

Domain-dependent surface adhesion in twisted few-layer graphene: Platform for moiré-assisted chemistry

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Abstract

Twisted van der Waals multilayers are widely regarded as a rich platform to access novel electronic phases, thanks to the multiple degrees of freedom available for controlling their electronic and chemical properties. Here, we propose that the stacking domains that form naturally due to the relative twist between successive layers act as an additional "knob" for controlling the behavior of these systems, and report the emergence and engineering of stacking domain-dependent surface chemistry in twisted few-layer graphene. Using mid-infrared near-field optical microscopy and atomic force microscopy, we observe a selective adhesion of metallic nanoparticles and liquid water at the domains with rhombohedral stacking configurations of minimally twisted double bi- and tri-layer graphene. Furthermore, we demonstrate that the manipulation of nanoparticles located at certain stacking domains can locally reconfigure the moiré superlattice in their vicinity at the μm -scale. Our findings establish a new approach to controlling moiré-assisted chemistry and nanoengineering.

keywords: twisted graphene moiré, rhombohedral and Bernal stacking domains, nanoengineering, surface chemistry

Twisted van der Waals (vdW) heterostructures have drawn growing interest in recent years.^{1–7} The twist angle creates a moiré pattern at a larger length scale than the periodicity in each independent layer, leading to spatially-dependent interactions between the layers. As such, the twist angle has been used as a continuous knob to effectively tune the Hamiltonian of the system and realize novel electronic phases.^{8,9} At small twist angles, large domains of lowest energy stacking configurations separated by sharp domain walls form through the atomic relaxation process,^{10–14} resulting in multiple different stacking orders coexisting within the multilayer.

Recently, several works have explored how the twist angle and the consequent changes to the global electronic structure of the material, such as the emergence of flat bands or modified density of states, affect the surface chemistry and catalytic properties of moiré systems.^{15–18} Additionally, preferential adsorption of chemical species due to corrugation and strain effects on the moiré surface has also been observed.¹⁹ In general, strain has been utilized as a means of manipulating the moiré patterns in twisted vdW heterostructures.^{20–22} However, until now, no investigation has focused on the local stacking order as a platform for controlling the surface chemistry of this class of materials.

Here, we demonstrate domain-dependent surface adhesion as a new avenue to observe and manipulate the surface chemistry of moiré systems, a phenomenon we call moiré-assisted chemistry. We first show selective adhesion of metallic film on the rhombohedral domain of twisted double bilayer graphene (tDBG) while avoiding the Bernal domain. We then explore the robustness of this observation in twisted double trilayer graphene (tDTG) under varying ambient conditions and through domain-dependent adhesion of another material, namely, water droplets. Finally, we demonstrate how the particles that selectively adhere to the surface can be used to reshape the underlying moiré lattice. Overall, our results establish a basis for the development of capabilities in moiré engineering via domain-dependent surface interactions.

In tDBG, the relaxed moiré superlattice is composed of two triangular stacking order domains: the lowest energy Bernal configuration (ABAB) and the metastable rhombohedral configuration (ABCA).^{11,23} Due to the small but finite energy difference between the Bernal phase and the rhombohedral phase, the rhombohedral domain walls exhibit a finite inward curvature.¹¹ This characteristic curving allows us to unambiguously identify the rhombohedral domains with scanning probe microscopy techniques (see Supporting Note A). Fig. 1a shows a mid-IR scanning nearfield optical microscopy (mIR-SNOM) map measured on a tDBG sample with a 0.3-degree twist angle. The rhombohedral and Bernal stacking domains are identified by the light and dark contrast respectively (see Fig. 1a insets); simultaneous topography imaging (lower left inset of Fig. 1a) shows that the sample is featureless on the surface. We deposit Field’s metal onto this surface using a mechanical ablation technique (see Supporting Note A). This process breaks the molten metal and scatters its nanoparticles on the surface. One would naively expect the formation of random patches of metallic film on the surface, with no correlation to the moiré superlattice. Instead, the tapping mode atomic force microscopy (AFM) map of Fig. 1b reveals the formation of metallic films with complete correlation to the rhombohedral domains of the moiré superlattice, as imaged in mid-IR nearfield. The triangular metallic film domains exhibit sharp corners and fine features, as narrow as the thin double domain walls separating two Bernal phases,¹¹ as shown in Fig. 1b (diagonal lines along the top right part of the panel).

After 45 days of storage within vacuum, the same AFM measurement over the surface of the sample shows degradation of fine nanoparticle features. Particularly along double domain walls, previously visible nanoparticle features no longer appear in subsequent topography scans (Fig. 1c). After 173 additional days and again 302 days of storage in a dry N₂ environment, no further degradation is observed (Fig. 1d). Robust coverage of the larger triangular regions persists regardless of the storage conditions of the sample. Theoreti-

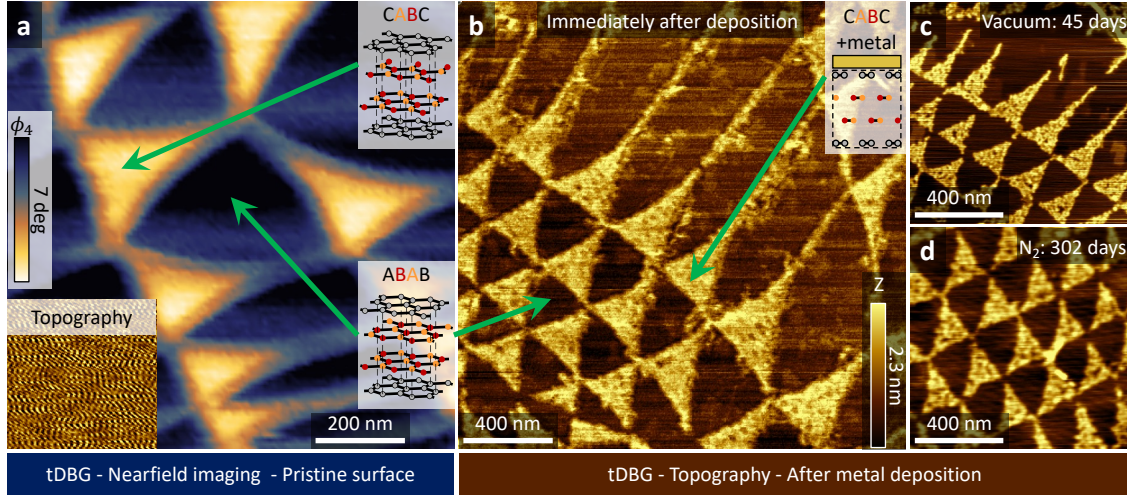


Figure 1: Moiré-assisted chemistry in twisted double bilayer graphene (a) Mid-IR nearfield imaging of the twisted double bilayer moiré superlattice (tDBG) moiré superlattice (phase, 4th harmonic). Image taken on pristine sample before deposition. No topographic features were observed (see simultaneously taken tapping mode scan in inset). The Bernal (ABAB) and rhombohedral (ABCA) domain configurations are illustrated over the image. (b) Metal nano-structures forming over the rhombohedral phase of tDBG, revealed by tapping mode atomic force microscopy (AFM). The patterns formed after mechanical ablation deposition of Field’s metal over a minimally twisted open phase tDBG (see Supporting Note A). (c) The same measurement performed after 45 days during which the sample was stored in vacuum showing degradation of fine features. (d) Storing the sample in dry N₂ at room temperature stopped further degradation, even after more than 300 days. All tapping mode scans share the same color-bar.

cal investigations using first principles density-functional theory (DFT) calculations indicate that domain-dependent adhesion is not intrinsic to the tDBG system. The calculated energy difference between the Bernal and rhombohedral double bilayer graphene changes only slightly with the addition of the metal, on the order of a few meV/unit cell, depending on the details of the metal nanoparticle model (see Supporting Note B). This energy difference is too small to account for the robust experimental observations. We therefore suspect that there is likely a mediating agent that plays a critical role in the emergence of selectivity. We hypothesize that the mediating agent is most likely a carbon-based organic substance, used either in the mechanical exfoliation of our materials or in the fabrication of the sample (see Supporting Notes A and D).

To probe whether the previously seen selectivity is unique to the tDBG system, we investigate a different multilayered graphene system, tDTG. Like tDBG, the tDTG system has

two nearly degenerate low-energy stacking configurations. We observe domain-dependent adhesion in tDTG as well, further supporting that the phenomenon is universal, with an apparent selectivity determined by stacking domain. As shown recently,²⁴ in tDTG, energy considerations lead to a global layer translation, resulting in the moiré superlattice being ABABAB/BCBACA (see Fig. S3a). The ABABAB is a pure Bernal stacking configuration and the BCBACA phase can be thought of as a 4-layered rhombohedral graphene encapsulated by two Bernal interfaces.

Similar to tDBG, we investigate the tDTG sample using mid-IR nearfield imaging and verify the existence of moiré patterns with domains of BCBACA and Bernal stacking order (see Fig. S3b). A simultaneously taken tapping mode AFM scan (see Fig. S3c) reveals no topographic features corresponding to the moiré pattern. We then place the tDTG sample into a humid environment (see Supporting Note D). Upon removal, we repeat simultaneous mid-IR

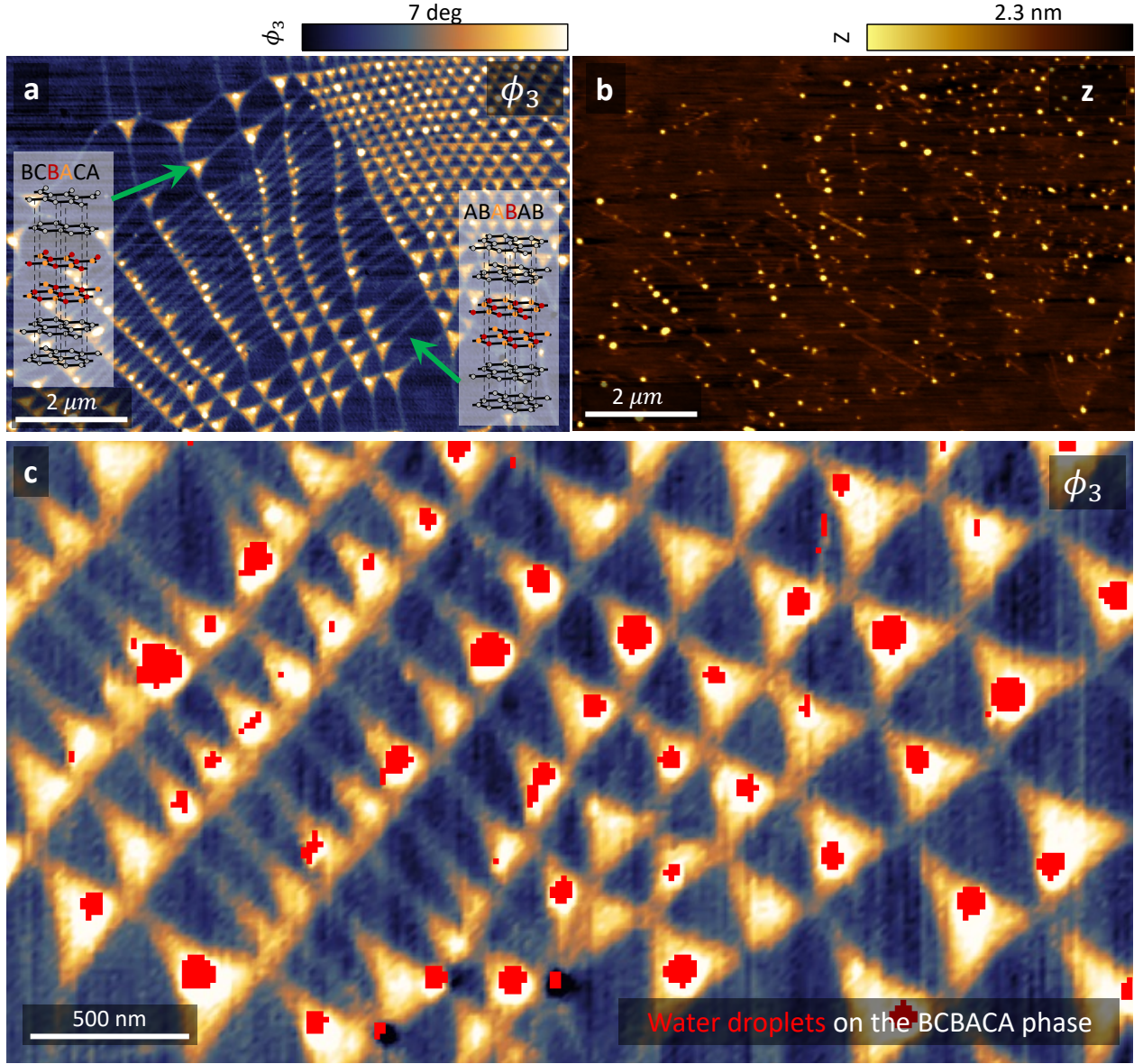


Figure 2: **Moiré-assisted chemistry in twisted double trilayer graphene** (a) For small twist angles the twisted double trilayer graphene system (tDTG) forms domains of ABABAB and BCBACA stacking configurations.²⁴ Mid-IR nearfield phase (3rd demodulation harmonic) of the tDTG, revealing the moiré superlattice. (b) A topography map of the moiré superlattice after wetting of the surface (see text for details), showing emergent topographic features. The scan was taken simultaneously with (a). (c) Finer resolution nearfield phase map of the tDTG sample, with location of water droplets (from simultaneously acquired topographic map) overlaid in red. The correlation between water droplets and one phase of the moiré superlattice demonstrates moiré selective wetting.

nearfield and tapping mode AFM scans. Remarkably, while mid-IR scans reveal the moiré are unchanged from exposure to humid conditions (Fig. 2a), tapping AFM scans (scan of Fig. 2b) show topographic features that correlate with the BCBACA phase of tDTG, as reflected in Fig. 2c. The preferential adhe-

sion of water molecules to the rhombohedral domain appears to be analogous to the tDBG system, albeit with qualitative differences. For instance, while the metallic film in tDBG completely covered the rhombohedral phase, the water molecules in tDTG form droplets which encompass the BCBACA phase, but do not ex-

tend to the domain boundary.

We note that the domain-preferential adhesion of water in the tDTG sample is a transitory effect, as repeating the same experiment several months later does not reproduce the results. Our *ab initio* molecular dynamics and continuum solvation model calculations confirm that liquid water alone does not exhibit domain-dependent adhesion, and thus, it cannot serve as a sole mediating agent for domain dependence (see Supporting Note E). Although our theoretical calculations model focus on the double bilayer graphene system and not on the double trilayer graphene system, these calculations confirm that liquid water is too distant from the surface to experience an electrostatic difference between the rhombohedral and Bernal phases, a result which should be analogous in both tDBG and tDTG systems. The transitory nature of this phenomenon further indicates that it is not intrinsic to the studied system. A comprehensive description of the history of a tDBG sample displaying off-to-on selectivity of Field’s metal adhesion is provided in Supporting Note F.

Having demonstrated moiré-assisted chemistry via domain-dependent surface interaction in twisted graphene heterostructures, we then explore whether this effect can be used to control the underlying moiré superlattice, for example, by mechanical manipulation of nanoparticles on the surface. Field’s metal nanoparticles on a minimally-twisted tDBG sample demonstrate preferential adhesion to the rhombohedral phase, as shown in the mid-IR nearfield scan of Fig. 3a. The nanoparticles form over and tile the rhombohedral domain in a characteristic pattern; a single larger nanoparticle forms in the center of the domain, surrounded by smaller nanoparticles that are all bounded by the rhombohedral domain. A histogram of the nanoparticle sizes after ablation deposition (Fig. 3a inset) further demonstrates this bimodal distribution, with a majority of particles tending on the sub-100 nm scale. The initial deposition of these nanoparticles on the graphene surface does not appear to affect the rhombohedral domain size. In contrast, in Fig. 3b-g we demonstrate that mechanical manipulation of an individual nanoparticle can re-

versibly impact the local stacking configuration of the tDBG structure in its vicinity.

A nanoparticle at the intersection of three double domain walls (Fig. 3b) is pushed by an AFM tip in contact mode towards a nearby rhombohedral domain (as indicated by arrow direction in Fig. 3b). When the particle is displaced to the center of the rhombohedral domain, we observe that the domain has collapsed to a point around the location of the nanoparticle (Fig. 3d-e). When the particle is pushed further away from the region, the rhombohedral domain appears to recover. Hence, by pushing a nanoparticle towards and away from the rhombohedral domain, we reversibly alter the size of the domain. We therefore demonstrate that the effect of domain-dependent adhesion is not passive but can actually cause a non-local reconfiguration of the lattice and the symmetry of the system by altering the stacking order around the nanoparticle. Our experimental results imply that the proximity of the nanoparticle appears to affect the stacking energy difference between rhombohedral and Bernal phases of tDBG. This causes a local perturbation, driving a local rearrangement of the domain to the extent of full collapse of the rhombohedral domain, which we expect to be reversible upon removal of the nanoparticle (as observed in Fig. 3 b-g).

We demonstrate three robust examples of domain-dependent adhesion under varying conditions. While qualitative differences exist among these three demonstrations, we attribute these differences to the conditions under which the experiments were performed. The surface temperature of the graphene systems during metallic nanoparticle dispersion may affect the morphology of the dispersion, resulting in either a thin film (Fig. 1) or discrete distributions of nanoparticles on the rhombohedral phase (Fig. 3). In the case of water (Fig. 2), we conjecture that the coverage of a certain domain will be additionally affected by the surface tension of the adsorbent. Thus, while we expect it would be possible to observe the domain-dependent adhesion of water and metal on both tDBG and tDTG systems, a clearer illustration of these phenomena would require the correct combina-

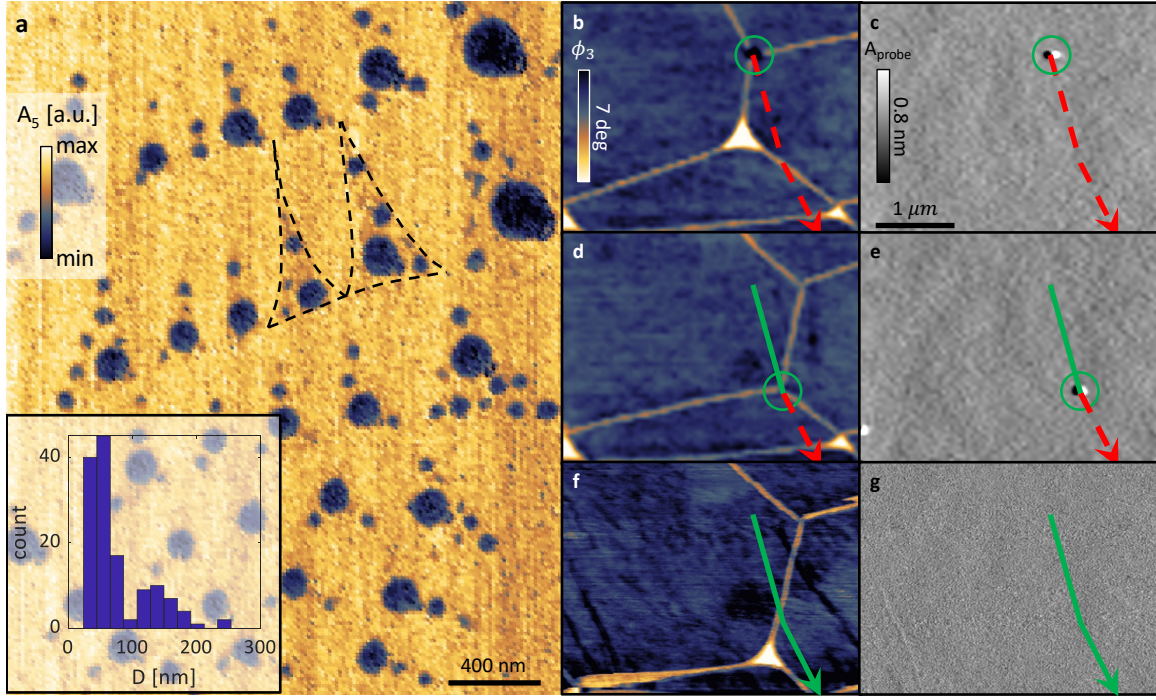


Figure 3: Moiré nanoengineering via metallic nanoparticles acting on the moiré superlattice (a) Mid-IR nearfield imaging (amplitude, 5th harmonic) after ablation deposition of Fields metal on tDBG, resulting in the distribution of nano-particles. The metallic nano-particles appear as dark and densely cover the rhombohedral phase of the moiré superlattice (schematically outlined by dashed black contours). Inset: Diameter histogram of the nano-particles appearing in the scan. (b) Mid-IR nearfield phase and (c) AFM tapping-mode probe-amplitude maps taken simultaneously over a tDBG surface. A metallic nanoparticle was found at the intersection of three double domain walls (DDWs). The particle (circled in (b,c)) seemed to be confined by the trapping potential of the DDWs. (d,e) A similar measurement performed after pushing the particle down toward the rhombohedral domain (bright triangle in (b)) using an AFM tip in contact mode. The particle settled in a new position (circled in (d,e)) at the center of the rhombohedral domain. The rhombohedral domain appears to have collapsed to a point due to proximity of the particle. (f) Nearfield and (g) probe amplitude maps after pushing the particle away from the rhombohedral domain. The rhombohedral domain seems to have recovered following removal of the particle. The overlaid path indicates the trajectory of the particle, where the already traveled vs the future parts are indicated by solid-green and red-dashed lines respectively. All panels share the scale-bar in (c). All nearfield panels share a color-scheme, and all topography panels share a color-scheme.

tion of surface temperature and adsorbent surface tension, as well as a more complete understanding of the mediating agent which is likely responsible for revealing domain-dependent adhesion.

Our experiments reveal a domain-dependent surface chemistry with robust global effects. We observe preferential adherence of various materials to the rhombohedral domains in tDBG and rhombohedral-analogous domains in tDTG systems with finely resolved features that span large areas of the twisted heterostructures.

Observation of samples both with and without domain-dependent adhesion, as well as the degradation of fine features in various environmental conditions, suggest that domain-dependent adhesion is not intrinsic to the materials studied. Our theoretical calculations indicate that domain-dependent adhesion is not an intrinsic phenomenon, which supports the suggestion that external mediating agents may play a role in the emergence of the domain dependence. Although the identity of this mediating agent remains an open question, we ex-

pect that it consists of an organic carbon-based substance to which the graphene systems are exposed during either the dry mechanical exfoliation of materials or the fabrication of the heterostructures (see Supporting Notes A and D).

More importantly, our findings reveal that domain-dependent interaction is so significant that it strongly affects the configuration of both the adsorbates and the surface itself. We observe that the local perturbation of a domain-preferentially adherent nanoparticle causes an extended, reversible reconfiguration of the moiré superlattice, namely, collapse and reappearance of a rhombohedral domain, around the manipulated nanoparticle. This allows us to propose a new approach, mechanical manipulation of the adsorbate, to reconfigure the underlying moiré superlattice. This additional level of control over stacking configuration may suggest a path towards additional control over the symmetry and electronic structure as well.

Further understanding of the domain-dependent adherent origin of nanoparticles could offer a process to manipulate systems of interest in-situ, a complementary capability to existing emerging techniques for the study and utilization of twisted heterostructure systems.^{20–22,25,26} Control of such surface manipulation mechanisms could lead to a dynamic approach towards fine control of moiré structures and more generally, to moiré engineering. Additionally, investigation of domain-dependent adhesion in non-graphene moiré systems could be an avenue for future research.^{27,28} In particular, moiré superlattices in transition metal dichalcogenide systems offer a rich platform for studying possible domain-dependent surface effects in stacking configurations not yet examined in our work.^{29,30}

Overall, our work showcases how moiré systems can serve as a scaffold to generate fine nanostructures in large scale without expensive and/or scale-limited fabrication techniques. Additionally, we demonstrate a varying surface chemistry through selective adhesion that is also controllable through nanoparticle manipulation. We believe that the moiré scaffold, to-

gether with moiré chemistry and engineering elements, holds great promise as a complete platform for future basic research and practical applications.

Supporting Information Available

- Experimental methods
- First-principles calculations of the effect of various metals on ABAB and ABCA graphene
- Mid-IR nearfield phase and topography of pristine twisted double trilayer graphene
- Detailed history of the samples
- Molecular dynamics simulations and continuum solvation model DFT calculations for effect of water on ABAB and ABCA graphene
- Demonstration of tDBG sample with and without moiré chemistry after surface treatment
- Demonstration of metallic-film coverage of the rhombohedral phase of tDBG at larger scales

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