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Approaching the Monolayer Limit of Carbon Layers by Pyrolysis of Polymer Films

Natalie Galfe¹  | Florian Herdl¹  | Sebastian Klenk¹  | Moritz Quincke²  | Cormac Ó Coileáin¹  | Kangho Lee¹  | Ute Kaiser²  | Georg S. Duesberg¹ 

¹Institute of Physics & Center for Integrated Sensor Systems (SENS), University of the Bundeswehr Munich, Neubiberg, Germany | ²Central Facility Electron Microscopy, Materials Science Electron Microscopy, Ulm University, Ulm, Germany

Correspondence: Georg S. Duesberg (duesberg@unibw.de)

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ABSTRACT

Research on monolayer materials remains at the forefront of materials research. Here, we present a systematic study of graphenic carbon layers focusing on their structural evolution and electrical properties as film thickness approaches the atomic limit. The ultrathin carbon films are obtained from the pyrolysis of photoresist films (PPF) directly on the target substrate, also allowing structuring by lithographic means. Thus, pre-defined graphenic structures can be realized with controlled thickness, down to the sub-nanometer scale as determined by atomic force microscopy. X-ray photoelectron spectroscopy confirms the predominant sp^2 hybridization of our films, transmission electron microscopy reveals domains with hexagonal atomic structure, and Raman spectroscopy shows signatures of evolving nanocrystallinity with decreasing film thickness until dimensional confinement imposes a lower limit. We further demonstrate the functionality of the sub-nanometric pyrolyzed polymer film as a chemiresistive NO_2 sensor. The films' scalability and patternability across multiple length scales, together with their chemical inertness and biocompatibility, make them promising candidates for future applications.

1 | Introduction

The structural diversity of pyrolyzed carbon materials, ranging from highly ordered graphite to fully amorphous carbon, has intrigued scientists for more than half a century. In the early 1950s, Rosalind Franklin distinguished between graphitizing and non-graphitizing carbon through X-ray diffraction and density measurements, establishing that the latter forms disordered, layered structures without long-range periodicity [1, 2]. This insight laid the foundation for materials such as glassy carbon, whose structure, particularly in the case of polymer-derived forms, was later described by Jenkins and Kawamura as a dense,

isotropic, conductive carbon formed from polymeric precursors under inert conditions [3–5].

Decades later, 2D carbon materials such as graphene or monolayer amorphous carbon (MAC) gained prominence for their exceptional electronic and mechanical properties [6, 7]. However, challenges in manufacturing devices with monolayer graphenic layers remain, including high temperatures, catalytic substrates, and multiple handling steps such as polymer-based transfers. Recent studies have explored scalable, process-compatible carbon layers derived from pyrolyzed polymer films (PPF) [8–10]. Here, we use the term “graphenic” to describe ultrathin carbon films

Abbreviations: AFM, atomic force microscopy; FFT, fast Fourier transform; (HR-)TEM, high resolution transmission electron microscopy; MAC, monolayer amorphous carbon; PF, polymer film; PPF, pyrolyzed polymer film; SNR, signal-to-noise ratio; TK, Tuinstra-Koenig [relation]; TLM, transfer length method; XPS, X-ray photoelectron spectroscopy.

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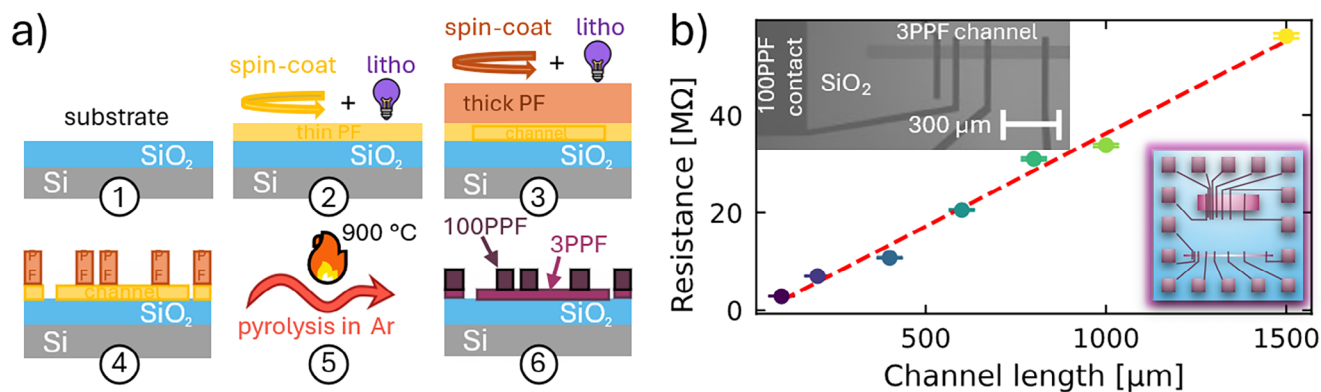


FIGURE 1 | (a) Workflow diagram for PPF-based devices. A thin polymer film (PF) is deposited and patterned lithographically ② to define channels. A thick PF is spun on top and patterned ③ to define contacts of transfer length structures with varying contact spacings. A joint development step ④ is followed by pyrolysis ⑤, which yields conductive devices composed of ultrathin 3PPF and thicker contact 100PPF ⑥. (b) The resistance of a narrow 3PPF channel depending on the length between contacts in a linear manner. The inset (top left) shows an electron micrograph of the shortest channels of the 100 μm wide device, and the other one (bottom right) shows the full layout of a representative die (out of 61) featuring two channels with different widths (1 mm and 100 μm).

containing predominantly graphene-like sp^2 domains without long-range crystalline order, distinguishing them from bulk “graphitic” materials, which require 3D stacking periodicity. These films are increasingly regarded as promising candidates for applications in nanoelectronics, sensing, and surface functionalization, where process integration is critical, and process simplicity can provide an added advantage.

In this work, we synthesize ultrathin PPFs by spin coating of diluted photoresist and subsequent pyrolysis. This two-step process avoids transfer steps and metal catalysts. The resulting carbon films are continuous over full 4-inch wafers and electrically conductive even at the monolayer limit. Characterization by atomic force microscopy (AFM), Raman spectroscopy, and transmission electron microscopy (TEM) reveals a nanocrystalline phase with lateral domains on the few-nanometre scale.

To demonstrate functionality and assess electrical performance, these sub-nm PPFs were structured directly by lithography and contacted with carbon leads. With this methodology, we also realized a chemiresistive sensor with high response to nitrogen dioxide (NO₂) measured at room temperature and atmospheric pressure.

2 | Material System and Fabrication Approach

Resistive PPF-based devices were manufactured using a highly scalable and integratable process that is compatible with current semiconductor production lines and has been previously reported to manufacture gate layers in high-performing planar hot electron emitters [11].

The fabrication sequence is summarized in Figure 1a, in which AZ nLOF 2070 is applied in a spin coating process with varying degrees of dilution. Table 1 lists the three PPFs employed in this work, including their designated names for distinction and the corresponding dilution levels. The resulting, already thin layer was patterned via UV lithography using a photomask. A post-exposure bake was performed to ensure crosslinking of the resist

TABLE 1 | Overview over studied pyrolyzed polymer films (PPFs) with corresponding precursor dilution and resulting thickness.

Name	Dilution in AZ EBR [wt % nLOF 2070]	Avg. thickness after pyrolysis [nm]
3PPF	3	0.34 ± 0.1
5PPF	5	1.1 ± 0.5
100PPF	100 (undiluted)	65 - 75 here, center: 73.5 ± 1

and stabilization of the polymer framework [12]. The second layer of undiluted AZ nLOF 2070 was spin-coated directly on top of the first and repatterned via UV lithography to define contact electrodes from 100PPF as shown in the process flow in Figure 1a. Due to their greater thickness, the resulting contact paths exhibit significantly increased conductance compared to the thinner channel regions. Both layers were developed simultaneously in AZ 2026 MIF. Full 4-inch Si/SiO₂ wafers were processed into devices featuring a transfer length method (TLM) layout, as illustrated by the inset on the bottom right in Figure 1b, exemplary for one out of 61 dies in total.

Pyrolysis of the patterned resist was conducted in a custom-made furnace as shown in Figure S3a, under argon flow (~1 mbar with 200 sccm Ar). To ensure controlled carbonization and gradual evaporation of volatile, non-carbonaceous components [13], the temperature was ramped at 5 K min⁻¹ to 500 °C and then at 10 K min⁻¹ to 900 °C, where it was held for 60 min. The temperature profile of the pyrolysis process is shown in Figure S3b. The resulting PPF thickness is defined by the dilution and the spin speed used during fabrication and, later, by pyrolytic shrinkage. Higher dilution of the resist results in proportionally greater shrinkage during pyrolysis, as a larger fraction of the initial material consists of volatile components such as the dilutor [11, 14].

For TEM, films were spin-coated with a polymethyl methacrylate (PMMA) layer at 3000 rpm and baked at 120 °C for 7 min.

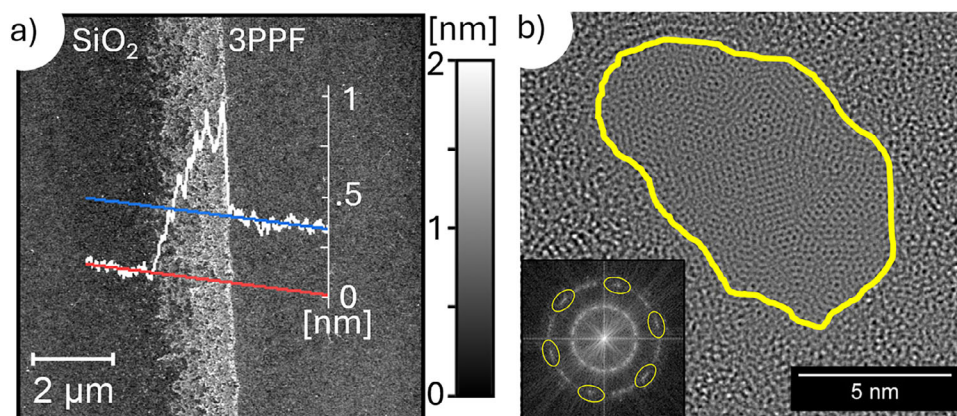


FIGURE 2 | (a) AFM of the edge of a channel of 3PPF with one exemplary line profile, revealing a height of 0.32 nm across. (b) High-resolution TEM of a 3PPF, revealing a nanocrystalline domain outlined in yellow. The inset shows the corresponding fast Fourier transform of the yellow region, with diffraction rings exhibiting a sixfold modulation of intensity maxima (highlighted by yellow ellipses), characteristic of local hexagonal ordering.

Subsequently, the die was cleaved around the region of interest, and the underlying SiO₂ layer was removed with buffered hydrofluoric acid (BHF) over approximately 1 day. After rinsing in DI water, the floating structure was transferred onto a TEM grid. The PMMA was subsequently dissolved by submersion in acetone and isopropanol.

For gas sensor tests, the device with 3PPF channels was annealed at 150°C in N₂ under ambient pressure for 2 hr to desorb gaseous surface molecules prior to placement in a custom-made sensing chamber. After purging, the devices were exposed to various gases while recording channel resistance. A source measure unit was used to monitor current variations during gas exposure at a constant voltage bias.

3 | Results and Discussion

AFM was used to determine the thickness of the films. The thinnest film, 3PPF, was measured at five locations across the wafer, as indicated by the squares in Figure 4a. On average, the 3PPF film exhibited a thickness of 0.34 nm, with slightly thinner films observed at the wafer periphery. Thus, these films represent a monolayer of sp² carbon across a full 4-inch wafer. A representative AFM scan of an outer die, centrally displaying the edge of the ultrathin 3PPF channel, is shown in Figure 2a. A representative line profile across the edge is superimposed over it. Between the SiO₂ substrate and the continuous PPF film, a rougher disorder border transition is typically observed. Likely, it originates from lithographic residues that are not ideally dissolved by the developer. Similar measurements across the 5PPF and for 100PPF only of the central die yielded a thickness of 1.1 and 73 nm on average, respectively, in good agreement with prior reports [11].

To investigate the structural ordering at the atomic level, high-resolution (HR-) TEM was performed on a free-standing 3PPF film, transferred from a die of the same wafer onto a TEM grid. The film is fully stable when suspended as previously reported for MAC films [7]. As shown in Figure 2b, a nanocrystalline domain of approximately 5 nm lateral size is observed, with hexagonal ordering visible in the corresponding fast Fourier

transform (FFT) [15, 16]. This sixfold modulation in the FFT resembles patterns reported for Ni-assisted “glassy graphene” films [17], yet is observed here without a catalyst. We note that quasi-periodic contrast may also arise from Moiré effects due to local overlap or bilayer regions, implying locally higher crystallinity. This structural motif is consistent with nanographitic sp² domains embedded in a disordered matrix [18–20]. Nevertheless, it is possible that such domains extend over larger areas throughout the film, but are being camouflaged by residual carbonaceous contamination [21, 22]. On the other hand, beam-induced graphitization during imaging cannot be completely ruled out; however, this scenario would be inconsistent with our AFM, X-ray photoelectron spectroscopy (XPS), and Raman results.

Outside the locally ordered regions, the film shows filamentous structures reminiscent of classic glassy carbon and pyrolyzed resins (Figure S4a) [5, 20, 23, 24]. Nevertheless, such contrast should be interpreted cautiously, as similar morphologies can also arise from polymeric residues (e.g., the PMMA) or environmental contaminants. FFTs of these regions show diffuse ring features (cf. Figure S4a inset), indicating short-range order without extended periodicity. In light of this, the interpretation of these filamentous regions as structural motifs of the PPF remains tentative. This apparent coexistence of ordered and disordered regions—whether intrinsic or artefactual—mirrors the structural duality described in previous TEM studies on pyrolyzed carbon systems [25, 26]. This interpretation remains preliminary, as the observed contrast features cannot be unambiguously assigned based on TEM alone. Taken together, these findings are consistent with the structural complexity frequently reported in pyrolyzed carbon systems and show strong similarity to features observed in recently reported MAC films [7]. Overall, our preliminary TEM findings align well with complementary data and suggest the presence of both nanocrystalline and disordered domains within the ultrathin PPF.

XPS was used to assess and to quantify the sp²/sp³ hybridization ratio across the PPFs of different thickness. C 1s core-level spectra (Figure 3a) were deconvoluted into sp² carbon, sp³ carbon (C–C, ~285.0 eV), and minor components corresponding to C–O, C=O, and originating from π – π^* transitions. All samples were

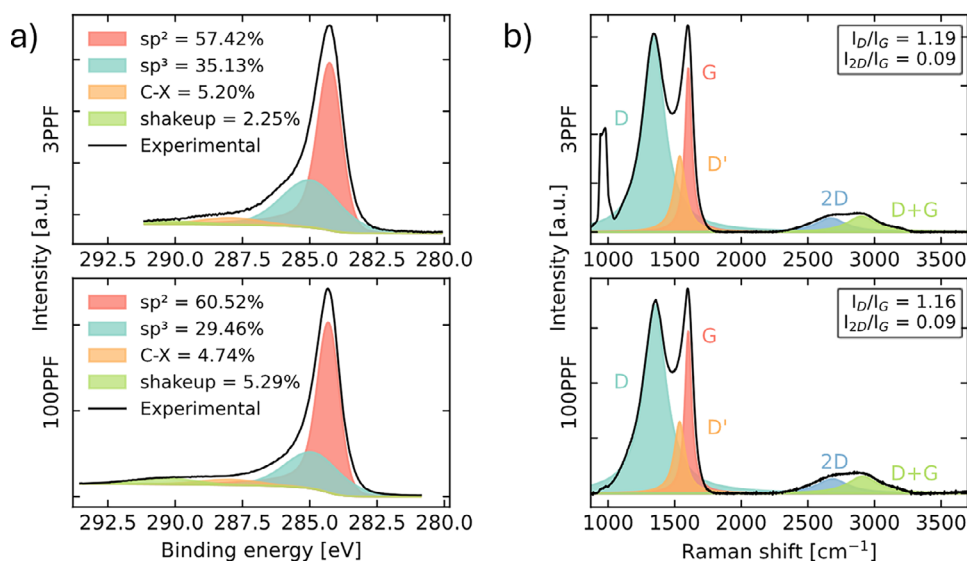


FIGURE 3 | (a) X-ray photoelectron spectra of the channel and contact regions of 3PPFs and 100PPF showing dominant sp^2 -hybridized carbon. (b) Raman spectra of 3PPF and 100PPF showing the characteristic vibrational modes of PPF fitted with Lorentzian peaks.

dominated by sp^2 bonding, indicating efficient graphitization. However, a modest but consistent decrease in sp^2 content was observed from the thicker 5PPF (cf. Figure S5 or [11]) to the thinner 3PPF. This trend brings us to the hypothesis that six-membered ring formation becomes increasingly hindered, both sterically and energetically, when approaching the monolayer limit, leading to a more chain-like carbon structure retained from the carbon backbone of the novolac-based precursor. As discussed below, this is echoed in the Raman-derived I_D/I_G ratios. Similar correlations between defect accumulation and reduced sp^2 fraction have been reported under controlled damage conditions in pyrolyzed films [27].

Observed trace oxygenated and sp^3 components likely originate from incomplete precursor conversion or minor environmental exposure. The comparatively weak $\pi-\pi^*$ shake-up feature observed in the thinner films suggests a reduced aromaticity of conjugated sp^2 rings and a higher degree of structural disorder. The relatively higher C-X contribution, which accounts for bonding situations not attributable to either sp^2 or to sp^3 components (see Supplemental Information), observed in the thinner films is plausibly linked to their increased surface-to-volume ratio, which amplifies the influence of surface-bound residues on spectral features. In ultrathin systems, polymeric or environmental contaminants can hardly be avoided, and even trace amounts can already affect surface properties – and thus device performance [22, 28].

Raman spectroscopy provides additional insight into structural ordering, defect density, and lateral domain size. Raman maps were acquired for each of the three PPF types. Raman spectra for 3PPF and 100PPF films measured on the same wafer are shown in Figure 3b. A more detailed description of the measurement and evaluation protocol is provided in the Supplemental Information.

The G band, associated with in-plane bond stretching of sp^2 atoms in both rings and chains, appeared consistently above 1600 cm⁻¹, a range typical for nanocrystalline graphite and denoting the upper end of “stage 1” in the Ferrari–Robertson disorder model, where nanocrystalline graphite with low-to-moderate disorder is characterized by an upward G shift and a dispersive D peak following the Tuinstra–Koenig relation [29]. In contrast, “stage 2” corresponds to more disordered sp^2 carbon, with the G peak downshifting (~1510 cm⁻¹) and I_D/I_G decreasing as the number of ordered rings diminishes [30, 31]. The highest G position was observed for the 5PPF film (1604 cm⁻¹), suggesting slightly enhanced short-range order and reproducing trends reported earlier [11], consistent with a larger domain size and its superior conductivity compared to the thinner 3PPF. The G-peak widths were nearly identical across all samples (~61 cm⁻¹), indicating similar levels of bond disorder, thereby confirming that thickness does not significantly impact bond-length fluctuations. These results are in good agreement with prior studies on pyrolyzed, sp^2 -rich carbon networks [5, 19, 20].

The D peak (~1350 cm⁻¹), activated by disorder and attributed to the breathing modes of six-membered rings, appeared in all spectra – indicating the preserved aromatic character despite structural disorder [5, 30]. The I_D/I_G ratio was consistently lowest in the 5PPF (1.08), and slightly higher in both 100PPF layers (1.15). Somewhat unexpectedly, the 3PPF showed the highest ratio (1.19), despite its thinner geometry and higher G-peak position. We attribute this to the aforementioned physical limitation in ring formation at monolayer-scale thickness: the novolac-based precursor contains chain-like carbon backbones, and spatial confinement during pyrolysis may hinder cyclization into closed six-membered ring structures. As a result, a higher I_D/I_G is observed, even as the G mode – activated by both chains and rings – remains sharp [32, 33]. The modest reduction in sp^2 content in 3PPF as observed via XPS (Figure 3a) further corroborates this interpretation.

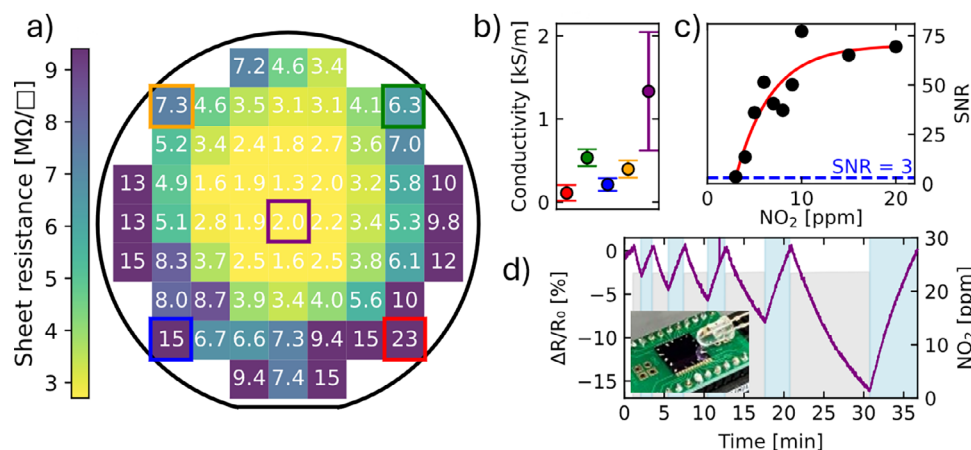


FIGURE 4 | (a) Wafer map of sheet resistance of 3PPF obtained by TLM measurements displaying lower values toward the center. (b) Conductivity derived from sheet resistance and thickness profiles measured at selected wafer positions, as indicated and color-coded by the squares in (a). (c) Signal-to-noise ratio (SNR) of sensor at various NO_2 concentrations (dots). The fitted curve (red) reveals a limit of detection of 2.99 ppm for NO_2 . (d) Resistance change versus time of an 3PPF gas sensor. Gray boxes indicate gas injections with varying exposure times, while the light blue region marks the recovery phase under UV illumination. The inset shows a 3PPF gas sensor mounted on a prototyping board with a UV-LED for gas sensor testing.

To estimate lateral crystallite size, the Tuinstra–Koenig (TK) relation was applied, which links I_D/I_G to the in-plane domain size L_a according to $I_D/I_G \propto 1/L_a$ [29]. Given all films lie at the end of “stage 1”, just above the 2 nm threshold, the relation is valid here [30, 32], and yields domain sizes of 3.7 and 4.1 nm for 3PPF and 5PPF, respectively, consistent with TEM findings.

Finally, all samples exhibited a weak but consistently present 2D band between 2670 and 2685 cm^{-1} . I_{2D}/I_G ratios remained below 0.09, much lower than for monolayer graphene. Although the 3PPF film approaches monolayer thickness, the weak and broadened 2D signal – together with its lower Raman shift at 2670 cm^{-1} – reflects disruption of coherent π -electronic band formation due to limited domain size and greater structural disorder, as expected from its isolated polymer-chain environment and confirmed by the lower L_a extracted via the TK relation. These second-order spectral features are characteristic of disordered, few-layer sp^2 carbon [19, 32, 34]. This interpretation aligns with Jurkiewicz et al., who proposed that the low intensity of the 2D bands for carbons from polymer pyrolysis, such as the PPFs studied here, arises from small lateral domain size and/or very high defect densities causing severe structural distortion [20]. Notably, the residual 2D band observed in all PPF films distinguishes them from truly amorphous carbon monolayers, such as MAC, where this feature is entirely suppressed due to the absence of long-range order and π -band coherence [7]. In summary, the elevated I_D/I_G ratio, the constant G-peak width, and the weak 2D band are consistent with the nanocrystalline character of the PPF [5, 20].

To assess the electrical performance and process uniformity of the ultrathin PPFs, wafer-scale sheet resistance measurements were performed using the TLM. Figure 4a shows the extracted sheet resistance of the 3PPF for the 61 dies of a 4-inch wafer. From each die the sheet resistance could be extracted, corresponding to a yield of 100 % which is astonishing given the monolayer nature of the channels. A slightly off-centered radial gradient is observed, with values decreasing from the edges toward the center – in line with the expected thickness variation induced by manual resist pipetting and our spin-coating setup, and

as previously reported for 5PPF on otherwise identical wafers [11]. This trend may be further reinforced by a center-to-edge temperature gradient during pyrolysis, as expected in radiatively heated cold-wall reactors. While the extracted sheet resistance spans approximately one order of magnitude in between the best and worst performing die, this variation is largely limited to few outermost dies. The overall distribution remains continuous and smooth and such experimental-stage variations could easily be eliminated through automation in an industrial setting.

Within the central 3×3 array of dies (Figure 4a), the sheet resistance averages to $(1.99 \pm 0.40) \times 10^6 \Omega/\square$. Using a film thickness of (0.34 ± 0.05) nm, as determined by AFM on the central die, the corresponding conductivity of the central array is estimated to be $(1.5 \pm 0.3) \times 10^3 \text{ S/m}$. Additional conductivity values for selected positions as indicated in Figure 4a distributed across the wafer are shown in Figure 4b for comparison. The resulting conductivities remain in reasonable agreement with earlier reports on thicker pyrolyzed resist films [14], albeit slightly reduced – by a factor of 2–3 – reflecting the incomplete aromatization and diminished charge transport pathways inherent to monolayer 3PPF. By contrast, 5PPF films have previously demonstrated conductivities approximately one order of magnitude higher [11].

To explore the gas-sensing capability of the 3PPF films, proof-of-concept experiments were carried out using one of the narrow-channel (100 μm) TLM devices. The device was manually wire-bonded onto a leaded chip carrier with silver paste and mounted into a sealed test chamber. The resistance changes of the structured ultrathin 3PPF films under atmospheric conditions were monitored by exposing it for various exposure time of 22.5 ppm NO_2 gas diluted in N_2 from 1 to 10 min. UV illumination (375 nm) was used to accelerate recovery speed of the sensors [35]. At a bias voltage of 10 V, real-time resistance changes of the 3PPF films can be displayed as

$$S = \frac{R_s - R_0}{R_0} \times 100 \text{ [%]} \quad (1)$$

where R_0 is the initial resistance before gas injection and R_s is the measured resistance after gas introduction. A typical relative percentile resistance change of sensors is depicted in Figure 4d. The 3PPF films reliably behave like a p-type material since the resistance decreased upon electron withdrawing molecule exposure [36, 37] and an empirical detection limit of 2.99 ppm was determined following periodic NO_2 exposure for 1 min (Figure 4c, as derived from Figure S2a), based on a signal-to-noise ratio (SNR) of 3, as defined by IUPAC definition [38]. While previous reports demonstrate ppt or even lower levels of NO_2 sensitivity, this detection limit is still promising considering exposure time, reliability, and stability [39–42].

The sensing results demonstrate that ultrathin PPFs can realize a robust, reversible, and repeatable gas sensor [37]. The 3PPF exhibited very limited sensitivity toward CO_2 and H_2S (cf. Figure S2b), in contrast to the heightened responsiveness observed in pristine graphene sensors [42–44]. This inherent selective sensitivity may suggest another route to the realization of selective gas sensors without additional functionalization [45].

4 | Conclusion

In this work, we have characterized PPFs and investigated the dependence of their structural properties on film thickness when downscaling to the monolayer limit for device applications. AFM has revealed layer thicknesses down to atomic scales for the thinnest 3PPF. We found that nanocrystallinity and conductivity improve as the thickness decreases when reducing from tens of nanometers to approximately one. However, further reduction into the sub-nm regime (3PPF) appears to reverse this trend: while film continuity is maintained, structural order and electronic performance slightly degrade. This structural change is consistently observed across complementary methods, including XPS, Raman, and electrical characterization. We believe that this originates from the onset of confinement-induced effects appearing in the monolayer limit.

In addition to the structural and electrical characteristics, we demonstrate the applicability of the ultrathin 3PPF film as a chemiresistive device in a sensing application for NO_2 gas detection. We demonstrated selectivity toward NO_2 over NH_3 and an excellent SNR. This proof-of-concept application highlights their practical relevance [11]. The ease of catalyst-free processing, resulting in scalability, and process reproducibility make PPFs promising candidates for future industrial applications. Additional favorable properties, such as chemical inertness and biocompatibility, pave the way for use in chemically challenging environments and biosensing applications [19].

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Conflicts of Interest

The authors declare no conflict of interest.

Data Availability Statement

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

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Supporting Information

Additional supporting information can be found online in the Supporting Information section.

Supporting File: aelm70245-sup-0001-SuppMat.docx.