Anisotropic strain induced soliton movement changes stacking order and bandstructure of graphene multilayers

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Abstract

Deterministic control over the electronic properties of solid-state matter is considered as highly rewarding to understand interaction interaction-driven electronic effects. For example, in graphene stacks the electronic properties are greatly affected by the lateral layer arrangement as the most stable configurations, Bernal and rhombohedral stacking, exhibit very different electronic properties. Nevertheless, they can coexist within one flake, separated by a strain soliton that can host topologically protected states. Clearly, accessing the transport properties of both stackings and the soliton is of high interest. However, the stacking orders can transform into one another and therefore, the seemingly trivial question whether reliable electrical contact can be made to either stacking order can a priori not be answered easily. Here, we show that manufacturing metal contacts to multilayer graphene can move solitons by several µm, unidirectionally enlarging Bernal domains due to arising mechanical strain. Using DFT-modeling we corroborate that anisotropic deformations of the multilayer graphene lattice increase the Bernal-stacking stability over the otherwise energetically favored rhombohedral stacking. Finally, we have devised systematics to avoid soliton movement, and how to achieve stable contacts to both stacking configurations.

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Main

Recent interest in graphene multilayers stems from their diverse (opto-)electronic properties that depend on layer thickness\(^1\), stacking order\(^2\)\textsuperscript{5} and twist angle of subsequent layers against one another\(^6\)\textsuperscript{8}. For example, Bernal-stacked bilayer graphene shows an electrically tunable bandgap\(^9\) and pseudospin polarizability\(^10\), unconventional quantum\(^11\) and fractional-quantum Hall effects,\(^12\)\textsuperscript{14} as well as a renormalization of the density of states near charge neutrality in the absence of a magnetic field\(^15\)\textsuperscript{20}. Furthermore, it has been recently shown that slightly twisting the two layers dramatically changes the band structure, allowing the observation of unconventional superconductivity\(^7\) and Mott insulating behavior\(^21\). An addition to these diverse properties of graphene bilayers are the recently identified topologically protected states at boundaries between AB and BA stacked graphene bilayers\(^22\)\textsuperscript{25}. In thicker graphene flakes, the physics can be expected to be even richer, since naturally two stable forms of stacking, Bernal (or ABA, Fig. 1a) and rhombohedral stacking (or ABC, Fig. 1b) exist, both with distinct bandstructures and electronic properties\(^2\)\textsuperscript{4}. For example, in thick rhombohedral graphene stacks flat electronic bands are present\(^26\)\textsuperscript{27}, which might support superconductivity\(^28\)\textsuperscript{30}. To explore the unique electronic properties of both stacking orders, precise knowledge of the local stacking type as well as stable electric contacting of both phases is necessary. This is even more important, since both stacking orders can occur within the same flake. Moreover, as recently shown, both types of stacking can be transformed into one another by applying an electrical field\(^31\), strain\(^32\), high temperatures\(^33\), doping\(^34\) or a mechanical force\(^35\) – some of which are also present during the patterning of electrical contacts. With the goal to assess the stability of the stacking orders under metal contact processing and to devise stable contacting schemes, we have investigated a series of multilayer graphene flakes.

Before electrically contacting our graphene multilayers, we identify the local stacking order using Raman spectroscopy of the 2D, G and M mode (Figs. 1c and d) as previously shown\(^36\)\textsuperscript{39}. Since within a single multilayer graphene flake Bernal and rhombohedral stacking order can be present, we spatially resolve the stacking domains using scanning Raman spectroscopy\(^37\). Fig. 2 shows in addition to an AFM (Fig. 2a) and optical image (Fig. 2b) a map of the full width at half maximum (FWHM) of the 2D peak of a tetralayer graphene flake after exfoliation (Fig. 2c), that reveals two different stacking domains. Knowledge of the local stacking order allows us to selectively pattern contacts on the Bernal and rhombohedral domains via standard electron beam lithography, metal evaporation and lift-off techniques. An optical image of the flake with fully processed contacts is shown in Fig. 2e and an AFM image in Fig. 2d. In the latter, new wrinkles between the contacts can be observed. The appearance of such folds can be caused by compressive and/or shear strain\(^40\)\textsuperscript{42}, induced by thermal expansion effects\(^42\)\textsuperscript{43}. 
At first sight, the appearance of strain and the occurrence of folds upon deposition of metal contacts does not seem worth a separate discussion, especially since numerous previous works have reported metal contacts to both rhombohedral and Bernal stacked multilayer graphene. Nevertheless, we have investigated the contacted flake again with scanning Raman spectroscopy. Surprisingly, we find that the rhombohedral domain has almost completely vanished (see Fig. 2f), even though it was covering an area of about 40 µm² prior to contact deposition. From the Raman 2D signal we can clearly identify that it has transformed to Bernal stacking (see Fig. 2j) and the M mode (Fig. 2j, inset). Finally, the D peak is negligible indicating that the transition has not introduced defects (data not shown). Since the lateral resolution of our Raman microscopy setup is about 1 µm, we cannot rule out that the rhombohedral part of the flake might have been broken up into several nanodomains of Bernal and rhombohedral stacking. We have therefore investigated the local nature of the transformed flake by infrared scattering-type scanning near-field optical microscopy (s-SNOM) at 20 – 30 nm spatial resolution. This technique is highly suitable in our case, as it allows (i) to distinguish Bernal from rhombohedral stacking owing to their different infrared responses, and (ii) to resolve boundaries between stacking domains in multilayer graphene (e.g. boundaries between ABA – BAB or ABA – ABC) due to reflections of surface plasmons. An s-SNOM image of the processed flake (Fig. 2h) confirms the spatial arrangement of the domains, as revealed by the Raman map (Fig. 2f). With its high resolution, the s-SNOM image corroborates that the transition from rhombohedral to Bernal stacking proceeds homogenously upon the contacting process. In other words, the transition does not nucleate at multiple points within the flake, but seems to be induced via movement of the strain soliton at the Bernal/rhombohedral stacking boundary. For a better visualization of the transition, a scheme including the rhombohedral, Bernal and transformed region is depicted in Fig. 2g.

This transformation from rhombohedral to Bernal stacking has been observed in about 50 % of the contacted samples, which are flakes with 3 to 7 graphene layers. The extent of the transition varies, ranging from an almost complete vanishing of the rhombohedral domain as in Fig. 2, to only a small movement of the domain wall by a few hundred nm. In flakes in which no transition occurs, the soliton might be pinned, as observed before by STM measurements. Further below, we discuss additional reasons why in some cases the soliton does not move.

We have made an attempt to clarify the detailed mechanism that causes the transition, with the aim to devise a way to avoid the transition or enhance it selectively. The most remarkable observation next to the stacking transition in the flake shown in Fig. 2 is the occurrence of wrinkles in the transformed region. This might imply that the transformation is directly correlated to or caused by the appearance of wrinkles. However, such wrinkles do not necessarily appear in the parts of the flake in which a
transition takes place. For example, in Fig. 3, we show details of a graphene trilayer that comprises a Bernal/rhombohedral stacking boundary. After fabricating metal contacts, as in the case of the tetralayer, the high-resolution s-SNOM image (Fig. 3g) shows that again the domain boundary has shifted, slightly increasing the Bernal-stacked region, whereas the wrinkles occur primarily in the untransformed rhombohedral part. This implies that both the stacking transformation and the topography changes appear simultaneously during processing and possibly originate from the same cause, however, the folds themselves do not trigger the transition.

To identify the cause of the transition, we apparently need to test if during our process steps effects occur that can lead to a stacking transformation, with special attention to mechanisms that are also known to induce folds. A few methods including applying an electrical field\textsuperscript{31}, strain\textsuperscript{32}, high temperatures\textsuperscript{33}, doping\textsuperscript{34}, an electron beam\textsuperscript{25} and recently also a mechanical force\textsuperscript{35} have been reported to cause a movement of solitons or a transformation between Bernal and rhombohedral graphene or vice versa. Since we do not apply an electric field across the flake, we can exclude this directly as possible trigger for a transformation. We next turn to doping as possible cause. In principle, metals, when in close contact with graphene, can lead to doping\textsuperscript{51}. However, our contacts are deposited locally at the edges of our multilayers, and screening lengths are well below 100 nm\textsuperscript{52}, whereas the transition occurs non-locally across several µm. Furthermore, we have deposited a few nm of titanium onto several multilayer flakes with both forms of stacking and have not observed a transition (data not shown). Finally, the contacts seem to rather hinder than foster the movement of the domain wall (see Fig. 3 and Supplementary Fig. S5). Additionally, we have corroborated that an electron beam does not cause the transition (for details see Supplementary Fig. S1). Consequently, strain and high temperature are left as possible explanations, both possibly occurring during processing. We analyzed the processing steps of cleaning, heating, spin-coating PMMA and softbake observing no transformation (see Supplementary Fig. S1). Thus, we conclude that the metal evaporation is the decisive step.

During the electron beam evaporation of metals, the substrates are held at about 10 – 20 °C by water-cooling the sample holder. Still, the graphene flakes can heat up locally caused by the thermal load of the condensing metals. Consequently, due to the large difference in the thermal expansion coefficients of graphene (about -8.0 x 10\textsuperscript{-6} 1/K, ref. \textsuperscript{53}), PMMA (about 100 x 10\textsuperscript{-6} 1/K, ref. \textsuperscript{54}) and the SiO\textsubscript{2} substrate (about 0.6 x 10\textsuperscript{-6} 1/K, ref. \textsuperscript{55}), mechanical strain can occur\textsuperscript{42}. To understand the combined effect of heating and mechanical strain during metal evaporation and to test if we can amplify the scale of the transformation, we deliberately increased the substrate temperature during metal deposition to 200 °C. AFM and s-SNOM images before and after metal evaporation at 200 °C and lift-off are shown for a tetralayer graphene flake in Figs. 4a – d (corresponding Raman data in Supplementary Fig. S2). Since
200°C is well above the glass transition of PMMA\textsuperscript{56}, one can assume that the corners of the patterned PMMA get softened and we consequently find that the metal contacts are torn off during the lift-off, resulting in an inhomogeneous surface of the processed flake (see Fig. 4c). Nevertheless, our measurements after processing show new wrinkles in the topography (see Fig. 4c) and a full transformation to Bernal stacking (see Fig. 4d). Apparently, the increase of the substrate temperature during metal evaporation has amplified the scale of the transformation in case that additionally the resist has been patterned. Therefore, we anticipate that the combination of thermal heating and local clamping of parts of the flake with metal contacts causes compressive strain and shear forces, which then lead to the observed folds\textsuperscript{40–42} and, more importantly, induce the transition.

To gain insight into the role of mechanical deformation of the graphene lattice on the relative stability of rhombohedral and Bernal stacked graphene multilayers, we have performed a detailed density-functional theory (DFT) study. More specifically, by atomistic calculations of the lattice of trilayer graphene, we have studied the relative stabilities of the stacking orders under compression and stretching. The unit cell for Bernal and rhombohedral stacking with the corresponding lattice vectors are shown in Figs. 1a and b, respectively. We have compared the stability by calculating the total energy per atom of rhombohedral- and Bernal stacked trilayers after fully relaxation of atomic positions and the lattice vectors. Unlike to what has been reported for bulk graphite\textsuperscript{57}, the rhombohedral stacking is slightly more stable, by about 7.8 \times 10^{-5} \text{ eV/atom}. This is our basis to test the impact of “homogenous” compression and stretching by applying a change of a factor $\delta$ to the lattice vectors, so that they are scaled as $\vec{a}_1' = (1 + \delta)\vec{a}_1$, $\vec{a}_2' = (1 + \delta)\vec{a}_2$. In this picture, $\delta < 0$ corresponds to homogeneous in-plane compression, and $\delta > 0$ describes stretching. In Fig. 5a, the difference in total energy per atom $E_B - E_R$, with the energy per atom for Bernal $E_B$ and rhombohedral stacking $E_R$, is plotted for different values of $\delta$. Rhombohedral stacking increases in stability upon stretching the trilayer. Under compression, the Bernal stacking was found to gain stability, however, the amount of compressive strain needed for Bernal to be more stable than rhombohedral stacking is well above $\delta > 15 \%$.

The response of the trilayer is different in the case of applying anisotropic, i.e. directional strain (strain). In Figs. 5b and c we show the difference of total energy for compression and stretching applied along the x- and y-direction, respectively. We find that in both directions for compressive and tensile strain of about 2 – 3 \%, Bernal stacking becomes more stable than rhombohedral stacking. To complete the picture of the stability under lattice deformation, we have also analyzed the case of anisotropic deformation that keeps the unit cell area constant. This means that compressing in x-direction leads to stretching in the y-direction and vice versa (for details see Supplementary Fig. S3). Again, we find that compressive and tensile strain slightly below 2 \% is enough to stabilize Bernal stacking. Although
we have considered only trilayer graphene in our DFT study, the results should be similar in the case of more layers where Bernal stacking is favored even more.

The DFT-calculations describe the experimental observations very well, as we discuss in the following. As previously reported, the stress transfer between graphene and PMMA is very good for small strain values, however, at strain of 0.6 % or higher, slippage between the two materials can occur. In the case that PMMA homogeneously covers a multilayer graphene flake (see Fig. 5d), upon heating and subsequent cooling the resist, the multilayer experiences homogeneous tension and/or compression, depending on whether slippage has occurred or not. Given the typical temperatures reached in our experiment (180 °C in the case of the PMMA softbake), the reached homogeneous expansion of PMMA and the resulting tension/compression of graphene is below 1.6 %. In agreement with Fig. 5a, such a homogeneous strain does not change the stability from rhombohedral to Bernal stacking, and we never have observed a change in stacking order under these circumstances. The situation is different in the case that e-beam lithography has been performed on the PMMA resist, as shown in Fig. 5e. Even though the detailed geometry of the sample with contacts is complicated, it seems plausible that partial local pinning of the flake in the heated condition and the subsequent cool-down (where parts of the flake might have slipped with respect to PMMA) lead to anisotropic strain, which significantly lowers the energy for the Bernal-stacked state (see Figs. 5b and c). Apparently, this anisotropic strain leads to folds and provides a driving force for the soliton movement causing the transition (in our calculations we find that 2 % of strain is needed in good agreement with the expected strain of up to 1.8 % in case the substrate is heated during evaporation). Since we do not observe a significant shift of the 2D Raman mode in the finished flakes, we conclude that after the resist has been removed, the graphene is unstrained – most probably due to the appearance of folds. Finally, we have also observed a rhombohedral-to-Bernal-stacking transition in graphene multilayers upon their dry transfer onto h-BN flakes during which apparently also an anisotropic strain pattern is present (details to be reported elsewhere).

In order to test if the transformed regions are stable under typical measurement conditions, we performed ex-situ measurements with the transformed samples. We cooled the samples in liquid nitrogen or helium and heated the contacted samples to 400 °C and performed AFM, Raman and s-SNOM measurements afterwards (see Supplementary Fig. S4). The folds moved, changed height or disappeared but the arrangement of the stacking domains remained unchanged.

While we have now established heating the sample during metal deposition as a method to induce soliton movement, to access the physics of rhombohedral domains or of states at the rhombohedral-to-Bernal-stacking boundary, it would be beneficial to devise a way to avoid soliton movement upon metal deposition. To this end, the applied strain during processing needs to be kept as small as
possible. This can be achieved by, firstly, assuring that the sample is cooled well during the evaporation, since a higher temperature amplifies the scale of the transition, and secondly, by choosing the right pattern of contacts. The latter can help to prevent a transformation, since the contacts seem to hinder the movement of the soliton. During the transition, the soliton at the Bernal/rhombohedral stacking boundary moves towards the rhombohedral part. If the contacts lie across the domain wall, the soliton shifts only slightly in between the contacts (see Fig. 3), thus, these contacts prevent a free movement of the soliton. In case a contact fully separates the rhombohedral domain from the Bernal part, the transformation stops (see Supplementary Fig. S5). Finally, we have found that a dense contact pattern around the edges of the flake effectively suppresses soliton movement (see Supplementary Fig. S6). Possibly, the high density of metal contacts clamps the flake and reduces anisotropic strain.

In summary, we have observed that anisotropic strain can induce a movement of stacking solitons at the rhombohedral/Bernal boundary in multilayer graphene. In our experiments, we induced this anisotropic strain by local metal deposition. Even though it is known that graphene layers can easily move against one another\textsuperscript{60}, the here reported transition is surprising since numerous literature reports have contacted rhombohedrally stacked multilayer graphene without reporting such a transition\textsuperscript{2,3,44,45}. The observation that fabricating metal contacts can lead to a lateral movement of van-der-Waals multilayers will also potentially be interesting for van-der-Waals heterostructures, where atomic lateral precision or twist angles are required\textsuperscript{7}.

Methods

Sample preparation. We mechanically exfoliated multilayer graphene flakes from an HOPG block onto a SiO\textsubscript{2}(300 nm)/Si substrate. The number of layers was determined by optical microscopy and Raman spectroscopy. The AFM images were recorded using an AFM (Dimension 3100, Veeco) in tapping mode.

Electron beam lithography. The samples were cleaned using acetone and isopropanol. Then, the resist (PMMA 950K with 4.5 % anisole, Allresist) was spin coated onto the substrates. A softbake was performed at 180 °C for 5 min. The resist was patterned using an electron beam (e-Line system, Raith). Afterwards, the resist was developed using a 1:3 mixture of MIBK and isopropanol. Finally, the metals were deposited using e-beam evaporation under high vacuum conditions (pressure about 10\textsuperscript{-7} mbar) while the substrates were cooled (10 – 20 °C). The experiments in which the samples are heated to 200°C during evaporation were performed in a thermal evaporator. For all samples, a thin titanium layer of about 1 nm was applied as adhesion layer. Subsequently, gold was deposited forming the 30 – 80 nm thick contacts. The evaporation rates were about 0.1 Å/s and 1.0 Å/s, respectively.
**Raman measurements.** The spectra were recorded using a Raman system (T64000, Horiba) with a laser excitation wavelength of 514 nm. The size of the laser spot on the sample was about 1 µm and the spectral resolution was 0.7 cm\(^{-1}\) (using a 1800 grooves/mm grating). The power of the laser spot was kept well below 1 mW to avoid local heating. The silicon peak at 521 cm\(^{-1}\) was used as reference for wavenumber calibration. In order to get a spatial resolution of the stacking domains, the method described by Lui et al.\(^{37}\) was used. The 2D mode is recorded every 1 µm. Then, a single Lorentzian peak is fitted to each spectrum and the FWHM is plotted. The spectra are background corrected to suppress the signal from nearby gold contacts. This procedure is further explained in the SI (see Supplementary Fig. S7).

**Infrared nano-imaging.** The infrared nano-imaging was performed using a commercial scattering-type scanning near-field microscope (s-SNOM, neaspec GmbH). Operating in intermittent contact AFM mode, topography and infrared nano-images of the graphene samples are obtained simultaneously. For infrared nano-images, an infrared CO\(_2\) laser beam with a wavelength of about 10.5 µm is focussed onto a metal-coated AFM tip (Pt/Ir, Arrow NCPT-50, Nanoworld). The tip oscillation frequency and amplitude were about 250 – 270 kHz and 50 – 80 nm, respectively. Acting as a nano-antenna, the AFM tip converts the incident infrared beam into a highly localized and enhanced electromagnetic field that is confined to its apex. This nanofocus creates a near-field interaction in the graphene underneath, whose magnitude depends on the local dielectric properties/optical conductivity of the graphene and thus is sensitive to layer number, stacking order and twist angle\(^{22,35,47,48}\). The near-field information is extracted from radiation back-scattered to a HgCdTe detector. We have also tested all-electronic Terahertz nanoscopy\(^{61}\) at 0.6 THz to map the local stacking order, but did not observe any difference between rhombohedral and Bernal stacking (see Supplementary Fig. S8).

**Computational Details.** All the atomistic calculations have been performed within density functional theory\(^{62-65}\) using the Vienna ab initio Simulation Package (VASP)\(^{66,67}\). To represent the dispersive interactions typical for trilayer graphene we use a VDW-DF2 functional\(^{68}\). The kinetic energy cutoff is set to 700 eV. The mesh in the first Brillouin zone is a fine grid of 30 x 30 x 1 k-points centered in the Gamma point as required for the hexagonal lattices. The electronic convergence of the total energy is taken to be 10\(^{-6}\) and for ionic optimization -2 x 10\(^{-4}\) eV. In addition, we use a PAW hard potential for carbon atoms, which is more suited in order to study multilayers\(^{62,69,70}\). The system of graphene multilayers is well separated from supercell images in the z-direction by a vacuum of 18 Å. The change in lattice parameters is performed keeping the cell volume so that the parameters used in calculations, such as grids and the number of plane waves, become fixed. We get the same trends and results when performing tests using other DFT methods with the same functional, such as SIESTA. We obtain slightly different results for the trilayer when testing another functional without accounting for van der Waals
forces, like LDA. In this case, especially the energy differences are larger, changing even their order of magnitude, a fact that is related to the smaller layer-layer distances well known to be obtained when using LDA. However, the functional that we chose for the calculations is better and more precise in principle, since it includes van der Waals interactions. Note that the interaction of multilayer graphene with substrates and PMMA is included by considering the effect of strain. Before testing the impact of lattice strain, we relax the atom positions and the lattice vectors. The geometry of the optimized stackings thus remains exactly hexagonal and defines the interlayer distance and the in-plane lattice vectors \( \vec{a}_1 \) and \( \vec{a}_2 \). The in-plane lattice constant for the two stackings is calculated as \( a_0 = 2.474 \, \text{Å} \), and the C-C distance is given by \( a_{CC} = 1.429 \, \text{Å} \). Since the interlayer distances between rhombohedral and Bernal stacking differ by less than 0.05 %, we use \( h = 3.554 \, \text{Å} \) for the calculations.

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Author contributions

F.R.G with the help of D.P., F.W. and J.L. fabricated the samples. F.R.G. performed the AFM and Raman measurements. F.R.G., T.D.G., Y.C.D. and F.K. performed the near-field infrared measurements. R.G., M.P. and A.A. carried out the DFT calculations. F.R.G, F.W., J.L., and R.T.W analyzed the data. R.T.W. conceived and provided advice on the project. All authors discussed the results and contributed to writing the manuscript.

Competing interests

T.D. Gokus, Y.C. Durmaz are an employee, and F. Keilmann is a cofounder of neaspec, producer of the s-SNOM microscope used in this study.

Materials & Correspondence
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Figure 1 | Crystal structure and Raman modes for trilayer graphene with Bernal (ABA) and rhombohedral (ABC) stacking order. a,b, Side and top view of the crystal structure of trilayer graphene with Bernal (a) and rhombohedral (b) stacking. Additionally, the in-plane lattice vectors \( \vec{a}_1 \) and \( \vec{a}_2 \) are shown. c,d, Raman spectra in trilayer graphene showing the 2D (c), G (d) and M mode (d inset) for Bernal (B-3LG, blue) and rhombohedral stacking (r-3LG, red). The 2D peaks are normalized. The M band is normalized to the G mode intensity and an offset is used for better visibility. An AFM image of the flake on which the spectra were acquired is shown in Fig. 3.
Figure 2 | Stacking transformation in a graphene tetralayer observed by Raman spectroscopy and infrared s-SNOM nano-imaging. a,d, AFM image of the top part of a tetralayer graphene flake before (a) and after (d) fabricating contacts. b,e, Optical image of the pristine (b) and processed (e) tetralayer, the orange and brown rectangles indicate the regions shown in (a),(d), and (c),(f), respectively. c,f, Map of the FWHM of the 2D mode in the pristine (c) and processed (f) flake. The rhombohedral and Bernal stacking domains are indicated. The green rectangle denotes the region shown in (h). g, Schematic representation of the arrangement of the stacking domains in the processed flake. h, s-SNOM amplitude image in the top part of the processed flake. Two domains of different stacking order are visible, with sharp boundary. i,j, Raman spectra recorded in the processed flake in the rhombohedral (r-4LG, red), Bernal (B-4LG, blue) and transformed region (TR, light blue) showing the 2D (i), G (j) and M mode (j, inset). The 2D peaks are normalized. The M band is normalized to the G mode intensity and an offset is used for better visibility. Scale bar in all images: 5 µm.
Figure 3 | Soliton movement in trilayer graphene. a,e, AFM image of a pristine (a) and processed (e) trilayer graphene flake. b,f, Spatial map of the width of the 2D mode recorded in the pristine (b) and processed (f) flake. The rhombohedral and Bernal stacking domains are indicated. Individual Raman spectra of the flake are shown in Fig. 1c and d. c,g, s-SNOM image showing the optical amplitude measured in the pristine (c) and processed trilayer (g). The white rectangle in (c) denotes the region shown in (d). The domains of different stacking order are marked. d, Zoomed-in image of the sharp stacking boundary. h, Scheme of the stacking domains in the processed flake. The rhombohedral, Bernal and transformed regions are indicated in red, blue and light blue, respectively. Scale bar: 5 µm in (a-c) and (e-g), 300 nm in (d).
Figure 4 | Forcing soliton movement. a,c, AFM image of a pristine (a) and processed (c) tetralayer graphene flake. b,d, s-SNOM images showing the optical amplitude measured in the pristine (b) and processed tetralayer (d). In b, domains with rhombohedral and Bernal stacking are marked. In d, two sections (part I and II) are indicated and explained in the SI. The ratio of the signal of the flake and the substrate changed compared to the ratio in the pristine flake, probably due to remaining residues of the resist. Scale bar in all images: 2 µm.
Figure 5 | Simulations of lattice deformation and suggested mechanism of transformation. a, b, c: Difference between the total energy per atom of Bernal and rhombohedral stacking under homogenous (a) and anisotropic (b, c) lattice deformation. The dashed lines are guides to the eye. Additionally, the corresponding deformations are schematically shown. d: Schematic illustration of heating and cooling the sample covered with unpatterned resist leading to homogeneous deformation. e: Proposed mechanism of how the combination of heating and local pinning during processing leads to anisotropic strain causing the preference of Bernal stacking.
Supplementary Information

Anisotropic strain induced soliton movement changes stacking order and bandstructure in graphene multilayers

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Impact of the processing steps of electron beam lithography on the stacking order

We have corroborated that no process steps prior to metal evaporation cause a rhombohedral to Bernal stacking change. This is shown in Fig. S1 for the first four processing steps for a pentalayer. Only a small decrease of the FWHM was found, indicated by the black arrow in Fig. S1c, which can be attributed to a relaxation of local strain variations during heating in agreement with previous reports for monolayer graphene. Furthermore, the electron beam exposure has as well no impact on the arrangement of stacking domains. Although it has been shown that solitons at stacking boundaries can move under electron irradiation, this happens only when using very high beam currents or comparable beam currents to the ones we use but at very high temperatures above 1000 °C.

Figure S1 | Impact of the first four processing steps on the stacking order in a graphene pentalayer. a, AFM image of a pristine graphene pentalayer. The brown rectangle indicates the region shown in (b). b, Map of the FWHM of the 2D mode in the pristine flake. The rhombohedral and Bernal domains are indicated. c, After each processing step, Raman maps were recorded and the extracted widths of the 2D mode are presented in form of histograms. From top to bottom, the histograms correspond to the Raman maps recorded in the pristine flake, after cleaning it in isopropanol/acetone, after heating it up to 180 °C, after spin coating resist onto the flake and after performing a softbake. In all histograms it is indicated to which stacking order the FWHM values correspond. Scale bar: 5 µm.
Raman measurements of the graphene tetralayer shown in Fig. 4 in the main text

Fig.s S2a and b show the spectra used for identifying the stacking order in the pristine flake. After processing, we see two parts within the flake with a small difference in the near-field amplitude (see Fig. 4d). Part II corresponds to a Bernal stacked tetralayer (see Figs. S2c and d). The 2D peak measured in part I (Fig. S2c) differs slightly in width and the G mode exhibits a lower intensity (Fig. S2d). However, the M bands (Fig. S2d, inset) have the same shape and intensity indicating that the entire flake consists of Bernal stacking. After thermally annealing the flake at 400 °C in vacuum both parts exhibit the same Raman signal (see Figs. S2e and f). Thus, the initial discrepancies could be caused by variations in local strain or doping, which are finally removed by annealing.

Figure S2 | Raman measurements of the tetralayer graphene flake shown in Fig. 4 before and after processing as well as after annealing. a,b, Raman spectra showing the 2D (a), G (b) and M mode (b, inset) for Bernal (B-4LG, blue) and rhombohedral stacking (r-4LG, red) in the pristine flake. c,d, shows the Raman spectra of the 2D (c), G (d) and M mode (d, inset) for part I (grey) and II (blue) in the processed flake. e,f, showing the 2D (e), G (f) and M mode (f, inset) for part I (grey) and II (blue) in the processed flake after annealing. All 2D peaks are normalized. The M bands are normalized to the corresponding G mode intensity and an offset is used for better visibility.
DFT of anisotropic deformation with constant unit cell area

Fig. S3 shows the difference between the total energy per atom of Bernal and rhombohedral stacking under deformation with constant unit cell area. $\delta > 0$ means stretching in x-direction and compressing in y-direction, and vice versa for $\delta < 0$:

$$\vec{a}_{1,2}' = \left( (1 + \delta) a_{1,2x}, \frac{a_{1,2y}}{1 + \delta} \right).$$

For deformations of about $-5 \% < \delta < -1.5 \%$ and $1 \% < \delta < 7.5 \%$, Bernal stacking is more stable.
Ex-situ measurements showing the stability of the transformed regions

Fig. S4a shows an AFM image of the processed tetralayer shown in Fig. 2 in the main text after cooling the sample in liquid helium. The wrinkles in between the contacts have vanished and a new fold crossing the entire sample in the middle part of the flake has appeared, however, the arrangement of the stacking domains remained the same (data not shown). Additionally, the flake was heated to 400 °C, however, no changes were observed (data not shown). Figs. S4b and d show an AFM image of the flake shown in Fig. 3 in the main text before and after cooling the sample in liquid nitrogen, respectively. The wrinkles occurring after processing (Fig. S4b) have almost vanished after cooling (see Fig. S4d). The new features appearing on the surface of the flake are dirt. Figs. S4c and e show s-SNOM amplitude images of the processed flake before and after cooling, respectively. The arrangement of the stacking domains remained the same. Thus, ex-situ cooling or heating does not affect the stacking order.

![Figure S4](image)

Figure S4 | Ex-situ measurements performed with the processed samples. a, AFM image of the processed tetralayer shown in Fig. 2 in the main text after cooling the sample in liquid helium. b,d, AFM image of the processed trilayer shown in Fig. 3 in the main text before (b) and after (d) cooling the flake in liquid nitrogen. c,e, Image of the s-SNOM amplitude measured in the processed flake before (c) and after (e) cooling the flake in liquid nitrogen. Both the rhombohedral and Bernal parts are marked. In all images, the scale bar is 5 µm.
Preventing a transition with the right choice of contact patterning

A graphene flake with 6 layers is shown before (Fig. S5a and b) and after processing (Fig. S5c – e). Here, again, the Bernal domain grows, commencing from the left, thicker part of the flake. However, the transformation stops at the big contact lead (see Fig. S5e, labeled with ‘Contact’). The contact lead hinders the soliton to move and thus prevents a transition in the right-hand region.

Figure S 5 | Multilayer graphene flake (6 layers) before and after processing. a,c, AFM image of a pristine (a) and processed (c) multilayer graphene flake. b,d, Spatial map of the width of the 2D mode recorded in the pristine (b) and processed (d) flake. The rhombohedral and Bernal stacking domains are indicated. e, Image of the s-SNOM amplitude measured in the processed flake. The domains of different stacking order are marked. Scale bar in all images: 5 µm.
Dense contact pattern to suppress soliton movement

Fig. S6a and c show an AFM image of a tetralayer graphene flake before and after processing. The corresponding s-SNOM phase images are shown in Fig. S6b and d, respectively. A high density of metal contacts was used to prevent soliton movement and indeed, the arrangement of the stacking domains remained the same.

Figure S4 | Tetralayer graphene flake before and after fabricating a dense pattern of contacts. Topography and s-SNOM phase image of a graphene tetralayer before (a,b) and after (c,d) patterning of metal contacts. The green rectangle in (c) marks the region shown in (d). In all images, the scale bar is 5 µm.
Background correction used for Raman 2D modes

Fig. S7a and b show the 2D mode for a Bernal graphene tetralayer with and without background correction, respectively. In Fig. S7c, the spectrum measured on a gold contact in the same spectral range is shown. Since the spectrum exhibits a linear behavior, a background function consisting of a straight line can be fitted. This is done by using the first and last ten data points of an uncorrected spectrum (Fig. S7a). Then, the fitted background function is subtracted (Fig. S7b). We could not observe any impact of the correction on the general shape of the 2D mode.

Figure S5 | Background correction. a, b, Uncorrected (a) and background corrected (b) 2D mode for a tetralayer graphene flake. Both spectra are normalized. c, The Raman signal measured on a gold contact in the spectral range of the 2D mode (2600 – 2800 cm⁻¹). The spectrum is normalized to the highest occurring value in (a).
Comparing THz and infrared s-SNOM nano-imaging in a multilayer graphene flake

Fig. S8a shows an optical image of a multilayer graphene on SiO₂. It consists of regions with one, three and five graphene layers, indicated by 1LG, 3LG and 5LG, respectively. The corresponding s-SNOM amplitude image taken at 10.5 µm is shown in Fig. S8b. Domains of rhombohedral and Bernal stacking are visible in the trilayer part (confirmed by Raman measurements, data not shown). Using all-electronic terahertz nanoscopy (tip oscillates with 70 kHz and 200 nm amplitude, laser wavelength of 0.6 THz, for further details see ref. 3), we have recorded an THz-amplitude image of the same region (see Fig. S8c). The multilayer flake itself is well distinguishable from the substrate, exhibiting a higher THz-amplitude. Additionally, we can see a difference between the monolayer and the parts with more layers. However, we cannot distinguish between three and five layers and also, we cannot resolve the domains of different stacking. The reason for this is the high conductivity of graphene, to be further discussed elsewhere.

Figure S8 | THz and infrared s-SNOM nanoscopy in multilayer graphene. a, Optical image of a graphene flake on SiO₂ with parts of 1, 3 and 5 layers. b, Infrared s-SNOM amplitude image. The rhombohedral and Bernal stacking domains in the trilayer part are indicated. c, THz s-SNOM amplitude image. The scale bar is 5 µm.
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