Growth mechanisms of GaAs/graphene/InAs double heterostructures

JO, J.[1], Tchoe, Y.[2], Ryu, J.[1], Yi, G.[2] and Kim, M.[1]

[1] Department of Materials Science and Engineering, Seoul National University, Republic of Korea, [2] Department of Physics and Astronomy, Institute of Applied Physics, Seoul National University, Republic of Korea

Atomically thin layered crystals isolated by mechanical exfoliation method have exhibited new physical properties and provided novel applications [1]. Moreover, hybrid heterostructures of these two-dimensional (2-D) layered materials with semiconductor thin films and nanostructures offer additional functionalities, such as flexibility and transferability, thereby greatly extending their applicability to the future electronic and optoelectronic devices [2]. One of the interesting aspects concerning the hybrid heterostructures is the fabrication of double heterostructures composed of two different material systems grown on the either side of 2-D material. Double heterostructures are the next-generation material systems that have a great potential for the large scale integration and multifunctionality of devices, compared with the conventional device structures. Accordingly, it is important to develop the capability of growing high-quality hybrid heterostructures. The growth of nanomaterials on 2-D materials is governed by a new mechanism called van der Waals epitaxy, which features weak van der Waals interactions that bind the growing material to substrates, *i.e.* 2-D materials. The van der Waals epitaxy thus eliminates any necessity to satisfy the lattice matching requirements, as opposed to the conventional epitaxy, which allows incommensurate epitaxy that enables coherent growth of materials despite a large lattice mismatch with 2-D materials. Additionally, atomically thin nature of 2-D materials may transfer the potential field of the growing materials to the opposite side through the ultrathin monoatomic layer [3], which implies the possibility that the materials grown on 2-D materials can influence the nucleation and growth behavior on the other side of double heterostructures. These unique growth processes could offer a great opportunity for achieving high-performance functional devices based on double heterostructures. In order to fabricate such high-performance double heterostructures with desired shapes and physical properties, the study on the growth mechanisms, such as nucleation, nuclei growth, and orientational relationship with a substrate, should be accompanied first in detail. However, the systematic study focusing on the fundamental mechanisms of the van der Waals epitaxy occurring on both sides of 2-D material has rarely been performed.

Here, we report on the growth behavior of GaAs/single-layer graphene (SLG)/InAs double heterostructures to verify the role of graphene in the van der Waals epitaxy. Two different types of nanomaterials were grown directly on both sides of free-standing SLG to avoid unintentional damages arising from conventional transmission electron microscopy (TEM) sample preparation processes (Fig. 1). TEM measurements revealed that the growth behavior of GaAs/SLG/InAs double heterostructures was different from that of GaAs/InAs heterostructures without any materials underneath SLG; the growth direction of GaAs was largely flipped in the direction normal to the surface of SLG by the interaction with InAs nanorods underneath SLG (Fig. 2). This indicates that InAs nanorods already present underneath SLG can affect the nucleation and growth behavior of GaAs on the opposite side through the atomically thin SLG (Fig. 3). We believe that this study will lay down the building block for fundamental understanding of how 2-D materials influence the growth behavior of double heterostructures.

[1] Novoselov K.S. *et al. Science*, **306,** 666 (2004)

[2] Chung K. *et al. Science*, **330,** 655 (2010)

[3] Kim Y. *et al. Nature*, **544,** 340 (2017)

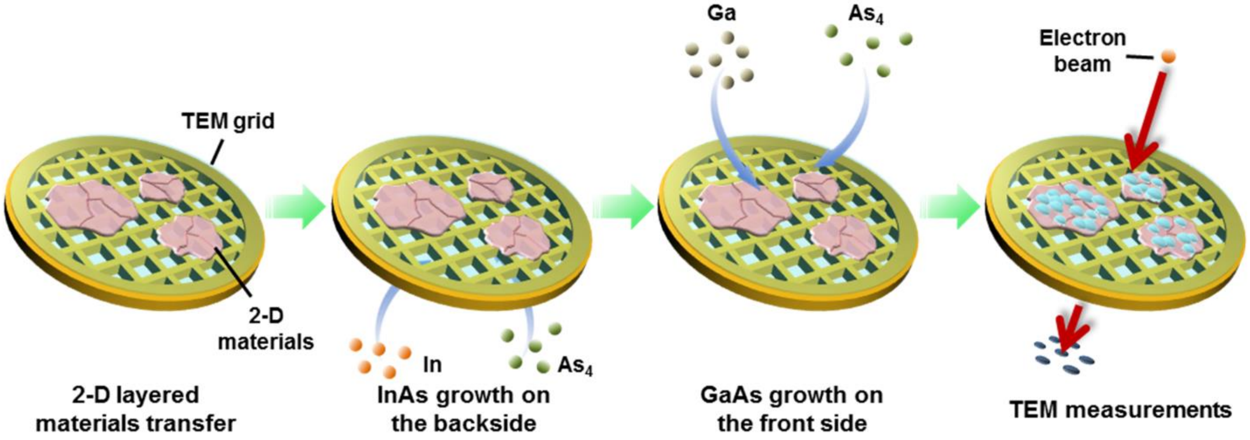

Figure 1. Schematic diagrams of the experimental technique for growing GaAs/graphene/InAs double heterostructures and performing TEM measurements. Two-dimensional materials were transferred onto a TEM grid. InAs was grown first on the backside of graphene by catalyst-free MBE method. Afterward, GaAs was deposited on the front side of graphene. After the growth, TEM measurements were performed without conventional TEM sample preparation processes.

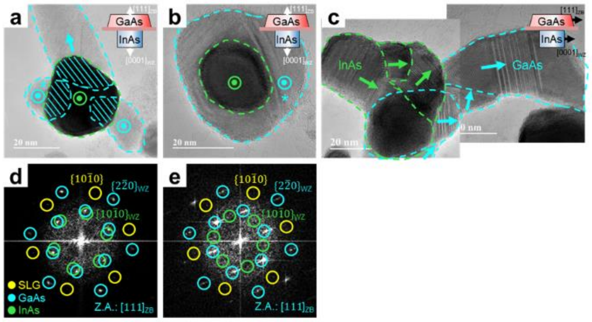

Fig. 2 | Growth behavior of GaAs clusters on SLG/InAs nanorods. (a–c) HR-TEM images showing GaAs clusters grown on SLG/InAs nanorods with both GaAs $[111]_{ZB}$ and InAs $[0001]_{WZ}$ directions pointing (a–b) vertically and (c) laterally with respect to the surface of SLG, respectively. Schematics in the insets represent the directions of GaAs $[111]_{ZB}$ and InAs $[0001]_{WZ}$. The grains of GaAs and InAs in the clusters are identified by cyan and green dashed lines, respectively. The $[111]_{ZB}$ and $[0001]_{WZ}$ direction of each grain is identified by an arrow. (d–e) Corresponding FFT patterns obtained from GaAs grains marked with asterisks in a and b, respectively.

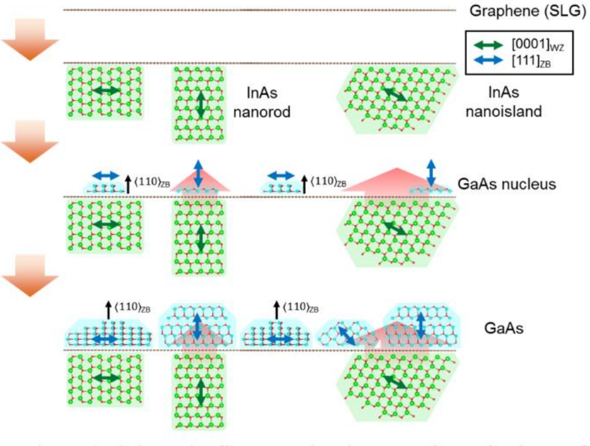

Figure 3. Schematic diagrams showing growth mechanisms of GaAs/SLG/InAs double heterostructures. GaAs and InAs nanostructures are represented by blue and green polygons, respectively, with their atomic configurations observed along $[11\bar{2}0]_{WZ}/[1\bar{1}0]_{ZB}$ directions. The directions of GaAs $[111]_{ZB}$ and InAs $[0001]_{WZ}$ are identified by blue and green arrows, respectively.