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Editorial

Special Issue "Fundamentals and Recent Advances in Epitaxial Graphene on SiC"

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Abstract: The aim of this Special Issue is to provide a scientific platform for recognized experts in the field of epitaxial graphene on SiC to present their recent studies towards a deeper comprehension of growth mechanisms, property engineering and device processing. This Special Issue gives readers the possibility to gain new insights into the nature of buffer layer formation, control of electronic properties of graphene and usage of epitaxial graphene as a substrate for deposition of different substances, including metals and insulators. We believe that the papers published within the current Special Issue develop cumulative knowledge on matters related to device-quality epaxial graphene on SiC, bringing this material closer to realistic practical applications.

Keywords: epitaxial graphene; sublimation; SiC; buffer layer; electronic properties; material engineering; deposition

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1. Introduction

For more than a decade, investigations of epitaxial graphene on SiC have gained special urgency in view of its possible applications in many fields, including metrology, electronics and sensorics. Further progress in the development of related technologies requires both rethinking of already existing knowledge and discovery of innovative solutions. This was the primary motivation for opening the call for papers within the Special Issue "Fundamentals and Recent Advances in Epitaxial Graphene on SiC".

In total, the Special Issue encompasses four research papers and three review papers. Two research works touch on crucial aspects of early stage of graphene growth, namely buffer layer formation [1], and graphene quality estimation [2]. Kaushik et al. [3] reported on a principal possibility to tune structural and electronic properties of epitaxial graphene through nitrogen ion implantation. Fundamental knowledge on both copper electrodeposition and atomic layer deposition of high-*k* insulators on epitaxial graphene/SiC is provided by the authors of [4,5]. These results suggest that epitaxial graphene is a stable support for metal and metal oxides, which is important in the context of metal contacts, gating, etc. Concomitantly, the interaction between epitaxial graphene and its environment, including metal contacts may limit, to some extent carrier transport in epitaxial graphene and therefore needs to be considered in detail. The role of such interaction has been a research subject of the review paper by Pradeepkumar et al. [6]. Finally, Wu et al. [7] critically reviewed recent advances in graphene twistronics and identified epitaxial graphene on SiC as the most promising platform for twistronics.

2. Critical Aspects of Epitaxial Graphene Growth: Recipes, Properties, and Quality

The quality of the buffer layer (also known as C-rich surface reconstruction of SiC and zero graphene layer) is identified as one of the most important factors determining the quality of the epitaxial graphene monolayer on SiC grown via Si sublimation approach. In other words, the fabrication of a large-area epitaxial graphene layer with high thickness uniformity requires pre-formation of high-quality continuous buffer layer on large areas.

Thus, an optimization of the growth regime with respect to the buffer layer formation during early stage graphenization process as well as a complete understanding of the growth mechanism are key ingredients to fabricate device-quality graphene. In fact, a successful graphenization process may occur only in a very narrow operational temperature/pressure window which imposes significant restrictions on the growth regime tunability. This makes the optimization of growth conditions quite a challenging task. Despite the large research efforts to tackle this task, it still requires more systematic consideration. In this regard, the critical study by the authors of [1] on optimizing the formation conditions of buffer layers through control of the graphite crucible temperature and varying the Ar gas pressure is a recent contribution to the process. It was revealed that the buffer layer coverage is strongly dependent on the temperature at which Ar gas is introduced, demonstrating a gradual decrease as the temperature increases. The mechanism behind this behavior has been discussed. In the same paper, the relationship between the growth temperature and electronic properties (carrier mobility, carrier density) of quasi-freestanding graphene monolayer and epitaxial graphene monolayer, respectively, were discussed. It was also illustrated that the conductivity type and free carrier density for graphene are extremely sensitive to ambient conditions which was observed by many researchers earlier. In line with this, the review paper by Pradeepkumar et al. [6] provides a more general picture of the effect of epitaxial graphene-ambient interaction on the carrier transport in SiC-supported epitaxial graphene. The authors highlight the adsorption of different molecules (O_2, H_2O, NO_2, H_2O_2, CO_2, NH_3, CO, NO, N_2O_4) as a main reason that underlie conductivity type flipping, transport properties fluctuations and carrier density saturation.

Apart from the unintentional doping of epitaxial graphene by environmental gases and other molecules, the electronic properties of epitaxial graphene on SiC can be modulated by the intentional incorporation of external dopants, as was demonstrated in another study [3]. Nitrogen ion implantation was proposed as an instrumental approach to stabilize the *n*-type conductivity in epitaxial graphene without serious structural damage. However, a balance between graphene quality and implantation dose must be reached. In that light, the mentioned paper dealt with finding the correlation between the fluence value and epitaxial graphene properties such as fragmentation degree, and defect density.

It is instructive that all mentioned works exploit Raman Spectroscopy to estimate the quality of epitaxial graphene. More specifically, the relationship between the intensities of $2D$ and G characteristic peaks is used to determine the number of graphene layers, while D/G amplitude ratio is employed to calculate the defect density. Although the graphene Raman spectroscopy is a mature field, it continues to evolve especially in the direction of signal processing (for example, peak fitting quality). In this regard, the work by Kunc and Rejhon [2] originally offers a Voigt line shape fitting approach for analysis of $2D$ peak line shape for epitaxial graphene, which includes both the inhomogeneous and homogeneous broadening. They also interpreted the physical nature of each term by ascribing the homogeneous broadening to intrinsic lifetime and inhomogeneous broadening to strain fluctuations, respectively.

3. Epitaxial Graphene as a Host for Material Deposition

Epitaxial graphene on SiC is of great interest because it not only has extraordinary intrinsic properties but also can be used as an atomically flat robust support for non-hybridized growth of different materials, especially metals and metal oxides. Such an integration may expand the functionality of epitaxial graphene and boost the development of innovative technologies in conceptually new fields, like catalysis, plasmonics, and spintronics. Thus, research efforts to contribute to this field and to enrich the existing knowledge capital are in high demand. In response to this demand, the group at Linköping University [4] launched a systematic study of metal electrodeposition on epitaxial graphene on SiC, choosing copper as a model metal at the first stage. This work sheds light on fundamental aspects of copper electrochemistry on epitaxial graphene and shows that copper electroreduction occurs via two subsequent single-electron transfer steps. The instantaneous nucleation mechanism was identified as a dominating mechanism during copper electrodeposition. The present results provide a deep understanding of the nature of copper–epitaxial graphene interaction, thereby facilitating the design of novel copper–graphene nanohybrid materials.

At the same time, Giannazzo et al. [5] in their work gave an overview of the recent results on the growth of high-k insulators on epitaxial graphene on SiC, focusing on atomic layer deposition of Al_2O_3 thin layers, which are important for fabrication of epitaxial graphene-based devices. It was argued that the monolayer epitaxial graphene uniformity is a key factor to achieve a homogeneous Al_2O_3 coverage via direct deposition, the latter has not been successful before in other studies. The role of different seeding layers and surface pre-functionalization in atomic layer deposition processes on epitaxial graphene is critically discussed. Finally, the authors explained the effect of pre-treatment and grown layers on the quality and electronic properties of epitaxial graphene. As was discussed in [6], this issue requires careful consideration, since the interaction between graphene and deposited layers may significantly affect the electron transport in graphene.

4. A New Look at Possible Applications of Epitaxial Graphene on SiC

Although the epitaxial graphene on SiC is nowadays reasoned to be utilized in electronics, quantum metrology, and gas/liquid sensing, the unique properties of this material make it promising for use in other non-conventional fields. Wu et al. [7] claim that epitaxial graphene on SiC could be regarded as an excellent platform for formation of twisted few-layer graphene with a magic twist angle that might be useful to control spin orders, ferromagnetism, and superconductivity. The authors have substantiated this claim by the fact that owing to its natural compatibility with the semiconductor technologies, epitaxial graphene-based device processing requires no intermediate graphene transfer steps and thus is more attractive from a technological point of view in comparison to transferred graphene. In this regard, there is a plenty of room for manipulation of the twist angle and for formation of twisted graphene layers on SiC with the desired angle through adjusting the sublimation growth conditions.

5. Concluding Remarks

The Guest Editors consider the current collection of papers as an important piece of the puzzle needed to boost both the more rational implementation of epitaxial graphene into traditional devices and the development of non-conventional innovative technologies. Furthermore, the new results reported in the frame of the Special Issue complement the existing knowledge on buffer layer formation, material preparation–property relationships, and growth mechanisms of different materials on epitaxial graphene. We believe that this information input will provide the driving force behind future experimental efforts to improve the epitaxial graphene quality and to design sophisticated devices exploiting epitaxial graphene as active and passive components.

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