Surface plasmons induce topological transition in graphene/$\alpha$-MoO$_3$ heterostructures

Francesco L. Ruta$^{1,2,5}$, Brian S. Y. Kim$^{3,5}$, Zhiyuan Sun$^1$, Daniel J. Rizzo$^1$, Alexander S. McLeod$^1$, Anjaly Rajendran$^3$, Song Liu$^3$, Andrew J. Millis$^{1,4}$, James C. Hone$^3$ & D. N. Basov$^1$

Polaritons in hyperbolic van der Waals materials—where principal axes have permittivities of opposite signs—are light-matter modes with unique properties and promising applications. Isofrequency contours of hyperbolic polaritons may undergo topological transitions from open hyperbolas to closed ellipse-like curves, prompting an abrupt change in physical properties. Electronically-tunable topological transitions are especially desirable for future integrated technologies but have yet to be demonstrated. In this work, we present a doping-induced topological transition effected by plasmon-phonon hybridization in graphene/$\alpha$-MoO$_3$ heterostructures. Scanning near-field optical microscopy was used to image hybrid polaritons in graphene/$\alpha$-MoO$_3$. We demonstrate the topological transition and characterize hybrid modes, which can be tuned from surface waves to bulk waveguide modes, traversing an exceptional point arising from the anisotropic plasmon-phonon coupling. Graphene/$\alpha$-MoO$_3$ heterostructures offer the possibility to explore dynamical topological transitions and directional coupling that could inspire new nanophotonic and quantum devices.
Hypersonic optics first rose to prominence with the development of hypersonic metamaterials constructed by periodically alternating metallic and dielectric media. The term "hypersonic" refers to the shape of the isofrequency surface, which is hyperboloidal rather than ellipsoidal like in common anisotropic media, because of the opposite signs of the real dielectric permittivities of metallic and dielectric media. Hyperbolic isofrequency surfaces give rise to an increased photonic anisotropic media, because of the opposite signs of the real signed permittivities and support in-plane hyperbolic modes with direction7,10,11.

Integrated nanophotonics requires dynamical tuning of hyperbolic polaritons, but this is challenging since hyperbolicity in the two most widely studied systems (h-BN and α-MoO₃) is rooted in crystal structure. Heterostructuring and hybridizing hyperbolic modes with other modes can modify their properties16-22, but adjusting tuning parameters typically requires altering device design. With α-MoO₃, for example, in-plane hyperbolic phonon polaritons can be modified by fabricating double-layer structures with a predefined twist angle as the tuning parameter16-19. The isofrequency contour (IFC) characterizing the propagation-direction-dependent momenta of in-plane hyperbolic modes can be tuned through a topological transition23 from an open hyperbola to a closed curve by sweeping twist angle. On the other hand, dynamical tuning of h-BN phonon polaritons has been demonstrated by means of hybridization with graphene surface plasmons20. Surface plasmon-phonon polariton (SP³) and hyperbolic plasmon-phonon polariton (HP³) hybrid modes have gate-tunable dispersions enabled by the carrier-density-dependent Fermi energy E_F of graphene. However, graphene/h-BN admits only modest electronic tunability without topological transitions.

Here, we present a graphene on α-MoO₃ heterostructure that can be tuned electronically through a topological transition as evidenced by direct imaging of propagating polaritons. At charge neutrality, graphene does not host plasmons and the IFC is a hyperbola. Upon electronic doping, the IFC transitions to a closed curve by modifying E_F and laser frequency ω. Furthermore, calculations corroborated by experiments reveal that, by rotating the polaritonic wavevector, the hybrid mode characterized by the closed IFC is tuned from HP³ to SP³ as its plasmon-phonon coupling is modified. Graphene/α-MoO₃ is thus a platform for studying the effect of anisotropic plasmon-phonon coupling on properties of hybrid hyperbolic polaritons. In particular, we identify an exceptional point in the direction-dependent polariton dispersion.

**Results and discussion**

**Hybrid polariton theory.** In this work, we focus on frequencies in the middle reststrahlen band of α-MoO₃ (820–972 cm⁻¹) where the topological transition occurs. Finite difference time domain simulations (Methods) show that, without doping, graphene is devoid of plasmonic response and the heterostructure is optically equivalent to bare α-MoO₃ (neglecting losses in charge-neutral graphene). In the cleavage plane of the [001] (x) and [001] (y) crystal axes (Fig. 1a), the phonon polariton wavefront is a hyperbola (Fig. 1b). The IFC, related to the wavefront by a Fourier transform, is likewise a hyperbola (Fig. 1c). Multiple hyperbolas appear: corresponding to higher-order, short wavelength hyperbolic modes. Upon doping the graphene to appreciable E_F, the wavefront becomes a closed curve with anisotropic polariton wavelengths as a result of plasmon-phonon hybridization (Fig. 1d). The first-order IFC undergoes a topological transition to a closed peanut shape (Fig. 1e).

The direction-dependent dispersion q(ω, θ) and dissipation γ(q, ω, θ) of hybrid modes in graphene with optical conductivity σ_x of a biaxial α-MoO₃ slab of thickness d at arbitrary frequency ω can be computed by solving Equation S5 (Supplementary Note 1). Figure 1f shows that calculated IFCs at E_F = 0 eV and 0.4 eV in the middle reststrahlen band are consistent with simulated IFCs in Fig. 1c, e, respectively. The angle θ specifies the in-plane propagation direction with respect to the [100] direction. The out-of-plane propagation constant inside α-MoO₃ is k_z ≈ ±q√−ε_y/ε_x where ε_x = ε_y cos²θ + ε_z sin²θ. In the doped structure, the wave launched by the tip is a surface wave (SP³: Im k_z ≫ Re k_z) along the [001] direction; and a waveguide mode (HP³: Re k_z ≈ Im k_z) along the [100] direction. Along intermediate directions (Fig. 1g), the polariton is an HP³ up to the open angle θ_i = arctan(1/√−ε_y/ε_z), i.e., the asymptote of the undoped hyperbola. Above θ_i, it is an SP³. Note that HP³ and SP³ in this work are only defined by their k_z. At θ_i, both real and imaginary components of k_z drop precipitously. Neglecting in-plane losses (dashed lines), k_z = 0 at θ_i: the polariton is an ideal plane wave propagating in-plane with finite momentum.

Along the [001] direction in the middle reststrahlen band, the α-MoO₃ phonon is absent and the plasmon-phonon coupling strength must be zero. Along the [100] direction, graphene plasmons and α-MoO₃ phonons hybridize due to strong coupling: that is, the plasmon mode splits into plasmon-phonon polariton branches above and below the a-axis transverse optical (TO) phonon frequency ω_TO. Tuning the in-plane propagation angle θ must then modify the plasmon-phonon coupling, which transitions at some point between strong and weak coupling regimes. Prior work24 has recognized that the transition occurs at an exceptional point (EP), above which plasmons experience Rabi splitting from strong coupling to phonons. Below the EP, there is no mode splitting: plasmons and phonons are weakly coupled in a manner akin to electromagnetically-induced transparency.

We now show quantitatively that tuning θ can be expected to modify plasmon-phonon coupling. Consider two-dimensional α-MoO₃ with high-frequency permittivities set to unity. The total dielectric function of Drude-like graphene with electron coupling to in-plane hyperbolic phonons modulated by the quantity α(q, θ) ≡ ω_F² cos²θ is then:

\[
ε_{2D}(ω, q, θ) = 1 - \frac{ω_F²(q)}{ω² + iωγ} + α(q, θ) \frac{ω² - ω_TO² - iωγ}{ω_TO² - ω² - iωγ},
\]
where $\omega^2(q) = g|E_F|^2/2\pi\hbar^2 \epsilon_0$ and $y_s$ and $y$ are the plasmon and phonon scattering rates, respectively (Supplementary Note 2). The real and imaginary roots of Eq. 1 are plotted in Fig. 1h, i, respectively. Larger $\alpha$ near the [100] direction split plasmon modes at $\omega_{pl}$ into two branches above and below $\omega_s'/\epsilon_0$ labeled $\omega_+$ and $\omega_-$, respectively (Fig. 1h). $\omega_s$ and $\omega$ have anticorrelated scattering rates, $\Gamma_+$ and $\Gamma_-$ (Fig. 1i). Splitting is reduced as $\theta$ rotates toward the [001] direction until a complex degeneracy, beyond which $\omega_+$ and $\omega_-$ coalesce and $\Gamma_+$ and $\Gamma_-$ split. This degeneracy is in fact the exceptional point discussed previously that marks the crossover from strong to weak coupling. The doped IFC from Fig. 1f is a curve (solid red line) on the $\omega_s$ surface, passing about the EP and traversing from strong to weak coupling (white dotted lines) in the two-dimensional limit as the mode evolves from HP3 to SP3 in the finite slab.

Nano-imaging and tunability. A scattering-type scanning near-field optical microscope (s-SNOM) was used to image polaritons and evaluate their energy-momentum ($\omega$, $q$) dispersions (Methods). s-SNOM is a tapping-mode atomic force microscope with a metallized tip illuminated by laser light. The sharp metallic s-SNOM tip launches propagating polaritons (Fig. 1a) that reflect off of sample edges and defects. The launched and reflected waves interfere and form standing waves, which are imaged by the scanning probe. Polariton momentum and dissipation, quantified by real and imaginary $q = q' + iq''$, respectively, were extracted from observed wavelengths $\lambda = \pi q' / q''$ and propagation lengths $L = 1/q''$ (Methods). We used oxidized tungsten diselenide (WO$_x$) to dope graphene and bilayer WSe$_2$ samples with $E_F \approx 0.60$ eV and $E_F \approx 0.45$ eV are shown in Fig. 3a, b, respectively. The [001] direction, which prohibits polariton propagation in bare $\alpha$-MoO$_3$ at this

Fig. 1 Doping-induced topological transition by plasmon-phonon hybridization. a Graphene/$\alpha$-MoO$_3$ device schematic in a scattering-type scanning near-field optical microscope (s-SNOM). Inset shows an optical microscope image of a sample. Graphene doping is achieved via charge transfer with WO$_x$. The s-SNOM tip acts as a dipole antenna, launching polaritons in all permitted directions with in-plane angle $\theta$. Without doping, there is no propagation along [001] direction and the, b wavefront and, c isofrequency contour (IFC) are hyperbolic. Upon doping, propagation is allowed along the [001] direction and the, d wavefront and, e isofrequency contour of the first-order mode are closed curves. f Semi-analytical IFC undergoing topological transition upon doping. g Along the [100] direction, polaritons have out-of-plane propagation constant $k_z$ with Re $k_z \gg$ Im $k_z$ such that the out-of-plane electric field is oscillatory like a waveguide mode (HP3, left inset). Along the [001], Im $k_z \gg$ Re $k_z$, and polaritons are surface waves with decaying electric field (SP3, right inset). h Plasmon mode splitting ($\omega_{pl} \rightarrow \omega_+ + \omega_-$) caused by the [100]-$\alpha$-MoO$_3$ phonon as a function of $q$ and $\theta$. The plot reveals an exceptional point (EP). The doped IFC from f is a curve on the $\omega_s$ surface (red line). The IFC passes about the EP, indicating a transition from strong to weak coupling. White dotted lines are guides to the eye. i Scattering rates $\Gamma_+$ and $\Gamma_-$ of $\omega_+$ and $\omega_-$ modes are anticorrelated (compare red and pink lines).
frequency, now hosts an SP$^3$. We observe a significant tunability of the [001] mode by modifying $E_F$ (Fig. 3c), while the wavelength of the [100] mode changes only slightly (compare blue and red lines). The degree of anisotropy between orthogonal directions can thus be tuned dramatically by modifying $E_F$. Both [100] and [001] modes disperse as $\omega$ is tuned from 875–915 cm$^{-1}$ (Fig. 3d), in agreement with the calculated dispersion (Fig. 3e). Note that SP$^3$ modes merge with the first-order hyperbolic phonon mode of the lower reststrahlen band below $\omega_{LO}$, becoming HP$^3$ modes and distinguishing them from pure plasmons. We remark that [001] modes in Fig. 3b are reflected from a physical boundary of the WO$\_x$/graphene layer that is 8° from the [001] direction.

We now expound on the exact topological transition point in graphene/$\alpha$-MoO$_3$ and the role of dissipation. Figure 4a shows the IFC evolving with doping from hyperbola to peanut to extended circle. The peanut-shaped IFC (blue contour in Fig. 4a) goes from concave to convex shape around the open angle $\theta_\ell$. Divergence of the wavevector $q_\ell$ at $\theta_\ell$ marks the topological transition, assuming no other $q$ diverges first, since bounded and unbounded curves cannot be homeomorphic. $q_\ell$ increases as the graphene conductivity decreases (Fig. 4b), diverging at $\sigma_g = 0$ in an approximate analytical equation neglecting in-plane losses (dashed lines):

$$q_\ell = \frac{1}{4} \left( 1 - \zeta + \sqrt{(1 - \zeta)^2 + 8\zeta} \right), \quad \zeta = \frac{2e^2}{\hbar d q_p},$$

where $q_p = i\omega/2\pi\sigma_g$ (Supplementary Note 1). Upon incremental doping away from zero conductivity, the IFC changes topology as momenta above $\theta_\ell$ gain large but finite values. In this picture, the topological transition point is exactly at $\sigma_g = 0$ since even marginally-conducting IFCs will be bounded and only the hyperbola at $\sigma_g = 0$ will be unbounded. Full numerical calculations (squares in Fig. 4b, Equation S5) support this trend up to low conductivities.

In practice, [001] modes will not be observable until appreciable Fermi energy $E_F$ with modest $\sigma_g$ because propagation lengths become shorter, or $q''$ increases, as $E_F$ decreases. In Fig. 4c, $q''$ plotted in polar coordinates is the imaginary counterpart of the IFC referred to as the loss contour. Loss contour calculations consider finite thickness $\alpha$-MoO$_3$ with intrinsic material scattering rate $\gamma = 4$ cm$^{-1}$ and intraband-only graphene with doping-independent scattering rate $\gamma_g = 40$ cm$^{-1}$. Loss contours show $q''$
hole doping and, there is a splitting at the imaginary part of the doped sample. The exact topological transition and the existence of SP3/HP3 hybrid modes will thus depend on the spatial resolution and the thickness of the graphene layer.

Fig. 9). Additionally, at very low doping, plasmons pass the interband transition threshold and are overdamped. Nonlocal scattering rate in the two-dimensional approximation (red line) moves from stronger coupling where \( \frac{\omega}{\Gamma} \approx \Gamma_{\alpha} \), to weaker coupling (blue line) where \( \frac{\omega}{\Gamma} \approx \Gamma_{g} \). This is consistent with weakness of plasmon-phonon coupling and the properties of hybridized polariton modes by modifying doping level and excitation frequency. Further, our nano-imaging data augmented with calculations reveal an intimate relationship between plasmon-phonon coupling and the properties of hybridized hyperbolic polaritons. Graphene/\( \alpha \)-MoO3 integrated into a gated structure will serve as a nanophotonic platform for engineering devices with dynamical isofrequency topologies and directional plasmon-phonon coupling. Furthermore, patterning WOx/graphene on \( \alpha \)-MoO3 could inspire new forms of photonic crystals with subwavelength periodic changes in isofrequency topology. Also, combining a graphene/\( \alpha \)-MoO3 device with an ultrafast laser may permit picosecond switching of isofrequency topology for time-varying metasurfaces.

Final note: the authors became aware of the following relevant works by Álvarez-Pérez et al.32; Bapat et al.33; Hu et al.34; and Zeng et al.35 after completion of this work.

Methods

Sample fabrication. High-quality WOx-doped graphene/\( \alpha \)-MoO3 heterostructures were fabricated using a polycaprolactone polymer-based dry transfer technique. First, we exfoliate \( \alpha \)-MoO3 bulk crystals purchased from a materials supplier (HQ Graphene) onto SiO2/Si substrates. A monolayer or bilayer WSe2 on graphene heterostructure is then transferred onto a predetermined \( \alpha \)-MoO3 flake. Finally, we treat samples in a UV-ozone generator (Jelight UVO-cleaner) at 300 K to oxidize the topmost WSe2 monolayer into WOx.

Scanning near-field optical microscopy. A Neaspec neaSNOM near-field microscope was used with a DRS Daylight Solutions Hedgehog tunable quantum

increasing along all directions as \( E_p \) decreases, consistent with finite-difference time-domain simulations (Supplementary Fig. 8). The exact \( E_p \) at which [001] modes are first observable, initiating this practical topological transition, will thus depend on the spatial resolution and the thickness of the \( \alpha \)-MoO3 slab (Supplementary Fig. 9). Additionally, at very low doping, plasmons pass the interband transition threshold and are overdamped. Nonlocal and other corrections to the graphene conductivity will also become important.

Loss contours at intermediate doping (e.g., solid blue line in Fig. 4c) reveal that \( \gamma' \) decreases as the wavevector rotates away from the [001] direction (imagine black arrow rotating with length matching the loss contour). Dissipation can thus be reduced relative to unhybridized graphene (dashed blue line) by changing propagation direction since \( \gamma < \gamma' \). The same holds for the scattering rate \( \Gamma = v_g \gamma' \), where \( v_g \) is the group velocity (Fig. 4d). The scattering rate in the two-dimensional approximation (red line in Fig. 11) provides some intuition on where the excess dissipation goes. Recall that the \( \alpha \)-MoO3 phonon splits the graphene plasmon into two modes \( \omega_{g} \) and \( \omega_{\alpha} \) with respective scattering rates \( \Gamma_{g} \) and \( \Gamma_{\alpha} \). As the wavevector angle \( \theta \) rotates in Fig. 11, the \( \omega_{g} \) loss contour (red line) moves from stronger coupling where \( \Gamma_{g} < \Gamma_{\alpha} \), through a degeneracy where \( \omega_{g} = \omega_{\alpha} \), to weaker coupling where \( \Gamma_{g} > \Gamma_{\alpha} \). The \( \omega_{g} \) loss follows the opposing path (pink line) with \( \Gamma_{g} \) increasing when \( \Gamma_{\alpha} \) decreases and vice versa such that \( \Gamma_{g} + \Gamma_{\alpha} = y' \gamma_{g} \) for fixed \( y' \) and \( y_{g} \).

In summary, we have demonstrated an electronically-induced topological transition and the existence of SP3/HP3 hybrid modes in a doped graphene/\( \alpha \)-MoO3 heterostructure by direct imaging of polaritons using an s-SNOM. Our observations are consistent with full calculations for the electrodynamics of coupled plasmon-phonon modes. Also, we demonstrated the tunability of hybrid polariton modes by modifying doping level and excitation frequency. Further, our nano-imaging data augmented with calculations reveal an intimate relationship between plasmon-phonon coupling and the properties of hybridized hyperbolic polaritons. Graphene/\( \alpha \)-MoO3 integrated into a gated structure will serve as a nanophotonic platform for engineering devices with dynamical isofrequency topologies and directional plasmon-phonon coupling. Furthermore, patterning WOx/graphene on \( \alpha \)-MoO3 could inspire new forms of photonic crystals with subwavelength periodic changes in isofrequency topology. Also, combining a graphene/\( \alpha \)-MoO3 device with an ultrafast laser may permit picosecond switching of isofrequency topology for time-varying metasurfaces.
occurs when conductivity modulated by the Fermi energy. The topological transition and can be tuned slightly by moderate doping.

θ away from the [001] direction. The undoped loss contour (purple) diverges at contour at 0.4 eV shows in a pseudoheterodyne detection scheme38.

mode scattering rate

lumerical.com/products/fdtd/). We used a point dipole source positioned 20 nm of about 40 nm in contact. The signal localized under the apex of the tip is isolated Arrow tips with 75 kHz resonance frequencies were used with tapping amplitudes 6 NATURE COMMUNICATIONS | https://doi.org/10.1038/s41467-022-31477-z | www.nature.com/naturecommunications cascade laser TLS-SK-41112-HHG (set the zero-frequency component to the center. The boundary conditions were set to the perfectly matched layer option. The NumPy fast Fourier difference time domain (FDTD) solver for Maxwell

Electrodynamics simulations. Simulations of the electric field distributions on graphene/MoO3 heterostructures were performed with the Ansys Lumerical

data fitting procedure. To extract polariton momenta \(q\) and dissipation \(\gamma\) from near-field amplitude images, it is necessary to fit a damped oscillatory function to line profiles. In this work, we use a zeroth-order Hankel function of the first kind \(H_0^\alpha\) of the following form:

\[
S_x = |AH_0^\alpha(2(q' - i\gamma)(x - x_0))| + B(x - x_0) + C, \tag{M1}
\]

where the fitting parameters are \(A, B, C, q'\) and \(\gamma\). The factor of two in the argument of the Hankel function comes from the fact that we are observing interference fringes between tip-launched and edge-reflected polaritons. The \(x_0\) value was fixed to match the position of the first peak, which was then cut off during fitting so that contributions from higher-order modes may be neglected.

Raman spectroscopy. Raman measurements were conducted on a commercial Bruker Senterra system using an excitation wavelength of 532 nm at 2 mW with a 60 second integration time. A grating of 1200 line/mm was used to attain an energy resolution of 3–5 cm\(^{-1}\).

Data availability

Relevant data supporting the key findings of this study are available within the article and the Supplementary Information file. All raw data generated during the current study are available from the corresponding authors upon request.

Received: 17 August 2021; Accepted: 31 May 2022; Published online: 28 June 2022

References

1. Poddubny, A., Iorsh, I., Belov, P. & Kivshar, Y. Hyperbolic metamaterials. Nat. Photonics 7, 948–957 (2013).

2. Jacob, Z., Smolyninov, I. & Narimanov, E. Broadband Purcell effect: radiative decay engineering with metamaterials. Appl. Phys. Lett. 100, 181105 (2012).

3. Guo, Y., Cortes, C. L., Molesky, S. & Jacob, Z. Broadband super-Planckian thermal emission from hyperbolic metamaterials. Appl. Phys. Lett. 101, 131106 (2012).

4. Liu, Z., Lee, H., Xiong, Y., Sun, C. & Zhang, X. Far-field optical hyperlens magnifying sub-diffraction-limited objects. Science 315, 1686 (2007).

5. Abedini Dereshgi, S. et al. Lithography-free IR polarization converters via orthogonally in-plane phonons in α-MoO3, flakes. Nat. Commun. 11, 5771 (2020).

6. Yao, J. et al. Optical negative refraction in bulk metamaterials of nanowires. Science 321, 930 (2008).

7. Dai, W. et al. Tunable phonon polaritons in atomically thin van der Waals crystals of boron nitride. Science 343, 1125–1129 (2014).

8. Caldwell, J. et al. Sub-diffractional volume-confined polaritons in the natural hyperbolic material hexagonal boron nitride. Nat. Commun. 5, 5221 (2014).

9. Ambrosio, A. et al. Mechanical detection and imaging of hyperbolic phonon polaritons in hexagonal boron nitride. ACS Nano 11, 8741–8746 (2017).

10. Ma, W. et al. In-plane anisotropic and ultra-low-loss polaritons in a natural van der Waals crystal. Nature 562, 557 (2018).

11. Zheng, Z. et al. A mid-infrared biaxial hyperbolic van der Waals crystal. Sci. Adv. 5, eaaw6905 (2019).

12. Basov, D. N., Fogler, M. M. & Garcia, F. J. de Abajo, Polaritons in van der Waals materials. Science 354, aag1992 (2016).

13. Low, T. et al. Polaritons in layered two-dimensional materials. Nat. Mater. 16, 182–194 (2017).

14. Basov, D. N., Asevero-Garcia, A., Schuck, P. J., Zhu, X. & Rubio, A. Polariton panorama. Nanophotonics 10, 549–577 (2020).

15. Sun, F. et al. Polariton waveguide modes in two-dimensional van der Waals crystals: an analytical model and correlative nano-imaging. Nanoscale 13, 4845–4854 (2021).

16. Hu, G. et al. Topological polaritons and photonic magic angles in twisted α-MoO3 bilayers. Nature 582, 209–213 (2020).

17. Chen, M. et al. Configurable phonon polaritons in twisted α-MoO3. Nat. Mater. 19, 1307–1311 (2020).

18. Duan, J. et al. Twisted nano-optics: manipulating light at the nanoscale with twisted phonon polariton slabs. Nano Lett. 20, 5323–5329 (2020).

19. Zheng, Z. et al. Phonon polaritons in twisted double-layers of hyperbolic van der Waals crystals. Nano Lett. 20, 5301–5308 (2020).

20. Basov, D. et al. Graphene on hexagonal boron nitride as a tunable hyperbolic metamaterial. Nat. Nanotechnol. 10, 682–686 (2015).

21. Duan, J. et al. Enabling propagation of anisotropic polaritons along forbidden direction via a topological transition. Sci. Adv. 7, abf2690 (2021).

22. Zhang, Q. et al. Hybridized hyperbolic surface phonon polaritons at α-MoO3, and polar dielectric interfaces. Nano Lett. 21, 3112–3119 (2021).

23. Krishnamoorthy, H. N. S., Jacob, Z., Narimanov, E., Kretschmar, I. & Menon, V. M. Topological transitions in metamaterials. Science 366, 205–209 (2021).

24. Fei, Z. et al. Gate-tuning of graphene plasmons revealed by infrared nano-imaging. Nature 487, 82–85 (2012).

25. Park, S. H., Xia, S., Oh, S.-H., Avouris, P. & Lou, T. Accessing the exceptional points in a graphene plasmon-vibrational mode coupled system. ACS Photonics 8, 3241–3248 (2021).

26. Choi, M. et al. High carrier mobility in graphene doped using a monolayer of tungsten oxyxenide. Nat. Electron. 4, 731–739 (2021).

27. Kim, B. S. Y. et al. Atomically imprinted graphene plasmonic cavities. Preprint at https://arxiv.org/abs/2206.12754 (2022).

28. Rizzo, D. J. et al. Charge-transfer plasmon polaritons at graphene/α-RuCl3 interfaces. Nano Lett. 20, 8438–8445 (2020).

29. Wunsch, B., Stauber, T., Solis, F. & Guinea, F. Dynamical polarization of graphene at finite doping. N. J. Phys. 8, 318 (2006).
Acknowledgements
The authors wish to thank S.H. Park and T. Low at the University of Minnesota for helpful discussions related to exceptional points in plasmon-phonon coupled systems. Research at Columbia University is solely supported as part of Programmable Quantum Materials, an Energy Frontier Research Center funded by the U.S. Department of Energy (DOE), Office of Science, Basic Energy Sciences (BES), under award DE-SC0019443. WSe$_2$ synthesis was supported by the Center on Precision-Assembled Quantum Materials, funded through the US National Science Foundation (NSF) Materials Research Science and Engineering Centers (award no. DMR-2011738). D.N.B. is Moore Investigator in Quantum Materials EPIQS #9455. The Flatiron Institute is a division of the Simons Foundation.

Author contributions
F.L.R. and B.S.Y.K. contributed equally to this work. F.L.R. performed the analysis and s-SNOM measurements. B.S.Y.K., A.R., and S.L. fabricated the heterostructures. Z.S. developed the semi-analytical model. D.J.R. and A.S.M. assisted with the analysis, and D.J.R. performed Raman measurements. A.J.M., J.C.H., and D.N.B. are the principal investigators who helped direct the course of the research.

Competing interests
The authors declare no competing interests.

Additional information
Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s41467-022-31477-z.

Correspondence and requests for materials should be addressed to Francesco L. Ruta or D. N. Basov.

Peer review information Nature Communications thanks the anonymous reviewers for their contribution to the peer review of this work.

Reprints and permission information is available at http://www.nature.com/reprints

Publisher’s note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made. The images or other third party material in this article are included in the article’s Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article’s Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this license, visit http://creativecommons.org/licenses/by/4.0/.

© The Author(s) 2022