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Concentric and Spiral Few-Layer Graphene: Growth Driven by Interfacial Nucleation vs Screw Dislocation

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ABSTRACT

Spiral growth of various nanomaterials including some two-dimensional (2D) transition metal dichalcogenides had recently been experimentally realized using chemical vapor deposition (CVD). However, such growth that is driven by screw dislocation remained elusive for graphene and are rarely discussed because of the use of metal catalysts. In this work, we show that formation of few-layer graphene (FLG) with a spiral structure driven by screw dislocation can be obtained alongside with FLG having a concentric layered structure formed by interfacial nucleation (nucleation at the graphene/Cu interface) using a Cu-catalyzed ambient pressure CVD. Unlike commonly reported FLG grown by interfacial nucleation where the second layer is grown independently beneath the first, the growth of a spiral structure adopts a top growth mechanism where the top layers are an extension from the initial monolayer which spirals around an axial dislocation in self-perpetuating steps. Since the same atomic orientation is preserved, the subsequent spiraling layers are stacked in an oriented AB-stacked configuration. This contrasts with FLG formed by interfacial nucleation where turbostratic stacking of the entire adlayer may exist. In both growth scenarios, the second layer (either top or bottom) can grow across the grain boundaries of the initial monolayer domains, forming partial regions with turbostratic stacking configuration due to weak interlayer van der Waals interactions. The unique interlayer coupling of FLG spirals which enable superior conductivity along the normal of the 2D crystal with spiraling trajectories, are expected to have new and interesting nanoscale applications.

INTRODUCTION

The extraordinary properties of graphene have led to over a decade of intensive research since its discovery in 2004.¹ Having an extremely diverse spectrum of potential applications, it is vital that this material can be produced artificially in large area for practical usage. The synthesis of large graphene films was first pioneered by the Ruoff's group utilizing chemical vapor deposition (CVD) on Cu foils.² To date, this is still the method of choice to obtain large-area graphene of relatively high quality. Many research groups have since then used CVD for graphene synthesis with slight modifications to the growth parameters to obtain controllable domain sizes, thicknesses, grain orientations and stacking configurations.³⁻⁷ The success of this technique is substantiated by the fact that it is simple, cost-effective, highly reproducible and able to meet the industrial standards for mass manufacturing.⁸ In order to further enhance this growth technique, in-depth knowledge of graphene's growth mechanisms must be established. Although there is a common understanding on the growth of monolayer graphene, the disparities in experimental observations of few-layer graphene (FLG) grown by Cu-catalyzed CVD have baffled researchers over the recent years (i.e., is the second layer grown on top or beneath the initial monolayer?).

Unlike metals with relatively higher C solubility such as Ni, where FLG is formed by segregation and precipitation,⁹⁻¹¹ metals with low C solubility such as Cu restrict growth by segregation resulting in a surface-mediated and self-limiting growth process where the graphene domain grows laterally by edge attachments.^{2, 7, 10} Hence, it offers more control towards achieving single-crystalline graphene with large domain sizes. Recently, it has been unambiguously identified, using a combination of low energy electron diffraction (LEED) and Raman characterization on misaligned and isotopically labeled graphene, that the bi- to few- layers grown by low pressure (LP)CVD adopt a bottom-growth mechanism.¹²⁻¹⁵ In these cases, the growth of

the monolayer crystals were designed to be relatively slow and as such, carbon atoms were allowed to diffuse along the graphene/Cu interface and nucleate on the Cu to form the second layer.¹³ Subsequent layers grow beneath the existing monolayer at the same nucleation center following Volmer-Weber (VW)-like growth mode resulting in an inverted pyramid structure. Further growth is ceased when the monolayer fully covered the Cu surface due to the lack of reactive species resulting from the termination of catalytic activity. However, the continual growth of new layers on top of the monolayer can be overcome by increasing the partial pressure of the carbon source or using atmospheric pressure (AP) CVD,¹⁶⁻¹⁸ or by supplying fresh catalysts.¹⁹ These techniques aid the generation of additional reactive species at the surface and multilayer graphene domains are able to nucleate on top of the existing layer following Stranski-Krastanov (SK)-like growth mode.

Although both VW and SK growth modes are well-documented to explain the formation of FLG, another important growth mode, that is, the screw dislocation-driven growth,²⁰ which exists naturally in crystals, including graphite,²¹ is still surprisingly lacking. Moreover, screw dislocation-driven growth has also been extensively reported on a wide range of nanostructures such as nanoplates,^{22, 23} nanotubes,^{24, 25} nanowires^{24, 26} and, most recently, on some two-dimensional layered materials transition metal dichalcogenides.²⁷⁻³² Here, we show that the growth of FLG can be formed by nucleation at the graphene/Cu interface following a bottom layer-by-layer (LBL) growth mechanism and on a screw dislocation which spirals above the initial monolayer in self-perpetuating steps. We observed that these additional layers (either on top or beneath the first layer) can be formed within a single-crystal domain or grown across grain boundaries of the initial merged monolayer domains. In the case of FLG that formed at the graphene/Cu interface, the stacking sequence of the adlayers can be either oriented (AB) or

turbostratic. For screw dislocation-driven growth, however, the top spiraling layers (excluding the initial first layer) are stacked in a purely oriented configuration following the orientation of the uplifted monolayer edge.

EXPERIMENTAL SECTION

APCVD Growth. Electropolished Cu foils (Alfa Aesar, 25 μm thick) were used as growth substrates. A piece of Cu foil was loaded into a 2 inch quartz tube under a constant H_2 flow of 500 sccm. The furnace was ramped up to 1035 $^{\circ}\text{C}$ in 1 h and kept constant for another 30 min to anneal the Cu and to remove the surface oxide. After annealing, 4.6 sccm of methane gas was flowed into the system to initiate graphene growth. The furnace was kept at 1035 $^{\circ}\text{C}$ for another 4 h for film growth and the lid of the furnace was lifted for quick cooling after growth.

Transfer. An electrochemical delamination process was used to transfer the FLG onto SiO_2/Si substrates. First, the as-grown graphene on Cu was spin coated with poly(methyl methacrylate) (PMMA) at 3000 rpm for 30 s. 1 M of NaOH was used as the electrolyte. A Pt foil was used as the anode and the spin-coated sample was used as the cathode. A constant voltage of 5 – 10 V was applied until the PMMA coated sample is completely detached from the Cu. The sample was then rinsed in DI water for several times and transferred onto a desired substrate. The as-transferred sample was baked at 80 $^{\circ}\text{C}$ until it is completely dried. Finally, the PMMA is removed by submerging the sample in acetone for several hours.

Characterization. Micro-Raman spectroscopy (Witec) with a laser excitation wavelength of 532 nm was used to map the various FLG domains at room temperature. TEM analysis was conducted on an aberration-corrected transmission electron microscope (FEI Titan3 G2 60-300) operated at 80 kV.

RESULTS AND DISCUSSION

Partially covered graphene films with a mixture of mono- and few-layer domains were grown by APCVD on electropolished Cu foils using methane (CH_4) as the carbon source (see methods). For growth by APCVD, there is an extremely narrow window to obtain sporadic monolayer crystals with the presence of few-layer domains. This is obtained by precisely controlling the partial pressure of CH_4 in an H_2 environment during growth. For our samples, we used a gas ratio of 500:4.6 sccm of H_2/CH_4 and a growth duration of 4 h. The graphene samples were then transferred onto 285 nm SiO_2/Si substrates for our study by micro-Raman characterization (Figure 1a). The magnified optical image in Figure 1b shows that the graphene grains comprise of mainly two different orientations with 30° rotational difference as outlined in cyan and red. We found two vastly different growth modes for FLG that co-exist in our samples: (i) nucleation at the graphene/Cu interface (referred to as interfacial nucleation) resulting in concentric stacking of the adlayers and (ii) screw dislocation-driven growth which spirals vertically upwards on top of the first layer as depicted in Figure 1c. We surveyed over 100 individual FLGs and found that the ratio of the bottom-growth concentric FLGs and the top-growth FLG spirals is close to 1:1.

Atomic resolution transmission electron microscopy (TEM) combined with dark-field (DF) imaging was further employed to study the orientation and stacking order of our graphene grains. Figure 1d shows a typical selected area electron diffraction (SAED) taken over the graphene film with partial bilayer regions. In line with our observation in Figure 1b, two sets of hexagonal diffraction patterns with 30° rotational difference were commonly observed throughout our graphene samples, indicating that there are two major orientations that exist in the polycrystalline film. This is attributed to the alignment of graphene grains with respect to the Cu facet/lattice. For graphene films that were grown on Cu foils, it is known that a lower-index facet such as Cu(100)

can accommodate two rotational orientations (30° rotational difference) of graphene.⁵ Figure 1e-g shows the false-colored DF-TEM images and their composite of a bilayer with two differently oriented graphene grains. By imaging using the diffraction spots, $\Phi 1$ and $\Phi 2$, the orientation and the number of graphene layers were revealed. From the composite image in Figure 1g, one can locate the AB-stacked 2-layer region (left side) following the orientation of $\Phi 1$ by the brighter contrast due to constructive interference of the diffracted electrons, and the turbostratic-stacked region (in the middle) by pairing up 1-layer from each orientation, $\Phi 1$ and $\Phi 2$. Figure 1h shows the atomic resolution TEM image at the region over the AB-stacked and turbostratic-stacked bilayer. By performing fast Fourier transform (FFT) analysis, the left region in Figure 1h is identified to be AB-stacked (Figure 1h(ii)) while the region on the right shows an elaborate Moiré pattern (Figure 1h(iii)) with an interlayer twist of 30° . Notably, monolayer regions (Figure 1h(i), regions over the blue dotted lines) are observed along the boundary between the AB-stacked and turbostratic-stacked regions. This is caused by the etching of graphene atoms when exposed to electron beam irradiation during TEM imaging which were initiated at the grain boundary due to weaker bonding structures with lower knock-on energy threshold.

Figure 1. Structural characterization of FLG domains grown using APCVD. (a) Optical image of a partially covered graphene film with a mixture of mono- and few-layer domains. The inset shows a photograph of the as-transferred graphene film on Si/SiO₂ substrate. (b-c) Magnified optical images of the white boxed region in (a). The cyan and red outlines in (b) are the hexagonal edges of the monolayer domains with 30° rotational difference. The white dotted outlines in (c) depicts the edges of the FLG domains. (d) SAED of the polycrystalline graphene film showing two major orientations with 30° rotational difference. False-colored DF-TEM images taken using (e) $\Phi 1$ and (f) $\Phi 2$, and their (g) composite of a bilayer with two differently oriented graphene grains. (h) Atomic resolution TEM image of the white boxed region in (g). Magnified TEM images from (h) showing the atomic configurations of (i) monolayer, (ii) AB-stacked and (iii) turbostratic-stacked regions. Insets in (h) show the FFTs taken at the AB-stacked (left) and turbostratic-stacked (right) regions.

Raman spectroscopy is a versatile and non-destructive characterization technique that can accurately identify the layers and stacking order of FLG owing to the provision of an immense database from many previous studies.³³⁻³⁵ In this work, we mapped out the intensity ratio of the 2D to G peaks ($I(2D)/I(G)$), G peak position ($Pos(G)$) and full width at half maximum of the 2D peak ($FWHM(2D)$) of the FLG domains as they are known to provide intimate details to the layering and stacking structures.³³ Based on available Raman information of graphene in the literature, we first establish the various ways to identify the regions with mono- to few-layer and their stacking configurations as shown in Figure 2. (i) The $I(2D)/I(G)$ is typically $\sim 2 - 3$ for 1-layer, while it decreases to ~ 1 for 2-layer and continue to decrease linearly for 3 layers and above when they are AB-stacked (oriented stacking).³³ For turbostratic stacking at high interlayer twist angle ($> 20^\circ$), the $I(2D)/I(G)$ is much higher of $\sim 3.5 - 4$.³⁶ For our graphene samples, most turbostratic-stacked regions has an interlayer twist angle of 30° . In the case of a combination of turbostratic and subsequent oriented stacking configuration, the $I(2D)/I(G)$ of a 3-layer is ~ 2 and decreases linearly with increasing AB-stacked layers (Figure 2a). (ii) $Pos(G)$ down-shifts with increasing number of layers regardless of the stacking configurations (Figure 2b).³³ Note that $Pos(G)$ varies linearly to the inverse of the number of layers.³⁷ (iii) The $FWHM(2D)$ for 1-layer is $\sim 31 \text{ cm}^{-1}$ and broadening of the 2D band occurs with increasing AB-stacked layers ($\sim 52 \text{ cm}^{-1}$ for 2-layer and $\sim 59 \text{ cm}^{-1}$ for 3-layer) caused by the splitting of electronic bands with increasing layers.^{33, 38} In the case for a turbostratic stacked 2-layer with high interlayer twist of $>20^\circ$, the $FWHM$ is slightly narrower than a monolayer measuring at $\sim 24 \text{ cm}^{-1}$,³⁶ and subsequent broadening occurs with additional AB-stack layers (Figure 2c).

Figure 2. Identification of the number of graphene layers and their stacking configuration by Raman spectroscopy. Plots of (a) $I(2D)/I(G)$ and (b) $Pos(G)$ as a function of number of graphene layers. (c) Raman spectra of the 2D band of various graphene layers with different stacking configuration.

Figure 3 shows the optical images, $I(2D)/I(G)$ Raman maps and their corresponding Raman spectra of various concentrically-stacked FLG pyramids. Namely, we found purely oriented-stacked FLGs (Figure 3a-c), turbostratic-stacked FLGs (Figure 3d-f) and FLGs with a mixture of oriented- and turbostratic-stacked regions (Figure 3g-i). From all the Raman maps in Figure 3, one can observe that the adlayers, which are in the form of compact hexagons, were grown independently from one another (unconnected white dotted lines) but were likely initiated at the same nucleation center. This is the same growth scenario as many reported bilayer graphene grown using LPCVD.¹³⁻¹⁵ For turbostratic-stacked FLG (Figure 3d-f), the second layer was grown in an

entirely different orientation with respect to the first layer. This shows that the orientation of such adlayer is not dependent on the existing graphene layer but adopt its orientation from its nucleation seed by aligning to the underlying Cu lattice (i.e., arising from the two rotational difference on Cu(100)). For FLG with regions comprising both oriented- and turbostratic-staking configurations (Figure 3g-i), the turbostratic-stacked regions of the FLG (magenta/green spots) are caused by the interlayer rotational difference relative to the initial monolayer. As observed by the white dotted lines which outline the perimeters of each adlayer, the hexagons are all aligned indicating that the 2-, 3- and 4-layer follow an oriented stacking order. Hence, this shows that the adlayers were able to grow across the grain boundaries of the initial monolayer without disrupting its crystal orientation due to weak interlayer van der Waals interactions.

FLG spirals with hexagonal edges were also readily observed in our CVD-grown samples. Figure 4 shows the optical images, I(2D)/I(G) Raman maps and their corresponding Raman spectra of various FLG spirals. In Figure 4a, the FLG spiral was grown on a single-crystal monolayer graphene (outlined in black dotted lines) indicating that a screw defect can be form within a single-crystalline structure and does not necessarily initiate at grain boundaries. From the Raman map in Figure 4b, we can unambiguously identify that the FLG spirals up one layer per step following the white dotted lines where the red spot marks a 2-layer region and the blue spot marks a 3-layer region. In screw dislocation-driven growth, this corresponds to one elemental burgers vector for graphene. Unlike the growth by interfacial nucleation where the adlayers can have different orientations, the additional layers grown from a screw dislocation always remain aligned following the orientation of the initial uplifted edge resulting in oriented AB-stacked layers. However, as shown in Figure 4d-f, the FLG spirals were also able to grow across the grain boundaries of the initial merged monolayer domains forming partial regions with turbostratic-stacked configuration.

Figure 3. Raman characterization of concentrically-stacked FLG pyramids. Optical images (left panel), $I(2D)/I(G)$ Raman maps (middle panel) and their Raman spectra corresponding to the colored dots (right panel) of a (a-c) 3-layer graphene with purely AB-stacked configuration, (d-f) 3-layer graphene with turbostratic-stacked configuration and (g-i) 4-layer graphene with regions comprising both AB-stacked and turbostratic-stacked configurations.

Figure 4. Raman characterization of FLG spirals. Optical images (left panel), I(2D)/I(G) Raman maps (middle panel) and their Raman spectra corresponding to the colored dots (right panel) of a (a-c) single-crystalline FLG spiral with purely AB-stacked configuration, and (d-f) FLG spiral with regions comprising both AB-stacked and turbostratic-stacked configurations.

To identify the growth mechanisms (i.e., top- or bottom-growth) of the different FLG structures, we mapped out a region where the two structures were grown in close proximity and intersecting each other (Figure 5). From the optical image in Figure 5a, it can be observed that the edges of the spiral structure (left) and concentric structure (right) are still visibly defined. Raman spectroscopy was used to confirm this overlapping structure by identifying the number of layers at the intersected region (Figure 5b). From the I(2D)/I(G) Raman map in Figure 5c, it is evident that the overlapping regions increased by one layer and the hexagonal edges from each individual structure are not affected by the existence of the other. Figure 5d shows the schematic translation of the Raman map for easy visualization. Note that a different scenario is observed when two of the same structure intersects each other (i.e., when a spiral meets another spiral or a concentric pyramid

meets another concentric pyramid). Instead of overlapping, their layers will merge along the same basal planes (Supporting Information Figure S1 and S2). Hence, the overlapping structure between the spiral and concentric pyramid indicates that the additional layers grown from a screw dislocation and by interfacial nucleation were formed along different basal planes.

Figure 5. Overlapping boundary of a spiral and a concentric FLG pyramid. (a) Optical image of the overlapped FLGs. (b) Raman spectra corresponding to the colored dots marked in (a). (c) $I(2D)/I(G)$ Raman map showing the overlapped region. (d) Schematic translation of the Raman map in (c).

From previous reports on FLG growths with concentric pyramid structure, they adopt a bottom-growth mechanism where new layers are form at the Cu interface.¹³⁻¹⁵ This is true for our case as well through the fact that a concentric pyramid structure may comprise of entirely turbostratic stacking and hence, the new layers which nucleate on the Cu surface may result in a possible orientational change aligning to the Cu lattice. We further confirmed this bottom-growth mechanism by performing Raman mapping on partially torn regions where part of the top layer is delaminated together with the Poly(methyl methacrylate) coating during lift-off while the adlayers at the bottom remains intact (Supporting Information S3 and S4). Since, the FLG with concentric structure does not grow along the same basal plane as the spiral structure, it is reasonable to conclude that the spiraling layers grown from a screw dislocation adopt a top-growth mechanism. Hence, the spiraling layers on top were able to grow over the buried concentric layers (and vice versa) because of the smooth steps created by the initial monolayer which separates the two structures.

While the growth of FLG by interfacial nucleation is predominant when using LPCVD, the growth of spiraling structures from screw dislocations can occur simultaneously when APCVD is employed. This indicates that although the thermodynamics of graphene synthesis is the same for both systems, the variation in kinetics caused by the difference in pressure resulted in significantly different growth mechanisms of the multilayers.¹⁸ Figure 6 shows the growth scenario of FLG domains in a Cu-catalyzed APCVD process. First, the carbon precursor (CH_4) diffuses through the boundary layer and dehydrogenation occurs at the Cu surface producing active carbon species.¹⁸ The active carbon species is then absorbed onto the Cu surface and C atoms which are completely dehydrogenated diffuses into the Cu.¹³ Nucleation of the first layer of graphene begins on the Cu surface and the monolayer graphene domains grow laterally by edge attachments. Neighboring

domains coalesce together where their edges meet, forming a partially continuous polycrystalline film.^{7, 10} During the growth, desorption of by-products such as H₂ are transported away from the Cu surface. The growth of the second layer occurs concurrently even when the first layer is incomplete. In the case of a bottom-growth concentric pyramid structure, C atoms diffuse along the graphene/Cu interface from beneath the Cu bulk and nucleation occurs on irregular sites on the Cu surface such as impurities and defects.¹³ Similar to the growth of the first layer, the second layer domain grows laterally by edge attachments and subsequent additional layers are likely to grow at the same nucleation center at the graphene/Cu interface. Since the growth of each layer is independent from one another, the stacking configuration can be purely turbostratic as their orientation(s) are confined to the underlying Cu lattice.

Figure 6. Schematic illustration depicting the different growth modes of FLGs in a Cu-catalyzed APCVD.

For a spiral growth driven by a screw dislocation, it adopts a top-growth mechanism where the graphene layers spiral around an axial dislocation in self-perpetuating steps. However, to the best of our knowledge, there is no reported growth of such structure using Cu-catalyzed LPCVD. From

previous studies of graphene growth using low carbon solubility catalyst, it is known that the graphene grown at AP is much more non-uniform in thickness and contain a higher concentration of defects.^{16, 18} This is because the growth is governed by the mass-transport limited regime where the geometric effects of the gas flow, substrate and CVD chamber plays an important role during the synthesis. Due to the variation in the amount of active carbon species diffusing through the boundary layer, the amount of carbon absorbed onto the Cu surface varies and thus resulting in thickness non-uniformity. In addition, as the gas flow rate is much higher as compared to LPCVD, there are more collisions between the gas molecules/radicals leading to gas phase reactions in the bulk gas flow.¹⁸ This may give rise to more active carbon species to be absorbed onto the existing monolayer graphene and thus enabling a top-growth mechanism. Since the growth rate of graphene is relatively faster in APCVD, formations of defects such as dislocations are more likely to occur. As shown in Figure 4a, a screw defect can exist within a single-crystal monolayer graphene. Furthermore, dislocations are also known to form along the grain boundaries of merged monolayer domains.³⁹ During the growth of FLG spirals, the C atoms diffuse along the existing graphene layer and attach to the active edge at the dislocation.²⁰ The FLG then propagates by spiraling up one layer per step, growing in both the vertical and horizontal directions by edge attachment and hence retaining its orientation and resulting in a purely AB-stacked configuration with the exclusion of the first layer from other neighboring domain(s) with different orientation.

In classical crystal growth theory, there are mainly three types of growth mechanisms: screw dislocation, LBL and dendritic growth.⁴⁰ Each of these growth modes are dependent on the degree of supersaturation, σ , which, in this case, is related to the concentration of the precursor in a CVD system, as defined by, $\sigma = \ln\left(\frac{c}{c_o}\right)$, where c and c_o are the concentration and equilibrium concentration in the system. Burton-Cabrera-Frank (BCF) theory states that as the supersaturation

increases, the crystal growth will be progressively be dominated by dislocation, LBL and dendritic growth, respectively.^{41, 42} In our CVD process, we have exploited low supersaturation condition by feeding in very little amount of precursor (CH_4) which enabled multilayer growth driven by screw dislocation as evidenced by the spiral structures. Although the concentric pyramid structure follows an LBL-like growth mode, this occurrence is triggered by the presence of the Cu catalyst which aided the dehydrogenation of the active species enabling C atoms to enter the Cu bulk and nucleate at the graphene/Cu interface from beneath. Further growth of adlayers will hence be ceased when the graphene layer fully covers the Cu surface terminating further catalytic activity. It should be noted that further increasing the partial pressure of CH_4 will increase the supersaturation which should lead to LBL growth where new layers are nucleated on top of the existing graphene layer as observed before in other CVD growths.¹⁶

CONCLUSIONS

In conclusion, we elucidate co-existing two different growth modes of FLG using Cu-catalyzed APCVD at low supersaturation condition. The first, which has been widely reported, is by interfacial nucleation. In this growth mode, it adopts a bottom-LBL-growth mechanism where additional layers are nucleated at the graphene/Cu interface. The hierarchical growth of the adlayers are independent from one another resulting in an inverted concentric pyramid structure. This also creates the possibility of forming an entirely turbostratic-stacked adlayer because of the underlying Cu lattice. The second is by screw dislocation-driven growth where the additional layers are an extension from the first layer. In this growth mode, it adopts a top-growth mechanism where the graphene layer spirals around an axial dislocation in self-perpetuating steps. Since the orientation of the spiraling layers remained unchanged from the first layer, they preserve an oriented AB-stacking configuration. However, in both cases, the second layer (either top or

bottom) is able to grow across the grain boundaries of the initial monolayer due to weak interlayer van der Waals forces, forming partial regions with turbostratic stacking. The FLGs with different structures are expected to have significantly differing properties. Of the most apparent are their electronic and thermal conductivities along the normal direction of the 2D crystal. In this essence, a spiral structure will have superior conductivity because all the layers are covalently bonded together. It has been predicted that such graphene spirals which resemble Riemann surfaces can act as nanosolenoids that are able to generate very high magnetic field,⁴³ or as nanosprings with extraordinary high tensile deformation capabilities.⁴⁴ Furthermore, because of its unique interlayer coupling, optoelectronic and spintronic phase transitions are expected.^{45, 46} Hence, in addition towards understanding the various growth mechanisms and its structures of FLG, this work further opens up experimental feasibility for new and interesting nanoscale applications.

ASSOCIATED CONTENT

Supporting Information. Figures S1–S4 showing additional Raman maps of various FLGs. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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Notes

The authors declare no competing financial interest.

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TOC graphic

Supporting Information

Concentric and Spiral Few-Layer Graphene: Growth Driven by Interfacial Nucleation vs Screw Dislocation

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Figure S1. (a) Optical image of a bilayer graphene with merged adlayers. The inset shows the $I(2D)/I(G)$ Raman map. (b) Raman spectra corresponding to the colored dots marked in (a).

Figure S2. (a) Optical image, (b) $I(2D)/I(G)$ Raman map and (c) Raman spectra corresponding to the colored dots of a FLG with merged spiral structures.

Figure S3. (a) Optical image of a concentric FLG pyramid with a partially torn region. (b) Raman spectra corresponding to the colored dots marked in (a). (c) I(G) and (d) I(2D) Raman maps of the corresponding FLG.

Figure S4. (a) Optical image of a concentric FLG pyramid with a partially torn region. (b) Raman spectra corresponding to the colored dots marked in (a). (c) $I(G)$ and (d) $I(2D)$ Raman maps of the corresponding FLG.