

Remote-epitaxially grown single-crystalline complex oxide for the integrated system

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Using the conventional epitaxy technology, various single-crystalline materials can be directly grown on single-crystalline substrates. The grown epitaxial materials exhibit excellent performance similar to the theoretical values, but there is a big challenge to independently operate from the seed substrate due to a strong atomic bonding. In order to fabricate the freestanding thin films, new concept growth methods using weak van der Waals force on two-dimensional materials (van der Waals epitaxy) and chemical etching/buffer layer (chemical lift-off) for oxide materials. However, the crystal structure of two-dimensional material and the insertion of the sacrificial layer could be limited in growing single-crystalline materials and their application for the future integrated system.

Here, we introduce a novel epitaxy method (remote epitaxy) for growing single-crystalline oxide films through the graphene on the substrate as a seed layer. Several important oxide structures including perovskite (SrTiO_3 , BaTiO_3 , $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$), spinel (CoFe_2O_4), and garnet ($\text{Y}_3\text{Fe}_5\text{O}_{12}$) are successfully demonstrated via remote epitaxy technologies. In contrast to III-V compound^[1] and III-Nitride^[2] material that can stably exist in graphene during the growth process, monolayer graphene is very susceptible to physical damage and structural deformation due to high oxygen partial pressure and high temperature conditions. Because of these critical issues such as deformation and oxidation of graphene, the grown single-crystalline oxide films cannot be peeled off from the substrate or partially shows polycrystalline. The oxygen partial pressure was gradually increased to minimize the oxidation of graphene, and two layers of graphene were used to prevent the destruction of graphene at the same time. Based on these modifications, magnetoelectric coupling has been observed by stacking CoFe_2O_4 and PMN-PT structures^[3]. These findings provide tremendous opportunities to fabricate various functional oxide materials, which could be eventually integrated for the future electronics.

[1] Bae, S.-H. et al. Graphene-assisted spontaneous relaxation towards dislocation-free heteroepitaxy. *Nature Nanotechnology* 15, 272-276 (2020)

[2] Kim, Y. et al. Remote epitaxy through graphene enables two-dimensional material-based layer transfer. *Nature* 544, 340-343 (2017)

[3] Kum, H. S. et al. Heterogeneous integration of single-crystalline complex-oxide membranes. *Nature* 578, 75-81 (2020)