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Graphene-Assisted Heteroepitaxy Towards Dislocation-Free Materials

Sungkyu Kim*

Sejong University, Korea

Although high-quality single-crystalline thin films can be grown onto the substrate via conventional homoepitaxy, material systems are limited by crystal structure and lattice mismatch. In addition, conventional heteroepitaxy of lattice-mismatched systems generates dislocations to release the strain energy between epilayer and substrate. Since the formation of dislocations in the epilayer degrades device performance, it is essential to control the formation and growth of defects. Based on the recently reported remote epitaxy, high-quality single-crystalline materials are obtained by an inserted atomically thin graphene layer between the epitaxial layer and the substrate.

Here, we demonstrate the role of a graphene layer that allows the heteroepitaxy materials of the highly-mismatched systems with reduced dislocation density. Van der Waals field generated onto graphene film effectively controls ionic force between the seed atoms of epilayer and substrate at the early stage of growth, which enables spontaneous relaxation of the epilayer before the accumulated elastic energy created by the lattice mismatch induces misfit dislocations. From the system with a small lattice mismatch (InGaP on GaAs with 0.74% misfit), we observed that conventional heteroepitaxy gradually relaxes the misfit strain via the introduction of dislocation, while heteroepitaxy on graphene-coated substrates abruptly relaxes the strain in the epilayer via interface displacement on the graphene's slippery surface. This effect becomes more prominent in a highly mismatched system (GaP on GaAs with 3.7% misfit), where a substantially reduced dislocation density is seen. In addition, this innovative technique is successfully applied to the heterogeneous integration of single-crystalline complex-oxide membranes. The complex oxide materials including perovskite, spinel, and garnet crystal formed by remote homoepitaxy and heteroepitaxy are successfully exfoliated from the graphene-coated substrate to fabricate freestanding single-crystalline membranes. Furthermore, the modified heteroepitaxy method with a buffer epilayer allows the physical lift-off of grown single-crystalline oxide materials via regularly generated dislocations.

The spontaneous relaxation technique could enable the monolithic integration of largely lattice-mismatched systems with minimized dislocation density, which could eventually broaden the material spectrum for advanced electronics and photonics. Also, the combination of remote heteroepitaxy and engineered heteroepitaxy with the buffer layer establishes a platform for the future stacking three-dimensional structures.

*Corresponding author	Sungkyu Kim
Affiliation	Sejong University, Korea
E-mail address	sungkyu@sejong.ac.kr

Submitter's Information

Name	Prof. Sungkyu Kim
E-mail	sungkyu@sejong.ac.kr

▪ Affiliation	Sejong University
▪ Department	Department of Nanotechnology & Advanced Materials Engineering
▪ Country	Korea
▪ Tel	+82-10-9707-1030

▪ Author(s)

Author 1	**Presenter**	Yes	**Corresponding Author**	Yes
	▪ Name	Prof. Sungkyu Kim		
	▪ Email	sungkyu@sejong.ac.kr		
	▪ Affiliation	Sejong University, Korea		

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