

Chemical and structural inhomogeneity in chemical vapor deposited graphene and molybdenum disulfide based moiré heterostructures

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Van der Waals (vdW) heterostructures are attracting enormous attention due to their remarkable physical properties governed by moiré superlattices (MSLs) directly controlled by interlayer twist angles or a lattice mismatch. We investigated surface properties of moiré heterostructures made up of commercially available chemical vapor deposited (CVD) graphene and exfoliated MoS₂ layers on silicon oxide, by means of spectroscopy, microscopy, and diffraction. High resolution Auger elemental maps and spectra revealed composition inhomogeneity due to thickness variations in CVD graphene and signatures of oxidation of bare MoS₂ surface exposed to the ambient atmosphere. Low-energy electron microscopy and diffraction disclosed local stacking and crystallographic information in the graphene-MoS₂ moiré heterostructure, including evidence for small-angle twisted bilayer MSL regions in CVD monolayer graphene. This systematic study helps us to understand the overall configuration of the vdW heterostructures and correlate their structure and performance in transport experiments.

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I. INTRODUCTION

Van der Waals (vdW) heterostructures exploit the possibility of stacking atomically thin crystals of vdW materials and thus inherit the advantages of its constituting two-dimensional isolated layers. Due to a slight lattice mismatch or a rotational misalignment between adjacent layers, moiré superlattices (MSLs) are formed, whose electronic band structures display existence of moiré bands [1]. In twisted vdW structures, the band structures can be engineered by controlling the interlayer twist or orientation angles (θ). For certain “magic” angles, novel physical properties such as the emergence of superconductivity, magnetism, or a correlated insulator phase are reported for twisted bilayer graphene (BLG) [2–4] and the transition metal dichalcogenide (TMDC) WSe₂ [5]. Very recently, the occurrence of moiré mini bands in twisted MoS₂ has also been proven [6]. These “magic” interlayer twist an-

gles lie within a constrained range—for example, below or close to $\sim 1.1^\circ$ for BLG and extending to $\sim 5^\circ$ for TMDCs WSe₂ and MoS₂. Huge efforts to achieve maximum possible control on θ during the fabrication of moiré heterostructures have led to a rapidly advancing field of twisted electronics [7,8]. When graphene monolayer is stacked in the proximity of an insulating vdW layer of hexagonal boron nitride (hBN) or semiconducting layer (MoS₂, WSe₂), increased carrier mobility is achieved [9–11]. Graphene-MoS₂ heterostructures are of particular interest, on which further interesting proximity-induced effects have been reported: spin orbit coupling in graphene [12] and tuning of the MoS₂ band gap (direct-indirect transitions) via interlayer twist angles [13,14]. Therefore, for a comprehensive study of the structure-property correlations in twisted moiré heterostructures, a precise determination of θ is of prime importance.

Exfoliation of vdW bulk materials facilitates the way for mix-and-match fabrication of a wide variety of layered structures, keeping their high-purity material properties, but are limited by their overall lateral sizes. In parallel, the deposition of large-area mono/bi- or few layers graphene or other vdW materials has caught enormous attention. The growth of monolayer graphene via chemical vapor deposition (CVD) has been claimed to be well controlled over the size of a few cm on copper foils. However, the deposited layers are needed to be transferred to the substrates of interest, which

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is not a simple process. Moreover, analogous to most of the production processes in development, the material properties might vary from batch to batch and it becomes necessary to examine the quality of materials after its transfer to the required substrates for its reliable use in research or device applications.

Raman spectroscopy is a standard quality control tool for verifying the number of layers in graphene and investigating the presence of defects, edge types, and a rough estimate of interlayer orientations [15–19]. High-resolution methods such as transmission electron microscopy (TEM) or scanning tunneling microscopy (STM) reveal precise interlayer orientations in vdW layers, however, requiring nanoscale probes and destructive postprocessing or cooling to cryogenic temperatures. To enable the characterization of vdW heterostructures on length scales that are expected to be relevant to their transport properties, microscopy or structural methods of mesoscopic nature become highly important.

Low-energy electron microscopy (LEEM) is a powerful, surface-sensitive technique that combines rapid image acquisition with few-nanometer lateral resolution and a field of view ranging from several micrometers to several tens of micrometers [20]. In contrast to STM and TEM, LEEM can be readily employed to characterize as-prepared, micrometer-scale vdW structures at noncryogenic temperatures. It also allows the insertion of an illumination aperture into the incident electron beam, enabling low-energy electron microdiffraction (μ LEED) of areas down to 250 nm in diameter. This enables determination of local twist angles between stacked layers with subdegree accuracy, which is challenging to achieve with microscopic methods. LEEM has been successfully used for real-time observations of the growth of vdW layers on various substrates [21–24]. However, to date, only a small number of studies have relied on LEEM to characterize exfoliated or transferred vdW layers [25,26].

In addition to the structural properties, knowledge of chemical homogeneity in vdW layered structures is also desired. Scanning Auger microscopy (SAM) combines the imaging functionality of a scanning electron microscope (SEM) down to submicrometer resolution with highly surface sensitive chemical analysis by Auger electron spectroscopy (AES), which enables it to image the elemental composition and corresponding chemical states with a high lateral resolution ≤ 10 nm [27]. The potential use of SAM in the study of vdW layered structures is still developing; few reports exist, which are limited to transferred or exfoliated graphene layers [28,29].

Here we report on a systematic study conducted on vdW heterostructures formed by transferring commercial CVD graphene monolayers to the surface of exfoliated MoS₂ layers on silicon oxide. We employed Raman and atomic force microscopy to confirm the quality and topography of graphene and MoS₂. The chemical homogeneity in vdW layers was checked by means of Auger transitions mapping and spectroscopy, which indicate thickness variations within CVD graphene and surface oxidation of unprotected MoS₂ under ambient conditions. LEEM with selected area μ LEED reveal moiré patterns formed by the rotated monolayer graphene on MoS₂ and also disclose the occurrence of twisted bilayer graphene MSLs within the monolayer graphene. In the end,

an atypical transport behavior observed on such a graphene-MoS₂ (Gr-MoS₂) heterostructure based device is discussed, which could be related to the structural and chemical inhomogeneities detailed in this study.

II. METHODS

We used commercial *n*-doped MoS₂ (natural mineral) to exfoliate few layers on *p*-doped silicon wafers with 300 nm thick thermally grown oxide [6,31]. Commercially available CVD-grown graphene was placed on top using a PMMA assisted wet transfer technique as detailed elsewhere [12,32]. The transfer of the graphene monolayers was confirmed by confocal Raman spectroscopy using a laser with 532 nm light and 1 mW power.

Scanning Auger microscopy experiments were carried out at the DESY NanoLab [33] with the help of a scanning Auger microscope equipped with a cylindrical mirror analyzer. The areas of interest were selected by SEM imaging in order to locally measure the Auger transitions. Survey scans and elemental area maps were recorded using a primary electron beam at 10 kV and 10 nA current. The elemental maps have a pixel resolution of 256×256 , with each pixel representing the integrated intensity for the corresponding Auger transition. For quantitative analysis, high resolution Auger spectra $N(E)$ were acquired with 0.5 eV step size at the positions selected from the maps and first differentiation with respect to energy was performed to retrieve the background subtracted $dN(E)/d(E)$ spectra.

Low-energy electron microscopy experiments were carried out using a commercial LEEM microscope at the University of Bremen, Germany, which operates at a base pressure of 10^{-10} mbar. The vdW heterostructures were annealed up to 600 °C in UHV prior to LEEM measurements. LEEM [20] allows full-field imaging with lateral resolution down to a few nanometers at several frames per second. For imaging, electrons with kinetic energies between 0 eV and 50 eV are used to ensure surface sensitivity and high electron reflectivity. The optics of the microscope allow the projection of either the image or the back-focal plane of the objective onto a multi-channel plate, thereby switching between imaging (LEEM) and diffraction (LEED) modes. By inserting apertures into the electron path, specific areas of 1 μ m in diameter can be selectively illuminated, allowing the acquisition of microspot LEED patterns from well-defined regions of interest. By adjusting the incident electron energy, spatially resolved intensity-voltage [I(V)-LEEM] curves can be obtained that can serve as a material fingerprint [34]. Surface topography was investigated by means of a commercial atomic force microscope operating in tapping mode.

For measuring transport properties, the graphene was etched in the shape of a Hall bar using oxygen plasma in a reactive ion etcher. Ti/Au (4 nm/70 nm) contacts were fabricated on top of the graphene using standard electron beam lithography techniques and metallization. Doped silicon with SiO₂ was used as a backgate. The resistance was measured by passing a 10 nA AC (13.13 Hz) current through the sample and measuring the voltage through lock-in technique. The backgate voltage was applied using a source measure unit. It was

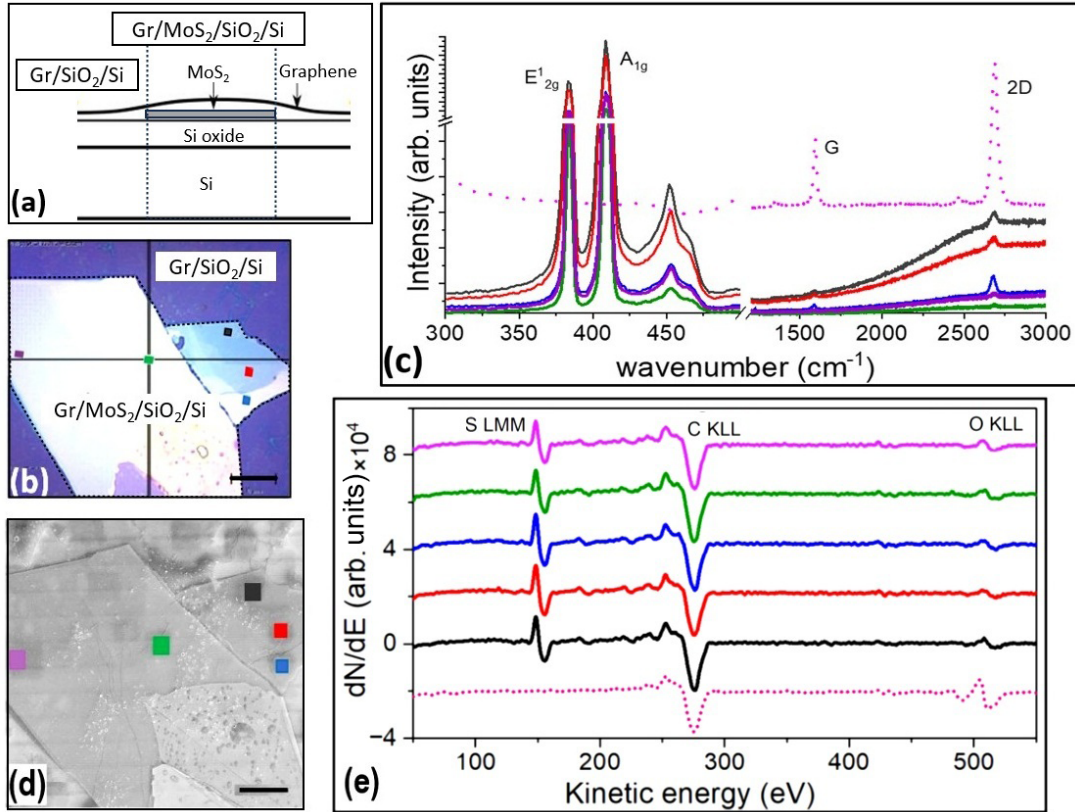


FIG. 1. (a) Sketch of the vdW layered structures presented in this work: CVD graphene layer transferred on top of a MoS₂ flake placed on SiO₂ surface; (b) optical image of the MoS₂ flake exfoliated on SiO₂/Si and covered by the CVD graphene, the boundary of the MoS₂ flake is marked by dotted lines and graphene is supposed to be everywhere; (c) Raman vibrational modes of graphene on MoS₂, for 532 nm excitation, measured at different positions marked with colored squares in (b), the dotted curve is a reference for graphene on SiO₂/Si recorded in close vicinity of the same area; (d) SEM of the graphene covered MoS₂ on SiO₂/Si given in (b); (e) differentiated Auger electron survey spectra from the marked regions in (d) and derived from the collected direct spectra shown in Fig. S1, the colors of the illustrated spectra correspond to those of the marked squares in (d) and the dotted spectrum is a reference for graphene on SiO₂/Si in a close vicinity of the discussed area; the scale bar in (b) and (d) is 10 μ m and MoS₂ layer thicknesses on the marked squares vary from 7 nm (black), 8 nm (red), 11 nm (purple), and 13 nm (blue) to 17 nm (green)—as measured by AFM (Fig. S1 in Supplemental Material [30]).

made sure that the gate-leakage current was less than 100 pA throughout the measurement.

III. RESULTS AND DISCUSSIONS

Our investigated vdW heterostructures of graphene and MoS₂ on SiO₂/Si are shown schematically in Fig. 1(a). An optical microscopy image of a typical exfoliated few-layer MoS₂ flake covered by the transferred CVD graphene layer is given in Fig. 1(b). Different contrasts suggest thickness variations in the exfoliated MoS₂ flake. The thicknesses of MoS₂ over the marked positions are revealed by AFM height profiles (Fig. S1a in Supplemental Material [30]), showing a variation from 7 nm (black), 8 nm (red), 11 nm (purple), and 13 nm (blue) to 17 nm (green) to contain more than ten atomic layers of S-Mo-S stack by considering 0.62 nm of interlayer spacing [35]. These spots were selected for Raman measurements. The characteristic vibration modes G and 2D of monolayer graphene at ~ 1580 and ~ 2690 cm⁻¹ [15], respectively, are observed in the reference Raman spectra recorded on the covered SiO₂/Si—displayed by a dotted curve in Fig. 1(c). This confirms a successful transfer of monolayer graphene.

On MoS₂, additional excitations E_{2g}^1 and A_{1g} are observed at ~ 383 and ~ 408 cm⁻¹, respectively, and the occurrence of an asymmetric Raman excitation at ~ 453 cm⁻¹ is also in accordance to the literature [36,37]. For graphene covering MoS₂, the associated G and 2D peaks sit on top of a high background arising from photoluminescence of the latter and the intensity of both peaks is much weaker compared to that of graphene directly on SiO₂/Si. Therefore, other methods are necessary to analyze the quality of transferred graphene on MoS₂.

We employed scanning Auger microscopy on the above described graphene covered MoS₂ structure. Several Auger survey spectra were collected at the preselected sites marked in the SEM image of the MoS₂ [Fig. 1(d)], which coincide with the regions investigated by Raman [Figs. 1(b) and 1(c)]. The measured Auger transitions intensities $N(E)$ are presented in Fig. S1(b) (Supplemental Material [30]), which clearly depict the prominent S LMM and C KLL Auger electron transitions originating from MoS₂ and graphene, respectively [27]. To remove a constant background, we differentiated the raw $N(E)$ Auger spectra with respect to energy and the first derivative $d(N)/d(E)$ Auger spectra given in Fig. 1(e). The C

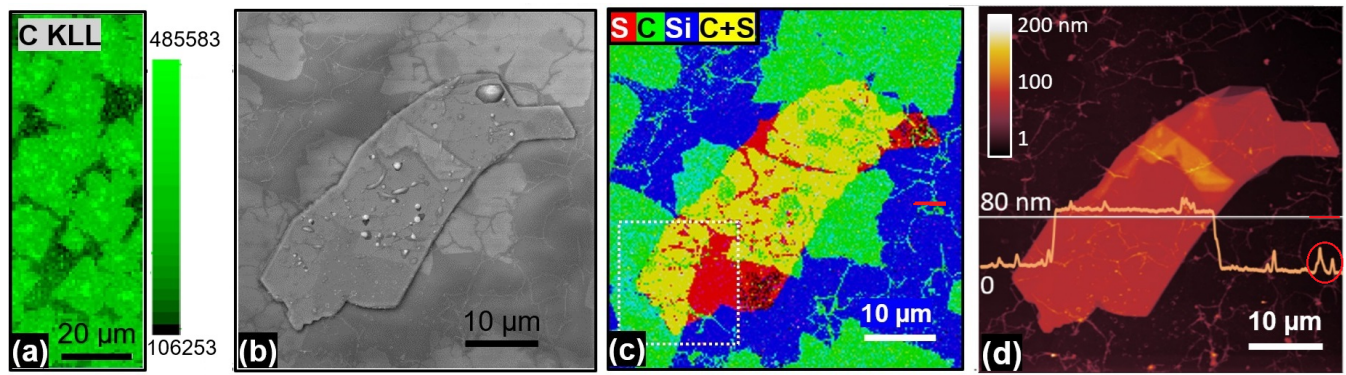


FIG. 2. Morphology and coverage of CVD graphene after transferring onto bare (a) and MoS₂ containing (b)–(d) SiO₂/Si surfaces: (a) Auger elemental map of C KLL transition (scale bar intensity in arbitrary units); (b) SEM of a MoS₂ multilayer flake exfoliated on SiO₂/Si and covered by graphene; (c) RGB Auger elemental overlay of the region in (b), with Si in blue, S in red, C in green, and a false color yellow for an overlap of C on S; (d) AFM of the region in (b), (c), including a height profile along the marked line. The red marked part of the height profile corresponds to the line marked in (c) over graphitic wires on Si.

KLL peak-to-peak intensities for graphene on MoS₂ are comparable to that of graphene on SiO₂/Si and the corresponding peak shapes are similar to those reported for exfoliated graphene on SiO₂/Si [28] and epitaxial CVD graphene deposited on metal surfaces [38,39]. Moreover, almost equally intense C KLL Auger transitions are observed for graphene on underlying MoS₂ layers of varying thicknesses ranging from 7 nm to 17 nm. These observations suggest that Auger spectroscopy can be exploited for a detailed and reliable surface study of graphene layers on various substrates.

A. Morphology and chemical homogeneity of transferred CVD graphene

To study the compositional homogeneity of the transferred graphene, we employed SAM as an elemental mapping tool and investigated several samples prepared using the same batch of as-purchased CVD graphene and MoS₂. Figure 2(a) shows a typical C KLL Auger transition map representing the transferred graphene coverage on SiO₂/Si. The carbon distribution over a 110 μm × 40 μm large area on the silicon oxide demonstrates the topography of a transferred graphene in the form of stitched patches of size typically >100 μm², whereas the dark regions represent the bare oxide surface. The observed morphology provides clear evidence of the domain microstructure in the CVD-grown graphene layer. Bright spots in the C KLL map are observed within the domains of the transferred graphene all over the sample. To investigate their origin, we have recorded higher resolution SAM elemental maps and performed LEEM/μLEED experiments at selected regions, which will be discussed in the following sections.

A graphene-MoS₂ heterostructure was investigated in detail, which was heated to 600 °C in UHV for the LEEM experiments (to be discussed later in section B) and again exposed to the environment for *ex situ* SAM measurements. A secondary electron micrograph in Fig. 2(b) illustrates a bird-shaped MoS₂ multilayer flake exfoliated on the SiO₂/Si substrate after transferring the CVD graphene monolayer on its top. Figure 2(c) shows an RGB overlay color map of Auger elemental transitions from the same region with S

LMM in red, C KLL in green, and Si LMM in blue, together with a false color yellow representing carbon on sulfur for graphene covered MoS₂. The individual maps are given in Fig. S2 of Supplemental Material [30]. This particular Gr-MoS₂ heterostructure is of lateral size >500 μm² and almost uniform thickness (~80 nm) as shown in the AFM height map by the height profile [Fig. 2(d)]. This suggests that the multilayer MoS₂ flake contains more than a hundred atomic layers of S-Mo-S stack and therefore should exhibit bulklike semiconducting properties. The graphene coverage is visible as patches of C-rich area shown in green on Si and in yellow on the top of the MoS₂. The morphology of the graphene placed on the MoS₂ follows a pattern similar to that on the SiO₂/Si surface. Many wirelike features interconnecting graphene patches are distributed over the bare silicon oxide surface, which exhibit excess C intensities in the Auger maps and are about a few tens of nanometers high as depicted from the part of the AFM height profile, marked in red. These graphitic wires were observed to be stable and unchanged during various microscopy inspections. Their origin is supposed to be from the CVD growth process because of their presence in the as-received CVD graphene on Cu foils.

To take a closer look at the chemical homogeneity of the graphene-MoS₂ heterostructures, additional Auger measurements were performed. C KLL Auger transition map with a higher lateral resolution was recorded from selected regions of Fig. 2(c), which is shown in Fig. 3(a) and depicts a partial coverage of the MoS₂ by graphene. Surface topography and morphology from the same region are presented in the AFM [Fig. 3(b)] and SEM [Fig. 3(c)] images, which show a highly rough surface of the bare MoS₂ compared to that with graphene coverage. This is discussed later in more detail under the section of surface modification in MoS₂ exposed to atmosphere. Moreover, the AFM height profile further reveals two marked steps of 1 nm and 2 nm, which can be associated to a monolayer graphene edge thickness and three atomic layers of MoS₂, respectively.

We recorded several Auger spectra at different positions marked in Fig. 3(a) to cover the kinetic energies of all existing surface elements: Si, S, Mo, C, and O. The Auger spectra $N(E)$ given in Fig. S3 of the Supplemental Material were obtained

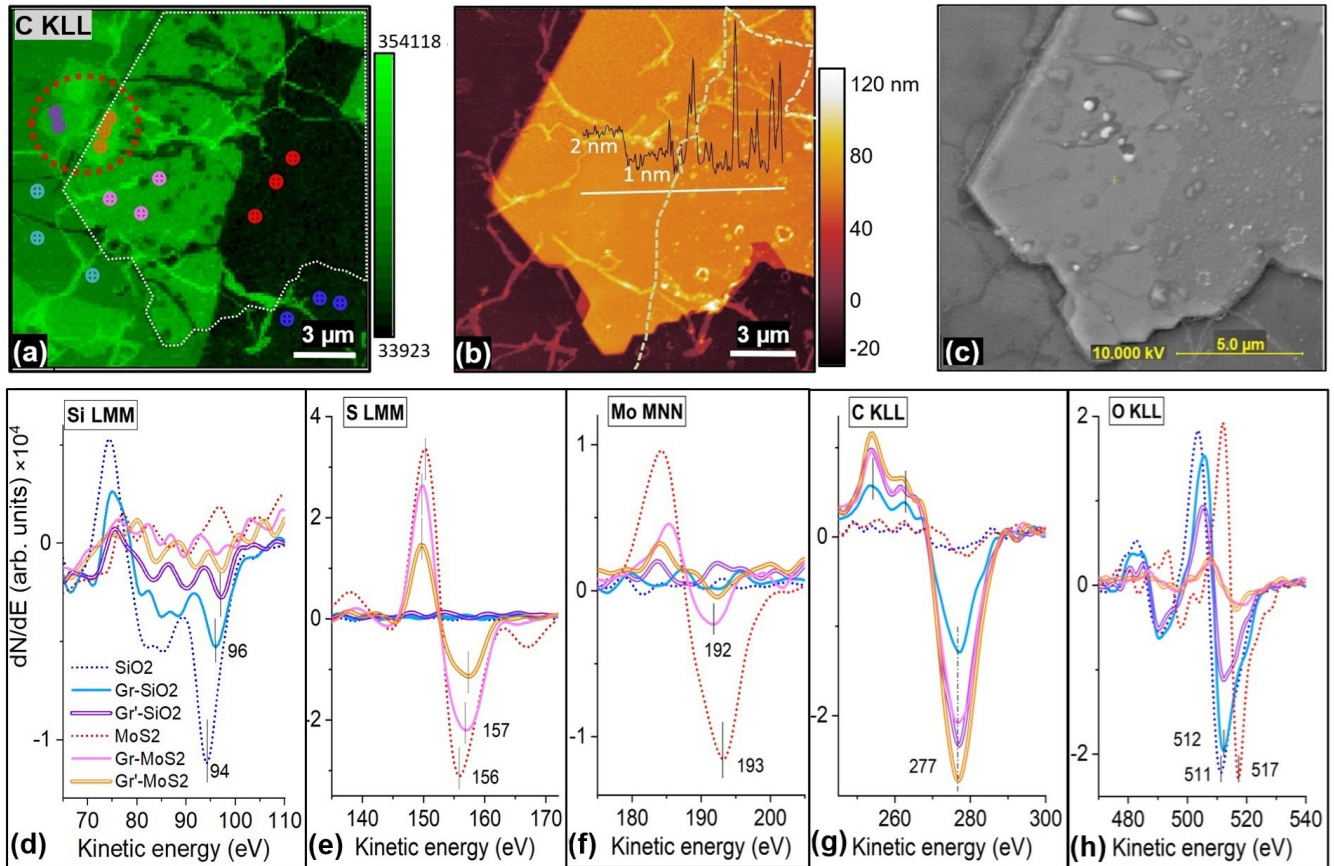


FIG. 3. (a) High resolution C KLL map of the marked square in Fig. 2(c) to show the partial graphene coverage on the exfoliated MoS₂ on SiO₂/Si with its boundary shown by the dotted white line; (b) AFM including a height profile along the marked line; the dashed line indicates the border of graphene as in (a); (c) SEM of region in (a), (b); (d)–(h) Auger spectra (dN/dE) for Si, S, Mo, C, and O Auger transitions, respectively, derived from the $N(E)$ spectra (Fig. S3 in Supplemental Material [30]) collected on different points selected in (a) and averaged over the same colored points to represent the monolayer (Gr) and few-layer (Gr') graphene. Blue (or red) curves represent the Auger signal from SiO₂ (or MoS₂): dotted, single line, and double lines correspond to the bare, monolayer graphene covered, and few-layer graphene covered surfaces, respectively.

by averaging the spectra recorded on the regions of similar contrasts in the C KLL map, marked by different colors on MoS₂ and SiO₂ regions [30]. The corresponding differential spectra $dN(E)/dE$ are shown in Figs. 3(d)–3(h). The presence of graphene is confirmed by strong C KLL transitions with a characteristic shape resembling that reported for exfoliated graphene on Si [28], while the Mo MNN Auger transitions lie in a close vicinity for the underlying MoS₂, as marked in Fig. S3. A similar observation is made in Fig. S4 associated with a nearby region of the graphene-MoS₂ heterostructure [30]. The dark regions in the C KLL map on MoS₂ (or SiO₂) exhibit the most prominent Mo and S (or Si and O) transitions with negligible C KLL intensities, suggesting that graphene is not present in these areas. However, the regions of intense C KLL transitions show attenuated Mo and S (or Si and O) transitions and thereby confirm the coverage by graphene. The overall enhanced intensities of C KLL transitions on MoS₂ compared to that on the SiO₂ suggest the influence of the underlying material with a higher electron density causing more backscattered electrons [40], which could lead to additional Auger electrons released from the surface.

The graphene-covered MoS₂ regions display rather homogeneous C KLL intensities except for a few regions of slightly enhanced intensities, for example, at the lowest left corner and the marked circle in Fig. 3(a). These represent a typical geometry of the bright spots observed in the C KLL map given in Fig. 2(a), which are frequently present in the CVD graphene layer. A well-defined geometry of these features and a fairly smooth surface evident from the AFM [Fig. 3(b)] exclude any possibility of PMMA residues or bubbles in the graphene at these areas. In such regions, an occurrence of more than one layer of graphene is possible because the stronger C KLL transitions retain their characteristic shapes associated to a graphene layer and at the same time further suppression of Auger transitions of the underlying layer elements is evident. The same was also observed in the other regions of this graphene-MoS₂ heterostructure, which is presented in Fig. S4 [30].

1. Local graphene thickness determination

The occurrence of minor regions of bi/few-layer graphene grown within large monolayer graphene by CVD on Cu or

other metal substrates is already known and has been reported to vary with growth conditions [41,42]. Therefore, for using CVD grown graphene layers in the device fabrication, one should have prior knowledge of accurate material properties of the constituting layers to understand the final device performance. Here we consider retrieving graphene thicknesses at the areas of higher intensities observed in the C KLL map in Fig. 3(a) with reference to the monolayer graphene covered MoS₂—for example, at the upper left edge of the MoS₂, marked by a red dotted circle, where a small region of few-layer graphene continues to extend from MoS₂ to SiO₂. The corresponding Auger spectra in Figs. 3(d)–3(h) show that Si LMM and O KLL decrease gradually with strengthening of the C KLL intensities on Si (blue spectra) for successively higher graphene coverage. Analogously, the S LMM and Mo MNN Auger transitions from MoS₂ (red spectra) were attenuated due to graphene coverage.

Based on the overlayer method of observing a reduction in the peak-to-peak height of the Auger transition originating from the underlying layer [43], we calculated the graphene thicknesses d by using the formula

$$d = -\lambda \sin \alpha \log_e(I_n/I_0),$$

where I_0 and I_n are peak-to-peak intensities of the Auger transitions from the mono- and few-layer graphene covered surfaces, respectively; λ is the electron's effective attenuation length for graphene at the corresponding kinetic energy and acceptance angle α for the SAM coaxial detector is 42°. Here we considered only Si and S LMM transitions for our calculations, because the Mo and O KLL transitions from MoS₂ are completely suppressed when covered by more than a monolayer of graphene thickness. The effective attenuation length (EAL) for graphene was taken as 6 Å for Si LMM (kinetic energy of 75 eV) and 8.6 Å for S LMM (kinetic energy of 150 eV), as derived from the energy and EAL correlation curve (Fig. S5 in Supplemental Material [30]) suggested in a recently reported study [29,44]. We retrieved the additional graphene thickness above the monolayer to be 3.3 Å and 3.6 Å on SiO₂ and MoS₂, respectively. This is in good agreement with the interlayer distance of 3.35 Å in graphite and therefore confirms our hypothesis that the high intensity regions in the CKLL maps of the transferred graphene contain more than one layer of graphene, typically observed for CVD graphene grown on metal substrates.

2. Surface modification in MoS₂ exposed to atmosphere

The stability of the MoS₂ surface exposed to the environment for a long time is important for reliable device applications. However, only a few reports with surface sensitive chemical studies are available. Looking on the topography in Fig. 3(b) and the corresponding morphology in Fig. 3(c), it is evident that the graphene-protected MoS₂ is almost homogeneous and smooth. On the other hand, many sub-μm size clusters/islands are distributed throughout the bare MoS₂ surface, resulting in a significantly rough appearance. This is further confirmed by the height profile along a marked line running over the graphene covered and bare part of the MoS₂ surface in Fig 3(b). Most probably, the surface of MoS₂ not covered with graphene develops defects, which could help

oxidation due to exposure to humidity or the water layer during the fabrication process and thus causes a rough surface [45,46].

The chemical composition details of the air-exposed MoS₂ surface can be obtained by analyzing the O KLL transitions together with the S LMM and Mo MNN transitions presented in Fig. 3. It is remarkable that the peak-to-peak oxygen intensity for the uncovered MoS₂ is significantly high and even comparable to that of the bare silicon oxide but with a shift to higher kinetic energy; however, very weak oxygen transitions were recorded from all graphene-covered MoS₂ regions. On the other hand, the O KLL intensities from SiO₂ gradually decreased for mono- and bilayer graphene coverage.

Besides the suppression of Mo MNN and S LMM intensities due to the graphene overlayer on the MoS₂ surface, we observe a slight change in the energy values of the peak minima as marked in Figs. 3(e) and 3(f). However, a much broader Mo MNN transition is evident for the air-exposed MoS₂ surface. To get a closer look on the Mo Auger transitions, a zoomed-in view of the Auger spectra in the 160–300 eV range is presented in Fig. S6 (Supplemental Material [30]), which shows that Mo MNN transitions exhibit three peaks in the energy range of 160 to 240 eV, with S LMM and C KLL peaks lying closely at the lower and higher energy ends, respectively. The corresponding energy values of the Auger transitions are given in Table S1 (Supplemental Material [30]), which agree very well with the literature [47,48]. All three Mo MNN peaks for the bare MoS₂ surface are significantly broader compared to that for the graphene covered one, which can be associated with (i) higher oxidation states of Mo [49] and/or (ii) a coexistence of different Mo oxides and suboxides on the unprotected MoS₂ [50]. Moreover, an appreciable difference in energy gaps between the Mo MNN Auger peaks is identified, suggesting a change in its chemical state. Based on the relative peak-to-peak intensities of Mo, S, and O, atomic concentration ratios of respective elements can be calculated for the bare and graphene covered MoS₂ surfaces. The details of the relative intensities and calculations of atomic concentration ratios are given in Table S1 [30]. For the graphene covered MoS₂ surface, an S:Mo ratio of 1.82 is derived, which is in excellent agreement to a recent *in situ* XPS study on UHV-exfoliated and annealed MoS₂ layers [51] as well as to an AES study of a sputter-cleaned MoS₂ surface [52]. On the bare MoS₂ surface, a lower value of the S:Mo ratio (0.8) and a higher value of the O:Mo ratio (1.6) suggest the existence of an oxygen-rich and sulfur-deficient phase due to the oxidation of MoS₂ on its unprotected surface.

A most plausible explanation could be that, because graphene is impermeable to the gases or water/OH- layer, it protected the underlying MoS₂ surface very well from humidity/adsorbates/surface oxidation; however, the existence of surface defects on the exfoliated MoS₂ surface is possible [45], which might facilitate the oxidation process in air. Formation of MoO₃ and its derivative oxide nanoparticles/clusters on the surface of exfoliated MoS₂ has been reported when heated in an air or humid environment up to mild temperatures [46,53–55]. The presence of small islands/clusters of insulating molybdenum oxides distributed over the MoS₂ surface can lead to charging during Auger measurements, which might explain the observed shift of the

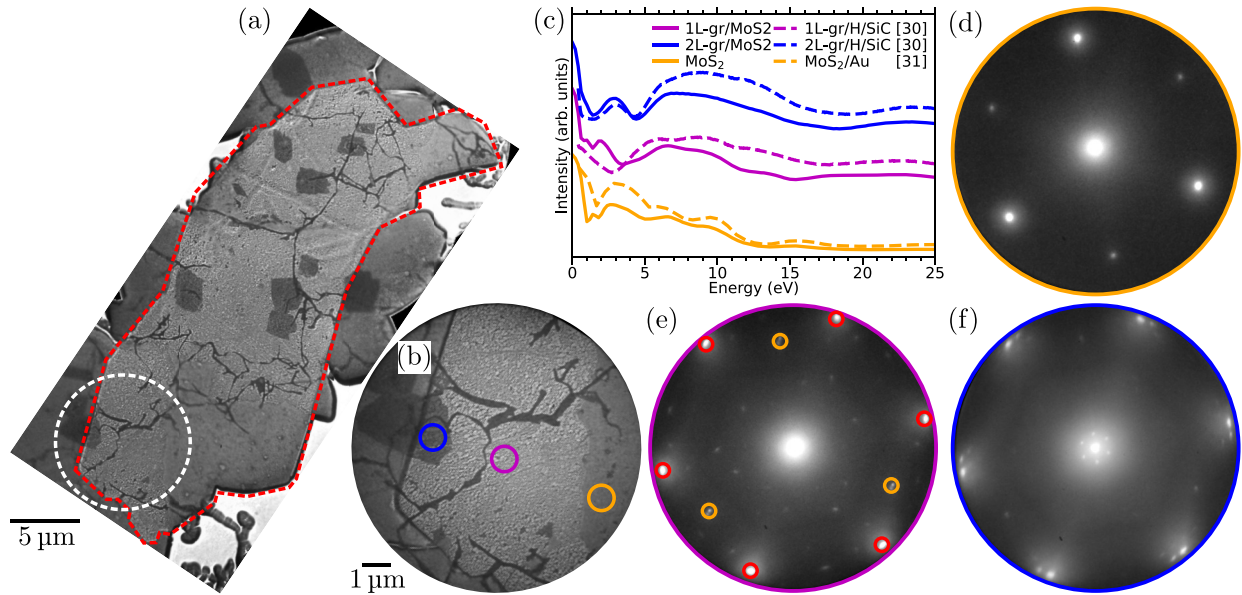


FIG. 4. LEEM/ μ LEED measurements on an exfoliated MoS₂ layer after graphene transfer: (a) LEEM image of MoS₂ partially covered by graphene, as shown in Fig. 2. The image is created by stitching together multiple LEEM images. The red dashed line indicates the outline of MoS₂. (b) Magnified view of the MoS₂, taken in the area marked by the white dashed circle in (a). The field of view is 10 μ m. (c) [I(V)]-LEEM spectra of bilayer graphene/MoS₂ (blue), monolayer graphene/MoS₂ (magenta) and MoS₂ (orange), extracted from the regions marked with circles in (b) with the same color code. For reference, I(V)-LEEM spectra of hydrogen-intercalated monolayer and bilayer graphene on SiC [56] and MoS₂/Au [57] are shown (dashed lines). The I(V) spectra are shifted vertically for clarity. (d)–(f) μ LEED images of (d) MoS₂, (e) monolayer graphene/MoS₂, and (f) bilayer graphene/MoS₂, acquired in the same regions. Integer reflections of graphene and MoS₂ are marked with red and orange circles in (e), respectively. The imaging electron energy for LEEM (LEED) images was 2 eV (40 eV).

oxygen transition to higher energy by 6 eV. This strengthens our hypothesis that air oxidation of the bare MoS₂ surface studied here occurred under ambient conditions, resulting in a rough surface due to the formation of nanoparticles/clusters rich in MoO_x phases.

B. Structure configuration in moiré superlattices of twisted graphene and MoS₂

For further structural characterization of the graphene-MoS₂ heterostructure, a combination of LEEM and μ LEED was employed (Fig. 4). Figure 4(a) shows an overview of the same MoS₂ flake as characterized by SAM in Figs. 2 and 3. A prior annealing to 600 °C in UHV should have facilitated the sublimation of MoO_x clusters/islands on an unprotected MoS₂ surface [46] and hence the charging effects, as observed in the SAM investigations, could be overcome. Within the MoS₂, different contrast levels are visible. Although most of its structure has a light gray contrast, a network of black lines and dark gray patches can be discerned. Furthermore, a slightly darker contrast is also present in the lower-right area of the MoS₂. More details of the graphene-MoS₂ heterostructure are observed in Fig. 4(b), which is a LEEM image of area in Fig. 3(a) obtained at higher magnification. A comparison with the SAM maps in Fig. 3(a) shows that the light-gray area, black lines, and dark-gray patches correspond to areas of high C KLL intensity and therefore graphene-covered MoS₂ regions, while the area in the lower-right region is not covered by graphene. Further classification is possible by analyzing the intensity vs energy [I(V)-LEEM] spectra acquired in different regions of the exfoliated MoS₂, which are the light gray

(blue) and dark gray (magenta) graphene covered regions, and the uncovered region (orange) in Fig. 4(b). The corresponding I(V)-LEEM spectra are shown in Fig. 4(c). The I(V) spectrum extracted from the uncovered regions is in good agreement with the reference curve for MoS₂ on Au [57], indicating that this region is indeed bare MoS₂. The I(V) spectrum extracted from the graphene-covered dark gray area (blue) shows two minima in the energy range between 2 eV and 7 eV, whereas the I(V) spectrum from the light gray area (magenta) only exhibits one minimum in the same energy range. It is well established that the number of minima observed for the reflected electron intensity curves in the low energy range is a direct measure of the number of graphene layers [21,23]. The comparison of the measured I(V)-LEEM spectra with those of H-intercalated monolayer and bilayer graphene on SiC [56] reveals a good agreement of the bilayer graphene spectra. For the monolayer graphene spectra, the minimum appears to have shifted to higher energies, likely due to the presence of MoS₂ underneath, which is not fully screened by the monolayer graphene. This further indicates that the light and dark gray areas correspond to monolayer and bilayer graphene, respectively, as previously suggested by SAM.

In order to characterize graphene on SiO₂ via low-energy electrons, we chose a part of the monolayer and bilayer graphene crossing over the edge of the MoS₂ flake to the SiO₂/Si support, as depicted in Fig 4(b). Unlike graphene on MoS₂, monolayer graphene on SiO₂ does not show a minimum in the I(V)-LEEM spectrum (Fig. S7 [30]) and exhibits only a single minimum around 3 eV for the bilayer graphene. These differences indicate the influence of the graphene-support interface, where exfoliated MoS₂ has a nearly perfect

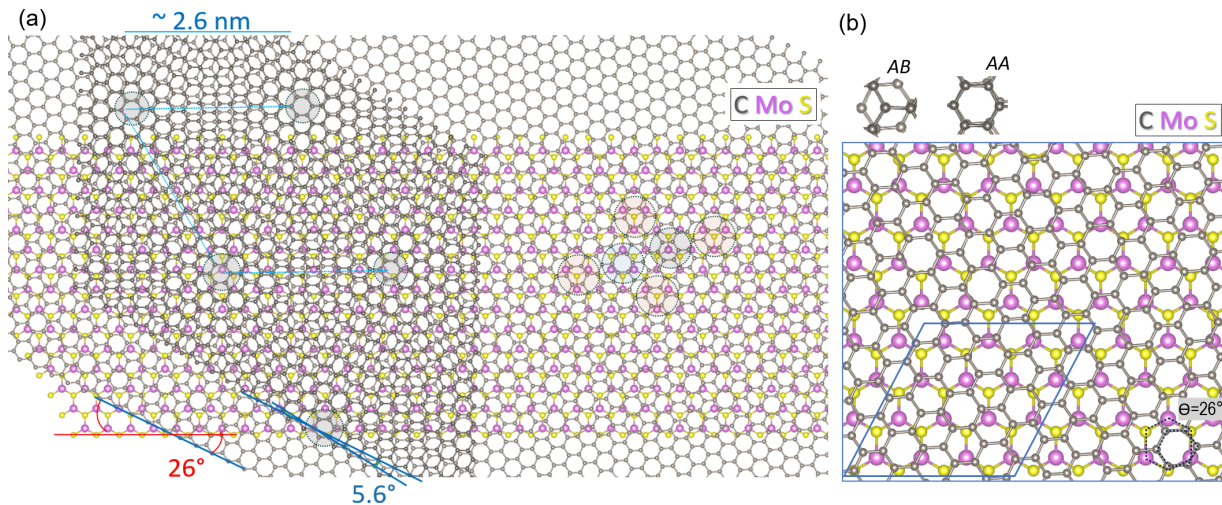


FIG. 5. (a) Illustration of the moiré superlattice formed by a 5.6° twisted bilayer graphene region, included within the monolayer graphene layer, which is placed over the MoS₂ surface with an orientation of 26°; the orientation (red) and twist (blue) angles are according to the LEED patterns in Figs. 4(e) and 4(f), respectively; (b) zoomed-in view of stacking AA and AB in the twisted BLG and the graphene/MoS₂ heterostructure in (a). C, Mo, and S atoms are shown in black, magenta, and yellow colors, respectively.

crystalline surface, while thermally grown silicon oxide is known to be prone to a high defect density on its surface.

Selected-area μ LEED patterns were recorded on bare regions of MoS₂, Fig. 4(d), monolayer graphene-covered regions, Fig. 4(e), and bilayer graphene-covered regions, Fig. 4(f), at the positions indicated in Fig. 4(b). The diffraction spots marked by orange and red circles in Fig. 4(e) correspond to the MoS₂ and graphene surfaces, respectively. The μ LEED pattern acquired on bare MoS₂ exhibits threefold symmetry, which, consistent with the I(V) spectrum, indicates bare, single-crystalline MoS₂. The μ LEED pattern acquired on monolayer graphene shows the integer reflections of both graphene and MoS₂, as well as additional spots due to multiple scattering. The angle between the integer reflections of graphene and MoS₂ indicates a 26° rotation angle between the vdW layers. The μ LEED pattern acquired on bilayer graphene shows two sets of graphene integer reflections and a well-resolved moiré pattern in the center. The MoS₂ reflections appear considerably weaker due to screening by the overlaying graphene layers. The moiré pattern originates from the twisted layers of bilayer graphene, which exhibit an interlayer rotation angle of 5.6°. These results confirm that commercial CVD-grown graphene contain minor regions of bilayer graphene in which the upper layer is not always aligned with the bottom layer, thus forming minor moiré superlattice regions.

μ LEED patterns are sensitive to the local registry of atoms on the surface and thereby reveal the stacking in surface layers. An atomic model to illustrate the above discussed graphene/MoS₂ heterostructure is constructed using VESTA software [58], as given in Fig. 5(a), which depicts an incommensurate moiré structure formed by 26° oriented graphene on an MoS₂ surface, containing a region of 5.6° twisted BLG moiré superlattice with a period of ~ 2.6 nm. Another moiré heterostructure observed by means of μ LEED on the same sample is presented in Fig. S8 of the Supplemental Material

[30]. Various possible stacking exist in these twisted BLG and graphene-MoS₂ incommensurate moiré heterostructures. The most distinct stacking regions are presented in Fig. 5(b) and Fig. S8(c). “AA” stacking in twisted BLG correspond to the overlapped positioning of C-C hexagonal rings in the upper and lower graphene layers, while in “AB” (Bernal) stacking, the lower carbon layer is shifted such that only alternating carbon atoms’ positions match with intersecting C-C hexagonal rings in both layers. The interlayer distance in twisted BLG for $\theta \leq 10^\circ$ shows a variation with stacking: the interlayer distance is maximum at “AA” stacking and minimum at “AB” stacking regions, which results in atomically corrugated graphene layers [59]. According to the DFT calculations from Ref. [59], for 5.6° twisted BLG, the “AA” and “AB” interlayer distances are approximately 3.55 Å and 3.37 Å, respectively, which would result in an average interlayer distance of 3.46 Å. It is interesting to note that this value is closer to the graphene thickness derived from the Auger analysis discussed earlier, compared to the 3.35 Å interlayer distance in natural graphite.

In the graphene-MoS₂ heterostructure depicted in Fig. 5(b), a 4×4 MoS₂ cell can be matched closely to a $3\sqrt{2} \times 3\sqrt{2}$ graphene unit cell (with an offset of 4°). Here, most distinct stacking regions are (i) alternating C atoms directly over S (Mo) atoms with Mo (S) atom below the center of the graphene C-C hexagonal ring structure and (ii) all C atoms positioned over Mo-S hexagonal ring structure with no atoms below the center of the graphene C-C hexagonal ring. In the proximity of graphene, S-S interplanar spacing in MoS₂ have been reported to be influenced by the interlayer orientation angle θ [13]. To utilize extraordinary properties of twisted moiré heterostructures, micro/nanoelectronic devices are commonly fabricated using the tear-and-stack method but can contain twist-angle disorders and strains [60]. The presented approach of using μ LEED can be exploited further to probe local stacking of vdW layers for controlling the interlayer

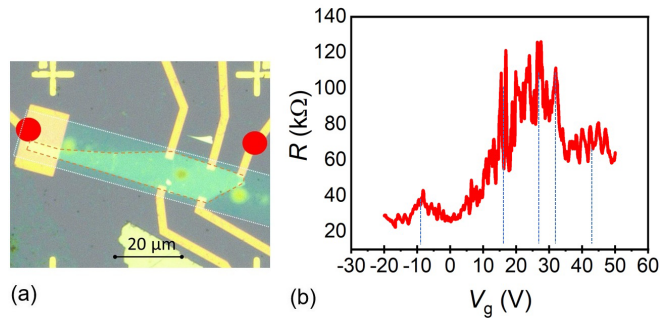


FIG. 6. (a) Optical image of a graphene-MoS₂ heterostructure device used to measure its transport properties between two contacts marked with red circles, white dots, and orange dashes represent the boundaries of graphene and MoS₂, respectively. (b) Two-probe resistance of the CVD graphene on MoS₂ at 1.5 K as a function of the gate voltage.

orientation in twisted moiré heterostructures with a high accuracy.

C. Transport characteristics

We performed transport measurements at low temperature of one such graphene-MoS₂ heterostructure fabricated identically. Figure 6(a) shows the optical image of the device. The graphene monolayer can be seen in light blue color on top of which gold contacts are fabricated and the edges of the underlying MoS₂ flake are marked with orange dashes for clarity. The transport characterization was restricted to two-probe measurements between the contacts marked by red dots, because other contacts were not functional. The two-probe resistance of the device at 1.5 K as a function of the backgate is shown in Fig. 6(b). For a standard monolayer graphene device, this curve is expected to be smooth and the resistance peaks at the charge neutrality point, i.e., when the Fermi energy is at the meeting point of the two Dirac cones [12]. On the contrary, we observe a very noisy behavior with multiple peaks. We have marked the positions of some of the prominent peaks by vertical lines in the plot. The noisy behavior is expected to arise from the grain boundaries [61]. Twisted bilayer graphene devices are known to show multiple resistance peaks as a function of the gate voltage [62]. We would like to emphasize here that the structural characterizations were performed on different samples; however, we expect the structural properties to be similar. Since this is a large sample area ($\sim 1500 \mu\text{m}^2$ between the contacts), it contains multiple grain boundaries and small-sized ($\lesssim 10 \mu\text{m}^2$) MSL regions with many angles and therefore does not show signatures specific to a certain angle or of the monolayer but some dominant ones give rise to such an atypical transport behavior.

IV. CONCLUSIONS

The present study suggests the use of powerful surface sensitive methods in investigating vdW moiré heterostructures to address current challenges in accessing local information on their compositional homogeneity, accurate interlayer layer twist angles, and structural configuration. The discussed methods are not yet common in practice for characterizing stacked vdW MSL heterostructures.

We employed scanning Auger microscopy to study the chemical (in)homogeneity in vdW layers with high lateral resolution revealing thickness variations in CVD graphene and signatures for MoS₂ surface oxidation under ambient conditions, which remained protected if covered by a graphene layer. We exploited LEEM/ μ LEED to determine the local interlayer twist angles precisely and associated stacking in the graphene-MoS₂ moiré heterostructures. We demonstrated that commercially available CVD monolayer graphene contained few μm sized regions of twisted bilayer graphene moiré superlattice. The combined effects of grain boundaries and moiré superlattice in graphene are expected to be responsible for the observed atypical transport properties on such structures that are normally assumed to be a monolayer. Although CVD is the most promising method to achieve BLG with homogeneous θ in the twisted area but control to achieve the desired θ is not trivial and needs further advancements for its commercial production.

The presented methodology can be extended to other vdW heterostructures for a better understanding of the structure-property correlations in a broad range of applications and thus help to increase reliability and reproducibility of device fabrication.

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L.B. and C.H.S. contributed equally to this work.

DATA AVAILABILITY

The data that support the findings of this article can be provided upon reasonable request.

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