SORBONNE UNIVERSITÉ

CHINA SCHOLARSHIP COUNCIL

Appel à projets
Campagne 2022
https://www.sorbonne-universite.fr

Title of the research project :

Monte-Carlo simulations of the growth of 2D materials by epitaxy

Thesis supervisor (HDR) :

Name : AQUA Surname : Jean-Noël

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(site, dresse, bulding, office…) Jussieu, 4 place Jussieu, 22-12-415

Research Unit

Name : INSP

Code *(ex. UMR xxxx)* :

UMR 7588

Doctorate School

Thesis supervisor's doctorate school (candidate's futur doctoral school) : ED 397

PhD student currently supervised by the thesis supervisor (number, year of the first inscription) :

none

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Description of the research project (*ENGLISH*)*:*

Ce texte sera diffusé en ligne : il ne doit pas excéder 3 pages et est écrit en interligne simple.
Ce texte est à l'adresse d'étudiantes et étudiants chinois, il doit donc être rédigé en anglais.

Détailler le contexte, l'objectif scientifique, la justification de l'approche scientifique ainsi que l'adéquation à l'initiative/l'Institut.

Le cas échéant, préciser le rôle de chaque encadrant ainsi que les compétences scientifiques apportées. Indiquer les publications/productions des encadrants en lien avec le projet.
Préciser le profil d'étudiant(e) recherché.

Merci d'enregistrer votre fichier au format PDF et de le nommer :
«CSC_22_Projet NOM Porteur.euse projet »

Monte-Carlo simulations of the growth of 2D materials by epitaxy

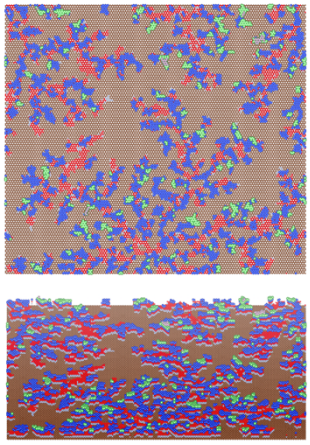

Figure 1: Kinetic Monte-Carlo simulations of 2D silicon on graphene

The recent discovery of two-dimensional materials (2DMs) has revolutionized solid-state physics thanks to their ability to confine carriers. It opened up novel potential applications related to their outstanding electronic and optical properties. Since the discovery in 2004 of graphene thanks to exfoliation, these materials have aroused an ever-growing interest since different materials form 2D layers: carbon, silicon, transition metal dichalcogenides[1]. Nowadays, 2DMs are expected to potentially surpass all previous technologies and their integration in devices has attracted great interest. Graphene has shown remarkable properties, such as the Dirac-cone-shaped energy band and high carrier mobility[2]. However, despite significant efforts, there has been no reproducible method to open up its bandgap while preserving high carrier mobility. 2DMs based on group IV elements such as Si (silicene) and Ge (germanene) are promising alternatives. They are predicted to be Dirac materials in which electrons behave as relativistic massless particles. More importantly, they are better suited than Gr in the race for ultimate thickness scaling of nanoelectronic devices. In addition, the larger buckling, as well as the resulting larger spin-orbit coupling, make it easier to create a bandgap in Si and Ge 2D crystals without degrading electronic Dirac properties. Manufacturing good quality, quality-controlled 2DMs of different elements is of major importance from both fundamental and applied perspectives. A huge experimental effort has been dedicated to the growth of such 2DMs. Yet, no reliable, high-quality and large-scale method is available. The goal of this numerical study is to put forward a new understanding of this growth to eventually guide experiments.

Obtaining 2DMs by the common and classical exfoliation technique has intrinsic limitations, notably on the size and quality of the 2D crystals. Moreover, it is not possible for silicene or germanene that do not exist in nature as allotropes of Si or Ge. The only remaining alternative is their production by molecular beam epitaxy that is a technological lock for any potential industrial application. The synthesis of silicene and germanene by epitaxy has been first reported on metallic substrates[3]. However, their properties were found to be strongly affected by the coupling with their substrate and mixing effects[4]. In addition, fabrication of functional electronic devices requires non-metallic supports. One alternative is to introduce a buffer layer for decoupling the 2DMs from the substrate[5]. Recent experiments concerning the epitaxial growth of Si on highly oriented pyrolytic graphite (HOPG) revealed the possibility to grow either 2D flakes with rather small sizes, or dewetted fractal islands. In all cases, the understanding and control of the epitaxial growth of 2D materials is largely insufficient. Today's progress is therefore limited by the lack of control of dynamical mechanisms.

This PhD study aims at developping a new numerical analysis based on kinetic Monte-Carlo (kMC) simulations. This analysis will be done in constant interaction with experimental groups in order to validate simulation schemes. Only a few theoretical works have been devoted to the study of the

[1] C. R. Dean et al., Nature Nanotechnol., 5 (2010) 722 ; A. K. Geim, I. V. Grigorieva, Nature, 499 (2013) 419
[2] K. S. Novoselov, A. K. Geim, et al., Science 306 (2004) 666
[3] B. Lalmi, H. Oughaddou et al., Appl. Phys. Lett. 97 (2010) 223109
[4] R. Bernard et al., Phys. Rev. B 88 (2012) 121411
[5] F. d'Acapito et al. J. Phys. Chem. Lett. 7 (2016) 11163

epitaxial growth of 2DMs [6], and even less on silicene or germanene. First principles-calculations have revealed the stability of some structures, but the dynamical description of their growth is sparse due to major difficulties. The challenge is to simulate out-of-equilibrium systems of sufficient size (typically of the order of a hundred nanometers) yet incorporating atomic details, over sufficiently long times (typically of the order of hours) yet describing atomic events (diffusion, incorporation, exchange…). Indeed, the use of silicene in devices requires a control of the good quality of the crystals on such large areas. Moreover, typical growth times in molecular beam epitaxy are precisely of the order of minutes in order to obtain systems with epitaxial quality. These space and time scales are by far unreachable with ab-initio or molecular dynamics tools. Kinetic Monte-Carlo simulations allow to meet all these challenges and are therefore a tool of choice for the theoretical description of these systems. The challenge of this project is to derive a coherent and realistic framework for kinetic Monte Carlo simulations describing the epitaxial growth of 2DMs such silicene and germanene on different substrates (HOPG, Gr, Ag…).

The host group has a vast experience in developing Monte Carlo simulations for epitaxial growth. The initial part of the work will be spent on developing an algorithm relevant for the systems of interest. It will be implemented for the description of the epitaxy of silicene or germanene on different substrates such as HOPG, Gr or Ag. This choice of substrates is both dictated by (i) the fact that experimental results are available to guide the modelization work and to test its outcome, (ii) that they are promising candidates for the realization of good quality and large scale 2D crystals and (iii) ongoing collaborations with experimental groups (G. Prévot's group at INSP and I. Berbezier's group at IM2NP).

Objective 1: KMC simulations of silicene and germanene growth

The numerical part will include the derivation of a rejection-free kinetic Monte-Carlo following the Bortz-Kalos-Lebowitz algorithm that is suitable for simulations where transition rates are all known initially. We will study an on-lattice solid-on-solid (SOS) model that describes a crystal without voids and that allow to simplify the description of atomistic vibrations. This model will include different atomic events starting with the fundamental deposition, diffusion and attachment/detachment processes. These processes can be made dependent on different configurations (local height, local configuration …) to account for different effects (segregation, alloying, wetting etc). The inputs to the kMC simulations will be the rates of various fundamental processes that will be obtained either from experiments outcomes or from ab-initio calculations. We will generalize these models on lattices with relevant anisotropies, essentially face-centred cubic substrates and honeycomb films. In addition, we want to investigate the influence of different kind of film/substrate interactions that may be found experimentally. We will start with the weak van der Waals interactions that allow the epitaxial growth of 2D materials with the appropriate Dirac properties. We will also allow for the description of long-range elastic interaction that may occur in lattice-mismatched coherent conditions, but also when buckling occurs (as was theoretically predicted for silicene and experimentally revealed). Accounting for elasticity is a challenge due to its long range but the supervisor has a long-standing experience in its description (using Green functions or Frenkel-Kontorova models). Moreover, and especially as concerns the deposition on metallic substrates such as Ag(111), it is necessary to describe alloying effects that where revealed in some conditions. To this end, we will include a two species model with extra possible atomic processes, and especially intermixing. By investigating a variety of systems and substrates, the results will not only be relevant for different experimental groups, but will help identifying the universal mechanisms and the most relevant growth parameters. In addition, the theoretical framework will be a suitable starting point for further theoretical investigations, that can describe the growth dynamics in a mean-field way.

[6] H. Jiang, Z. Hou, J. Chem. Phys. 143 (2015) 084109 ; Y. Nie et al., Sci. Rep. 7 (2017) 2977

Objective 2: Theory/experiment comparison

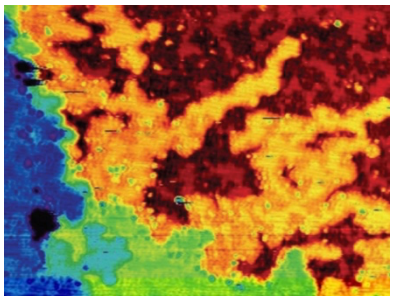

Figure 2: Growth of Si on Ag(111) done at INSP

The modelization part of the project will be dedicated to the connection between theory and experiments. The work will be done in a back-and-forth approach, where the models will include first the main mechanisms revealed in the experimental literature and by the collaborating groups. We will dedicate our studies to the case of Silicene and Germanene epitaxy on the three substrates, the most promising HOPG and Gr, but also on the extensively studied Ag(111). Kinetic energy barriers are crucial parameters in kinetic Monte-Carlo simulations, and their absolute values may trigger different regimes (e.g. with fractals or compact growth shapes as a function of the nearest-neighbour interactions). Hence, we will consider first typical values for these barriers, already derived in some experimental studies (e.g. 2D flakes densities, shapes, amount of alloying etc) and first-principles calculations (e.g. considering energy barriers in energy landscapes). We will then systematically analyse the outcomes of the simulations depending on these barriers and will compare with experimental results in order to derive typical values and bounds. The aim of this coupling between theory and experiment is to gain control on the growth procedure in order to obtain two-dimensional epitaxial deposits of large size and good crystalline quality.

The project is well embedded with the host institute research focus. It concerns the growth of nanomaterials that is precisely a focus at INSP. Moreover, the hosting team is currently developing experiments on the epitaxial growth of germanene on metallic substrates. Hence, the simulation approach here will be directly compared with the experimental outcomes. It will be of first importance both for the theoretical approach and for guiding the experimental procedures. The project thence belongs to the scientific priorities of the host.

The typical desired student should have a basic knowledge of condensed matter and statistical physics, together with basic abilities for scientific computing.

Publications of the supervisor related to the project:

- Growth and Self-organization of Si-Ge nanostructures, J.-N. Aqua, I. Berbezier, L. Favre, T. Frisch, A. Ronda, *Physcis Reports* **522** (2013) 59
- Kinetic Monte Carlo simulations of the growth of silicon germanium pyramids, P. Gaillard, J.-N. Aqua, T. Frisch, *Physical Review B* **87** (2013) 125310
- Elastic interactions and kinetics during reversible submonolayer growth: Monte Carlo simulation, J.-N. Aqua, T. Frisch, *Physical Review B* **78** (2008) 121305(R)
- New strategies for producing defect free SiGe strained nanolayers, T. David, J.-N. Aqua et al, *Scientific Report* **8** (2018) 2891

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CALENDRIER DE LA CAMPAGNE

26 juillet

Lancement de la campagne
Diffusion de l'appel à projets par les écoles doctorales auprès de leurs encadrantes et encadrants.

Jusqu'au 17 septembre

Les chercheurs/enseignants-chercheurs et chercheuses/enseignantes-chercheuses de Sorbonne Université soumettent des propositions de projets de recherche doctoraux à leur directeur et directrice d'école doctorale (en utilisant le formulaire joint) et à l'adresse suivante : **https://inscriptions.sorbonne-universite.fr/lime25/index.php/344242?lang=fr**

Jusqu'au 24 septembre

Les écoles doctorales valident le cas échéant les projets et notifie le collège doctoral de leur décision à l'adresse suivante : **csc-su@listes.upmc.fr**

1er octobre

Mise en ligne des projets validés sur le site web de Sorbonne Université et ouverture des candidatures
https://www.sorbonne-universite.fr
Les candidats chinois prennent contact avec les porteurs et porteuses de projets et leur envoient un dossier de candidature.
Les candidates et les candidats déposent leur dossier à l'adresse suivante :
https://inscriptions.sorbonne-universite.fr/lime25/index.php/383154?newtest=Y&lang=fr

31 janvier

Fermeture des candidatures
Les porteurs et porteuses de projet ont transmis la candidature retenue après audition des candidates et candidats à leur école doctorale

Jusqu'au 21 février

Après examen, les écoles doctorales envoient les lettres de pré-admission signées et tamponnées des candidats et candidates retenues au collège doctoral

28 février

Le collège doctoral envoie les lettres de pré-admission aux candidates et candidats

Jusqu'au 31 mars

Les candidates et candidats retenus par Sorbonne Université candidate sur le site internet du CSC

31 mai

Diffusion des résultats par le CSC auprès de SU
Envoi résultats aux candidates et candidats ainsi qu'aux porteurs et porteuses de projet

SORBONNE UNIVERSITÉ

Joint supervisor :

Name : Surname :

Title :

email :

Professional adress :
(site, dresse, bulding, office…)

Research Unit

Name :

Code *(ex. UMR xxxx)* :

École doctorale

Joint supervisor's doctorate school : Or, if non SU :

PhD student currently supervised by the joint supervisor (number, year of the first inscription) :

Joint supervisor :

Name : Surname :

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