**Supplementary Materials**

# *In situ* Nanoindentation Study of Plastic Co-deformation in Al-TiN Nanocomposites

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Figure S1. Σ3{112} ITBs in the Al layers

Figure S2. HRTEM images of the 5 nm Al – 5 nm TiN multilayers

Figure S3. HRTEM images of the 2.7 nm Al – 2.7 nm TiN multilayers

Figure S4. Stress field in the TiN layer predicted by finite element method

Movie I: *in situ* indentation of the 50 nm Al – 50 nm TiN multilayer, played at 15x speed (3 frames/sec)

Movie II: *in situ* indentation of the 5 nm Al – 5 nm TiN multilayer, played at 15x speed (3 frames/sec)

Movie III: *in situ* indentation of the 2.7 nm Al – 2.7 nm TiN multilayer, played at 8x speed (3 frames/sec)

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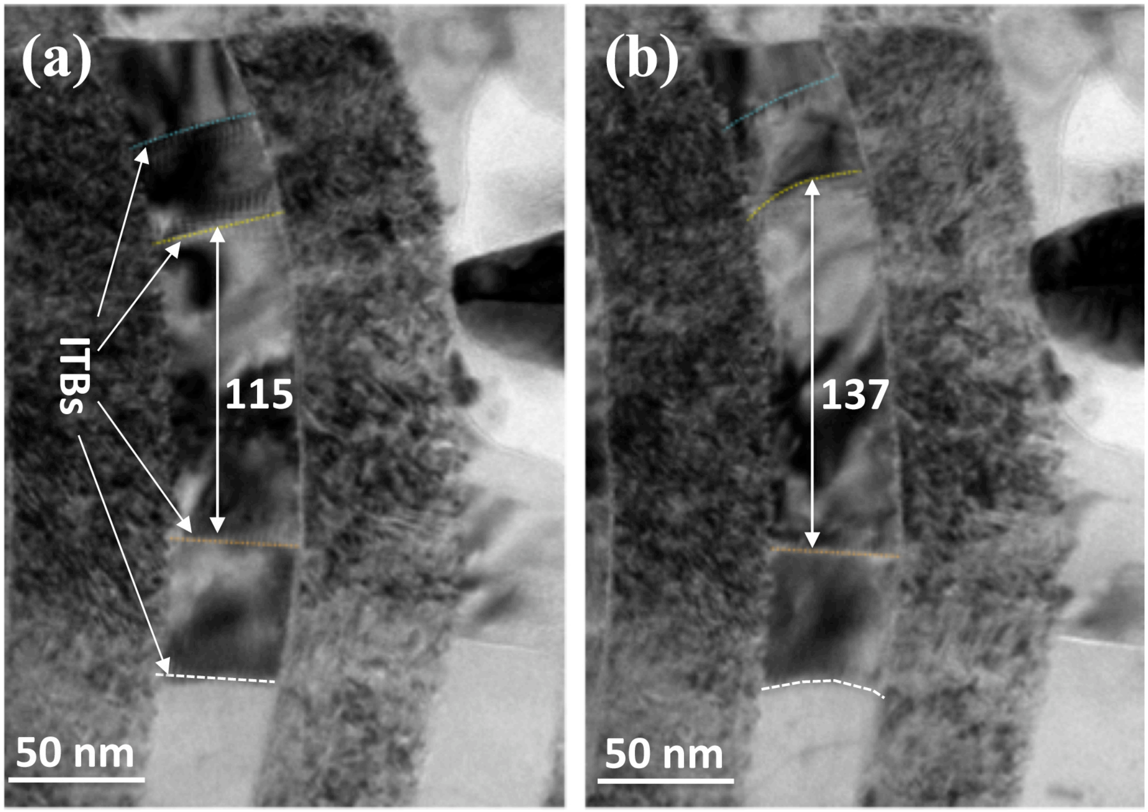


Figure S1. TEM images show Σ3{112} ITBs in the Al layer and their migration during indentation. The distance between the two ITBs increases from 115 nm to 137 nm.

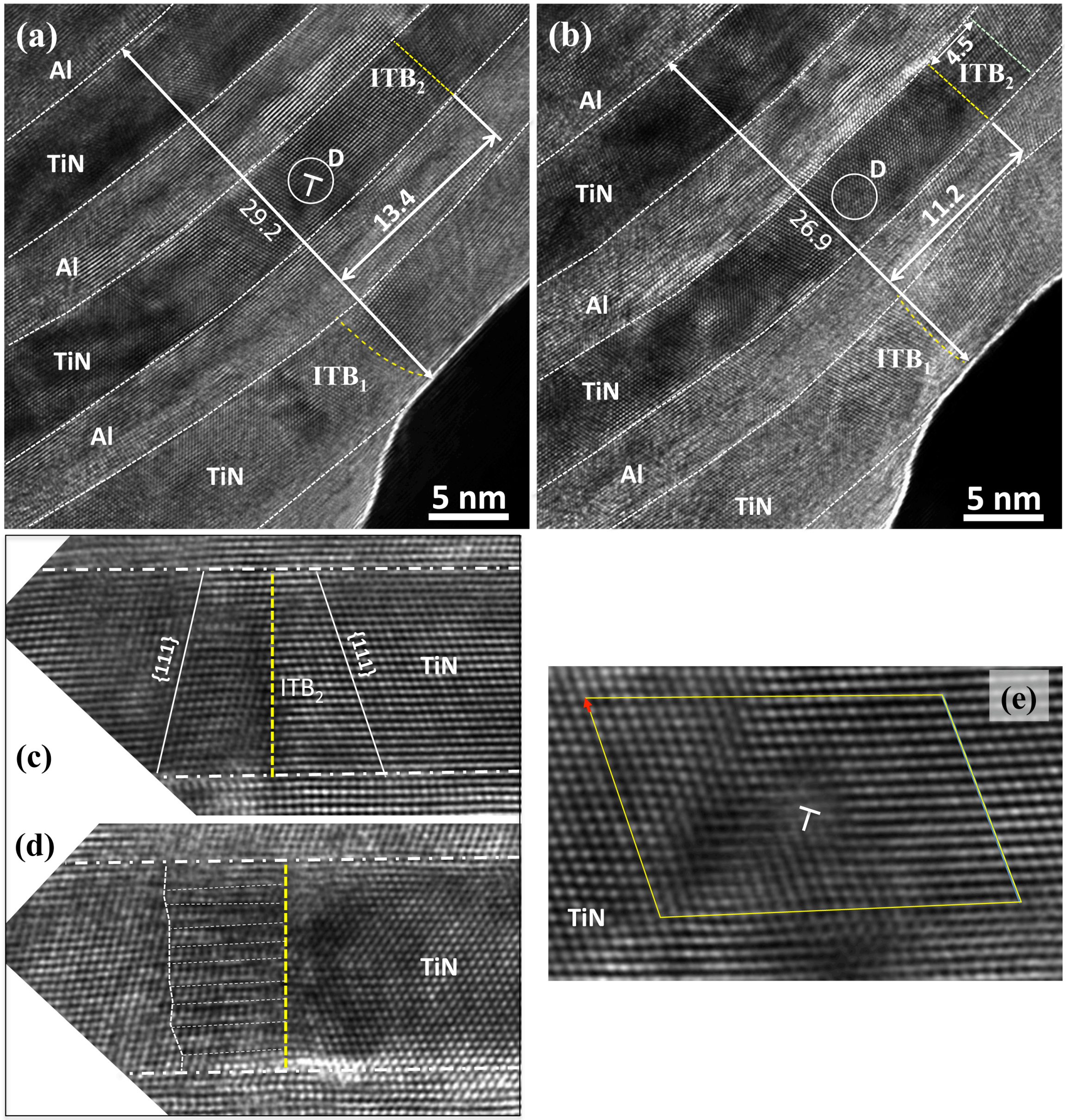


Figure S2. (a) and (b) HRTEM images of the 5 nm Al -5 nm TiN multilayer during indentation. The first five layers reduce the thickness from 29.2 nm to 26.9 nm, corresponding to a compressive strain of 8%. The yellow dashed lines indicate Σ3{112} ITBs in the TiN layers. The distance between the two ITBs reduces from 13.4 nm to 11.2 nm, indicating the migration of the $ITB_1$. (c) The straight $ITB_2$ and (d) the dissociated $ITB_2$. (e) A dislocation is characterized in the region D in (a) by the Burgers circuit, but disappears in (b). This implies the dislocation motion/activity in the TiN layer during indentation.

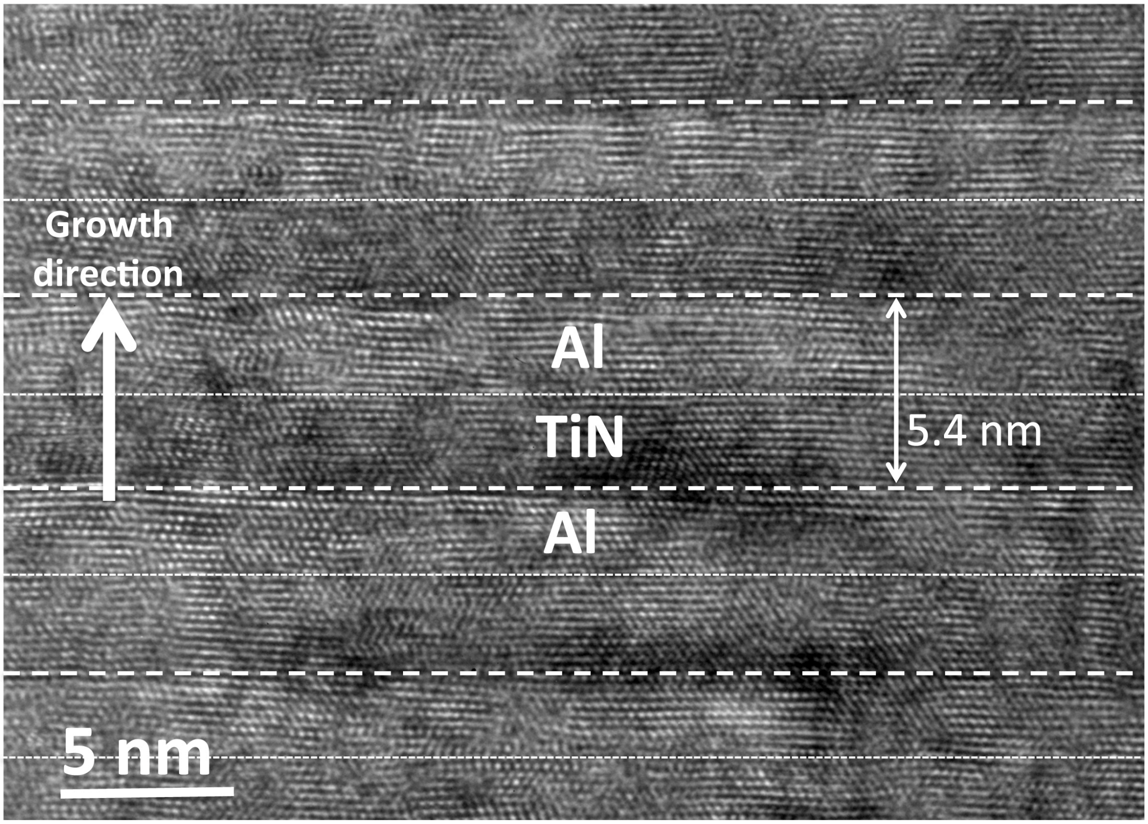


Figure S3. A HRTEM image of the 2.7 nm Al – 2.7 nm TiN multilayer. The bold dashed lines indicate the sharp and flat Al-TiN interface and the thin dashed lines indicate the relatively rough TiN-Al interface.

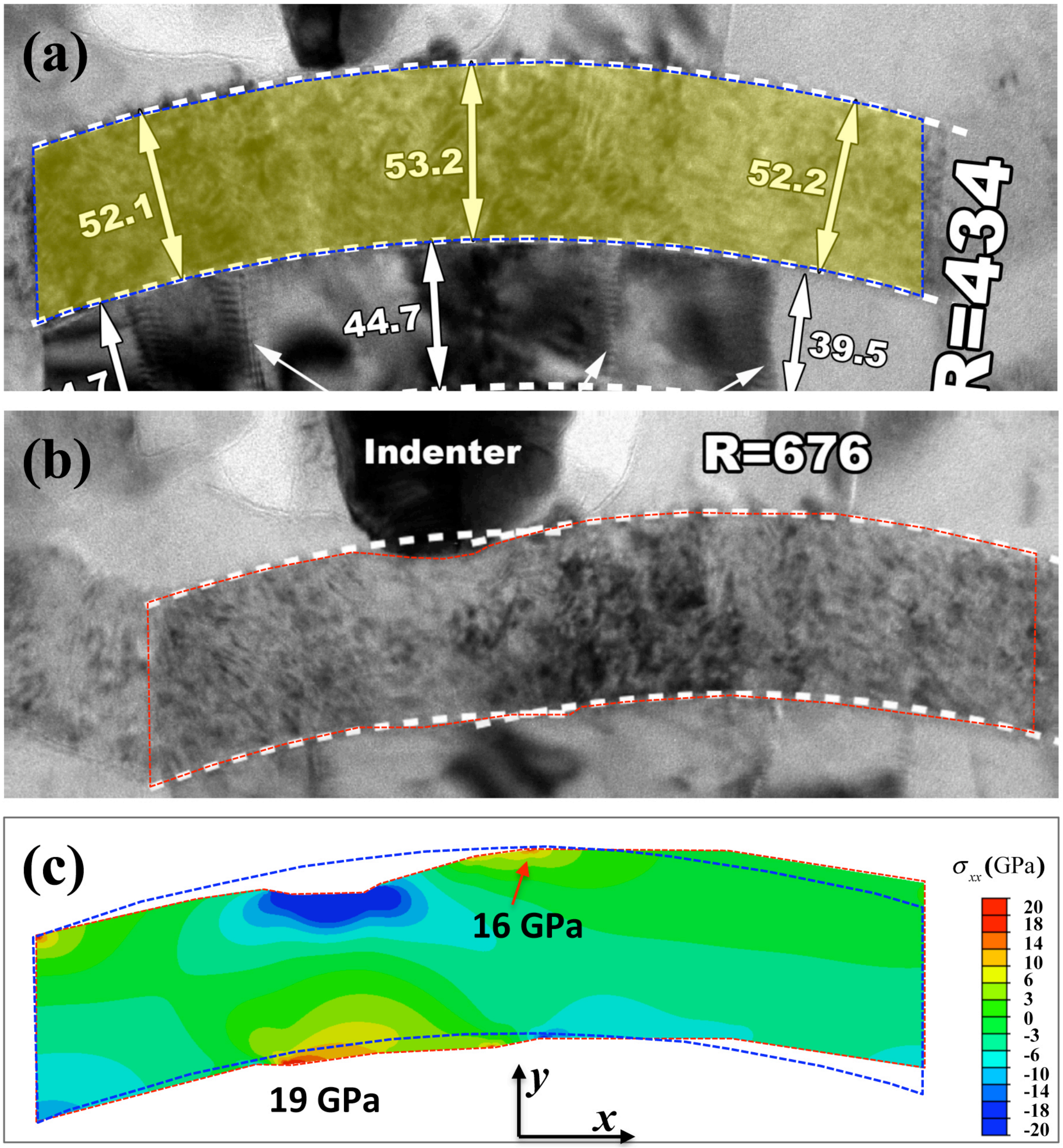


Figure S4. The change in the morphology of the first TiN layer during indentation. (a) The blue dashed line outlines the initial shape of the TiN layer before indentation, (b) the red dashed line outlines the deformed TiN layer before cracking. The stress field was solved in the TiN layer with the displacement boundary condition which is determined according to the morphology change from (a) to (b). The Young's modulus is 260 GPa and the Poisson's ratio is 0.30. Two high tensile stress regions are corresponding to the crack initiation.