<u>Name and DOI of manuscript:</u> A quasi-2D polypyrrole film with band-like transport behavior and high charge carrier mobility, https://onlinelibrary.wiley.com/doi/full/10.1002/adma.202303288

Data availability

Data measured by the Banerji group in agreement with data management policies (SNF, Horizon 2020...) and shown in the main manuscript is made publicly available in the BORIS repository of the University of Bern. For raw data, S.I. data and data acquired by collaborators, please contact the authors (available upon request).

<u>Data acquisition</u>: Details on the methods of data acquisition are described in the above manuscript and the corresponding S.I.

Data and analysis:

Figure 1

Figure 1. Schematic illustration and reaction scheme of the preparation of q2DPPy film on a concentrated H2SO4 surface.

Figure 1 data: no data. The figure contains a reaction scheme made by Chemdraw by Kejun Liu, and a schematically drawing of a 3D layered structure made by Peng Zhang Person who made it: Kejun Liu and Peng Zhang Reference to lab book: null

Figure 2

Morphological and structural analysis of the q2DPPy film. (a) SEM image from a top-view direction. (b) SEM image and (c) AFM image from a cross-section direction with inserted images of exfoliated flake in the top-right coner of the images. (d) 2D GIWAXS pattern of q2DPPy. (e) AC-HRTEM image of q2DPPy with inserted SAED pattern (upper-left); and a simulated (bottom-left) HRTEM image matching the experimental image; the proposed chemcial quinodal PPy chain structure is overlapped in the image with lattice parameters. (f) SAED pattern and (g) PPy chain stacking schematic from the cross-section direction of the film.

Figure 2 data: a) SEM image from a top-view direction. From TU Dresden by Kejun Liub) SEM image from a cross-section direction. From TU-Dresden. By Kejun Liu

c) AFM image from a cross-section direction from TU-Dresden by Kejun Liu

d) 2D GIWAXS pattern of q2DPPy from TU Dresden by Mike Hambsch using beamline XRD1 at Elettra Sincrotrone Trieste.

e) and (f) and (g)AC-HRTEM image and simulation: from University of Ulm, by Haoyuan Qi

Person who measured: Kejun Liu, Mike Hambsch, Haoyuan qi Reference to lab book: in lab journals of people from other institutes, no exact information.

Figure 3

Characterization of the chemical composition of q2DPPy (a) FTIR spectrum showing characteristic peaks of functional groups of q2DPPy. (b) Raman spectrum of q2DPPy. (c) Ultraviolet-visible-near-infrared (UV-VIS-NIR) absorbance spectrum obtained with an integrating sphere to minimize scattering effects during our measurements. (d) High-resolution XPS spectra of C1s and (e) of N1s.

Figure 3 data: a) Fig3a_FTIR.txt

b) Fig3b_Raman.txt

c) Fig3c_UV-VIS-NIR ABS.txt

Person who measured: Kejun Liu (FTIR, UV-VIS-ABS), Raman (Sezer Seçkin), XPS (Xiaodong Zhuang, shanghai jiao tong university)

Reference to lab book: record in Dresden

Figure 4

Charge transport study of the q2DPPy film. (a) Semilogarithmic plot of electrical conductivity measured by the vdP method versus T^{-1/4} over the measured temperature range, with a linear fit using the Mott law variable-range hopping model between 300K and 250K. (b) Temperature-dependent conductivity from the THz and vdP methods. For clarity, one data point was selected for every 2K interval by extracting the real part of THz-TDS conductivity between 0.8 and 1 THz. (c) Electronic band structure along the Γ -X path line in the first Brillouin zone for a single protonated fully-extended quinoidal chain. (d) THz-TDS traces of the time-resolved electrical field intensity (solid lines) and reference THz pulse (blank, dashed line) at temperatures ranging from 240 K to 298 K under inert gas (N₂) conditions. (e) Complex THz conductivity spectra at 299K, 290K, 280K, 269K, 250K and 241K shown in different colours. The Drude-Smith model fits to their real (square symbol **I**) and imaginary (circle symbol •) parts are represented as dashed lines. (f) Effective THz mobility at various temperatures according to $\mu_{eff} = q\tau/m_o(1 + c_1)$, with the inserted figure showing the c_1 value at different temperatures.

Figure 4 data: a) Figure4a_Semilogarithmic plot of electrical conductivity .txt

b) Figure4b_Conductivity by vdP method at various temperatures and THz.txt

c) from Yingying Zhang, just a image no data

d) Figure4d_THZ absorption.txt

e) Figure4e_THZ_conductivity.txt

f) fitting parameters

Person who measured: Kejun Liu , Julien Réhault (THZ), vdP (YX Zhou)

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