Supplementary Information for

Nucleation preference and lateral growth of monolayer tin disulfide on graphene

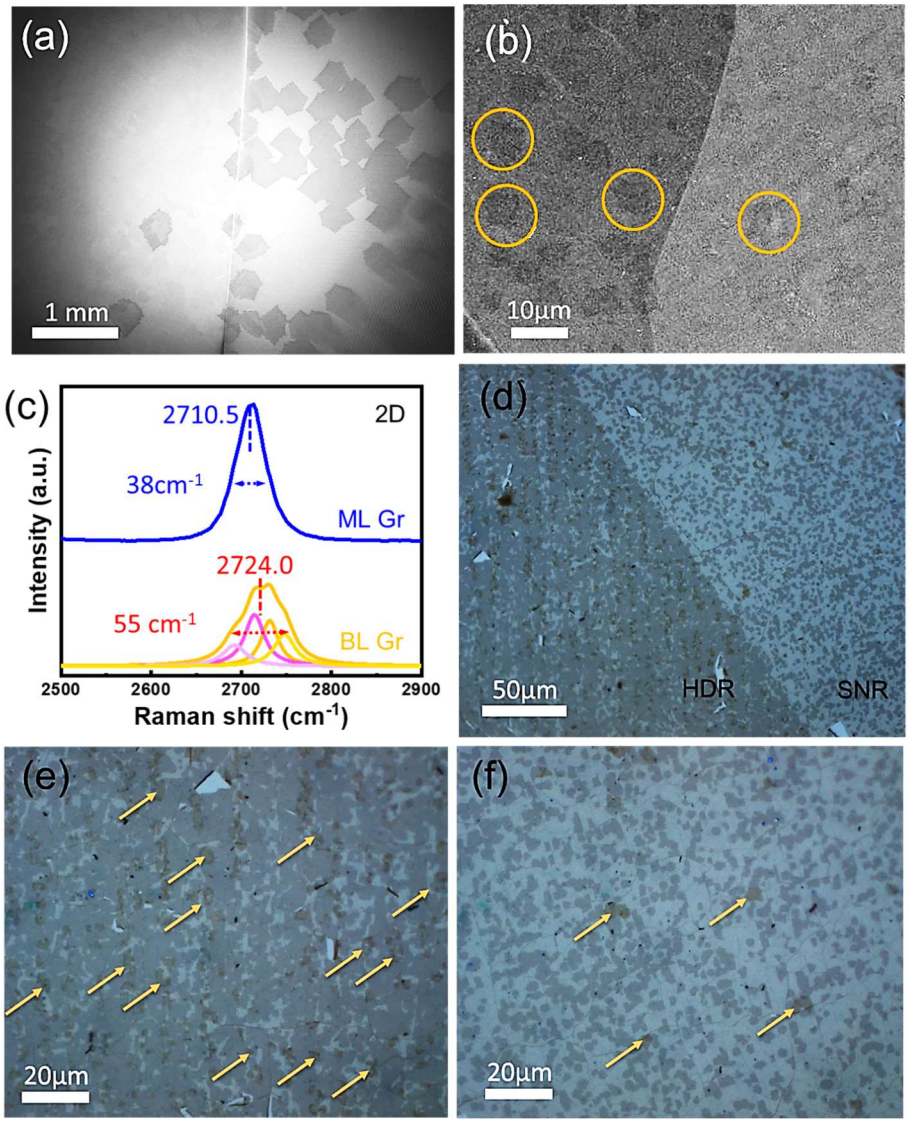

Figure S1. (a) SEM image of single crystal graphene synthesized on different Cu grains. (b) The distribution of adlayer graphene, highlighted by orange circles, on different Cu grains. (c) Comparison of Raman 2D band in monolayer and adlayer graphene. (d-f) Optical images of SnS_2 grown in HDR and SNR. The distribution of adlayer graphene, marked with yellow arrows, in (e) HDR and (f) SNR regions, respectively.

The synthesis of graphene is significantly influenced by the Cu substrate. Fig. S1a shows the nucleation difference of single-crystal graphene on different Cu grains. The bright line in the image depicts the Cu grain boundary, and the graphene domains appear

as darker patches. Clearly, on the left area of the boundary, there are only a few graphene domains. In contrast, on the right, the number of graphene grains increases significantly, with a coverage of more than half of the area. The graphene film with adlayers was also synthesized on Cu foil. The adlayer graphene can be distinguished from the monolayer graphene by the color contrast in the image, as indicated by the orange circles in Fig. S1b. The density of the adlayer graphene on the left side is much higher than that on the right. This is likely due to difference in the amount of subsurface carbon inside the Cu grains, which leads to the variation in the number of additional graphene nuclei.

Fig. S1c presents the Raman spectra collected from the regions of single-layer and adlayer graphene. The 2D peak of monolayer graphene shows a full width at half maxima (FWHM) of ~38 cm^{-1} and appears as a single and symmetric Lorentzian peak. This is consistent with the highly ordered structure and simplified vibrational modes within the single-layer graphene. The 2D Raman peak of adlayer graphene shows a significant blue shift and an FWHM of ~55 cm^{-1}, which could be decomposed into four Lorentzian peaks. The 2D band arises from a two-phonon resonance process, which is closely related to the band structure of graphene. For the bilayer graphene with AB stacking, the conduction and valence bands are composed of two parabolas, resulting in four possible double resonance scattering processes. Therefore, the adlayer regions have the features of strongly coupled bilayer graphene.

The boundary between HDR and SNR of SnS_2 nuclei is clearly seen in the optical image (as shown in Fig. S1d), due to the noticeable difference in nucleation density and the resulting difference in color contrast. Fig. S1e and S1f display the enlarged images of these two regions, respectively, in which the graphene adlayers can be distinguished from monolayer graphene and SnS_2 domains based on the darker contrast, as indicated by yellow arrows. The statistical results show that in the HDR with high-density SnS_2 grains shown in Fig. S1e, the coverage of adlayer graphene exceeds 31.2%. In contrast, in the SNR where SnS_2 grains are sparsely distributed, shown in Fig. S1f, the number of adlayer graphene is only 11 locations (about 4.2% in coverage). It shows a strong correlation between the distribution of adlayer graphene and the nucleation preference of SnS_2.