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Van der Waals Epitaxy: A new way for growth of III-nitrides

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Inorganic semiconductor plays a key role for today's technological progress [1–4]. Comparing with other organic semiconductors [5,6] and two-dimensional (2D) metal sulfides [7–10], III-nitrides as direct bandgap semiconductors have achieved enormous success in commercial applications. Owing to their excellent physical properties and adjustable bandgap from ultraviolet to near-infrared range by composition design in their family [11–13], III-nitrides have shown great potential in many fields including but not limited to ultraviolet (UV) light-emitting diodes (LEDs) and photodetectors. Due to the deficiency of suitable substrate, III-nitrides are usually epitaxial on foreign substrates by metal-organic chemical vapor deposition (MOCVD) [14,15]. The intrinsic difference between epitaxy layer and substrate in lattice constant and thermal expansion coefficient induces large residual stress and high defect density, and therefore degrading the performance of III-nitride based devices [16]. In spite of great achievement in reducing defect density in III-nitrides, further requirement for improvement in crystal quality is still remained. In addition, the strong covalent bond between substrate and epitaxy layer makes it difficult to exfoliate III-nitride films from substrate for further fabrication in flexible and wearable devices.

Recently, a novel method termed as "van der Waals (vdW) epitaxy" provides promise to solve both above-mentioned difficulties. By introducing 2D materials between substrate and epitaxy layer, the crystal quality could be improved due to the 2D buffer layer and the release of epitaxy layer is facile by the mechanical exfoliation owing to the weak vdW interaction. Among 2D material family, graphene is the most extensively investigated material for vdW epitaxy.

Figure 1(a) shows ZnO-coated graphene as seed layer to grow GaN epilayer and subsequent LEDs structure, which could be easily transferred onto other foreign substrates [17]. The thin layer graphene could not screen the strong potential field of some substrate like GaAs and the growth of epilayer is still controlled by the underneath homoepitaxial substrate [18]. High-quality GaN and AlN grown on graphene-coated sapphire and amorphous silicon oxide have been realized by vdW epitaxial process [19–21]. Thanks to the weak vdW interaction between III-nitrides and graphene, epilayer suffers bare restriction from the underneath substrates then realizes low defect density and relaxed stress. As pristine graphene is an integrated 2D flat structure which lacks of dangling bonds, resulting in low nucleation density of III-nitrides and three-dimensional island growth mode. The N_2 plasma or NH_3 treatment (in MOCVD system) are introduced for pristine graphene to create C-N bonding as III-nitride nucleation sites (Figure 1(b)).

In addition to graphene, hBN has also been experimentally applied as buffer layer to grow stress-free and transferable III-nitrides epilayer [22]. According to recent reports, the crystal quality of III-nitrides vdW epitaxy on hBN is inferior to that grown on graphene [23,24].

To explore efficient vdW epitaxy strategy, the construction of 2D materials' substrates matched with III-nitrides vdW

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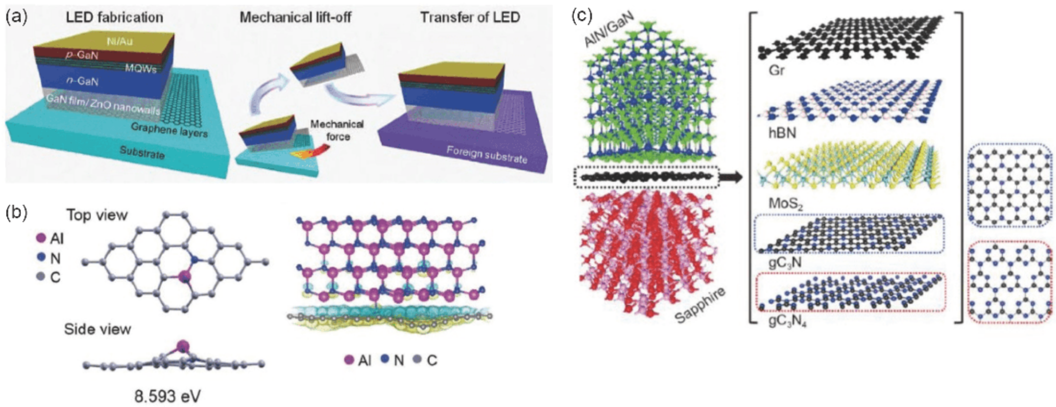

Figure 1 (Color online) (a) Schematic illustration of the fabrication and transfer processes for thin-film LEDs grown on graphene-layer substrates [17]; (b) atomic model structures of AlN on N_2-plasma-treated graphene via Al-N bond [20]; (c) scheme of GaN or AlN epilayer stacking on sapphire covered with different 2D materials [25].

epitaxy is crucial. Lots of materials in 2D family are still remained to be explored. In our recent work, we have proposed five existing 2D materials for vdW epitaxy of III-nitrides, including graphene, hBN, MoS_2, gC_3N, and gC_3N_4 (Figure 1(c)), providing the physical insights by first-principle calculations [25]. The calculated results indicate that MoS_2 and gC_3N are better candidates than graphene and hBN for vdW epitaxy because of their suitable binding strength and diffusion barrier. This finding is helpful to obtain high-quality III-nitrides and develop new criterions to discover effective 2D materials for vdW epitaxy.

Although vdW epitaxy of III-nitrides on 2D materials has achieved apparent progress in lower defect density, released stress and transferable devices, the inherent nucleation mechanism and specific growth kinetics process are still unclear. III-nitrides grown on N-doped graphene which needs bonding between Ga/Al and N is more likely quasi-vdW heteroepitaxy, complete vdW epitaxy is only achieved on homoepitaxial substrates. Further systemic research should be carried out to definite its kinetics process and answer these question properly. Otherwise, 2D materials for vdW epitaxy have been limited to graphene and hBN, metal sulfides/selenides with low temperature stability are not successfully applied in actual MOCVD condition. The III-nitrides vdW epitaxy on these substrates also needs more attention, aiming at enlarging the feasibility of vdW epitaxyon various 2D substrates.

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