Supplemental Material

Biaxial strain modulated electronic structure of layered two-dimensional $MoSiGeN_4$ Rashba systems

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(a) The change of Geometry Structures under biaxial strain

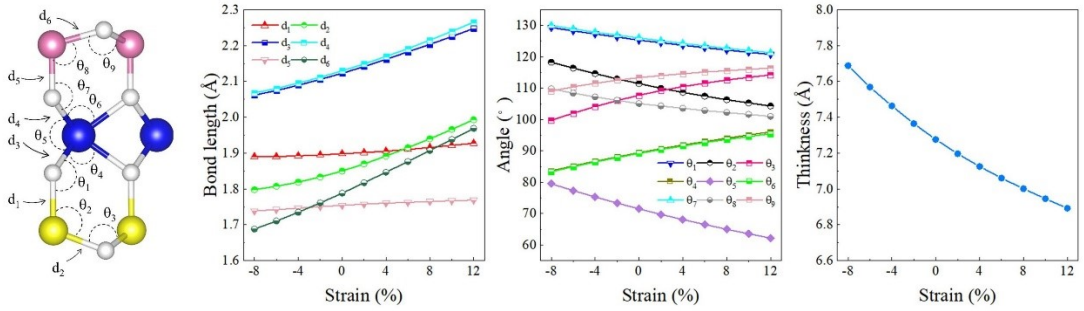

Fig. S1. The schematic diagram of structural parameters and the changes of band length, band angle and thickness under biaxial strains. The thickness and θ_1, θ_2, θ_5, θ_7, θ_8 of $MoSiGeN_4$ increases with the decrease of compressive strain and decreases as tensile strain increase, a trend opposite to that of bonds length and θ_3, θ_4, θ_6, θ_9.

(b) Band structure of $MoSi_2N_4$ and $MoSiGeN_4$ by using HSE and PBE

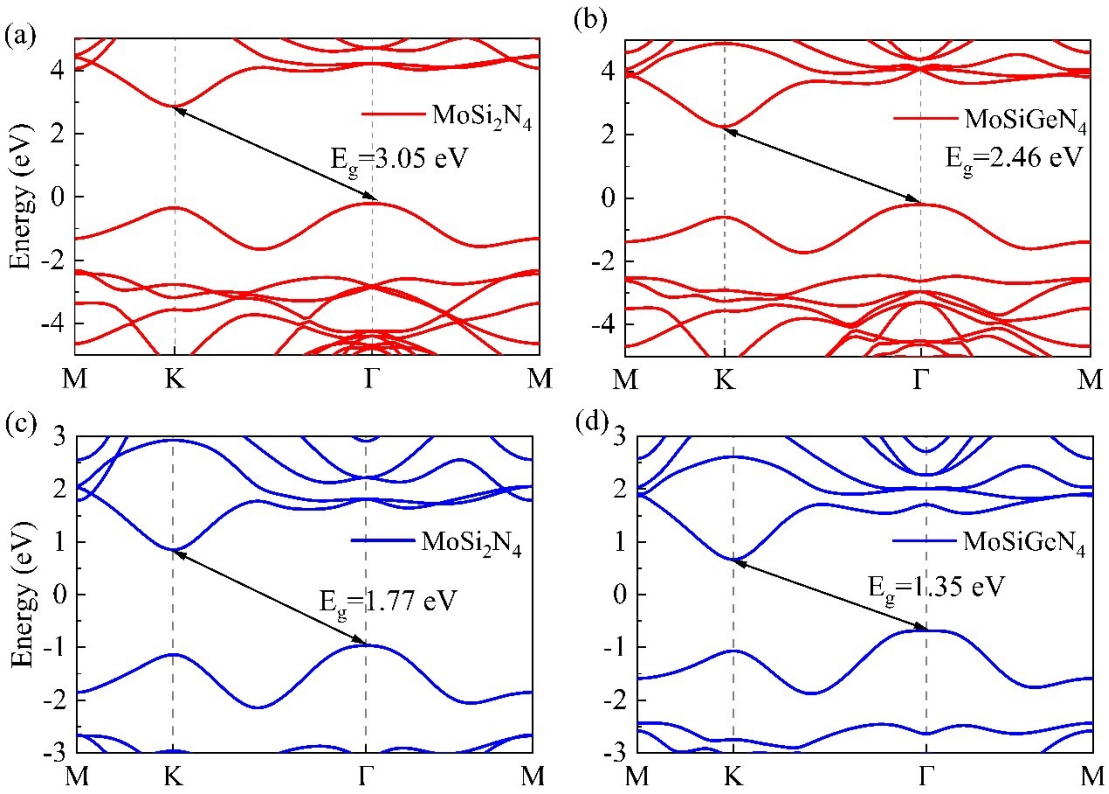

Fig. S2. The band structure of $MoSi_2N_4$ (a) and $MoSiGeN_4$ (b) by using HSE functional (red line). The band structure of $MoSi_2N_4$ (c) and $MoSiGeN_4$ (d)by using PBE functional (blue line).

(c) The Geometry Structures of bilayer $MoSiGeN_4$

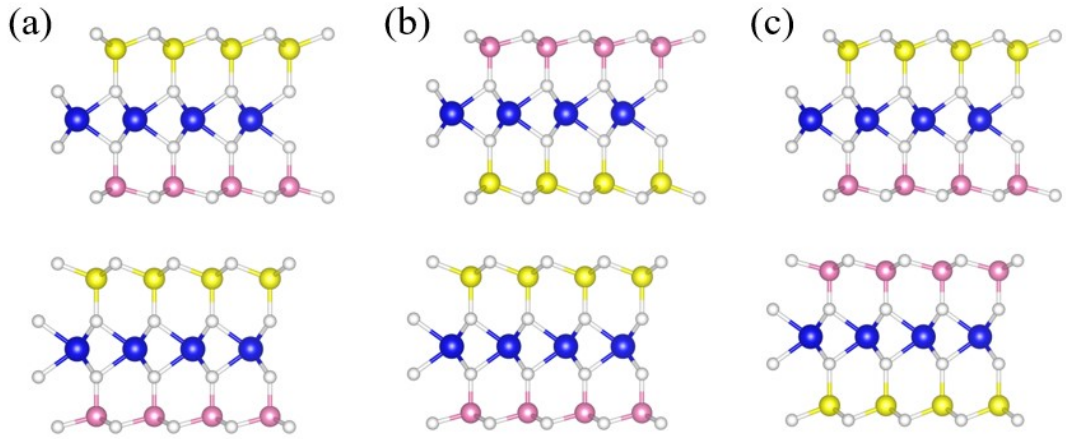

Fig. S3. The three stacking structures of (a) Si_{top}-Ge_{bot} (b) Ge_{top}-Ge_{bot} and (c) Si_{top}-Si_{bot} for bilayer $MoSiGeN_4$.

(d)Ab-initio Molecular Dynamics

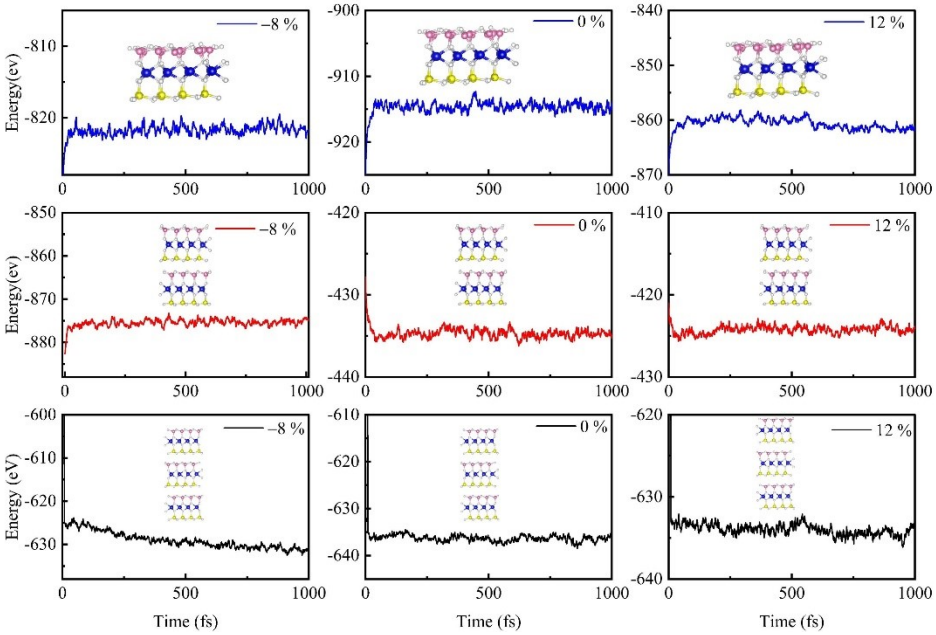

Fig. S4. The ab initio molecular dynamics (AIMD) of $MoSiGeN_4$ monolayer (blue line), bilayer (red line) and trilayer (black line) under different strains (−8%, 0% and 12%).

(e) The orbital-projected band structure of $MoSiGeN_4$ monolayer

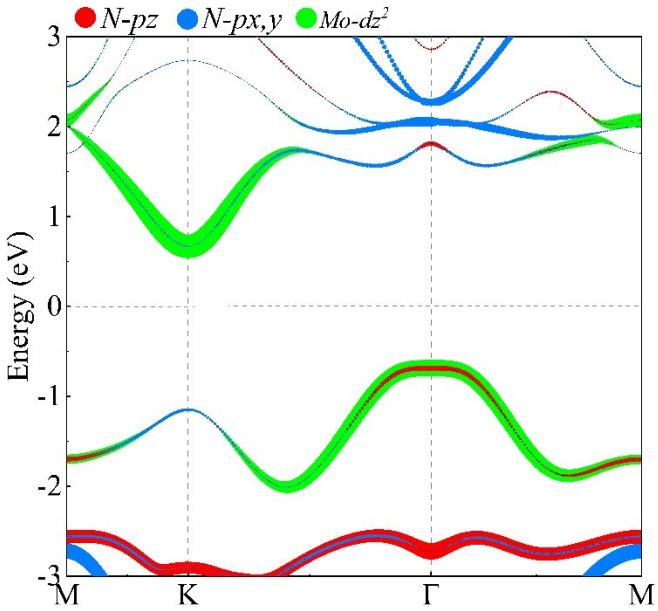

Fig. S5. The orbital-projected band structures of $MoSiGeN_4$ monolayer. The color of red, blue and green are the contribution of N-*p*, N-$p_{x,y}$ and Mo-dz^2, respectively.

(f) The orbital-projected band structures of $MoSiGeN_4$ mutilayer

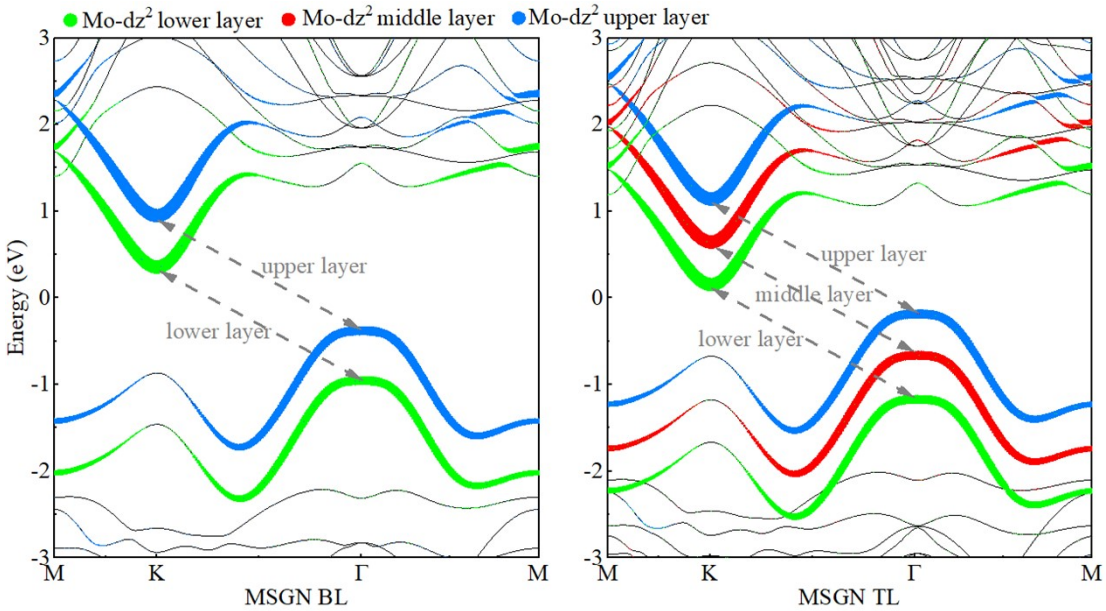

Fig. S6. The orbital-projected band structures of $MoSiGeN_4$ bilayer (MSGN BL) and $MoSiGeN_4$ trilayer (MSGN TL). The color of green, red and blue are the contribution of Mo-dz^2 in the upper layer, middle layer and the lower layer, respectively.

(g) The band structure of trilayer $MoSiGeN_4$

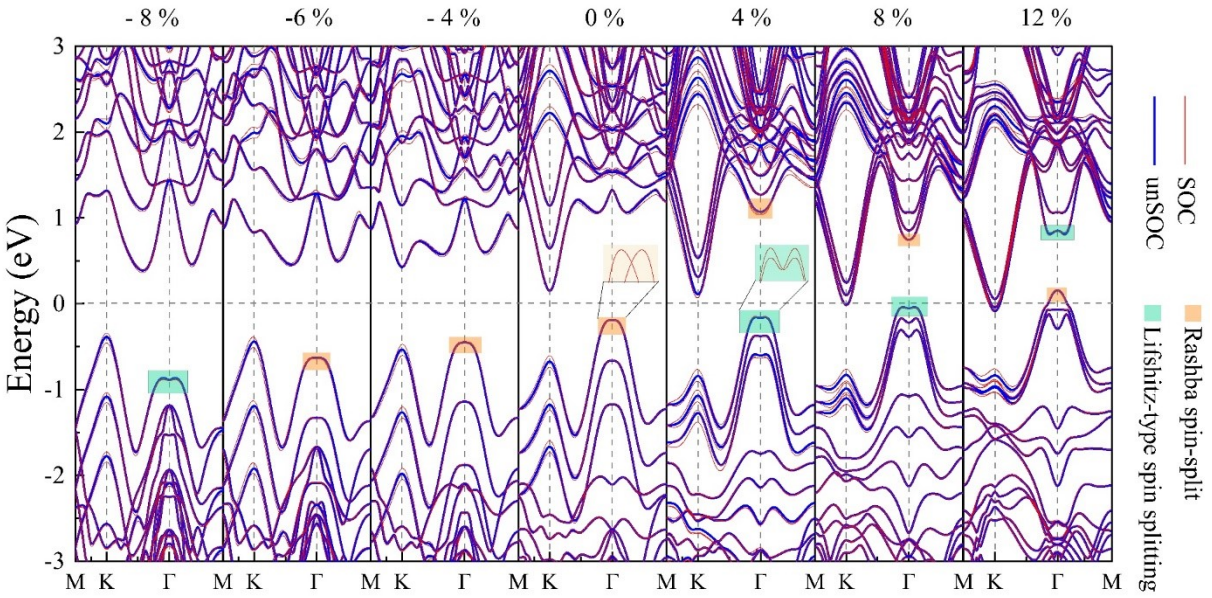

Fig. S7. Band structure of trilayer $MoSiGeN_4$ under biaxial strains considering without (blue lines) and with SOC (red lines). The enlarged drawing of Rashba spin splitting (orange patch) and Lifshitz-type spin splitting (green patch) are shown in the inset.

(h) The dipole momemt and electronic structures of $MoSiGeN_4$ monolayer and mutilayer

Table SI: The dipole moment P, bang gap E_g, Rashba coefficient α_R, Mexican coefficient M, L coefficient $E_{1\text{-}2}$, $E_{2\text{-}3}$, k_f and k_L of $MoSiGeN_4$ monolayer, bilayer and trilayer. Dip. denotes include dipole corrections in calculations with VASP.

Structure	Monolayer		Bilayer		Trilayer	
Strain	0 %	8 %	0 %	8 %	0 %	8 %
P (e×Å)	0.026	0.009	0.054	0.021	0.064	0.027
E_g (eV)	1.35 (Dip.) 1.35	0.29(Dip.) 0.30	0.64 (Dip.) 0.76	0.005(Dip) 0.01	0.48(Dip.) 0.38	0.003 0.001
M (eVÅ)	--	0.87(Dip.) 0.68	--	0.30(Dip.) 0.52	--	0.24(Dip.) 0.39
α_R (meVÅ)	63.025 (Dip.) 61.073	--	35.026 (Dip.) 37.424	--	24.395(Dip.) 19.968	--
$E_{1\text{-}2}$	--	0.006(Dip.) 0.006	--	0.007(Dip.) 0.007	--	0.006(Dip.) 0.006
$E_{2\text{-}3}$	--	0.17(Dip.) 0.13	--	0.038(Dip.) 0.035	--	0.006(Dip.) 0.007
k_f	--	0.02(Dip.) 0.02	--	0.04(Dip.) 0.04	--	0.03(Dip.) 0.03
k_L	--	0.20(Dip.) 0.20	--	0.15(Dip.) 0.14	--	0.12(Dip.) 0.12

(i) The Rashba constants of $MoSiGeN_4$ monolayer and mutilayer

Table SII: The k_0, E_R, α_R and position of Rashba effect in $MoSiGeN_4$ monolayer, bilayer and trilayer under biaxial strains. V_1 denotes the highest valence band at Γ point, and C_1 denotes the lowest conduction band at Γ point.

Structure	*Strain (%)*	*Position* (Γ)	k_0 (Å^{-1})	E_R (meV)	α_R (meVÅ)
Monolayer	-6	V_1	0.150	4.378	58.380
	-4	V_1	0.070	1.700	48.220
	-2	V_1	0.043	0.710	32.302
	0	V_1	0.070	2.206	63.025
	2	C_1	0.036	2.360	13.038
	4	C_1	0.019	2.800	28.448
	6	C_1	0.041	3.130	149.540
	8	C_1	0.102	16.500	324.450
Bilayer	-6	V_1	0.091	4.000	87.912
	-4	V_1	0.013	0.100	15.384
	-2	V_1	0.072	0.560	15.448
	0	V_1	0.024	0.420	35.026
	2	C_1	0.006	1.151	381.12
	4	C_1	0.018	1.423	240.37
	6	C_1	0.016	1.842	173.72
	8	C_1	0.016	9.598	327.15
Trilayer	-6	V_1	0.075	2.370	62.434
	-4	V_1	0.042	0.405	19.148
	-2	V_1	0.024	0.250	20.815
	0	V_1	0.062	0.627	24.395
	2	C_1	0.059	3.552	118.459
	4	C_1	0.017	1.307	154.583
	6	C_1	0.019	1.563	163.152
	8	C_1	0.093	15.108	324.449