Spectroscopy of a tunable moiré system with a correlated and topological flat band

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Moiré superlattices created by the twisted stacking of two-dimensional crystals can host electronic bands with flat energy dispersion in which enhanced interactions promote correlated electron states. The twisted double bilayer graphene (TDBG), where two Bernal bilayer graphene are stacked with a twist angle, is such a moiré system with tunable flat bands. Here, we use gate-tuned scanning tunneling spectroscopy to directly demonstrate the tunability of the band structure of TDBG with an electric field and to show spectroscopic signatures of electronic correlations and topology for its flat band. Our spectroscopic experiments are in agreement with a continuum model of TDBG band structure and reveal signatures of a correlated insulator gap at partial filling of its isolated flat band. The topological properties of this flat band are probed with the application of a magnetic field, which leads to valley polarization and the splitting of Chern bands with a large effective g-factor.

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A moiré superlattice with flat electronic bands was first discovered by stacking two layers of graphene on top of each other at a precise angle\(^1\). The flat band in magic angle twisted bilayer graphene (MATBG) emerges from the interplay between interlayer hybridization and monolayer graphene electronic structure and displays a plethora of correlated and topological phenomena. At partial band filling, MATBG shows a cascade of correlated insulating and superconducting phases, the underlying mechanisms of which are still being investigated\(^2\). A number of different Chern insulator phases, driven by either alignment of the MATBG with hexagonal BN or intrinsic strong correlations in such flat bands have also been found\(^25\). These discoveries motivate interest in other moiré systems and search for novel ways to create and control flat bands within them. A particularly promising moiré system is twisted double bilayer graphene (TDBG), in which the tunability of Bernal bilayer graphene band structure with an electric field can be exploited to create a tunable moiré system\(^22\). Previous theoretical and experimental studies have demonstrated that application of an electric field can isolate a flat band with a bandwidth of \(10–20\) meV\(^23\) in TDBG. The electric field tunability makes it possible to realize this flat band in a wide range of twist angles (0.8°–1.53°) between the stacked bilayers, instead of at a specific angle as for the case of MATBG\(^23,24\).

Corroborating this picture, previous transport experiments have shown that a robust correlated insulator appears in a confined electric field range at densities that correspond to half filling of a flat band in TDBG\(^22\). However, direct measurements of the band structure of this system and direct demonstration of its tunability, which underlies the interpretation of transport studies, has not been thus far performed.

We characterize the electronic structure of TDBG using scanning tunneling microscopy (STM) and spectroscopy as a function of carrier density and in the presence of a magnetic field. STM and gate-tuned scanning tunneling spectroscopy (GT-STS) have become a powerful tool to probe the properties of moiré systems, as its application to MATBG has provided critical information on the nature of flat bands in that system. STM experiments have not only confirmed the theoretical picture of the single particle band structure of MATBG but also have uncovered signatures of strong electronic correlation and topology at partial band filling of this system\(^7\)–\(^10,19\). Applying these techniques to TDBG not only demonstrates its tunability but also provides spectroscopic signatures of correlation and topology of its flat bands that complements transport studies.

**Results and discussion**

Figure 1a shows the schematic diagram of our device geometry, which we use to probe the electronic properties of TDBG samples, in a home built ultra-high vacuum STM at a temperature \(T = 1.4\) K (see Methods for sample fabrication and STM measurement). Using STM imaging, we first obtain information on the structure and symmetry of the TDBG’s moiré superlattice in our devices. Figure 1b shows an STM topography image, from which we can measure the periodicity and distortion of the moiré lattice to determine the twist angle of 1.48° and a very small hetero-strain of 0.1% for this TDBG sample. Higher resolution STM images of TDBG samples (Fig. 1b inset) show the topmost graphene layer’s triangular atomic lattice, which signifies the sensitivity of our experiments to the electronic properties of this layer (out of four) in this multilayer system. STM images also reveal the breaking of \(C_3\) symmetry—a salient feature of TDBG, which is in contrast preserved in MATBG. This broken symmetry can be visualized by filtering the atomic lattice from STM images (Fig. 1b) and displaying the results in the moiré unit cell, as shown in Fig. 1c. From the STM topographic height in such images, we identify three different locations, ABCB, ABCA, and ABAB, within the unit cell that correspond to different stacking configurations of the atoms between the layers in our sample. While the locations corresponding to the AA stacking between the middle layers appear higher in STM images (Fig. 1b, c), there is also a measurable contrast in the height between AB or BA stacking locations as well. These locations are related by the \(C_2\) symmetry of the moiré lattice, which is broken in TDBG. The broken \(C_2\) symmetry is a distinguishing property of TDBG and is responsible for the gap between the conduction and the valence band at the Dirac points of its band structure, as well as the unique band topology of TDBG.

Spectroscopy and visualization of the electronic states of TDBG over a wide range of energies reveal the basic band structure of this system and validates the continuum model that has been used to understand its properties. Figure 1e shows the STS measurement at the charge neutral point (CNP) that displays four distinct peaks corresponding to the van Hove singularities (vHs) of the four lowest energy bands of this system. The excellent agreement between these measurements and the density of states calculated by the continuum model (Fig. 1f and Supplementary Material) allows us to attribute the four identified bands to the first conduction and valence bands: C1, V1, and the second conduction and valence band: C2, V2 (Fig. 1d). Further corroborating for this assignment can be found by comparing the spatial dependence of the electronic states measured using STM conductance maps at energies corresponding to vHs of these bands and the maps of the electron distribution on the top graphene layer for the corresponding bands from the continuum model (Supplementary Material). Figure 2 shows an example of this comparison illustrating that the points of high density of states for V2, C1, and C2 bands are located on the ABCB moiré sites, while V1 displays a different pattern with its maximum density of states on ABCA sites. There is not only good agreement between the experimental results and the continuum model but also such maps clearly visualize the broken \(C_2\) symmetry in TDBG by displaying the difference between the density of states at ABAB and ABCA sites.

The tunability of TDBG’s band structure can be demonstrated by the measurements of its spectroscopic properties as we vary the gate voltage of the silicon gate \(V_{SiG}\) in our devices. In similar previous experiments on gated devices with STM, such as those on MATBG (for example ref. \(^{11}\)), varying \(V_{SiG}\) allowed us to study the properties of such two-dimensional systems as a function of carrier concentration. However, for studies of TDBG, it is important to recognize \(V_{SiG}\) tunes both the carrier concentration \(n\) and the electric field \(D\) simultaneously: \(n = C V_{SiG}/e + n_0, D = C V_{SiG}/2 + D_0\), where \(C\) is the capacitance per area between TDBG and the silicon gate, \(n_0\) and \(D_0\) are the intrinsic doping and the built-in electric field. Measurements of the tunneling spectra display a rich array of features that shift systematically as a function of \(V_{SiG}\), as shown in Fig. 3a. First, we highlight that changes of doping in TDBG caused by the \(V_{SiG}\) results in clear jumps of the vHs marked by dashed white lines, indicating the Fermi energy is passing through a band gap. From these jumps, we identify the gate voltages corresponding to CNP (\(V_{SiG} = -3.5\) V), full occupancy of the conduction band (72.5 V), and full occupancy of the valence band (–79.5 V). The appearance of CNP near zero gate voltage reveals that samples are not doped by impurities or by a significant work function difference between the sample and the STM tip. Furthermore, we find the gate voltage ranges required to occupy the conduction band and the valance band are identical: \(\Delta V_{SiG} = 76\) V, which is also consistent with the carrier density needed to fill a moiré band of the 1.48° TDBG: \(n_S = 5.08 \times 10^{12} \text{cm}^{-2}\) based on a parallel plate capacitance.
Fig. 1 STM on TDBG. **a** Schematic of the STM measurement setup on TDBG devices. **b** STM topograph of TDBG, obtained with bias setpoint $V_{\text{set}} = -400$ mV, current setpoint $I_{\text{set}} = 20$ pA and gate voltage $V_{\text{GC}} = -88$ V. Top right inset shows magnified image (1 nm size) with top graphene layer lattice in a Bernal bilayer. The bright spots in the inset are carbon atoms. **c** Filtered topograph removing atomic lattices to emphasize different stacking orders of TDBG plotted in the moiré unit cell. The colored circles on the right represent carbon atoms on A and B sublattices of each layer. **d** Calculated band structure of TDBG with a twist angle of $\theta = 1.48^\circ$ under zero potential difference between the top and bottom graphene layers ($U = 0$). **e** Tunneling spectrum of 1.48° TDBG device at charge neutrality, measured with $V_{\text{set}} = -400$ mV, $I_{\text{set}} = 400$ pA and AC modulation of $V_{\text{mod}} = 3$ mV. **f** Density of state on the top graphene layer calculated by a continuum model under zero electric field.

Fig. 2 Visualizing Electronic States of TDBG. **a** Tunneling spectrum at $V_{\text{SiG}} = -50$ V and $T = 1.4$ K, measured with $V_{\text{mod}} = 5$ mV, $V_{\text{set}} = -200$ mV, $I_{\text{set}} = 400$ pA. **b**–**e** Conductance maps for V2, V1, C1 and C2 bands, taken at $T = 1.4$ K, $V_{\text{SiG}} = -50$ V, $V_{\text{mod}} = 5$ mV. The voltage setpoint of each map is set to the spectrum peaks in **a** at $V_{\text{set}} = -103$, $-35$, $32$, and $91$ mV, while the current setpoint is chosen to keep the tip at roughly the same height as **a**. The plotted data was smoothed with a 0.8 nm radius filter. The diamond shapes represent a moiré unit cell. **g**–**j** Calculated electron density distribution on the top graphene layer for the four bands. $U = -40$ meV is used for this calculation. Shown electron densities are summed over each band (Supplementary Material), which appears to capture the data under these STM setup conditions. The diamonds match with the ones in **b**–**e**. Different stacking orders are shown by different colored symbols.
almost eliminated by large electric fields, as displayed at the top and bottom portions of Fig. 3a. These gaps reduce in a symmetric manner toward the positive and negative gate voltages, signifying that the built-in electric $D_0$ is close to zero. These findings are consistent with previous transport studies of TDBG and show that the band structure of TDBG can be tuned with an electric field. In addition to these gaps, we identify a smaller gap between C1 and V1 bands at charge neutrality $A_{\text{CNP}}$, which behaves in an asymmetric fashion with respect to the sign of the $V_{\text{SiG}}$. At positive values of $V_{\text{SiG}}$, $A_{\text{CNP}}$ is first reduced in magnitude to zero but it reopens again with increasing $V_{\text{SiG}}$ (Fig. 3a, d), in agreement with transport experiments at this twist angle.\(^{23}\) In contrast, $A_{\text{CNP}}$ monotonously increases in magnitude as $V_{\text{SiG}}$ is made more negative (Fig. 3a, c). This asymmetry with respect to $V_{\text{SiG}}$ is due to the polarizing effect of the electric field, which induces changes in the spatial distribution of electronic states across the different atomic layers of TDBG. Our experiments are sensitive to this asymmetry because they are sensitive to the electronic properties of the topmost graphene layer in TDBG. In fact, comparing our experimental results of the gate-tuned measurements of tunneling spectra to the continuum model (Supplementary Material), we find the calculated density of states for the top graphene layer (Fig. 3b) from this model shows a remarkable resemblance to our data (Fig. 3a), including the observed asymmetric behavior with respect to $V_{\text{SiG}}$. In these calculations, the tuning of electric field $D$ is modeled by the adjustable parameter $U$, which is the onsite potential difference between the top and bottom graphene layer, spread across the first graphene layers (Supplementary Material). Furthermore, the continuum model allows us to understand why we do not observe the closing of $A_{\text{CNP}}$ at negative $V_{\text{SiG}}$—it actually closes, but the signature appears only in the density of states of the bottom layer, which is not being probed by our experiment (Supplementary Fig. 1). One might notice a slight discrepancy between the model and the experiment, that there is a small overlap between V1 and C1 in the calculated band structure (Figs. 1d and 3b), in contrast to the clear CNP gap seen in our experiment and transport experiment.\(^{23}\) In fact, with our choice of model parameters, a CNP gap at $D = 0$ and its closing and opening with the application of $D$ is predicted for a twist angle larger than 1.52°. This minor discrepancy can thus be explained by electron interactions, coupling with the substrate or inaccurate model parameters. Lastly, we find that the bandwidth of C1 in both experiment and model calculation is smaller than that of V1, which explains why correlated insulators have only been discovered in the conduction band and establishes the setting for the correlation signatures to emerge in our experiments.

The influence of strong electronic correlation in the TDBG system can be examined in higher resolution gate-tuned spectroscopy measurements of the C1 and V1 bands. Figure 4a shows such measurements in which we find a number of different features as the occupation of the C1 band is tuned. First, we note the abrupt jump in vHs of C1 at CNP due to the CNP gap, the closing of which can be identified near $V_{\text{SiG}} \approx 20 \, \text{V}$ in this higher resolution measurement. Second, as the C1 is being occupied, we also observe a sudden increase in the broadening of vHs of the fully occupied V1 band, reminiscent of similar features seen in such measurements of MATBG and understood as a results of strong correlation in that system.\(^8\) Third and most intriguingly, increasing the occupation of C1 to half filling (at $V_{\text{SiG}} = 34.5 \, \text{V}$), where transport studies have uncovered a correlated insulator, we observe a sudden jump of the vHs of this band (Fig. 4a–c and Supplementary Figs. 2 and 3). This jump sometimes appears together with a suppression of the density of states at the Fermi level, the measurements of which we find to be sensitive to the location of the tip and also likely to its work function (inset Fig. 4a and discussion on Figs. 2 and S3 in Methods). Examining the behavior of vHs more closely by tracking its location in energy at different $V_{\text{SiG}}$, we extract the magnitude of its jumps $\Delta$ at different twist angle areas. Consistent with correlation gaps extracted from transport studies at half filling of the C1 band, we find the magnitude of $\Delta$ increases from 0.38 to 1.3 meV as the twist angle decreases from 1.48° to 1.43°. The observation of jump-like features in our data is not expected from a simple insulating gap, such as observed single particle gaps observed at CNP and full occupancy of C1 and V1 bands. Its shape is however reminiscent of similar measurements on MATBG near every
quarter fillings of that system\textsuperscript{11,12} and requires further measurements and theoretical analysis.

Lastly, we show that topology of the band structure of the TDBG band can be revealed by examining its spectroscopic properties in a magnetic field. In the presence of an electric field $D$, the C1 band of TDBG in our theoretical model carries a topological valley Chern number $C_{\text{v}} = 2$, meaning Chern number $C = 2$ in one valley and $C = -2$ in the opposite valley. Since a Chern band generates orbital magnetic moments, it would couple strongly to a perpendicular magnetic field. In Fig. 5a, we show a gate-dependent STS measurement under a perpendicular magnetic field of 4 T. Compared with zero field data (Fig. 4a), the most salient feature is the splitting of the vHs of the C1 band. This splitting persists over a large range of densities and occurs below Fermi level, thus is likely due to single particle band effect. It is in contrast to the splitting induced by interactions, which occurs on the Fermi energy near half filling. The behavior of the splitting broadly agrees with the expectation of a topological valley Chern band. Since the two valleys of the C1 band carry opposite Chern numbers and orbital g-factors, they shift in energy under a magnetic field in the opposite directions by $\pm g_{\text{orb}} B$, where $g_{\text{orb}}$ is the Bohr magneton and $g$ is the magnitude of the orbital g-factor (Fig. 5d, e). While in general g-factors depend on crystal momentum, here for simplicity we discuss the average g-factor\textsuperscript{26}. Thus, the magnetic field split the valley-degenerate vHs of the C1 band under zero magnetic field into two vHs of different valleys separated by $2g_{\text{orb}} B$. Figure 5b and Supplementary Fig. 4 shows the evolution of C1 splitting as a function of perpendicular magnetic fields. Although the energy of the C1 peaks move irregularly between different fields, possibly caused by the tip position change when changing the magnetic field, the separation between the two peaks evolves consistently. Extracting this separation, we find they collapse onto lines with very large g-factors of $g = 13$ and 21 for half filling and three-quarter filling (Fig. 5c). The large g-factor rules out the possibility that the splitting is caused by spin Zeeman effect, since transport studies under in-plane magnetic field consistently find spin g-factor close to two\textsuperscript{22-26}. From the continuum model calculation, orbital g-factor averaged in the mini-Brillouin zone is between 6 and 7 for most U (Supplementary Material), which is a factor of two times smaller than the experimental value. However, the orbital g-factor calculation sensitively depends on the energy landscape within the narrow C1 band, which may be altered by interactions that are not accounted for in the single particle picture. Without knowing the exact shape of the band, a simple picture below may provide a rough estimate of the g-factor amplitude for a general $C_{\text{v}} = 2$ band. Since the CNP gap possesses valley Chern numbers, at the $K_1$ ($K_2$) valley, the CNP gap would shift in energy from the bottom (top) edge of C1 (V1) band at zero magnetic field to the top (bottom) of C1 (V1) band at half flux quantum per moire unit cell\textsuperscript{33} (Fig. 5e). If the vHs of C1 roughly shift along with the CNP gap, and if we assume the CNP gap is much smaller than the bandwidth of C1 and V1, the splitting between vHs of the C1 band in two valleys produced by the field would be on the order of $B/B_{1/2} \approx (w_c + w_v)$, where $B_{1/2} = 26.3$ T is the magnetic field of half flux quantum per moire unit cell and $w_c (w_v)$ is the bandwidth of the C1 (V1) band. Assuming the bandwidth of C1 and V1 are on the order of 20 meV, this formula produces a g-factor $g = 13$, roughly agreeing with our observation. Beside the splitting, we also resolve a discontinuity of the vHs just below $V_{\text{sig}} = 20$ V, where the CNP gap collapses in Fig. 4a, and a jump of vHs near three-quarter filling of the C1 band. These features and other topological signatures of this system that are revealed in the present of a magnetic field\textsuperscript{33}, can be explored in future studies of this system.

**Methods**

**Sample preparation.** TDBG samples are stacked on a hexagonal Boron Nitride (hBN) layer on SiO$_2$/silicon substrate and electrically contacted with gold electrodes.
The samples are fabricated by tearing and stacking an exfoliated Bernal stacked bilayer graphene. We fabricated our samples in similar ways to published STM results on MATBG8,11. Our pickup stamp is made from a polyvinyl alcohol (PVA) coated transparent tape, which cover a polydimethylsiloxane (PDMS) block on a glass slide by standard lock-in method with AC modulation at 4 kHz.

STM measurements. The experiment is done in a home built UHV STM with (6,1,1) vector magnet operating at T = 1.4 K. The measurements are performed with a tungsten tip prepared and characterized on a Cu(111) single crystal. Through controlled indentation, we shape the tip until its poke mark is confirmed and its spectrum featuring the Cu(111) surface state at the right energy. We then locate the TDBG sample with the capacitance guiding technique34.

The topography is taken under constant current mode, and the differential conductance is acquired by standard lock-in method with AC modulation at 4 kHz.

All spectrums presented in this study with the exception of Fig. 2a are measured with a freshly prepared tip on the first landing spot before any scanning. We note that the current generation of samples have some residue contamination, which sometimes modifies the spectrums after scanning to obtain images of the sample. Supplementary Fig. 5a is spectrum measured with a freshly prepared tip on the SiO2 substrate.

Discussion on Supplementary Figs. 2 and 3. In Fig. 4a–c and Supplementary Figs. 2 and 3, we show several GT-STS measurements that provide information on the variation of such measurements at different areas of the same device as well as different locations within the moiré unit cell. Supplementary Fig. 2 shows a different type of data showing the signatures of correlations in the broadening and the visibility of the gap at the Fermi level in C1 can be different when measured on different tops of the same device.
different locations or with different tip conditions. In Supplementary Fig. 3, we include more GT-STS measurements presented together with STM topographs that show areas 1–4. The location of the corresponding measurements within the unit cell are marked on these topographs (Supplementary Fig. 3a–d). Spectrums in Supplementary Fig. 2 (Supplementary Fig. 3b), which shows two peaks of C1 band, with one peak crossing the Fermi level and further split around the half filling, appear to the very different from other data set (Supplementary Fig. 3a–c). The difference could be caused by the different locations where the data is taken, but it could also be caused by anomalous tip condition in acquiring Supplementary Fig. 2.

Data availability
The data that support the findings of this study are available from the corresponding author upon reasonable request.

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References
1. Suárez Morell, E., Correa, J. D., Vargas, P., Paccheco, M. & Barticevic, Z. Flat bands in slightly twisted bilayer graphene: Tight-binding calculations. Phys. Rev. B 82, 121407 (2010).
2. Bistritzer, R. & MacDonald, A. H. Moiré bands in twisted double-layer graphene. Proc. Natl Acad. Sci. USA 108, 12333–12337 (2011).
3. Cao, Y. et al. Correlated insulator behaviour at half-filling in magic-angle graphene superlattices. Nature 556, 80–84 (2018).
4. Cao, Y. et al. Unconventional superconducting in magic-angle graphene superlattices. Nature 556, 43–50 (2018).
5. Yankowitz, M. et al. Tuning superconductivity in twisted bilayer graphene. Science 363, 1059–1064 (2019).
6. Lu, X. et al. Superconductors, orbital magnets and correlated states in magic-angle bilayer graphene. Nature 574, 653–657 (2019).
7. Choi, Y. et al. Electronic correlations in twisted bilayer graphene near the magic angle. Nat. Phys. 15, 1174–1180 (2019).
8. Xie, Y. et al. Spectroscopic signatures of many-body correlations in magic-angle twisted bilayer graphene. Nature 572, 101–105 (2019).
9. Jiang, Y. et al. Charge order and broken rotational symmetry in magic-angle twisted bilayer graphene. Nature 573, 91–95 (2019).
10. Kerelsky, A. et al. Maximized electron interactions at the magic angle in twisted bilayer graphene. Nature 572, 95–100 (2019).
11. Wong, D. et al. Cascade of electronic transitions in magic-angle twisted bilayer graphene. Nature 582, 198–202 (2020).
12. Zondiner, U. et al. Cascade of phase transitions and Dirac revivals in magic-angle graphene. Nature 582, 203–208 (2020).
13. Stepnov, P. et al. Untying the insulating and superconducting orders in magic-angle graphene. Nature 583, 375–378 (2020).
14. Saito, Y., Ge, J., Watanabe, K., Taniguchi, T. & Young, A. F. Independent superconductors and correlated insulators in twisted bilayer graphene. Nat. Phys. 16, 926–930 (2020).
15. Arora, H. S. et al. Superconductivity in metallic twisted bilayer graphene stabilized by WSe2. Nature 583, 379–384 (2020).
16. Liu, X. et al. Tuning electron correlation in magic-angle twisted bilayer graphene using Coulomb screening. Science 371, 1261–1265 (2021).
17. Sharpe, A. L. et al. Emergent ferromagnetism near three-quarters filling in twisted bilayer graphene. Science 365, 605–608 (2019).
18. Serlin, M. et al. Intrinsic quantized anomalous Hall effect in a moiré heterostructure. Science 367, 900–903 (2020).
19. Nuckolls, K. P. et al. Strongly correlated Chern insulators in magic-angle twisted bilayer graphene. Nature 588, 610–615 (2020).
20. Wu, S., Zhang, Z., Watanabe, K., Taniguchi, T. & Andrei, E. Y. Chern insulators and topological flat-bands in magic-angle twisted bilayer graphene. Nat. Mater. https://doi.org/10.1038/s41563-020-00991-2 (2021).
21. Saito, Y. et al. Hofstadter subband ferromagnetism and symmetry-broken Chern insulators in twisted bilayer graphene. Nat. Phys. https://doi.org/10.1038/s41567-020-01129-4 (2021).
22. Shen, C. et al. Correlated states in twisted double bilayer graphene. Nat. Phys. 16, 520–525 (2020).
23. Liu, X. et al. Tunable spin-polarized correlated states in twisted double bilayer graphene. Nature 583, 221–225 (2020).
24. Cao, Y. et al. Tunable correlated states and spin-polarized phases in twisted bilayer–bilayer graphene. Nature 583, 215–220 (2020).
25. Burg, G. W. et al. Correlated insulating states in twisted double bilayer graphene. Phys. Rev. Lett. 123, 197702 (2019).

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Author contributions
X.L., C.C., and A.Y. designed the experiment. X.L. and C.C. fabricated the samples. X.L., C.C., and G.F. performed the STMs measurements and analyzed the data. J.Y.L., A.V., and X.L. conducted the theoretical calculations. K.W. and T.T. provided hBN crystals. X.L., C.C., J.Y.L., and A.Y. wrote the paper with input from all authors.

Competing interests
The authors declare no competing interests.

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