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Fig. S1 Representative SEM images of a AlTiN (~11%) film on a SiO₂ substrate for (a) low and (b) high magnification. To demonstrate that the surface of the film was in focus, a contamination on the film was intentionally included in the pictures.



Fig. S2 Models for *ab initio* calculation. (a) Wurtzite AlN and (b) wurtzite AlTiN where a Ti atom substitutes an Al atom. Large light blue balls are Al atoms and small dark blue balls are N atoms. Large yellow ball is the Ti atom.



Fig. S3 Tauc plots for the AlTiN films corresponding to (a) allowed direct and (b) allowed indirect transitions. (c) The estimated E_g values.



Fig. S4 Tauc plots for estimation of "Eg" plotted in Fig. S3(c).



Fig. S5 Scanning TEM images of an AlTiN (~11%) film on Ti substrate for (a) low and (b) high magnification near the surface.

(a)



Fig. S6 XRD profiles of AlTiN films with various Ti concentrations in out-of-plane (a) and inplane (b) modes.