Substrate effect on the electronic properties of graphene on vicinal Pt(111)

M N Nair, A Celis, F Nicolas, S Kubsky, A Taleb-Ibrahimi, Antonio Tejeda

► To cite this version:

M N Nair, A Celis, F Nicolas, S Kubsky, A Taleb-Ibrahimi, et al.. Substrate effect on the electronic properties of graphene on vicinal Pt(111). Applied Surface Science, 2021. hal-03407576

HAL Id: hal-03407576

<https://hal.science/hal-03407576>

Submitted on 28 Oct 2021

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Substrate effect on the electronic properties of graphene on vicinal Pt(111)

M. N. Nair,^{1,2} A. Celis,^{2,3} F. Nicolas,² S. Kubsky², A. Taleb-Ibrahimi,² and A. Tejeda^{2,3}

¹ CUNY Advanced Science Research Center, 85 St. Nicholas Terrace, New York, NY 10031, USA.

² Synchrotron SOLEIL/UR1 CNRS, Saint-Aubin, 91192, Gif-sur-Yvette, France

³ Université Paris-Saclay, CNRS, Laboratoire de Physique des Solides, 91405, Orsay, France

Abstract

The atomic structure and electronic properties of monolayer graphene on a curved multi-nano-vicinal Pt (111) substrate are investigated with a low-energy electron diffraction, scanning tunneling microscopy and angle-resolved photoemission spectroscopy. Despite graphene grows continuously over the terraces and step bunching areas, the spatial periodicity also varies by varying the vicinal angle of the substrate. This superperiodicity has been evidenced by splitting of first order Pt spots in LEED and from STM. The photoemission spectroscopy unravels that the linearly dispersing π band of graphene is affected by the step periodicity as evidenced by the opening of bandgaps. The gaps opening occurs at the intersection of the main graphene band with Umklapp bands due to the superperiodicity of the one-dimensional nanostructured substrate. The energy and momentum locations of the minigaps change with the superperiodicity, which is related to the spatial periodicity and vicinal angle of the substrate. Our results show a simple way to tune the electronic properties of epitaxial graphene by tuning the substrate vicinality.

Keywords: Graphene, bandgaps, electronic properties, substrate vicinality, atomic structure

1. Introduction

Efforts to control the electronic properties of atomically thin two-dimensional materials (2D), either by twisted stacks, or by deposition on a patterned substrate, are an intense research line nowadays[1]. Tuning the electronic properties of graphene by periodic potentials has resulted in interesting properties such as Fermi velocity renormalization[2–4], extra Dirac points[5], tunable bandgap[6], minigap formation[7–10] and quantum fractal spectrum, the so called Hofstadter’s butterfly, due to the interplay between superlattice potential and magnetic field[11,12]. However, it is still challenging to couple graphene to a lithographic structure with a periodicity of few nanometers[13]. A very promising option consists of growing graphene on an appropriate periodical substrate, a natural route to induce periodic potentials in graphene. Sometimes the interaction of graphene with the substrate can modify the electronic band structure; i.e. to open a gap at the Dirac cone as in the case of graphene on Ni(111)[14], Ru(111)[15], Co(111)[16] and the initial buffer layer on SiC(0001)[17][18]. For weak interaction, minigaps can be induced in the graphene band structure, as it is the case for graphene on Ir(111)[9] and Pt(111)[19]. Pt(111) substrate is particularly interesting because it exhibits a very weak graphene-substrate interaction[20], so

the effect on the band structure as a function of the periodicity could exhibit a transition from mild coupling towards no coupling at all. Several studies on the structural and electronic properties of graphene on Ir(111) and Pt(111) have been carried out so far, which report different moiré superstructures, different rotational domains and nearly unaffected π band dispersion[7,8,19,21–25]. It has also been observed that the growth temperature can drastically affect the quality of graphene on Ir(111)[26].

The vicinal faces of single crystals allow to study the confinement effects through the electronic dispersion along two directions of interest, parallel and perpendicular to the step edges. Vicinality also offers a flexible way to control structural superperiodicities on graphene, introducing a one-dimensional potential which affects the electronic properties of graphene. Several studies have focused on graphene grown on vicinal silicon carbide substrates, although the strongly interacting graphene buffer layer beneath the monolayer graphene is the responsible there of the modification of graphene's electronic structure[13,27–30]. It is known that some growth procedures of epitaxial graphene grown on Pt(997), a (2x2) multidomain superstructure of graphene with weakly affected π bands and superperiodicity replicas[31]. In other substrates, graphene band structure can be affected by the vicinal superperiodicity. When graphene grown on Ir(332) substrate by chemical vapor deposition (CVD), a high level of ordering can be achieved, an essential factor for observing the confinement bandgap opening due to the superperiodicity [8].

In this paper, we study a monolayer graphene grown on a curved multivincinal Pt(111) substrate with nanometric-width steps, in order to compare the same growth procedure of graphene onto different surface periodicities. We analyze the detailed atomic and electronic structure by low-energy electron diffraction (LEED), scanning tunneling microscopy (STM) and angle-resolved photoemission spectroscopy (ARPES). With LEED and STM, we identify the moiré structures with different prevalent rotational domains of graphene. STM shows that graphene grows continuously on the flat (111) terraces and the step bunching areas. Moreover, vicinals with A-type steps promote more homogeneous step arrays, while the B-type vicinals give rise to wiggly edges with no long-range ordering. The effect of the periodically ordered nanostructured substrate on graphene electronic band structure has been studied by ARPES. ARPES shows the appearance of minigaps where graphene π -band crosses with the first and or the second order Umklapp bands originated by the substrate superperiodicity. The band gaps differ in their value and their energy and momentum positions with respect to the vicinality. This implies that by tuning the vicinality of the substrate, it is possible to tune the superperiodicity and thus the bandgap in its energy and momentum locations.

2. Experimental methods

Low energy electron diffraction (LEED) and room temperature scanning tunneling microscopy (STM) measurements were performed at the Surface Laboratory of SOLEIL and angle-resolved photoemission spectroscopy (ARPES) measurements were performed at CASSIOPEE beamline of SOLEIL synchrotron. The beamline spot size has 50 microns when using photon energies of 150 eV and 36 eV. ARPES measurements were performed with a photon energy of 36 eV at 77 K. Multivincinal Pt(111) crystal was purchased from Bihur Crystal. It was prepared by four-step cycles consisting of Ar⁺ sputtering at 1 keV, annealing at 970 K, Ar⁺ sputtering at 1 keV and annealing under oxygen atmosphere (1×10^{-7} mbar) for 10 min. A final flash at 1220 K ends the preparation. Graphene is grown by 10 min ethylene exposure to the substrate kept at

1070 K. This method is self-limited to one monolayer, as the growth needs the catalytic substrate to dissociate ethylene[19,38].

3. Results and discussion

3.1 Structural Characterization

Figure 1(a) shows the image of the curved Pt (111) crystal. The macroscopic normal of multivincinal Pt(111) ranges from 0° on the [111] surface up to 16° toward the $[\bar{1}\bar{1}2]$ and $[11\bar{2}]$ directions. The steps of the vicinal surfaces run perpendicular to $[\bar{1}\bar{1}2]$ or $[11\bar{2}]$ directions. A steps and B steps are referred as the vicinal steps measured at different vicinal angles with respect to [111] direction. On both steps sides, the atomic edges are arranged in different way. On the A-step terminated edges, the last atomic row of the upper terrace is aligned with the first atomic row of the lower terrace by a square disposition of edge atoms. While on the B-step terminated edges, the last atomic row of the upper terrace is shifted by half a lattice parameter parallel to the step with respect to the first atomic row of the lower terrace by a triangular disposition. The different atomic coordination at these step edges varies its reactivity.

Figure 1.(a) Image of the curved Pt crystal. The Pt(111) orientation is found at the center (vicinal angle = 0° , $x=0$ mm), and as we move further away from the center up to a tilt of 16° towards the $[\bar{1}\bar{1}2]$ or $[11\bar{2}]$ directions, the vicinal surfaces present A or B step edges respectively. The steps of the vicinal surfaces run perpendicular to $[\bar{1}\bar{1}2]$ or $[11\bar{2}]$ directions. (b-f) LEED pattern (at 128eV) shows the first order Pt spots for vicinal angles -11° , -7° , 0° , $+7^\circ$ and $+11^\circ$ respectively. The LEED splitting associated to the vicinality is sharp on the A-steps side and blurred on the B-steps side.

The $9\text{ mm} \times 9\text{ mm}$ curved Pt crystal consists of terraces of different widths, depending on the macroscopic vicinal angle, that run parallel to the $[1\bar{1}0]$ direction. The vicinal Pt(111) surface was prepared by a series of annealing and sputtering cycles. The angle of the macroscopic surface is figured out from the spatial translation made away from the sample centre. As detailed below, graphene was grown on top afterwards. The major advantage of the curved crystal is that a single graphene growth process allows to probe how it is affected by several vicinal surfaces. By studying different sites across the green dashed line shown in figure 1a, each of them corresponding to various terrace sizes, we can correlate the electronic structure

to the terrace atomic structure. We probed by LEED five different points 1mm apart each on both sides of the (111) surface. These positions present a macroscopic vicinal angle with respect to the (111) surface of -11° , -7° , 0° , $+7^\circ$, and $+11^\circ$ respectively. The LEED pattern of the clean Pt(111) surface is characterized by well-defined Pt spots and no background intensity due to carbon segregation during annealing or from residual atmosphere in UHV, as shown in fig. 1d. When moving away from the center towards the A-step region, LEED shows a sharp splitting of the Pt(111) spots along the $[11\bar{2}]$ direction (fig.1c and fig.1b). This splitting is due to the periodic nature of the vicinal surface along the $[11\bar{2}]$ direction. Taking the Pt (1×1) spots as reference, the terrace size calculated from the splitting is $L = 2.1 \pm 0.1$ nm and $L = 1.2 \pm 0.1$ nm for the -7° and -11° macroscopic vicinal angles respectively, in agreement with the expected vicinal surface. The sharp nature of the splitting spots indicates the high-quality ordering of the terraces. The terrace size decreases away from the center. When moving from the center towards the B-steps side, the LEED shows a blurred splitting of the Pt(111) spots (fig.1e and fig.1f). The blurred splitting indicates a wider distribution of terrace sizes and a lower ordering of the surface. The calculated terrace size by LEED is $L = 1.7 \pm 0.3$ nm and $L = 1.2 \pm 0.2$ nm for the $+7^\circ$ and $+11^\circ$ vicinal angles respectively, in agreement with the expected vicinal surfaces.

The Chemical Vapor Deposition (CVD) technique was chosen to grow high quality graphene on the whole surface at once. After the graphene growth, the surface was characterized by LEED and room temperature STM. Figure 2 shows the LEED patterns and the STM images of different vicinalities obtained as a function of the vicinal angle. The LEED pattern of Pt(111) surface at vicinal angle 0° (fig.2b) shows different features of graphene on Pt such as a sharp R30 domain and an arc around it. The inset shows the (1×1) spot of platinum and two graphene rotations at $\pm 5^\circ$ (R5, R5'). When graphene grows on Pt(111), it forms different rotational domains (R0, R30, R19), whose signature in diffraction techniques are well-defined spots rotated with respect to those of the substrate[19,32,33]. On the other hand, diffraction arcs also form because of indistinguishably close rotational domains. The formation of sharp dots or arcs depends on the growth parameters. Depending on the preparation procedure, a variety of moiré-like superstructures and multiple domains have been found on Pt(111)[22,23,33]. It is evident from LEED data that the intensity of these graphene orientations decreases away from the (111) surface. The corresponding STM image on vicinal angle 0° (fig.2j) shows non-ordered large terraces. The different rotations of graphene with respect to platinum give rise to several moiré patterns that can coexist as shown in fig. 2j. The two different moiré structures on atomically resolved images marked by the two unit cells (white rhombus in fig. 2j).

When exploring the A-steps side, we have evidenced that the step array is homogeneous for the small vicinal angles -2° and -3.5° (fig. 2(a, i, c, k)). Graphene grows over the step edges, as demonstrated by the continuity of the honeycomb network (white hexagons) on both sides of the steps, in agreement with previous work[19,23,33]. A clear splitting of the (1×1) Pt spots is observed on the LEED inset (fig. 2c), which corresponds to a periodicity of $L = 5.8 \pm 0.8$ nm. The STM images show a homogenous periodic structure composed of (111) terraces and step bunching areas (fig. 2(i, k)). Step bunching has also been observed for graphene grown on other metal substrates such as Ir(111)[34], Ir(332)[8][35], Ru(0001)[36] and SiC substrate[37]. The analysis of STM images perpendicularly to the steps allows to determine the surface periodicity L (from the terrace and the step bunching), indicating a period of $L = 6.8 \pm 1.8$ nm.

For the vicinal angle of -7° , we find the best ordered surfaces (fig. 2(e, m)). We observe replicas of graphene arcs, resulting from the high ordered periodic structure. The period here is $L = 5.8 \pm 0.5$ nm as calculated from LEED and STM, showing narrower terraces as the result of a wider step bunching which is clearly visible. The calculated period from STM is

$L = 5.8 \pm 0.5 \text{ nm}$. Here, graphene is also continuous, as suggested by the honeycomb on the terraces (blue region) at both sides of the step bunching areas (orange region) (fig. 2m).

Figure. 2. LEED (left) and details of the STM images (right) for vicinal angles for -2° (a, i), -3.5° (c, k), -7° (e, m), -11° (g, o), 0° (b, j), $+3.5^\circ$ (d, l), $+7^\circ$ (f, n), $+11^\circ$ (h, p). Tunnelling conditions: -2° [70 mV, 2.6 nA], -3.5° [70 mV, 2.6 nA], -7° [100 mV, 2.8 nA], -11° [1.14 V, 0.7 nA], 0° [360 mV, 2.6 nA], $+3.5^\circ$ [1 V, 0.8 nA], $+7^\circ$ [1 V, 0.5 nA] and $+11^\circ$ [1 V, 0.8 nA]. Three main graphene features appear: R30, equivalent R5/R5' rotations and multidomain arcs. The A-steps side (a,c,e,g) presents a gradual splitting on the Pt spot, indicating the formation of a well ordered periodic system, whose period L (terraces + step bunching) varies from 6.8 to 5.7 ± 1.4 nm obtained from STM (i,k,m,o). The B-steps side (b,d,f,h) does not show the Pt spot splitting in LEED, although STM images confirm that there is a certain degree of order (j,l,n,p). Atomic resolution images suggest the continuity of graphene, as the graphene honeycomb on the blue terraces is oriented in the same way at both sides of the orange step bunching area. The white rhombus highlights the unit cells of moirés.

Finally, for -11° , the highest macroscopic vicinal angle that we studied, the LEED spot splitting of Pt is similar to that observed for -7° , so it corresponds to $L = 4.8 \pm 1.5$ nm (fig 2g). On the other hand, STM indicates a periodicity of $L = 5.7 \pm 1.8$ nm. The large error bars are due to a less homogeneous surface, in agreement with the broad Pt splitting spot in LEED. On the contrary, in the B-step side, LEED patterns of vicinal angle $+2^\circ$ (not shown) and $+3.5^\circ$ (fig. 2d) are very similar to that of the flat surface. STM images show the apparition of a preferential orientation for steps and the development of step bunching areas (orange on the atomic resolution image in (fig.2l)), leading to narrow terraces. The periodicity from STM is $L = 6.6 \pm 2.7$ nm. Such large error bar is due to the significant variation in terrace width ranging between 4.6 and 6.6 nm. Such an observation explains why the Pt spot splitting is not observed in LEED. Anyway, graphene grows in a continuous way over these terraces and graphene rotational domains (R5 and R14) are preserved across the steps, as highlighted by the continuity of the moiré unit cell on both domains (fig. 2l).

For higher vicinal angles on the B-type step, i.e., for $+7^\circ$ and $+11^\circ$ (fig. 2(f, n and h, p)), platinum spots in the LEED pattern lose intensity and the splitting is less defined, indicating the weaker ordering of the step array, as shown also in STM images. STM shows a broad dispersion in widths, ranging from 2 to 15 nm for $+7^\circ$ and from 2.5 nm to 13 nm for $+11^\circ$. These areas will be discarded from the electronic structure analysis due to the structural inhomogeneity. The growth of graphene on A and B steps is possibly different because already the underlying substrate is different for each type of steps. We speculate the reason is that the annealing temperature of the crystal is optimal for A-type steps but not for B-type, inducing here more disordered steps that will impact in graphene growth order.

In conclusion, we observe that graphene grows more ordered in vicinal surfaces on the A-step side, as seen by LEED on the splitting of the Pt spot and directly by STM, that shows ordered step arrays. The quality of the step periodicity decreases for higher macroscopic miscut angles, namely for vicinal angles $+11^\circ$ and -11° . The new periodicity is different from the initial Pt periodicity before graphene growth. Graphene grows continuously across the sample between the terraces and step bunching areas, so a modification of the electronic properties could be expected, therefore we analyze the band structure of graphene by ARPES.

3.2 Substrate effect on graphene electronic properties

We performed detailed ARPES measurements at three different vicinal angles where well-ordered steps are developed. Figure 3a-c shows the ARPES intensity maps as a function of the vicinal angle obtained through graphene K point along a direction perpendicular to the steps. The first striking feature is that the linear π band dispersion of graphene is discontinuous, as the intensity decreases at certain energies and its $E(k)$ location depends on the vicinal surface. On fig. 3(d-f) there are guides-to-the-eye lines to better understand the electronic structure. Two different Dirac cones are observed, corresponding to R5 and R5' rotational domains (green dotted lines). Depending on the vicinality or eventually on varying surface doping, the binding energy of the Dirac point shifts from 320 meV to 250 meV, as in Pt(111)[19]. In addition to the graphene cones of R5 and R5' domains, a spectral intensity is appreciated around 2.5 eV. This corresponds to the saddle point of the π band from R30 rotational domain (cyan line). Close to the Fermi level, there is a Pt(111) bulk band indicated by a black line[19]. We now focus on the discontinuities of the graphene π band structure at the locations with no interference with other spectral features, i.e., on the 2nd gap on fig. 3(d-f) shown with black arrows. A detail of this region is shown in the Energy Distribution Curves (EDC) in fig. 3(g-i). The black lines in EDC curves follow the maxima of the bands, showing the

folding of the bands and consequently, band gap openings of 360 meV (fig. 3g), 290 meV (fig. 3h), and 200 meV(fig. 3i) respectively.

Figure 3. E(k) maps of graphene for the vicinal angles (a) -5.5° , (b) -2° and (c) $+2^\circ$. The same data with indications of the different spectral features are shown for vicinal angles (d) -5.5° , (e) -2° and (f) $+2^\circ$. The dotted green lines show the π bands from R5/R5' rotational domains with a different p-doping at every vicinal angle. The band at 2.5 eV (cyan line) is the saddle point of the π band from the R30 domain. The curved black line indicates Pt(111) bulk band. The 1st, 2nd and 3rd gap feature the three gaps due to the crossing between the main band and the Umklapps. However, the 1st and 3rd gap features are not clearly visible due to the high spectral intensity from other bands. The back arrow indicates the location of 2nd gap (g)-(i) EDC curves showing the gap opening of the second band gap for -5.5° , -2° and $+2^\circ$ respectively.

Figure 4. (a-d) show the 3D representation of STM topographic images of various vicinal angles (e) shows the average facet width versus vicinal angle obtained by various experimental techniques (LEED, STM and ARPES) (f) Scheme of crossing of graphene π band with Umklapps bands due to the superperiodicity induced by the step arrays, causing gap openings.

Figure 4(a-d) show the 3D representation of STM topographic images of various vicinal angles where a homogenous periodic structure composed of (111) terraces and step bunching areas can be appreciated (e) the average facet width versus the vicinal angle obtained by three different experimental techniques LEED, STM and ARPES. The average facet width decreases when increasing the vicinal angle away from the (111) surface. Figure 4f shows the scheme of Umklapps bands due to the superperiodicity induced by the step arrays. The origin of the ARPES observed band gap is illustrated with a scheme in fig. 4b. Due to the superperiodicity introduced by the nanostructured substrate, a first order Umklapp band (or replica band) appears shifted by $G = \frac{2\pi}{d}$, where d is the spatial periodicity of the system. The same process occurs for the second and third order Umklapp bands that are shifted by $2G$ and $3G$ respectively. The band gaps are originated at the crossing between the main graphene band and the Umklapp band. The 2nd bandgap corresponds to the crossing between the 2nd order Umklapp and the main graphene band. Other gaps are expected for crossings with the first and third order Umklapp, as indicated in fig.

3d-f. They are however less visible due to the proximity to platinum bulk bands or to the R30 graphene band. Therefore, we focus more attention to the 2nd bandgap. The E and k location of this gap changes with the vicinality (as shown with white dotted lines in fig.3). If we trace a line through the band gap energy, there is a shift from one surface to the other. There is also a shift in the k position where the band gap appears, meaning that the G vector depends on the vicinal angle. Indeed, the periodicities associated to the G vector observed in ARPES are 3.8 ± 0.9 nm (-5.5°), 3.9 ± 0.9 nm (-2°) and 4.5 ± 0.9 nm ($+2^\circ$). These values correspond roughly to half of the periodicity found by LEED and STM. In nutshell, by tuning the vicinal angles of the substrate, we can tune the spatial periodicity and thus modify the electronic properties of graphene.

4. Conclusions

In conclusion, we have modified the electronic properties of graphene by growing it on a nanostructured vicinal Pt(111) substrate. We have explored the effect of both A-steps and B-steps at vicinal angles between -11° to $+11^\circ$ with respect to the [111] surface normal. The side with A-steps results in more homogeneous step arrays, while most of the side with B-steps presents wiggly edges, giving rise to non-homogeneous width steps. In our growth conditions, for coverages close to the monolayer, we have grown a continuous layer of graphene. Graphene grows more ordered in the vicinal surfaces on the A-step side as evidenced by LEED and STM. The average facet width and the quality of the step array decrease when far away from the [111] direction.

Moreover, the effects of the structured substrate on the graphene electronic band structure have been studied by ARPES. We observed that the band gaps differing in their amplitude and their energy and momentum positions which are correlated with the vicinal angles of the substrate. The origin of the gaps comes from the superperiodicity induced by the step array. Finally, we show that how it is possible to control the superperiodicity and thus the bandgap by tuning the vicinality of the substrate, a simple way to engineer the electronic properties of graphene or other 2D materials.

Declaration of competing Interest

None

Acknowledgements

We acknowledge the financial support of the Agence Nationale de la Recherche (France) under contract CoRi-Graph and the support from F. Bertran and P. Le Fèvre and all other CASSIOPEE staffs during ARPES beam times.

References

- [1] A.K. Geim, I. V Grigorieva, Van der Waals heterostructures., *Nature*. 499 (2013) 419–25. <https://doi.org/10.1038/nature12385>.
- [2] C.H. Park, L. Yang, Y.W. Son, M.L. Cohen, S.G. Louie, Anisotropic behaviours of massless Dirac fermions in graphene under periodic potentials, *Nat. Phys.* 4 (2008) 213–217. <https://doi.org/10.1038/nphys890>.
- [3] M. Barbier, F.M. Peeters, P. Vasilopoulos, J.M. Pereira, Dirac and Klein-Gordon particles in one-dimensional periodic potentials, *Phys. Rev. B*. 77 (2008) 115446. <https://doi.org/10.1103/PhysRevB.77.115446>.
- [4] J.H. Ho, Y.H. Chiu, S.J. Tsai, M.F. Lin, Semimetallic graphene in a modulated electric potential, *Phys. Rev. B*. 79 (2009) 115427. <https://doi.org/10.1103/PhysRevB.79.115427>.

- [5] C.-H. Park, L. Yang, Y.-W. Son, M.L. Cohen, S.G. Louie, New Generation of Massless Dirac Fermions in Graphene under External Periodic Potentials, *Phys. Rev. Lett.* 101 (2008) 126804. <https://doi.org/10.1103/PhysRevLett.101.126804>.
- [6] R.P. Tiwari, D. Stroud, Tunable band gap in graphene with a noncentrosymmetric superlattice potential, *Phys. Rev. B.* 79 (2009) 205435. <https://doi.org/10.1103/PhysRevB.79.205435>.
- [7] A. Celis, M.N. Nair, M. Sicot, F. Nicolas, S. Kubsky, D. Malterre, A. Tejeda, Superlattice-induced minigaps in graphene band structure due to underlying one-dimensional nanostructuration, *Phys. Rev. B.* 195410 (2018) 1–6. <https://doi.org/10.1103/PhysRevB.97.195410>.
- [8] A. Celis, M.N. Nair, M. Sicot, F. Nicolas, S. Kubsky, A. Taleb-Ibrahimi, D. Malterre, A. Tejeda, Growth, morphology and electronic properties of epitaxial graphene on vicinal Ir(332) surface, *Nanotechnology.* 31 (2020) 285601. <https://doi.org/10.1088/1361-6528/ab866a>.
- [9] I. Pletikoscic, M. Kralj, P. Pervan, R. Brako, J. Coraux, A.T. N'Diaye, C. Busse, T. Michely, Dirac Cones and Minigaps for Graphene on Ir(111), *Phys. Rev. Lett.* 102 (2009) 056808. <https://doi.org/10.1103/PhysRevLett.102.056808>.
- [10] E. Starodub, A. Bostwick, L. Moreschini, S. Nie, F. El Gabaly, K.F. McCarty, E. Rotenberg, In-plane orientation effects on the electronic structure, stability, and Raman scattering of monolayer graphene on Ir(111), *Phys. Rev. B.* 83 (2011) 125428. <https://doi.org/10.1103/PhysRevB.83.125428>.
- [11] C.R. Dean, L. Wang, P. Maher, C. Forsythe, F. Ghahari, Y. Gao, J. Katoch, M. Ishigami, P. Moon, M. Koshino, T. Taniguchi, K. Watanabe, K.L. Shepard, J. Hone, P. Kim, Hofstadter's butterfly and the fractal quantum Hall effect in moiré superlattices, *Nature.* 497 (2013) 598–602. <https://doi.org/10.1038/nature12186>.
- [12] L.A. Ponomarenko, R. V. Gorbachev, G.L. Yu, D.C. Elias, R. Jalil, A.A. Patel, A. Mishchenko, A.S. Mayorov, C.R. Woods, J.R. Wallbank, M. Mucha-Kruczynski, B.A. Piot, M. Potemski, I. V. Grigorieva, K.S. Novoselov, F. Guinea, V.I. Fal'Ko, A.K. Geim, Cloning of Dirac fermions in graphene superlattices, *Nature.* 497 (2013) 594–597. <https://doi.org/10.1038/nature12187>.
- [13] A. Celis, M.N. Nair, A. Taleb-Ibrahimi, E.H. Conrad, C. Berger, W.A. de Heer, A. Tejeda, Graphene nanoribbons: fabrication, properties and devices, *J. Phys. D. Appl. Phys.* 49 (2016) 143001. <https://doi.org/10.1088/0022-3727/49/14/143001>.
- [14] A. Varykhalov, J. Sánchez-Barriga, A.M. Shikin, C. Biswas, E. Vescovo, A. Rybkin, D. Marchenko, O. Rader, Electronic and Magnetic Properties of Quasifreestanding Graphene on Ni, *Phys. Rev. Lett.* 101 (2008) 157601. <https://doi.org/10.1103/PhysRevLett.101.157601>.
- [15] P. Sutter, M.S. Hybertsen, J.T. Sadowski, E. Sutter, Electronic Structure of Few-Layer Epitaxial Graphene on Ru(0001), *Nano Lett.* 9 (2009) 2654–2660. <https://doi.org/10.1021/nl901040v>.
- [16] A. Varykhalov, O. Rader, Graphene grown on Co(0001) films and islands: Electronic structure and its precise magnetization dependence, *Phys. Rev. B.* 80 (2009) 035437. <https://doi.org/10.1103/PhysRevB.80.035437>.
- [17] M. N. Nair, I. Palacio, A. Celis, A. Zobelli, A. Gloter, S. Kubsky, J.-P. Turmaud, M. Conrad, C. Berger, W. de Heer, E.H. Conrad, A. Taleb-Ibrahimi, A. Tejeda, Band Gap Opening Induced by the Structural Periodicity in Epitaxial Graphene Buffer Layer, *Nano Lett.* 17 (2017) 2681–2689. <https://doi.org/10.1021/acs.nanolett.7b00509>.
- [18] M.S. Nevius, M. Conrad, F. Wang, A. Celis, M.N. Nair, A. Taleb-Ibrahimi, A. Tejeda, E.H. Conrad, Semiconducting Graphene from Highly Ordered Substrate Interactions, *Phys. Rev. Lett.* 115 (2015) 136802. <https://doi.org/10.1103/PhysRevLett.115.136802>.
- [19] P. Sutter, J.T. Sadowski, E. Sutter, Graphene on Pt(111): Growth and substrate interaction, *Phys. Rev. B.* 80 (2009) 245411. <https://doi.org/10.1103/PhysRevB.80.245411>.
- [20] J. Wintterlin, M.L. Bocquet, Graphene on metal surfaces, *Surf. Sci.* 603 (2009) 1841–1852. <https://doi.org/10.1016/j.susc.2008.08.037>.
- [21] I. Šrut, V. Mikšić Trontl, P. Pervan, M. Kralj, Temperature dependence of graphene growth on a stepped iridium surface, *Carbon N. Y.* 56 (2013) 193–200. <https://doi.org/10.1016/j.carbon.2013.01.026>.
- [22] A.B. Preobrajenski, M.L. Ng, A.S. Vinogradov, N. Mårtensson, Controlling graphene corrugation on lattice-mismatched

- substrates, *Phys. Rev. B.* 78 (2008) 073401. <https://doi.org/10.1103/PhysRevB.78.073401>.
- [23] M. Gao, Y. Pan, L. Huang, H. Hu, L.Z. Zhang, H.M. Guo, S.X. Du, H.-J. Gao, Epitaxial growth and structural property of graphene on Pt(111), *Appl. Phys. Lett.* 98 (2011) 033101. <https://doi.org/10.1063/1.3543624>.
- [24] S. Rajasekaran, S. Kaya, T. Anniyev, H. Ogasawara, A. Nilsson, Probing substrate effects in the carbon-projected band structure of graphene on Pt(111) through resonant inelastic x-ray scattering, *Phys. Rev. B.* 85 (2012) 045419. <https://doi.org/10.1103/PhysRevB.85.045419>.
- [25] P.A. Khomyakov, G. Giovannetti, P.C. Rusu, G. Brocks, J. van den Brink, P.J. Kelly, First-principles study of the interaction and charge transfer between graphene and metals, *Phys. Rev. B.* 79 (2009) 195425. <https://doi.org/10.1103/PhysRevB.79.195425>.
- [26] E. Loginova, N.C. Bartelt, P.J. Feibelman, K.F. McCarty, Factors influencing graphene growth on metal surfaces, *New J. Phys.* 11 (2009) 063046. <https://doi.org/10.1088/1367-2630/11/6/063046>.
- [27] J. Hicks, A. Tejada, A. Taleb-Ibrahimi, M.S. Nevius, F. Wang, K. Shepperd, J. Palmer, F. Bertran, P. Le Fèvre, J. Kunc, W. a. de Heer, C. Berger, E.H. Conrad, A wide-bandgap metal–semiconductor–metal nanostructure made entirely from graphene, *Nat. Phys.* 9 (2013) 49–54. <https://doi.org/10.1038/nphys2487>.
- [28] I. Palacio, A. Celis, M.N. Nair, A. Gloter, A. Zobelli, M. Sicot, D. Malterre, M.S. Nevius, W.A. de Heer, C. Berger, E.H. Conrad, A. Taleb-Ibrahimi, A. Tejada, Atomic Structure of Epitaxial Graphene Sidewall Nanoribbons: Flat Graphene, Miniribbons, and the Confinement Gap, *Nano Lett.* 15 (2015) 182–189. <https://doi.org/10.1021/nl503352v>.
- [29] K. Nakatsuji, T. Yoshimura, F. Komori, K. Morita, S. Tanaka, Uniaxial deformation of graphene Dirac cone on a vicinal SiC substrate, *Phys. Rev. B.* 85 (2012) 195416. <https://doi.org/10.1103/PhysRevB.85.195416>.
- [30] K.-K. Bai, Y. Zhou, H. Zheng, L. Meng, H. Peng, Z. Liu, J.-C. Nie, L. He, Creating One-Dimensional Nanoscale Periodic Ripples in a Continuous Mosaic Graphene Monolayer, *Phys. Rev. Lett.* 113 (2014) 086102. <https://doi.org/10.1103/PhysRevLett.113.086102>.
- [31] M. Pissarra, D. Pacilé, P. Moras, P.M. Sheverdyayeva, A. Sindona, M. Papagno, C. Carbone, Electronic structure of epitaxial graphene grown on stepped Pt(997), *Phys. Rev. B.* 89 (2014) 1–6. <https://doi.org/10.1103/PhysRevB.89.195438>.
- [32] M. Gao, Y. Pan, L. Huang, H. Hu, L.Z. Zhang, H.M. Guo, S.X. Du, H.J. Gao, Epitaxial growth and structural property of graphene on Pt(111), *Appl. Phys. Lett.* (2011). <https://doi.org/10.1063/1.3543624>.
- [33] H. Ueta, M. Saida, C. Nakai, Y. Yamada, M. Sasaki, S. Yamamoto, Highly oriented monolayer graphite formation on Pt(111) by a supersonic methane beam, *Surf. Sci.* 560 (2004) 183–190. <https://doi.org/10.1016/j.susc.2004.04.039>.
- [34] J. Coraux, A.T. N'Diaye, C. Busse, T. Michely, Structural coherency of graphene on Ir(111), *Nano Lett.* 8 (2008) 565–570. <https://doi.org/10.1021/nl0728874>.
- [35] I. Šrut Rakić, M. Kralj, W. Jolie, P. Lazić, W. Sun, J. Avila, M.-C. Asensio, F. Craes, V. Mikšić Trontl, C. Busse, P. Pervan, Step-induced faceting and related electronic effects for graphene on Ir(332), *Carbon N. Y.* 110 (2016) 267–277. <https://doi.org/10.1016/j.carbon.2016.09.024>.
- [36] S. Günther, S. Dänhardt, B. Wang, M.-L. Bocquet, S. Schmitt, J. Wintterlin, Single Terrace Growth of Graphene on a Metal Surface, *Nano Lett.* 11 (2011) 1895–1900. <https://doi.org/10.1021/nl103947x>.
- [37] M. Hupalo, E.H. Conrad, M.C. Tringides, Growth mechanism for epitaxial graphene on vicinal 6H-SiC(0001) surfaces: A scanning tunneling microscopy study, *Phys. Rev. B.* 80 (2009) 041401. <https://doi.org/10.1103/PhysRevB.80.041401>.
- [38] T.A. Land, T. Michely, R.J. Behm, J.C. Hemminger, G. Comsa, STM investigation of single layer graphite structures produced on Pt(111) by hydrocarbon decomposition, *Surf. Sci.* 264 (1992) 261–270. [https://doi.org/10.1016/0039-6028\(92\)90183-7](https://doi.org/10.1016/0039-6028(92)90183-7).