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Supplementary Information

Band gap controlling and transformation of monolayer MoS_2-based hetero-bilayers

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I: Crystal structure of MoS_2 bulk.

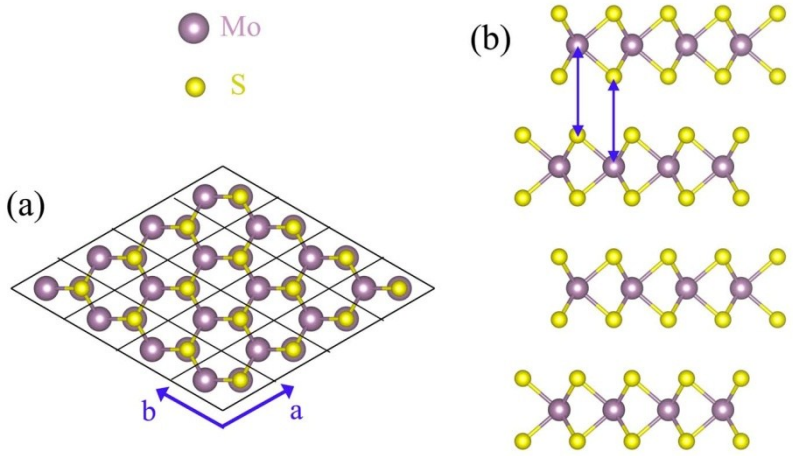

Fig. S1. (Color online). (a) Top and (b) side views of the MoS_2 bulk. The primitive vectors are marked with a and b.

II: How to find optimal structures for free-standing hetero-bilayers?

For bilayer systems, there are two important structure parameters need to be considered: the lattice constant (a) and the interlayer distance (d). The electronic properties of bilayer systems are sensitive to these two factors. Thus, it is very important to find their optimum

values. (1) At first, we take the lattice parameter of 5×5 MoS_2 monolayer $a' = 5a_{MoS_2}$ as the reference value. And around this reference value, we choose 5 different lattice constants ($0.95a'$, $0.97a'$, $1.00a'$, $1.03a'$, $1.05a'$). For each lattice constant, all the atomic positions of the structure were relaxed with a force tolerance of 10^{-2} eV/Å. We can get an E-a curve. And the optimized lattice constant can be gotten by using data fitting. At last, we built an initial structure based on the optimized lattice constant and relaxed it again to get the interlayer distance and also the corresponding total energies of each hetero-bilayer system.

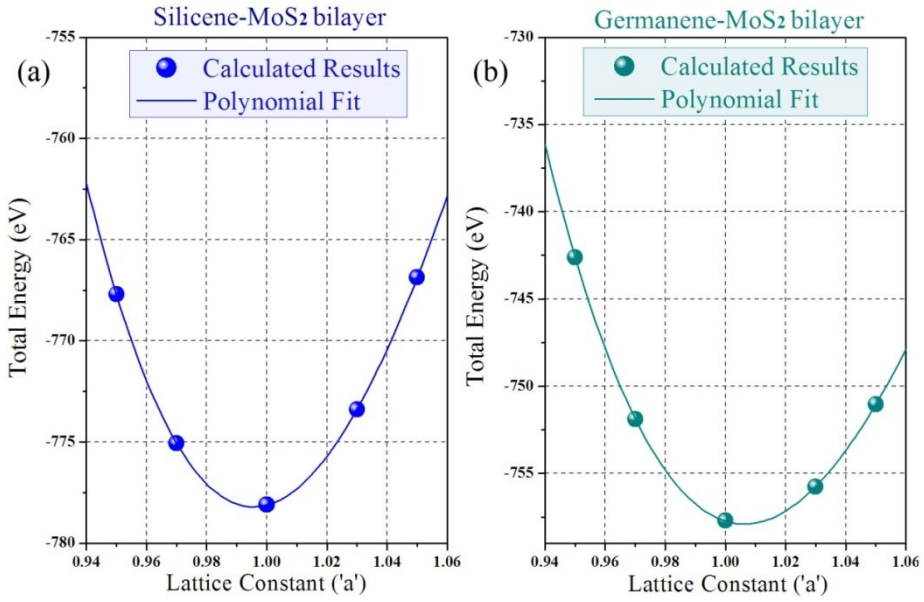

Fig. S2. (Color online). Total energies of hetero-bilayer systems as a function of the lattice constant.

III: The buckling values of silicene and germanene in hetero-bilayers.

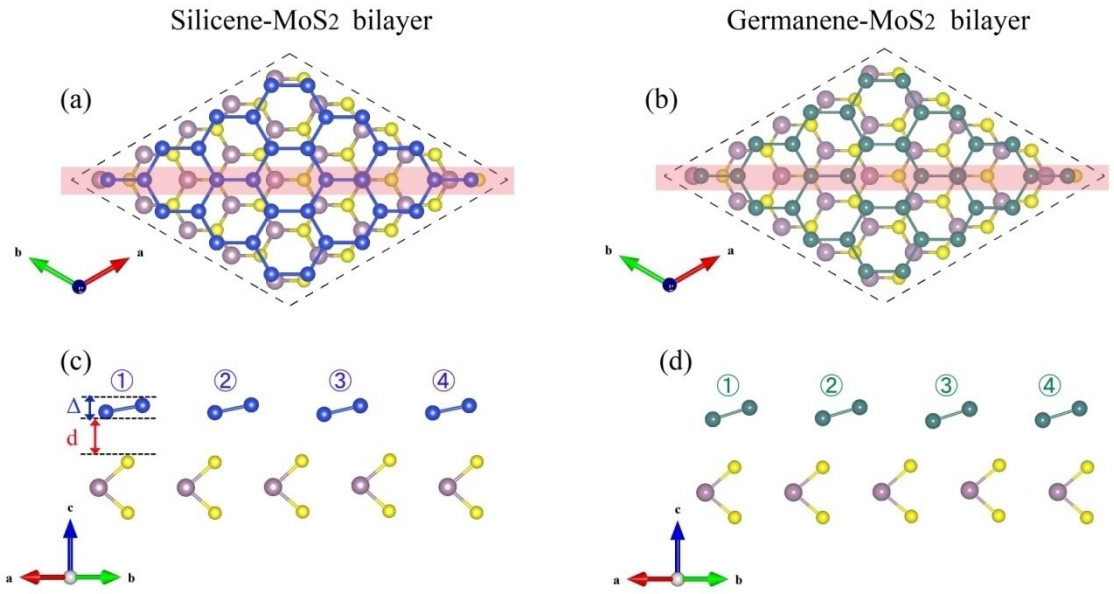

Fig. S3. (Color online). (a) and (b) are top views of Silicene-MoS_2 and Germanene-MoS_2 bilayers, respectively. (c) and (d) are side views of the red regions of corresponding hetero-bilayers.

Table S1. Buckling height (Δ) and interlayer distance (d) of different areas shown in Fig. S3.

Bilayer System	Parameter	①	②	③	④
Silicene-MoS_2	Δ(Å)	0.455	0.451	0.527	0.474
	d(Å)	3.050	3.056	2.866	3.015
Germanene- MoS_2	Δ(Å)	0.715	0.678	0.744	0.717
	d(Å)	2.953	3.028	2.776	2.902

IV: The atomic bond lengths of Mo-S within the hetero-bilayers.

As shown in Fig. S4, within each single layer MoS_2, one Mo atom layer sandwiched between two S layers. Then we pick a hexagonal unit (highlighted in blue shades) from the monolayer MoS_2. As shown in Fig. S4 (c), the lattice constant (a_{MoS_2}) is the in-plane distance between two S atoms. In order to analyze other configuration parameters, we further pick a

triangular unit (highlighted in red shades) from the hexagonal unit (Fig. S4 (e)). For the right-angled triangle, we have $m^2 + n^2 = s^2$, where s is the length of Mo-S bond ($s = d_{Mo-S}$), n is half the sheet thickness of monolayer MoS_2 ($n = \frac{1}{2}h_{S-S}$), m increases with an increasing lattice constant ($m = \frac{\sqrt{3}}{3}a_{MoS2}$). The lattice constant ($5a_{MoS2}$) of Silicene-MoS_2 is smaller than that of Germanene-MoS_2, while the sheet thickness (h_{S-S}) of Silicene-MoS_2 is bigger than that of Germanene-MoS_2, resulting in the "almost same" bond lengths of Mo-S in the MoS_2 sheets with different strains.

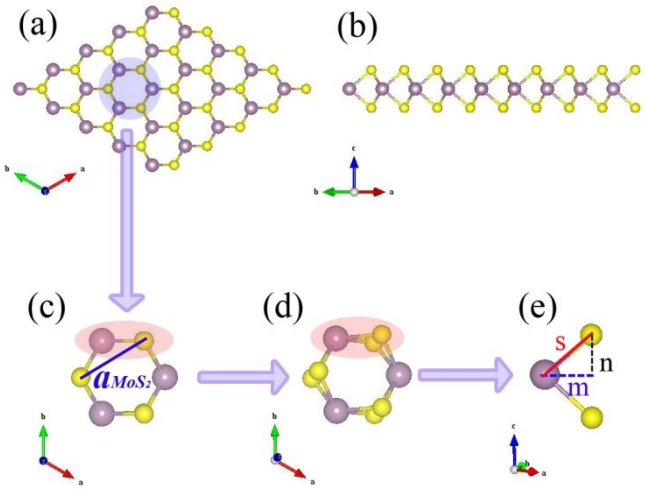

Fig. S4. (Color online). Configurations of monolayer MoS_2. (a) Top and (b) side views of monolayer MoS_2. (c) and (d): top view of single hexagonal unit (consist of 3 Mo atoms and 6 S atoms). (e) side view of single triangular unit (consist of 1 Mo atoms and 2 S atoms). Mo and S atoms are shown in pink and yellow circles, respectively.