Electronic Supplementary Information (ESI)

Direct characterization of intrinsic defects in monolayer $ReSe_2$ on graphene

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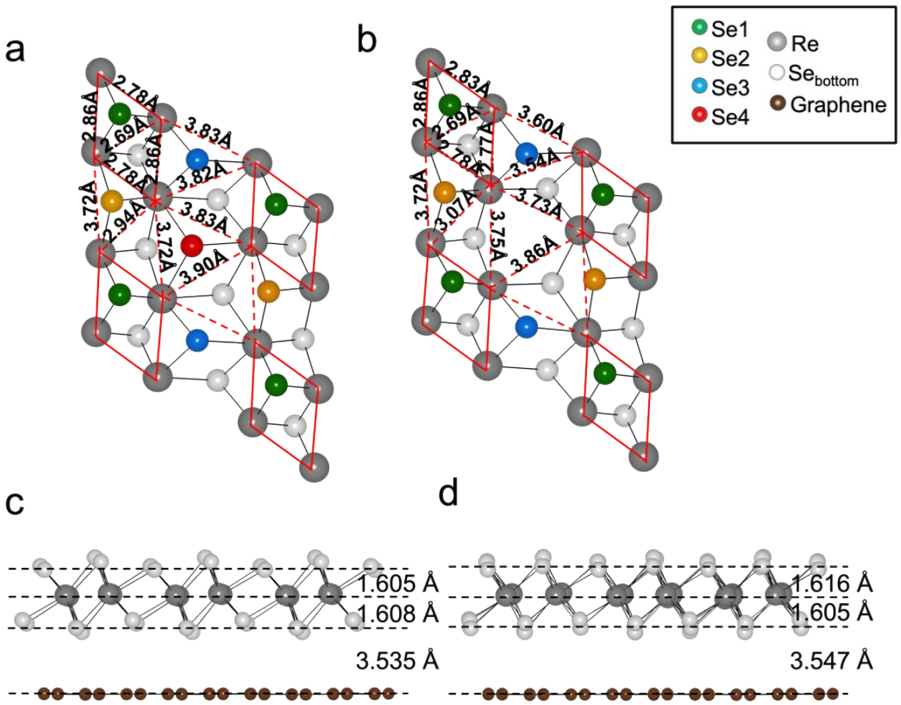

Figure S1. Top-view: Atomic details for (a) pristine case of $ReSe_2$/graphene and (b) V_{Se4} case of $ReSe_2$/graphene. Side-view: Vertical distances of the upper Se, Re, lower Se, and graphene layers for (c) pristine case of $ReSe_2$/graphene and (d) V_{Se4} case of $ReSe_2$/graphene.

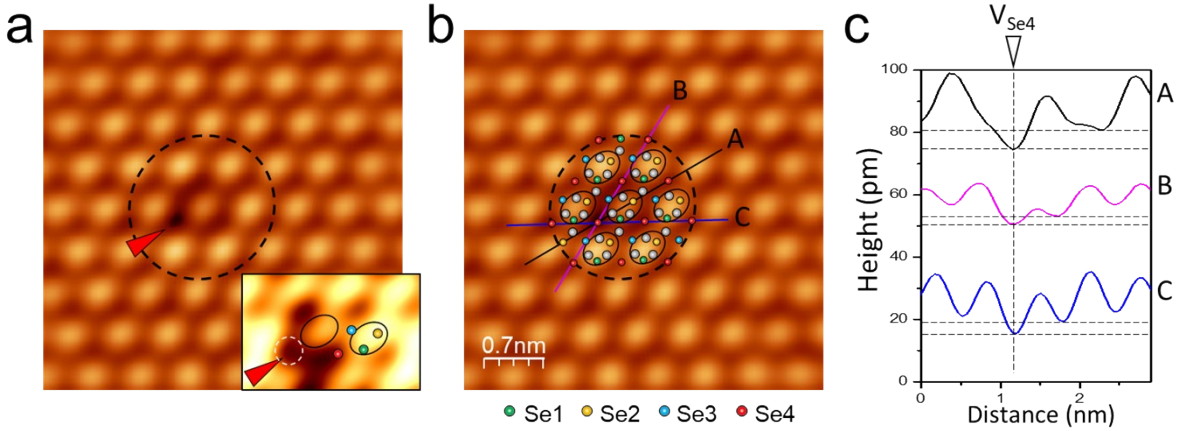

Figure S2. (a) The atomic STM image of a Se4 vacancy. The missing Se4 atom site (Se4 vacancy) is indicated by a red triangle. The inset in (a) is a DFT simulation image of a Se4 vacancy. (b) The

model structure of $ReSe_2$ is superimposed on (a). (c) Line profiles crossing the Se4 vacancy. A strong depression at one Se4 site implies the missing of a Se4 atom.

In Figure S2a, an atomistic image of a defect region clearly displays a prominent dark hole defect (marked by a red triangle). By comparing STM topography with the model structure used in DFT simulated images, as illustrated in Figure S2b, we have successfully identified the presence of a Se4 vacancy close to the Re layer. Furthermore, line profiles crossing the defect region, as depicted in Figure S2c, clearly exhibit a pronounced depression at the Se4 site, indicating the absence of a Se4 atom (i.e., the Se4 vacancy). This observation is further supported by the DFT-simulated STM image of a Se4 vacancy, which accurately reproduces the hole-like feature observed in the STM image (as shown in the inset of Figure S2a).

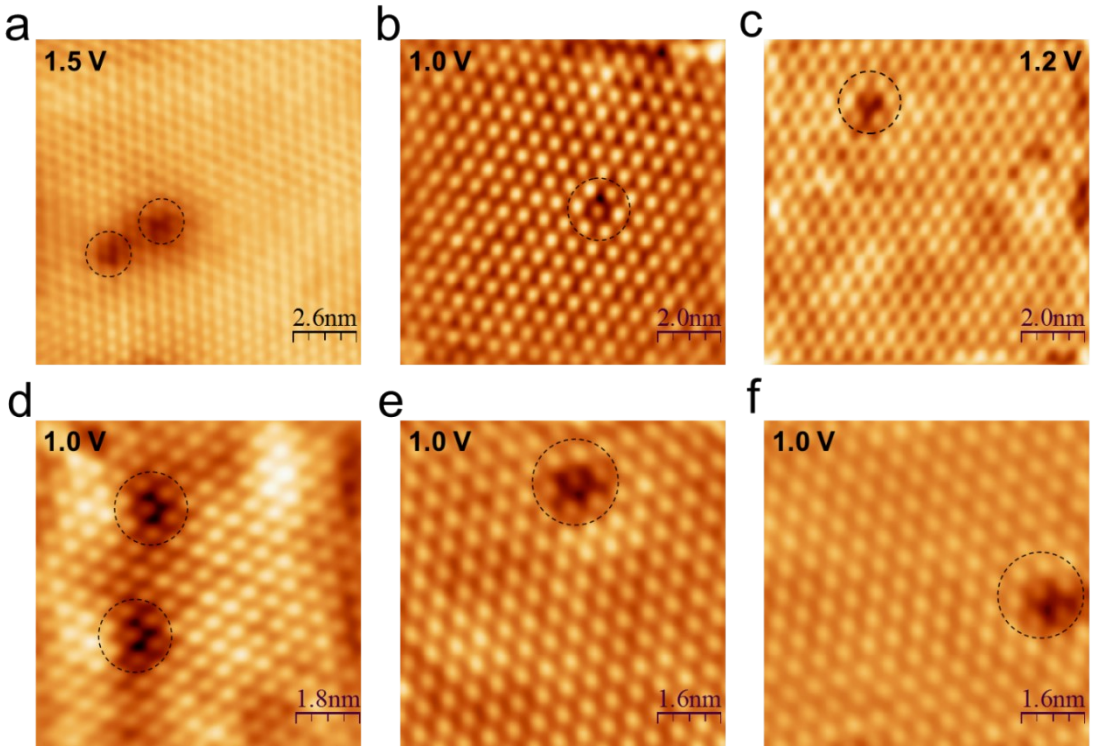

Figure S3. The STM images of Se4 vacancy in ML $ReSe_2$ on graphene substrate.

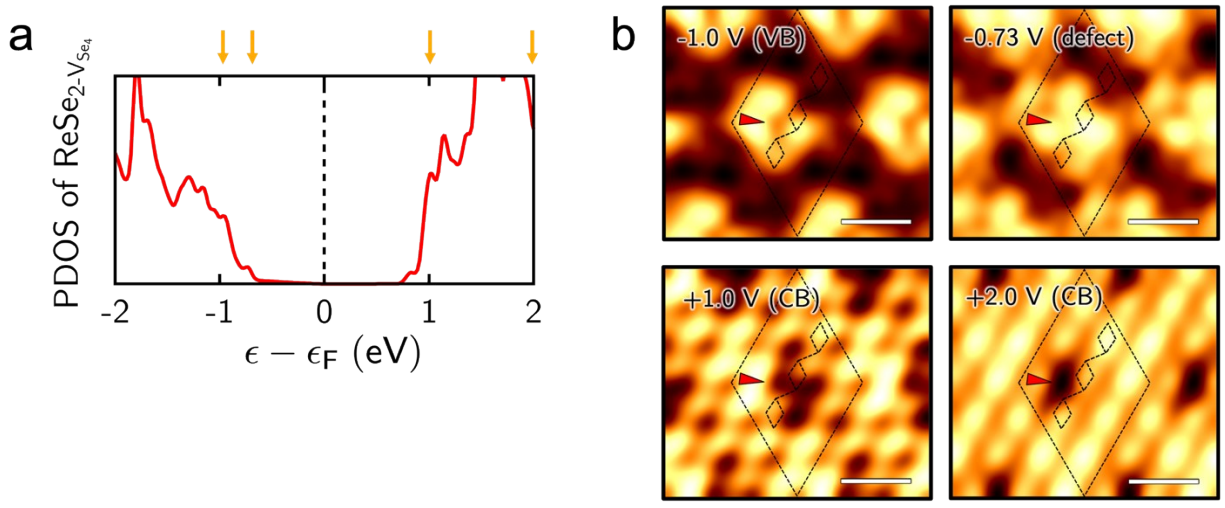

Figure S4. Voltage-dependent simulated STM images of Se4 vacancy in $ReSe_2$/graphene heterostructure. (a) The projected density-of-states (pDOS), which is aligned to the Fermi energy (ϵ_F) is shown. Orange vertical down-pointing arrows are used to indicate the specific energy level where the STM image of (b) is simulated. White color bars in the bottom right corner of the STM images indicate the 1 nm scale. The diamond-shaped motifs in each image depict the positions of the Re atoms (cf. Figure 1). The red filled arrow is used to denote the Se4 vacancy site.

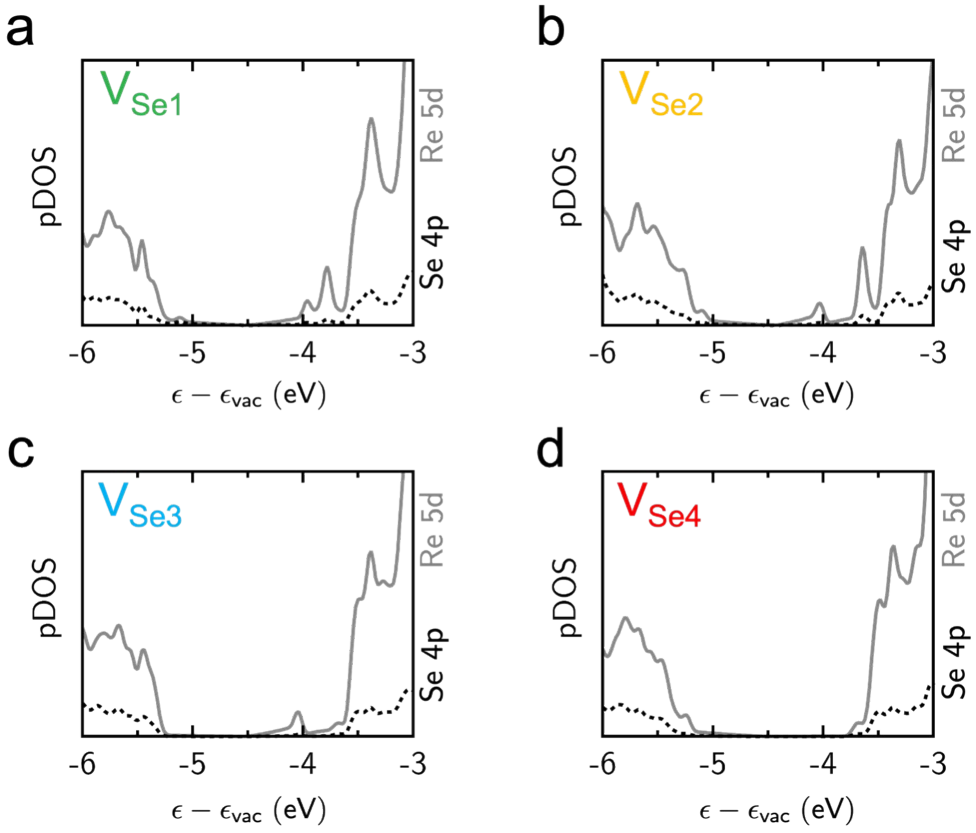

Figure S5. HSE06 partial density-of-states (pDOS) for (a) V_{Se1}, (b) V_{Se2}, (c) V_{Se3}, and (d) V_{Se4} cases in $ReSe_2$/graphene. The solid gray and dashed black curves correspond to the pDOS of Re and Se atoms, respectively.

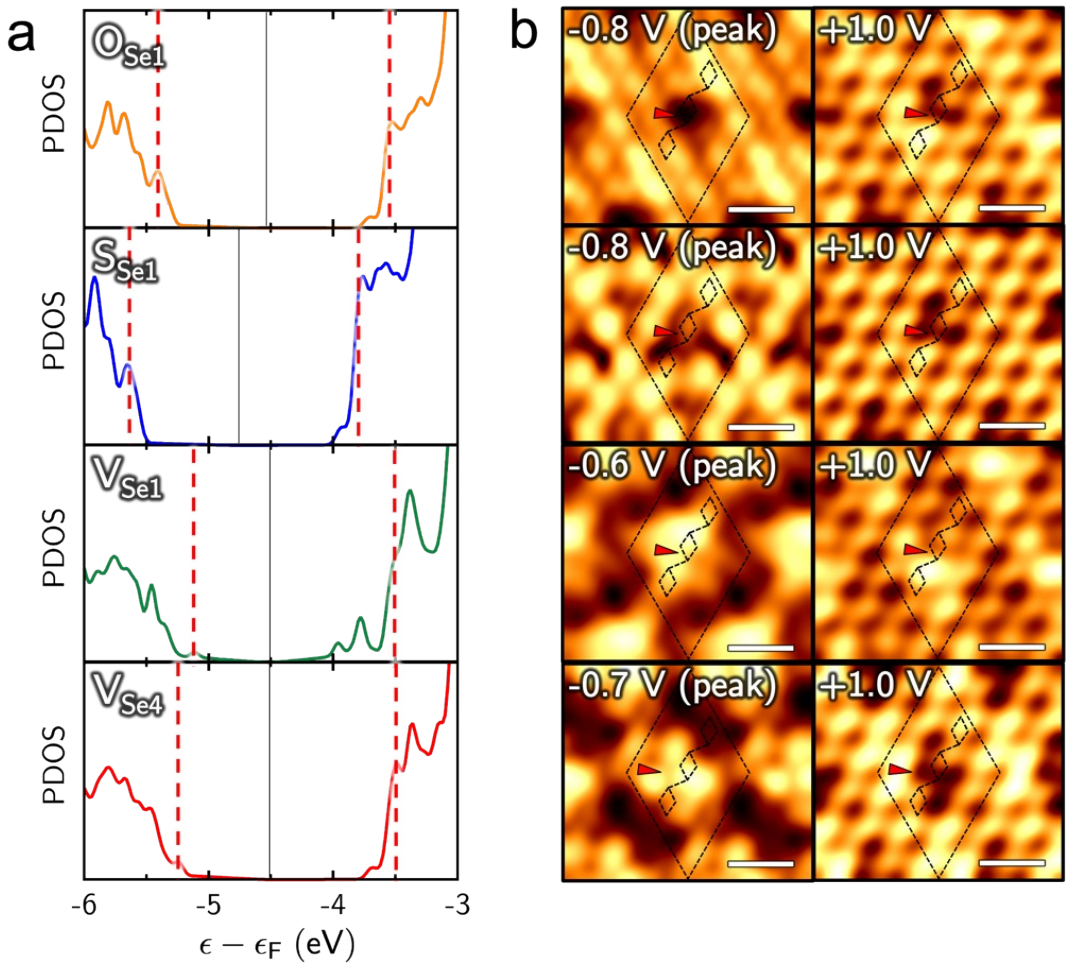

Figure S6. (a) Projected density-of-states (pDOS) for $ReSe_2$ layer of $ReSe_2$/graphene, aligned to the vacuum level (ϵ_{vac}). Each vertical solid grey line denotes the Fermi level. From the top panel, the DOS plot for the oxygen substitution at Se1, sulfur substitution at Se1, Se1 vacancy, Se4 vacancy, are shown. (b) The corresponding STM images for each system in (a) are displayed. Using vertical dashed red lines in (a) to indicate the chosen energy levels, on the left, we show the STM image of the energy level (denoted by the first peak in the valence band) while on the right, the STM image at +1.0 eV in the conduction band is shown accordingly. White color bars in the bottom right corner of the STM images indicate the 1 nm scale. The diamond-shaped motifs in

each image depict the positions of the Re atoms (cf. Figure 1). The red filled arrow is used to denote the corresponding defect sites.

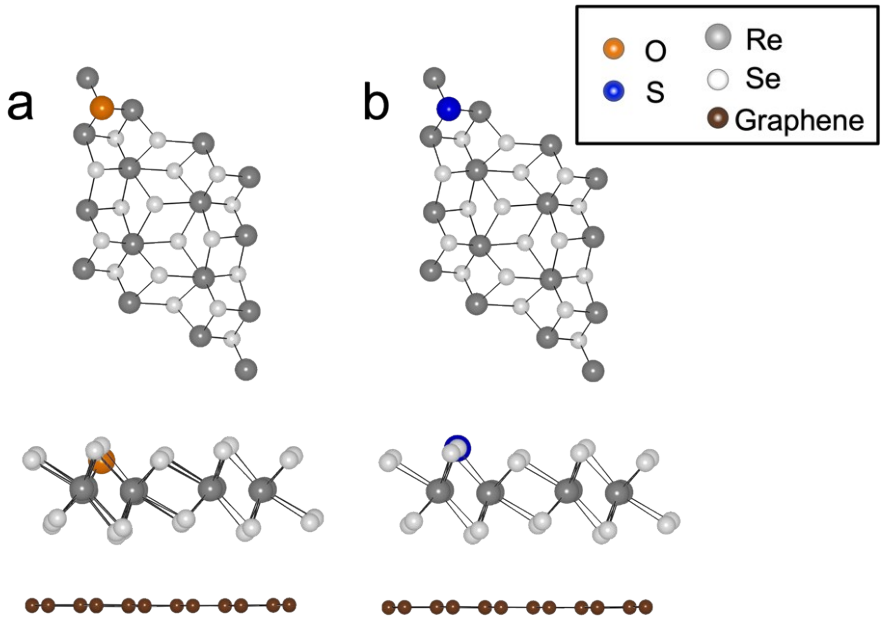

Figure S7. Atomic structures of the (a) oxygen substituted Se1 and (b) sulfur substituted Se1 sites in $ReSe_2$/graphene. The top panel shows the top-view of only the substituted $ReSe_2$ layers while the lower panel shows its side-view. The legend for labelling the atomic species are provided in the top-right hand corner of this figure.

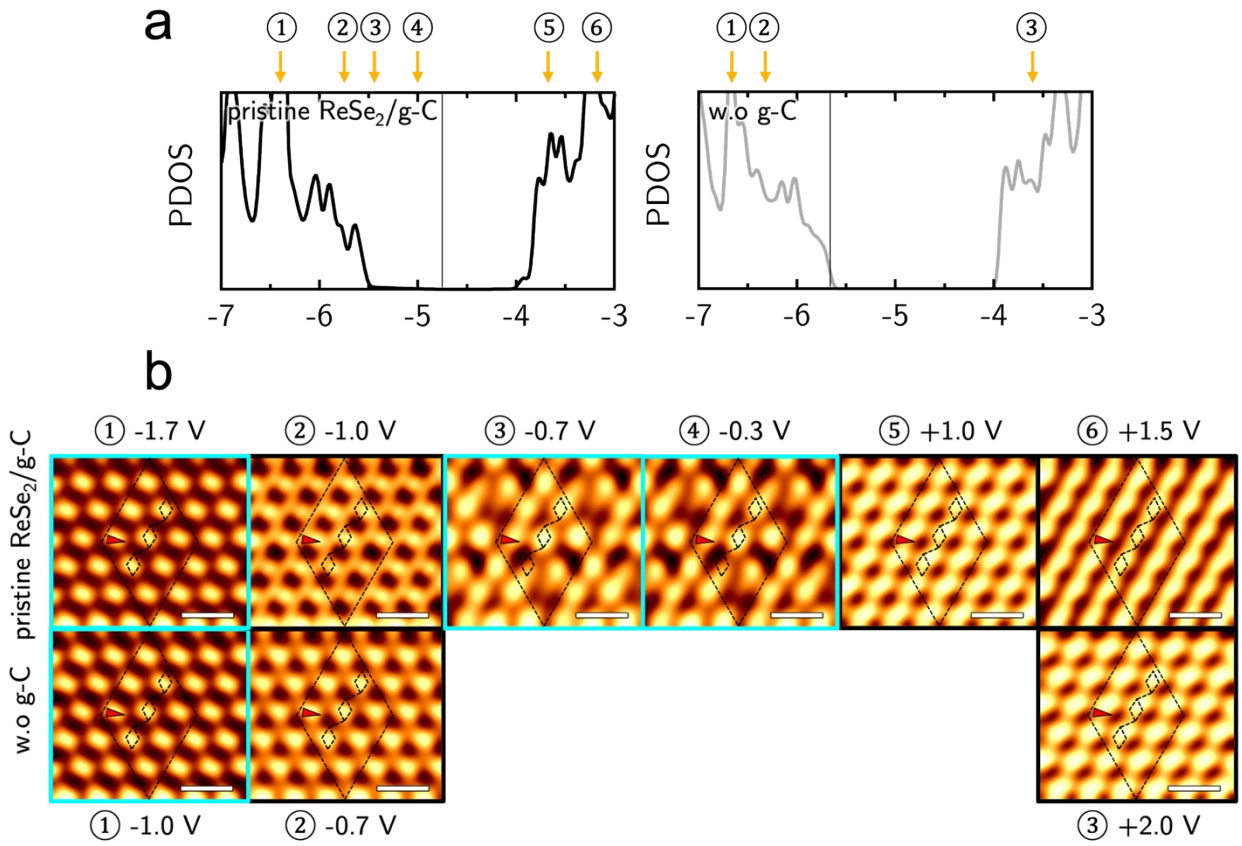

Figure S8. HSE06-derived pDOS (in (a) and aligned to the vacuum level (ϵ_{vac})) and its corresponding STM images (in (b)) for the defect-free (pristine) $ReSe_2$/graphene (where its pDOS is shown in (a) with the black solid line and the vertical line indicating the Fermi level) and the constrained $ReSe_2$ layer with the graphene support removed (where its pDOS is shown in (a) with the grey solid line and the vertical line indicating the valence band maximum). Orange vertical down-pointing arrows are used to indicate the specific energy level (numerically labelled) where the STM image of (a) is simulated.