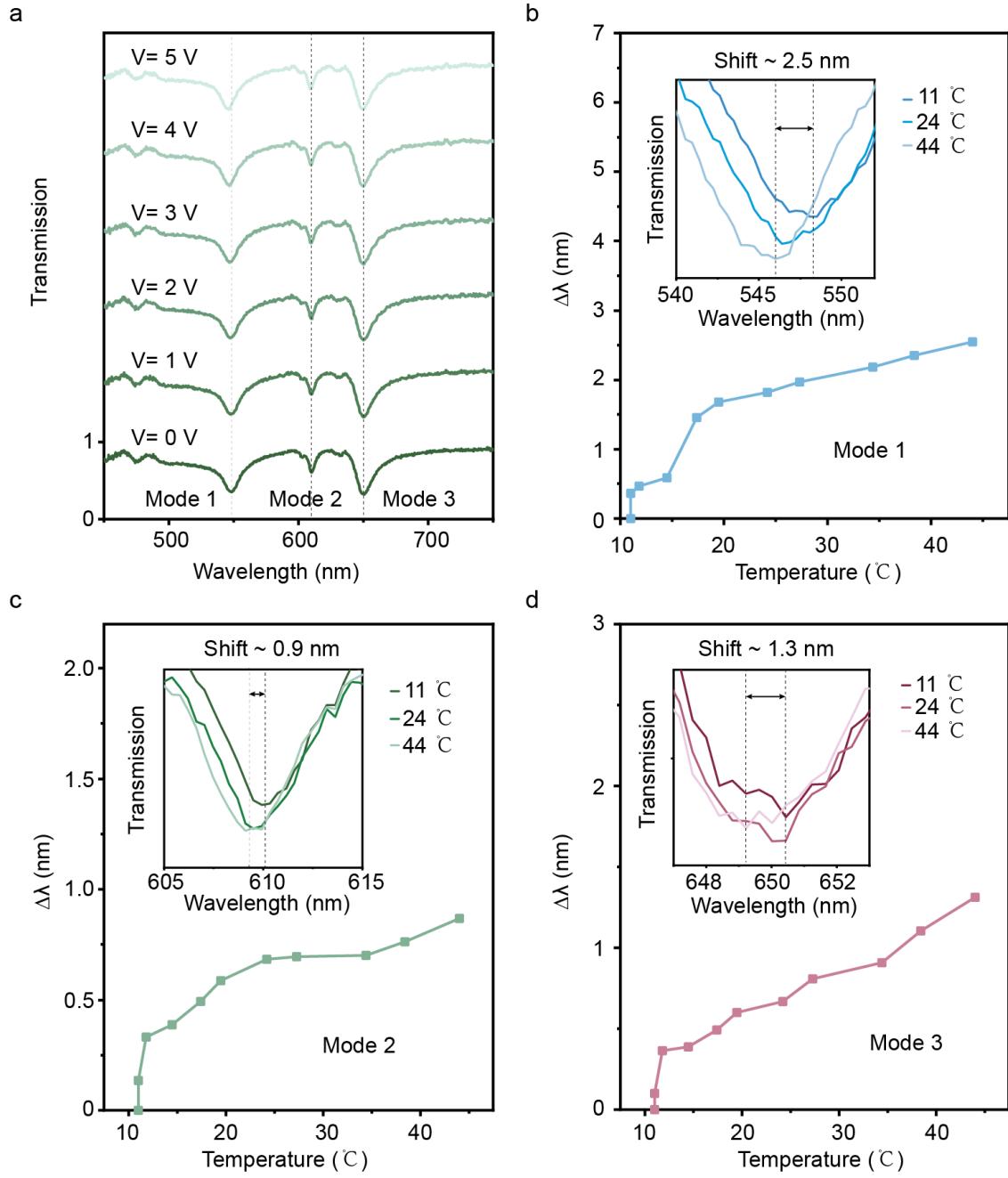


Figure S17. Electrical modulation performance of Al NP lattices with the ITGZO layer.
(a) The experimental transmission spectra of Al metasurface at different voltages. The variation of shifts with temperatures from 11 °C to 44 °C of **(b)** mode 1, **(c)** mode 2, and **(d)** mode 3. The insets show the transmission spectra of mode 1, mode 2, and mode with the temperature of 11 °C, 24 °C, and 44 °C in **(b-d)**.

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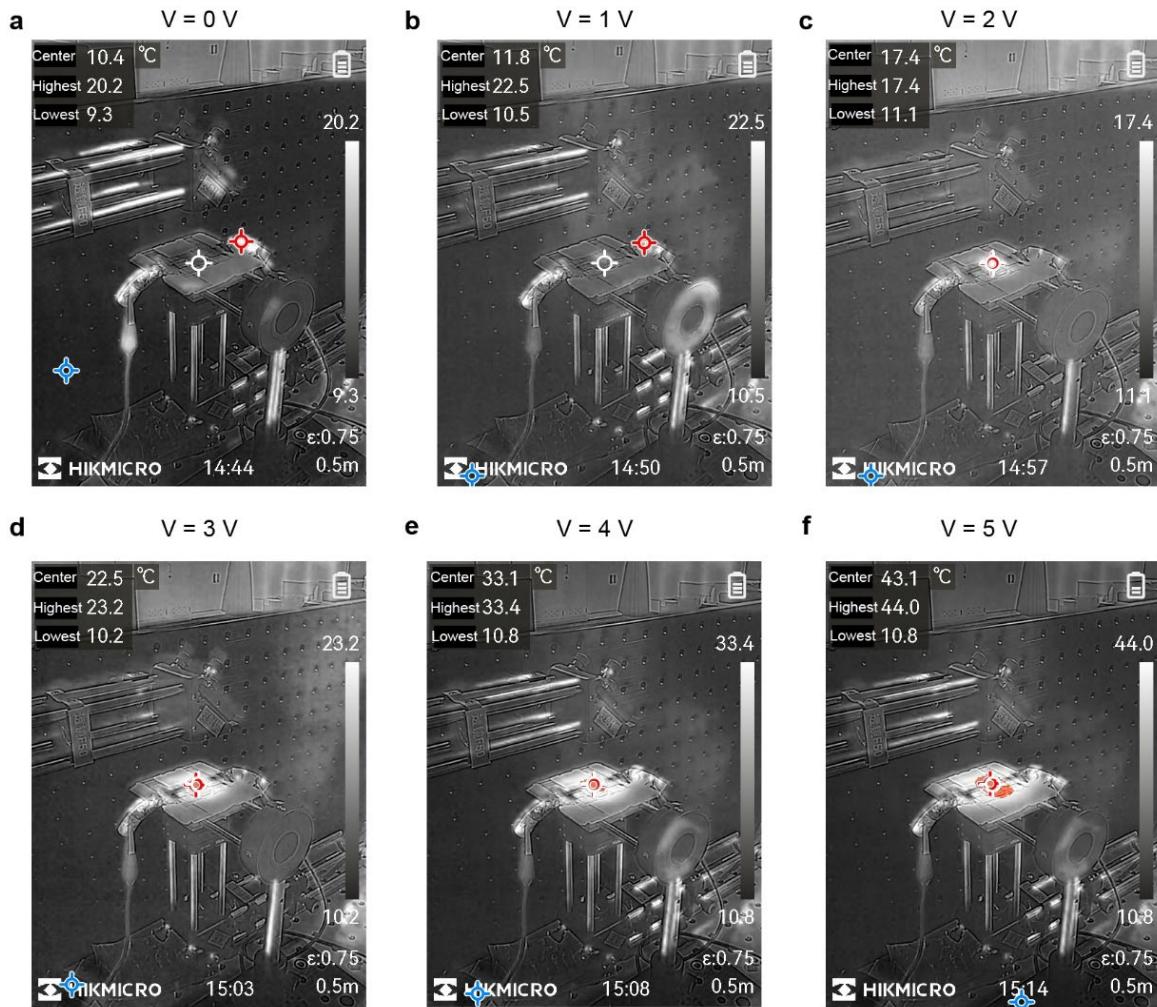
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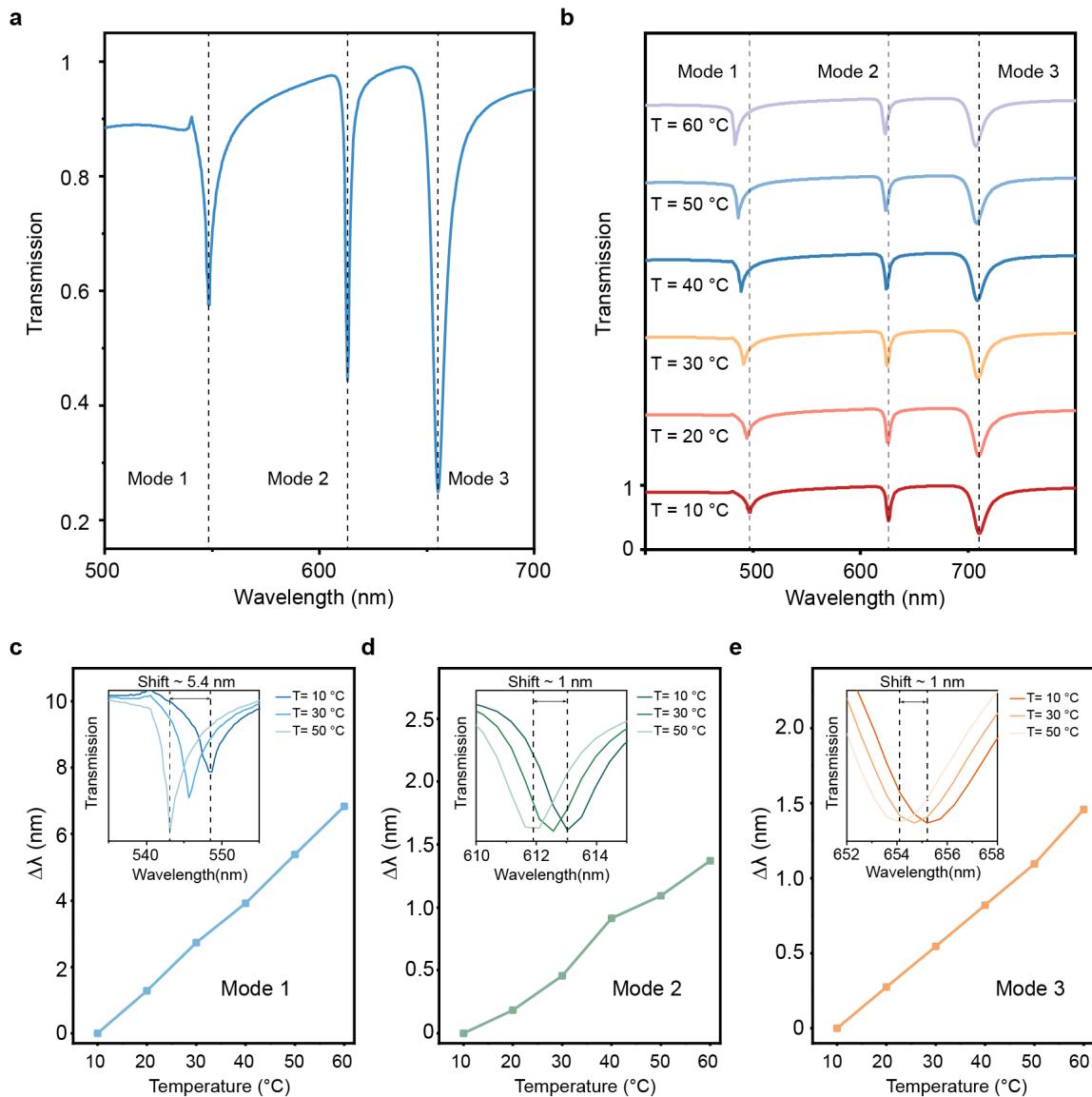
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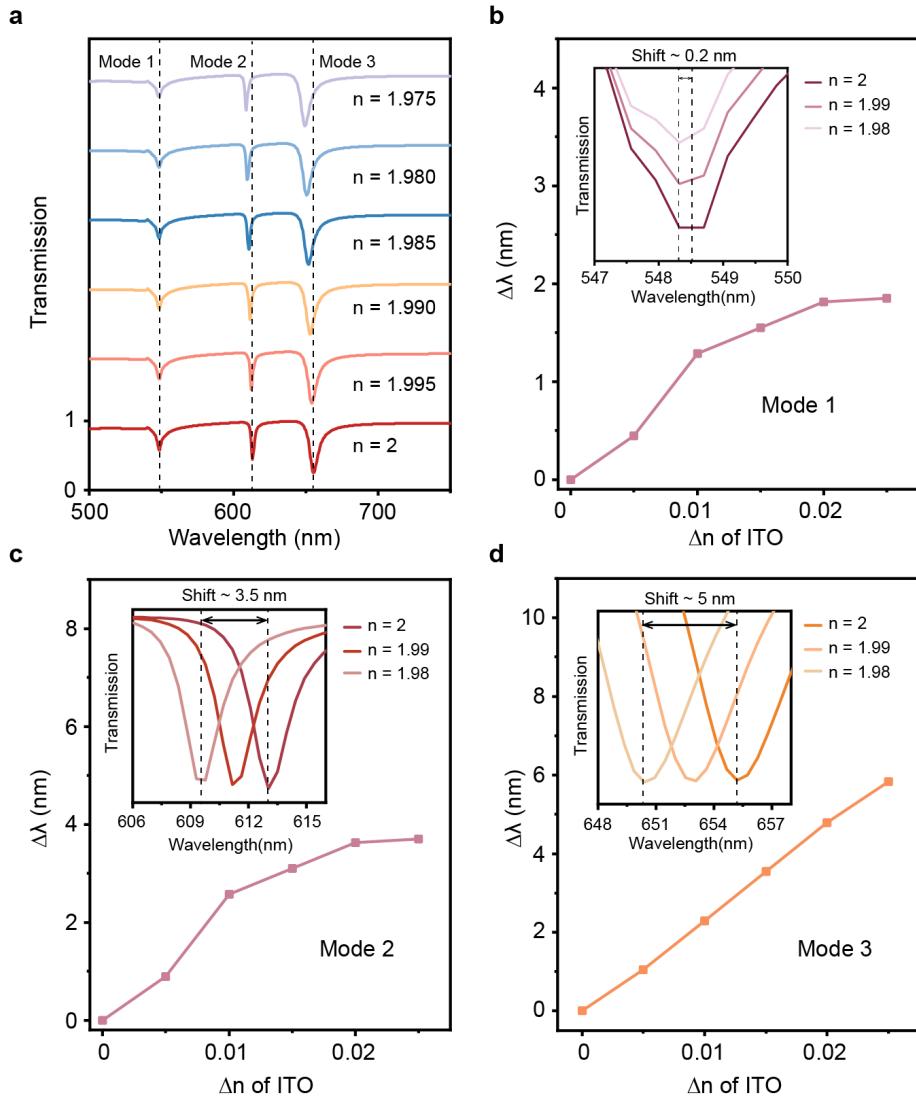
269 **Figure S18. Temperature distribution of the Al metasurface at different voltages**
270 **measured by the infrared thermal imaging camera.** The white circle indicates the location
271 of the sample, the blue circle indicates the area with the lowest temperature, and the red circle
272 indicates the region with the highest temperature.

273



275
276 **Figure S19. Simulated transmission spectra of the Al metasurface with different**
277 **temperatures. (a)** Simulated transmission spectra of the Al metasurface with 3 modes (gray
278 dashed lines). **(b)** Simulated transmission spectra with temperatures varying from 10 °C to
279 60 °C. The gray dashed lines label the initial wavelength peaks of three modes. The variation
280 of shifts with temperature from 10 °C to 60 °C of **(c)** mode 1, **(d)** mode 2, and **(e)** mode 3.

281 The Al metasurface has a period of 370 nm, an Al NP diameter of 80 nm, a height of 80
282 nm, and an ITGZO thickness of 150 nm, with mode 1 at 541 nm, mode 2 at 611 nm, and mode
283 3 at 653 nm. All three modes exhibit blue shifts with the temperature ranging from 10 °C to
284 60 °C, with distinct amounts of shifts. When the temperature was increased from 10 °C to 60 °C,
285 mode 1 exhibited a blue shift of ~ 5.4 nm, and modes 2 and 3 exhibited a blue shift of ~ 1 nm.

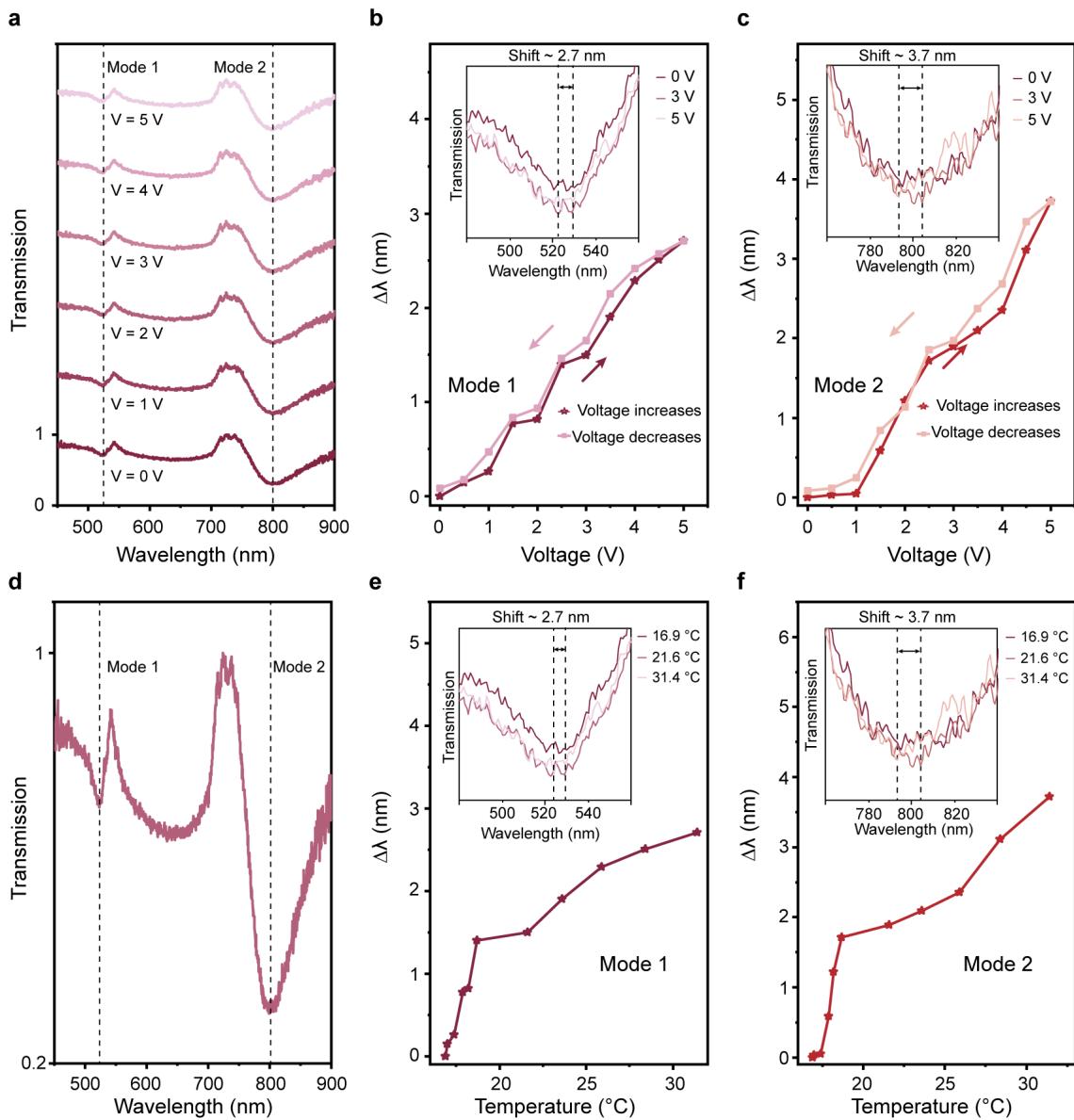


286

287 **Figure S20. Simulated transmission spectra of the Al metasurface at different refractive**
 288 **indices of ITO. (a)** Simulated transmission spectra with the refractive index of ITO varying
 289 from 2 to 1.97. The gray dashed lines label the initial wavelength peaks of three modes. The
 290 variation of shifts with Δn from 0 to 0.025 of (b) mode 1, (c) mode 2, and (d) mode 3. The
 291 insets show the transmission spectra with the refractive index of ITO of 2, 1.99, and 1.98 of 3
 292 modes in (b-d). The gray dashed lines show the largest shifts of the three modes.

293

294 **The electrical modulation performance of the Al metasurface and the Ag metasurface**
 295 **with the ITO layer**



296
 297 **Figure S21. Electrical modulation performance of the Al metasurface with the ITO layer**
 298 **with period of 500 nm.** (a) Electrically modulated transmission spectra of the Al metasurface
 299 with voltages from 0 V to 5 V. The gray dashed lines label the wavelength peak of mode 1 and
 300 mode 2. Resonance shifts with the voltage increases (dark line) and the voltage decreases (light
 301 line) of (b) mode 1 and (c) mode 2. The inset in (b-c) shows the transmission spectra with the
 302 bias voltage of 0 V, 3 V, and 5 V. (d) Transmission spectra of the Al metasurface at 0 V with 2
 303 modes (gray dashed lines). The variation of shifts with temperatures corresponding to the
 304 voltages from 16.9 °C to 31.4 °C of (e) mode 1 and (f) mode 2. The insets display the
 305 transmission spectra with the temperatures of 16.9 °C, 21.6 °C, and 31.4 °C. The gray dashed
 306 lines show the largest shifts of the two modes.

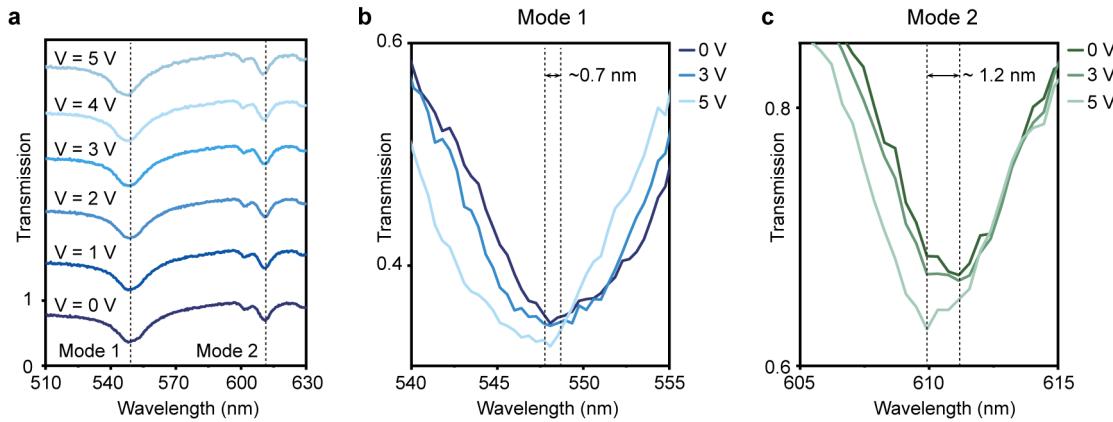
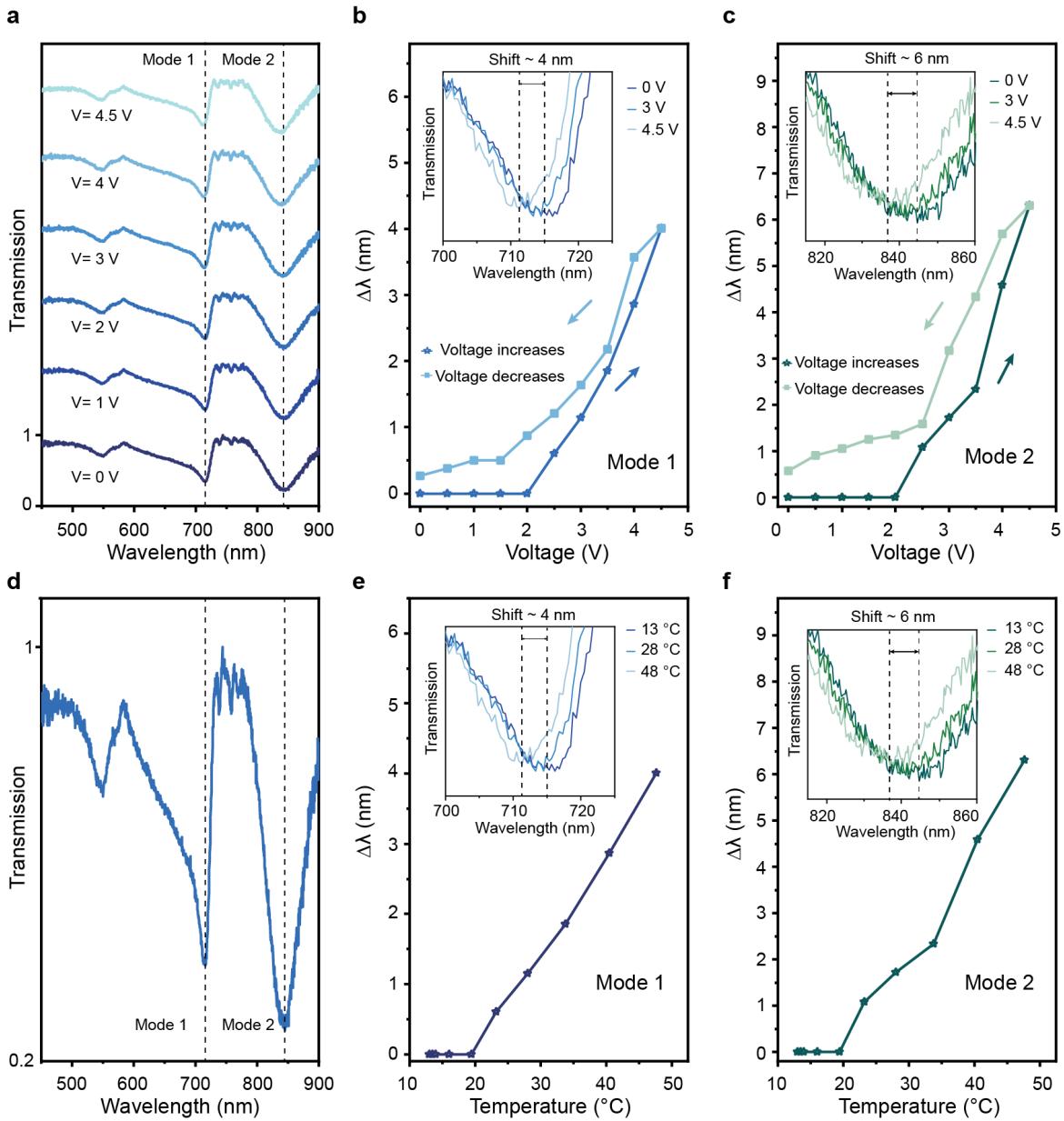


Figure S22. Electrical modulation performance of the Al metasurface on the ITO layer with a period of 370 nm. (a) Electrically modulated transmission spectra of the Al metasurface with voltages from 0 V to 5 V. The gray dashed lines label the wavelength peak of mode 1 and mode 2. The enlarged transmission spectra with the bias voltage of 0 V, 3 V, and 5 V of **(b)** mode 1 and **(c)** mode 2.

To further demonstrate the universality of electrical modulation in metasurfaces with varying geometries, we fabricated Al and Ag NP lattices with distinct lattice periods and NP sizes. The Al NP lattices with the ITO layer exhibit 2 resonance modes, characterized by a lattice period of 500 nm, an Al NP diameter of 150 nm, a height of 80 nm, and an ITO thickness of 150 nm. The maximum wavelength shifts observed for two modes are ~ 2.7 nm and ~ 3.7 nm with the voltage ranging from 0 V to 5 V and temperature ranging from 16.9 °C to 31.4 °C. **For an Al array with a period of 370 nm and a diameter and height of 80 nm, the wavelength shift is ~ 0.7 nm at 548 nm and 1.2 nm at 612 nm with a voltage of 5 V.**

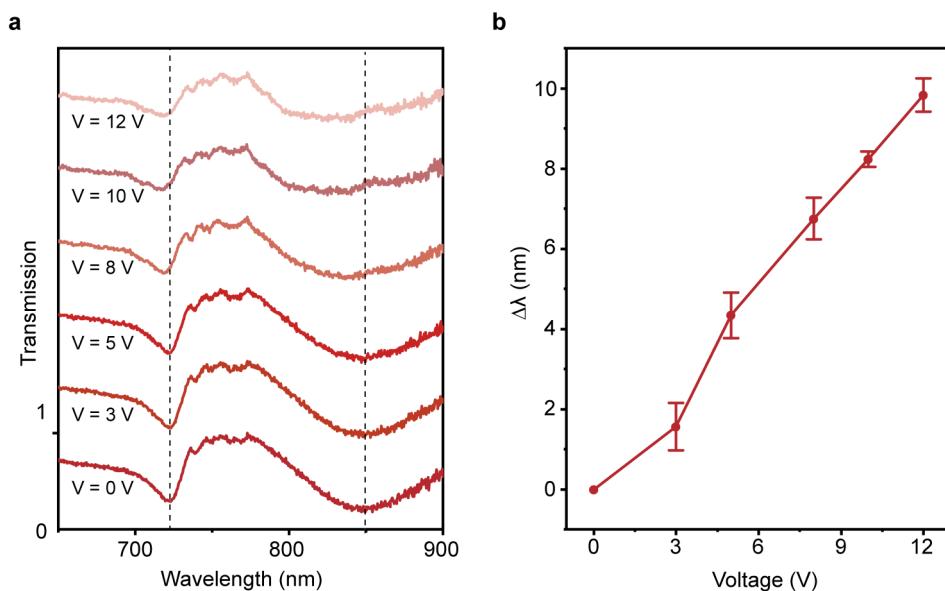
The Ag NP lattices with the ITO layer exhibit 2 resonance modes, with a lattice period of 500 nm, the diameter of Ag NP 150 nm, the height of 80 nm, and the thickness of the ITO layer of 150 nm. The maximum wavelength shifts observed for two devices are ~ 4 nm and ~ 6 nm, with the voltage ranging from 0 V to 5 V and temperature ranging from 13 °C to 48 °C.

Shifts of all modes in these two metasurfaces exceed the temperature-induced shifts obtained by simulation, signifying the existence of electrical synergistic modulation.

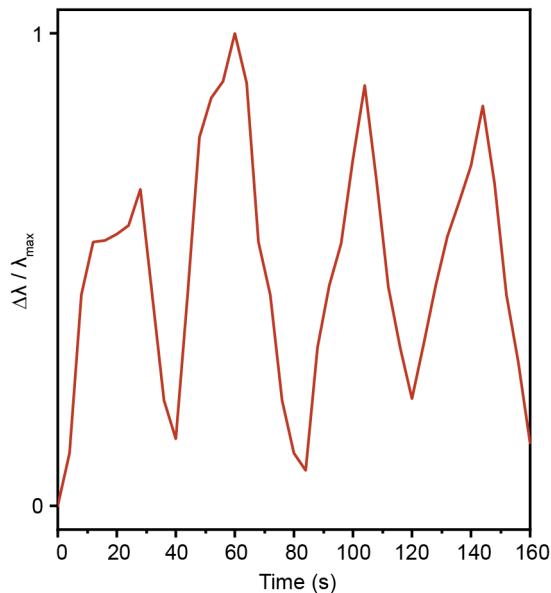


339 **Electrical modulation performance of Ag NP lattices on the ITGZO layer at a wider**
340 **voltage range.**

341 We have added the transmission spectra with the operation voltages up to 12 V. At voltages
342 exceeding 5 V, the resonance shift of Ag NPs on the ITGZO layer exhibits a linear relationship
343 with the voltage. This increased wavelength shift is primarily attributed to the large temperature
344 differential (approximately 50 °C) that occurs at these high voltages, which further reduces the
345 refractive index of DMSO. And the variation in the refractive index of the TCO is further
346 enhanced by the Seebeck effect. The observed broadening of the transmission spectrum at 10
347 V and 12 V may result from mismatched effective refractive indices with the substrate,
348 potentially due to the evaporation or denaturation of DMSO. Further, high voltages can cause
349 a rapid increase in temperature, leading to the evaporation or decomposition of DMSO.
350 Therefore, proper encapsulation of DMSO at high voltages to prevent its evaporation can
351 further improve the device performance.



352
353 **Figure S24. Electrical modulation performance of Ag NP lattices on the ITGZO layer. (a)**
354 **Electrically modulated transmission spectra at 0 V, 3 V, 5 V, 8 V, 10 V, and 12 V. (b)** Wavelength
355 **shifts at 722 nm with the voltage increases.**

357 **Alternating Current (AC) modulation performance of the Ag metasurface**

358

359 **Figure S25. AC modulation performance of Ag NP lattices with ITGZO layer.**

360 The signal generator we used produces a square wave signal with a value of peak-to-peak
 361 (VPP) of 4V and a period of 40s, with a rise time and a fall time of 20s each, and the
 362 spectrometer performs data acquisition every 4s for 200s.

363

364 **The electrical modulation performance of the TiO₂ metasurface on ITGZO/quartz
 365 substrate**

366 For dielectric metasurfaces, the reduction in DMSO refractive index upon voltage
 367 application, thereby causing a blue-shift of the resonant wavelength applies to dielectric
 368 metasurfaces. However, as the Seebeck effect occurs only in metals and semiconductors, it
 369 does not apply to the interface between the dielectric NP arrays and the TCO layer. We simulate
 370 a TiO₂ NP array with a period of 410 nm, a diameter of 130 nm, and a height of 80 nm, with a
 371 150 nm thick layer of ITGZO. We mainly focus on the resonance wavelength of approximately
 372 700 nm (Figure S26). As the temperature varies from 10 °C to 60 °C, the simulated
 373 transmission spectrum exhibits a continuous and pronounced blue shift, with the maximum
 374 shift of ~1.2 nm.

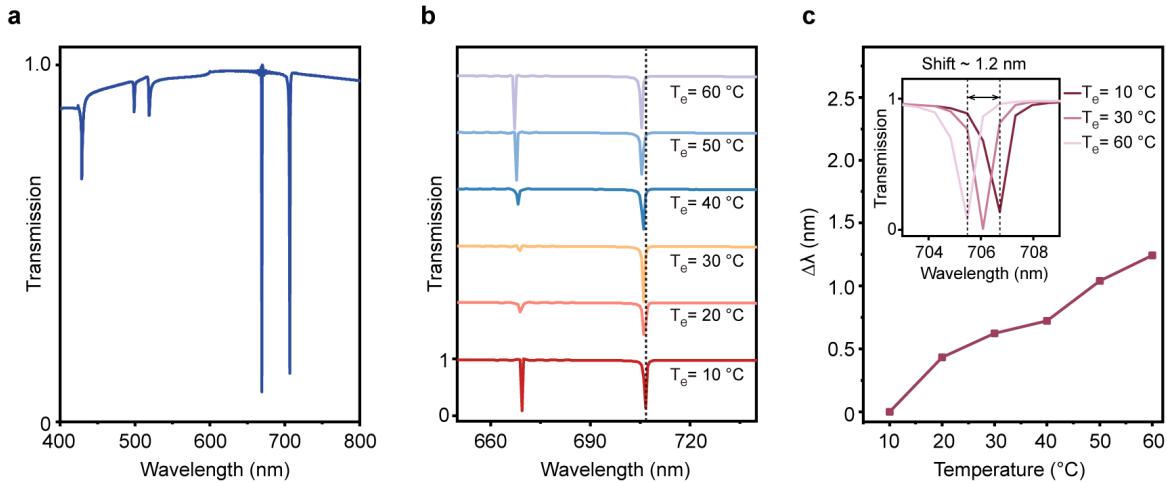
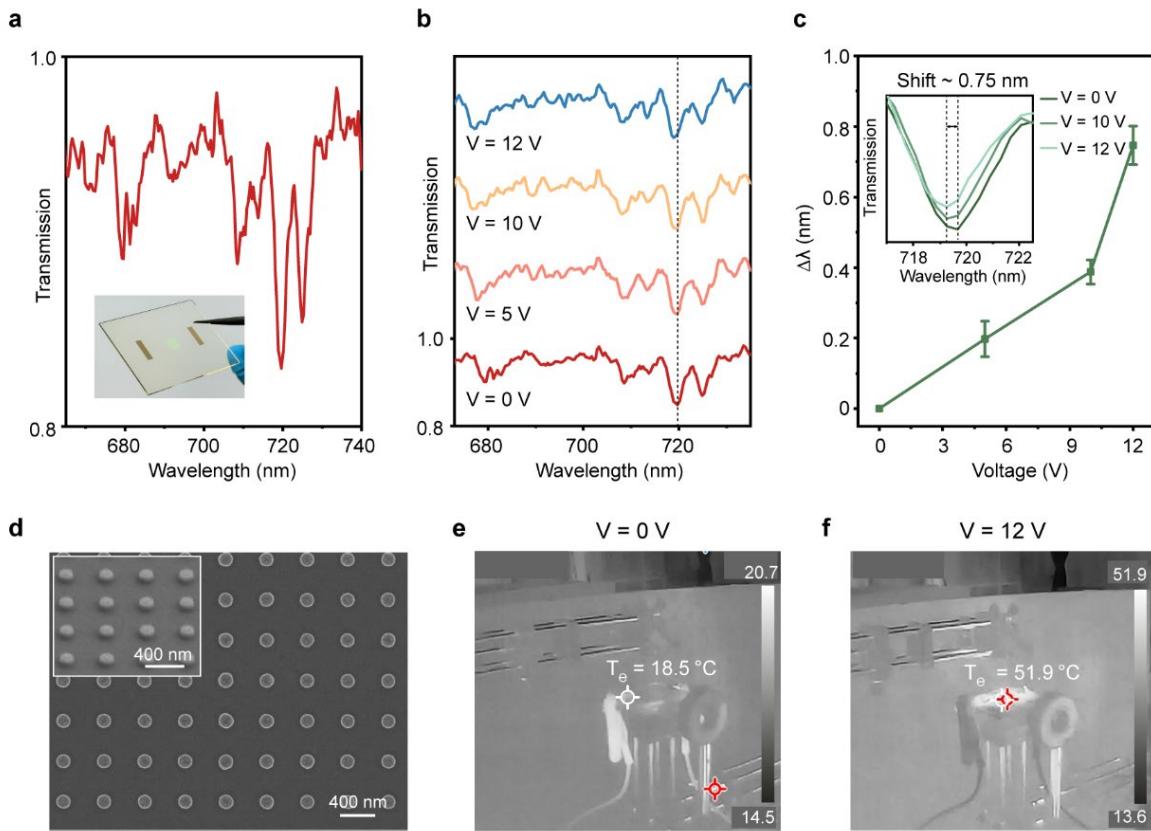


Figure S26. Simulated transmission spectra of TiO_2 metasurface. (a) Simulated transmission spectra of the TiO_2 metasurface. (b) Simulated transmission spectra with temperatures varying from 10 °C to 60 °C. The gray dashed lines label the initial wavelength peaks at ~700 nm. (c) The variation of shifts with temperature from 10 °C to 60 °C. The inset shows the resonance peaks at 10 °C, 30 °C, and 60 °C.

Further, we prepare a TiO_2 NP array with a period of 410 nm with the same device structure of metal NP lattices and measure the transmission spectra of TiO_2 at voltages varying from 0 V to 12 (Figure S27). The resonant wavelength at 720 nm with a linewidth of 2.5 nm exhibits a pronounced blue shift at 5 V, 10 V, and 12 V, with the maximum shift of approximately 0.75 nm (Figure S27 b-c). This shift is in reasonable agreement with the wavelength shift observed in simulations under temperature variations of 30-40 °C. Using an infrared thermal imaging camera, we measured the temperature distribution of the sample at 0 V and 12 V, showing a temperature difference of approximately 30 °C.

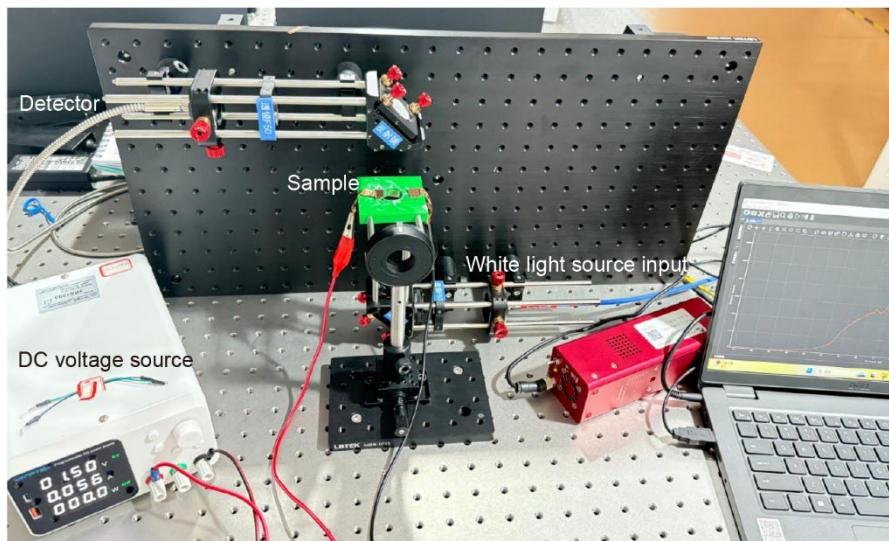
Therefore, this supplementary experiment further demonstrates that the wavelength blue shift of the plasmonic metasurface induced by voltage application is caused by the change in both the refractive index of DMSO and the Seebeck effect at the metal-TCO interface.



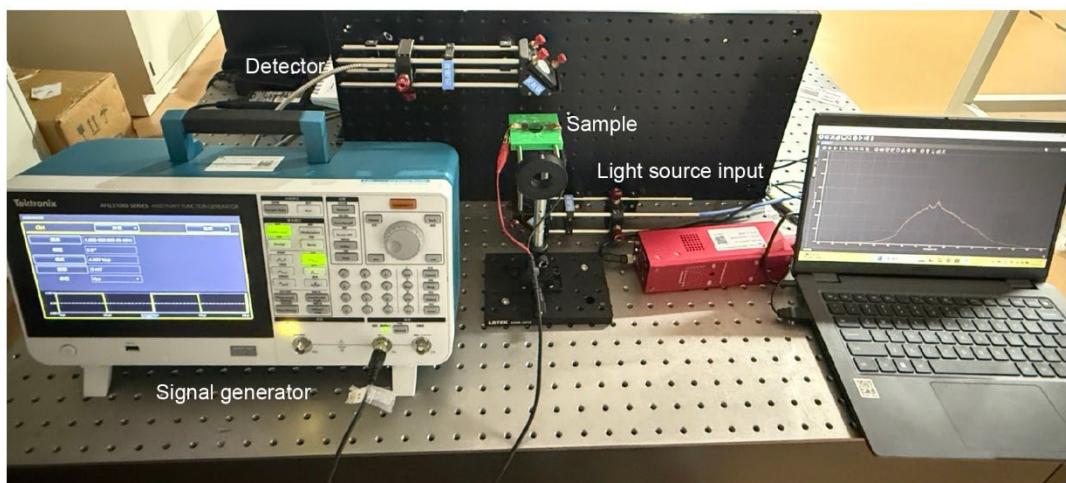
393
394 **Figure S27. Electrical modulation of tunable metasurfaces composed of TiO₂ NP lattices**
395 **on ITGZO layer. (a)** Measured transmission spectra of TiO₂ NP lattices on ITGZO/quartz
396 substrate. **(b)** Electrically modulated transmission spectra at 0 V, 5 V, 10 V, and 12 V. The grey
397 dashed line indicates the resonant wavelength of interest. **(c)** Resonance shifts of Ag NP lattices
398 on ITO with the voltage increases. The inset shows the resonance peaks with bias voltages of
399 0 V, 10 V, and 12 V. **(d)** SEM image of the top view of TiO₂ NP lattices. The inset shows the
400 SEM image at a 45-degree angle of TiO₂ NP lattices. The temperature distribution at **(e)** 0 V
401 and **(f)** 12 V measured by the infrared thermal imaging camera.

402

403 **Photo of the experiment setup for characterization of the electrical modulation**
404 **performance of metasurfaces.**



405
406 **Figure S28. Experiment setup for characterization of direct current (DC) performance.**
407



408
409 **Figure S29. Experiment setup for characterization of AC performance and image**
410 **transmissions.**
411

412 **References**

413 1. Lee MH, Huntington MD, Zhou W, Yang J-C, Odom TW. Programmable Soft Lithography:
414 Solvent-Assisted Nanoscale Embossing. *Nano Letters* **11**, 311-315 (2011).

415 2. Kogelnik H. Theory of Optical Waveguides. In: *Guided-Wave Optoelectronics* (ed Tamir T).
416 Springer Berlin Heidelberg (1988).

417 3. Mock JJ, Barbic M, Smith DR, Schultz DA, Schultz S. Shape effects in plasmon resonance
418 of individual colloidal silver nanoparticles. *The Journal of Chemical Physics* **116**, 6755-6759
419 (2002).

420 4. Palik, Edward D. Handbook of optical constants of solids. *Academic Press*, (1985).

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