Supplemental Document

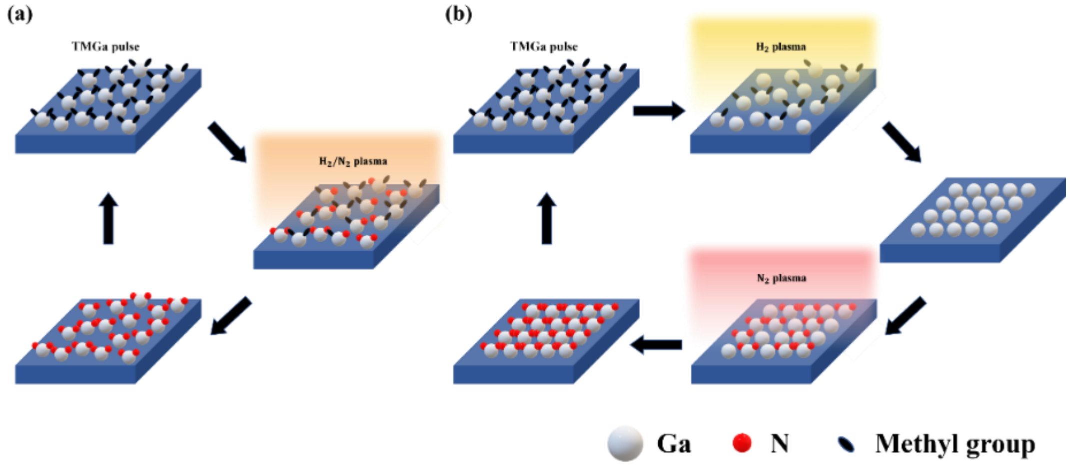

Figure 1. Schematic illustration of (a) the mixed H_2/N_2 plasma mode, where steric hindrance from residual ligands restricts surface reactions, and (b) the sequential H_2/N_2 plasma mode (HM-ALE), which enables a ligand-free surface template. The specialized H_2 plasma step not only effectively eliminates organic species but also facilitates surface reconstruction through energy transfer from hydrogen species.

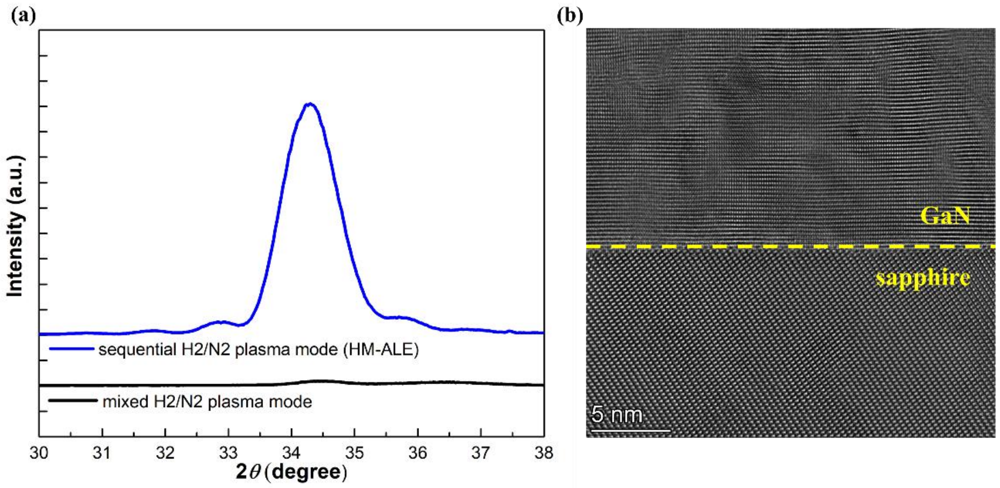

Figure 2. (a) ω-2θ XRD patterns of GaN films grown using the mixed H_2/N_2 plasma mode and the sequential H_2/N_2 plasma mode (HM-ALE). Both films exhibit a GaN (0002) diffraction peak at $2\theta \approx 34.3°$, indicating c-axis-oriented wurtzite GaN with the (0002) planes parallel to the substrate surface. The markedly enhanced peak intensity for the sequential H_2/N_2 plasma mode demonstrates a significant improvement in crystalline quality. (b) High-resolution TEM image of GaN grown on sapphire using the HM-ALE method, showing well-resolved lattice fringes.