Electronic Supplementary Information

1. Stability of GR/MoSe₂ heterobilayer proven via MD simulation

To prove the stability of our system, after system reaches 300 K in the NVT ensemble, we plot the temperature of GR, MoSe₂ and overall bilayer system as a function of simulation time, during the following 200 ps relaxation process in NVE ensemble. As shown in Fig. S1, the temperature of all layers converge at 300 K, which means the system is thermally stabilized.



Figure S1. Temperature evolutions of GR, $MoSe_2$ and the heterobilayer in a $20.2 \times 11.7 \text{ nm}^2$ ($x \times y$) system, during 200 ps NVE classical MD simulation process. The red solid line represents temperature 300 K.

The radial distribution function (RDF) of both supported and suspended GR and MoSe₂ layers are calculated to verify the structural stability of the bilayer system. The results are summarized in Fig. S2. The RDFs for suspended and supported GR layers soundly match each other, indicating that the existence of MoSe₂ layer does not affect the stability of GR lattice structure. The C-C bond length of GR layer is 1.42 Å for both cases. The first two peaks in the MoSe₂ RDF represent the average distance of Mo atom to adjacent Se and Mo atoms, respectively. In this case, the suspended and supported RDFs present a slight displacement due to the lattice mismatch. Besides the effect of lattice mismatch, the structure of supported GR and MoSe₂ layers stay non-deformed compared with suspended layers.



Figure S2. Radial distribution functions in supported and suspended GR and MoSe₂ layers.

2. Stability of GR/MoSe₂ heterobilayer proved by AIMD simulation

To further prove the stability of the bilayer structure, we performed *ab initio* molecular dynamics (AIMD) simulation using Vienna *ab initio* Simulation Package (VASP)², implementing density functional theory (DFT) calculations. The Perdew–Burke–Ernzerhof (PBE)³ of generalized gradient approximation (GGA) and vdW-corrected DFT approach are used as electron exchange-correlation functional. The energy cutoff for the plane-wave basis is set to be 520 eV. Before the AIMD simulation, geometry optimization is performed to the rectangular unitcell with 118 atoms based on Monkhorst-Pack⁴ *k*-mesh of $5 \times 5 \times 1$ for the summation in the first Brillouin Zone (BZ) and a maximum residual force of 10^{-2} eV/Å. The electron total energy self-consistent threshold is set as 10^{-4} eV. The vacuum spacing in the *z* direction is greater than 15 Å to avoid the interactions between periodic layers. Once bilayer structure is optimized, a Monkhorst-Pack *k*-mesh of $1 \times 1 \times 1$ is used for AIMD simulation to save computational cost. The system is putted into NVT ensemble at 800 K for 10 ps. The configurations before and after AIMD simulation are shown in Fig S3. As shown in Figs. S3, the bilayer structure is thermal dynamically stable at temperature as high as 800 K.



Figure S3. Configurations of GR-MoSe₂ bilayer (a) Top and (b) side views after geometry optimization; (c) Top and (d) side views after 10 ps NVT AIMD simulation at 800 K.

Energy evolution during the AIMD simulation are presented in Fig. S4. One can see that the energy is converged at the end of simulation period.



Figure S4. Energy evolution of GR-MoSe₂ bilayer during 10 ps NVT AIMD simulation at 800 K.

Supplementary References

- 1. G. Kresse and J. Furthmüller, *Physical Review B*, 1996, 54, 11169-11186.
- 2. J. P. Perdew, K. Burke and M. Ernzerhof, *Physical Review Letters*, 1996, 77, 3865-3868.
- 3. H. J. Monkhorst and J. D. Pack, *Physical Review B*, 1976, 13, 5188-5192.