Configure polaritons in twisted $\alpha$-MoO$_3$

M. Chen$^{1†}$, X. Lin$^{2†}$, T. Dinh$^3$, Z. Zheng$^3$, J. Shen$^1$, Q. Ma$^3$, H. Chen$^2$, P. Jarillo-Herrero$^3$, S. Dai$^{1*}$

$^1$Materials Research and Education Center, Department of Mechanical Engineering, Auburn University, Auburn, Alabama 36849, USA
$^2$Interdisciplinary Center for Quantum Information, State Key Laboratory of Modern Optical Instrumentation, ZJU-Hangzhou Global Science and Technology Innovation Center, Zhejiang University, Hangzhou 310027, China
$^3$Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA

†These authors contribute equally

*Correspondence to: sdai@auburn.edu

Moiré engineering as a configuration method to twist van der Waals materials has delivered a series of advances in electronics, magnetics and optics. Yet these advances stem from peculiar moiré superlattices which form at small specific twisting angles. Here we report the configuration of nanoscale light-matter waves – the polaritons – by twisting stacked $\alpha$-phase molybdenum trioxide ($\alpha$-MoO$_3$) slabs in the broad range of 0° to 90°. Our combined experimental and theoretical results reveal a variety of polariton wavefront geometry and topological transitions via the twisting. The polariton twisting configuration is attributed to the
electromagnetic interaction of highly anisotropic hyperbolic polaritons in stacked $\alpha$-MoO$_3$ slabs. The nano-polaritons demonstrated in twisted $\alpha$-MoO$_3$ hold the promise as tailored nano-light for on-demand nanophotonic functionalities.

**Main text**

Weakly bonded atomic layers in van der Waals (vdW) systems[1-3] offer the platform to configure physical properties via twisting and stacking. This configuration has been recently investigated for the special case of moiré superlattices – the interference lattice pattern by twisting stacked vdW materials at small angles $\delta$ – to configure electronic, magnetic, optical and mechanical properties. These important qualities start from electronics where the moiré superlattice led to Hofstadter’s butterfly[4-6], superconductivity[7], correlated insulating states[8] and many others[9-13]. Later on, ferromagnetism[14] and moiré excitons[15-18] were reported in vdW structures at small magic twisting angles. In nanophotonics and polaritonic nano-optics where the research involves highly confined light-matter waves called polaritons[19, 20], the moiré superlattice was shown to reflect plasmon polaritons in graphene[21-24] and produce nano-light photonic crystals[21]. These previous breakthroughs in configuring vdW materials remark the peculiarities in the formed moiré superlattice as a special case of vdW configuration at small specific twisting angles $\delta$ (typically $\delta < 5^\circ$). To further exploit the advances of vdW systems, it is worth exploring the configurability via twisting, stacking etc. beyond the small specific $\delta$ range. Yet, for polaritons in vdW systems, without evident moiré superlattice, they remain rigid at a broad range of twisting angles ($\delta > 5^\circ$). This lack of configuration effect stems from the spreading nature of polaritons in most vdW materials: once excited, the polaritons
propagate toward all directions in the basal plane. The insufficient electromagnetic anisotropy of the spreading propagation manner weakens the twisting configuration by rotating the components.

In this work, we report on the configuration of polaritons in twisted $\alpha$-phase molybdenum trioxide ($\alpha$-MoO$_3$) slabs. Different from other spreading polaritons, polaritons in $\alpha$-MoO$_3$[25, 26] are extremely anisotropic: they propagate only along certain directions in the basal plane. Using real-space infrared (IR) nano-imaging of the propagating polaritons, we demonstrated various wavefront geometry and polariton topological transitions by twisting stacked $\alpha$-MoO$_3$ slabs with an angle $\delta$ from 0° to 90° (Fig. 1(a)). The polariton twisting configuration originates from the electromagnetic hybridization between directional polaritons in the top and bottom $\alpha$-MoO$_3$ slab where the hybridization strongly depends on the twisting angle $\delta$. Since a variety of polariton wavefront geometry like open hyperbola and parenthesis, closed oval and squircle as well as their deformation including compression and stretching can all be obtained simply by varying the twisting angle $\delta$, twisted $\alpha$-MoO$_3$ in this work holds promises to offer tailorable IR nano-light for various nano-optical functionalities.

The IR nano-imaging of polaritons in twisted $\alpha$-MoO$_3$ were performed using scattering-type scanning near-field optical microscopy (s-SNOM). The s-SNOM is based on a tapping-mode atomic force microscope (AFM) which simultaneously yields the topography and nano-IR image of the scanned area (Fig. 1(a)). In the experiment, the AFM tip is illuminated with an IR laser (brown solid arrow) at the frequency $\omega = 1 / \lambda_0$ ($\lambda_0$ is the wavelength of IR light in free space), the topography and back scattered near-field signal (brown dashed arrow) are recorded during
the scan. The experimental observable near-field amplitude \( S(\omega) \) and phase \( \Phi(\omega) \) typically possess a spatial resolution \( \sim 10 \) nm, close to the radius of the AFM tip, and therefore can map nano-polaritons in the real-space\[19, 20, 27\]. The polaritons are typically imaged as fringes – standing wave interference oscillations – in the s-SNOM experiments.

At the IR frequency \( \omega = 915 \) cm\(^{-1}\), a representative s-SNOM amplitude \( S(\omega) \) image of the single \( \alpha\)-MoO\(_3\) slab is shown in Fig. 1(b). In the experiment, we fabricated an Au disk (thickness 100 nm, diameter 1 \( \mu \)m) on top of the \( \alpha\)-MoO\(_3\) slab. Two types of polariton fringes – the \( S(\omega) \) oscillations – can be observed: “X” shape hyperbolic fringes around the Au disk (red disk) and linear fringes close to the slab edge (white dashed line). The linear fringes originate from the interference between polaritons launched by the s-SNOM tip and those reflected at the crystal edges. These fringes are always parallel to the crystal edge\[28-30\] regardless of the polariton wavefront geometry. The “X” shape fringes around the Au disk are interference patterns between polaritons launched by the Au disk and the IR illumination (brown solid arrow in Fig. 1(a))\[29, 31\]. For highly confined polaritons in vdW materials\[29\], these fringes superposition with the wavefront of the launched polaritons and therefore are mainly discussed in this work. In contrast to spreading concentric fringes from other vdW materials, “X” shape fringes observed in \( \alpha\)-MoO\(_3\) indicate the extreme anisotropy of the directional polaritons. This polariton directionality stems from the hyperbolic response (\( \varepsilon_i \varepsilon_j < 0 \), where \( \varepsilon_i \) and \( \varepsilon_j \) are permittivity along different directions, \( i \) and \( j \) denote crystal axis [100], [001] or [010]) inside Reststrahlen bands of \( \alpha\)-MoO\(_3\)\[25, 26\]: Band 1 at \( \omega = 545 \) to 851 cm\(^{-1}\) for [001] phonon (\( \varepsilon_{[100]} > 0, \varepsilon_{[001]} < 0 \) and \( \varepsilon_{[010]} > 0 \)), Band 2 at \( \omega = 820 \) to 972 cm\(^{-1}\) for [100] phonon (\( \varepsilon_{[100]} < 0, \varepsilon_{[001]} > 0 \) and \( \varepsilon_{[010]} > 0 \)), and Band
3 at $\omega = 958$ to 1010 cm$^{-1}$ for [010] phonon ($\varepsilon_{[100]} > 0$, $\varepsilon_{[010]} > 0$ and $\varepsilon_{[001]} < 0$). Specifically, the directional “X” shape fringes (Fig. 1(b)) are directly attributed to the basal plane hyperbolic response ($\varepsilon_{[100]}\varepsilon_{[001]} < 0$) inside the Reststrahlen band 2: the isofrequency dispersion for the $\alpha$-MoO$_3$ slab is an open hyperbola (Fig. 1(c), green curve) in the basal plane and polaritons are propagating along fixed directions (green arrows in Fig. 1(c)) with an angle $\theta = \arctan \sqrt{|\varepsilon_{[100]}| / |\varepsilon_{[010]}|}$ to the [010] direction [26, 32].

The directional polaritons in the basal plane suggest the configurability by twisting stacked $\alpha$-MoO$_3$ slabs inside the Reststrahlen band 2. In Figures 1(d) –(h), we provided s-SNOM images of polaritons in twisted $\alpha$-MoO$_3$ at various twisting angle $\delta$. The s-SNOM images are oriented with all [001] axis of the bottom $\alpha$-MoO$_3$ slab aligned in the vertical direction (see the coordinate axis in Figure 1). At $\delta = 0^o$, the polaritons exhibit the “X” shape fringes (Fig. 1(d)) that correspond to the hyperbolic wavefront, very similar to that of the single slab $\alpha$-MoO$_3$ (Fig. 1(b)). At $\delta = 20^o$ (Fig. 1(e)), the hyperbolic wavefront can still be observed but is titled from that with $\delta = 0^o$ and the single slab. The polariton wavefront at $\delta = 63^o$ is different: the fringes can only be observed along a tilted line across the Au disk (Fig. 1(f)), the wavefront is therefore in a parenthesis geometry. With this parenthesis wavefront, polaritons are confined along the tilted line, the propagation along other directions are prohibited. This parenthesis wavefront is also revealed in polariton fringes launched by a defect (red * in Fig. 1(f)) where similar parenthesis fringes along the same tilted direction can be observed. At $\delta = 81^o$ (Fig. 1(g)), the polariton wavefront becomes oval as fringes observed around the Au dot. Finally, at $\delta = 90^o$ (Fig. 1(h)), the wavefront geometry appears as a squircle, rounded square polariton fringes are observed. In Fig.
1(h), the squircle fringes are less visible on the lower left side of the Au disk, due to the shadowing effect of the AFM cantilever[29].

s-SNOM images in Figure 1 reveal various wavefront geometry and topological transitions of the polaritons in twisted α-MoO3. As the twisting angle $\delta$ increases from 0° to 90°, the polariton wavefront exhibit a hyperbola (Fig. 1(d), $\delta = 0°$), tilted hyperbola (Fig. 1(e), $\delta = 20°$), parenthesis (Fig. 1(f), $\delta = 63°$), oval (Fig. 1(g), $\delta = 81°$) and squircle (Fig. 1(h), $\delta = 90°$). Note that at $\delta \sim 63°$, the polariton wavefront changes from an open hyperbola-like geometry (Figures 1(d)-(e)) to a closed ellipse-like geometry (diamond, squircle in Figures 1(g)-(h)), corresponding to the topological transitions[33, 34]. This continuous polariton twisting configuration in α-MoO3 over the broad range $\delta = 0°$ to 90° is in stark contrast to plasmons in twisted graphene and graphene/hBN heterostructures where plasmon polaritons are affected at specific and small twisting angles ($\delta < 5°$) by the moiré superlattice[21-23].

The observed polariton twisting configuration is supported both by our finite element method (FEM) simulation and electromagnetic wave theory in Figure 2. With the input of α-MoO3 slab thickness and IR frequency $\omega$ from our experiments and fitted α-MoO3 permittivity based on ref. [25, 26, 35], we simulated the real-space electromagnetic field $E_z$ and modelled momentum space ($k$-space) isofrequency dispersion in twisted α-MoO3. In the real-space simulation (see details in supplementary information Section 1), a vertical dipole was placed at the center (Figures 2(a)-(f), top) and above the twisted α-MoO3 as the polariton launcher. The real-space simulation reproduces the topological transition of polaritons observed in our experiments (Figure 1): the wavefront exhibit the hyperbola at the single slab and $\delta = 0°$ (Fig. 2 (a), (b), top),
tilted hyperbola at $\delta = 20^\circ$ (Fig. 2(c), top), parenthesis at $\delta = 63^\circ$ (Fig. 2(d), top), oval at $\delta = 81^\circ$ (Fig. 2(e), top) and squircle at $\delta = 90^\circ$ (Fig. 2(f), top). Our electromagnetic theory (see details in Supplementary Information Section 2) of the isofrequency dispersion in twisted $\alpha$-MoO$_3$ was developed from previous theoretical works on black phosphorus[36, 37] and graphene metasurface[33, 38]. The modeled $k$-space isofrequency curves (Figures 2(a)-(f), bottom) are in excellent agreement with the real-space images produced in our experiments (Figure 1) and FEM simulations (Figures 2(a)-(f), top).

We attribute the twisting configuration reported in Figures 1-2 to electromagnetic interaction between extremely anisotropic polaritons in the stacked top and bottom $\alpha$-MoO$_3$ slab. Specifically, the twisting configuration requires 1) extreme basal plane anisotropy of the polariton and 2) sufficient overlap of polariton field from stacked slabs. In Figure 3, we provide experimental data and calculation analysis to demonstrate these criteria for the twisting configuration. At a representative IR frequency $\omega = 980$ cm$^{-1}$ inside the Reststrahlen Band 3, the $k$-space polariton geometry is an ellipse (red curve, Fig. 1(c)) and polaritons propagate with an elliptical wavefront in the real-space (Fig. 3(a)). Compared with directional polaritons inside the Band 1 (Fig. 1(b)), polaritons at $\omega = 980$ cm$^{-1}$ lack sufficient anisotropy such that they spread towards all directions (Fig. 3(a)). The twisting configuration is ineffective at $\omega = 980$ cm$^{-1}$: polaritons from twisted $\alpha$-MoO$_3$ with $\delta = 20^\circ$ (Fig. 3(b)), $63^\circ$ (Fig. 3(c)) and $90^\circ$ (Fig. 3(d)) all exhibit elliptical wavefront similar to that in the single slab (Fig. 3(a)). These results are in stark contrast with the s-SNOM image at $\omega = 900$ cm$^{-1}$ (Fig. 3(f)) where the squircle and a series of other wavefront (Figure 1) can be configured by twisting $\alpha$-MoO$_3$ slabs supporting the
directional polaritons.

In addition to the extreme basal plane anisotropy, sufficient field overlap between polaritons in stacked vdW structures is also required to build adequate electromagnetic interaction to configure polaritons. Since polaritons exponentially decay out-side-of the $\alpha$-MoO$_3$ slab, this criterion corresponds to a moderate decay of the polariton field. In Fig. 3(g), we calculated the distribution of electromagnetic field $|E_z|$ away from the $\alpha$-MoO$_3$ slab (see Supplementary Information Section 2 for details) at three representative IR frequencies: $\omega = 900$ cm$^{-1}$ (green) and 935 cm$^{-1}$ (blue) inside the Band 2 and $\omega = 980$ cm$^{-1}$ (red) inside the Band 3. Away from the $\alpha$-MoO$_3$ slab, the polariton field $|E_z|$ decays quickly at $\omega = 935$ and 980 cm$^{-1}$ while moderately at $\omega = 900$ cm$^{-1}$: the field decay length where $|E_z|$ becomes $1/e$ of that at $z = 0$ is 14, 92 and 243 nm for $\omega = 980$, 935 and 900 cm$^{-1}$, respectively. Therefore, in stacked $\alpha$-MoO$_3$, the polaritons field overlap between the top and bottom $\alpha$-MoO$_3$ is sufficient at $\omega = 900$ cm$^{-1}$ while insufficient at $\omega = 935$ and 980 cm$^{-1}$. The sufficient polariton field overlap leads to adequate electromagnetic interaction and thus the twisting configuration at $\omega = 900$ cm$^{-1}$. Our s-SNOM data in Figures 3(d)-(f) support this theoretical expectation. Evident twisting configuration was observed at $\omega = 900$ cm$^{-1}$ (Fig. 3(f) and Figure 1) while the lack of twisting configuration was observed at $\omega = 935$ and 980 cm$^{-1}$. Note that at $\omega = 935$ cm$^{-1}$ (Fig. 3(e)), the polaritons in the top and bottom $\alpha$-MoO$_3$ slab fulfill the other criterion to be basal plane anisotropic (hyperbolic wavefront), yet these hyperbolic modes barely interact with each other due to their insufficient field overlap (blue curve in Fig. 3(g)). Adequate electromagnetic interaction as the criterion of polariton twisting configuration is further confirmed by our simulation results of separated $\alpha$-MoO$_3$ slabs
in Supplementary Section 3.

Combined experimental and theoretical results in Figures 1-3 report the configuration of nano-polaritons in twisted $\alpha$-MoO$_3$. The polariton wavefront can be altered by controlling the twisting angle $\delta$ between stacked $\alpha$-MoO$_3$ slabs in a broad range of 0° to 90°, thus demonstrating the extended vdW configurability of the nanoscale electromagnetic energy beyond Morié engineering with small twisting angles [4-13, 21, 22]. The observed twisting configuration of polaritons is attributed to electromagnetic interaction between anisotropic polariton fields where the interaction is highly dependent on the twisting angle. Note that by configuring the wavefront via twisting, the tuning of polariton propagation direction and wavelength surpasses previous modulation efforts through refractive index engineering[39, 40] or vdW heterostructuring[41-43]. Future work may be guided towards exploring the dynamics and reconfigurability of twisted polariton nano-light via nano-mechanical manipulation[2] or vdW photonic hybrids[1, 41-44]. Various polariton wavefront and topologies demonstrated in this work suggest the opportunity to offer polariton nano-light with tailored propagating properties and photonic density of states[45, 46] for on-demand nano-optical functionalities that can benefit light emission[46], quantum optics[47, 48] and exotic transitions[49] etc. The twisting configuration of hyperbolic polaritons in $\alpha$-MoO$_3$ also provides a prototype for the exploration of configuring other anisotropic physical properties in vdW materials via twisting, stacking and heterostructuring.
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FIG. 1. Real-space IR nano-images reveal the twisting configuration for nano-polaritons. (a) Experiment schematic. Solid and dashed brown arrows denote the incident and back-scattered IR light. (b) s-SNOM amplitude image of single slab $\alpha$-MoO$_3$. (c) Isofrequency curve of hyperbolic polaritons in Reststrahlen Band 2 and Band 3. Green arrows denote the momentum of polaritons in Band 2. (d)–(h) s-SNOM amplitude images of twisted $\alpha$-MoO$_3$ with $\delta = 0^\circ$, 20$^\circ$, 63$^\circ$, 81$^\circ$ and 90$^\circ$. IR frequency in b): 915 cm$^{-1}$, d): 915 cm$^{-1}$, e): 910 cm$^{-1}$, f): 920 cm$^{-1}$, g): 915 cm$^{-1}$, h): 905 cm$^{-1}$. Scale bar: 2 $\mu$m. Note that at the fixed frequency $\omega = 910$ and 915 cm$^{-1}$, the s-SNOM images (Supplementary Section 4) of twisted $\alpha$-MoO$_3$ show the same twisting configuration and topological transitions of polaritons as those in Figure 1.
FIG. 2. Finite Element Method (FEM) real-space simulation and electromagnetic theory of momentum-space (k-space) isofrequency dispersion of twisted polaritons. (a) – (f) top, real-space FEM simulation of polaritons in twisted α-MoO₃ where a dipole was placed above the center of the image to launch the polaritons. Scale bar: 2 μm. (a) – (f) bottom, electromagnetic theory of corresponding isofrequency dispersions. Scale bar: 15 $k_0$, $k_0$ is the momentum of IR photon $k_0 = 2\pi / \lambda_0$. 


FIG. 3. Electromagnetic interaction as the origin of twisting configuration in polaritons. (a) – (d) s-SNOM amplitude images of single slab $\alpha$-MoO$_3$ (a) and twisted $\alpha$-MoO$_3$ with $\delta = 20^\circ$, $63^\circ$ and $90^\circ$ at IR frequency $980$ cm$^{-1}$. (e) s-SNOM amplitude images of twisted $\alpha$-MoO$_3$ with $\delta = 81^\circ$ at IR frequency $935$ cm$^{-1}$. (f) s-SNOM amplitude images of twisted $\alpha$-MoO$_3$ with $\delta = 90^\circ$ at IR frequency $900$ cm$^{-1}$. Scale bar: 2 $\mu$m. (g), theoretical electromagnetic field distribution $|E_z|$ along the [100] direction away from the $\alpha$-MoO$_3$ slab. $\alpha$-MoO$_3$ thickness: 100 nm.