

Moving Beyond Scotch Tape: Scalable Transfer of Research-grade CVD Graphene

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Abstract

While scalable graphene synthesis by chemical vapor deposition (CVD) has been in widespread use since 2008, fundamental research requiring consistently high quality continues to rely upon exfoliated graphene. Here we use evaporated Ni to transfer ultrahigh quality CVD graphene from Cu(111), as continuous films or arrays of pre-defined shapes. By careful control over the evaporation conditions, we avoid damage to the graphene, preserving intrinsic quality. The dry transfer process minimizes strain and

doping. After hBN encapsulation, the CVD-grown graphene shows low-temperature magnetotransport behavior on par with the best exfoliated graphene devices. The CVD-grown graphene can be stacked to create magic-angle twisted bilayer graphene with low twist disorder. These results demonstrate that CVD-grown graphene can replace exfoliated flakes for even the most demanding applications.

Keywords: Graphene synthesis, Research-grade, Reproducible, Oxygen-free

Graphene and its moiré heterostructures are a fertile platform for fundamental condensed matter physics research, hosting phases of matter such as superconductivity [1, 2], correlated insulators [3, 4], and exciton condensates [5]; and exhibiting phenomena including hydrodynamic electrical transport [6–9], fractal bandstructure [10–12], fractional quantum Hall effects [13–15], and quantum anomalous Hall effects [16]. Because exploration of new phases and phenomena requires samples with the highest achievable quality, virtually all such research still utilizes graphene that is mechanically exfoliated from bulk crystals by the ‘Scotch tape’ method demonstrated 20 years ago [17].

Using chemical vapor deposited (CVD) graphene could circumvent this slow and labor-intensive process, provided that the sufficiently high quality can be achieved and that the graphene can reliably be transferred from the growth substrate without damage. Indeed, recent work has demonstrated that the quality of CVD graphene can approach that of exfoliated graphene. Key recent achievements include fabrication of CVD-derived devices with low-temperature free path exceeding $25\text{ }\mu\text{m}$ [18], the first observation of fractional quantum Hall effect in CVD graphene [19, 20], and moiré-induced transport in twisted bilayers [21].

We have recently demonstrated that oxygen-free chemical vapor deposition (OF-CVD) can be used to synthesize graphene with intrinsic quality matching that of exfoliated graphene [22]. OF-CVD graphene is free of contamination and shows defect density below $\sim 2 \times 10^9\text{ cm}^{-2}$. hBN-encapsulated devices fabricated by dry transfer from the Cu growth

substrate show intrinsic (phonon-limited) room-temperature mobility and fully developed fractional quantum Hall states at low temperature and applied magnetic field $B = 15$ T.

To take full advantage of OF-CVD graphene for research applications, a reliable and scalable transfer technique is required. Dry transfer requires aggressive oxidation that roughens the Cu surface, limiting device size and yield [18, 19, 23]. Metal-assisted transfer, which uses a strongly adherent metal to peel the graphene off of the Cu surface, is an alternative approach that does not require Cu oxidation. Previous reports show that metal-assisted transfer is effective, scalable, and can produce reasonably clean graphene films [24–26]. However, metal evaporation and etching can potentially damage the graphene, and no work to date has demonstrated that this method can yield graphene free of contamination and defects.

Here we establish a Ni-assisted method for damage-free transfer of OF-CVD graphene. The method is demonstrated for epitaxial graphene grown on sapphire-supported Cu(111) films. By multiple measures, the transferred graphene is equivalent in performance to exfoliated graphene. Atomic force microscopy indicates that it is flat and free of contamination. Raman spectroscopy indicates that it is free of strain and doping, with no detectable defects. h BN-encapsulated devices show low-temperature electronic transport performance equivalent to that of the best exfoliated devices, including ultra-low charge disorder and Landau level broadening. Under applied magnetic fields, the integer and fractional quantum Hall response matches or surpasses that of the best reports for exfoliated devices. Furthermore, the Ni-assisted method can create arrays of graphene pieces with arbitrary shapes and identical crystal orientation. We use these arrays to demonstrate assembly of magic-angle twisted bilayer graphene with a highly uniform moiré pattern.

Figure 1a provides an overview of the nickel-assisted graphene transfer method (see Methods for details). Graphene is first grown by OF-CVD on a sapphire-supported Cu(111) film. Immediately after synthesis, a thin (~ 40 nm) Ni film is evaporated onto the graphene surface (Fig. 1a(1)). Next, the Ni/Gr is peeled from the Cu with the assistance of polymers

and a rigid support (Fig. 1a(2)). The selective exfoliation of graphene by Ni relies on the increased binding energy between Ni/Gr (141 meV) compared to Cu/Gr (62 meV) [27]. Finally, the entire structure is laminated on the target substrate (here we use Si with a 285 nm SiO₂ epilayer). After transfer, the polymer is removed with solvents and a gentle reactive ion etch (RIE) process and Ni is etched in HCl (Fig. 1a(3)).

Key modifications to previously reported procedures for Ni-assisted transfer were required to achieve optimal results. Electron-beam evaporation of Ni was found to damage the graphene, as indicated by appearance of a D peak in the Raman spectrum. This damage was eliminated by adopting a pulsed deposition technique [28] (Fig. S1a-b) and placing a permanent magnet next to the sample to deflect secondary electrons (Fig. S1c-d). Using only polymer plus thermal release tape for transfer resulted in cracking of the Ni/Gr film. This was eliminated by adding a rigid epoxy support (Fig. S2) to minimize deformation during transfer. Finally, a thin layer of residue remains on the Ni after solvent removal of the polymer. This was removed by treatment with O₂/Ar plasma and etching in dilute Piranha solution (Fig. S4).

Figure 1b shows an optical microscope image of a large-area graphene film transferred to SiO₂. The graphene appears free of both tears and residues. A magnified view (inset) shows a clean, fold-free surface. Figure 1e shows an atomic force microscopy (AFM) topography image of a natural graphene/SiO₂ step within this sample (Fig. S5). The graphene exhibits conformal contact with a Gr/SiO₂ step height of 0.34 nm, indicating that the film is flat and free of contaminants such as polymer residue and amorphous carbon.

During the Ni-assisted transfer, any graphene not covered by Ni remains on the Cu (Fig. S3). Thus, the graphene can be patterned by depositing the Ni in the desired pattern prior to transfer. Here we pattern the Ni by evaporation through shadow masks. Figure 1c shows an optical image of an array of 25 μm graphene squares obtained by using a TEM grid as the shadow mask, as illustrated in the inset. More complicated shapes can be obtained using custom-designed masks. As an example, Fig. 1d shows an array of crowns obtained

using a patterned silicon nitride membrane. Figures 1f and g show AFM height maps of the patterned graphene. As for the continuous film, these samples are free of contamination and have roughness comparable to that of exfoliated graphene on SiO₂ (Fig. S6). The step height at the edge of each pattern is 2 ~ 3 nm, because the plasma descum process slightly etches the surrounding SiO₂ (Fig. S5); whereas the natural step seen in Fig. 1e was located at a local crack within the film boundary. Comparing Figs. 1f and g, it can be seen that the square pattern has rougher edges. These differences reflect the deposition masks used, with our customized shadow mask providing a smoother boundary than the as purchased TEM grids.

Raman spectroscopy was used to assess defect density, doping, and strain in the graphene after Ni-assisted transfer to Si/SiO₂. Figure 2a shows representative Raman spectra of graphene transferred to Si/SiO₂ by the Ni-assisted method. As shown in the left inset, no D peak is observed; from the noise level, we can conclude that $I_D/I_G < 0.013$. Likewise, in Raman maps of the patterned graphene, there is no detectable D peak, and I_D/I_G remains below ~ 0.03 (Figs. S7-8). This sets an upper bound on the defect density n_D of the graphene film: for the single spectrum $n_D < 2.9 \times 10^9 \text{ cm}^{-2}$, while for the map and $n_D < 6.8 \times 10^9 \text{ cm}^{-2}$ [29]. The right inset of Fig. 2a shows the Raman map of collected data shown in Fig. 2b-c displaying the 2D peak position (ω_{2D}). We note a low spatial variance in ω_{2D} ($\sim 1 \text{ cm}^{-1}$) over a 15 μm area.

Figure 2b shows Raman analysis of strain and doping over a large area. The solid and dashed lines show axes of varying strain at zero doping, and varying doping at zero strain [30]. The blue data points represent the G and 2D peak positions at each location for the Ni-transferred graphene film. These points are tightly clustered near the intrinsic (zero strain/doping) point. Wet-transferred OF-CVD graphene (yellow point [22]) is similarly strain-free, but with high doping ($n > 10^{13} \text{ cm}^{-2}$), suggesting residual contamination is present from the transfer process. The green and lavender points show the results of two recent studies that achieve the lowest strain/doping to date for CVD graphene transfer [31,

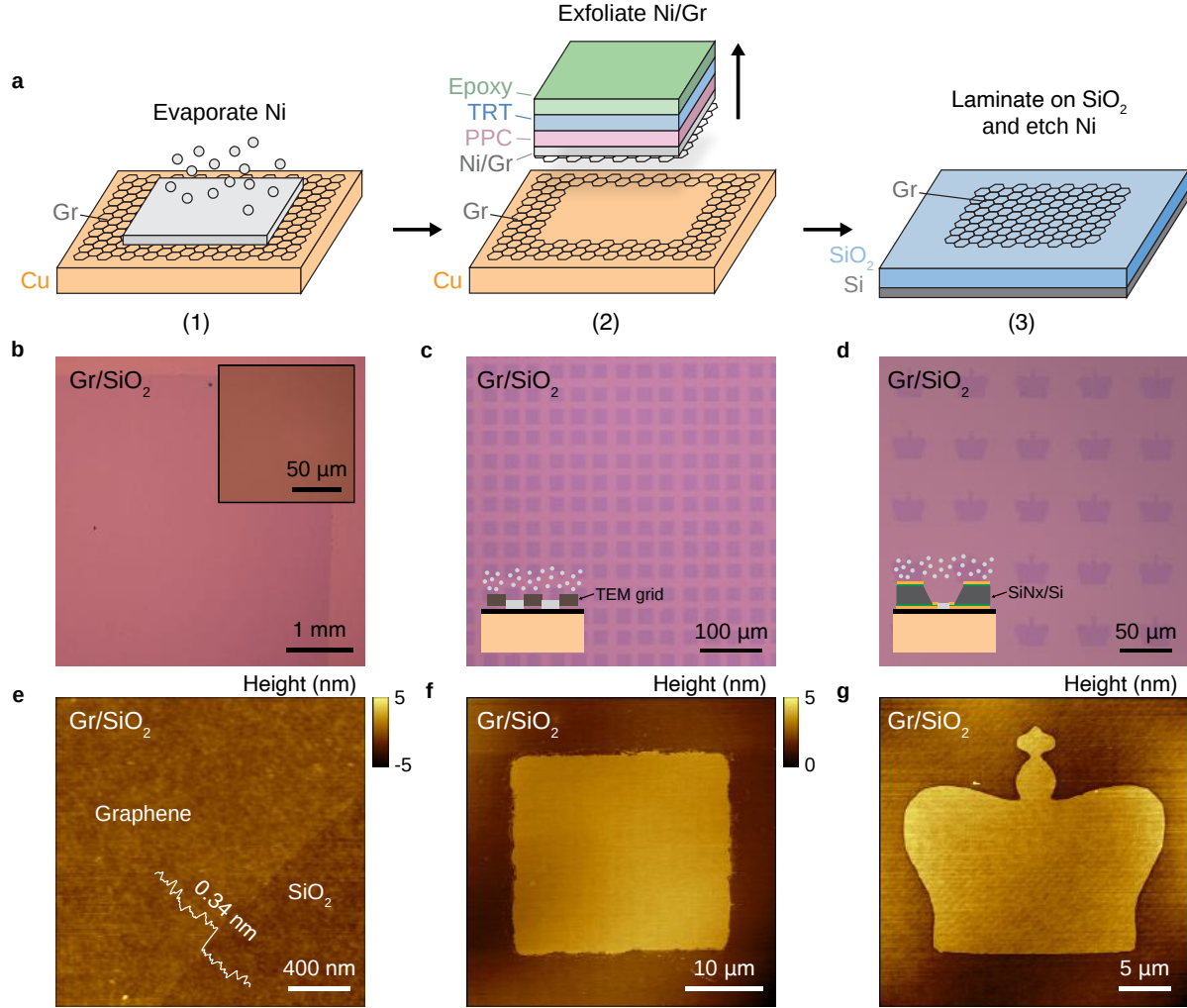


Figure 1: Nickel-assisted transfer of large-area/patterned graphene films onto SiO_2/Si . **a**, Schematic for nickel-assisted CVD graphene transfer: (1) Ni evaporation on as-grown Gr/Cu(111) (2) Spin-coating the metal surface with a layer of polypropylene carbonate (PPC) and thermal release tape (TRT) to exfoliate the PPC/Ni/Gr layers from the Cu. (3) Lamination onto a SiO_2/Si substrate, descum TRT/PPC residue, and etch Ni film with HCl. **b**, Optical image of a large-area graphene film transferred to SiO_2/Si . Inset of **(b)** shows a magnified view of the graphene film highlighting a lack of wrinkles, add-on layers, and voids. **c-d**, Optical images of graphene squares and crowns transferred to SiO_2/Si using a TEM grid and SiNx/Si shadow masks, respectively. **e**, AFM image of a natural Gr- SiO_2 step with a height of 0.34 nm pointing to a residue-free graphene surface. **f-g**, AFM images of representative patterned ultra-flat graphene squares and crowns.

32] along with a comparison to exfoliated graphene (peach points) [30].

Figure 2c shows a histogram of the measured strain, determined by projecting the data Fig. 2b onto the strain vector for exfoliated (top) and Ni-transferred CVD graphene (bot-

tom). We find that the strain disorder is exceptionally low with a mean and standard deviation (0.002 ± 0.007 %). These values are smaller than those reported for exfoliated graphene on SiO_2 (0.025 ± 0.030 %) [30]. We speculate that the Ni and rigid support help to minimize induced strain during transfer. OF-CVD synthesis coupled with Ni-assisted transfer is the only technique to consistently achieve simultaneously zero strain and residual charge doping after processing.

Reproducibility of these findings across a patterned array was assessed by Raman mapping of 24 graphene squares (Fig. S9). The I_{2D}/I_G ratio is highly uniform (Fig. S9b) and the D peak remains uniformly imperceptible (Fig. S9d). The variance in strain and doping across squares is comparable the variance within a given square (Fig. S9e).

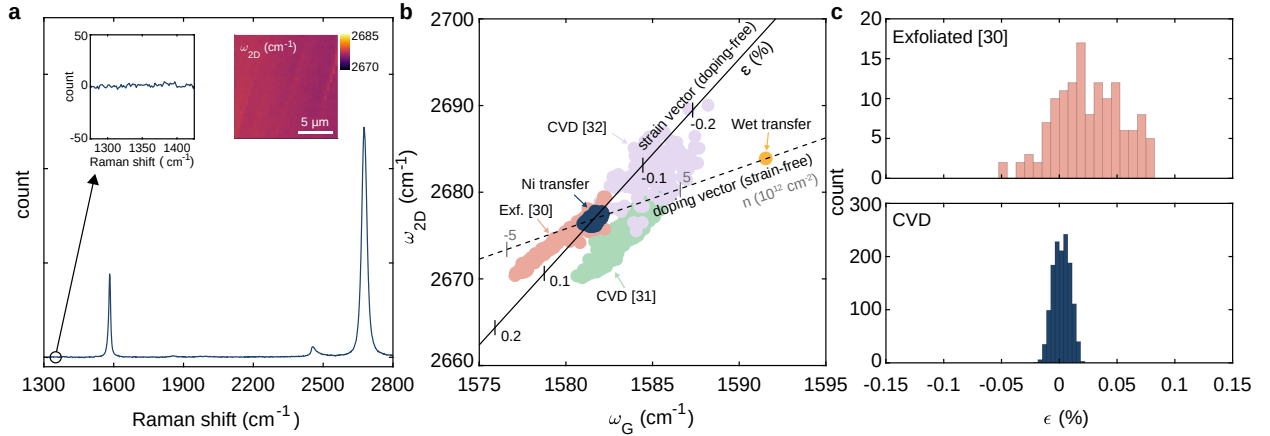


Figure 2: Suppressed doping and strain in CVD graphene films on SiO_2/Si . **a**, Typical Raman spectrum of graphene transferred by Ni-assisted technique collected with 0.3 mW, 60 second exposure, and 1 accumulation. Left inset shows the lack of an identifiable D peak in the nickel-based method. Right inset is a Raman map of the 2D peak position for the Ni-transferred graphene seen in **(b-c)**. Raman mapping for the Ni transfer was collected with 532 nm excitation (0.3 mW, 10 second exposure, 1 accumulation, $N = 1,348$). **b**, Plot of G-band versus 2D-band frequencies for various graphene transfer methods. The intersection of the dashed lines (frequency ω_G at $1,582 \text{ cm}^{-1}$, ω_{2D} at $2,677 \text{ cm}^{-1}$) is the value reported in the literature for suspended graphene free of both strain and doping. Exfoliated graphene [30] along with two recent studies using CVD graphene are provided as comparison [31, 32]. **c**, Histogram showing the spread of strain in the exfoliated (top [30]) and Ni-transferred CVD graphene (bottom). From **(c)**, we extract a low strain disorder of 0.002 ± 0.007 % for the CVD film.

For electrical transport measurements, the graphene was encapsulated within $h\text{BN}$ and

edge-contacted using the conventional van der Waals assembly process [33, 34]. (Fabrication details in Methods). A first device utilizing the Si^{++} wafer as the back gate showed behavior similar to that of exfoliated graphene in the same architecture (Fig. S10), where charge disorder from the $\text{Si}^{++}/\text{SiO}_2$ substrate can limit performance. Figure 3 summarizes magnetotransport measurements a second device employing graphite gates (Fabrication details in Fig. S11). Its room-temperature carrier mobility matches that of exfoliated graphene and dry-transferred OF-CVD graphene (Table 1, Fig. S12). Upon cooling to low T , the device shows ballistic transport, with mean free path limited by device size. Therefore, following previous work [35], quality was assessed in two ways: by measuring the zero-field resistivity at low density, which is sensitive to charge disorder; and by measuring the quantum Hall response, which is sensitive to Landau level broadening (Γ).

Zero-field transport measurements (Fig. 3a) reveal ultra-low charge disorder: the Dirac peak is narrow ($\text{FWHM} = 6 \times 10^9 \text{ cm}^{-2}$), and the residual carrier density found by extrapolating the linear trend in $1/R_{xx}$ is $n^* = 1.6 \times 10^9 \text{ cm}^{-2}$. These values are comparable to or better than the best reported values for exfoliated graphene [14, 36, 37].

Figure 3b plots longitudinal resistance (R_{xx}) *vs.* density (n) and magnetic field (B) in the low-field regime. Integer quantum Hall states (*e.g.* $\nu = 2$) emerge at fields less than 15 mT, consistent with the best values observed for exfoliated graphene [14, 15] (Fig. S13a). Interestingly, the data also show slight splitting and negative magnetoresistance near the central Dirac point. These features are consistent with valley splitting and bandgap opening due to graphene-hBN interaction [11], but further study is needed to confirm this assignment.

Figure 3c shows a high-field map of longitudinal conductivity σ_{xx} as a function of ν and B . Several distinguishing features are evident: symmetry-broken integer quantum Hall states emerge at less than $B = 1 \text{ T}$; prominent fractional states are resolvable below 4 T (Fig. S13b). The appearance of well-resolved integer and fractional states at low fields indicates very high quality.

To illustrate the fractional quantum Hall response in more detail, figure 3d plots σ_{xx} and

inverse Hall resistance ($1/R_{xy}$) *vs.* ν at $B = 15$ T between $\nu = 0$ and $\nu = 1$. Multiple two-flux composite fermion (CF) states centered around $\nu = 1/2$ (red lines) are observed. Fully developed states with $\sigma_{xx} = 0$ and corresponding plateaus in $1/R_{xy}$, are seen up to denominator 11, and incipient states are seen up to denominator 19. Four-flux CF states centered around $\nu = 1/4$ (blue lines) are also observed with denominators up to 17.

Figures S14 and S15 compare these data to previous results for dry-transferred OF-CVD graphene [22] and exfoliated graphene [14], respectively, at the same magnetic field and temperature. Ni-assisted transfer leads to substantially improved FQHE response *vs.* dry transfer: many more FQHE states are fully developed, and the response around $\nu = 1/2$ is more regular. The measured FQHE response is quite similar to that of exfoliated graphene, and in fact shows a larger number of developed FQHE states (both 2-flux and 4-flux). Similar results are seen in comparing to graphite-gated [15] and gate-defined [13] devices fabricated from exfoliated graphene.

For quantitative assessment of quality, we extract the gap sizes for fractional quantum Hall states from thermally activated transport measurements. Figure 3e shows a plot of the activation gap (Δ) for several FQH states. Under the assumption that the effect of disorder is to reduce all gaps by a fixed energy Γ [38, 39], Δ is expected to increase linearly with $B_{eff} = B|1 - 2\nu|$. Therefore, extrapolating a linear fit of Δ *vs.* ν to $\nu = 1/2$ provides an estimate of Γ . Here we obtain $\Gamma \sim 13.3$ K, comparable to the best value obtained for exfoliated graphene [14].

Fabricating moiré heterostructures such as twisted bilayer graphene from exfoliated flakes generally involves cutting or tearing a single flake into two pieces with identical crystal orientation, that can then be picked up at the desired twist angle [2, 40, 41]. Because graphene grows epitaxially on the Cu(111) surface, its crystal orientation remains uniform over large areas [32, 42–44]. This provides an opportunity to create moiré heterostructures by sequential pickup of graphene pieces within an array. Here we use this approach to fabricate twisted bilayer graphene (tBLG) at the ‘magic angle’ required to realize superconductivity [2]. Fabri-

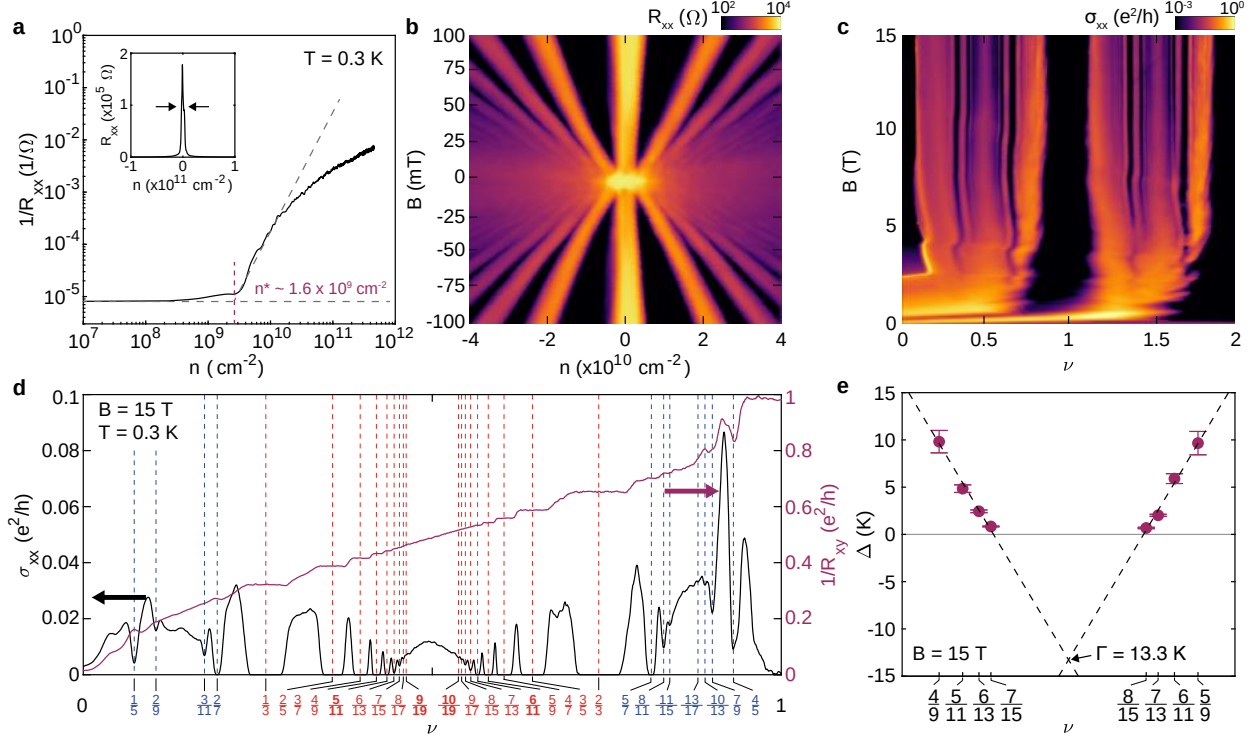


Figure 3: **Transport properties of the nickel-transferred graphene film.** **a**, Low-temperature (0.3 K) inverse four terminal resistance ($1/R_{xx}$) versus density at $B = 0$ T. The intercept of two lines following the regimes of constant slope provides an estimate of the residual charge carrier fluctuations $n^* = 1.6 \times 10^9 \text{ cm}^{-2}$. Inset of **(a)** plots the Dirac curve (R_{xx} vs. n) to extract a narrow FWHM value of ($\Delta n = 6 \times 10^9 \text{ cm}^{-2}$). **b**, Low field resistance (R_{xx}) plotted versus n and B field at $T = 0.3$ K. **c**, σ_{xx} versus ν and B at $T = 0.3$ K for $0 < \nu < 2$. **d**, σ_{xx} and $1/R_{xy}$ versus ν at $B = 15$ T and $T = 0.3$ K for $0 < \nu < 1$. Red lines and blue dashed lines identify fractional states belonging to the two-flux and four-flux CF sequences, respectively. **e**, Activation gaps for the observed fractional quantum Hall states in the $N = 0$ level as a function of filling fraction at $B = 15$ T.

cation of magic-angle twisted bilayer graphene (MATBG) is particularly challenging, because of the narrow angle window ($\pm 0.1^\circ$) and uniformity required to realize the superconducting state [1, 2].

The general process flow to generate a single tBLG structure can be seen schematically in Fig. 4a. Following the standard procedure for tBLG fabrication, a PC stamp (similar to the previous section) is used to pick up hBN, which is then used to pick up a graphene square from an array that had been previously transferred to SiO_2 using the Ni-assisted method. Next, the substrate is rotated by the desired twist angle (*i.e.* 1.1°), and a second

graphene square is picked up. Here we focus on characterization of the twist angle and moiré pattern, and therefore leave the sample ‘open-faced’ on the transfer slide for optical and AFM imaging.

Figure 4b shows an optical image of the tBLG stack (the squares are intentionally offset to make them easy to identify individually). Both optical and AFM imaging indicate a twist angle of approximately 1° . To demonstrate the advantage of utilizing scalable CVD graphene, we fabricated an array of tBLG structures by performing the process shown in Fig. 4a with now using a graphene array with a reduced square size and pitch (Fig. 4c). By stacking two separate graphene arrays with 1° global twist between the layers, we observe a distribution of twist angles throughout the sample (Fig. 4d).

Figure 4e shows a 450×450 nm lateral force microscopy (LFM) image of the tBLG shown in Fig. 4b. A clear and uniform moiré pattern is observed, confirming that the interface between the two graphene layers is clean. A fast Fourier transform (FFT) of the LFM scan is shown in the inset of Fig. 4e. The hexagonal diffraction pattern corresponds to twist angle of $\theta = 1.16^\circ$. Apart from the average twist angle from FFT characterization, we analyzed the real-space moiré pattern to extract the local twist angle by locating the center of each superlattice site (Fig. 4f). Twist angle inhomogeneity across a $1.8 \times 1.8 \mu\text{m}$ region (white box in Fig. 4b) was determined by scanning 16 individual windows. Fig. 4g shows a twist angle map, where the local value of θ was determined by processing the superlattice site position *via* image thresholding. Plotting this data as a histogram yields $\theta = 1.15^\circ \pm 0.02^\circ$ (mean $\pm$ standard dev.) (Fig. 4h). The twist angle variation within this device $\pm 0.02^\circ$ is below the previously discussed margin of $\pm 0.1^\circ$ thus pointing to a reliable method for fabricating tBLG superlattice devices. The average twist angle and variation across the measured array was $0.99 \pm 0.06^\circ$ (Fig. 4i). The low measured variation indicates that it should be feasible to use this technique to construct arrays of closely matched MATBG devices for fundamental studies and applications.

These findings mark a critical threshold at which CVD-grown graphene exhibits no dis-

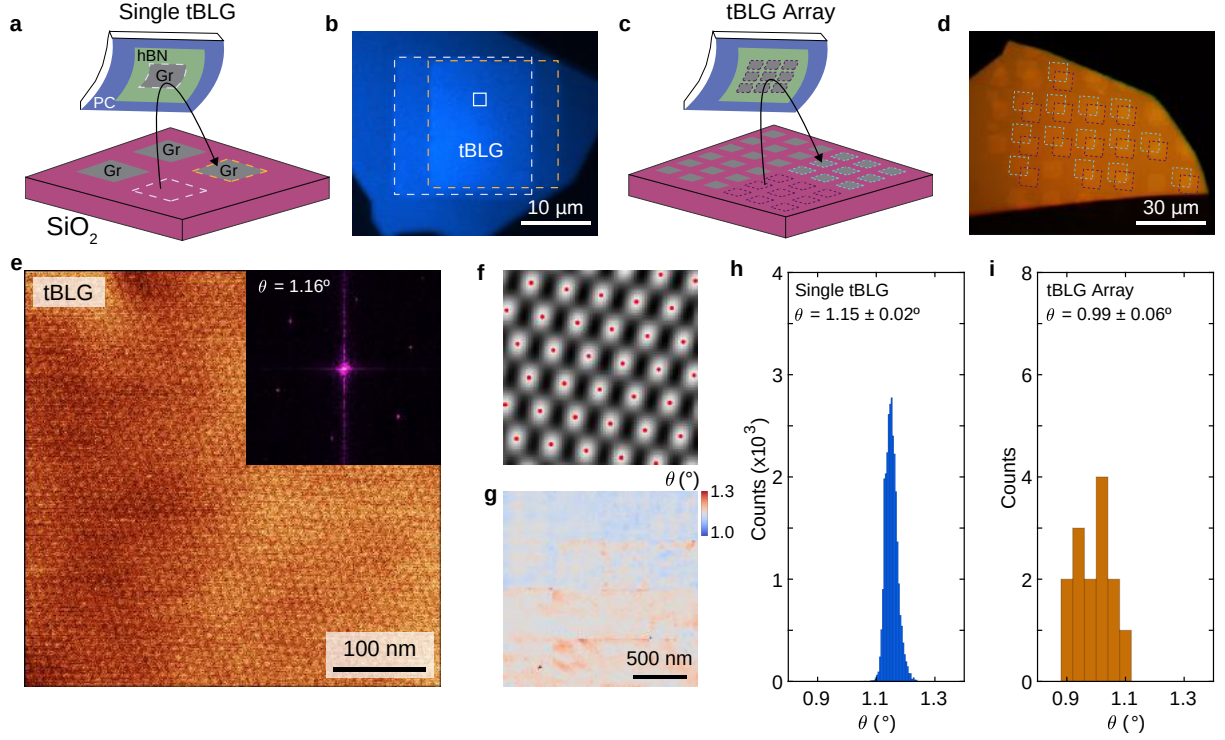


Figure 4: **Fabrication of magic-angle CVD tBLG using nickel-transferred graphene.** **a**, Schematic illustration of the sequential pickup-process to fabricate a single tBLG structure. First, hBN is picked up with PC. Next, a graphene square is picked up from the array using the hBN. Finally, the substrate is then rotated by the desired twist angle (e.g. 1.1°), while keeping the stamp stationary and the second square is picked up. **b**, Optical image of CVD tBLG on hBN/PC where two square graphene samples are twisted by θ during stacking. **c**, Schematic illustration of the sequential pickup-process to fabricate a tBLG array. The process is same as **(a)** except now using a smaller graphene square size and pitch. **d**, Optical image of CVD tBLG array on hBN/PC where 14 tBLG structures are generated with a single pickup. **e**, Large-area LFM scan of the CVD tBLG sample shown in **(b)**. Inset of **(e)** is the corresponding FFT where a global twist angle of 1.16° is extracted from the FFT peak positions. **f**, Denoised LFM scan, where the red dots indicate the center of the superlattice sites used for local twist angle characterization. **g**, Large-area map of the local twist angle calculated from the moiré sites positions of LFM scan area outlined from the white box in **(b)**. **h**, Twist angle statistics from the map derived in **(g)** yielding an intrinsic twist-angle disorder of 0.02° . **i**, Histogram displaying the statistics of global twist-angle, θ , for all measurable samples in the tBLG array seen in **(d)**. The average value for θ was found to be $\theta = 0.99 \pm 0.06^\circ$

cernible disadvantage relative to exfoliated graphene for fundamental research. In fact, the low strain disorder in Ni-transferred graphene can be advantageous for assembly of moiré heterostructures, and can lead to improved quantum Hall response, since strain gradients

induce pseudo-magnetic fields in graphene [45, 46].

Beyond savings of time and effort, the use of single-crystal CVD graphene presents several practical advantages over exfoliated graphene, including deterministic shape control and a well-defined crystallographic orientation [22, 42, 44]. Whereas exfoliated graphene is inherently stochastic and size-limited, this CVD platform enables the production of high-performance graphene over large areas, paving the way for integration into advanced device architectures. Utilizing pre-defined graphene arrays will also simplify the process of robotic heterostructure assembly [47, 48]. To achieve scalable fabrication of research-grade graphene devices, the key remaining challenge is to move ‘beyond Scotch tape’ for the hBN dielectric layers. This challenge can be addressed by continuing to improve the quality of hBN films, or by utilizing large-scale exfoliation [49] from high-quality single crystals. Alternatively, other low-disorder dielectrics [50, 51] may provide sufficient quality for many applications.

Supporting information

Detailed description of the experimental methods, Ni transfer considerations, atomic force microscopy, Raman spectroscopy, heterostructure assembly, transport measurements, and comparisons to literature

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Author contributions

J.A. and J.H. conceived the experiments. J.H. supervised the project. J.A. and X.Y. prepared the Cu(111)/sapphire samples. J.A. and X.Y. performed the OF-CVD graphene synthesis. J.A. and Y.B. developed the Ni evaporation process. T.C. fabricated the SiN_x/Si shadow masks. J.A. was responsible for Raman spectroscopy. J.A. and K.D. were responsible for the tBLG assembly. J.A. was responsible for AFM. A.M. was responsible for LFM. K.W. and T.T. supplied the hBN. J.A. fabricated the graphene transport devices and J.P., B.Y., and J.A. performed electronic transport measurements, which were supervised by C.R.D. J.A. and J.P. were responsible for data analysis. J.A. and J.H. drafted the manuscript. All

co-authors commented on the manuscript prior to its submission.

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