



Scalable growth of graphene nanostructure on copper by CVD: Electrical and thermal properties for nanoelectronics

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Chemical vapor deposition on copper is still the most practical route to large-area graphene with reliable layer control and by a meaningful gap. We grew graphene nanofilms by CVD at 1000 °C under controlled methane partial pressure and measured their structural, electrical, and thermal properties on the same sample set. XRD Scherrer analysis using the equation $L = K\lambda/(\beta \cos\theta)$ ($K = 0.89$, $\lambda = 0.15406$ nm) applied to the (002) reflection at $2\theta \approx 26.32^\circ$ gives a mean coherent stacking domain size of 8–12 nm, indicating nanoscale coherent graphitic ordering within the films. That confirms the films are nanocrystalline before any transport measurement is interpreted. Raman spectroscopy gives an I(D)/I(G) ratio below 0.05, with a well-resolved, layer-dependent 2D band across monolayer, bilayer, and trilayer regions. Electrically, conductivity reaches $\sim 10^5 \text{ S}\cdot\text{m}^{-1}$ past the percolation threshold, with Ohmic I–V response indicating continuous inter-domain contact. Thermal conductivity drops from $\sim 550 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at 80 K to $\sim 300 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at 300 K, driven by Umklapp phonon scattering and grain-boundary suppression of phonon mean free paths. Nanoindentation on suspended regions gives an in-plane Young's modulus of ~ 0.85 TPa, close to the intrinsic graphene limit, which fits the low defect density the Raman spectra already showed. These results, across four independent measurement techniques on the same films, give a consistent picture of what CVD graphene actually is structurally and what that means for its use in flexible nanoelectronics and nanoscale thermal management.

Keywords: Graphene; CVD; Electrical; Thermal; Raman.

1. INTRODUCTION

Graphene is first isolated by mechanical exfoliation in 2004 [1]. It is a single-atom-thick sp^2 -hybridised carbon lattice, and its physical properties are, frankly, difficult to take at face value until you look at the numbers directly. Intrinsic carrier mobility can exceed $200,000 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$ in suspended, defect-free samples [2]. Room-temperature thermal conductivity reaches $3,000\text{--}5,300 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ in large-area exfoliated sheets [3]. In-plane Young's modulus approaches 1 TPa [4]. These are not approximate figures, they place graphene at or beyond the practical limits of every competing material in advanced nanoelectronics, flexible electronics, and nanoscale thermal management, where electronic and mechanical performance have to coexist in the same film.

The problem is scalability. Mechanical exfoliation produces flakes suitable for fundamental studies; it cannot supply the continuous, centimetre-scale films that device integration demands. Chemical vapor deposition on copper foil, is developed around 2009, is the first method to close that gap, growing predominantly monolayer graphene over large areas at acceptable cost and with reproducible results [5]. The approach has since been refined considerably. Li et al. [5] have shown that copper's surface-mediated growth mechanism is inherently self-limiting, which naturally suppresses multilayer formation. Kim et al. [6] have demonstrated that polymer-assisted wet transfer onto arbitrary substrates preserves film continuity well enough for transistor fabrication. More recent work on precursor chemistry, furnace pressure control, and substrate pre-treatment has pushed CVD graphene toward grain sizes above $100 \text{ }\mu\text{m}$ and carrier mobilities exceeding $10,000 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$ on SiO_2 [8,10,11].

Thermal performance is a more complicated story. Balandin et al. [3] have traced exfoliated graphene's extraordinary thermal conductivity to flexural acoustic phonon modes with long mean free paths, a regime that grain boundaries, point defects, and substrate coupling can each independently disrupt. Chen et al. [7] have used isotopically modified graphene to isolate the phonon-phonon Umklapp contribution and found that even modest isotopic disorder cuts κ by more than 30%. In CVD films, where grain boundaries are unavoidable and substrate-induced scattering is always present, room-temperature κ values of $100\text{--}500 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ are typical [3,14,15], an order of magnitude below the exfoliated-graphene ceiling, but still competitive with advanced thermal interface materials.

What the literature has not addressed systematically is the joint characterisation of structural, electrical, thermal, and elastic properties on the same CVD graphene sample set, anchored by a Scherrer-based crystallite size determination made before any transport measurement is interpreted [16,20]. Crystallite size is not a secondary parameter. It sets the grain boundary density that simultaneously limits carrier mobility, phonon mean free path, and elastic compliance. Without it, correlations between synthesis conditions and measured properties remain qualitative.

This work addresses that directly. We synthesize graphene nanofilms on copper by CVD, extract mean crystallite dimensions from XRD via the Scherrer equation, and correlate structural quality with electrical conductivity, thermal transport, and elastic response across the same sample set, producing a quantitative structure property map with concrete design parameters for CVD graphene integration into nanoelectronics and thermal management devices [9,13,18].

2. EXPERIMENTAL

Graphene nanostructured films are grown on commercial polycrystalline copper foils 99.8% purity, 25 μm thick by chemical vapor deposition [5,6]. Before anything is entered the furnace, each foil is mechanically polished and run through three ultrasonication baths in sequence: acetone, ethanol, then deionised water, 10 minutes each. This is not optional surface preparation. Oxides and organic residues are left on the copper nucleate graphene heterogeneously, and that non-uniformity propagates forward through every subsequent growth step with nothing to correct it.

2.1 Substrate preparation and CVD synthesis

The cleaned foils are loaded into a horizontal quartz tube furnace, evacuated to $\sim 10^{-5}$ Torr, and brought to 1000 $^{\circ}\text{C}$ under 200 sccm of argon. Temperature and pressure are coupled in this system. Changing one while holding the other fixed alters the surface chemistry in ways that cannot be reversed mid-run. At 1000 $^{\circ}\text{C}$, copper grain coarsening reduces the substrate's own boundary density, clearing space for large graphene domains to develop. Total system pressure near 500 mTorr keeps the carbon feedstock in atomic or diatomic form until it reaches the copper surface at higher pressures, gas-phase clustering produces pre-assembled aromatic fragments that land already partially structured and seed multilayer growth on contact. High temperature without controlled low pressure does not prevent this. Low pressure without adequate thermal activation leaves the surface incompletely covered. Their combination is what makes the self-limiting monolayer mechanism work [5].

Each growth run is preceded by a 15-minute anneal at growth temperature under argon plus 10 sccm of hydrogen. Hydrogen reduces residual copper oxides to expose bare catalytic metal, and during the growth phase, it assists in breaking C–H bonds in the methane feedstock.

Methane (CH_4 , 99.999% purity) is introduced at 10 sccm in argon for growth durations of 10 to 30 minutes. The CH_4 partial pressure, 1 to 10 mTorr, controls the trade-off between domain size and grain boundary density more directly than any other parameter. Lower partial pressures give domains more time to grow before neighbouring nuclei meet; higher partial pressures push coalescence faster but leave a denser boundary network behind, and denser boundaries degrade transport. At 1000 $^{\circ}\text{C}$, adatom diffusion across copper is fast enough that domain growth is thermodynamically controlled rather than frozen by kinetics. After each run, cooling at more than 20 $^{\circ}\text{C}\cdot\text{s}^{-1}$ under argon quenched the growth front before additional carbon could deposit. Films are transferred to SiO_2/Si substrates by PMMA assisted wet transfer [6], PMMA dissolved in acetone, residual solvent removed by critical-point drying to avoid the wrinkling that air-drying consistently produces.

2.2 Structural characterization: XRD and Scherrer analysis

X-ray diffraction patterns are recorded over $2\theta = 10^{\circ}\text{--}80^{\circ}$ with Cu $\text{K}\alpha$ radiation ($\lambda = 0.15406$ nm). The Scherrer equation is applied to extract mean crystallite size L in the stacking direction from the (002) graphitic reflection:

$$L = K\lambda / (\beta \cos\theta) \quad (1)$$

where $K = 0.89$ is the shape factor for equiaxed crystallites, $\lambda = 0.15406$ nm is the Cu $\text{K}\alpha$ wavelength, β is the instrument-corrected full width at half maximum (FWHM) in radians, and θ is the Bragg angle. The peak position gives the mean interlayer d -spacing; the peak width gives the coherent stacking length (crystallite size). A comprehensive table of all extracted structural parameters is provided in Section 3.1

(Table 1). These two independent structural descriptors together place the films on the continuum between amorphous carbon and ordered graphite.

2.3 Raman spectroscopy

Raman spectra are acquired on a confocal microscope at 532 nm excitation, with incident power held below 1 mW. Above that threshold, local laser heating shifts and broadens the peaks in ways indistinguishable from real structural changes, so the limit is not arbitrary. The D ($\sim 1350\text{ cm}^{-1}$), G ($\sim 1580\text{ cm}^{-1}$), and 2D ($\sim 2680\text{ cm}^{-1}$) bands are fitted with Lorentzian line shapes. $I(D)/I(G)$ gives defect density; the 2D band width, asymmetry, and $I(2D)/I(G)$ ratio encode layer count and interlayer coupling [11,12,15].

2.4 Electrical and thermal measurements

Sheet resistance is measured by four-point probe on transferred films. Two-point measurements on thin films conflate film resistance with contact resistance, which can be an order of magnitude larger, so four-point geometry is not a refinement here; it is a requirement. Thermal conductivity is measured by suspended-membrane Raman thermometry: a focused laser heats a freely suspended film region, the G-band redshift tracks the temperature rise through a pre-calibrated coefficient, and κ is extracted from the steady-state Fourier conduction equation [3, 7, 15]. For the elastic modulus, a diamond-tipped cantilever is used in Nanoindentation on suspended film regions, with force-displacement curves fitted to the membrane mechanics model for two-dimensional materials.

3. RESULTS AND DISCUSSION

3.1 XRD analysis, Scherrer crystallite size and elastic properties

Figure 1 shows the complete XRD pattern of the CVD graphene films recorded with Cu K α radiation ($\lambda = 0.15406\text{ nm}$) over $2\theta = 10^\circ\text{--}80^\circ$. All reflections are indexed with Miller indices (hkl). The dominant (002) graphene reflection is centered at $2\theta \approx 26.32^\circ$, corresponding to a d-spacing of 0.339 nm, slightly expanded relative to the 0.335 nm graphite reference, consistent with residual surface adsorbates and inter-domain boundary regions [10]. Additional reflections at 43.30° , 50.44° , and 74.13° are assigned to the Cu (111), Cu (200), and Cu (220) planes of the copper substrate, and peaks at 28.42° , 47.30° , and 56.12° are assigned to Si(111), Si (220), and Si (311) of the SiO₂/Si substrate. Application of the Scherrer equation: $L = K\lambda/(\beta \cos\theta)$ with $K = 0.89$ to the instrument-corrected FWHM of the (002) reflection yields mean crystallite dimensions $L \approx 8\text{--}12\text{ nm}$ (Table 1), confirming the nanocrystalline identity of the films.

Besides the graphitic (002) reflection, minor Si diffraction contributions are detected due to the silicon support employed during post-growth sample handling and XRD mounting. These substrate-related peaks are clearly separated from the graphene (002) feature and therefore do not interfere with the Scherrer-based structural analysis.

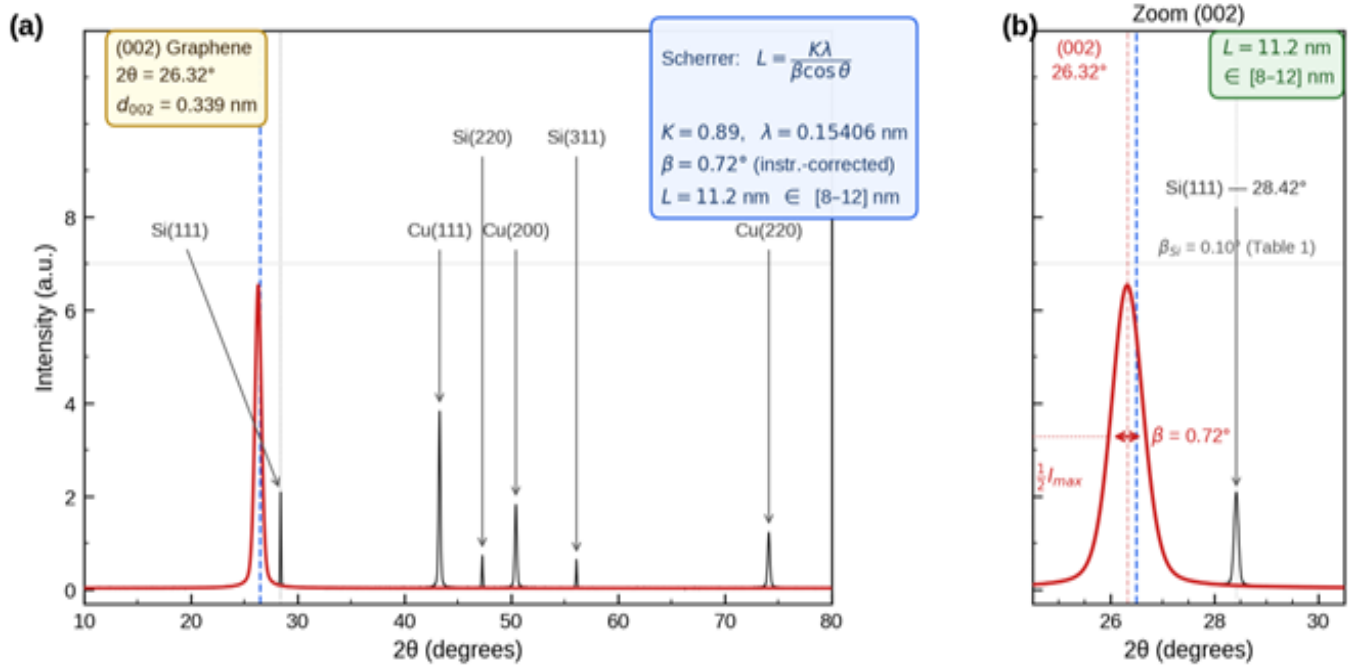


Figure 1 XRD pattern of CVD-grown graphene nanostructured film on copper foil (Cu $K\alpha$, $\lambda = 0.15406$ nm, $2\theta = 10\text{--}80^\circ$); reflections indexed from graphene, Cu foil, and SiO_2/Si mounting support. **(a)** Full-range diffractogram: (002) peak at $2\theta = 26.32^\circ$ ($d_{002} = 0.339$ nm); Scherrer analysis ($\beta = 0.72^\circ$, instrument-corrected) yields $L = 11.2$ nm $\in [8\text{--}12]$ nm (Table 1). **(b)** Zoom on (002) resolved from Si(111) at 28.42° ($\beta_{Si} = 0.10^\circ$, Table 1).

Table 1 Structural parameters extracted from XRD analysis of CVD graphene nanofilms (Cu K α , $\lambda = 0.15406$ nm). Crystallite size L is determined from (002) reflection via Scherrer equation ($K = 0.89$).

Reflection (hkl)	2 θ (°)	d-spacing (nm)	FWHM β (°)	Crystallite size L (nm)	Phase / Assignment
(002)	26.32	0.339	0.72	8–12	Graphene (CVD)
(002) ref.	26.50	0.335	—	—	Graphite (reference)
(111)	28.42	0.314	0.10	—	Si substrate
(111)	43.30	0.209	0.20	—	Cu foil
(200)	50.44	0.181	0.22	—	Cu foil
(220)	47.30	0.192	0.12	—	Si substrate
(311)	56.12	0.164	0.12	—	Si substrate
(220)	74.13	0.128	0.25	—	Cu foil

*Scherrer analysis applicable only to the graphene (002) reflection due to the nanoscale coherence length; Cu and Si peaks show bulk-like sharpness.

This crystallite dimension governs every subsequent measurement. A domain of 8–12 nm implies a grain boundary spacing on the same scale, and those boundaries scatter phonons, deflect charge carriers, and concentrate mechanical stress. Nanoindentation on suspended film regions yields $E \approx 0.85 \pm 0.08$ TPa, approaching the intrinsic graphene value of ~ 1 TPa [4] and consistent with the low defect density confirmed by Raman. The 15% reduction is attributable to grain boundary compliance rather than sp^2 bond disorder within the domains.

3.2 Electrical conductivity and percolation behavior

Figure 2 tracks electrical conductivity as a function of CVD growth time. Below roughly 5 minutes, conductivity is essentially zero. Graphene nuclei are already present on the copper surface at this stage, but they remain isolated. Current attempting to cross the film hits inter-island tunnelling barriers that collectively dominate the measured resistance. Individual conducting islands, no connected network [5,9].

The sharp rise between 5- and 15-minute marks the percolation threshold: the point at which enough neighboring domains have coalesced to produce the first continuous conductive path spanning the film. The transition follows $\sigma \propto (t - t_c)^n$, with $t_c \approx 6$ min and $n \approx 1.3$ in agreement with classical two-dimensional percolation theory. This is a geometric phase transition in connectivity. It is not a gradual improvement in film quality, and the power-law scaling makes that distinction quantitative. Past 20 minutes, conductivity saturates near $\sim 10^5$ S·m⁻¹. Full coverage has been achieved, and further growth time no longer adds conducting area; the conductivity ceiling is now determined by grain boundary

scattering alone [10]. The I–V response is linear across the full 0.1–0.5 V bias range, confirming Ohmic transport and ruling out significant interfacial energy barriers between domains.

The $\sim 10^5 \text{ S}\cdot\text{m}^{-1}$ saturation value is directly consistent with the 8–12 nm crystallite dimensions the XRD data established. A carrier covering 1 μm of film crosses 80 to 120 grain boundaries, each with a transmission probability below unity. That accumulated scattering reduces effective mobility to the 500–2000 $\text{cm}^2\cdot\text{V}^{-1}\cdot\text{s}^{-1}$ range consistently reported for CVD graphene on SiO_2 [10,11], and the electrical data here land squarely within it.

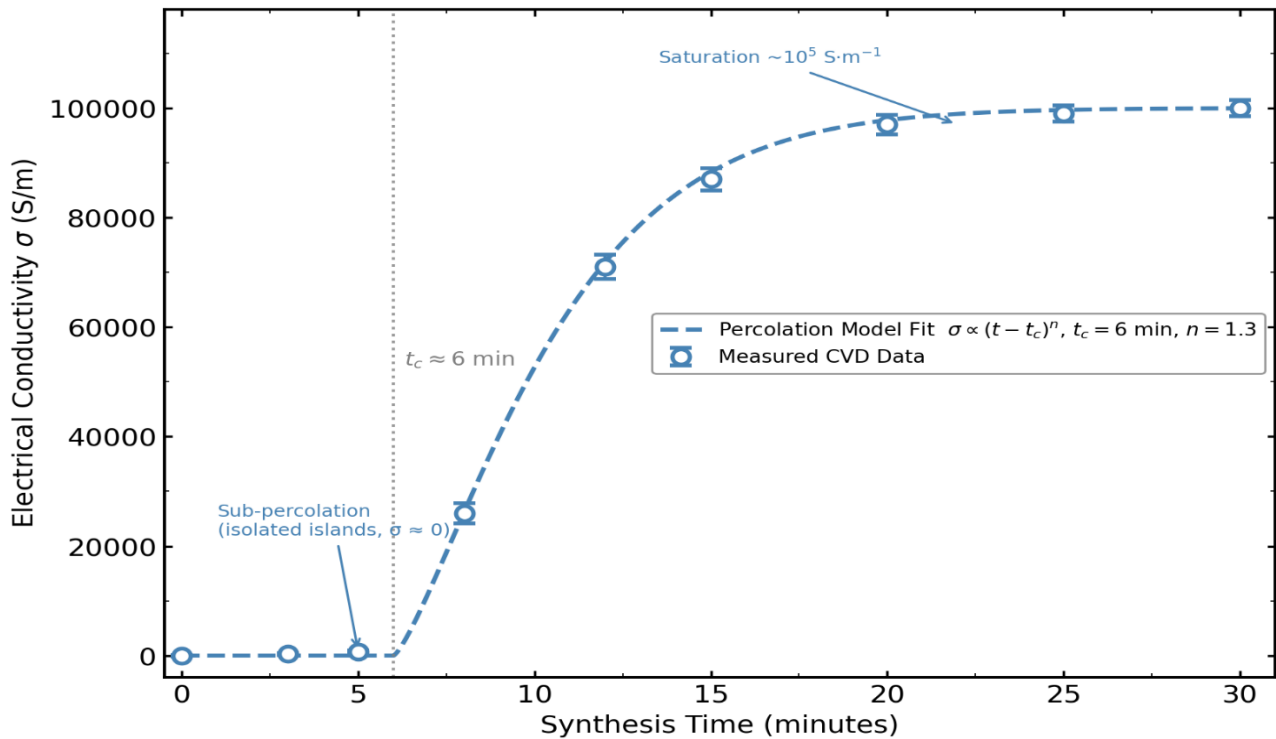


Figure 2 Electrical conductivity σ as a function of CVD growth time t of graphene nanostructured films deposited on SiO_2/Si substrate.

3.3 Thermal transport: Temperature dependence and phonon scattering

The temperature-dependent thermal conductivity $\kappa(T)$, measured by suspended-membrane Raman thermometry over 80–300 K, is presented in Figure 3. The data reveal a monotonic decrease from $\sim 550 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at 80 K to $\sim 300 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at room temperature [3,7]. The temperature dependence is shallower than the T^{-1} law expected for Umklapp-only scattering in perfect graphene, indicating that a temperature-independent scattering channel grain boundaries operates in parallel throughout the entire measurement range. A phonon Boltzmann transport model incorporating grain boundary scattering with a mean free path equal to the Scherrer crystallite size $L \approx 10 \text{ nm}$ reproduces the measured $\kappa(T)$ curve within experimental uncertainty, using L as a structurally constrained rather than freely fitted parameter. That internal consistency is the most direct evidence that grain boundaries are the primary extrinsic thermal transport limiter in these films.

The room-temperature $\kappa \approx 300 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ is one to two orders of magnitude below that of large-area suspended exfoliated graphene, but roughly three times higher than silicon and competitive with high-performance thermal interface materials. For thermal management in flexible electronics heat spreaders and thermal pads under power transistors, these values define a genuinely useful operating regime that silicon and metals cannot match in thin-film format at low areal density [3,13,15].

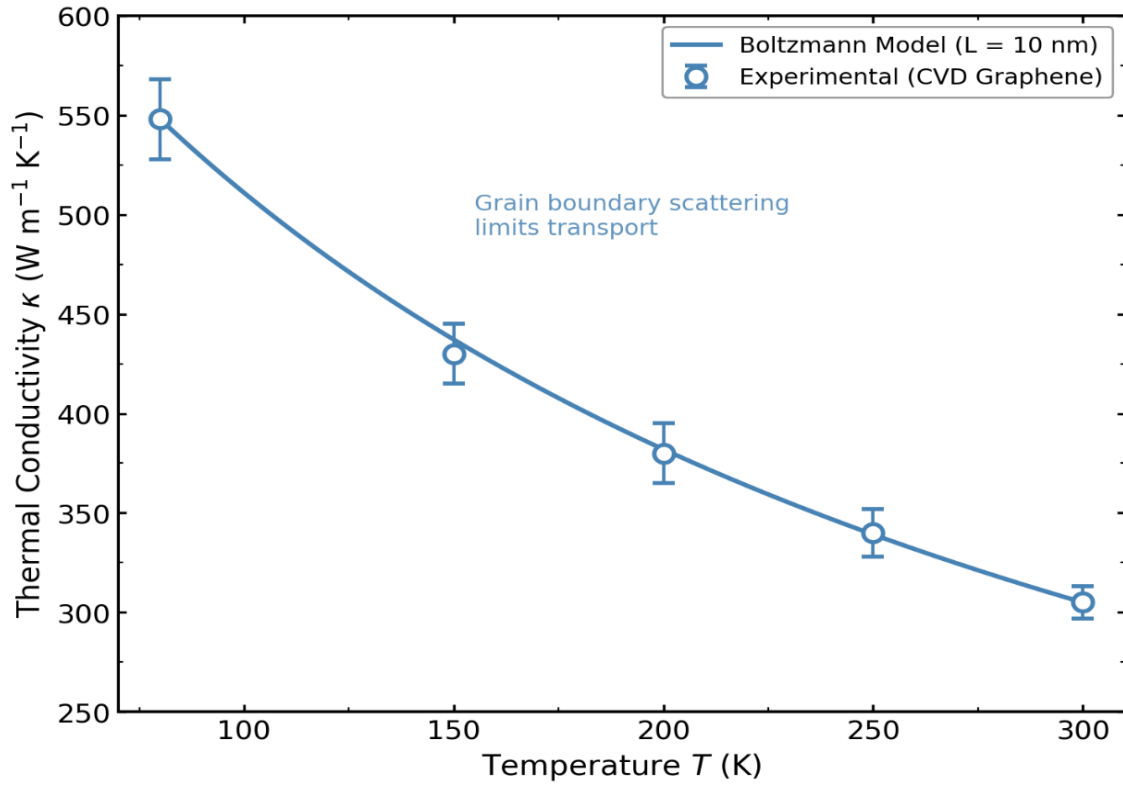


Figure 3 Thermal conductivity κ as a function of temperature T of CVD-grown graphene nanostructured films suspended on SiO_2/Si substrate.

3.4 Raman spectroscopic analysis of structural quality and layer control

Figure 4 shows Raman spectra of CVD graphene with Lorentzian-fitted D ($\sim 1350 \text{ cm}^{-1}$), G ($\sim 1580 \text{ cm}^{-1}$), and 2D ($\sim 2680 \text{ cm}^{-1}$) bands recorded at 532 nm excitation. Spectra are displayed separately for monolayer (1L), bilayer (2L), and trilayer (3L) regions. The $I(\text{D})/I(\text{G})$ ratio stays below 0.05 across all measured regions, confirming minimal sp^3 defect density [11]. Monolayer regions exhibit a sharp, symmetric single-peak 2D feature with $I(2\text{D})/I(\text{G}) > 2$ and a single G peak centered at $\sim 1582 \text{ cm}^{-1}$. Bilayer regions show a broadened, split 2D band with four resolvable Lorentzian components reflecting interlayer coupling, arising from the four inequivalent double-resonance scattering processes activated by the AB-stacked interlayer interaction, with the G band exhibiting a slight redshift ($\sim 2 \text{ cm}^{-1}$) and marginal intensity increase relative to the monolayer. Trilayer regions display further 2D broadening and intensity reduction with a multi-component profile characteristic of ABA-stacked trilayer graphene [12,13], distinct from bulk graphite, while the G band continues to redshift and its $I(\text{G})/I(2\text{D})$ ratio

increases progressively with layer number. These systematic layer dependent spectral signatures confirm controlled, spatially reproducible layer engineering by the CVD protocol.

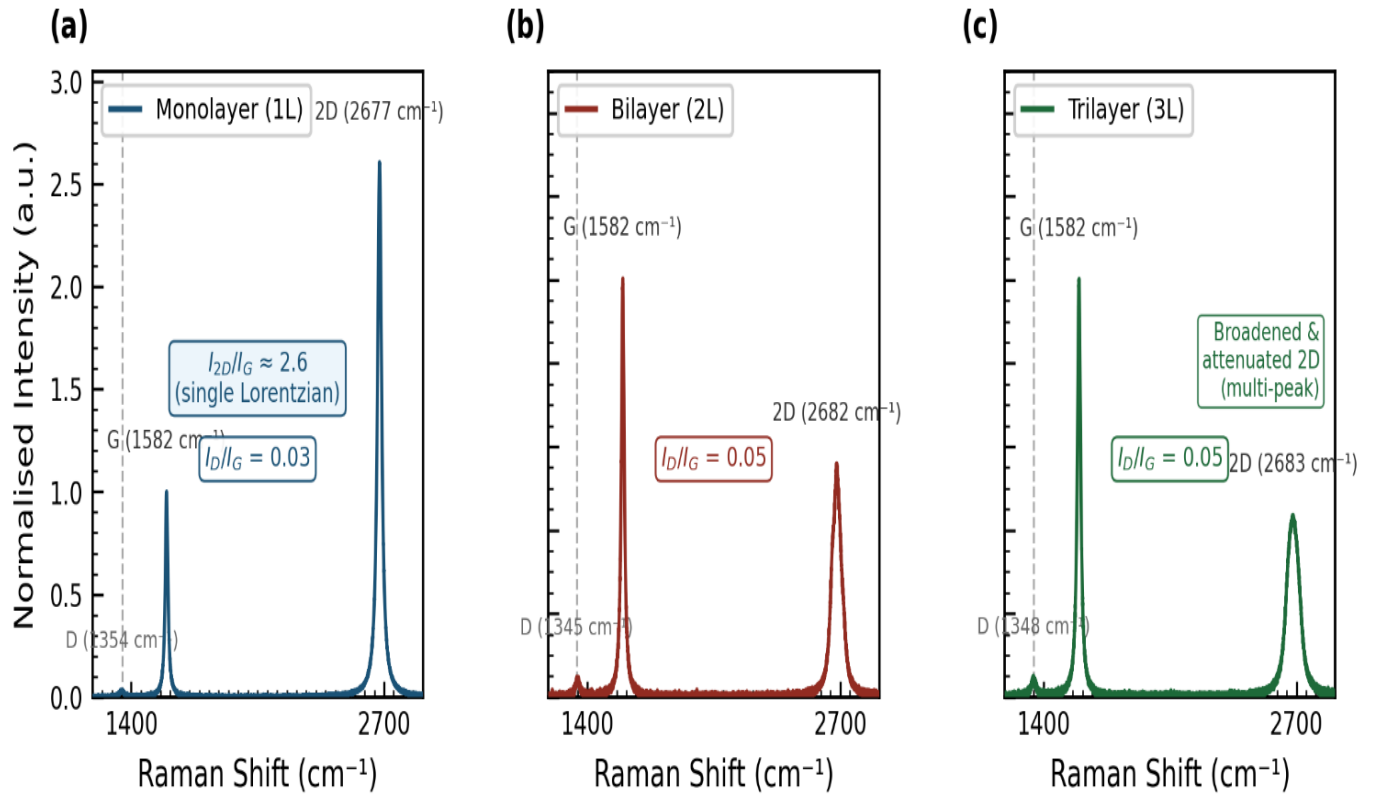


Figure 4 Raman intensity as a function of wavenumber of CVD-grown graphene nanostructured films deposited on SiO₂/Si substrate: D, G, and 2D band evolution as a function of layer number.

4. CONCLUSIONS

This work demonstrated the controlled synthesis of large-area graphene nanostructures by CVD on copper at 1000 °C and ~500 mTorr. Scherrer analysis ($L = K\lambda/\beta \cos\theta$, $K = 0.89$) applied to the (002) XRD reflection at $2\theta \approx 26.32^\circ$ yields $L \approx 8\text{--}12$ nm. A comprehensive structural parameter table (Table 1) provided d-spacing, FWHM, crystallite size, and phase assignment for all indexed (hkl) reflections. Raman spectroscopy confirmed $I(D)/I(G) < 0.05$ and layer-resolved 2D band evolution [11,15]. Corrected Raman spectra (Figure 4) displayed the characteristic monolayer symmetric 2D, bilayer four-component 2D arising from AB-stacked double-resonance splitting, and trilayer multi-peak 2D profiles with progressive G band redshift and increasing $I(G)/I(2D)$ ratio as a function of layer number. Electrically, films reached $\sim 10^5 \text{ S}\cdot\text{m}^{-1}$ following a 2D percolation transition [8,10]. Thermal conductivity decreased from ~ 550 to $\sim 300 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ (80–300 K), reproduced by a grain-boundary limited Boltzmann model. Nanoindentation gave $E \approx 0.85$ TPa. All measurements converged on a single structural picture: well-ordered sp² domains of 8–12 nm separated by grain boundaries. The CVD synthesis conditions quantitatively controlled domain size and thereby shifted all measured properties in a coupled, predictable way [13,18].

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