Supplementary Information (SI) for Physical Chemistry Chemical Physics.

Supplementary Information for Biaxial Strain-Mediated Magnetic Phase Transition and Anisotropy Engineering in MoSBr Monolayer: A First-Principles Approach for Room-Temperature Spintronics

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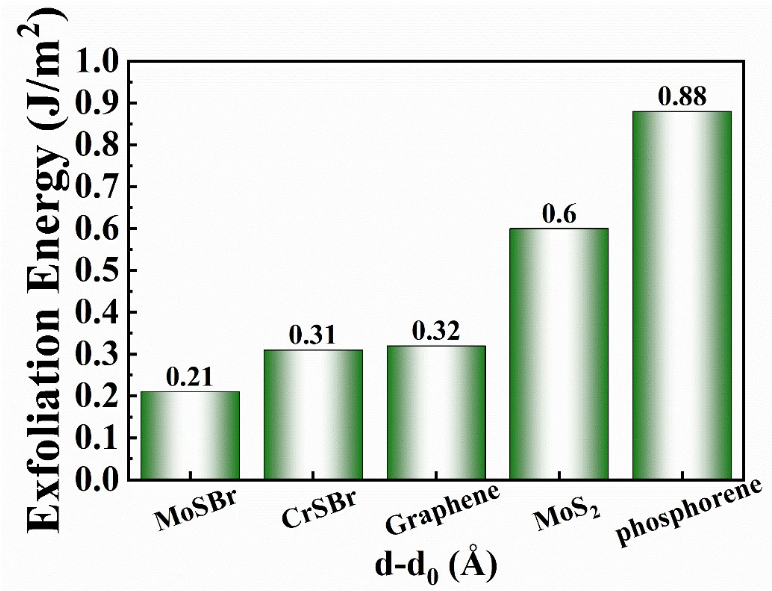

Fig. S1. Cleavage energies for the mechanically exfoliated MoSBr monolayers and in comparison with several other materials[66-69].

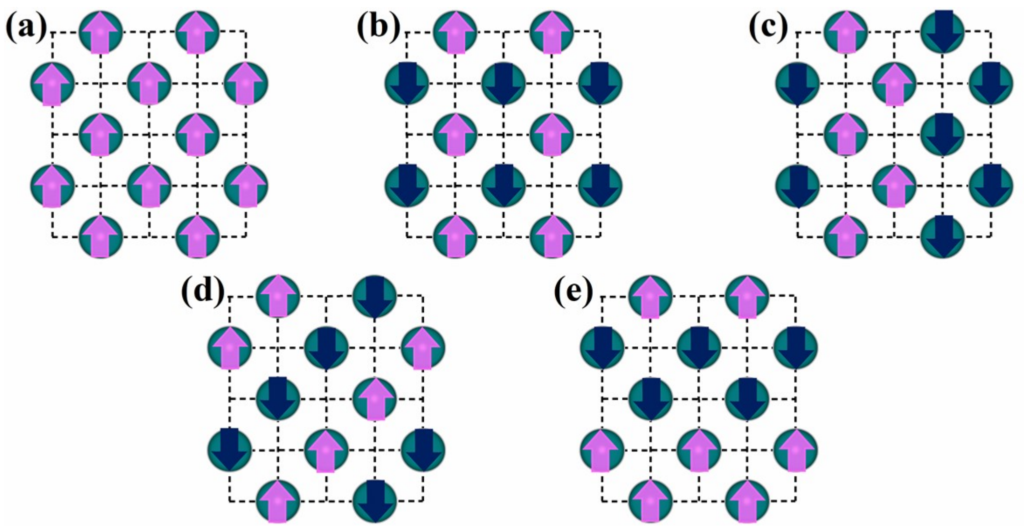

Fig. S2. The five possible magnetic configurations of the MoSBr monolayer: FM, AFM1, AFM2, AFM3, and AFM4. The purple (blue) arrows represent spin-up (spin-down).

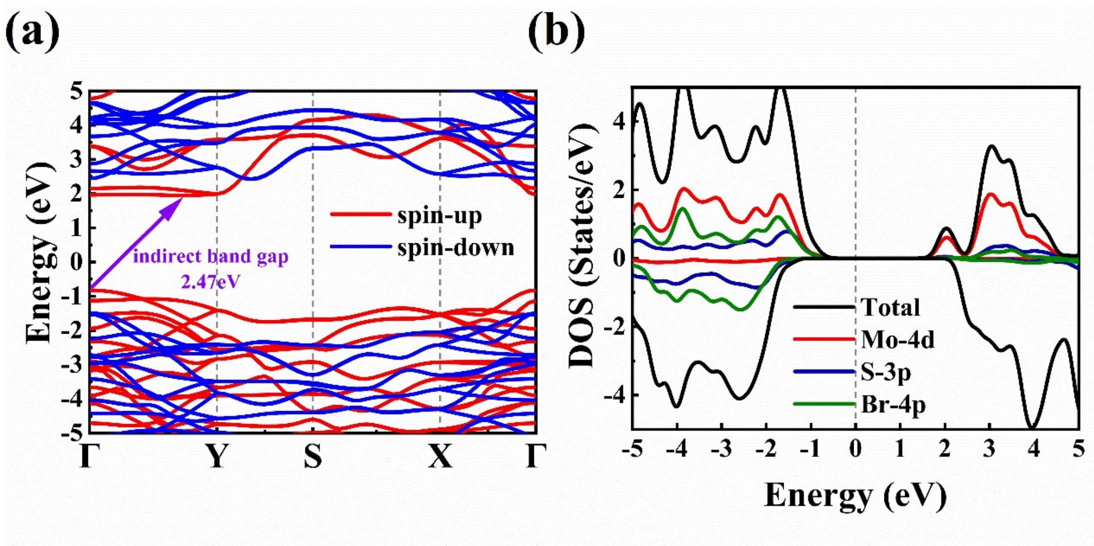

Fig. S3. (a) Band structures and (b) *DOS* of the MoSBr monolayer based on the HSE06 functional.

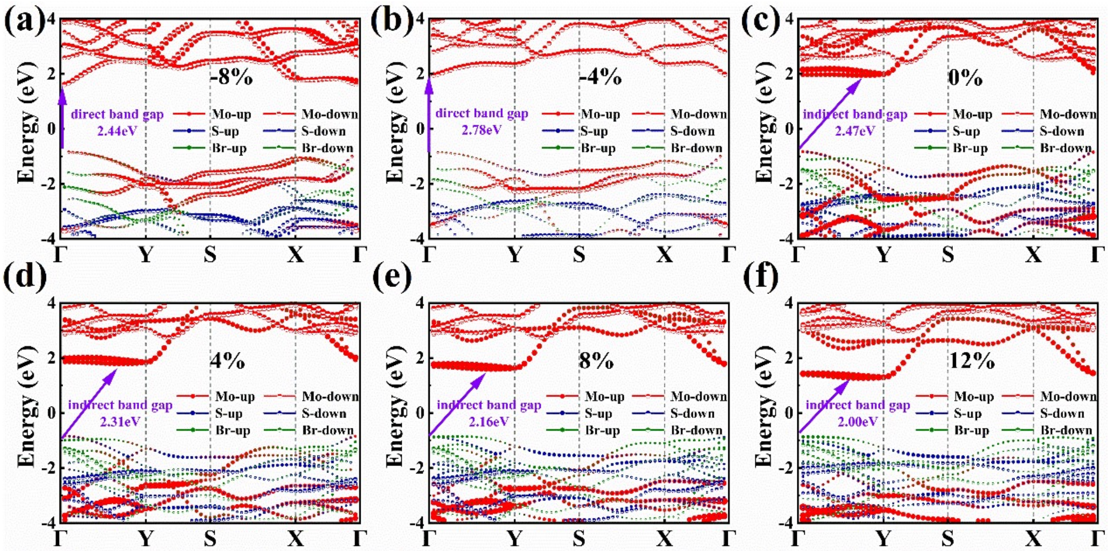

Fig. S4. (a)–(f) Energy band structures of the MoSBr monolayer under different biaxial strain values showing the transition from a direct-bandgap to an indirect-bandgap semiconductor.

Table. S1. Energies of the five possible magnetic structures of the MoSBr monolayer under different biaxial strains and corresponding energy differences between the FM and AFM states.

	Energy (atom)	**FM**	**AFM1**	**AFM2**	**AFM3**	**AFM4**
-8%	E (eV)	-123.805	-123.549	-124.540	-124.499	-123.604
	$\Delta E\ (meV)$	0	0.256	0.736	0.694	0.201
-6%	E (eV)	-126.073	-125.801	-126.508	-126.407	-125.807
	$\Delta E\ (meV)$	0	0.272	0.435	0.334	0.265
-4%	E (eV)	-127.535	-127.258	-127.741	-127.604	-127.234
	$\Delta E\ (meV)$	0	0.277	0.206	0.068	0.301
-2%	E (eV)	-128.311	-128.033	-128.346	-128.188	-127.993
	$\Delta E\ (meV)$	0	0.278	0.035	0.123	0.318
0%	E (eV)	-128.505	-128.226	-128.413	-128.245	-128.180
	$\Delta E\ (meV)$	0	0.278	0.092	0.259	0.324
2%	E (eV)	-128.197	-127.924	-128.019	-127.851	-127.879
	$\Delta E\ (meV)$	0	0.274	0.179	0.347	0.318
4%	E (eV)	-127.472	-127.202	-127.232	-127.069	-127.164
	$\Delta E\ (meV)$	0	0.270	0.240	0.404	0.309
6%	E (eV)	-126.393	-126.126	-126.111	-125.957	-126.097
	$\Delta E\ (meV)$	0	0.267	0.282	0.436	0.296
8%	E (eV)	-125.016	-124.753	-124.708	-124.565	-124.736
	$\Delta E\ (meV)$	0	0.263	0.308	0.451	0.281
10%	E (eV)	-123.393	-123.133	-123.069	-122.939	-123.128
	$\Delta E\ (meV)$	0	0.260	0.324	0.454	0.265
12%	E (eV)	-121.565	-121.308	-121.233	-121.116	-121.318
	$\Delta E\ (meV)$	0	0.257	0.332	0.449	0.247

Table. S2. SOC-MAE, shape-MAE, total MAE (in meV), and atomic MAE for the MoSBr monolayer with the (001) direction taken as reference.

	$E_{(100)}$ (meV)	**$E_{(010)}$ (meV)**	**$E_{(001)}$ (meV)**	**EMA**
SOC-MAE	0.269	-0.464	0	In-plane (010)
Shape-MAE	-0.065	-0.056	0	
Total-MAE	0.205	-0.520	0	
MAE_{Mo}	0.126	-0.076	0	In-plane (010)
MAE_{S}	-0.007	0.014	0	
MAE_{Br}	0.002	-0.163	0	

Table. S3. Orbital-resolved contributions to MAE from Mo *d*-orbitals and Br *p*-orbitals, with the magnetization aligned along (100) crystallographic directions.

	d_{xy}	d_{yz}	d_{z^2}	d_{xz}	$d_{x^2-y^2}$
d_{xy}	0.000	0.000	0.000	0.005	-0.530
d_{yz}	0.000	0.000	0.800	-0.005	-0.205
d_{z^2}	0.000	0.800	0.000	0.000	0.000
d_{xz}	0.005	-0.005	0.000	0.000	0.000
$d_{x^2-y^2}$	-0.530	-0.205	0.000	0.000	0.000

	p_y	p_z	p_x
p_y	0.000	0.107	-0.111
p_z	0.107	0.000	-0.001
p_x	-0.111	-0.001	0.000

Table. S4. Orbital-resolved contributions to MAE from Mo *d*-orbitals with the magnetization aligned along (010) crystallographic directions.

	d_{xy}	d_{yz}	d_{z^2}	d_{xz}	$d_{x^2-y^2}$
d_{xy}	0.000	0.000	0.000	-0.011	-0.517
d_{yz}	0.000	0.000	0.008	-0.015	0.005
d_{z^2}	0.000	0.008	0.000	0.000	0.000
d_{xz}	-0.011	-0.015	0.000	0.000	0.000
$d_{x^2-y^2}$	-0.517	0.005	0.000	0.000	0.000

	p_y	p_z	p_x
p_y	0.000	0.006	-0.107
p_z	0.006	0.000	0.019
p_x	-0.107	0.019	0.000

Table. S5. The difference between the square of the angular momentum matrix elements for the two magnetization directions $|\langle o^{-/+}|L_z|u^{-/+}\rangle|^2 - |\langle o^{-/+}|L_x|u^{-/+}\rangle|^2$.

u^+	o^+				
	d_{xy}	d_{yz}	d_{z^2}	d_{xz}	$d_{x^2-y^2}$
d_{xy}	0	0	0	1	−4
d_{yz}	0	0	3	−1	1
d_{z^2}	0	3	0	0	0
d_{xz}	1	−1	0	0	0
$d_{x^2-y^2}$	−4	1	0	0	0

$\boldsymbol{u}^{+}$	$\boldsymbol{o}^{-}$		
	p_y	p_z	p_x
p_y	0	0	−1
p_z	0	0	1
p_x	−1	1	0