

## Electronic Supplementary Information

### Two-dimensional natural hyperbolic materials: from polaritons modulation to applications

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#### Lorentz model for calculating anisotropic permittivities of HMs

To describe the infrared permittivities of  $\alpha$ -MoO<sub>3</sub> and  $\alpha$ -V<sub>2</sub>O<sub>5</sub> crystals in Fig. 3 in the main article, a three-parameter Lorentz oscillator model is used with

$$\varepsilon_{jj} = \varepsilon_{jj}^{\infty} \prod_m^N \frac{(\omega_{jm}^{LO})^2 - \omega^2 - i\omega\Gamma_{jm}^{LO}}{(\omega_{jm}^{TO})^2 - \omega^2 - i\omega\Gamma_{jm}^{TO}}, \quad j = x, y, z$$

where  $\varepsilon_{jj}$  denotes the principal components of the permittivity tensor,  $\varepsilon_{jj}^{\infty}$  is the high-frequency dielectric constant. The parameters  $\omega_{jm}^{LO}$  and  $\omega_{jm}^{TO}$  represent the longitude and transverse optical phonon frequencies, respectively. The factor  $\Gamma_{jm}$  is the broadening factor of the Lorentzian lineshape. The superscripts  $x$ ,  $y$ , and  $z$  indicate three principal axes of the crystal along the crystal directions [100], [001], and [010], respectively, and  $m$  is the mode index along three crystal directions. The detailed parameter values utilized in our calculation for  $\alpha$ -MoO<sub>3</sub> and  $\alpha$ -V<sub>2</sub>O<sub>5</sub> are shown in Table S1.

**Table S1** Parameter values used for calculating the anisotropic permittivities of  $\alpha$ -MoO<sub>3</sub> and  $\alpha$ -V<sub>2</sub>O<sub>5</sub>

HMs	Crystal directions	$m$	$\omega_{jm}^{LO}$ (cm <sup>-1</sup> )	$\omega_{jm}^{TO}$ (cm <sup>-1</sup> )	$\Gamma_{jm}^{LO}$ (cm <sup>-1</sup> )	$\Gamma_{jm}^{TO}$ (cm <sup>-1</sup> )	$\epsilon_{jj}^{\infty}$
$\alpha$ -MoO <sub>3</sub>	[100]	1	972	820	4.0	4.0	4.0
	[001]	1	851	545	4.0	4.0	5.2
	[010]	1	1004	958	2.0	2.0	2.4
$\alpha$ -V <sub>2</sub> O <sub>5</sub>	[100]	1	76.2	72.4	4.2	3.6	6.559
		2	265.5	261.0	8.0	13.0	
		3	390.5	303.0	12.2	15.0	
		4	586.0	411.0	30.0	5.0	
		5	959.0	767.5	50.0	30.0	
		6	982.0	980.5	15.0	10.0	
	[001]	1	490.0	473.0	15.0	18.0	6.142
		2	1038.0	975.5	2.5	2.5	
	[010]	1	225.0	212.0	7.5	10.5	3.899
		2	312.5	284.0	10.2	7.8	
		3	842.5	506.5	18.0	21.0	