Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Giant Magnetic Anisotropy of Adatoms on Graphane Surface

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1. Structure of 2√2 × 2√2 MgO supercell with Pb/Bi adsorbed

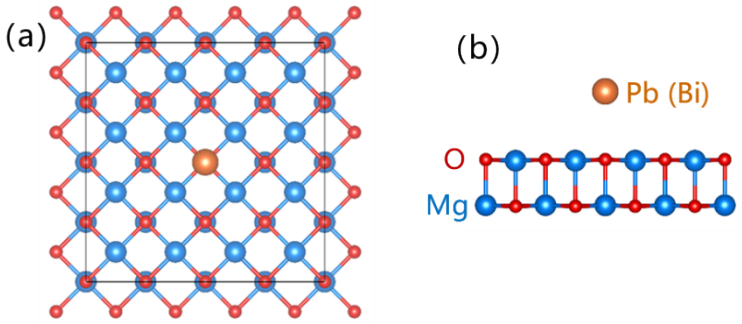

Figure S1. Schematic structure of 2√2 × 2√2 MgO supercell with Pb (Bi) adsorbed.

To examine the structural stabilities of above systems, the adsorption energy is calculated by $E_{ad} = -(E_{Total} - E_M - E_{MgO})$, where E_{Total} is the total energy of the system, E_M and E_{MgO} are the energies of the single Pb/Bi atom and MgO, respectively. The values 0.04/0.78 eV are obtained for Pb/Bi adatom, which means that the Pb adatom adsorption structure is susceptible to the external perturbations.

2. Calculated adsorption energy of M atom adsorbed on graphane

Table S1. Calculated adsorption energy of M atom adsorbed on graphane.

M	Bi	Pb	Sn	Re	Sb	Os	Ru	Co	Ge	Sc
E_{ad} (eV)	1.88	2.00	2.20	2.14	2.07	2.12	3.20	2.42	2.51	1.54
M	Ti	V	Cr	Mn	Fe	Ni	Cu	Y	Zr	Nb
E_{ad} (eV)	0.61	0.98	0.63	0.72	0.91	1.74	1.48	2.19	2.10	1.47
M	Mo	Tc	Rh	Pd	Ag	Hf	Ta	W	Ir	Pt
E_{ad} (eV)	2.43	1.37	1.97	1.54	1.36	2.64	2.48	2.38	2.81	3.10
M	Au	Zn	Ga	As	Cd	In	Hg	Tl	Li	Na
E_{ad} (eV)	2.57	0.71	2.63	2.41	0.54	2.30	0.49	2.11	1.84	1.38
M	K	Rb	Cs	Be	Mg	Ca	Sr	Ba	Se	Te
E_{ad} (eV)	1.27	1.23	1.31	2.31	1.11	1.65	1.46	1.80	2.94	2.46

The adsorption energies of M on graphane substrate were examined by the same method with E_{ad} = - ($E_{Total} - E_M - E_{graphane}$), where E_{Total} is the total energy of the system, E_M and $E_{graphane}$ are the energies of the single M atom and substrate graphane, respectively. The calculation results show that E_{ad} of those adatoms are in the range of 0.5 ~ 3.2 eV, indicating structural stability and possibility to synthesize and control in experiment.

3. Projected density of states

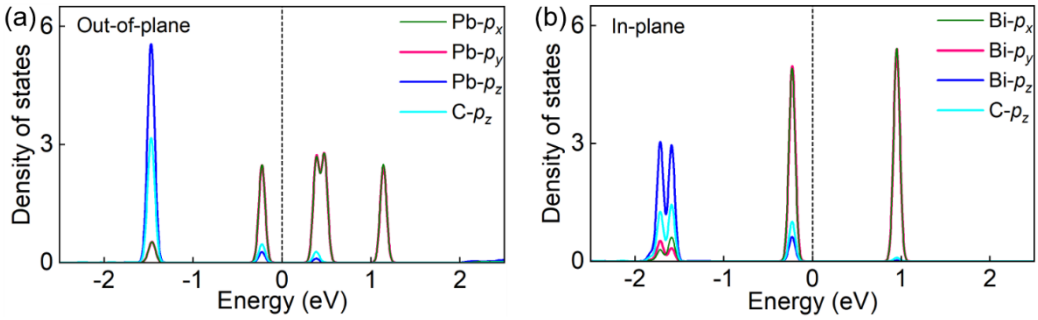

Figure S2. Projected density of states of (a) Pb and (b) Bi and C atom directly below, respectively.

4. Charge density difference

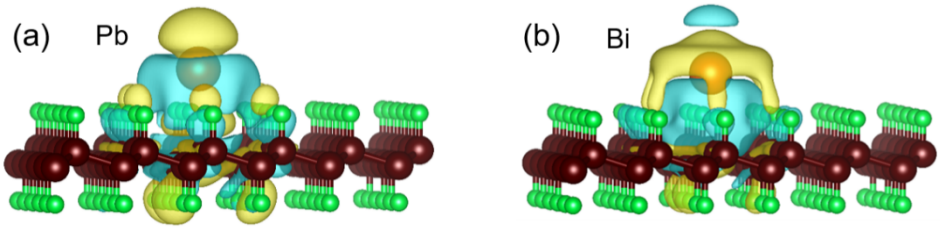

Figure S3. Charge density difference between single atom (a) Pb and (b) Bi and graphane substrate, where blue and yellow indicate electron depletion and accumulation, respectively. The isosurface value is taken as 0.0005 e/Bohr^3.

5. Geometric structures with different magnetic lattices

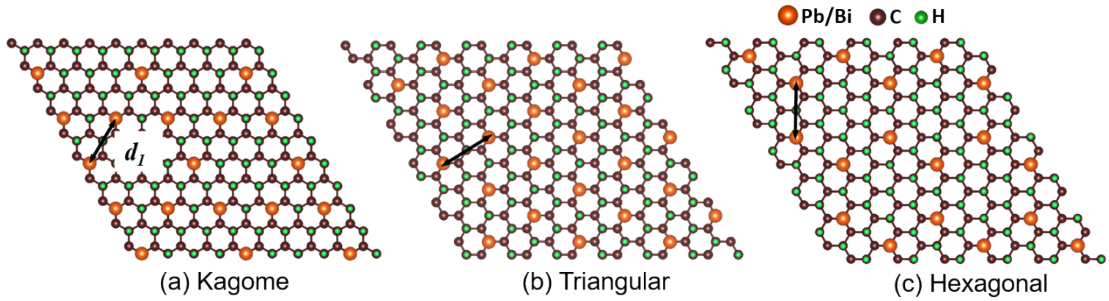

Figure S4. Structures with three different atomic magnetic lattices of Pb (Bi) adatoms, including (a) kagome, (b) triangular and (c) hexagonal lattice.

6. Electronic band structures of different magnetic lattices

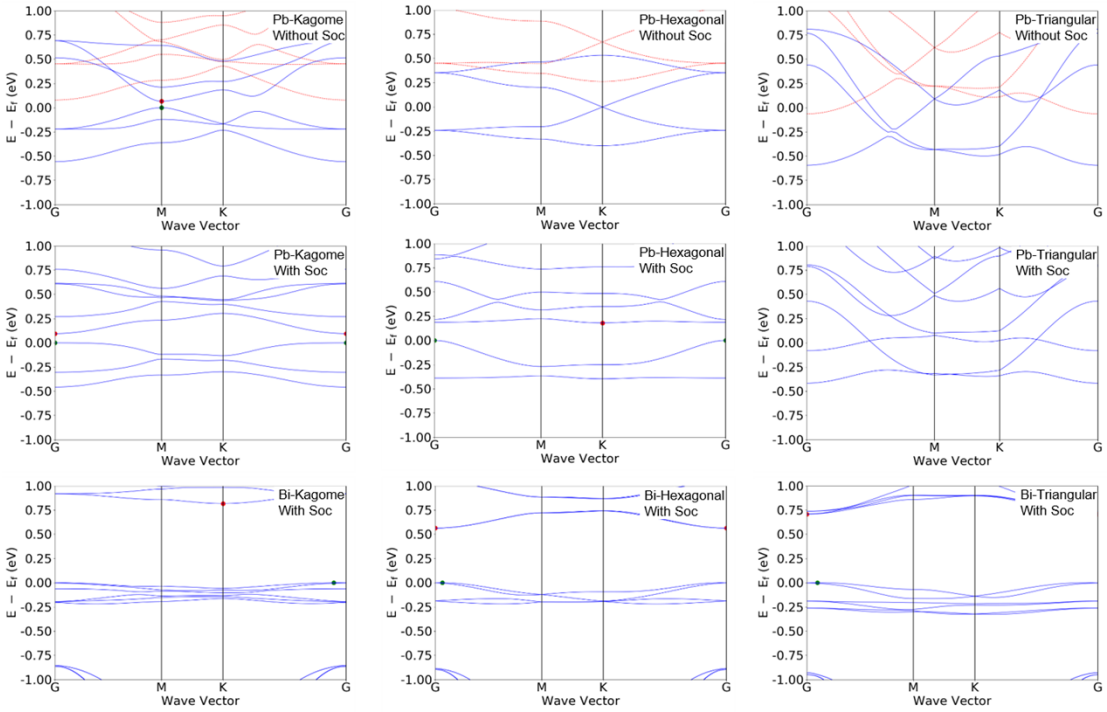

Figure S5. Electronic band structures at GGA and GGA+SOC level of three different Pb (Bi) magnetic lattices, respectively.

7. Calculated magnetic moment of Bi/Pb atom in different systems

Table S2. Calculated magnetic moment M (μ_B) of Bi/Pb atom in different systems, where H/K/T-lattice presents the hexagonal/kagome/triangular lattice, respectively.

	M (μ_B)					
	atom	adatom	metal	H-lattice	K-lattice	T-lattice
Bi	3	2	0	2	2	2
Pb	2	1	0	1	1	1

8. Projected density of states of Bi atom in different systems

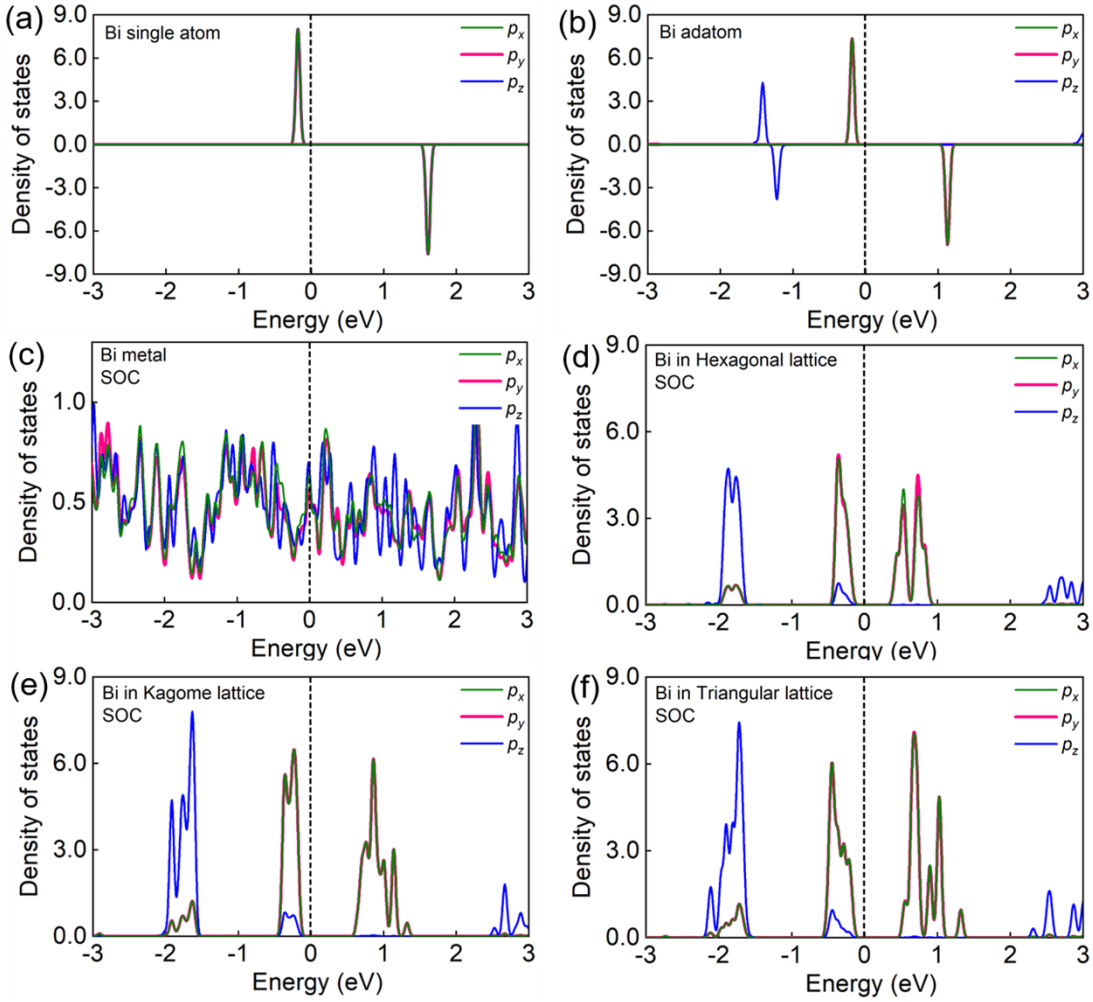

Figure S6. Project density of states of Bi atom in different systems, respectively.