

## Supporting Information

### Dual control of multi-band resonances with a metal-halide perovskite-integrated terahertz metasurface

Yuying Lu<sup>a,b,g,1</sup>, Tengteng Li<sup>e,1</sup>, Maosheng Yang<sup>c,d</sup>, Haiyun Yao<sup>d</sup>, Lanju Liang<sup>\*d</sup>, Xin Yan<sup>d</sup>, Kai Kai Lv<sup>d</sup>, Meng Wang<sup>d</sup>, Qili Yang<sup>d</sup>, Chaoyang Wei<sup>\*a,b,g</sup>, Jianda Shao<sup>\*b,f,g</sup>, Jianquan Yao<sup>e</sup>

<sup>a</sup> Precision Optical Manufacturing and Testing Centre, Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Shanghai, 201800, PR China. E-mail: siomwei@siom.ac.cn (C. Wei), E-mail: jdshao@siom.ac.cn (J. Shao).

<sup>b</sup> Key Laboratory for High Power Laser Material of Chinese Academy of Sciences, Shanghai Institute of Optics and Fine Mechanics, Shanghai, 201800, PR China

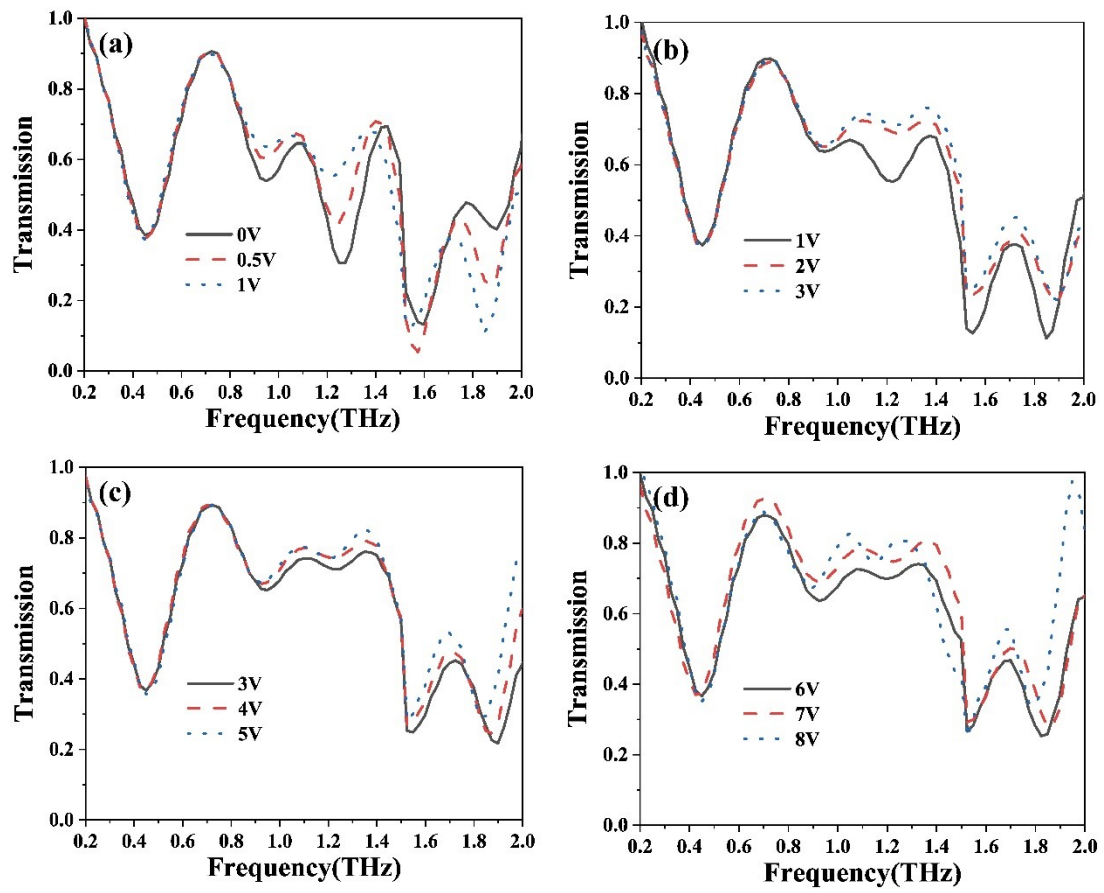
<sup>c</sup> Institute of Micro-nano Optoelectronics and Terahertz Technology and School of Mechanical Engineering, Jiangsu University, Zhen Jiang, 212013, China.

<sup>d</sup> School of Opto-electric Engineering, Zao Zhuang University, Zao Zhuang, 277160, China. E-mail: lianglanju123@163.com (L. Liang)

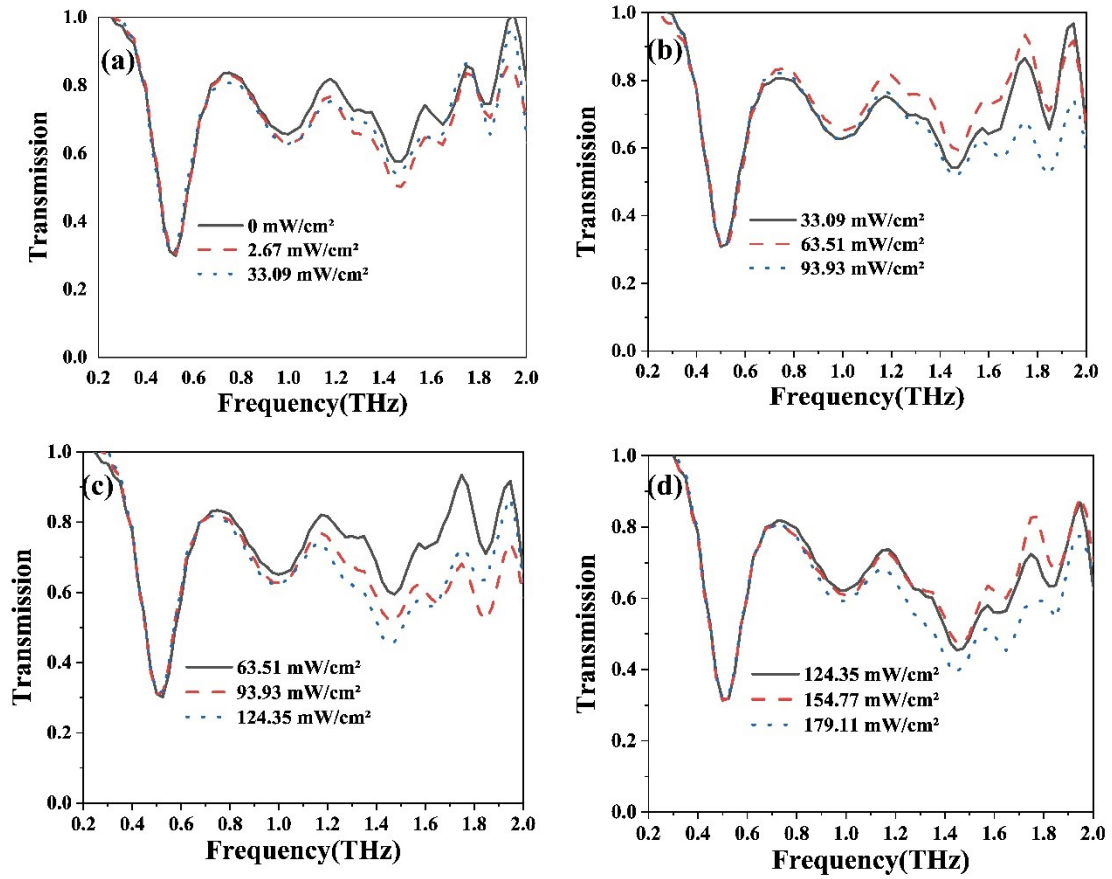
<sup>e</sup> College of Precision Instruments and Opto-electronics Engineering, Tianjin University, Tianjin, 300072, China.

<sup>f</sup> Hangzhou Institute for Advanced Study, University of Chinese Academy of Sciences, Hangzhou, 310024, PR China

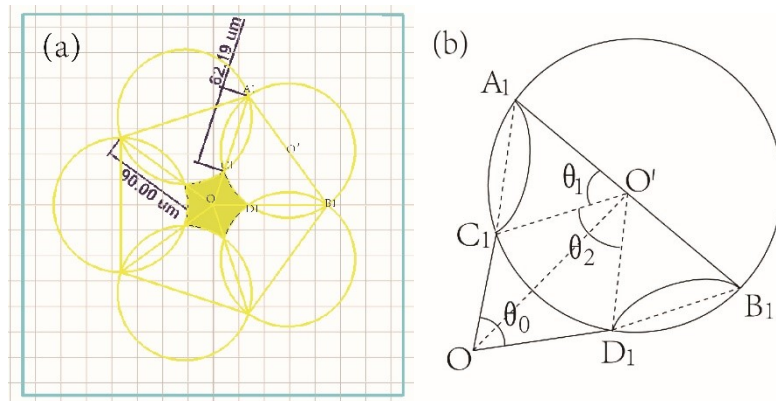
<sup>g</sup> Centre of Material Science and Optoelectronics Engineering, University of Chinese Academy of Sciences, Beijing, 100049, PR China.



**Fig. S1.** Transmission modulation of higher-order Fano resonances by the electronic method: (a)- (d) Y-polarized electrical modulation, also by tuning the bias voltage from 0 to 8 V.



**Fig.S2.** Transmission modulation of higher-order Fano resonances via the optical method: (a)-(d) Y-polarized optical modulation for the optical pump powers from 0 to 179.11 mW/cm<sup>2</sup>.



**Fig. S3.** The calculation process of the arcs (a) the macroscopic schematic diagram of the arc  $A_1C_1$  and  $C_1D_1$  (b) the microscopic schematic diagram of the arc  $A_1C_1$  and  $C_1D_1$ .

The coordinate point of O is (0, 0) and the coordinate point of O' is  $(x_0, y_0)$ .

Also,  $OO' = R = 90 \mu\text{m}$ .

In this picture,  $x_0 = R * \cos(\text{rot\_phi}) \approx 27.81 \mu\text{m}$

Here,  $y_0 = R * \sin(rot\_phi) \approx 85.59 \mu m$ .

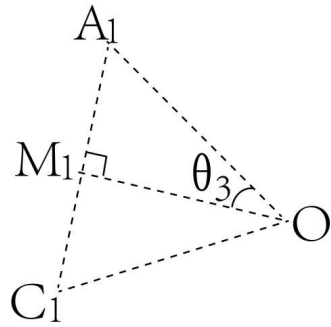
For  $n=5$ ,  $rot\_deg = 360 / n = 72^\circ$ .

And,  $rot\_phi = rot\_deg * 2 * \pi / 360$ .

We set  $R'' = \frac{A_1 B_1}{2} = D_1 O' = A_1 O'$  and  $R'' = \sqrt{(R - x_0)^2 + y_0^2}$ ,

After calculation, we can gain  $R'' \approx 53.91 \mu m$ ,

By measuring the length of the model of CST software,  $A_1 C_1 = 62.19 \mu m$



**Fig.S4.** The calculation process of the center angle corresponding to the arcs

For  $\theta_0 = 72^\circ$ , we set  $\theta_1 = 2 * \theta_3 = \angle A_1 O' C_1$ ,

And, we also set  $\theta_2 = \angle C_1 O' D_1$ ;  $\theta_2 + 2\theta_1 = \pi(rad)$ ,

$\theta_3 = \frac{1}{2} * \theta_1$  and  $A_1 M_1 = \frac{1}{2} A_1 C_1 \approx 31.095 \mu m$ .

$\sin \theta_3 = \frac{A_1 M_1}{A_1 O'} = \frac{A_1 M_1}{R''} \approx \frac{31.095}{53.91} = 0.57679$

$\theta_3 \approx \arcsin(0.57679) \approx 0.6148(rad)$ .

$\theta_1 = 2 * \theta_3 = 1.2296(rad)$ .

$\theta_2 = \pi - \theta_1 * 2 = 0.6824(rad)$

Thus, the center angle and radius corresponding to the arc  $A_1 C_1$  are  $53.91 \mu m$  and  $1.2296(rad)$ . While, the center angle and radius corresponding to the arc  $C_1 D_1$  are  $53.91 \mu m$  and  $0.6824(rad)$ . The curvature of these arcs can be more clearly described by arc lengths and radius.

**Table 1.** X-polarized electrical dynamic amplitude and its corresponding frequencies

Freq/Voltag	F <sub>0x</sub> (THz)/Amp	F <sub>1x</sub> (THz)/Amp	F <sub>2x</sub> (THz)/Amp	F <sub>3x</sub> (THz)/Amp	F <sub>4x</sub> (THz)/Amp
e					
0 V	0.4495/0.3789	0.9490/0.5928	1.2237/0.2964	1.5734/0.1959	1.7732/0.3027
0.5 V	0.4495/0.3859	0.9490/0.5725	1.2487/0.2248	1.5734/0.3598	1.7482/0.3976
1 V	0.4495/0.3826	0.9740/0.6209	1.2237/0.4259	1.5234/0.1420	1.8231/0.1305
2 V	0.4495/0.3730	0.9740/0.6514	1.2237/0.5916	1.5484/0.3701	1.8481/0.2573
3 V	0.4495/0.3657	0.9240/0.6939	1.1988/0.6913	1.5234/0.4561	1.8231/0.3097
4 V	0.4495/0.3712	0.9240/0.6699	1.1988/0.6450	1.4985/0.4168	1.8231/0.2555
5 V	0.4495/0.3673	0.9240/0.6982	1.1738/0.6928	1.4985/0.4829	1.7982/0.2732
7 V	0.4495/0.3617	0.8991/0.7147	1.1238/0.7237	1.4485/0.5751	1.7482/0.3371
8 V	0.4495/0.3601	0.8991/0.7229	1.0989/0.7427	1.4485/0.5826	1.7482/0.3644

**Table 2.** X-polarized optical dynamic amplitude and its corresponding frequencies

Power/mW/cm	G <sub>0x</sub> (THz)/Amp	G <sub>1x</sub> (THz)/Amp	G <sub>2x</sub> (THz)/Amp	G <sub>3x</sub> (THz)/Amp	G <sub>4x</sub> (THz)/Amp
0	0.5244/0.3207	0.9990/0.6198	1.4235/0.4959	1.5984/0.5648	1.7982/0.6841
2.67	0.4995/0.3224	0.9990/0.5735	1.4485/0.4580	1.5984/0.5224	1.8231/0.6054
33.09	0.4995/0.3172	0.9990/0.6161	1.4485/0.4953	1.5984/0.5582	1.7982/0.6346
63.51	0.5244/0.3235	0.9990/0.6648	1.4485/0.5881	1.5984/0.6219	1.7982/0.6582
93.93	0.4995/0.3147	0.9990/0.6554	1.4485/0.5920	1.5984/0.6489	1.7982/0.7193
124.35	0.4995/0.3228	0.9990/0.6324	1.4485/0.5196	1.5984/0.5709	1.7982/0.6422
154.77	0.4995/0.3222	0.9990/0.6408	1.4485/0.5106	1.5984/0.5795	1.7982/0.6652
179.11	0.5244/0.3187	0.9990/0.6628	1.4485/0.5926	1.5984/0.6635	1.7982/0.7132

**Table 3. X-polarized electrical dynamic calculated MD and its corresponding frequencies**

Freq/Voltage	F <sub>0x</sub> (THz)/MD(%)	F <sub>1x</sub> (THz)/ MD(%)	F <sub>2x</sub> (THz)/ MD(%)	F <sub>3x</sub> (THz)/ MD(%)	F <sub>4x</sub> (THz)/ MD(%)
0 V	0.4495/0	0.9490/0	1.2237/0	1.5734/0	1.7732/0
0.5 V	0.4495/1.847	0.9490/-3.42	1.2487/-24.15	1.5734/83.66	1.7482/31.35
1 V	0.4495/0.976	0.9740/4.74	1.2237/43.69	1.5234/-27.51	1.8231/-56.88
2 V	0.4495/-1.55	0.9740/9.88	1.2237/99.59	1.5484/88.92	1.8481/-14.99
3 V	0.4495/-3.48	0.9240/17.05	1.1988/133.23	1.5234/132.82	1.8231/2.31
4 V	0.4495/-2.03	0.9240/13	1.1988/117.61	1.4985/112.76	1.8232/-15.59
5 V	0.4495/-3.061	0.9240/17.78	1.1738/133.73	1.4985/146.50	1.7982/-9.74
7 V	0.4495/-4.53	0.8991/20.56	1.1238/144.16	1.4485/193.56	1.7482/11.36
8 V	0.4495/-4.96	0.8991/21.94	1.0989/150.57	1.4485/197.39	1.7482/20.38
MD range(%)	6.807	25.36	174.72	224.9	77.26

**Table 4. X-polarized optical dynamic calculated MD and its corresponding frequencies**

Freq/Power	G <sub>0x</sub> (THz)/MD(%)	G <sub>1x</sub> (THz)/ MD(%)	G <sub>2x</sub> (THz)/ MD(%)	G <sub>3x</sub> (THz)/ MD(%)	G <sub>4x</sub> (THz)/ MD(%)
0 mW/cm <sup>2</sup>	0.5244/0	0.9990/0	1.4235/0	1.5984/0	1.7982/0
2.67 mW/cm <sup>2</sup>	0.4995/0.53	0.9990/-7.47	1.4485/-7.64	1.5984/-7.50	1.8231/-11.50
33.09 mW/cm <sup>2</sup>	0.4995/-1.09	0.9990/-0.59	1.4485/-0.12	1.5984/-1.168	1.7982/-7.23
63.51 mW/cm <sup>2</sup>	0.5244/0.87	0.9990/7.26	1.4485/18.59	1.5984/10.10	1.7982/-3.78
93.93 mW/cm <sup>2</sup>	0.4995/-1.87	0.9990/5.74	1.4485/19.37	1.5984/14.89	1.7982/0.7193
124.35 mW/cm <sup>2</sup>	0.4995/0.65	0.9990/2.03	1.4485/4.77	1.5984/1.08	1.7982/5.14
154.77 mW/cm <sup>2</sup>	0.4995/0.46	0.9990/3.38	1.4485/2.96	1.5984/2.60	1.7982/-2.76
179.11 mW/cm <sup>2</sup>	0.5244/-0.62	0.9990/6.93	1.4485/19.49	1.5984/17.47	1.7982/4.25
MD range(%)	2.74	14.73	27.13	24.97	16.64



**Table 5. X-polarized electrical dynamic calculated frequency shifts and their corresponding frequencies**

Freq/Voltage	F <sub>0x</sub> (THz)/frequency shift(GHz)	F <sub>1x</sub> (THz)/frequency shift(GHz)	F <sub>2x</sub> (THz)/frequency shift(GHz)	F <sub>3x</sub> (THz)/frequency shift(GHz)	F <sub>4x</sub> (THz)/ frequency shift(GHz)
0 V	0.4495/0	0.9490/0	1.2237/0	1.5734/0	1.7732/0
0.5 V	0.4495/0	0.9490/0	1.2487/25	1.5734/0	1.7482/-25
1 V	0.4495/0	0.9740/25	1.2237/0	1.5234/-50	1.8231/49.9
2 V	0.4495/0	0.9740/25	1.2237/0	1.5484/-25	1.8481/74.9
3 V	0.4495/0	0.9240/-25	1.1988/-24.9	1.5234/-50	1.8231/49.9
4 V	0.4495/0	0.9240/-25	1.1988/-24.9	1.4985/-74.9	1.8231/49.9
5 V	0.4495/0	0.9240/-25	1.1738/-49.9	1.4985/-74.9	1.7982/25
7 V	0.4495/0	0.8991/-49.9	1.1238/-99.9	1.4485/-124.9	1.7482/-25
8 V	0.4495/0	0.8991/-49.9	1.0989/-124.8	1.4485/-124.9	1.7482/-25
Shift Range	0	74.9	149.8	124.9	99.9
(GHz)					

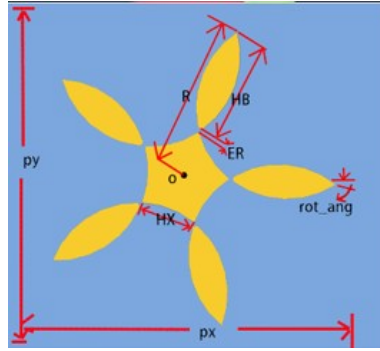
**Table 6. X-polarized optical dynamic calculated frequency shifts and their corresponding frequencies**

Freq/Optical	$G_{0x}$ (THz) /frequency shift(GHz)	$G_{1x}$ (THz) /frequency shift(GHz)	$G_{2x}$ (THz) /frequency shift(GHz)	$G_{3x}$ (THz) /frequency shift(GHz)	$G_{4x}$ (THz)/ /frequency shift(GHz)
0 mW/cm <sup>2</sup>	0.5244/0	0.9990/0	1.4235/0	1.5984/0	1.7982/0
2.67 mW/cm <sup>2</sup>	0.4995/-24.9	0.9990/0	1.4485/25	1.5984/0	1.8231/24.9
33.09 mW/cm <sup>2</sup>	0.4995/-24.9	0.9990/0	1.4485/25	1.5984/0	1.7982/0
63.51 mW/cm <sup>2</sup>	0.5244/0	0.9990/0	1.4485/25	1.5984/0	1.7982/0
93.93 mW/cm <sup>2</sup>	0.4995/-24.9	0.9990/0	1.4485/25	1.5984/0	1.7982/0
124.35 mW/cm <sup>2</sup>	0.4995/-24.9	0.9990/0	1.4485/25	1.5984/0	1.7982/0
154.77 mW/cm <sup>2</sup>	0.4995/-24.9	0.9990/0	1.4485/25	1.5984/0	1.7982/0
179.11 mW/cm <sup>2</sup>	0.5244/0	0.9990/0	1.4485/25	1.5984/0	1.7982/0
Shift Range (GHz)	24.9	0	25	0	24.9

**Table 7. The drawing process of the unit cell for the metasurface**

<p>Step1</p>		<p>First, we make a triangle with <math>R=90 \mu\text{m}</math> as the radius and the center angle of <math>360/n=360/5=72^\circ</math>. Then, we rotate the triangle five times with the Z axis to obtain a regular pentagon. One side of the triangle is parallel to the X axis. Five triangles are combined by Boolean operation (ADD) to obtain Part 1.</p>
<p>Step2</p>		<p>We took the midpoint of the hypotenuse of the triangle as the center of the circle. Its coordinate point is <math>(x_0, y_0)</math>. We also took the half of the hypotenuse of the triangle as the radius to draw a single circle.</p> <p>The intersection between the two adjacent circles is named as part 2.</p> $x_0 = R * \cos(\text{rot\_phi}) \approx 27.81 \mu\text{m}$ $y_0 = R * \sin(\text{rot\_phi}) \approx 85.59 \mu\text{m}$
<p>Step3</p>		<p>Rotate the single circle in the previous step for five times with the z-axis to obtain Part 3.</p>
<p>Step4</p>		<p>Add Part 1 and part 3 by Boolean operation to obtain Part 4.</p>
<p>Step5</p>		<p>Subtract Part 4 and part 3 by Boolean operation to obtain Part 5.</p>
<p>Step6</p>		<p>An intersection between two adjacent circles can be obtained as part 2. Add all the Boolean operations of part 2 to obtain part 6.</p>
<p>Step7</p>		<p>Part 5 and part 6 are added by Boolean operation to form part 7.</p>

Step 8



We rotate the obtained metasurface in the counterclockwise direction, In order to achieve the sharpest S-parameters curves at all the resonant frequencies, the rotated angle of the unit cell can be achieved by the scanning function in the CST software. By a lot of simulation process, the optimization goal was achieved when the rot\_angle equals to  $\pi/60(\text{rad})$  in Figure 1.

**The videos for the electric fields of the five resonant frequencies are attached as follows,**



0.61THz.mp4



0.8025THz.mp4



1.03THz.mp4



1.4175THz.mp4



1.5875THz.mp4