Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Structural, vibrational, photoelectrochemical, and optical properties of two-dimensional Ruddlesden-Popper perovskite BA2PbI4 crystals

Aochen DU [1,2], Debing, SHEN [3], Wenxiao ZHAO [1,2], Yongzhen Liu [1,2], Xinzhi QIN [1,2], Zexi LIN [1,2], Yun YE [1,2*], Enguo CHEN [1,2*], Sheng XU [1,2], Tailiang GUO [1,2]

1. College of Physics and Information Engineering, Fuzhou University, Fuzhou 350100, China

2. Fujian Science & Technology Innovation Laboratory for Optoelectronic Information of China, Fuzhou 350100, China

3. College of Chemistry, Fuzhou University, Fuzhou 350108, Fujian, China

The preparation process of the phase stability diagram of BA_2PbI_4

ΔH (**PbI_2**)=Energy/Unit cell number (**PbI_2**)-Energy/Unit cell number (**Pb**)-**2**×Energy/Unit cell number (**I**)

ΔH (**PbI_2**)=(-8.612658508)-(-3.55549016)-2×(-1.469965435)=-2.117237478

From this, **Eq. (3)** is obtained, **$\mu_{Pb}+2\mu_I<\Delta H\ (PbI_2)=-2.117$ eV**

ΔH (**BAI**)=Energy/Unit cell number (**BAI**)-Energy/Unit cell number (**BA**)-Energy/Unit cell number (**I**)

ΔH (**BAI**)=(-91.0312)-(-86.73996679)-(-1.469965435)=-2.821267775

From this, **Eq. (2)** is obtained, **$\mu_{BA}+\mu_I<\Delta H\ (BAI)=-2.821$ eV**

ΔH (**BA_2PbI_4**)=Energy/Unit cell number (**BA_2PbI_4**)-**2**×Energy/Unit cell number (**BA**)-Energy/Unit cell number (**Pb**)-**4**×Energy/Unit cell number (**I**)

ΔH (**BA_2PbI_4**)=(-192.4926372)-2×(-86.73996679)-(-3.55549016)-4×(-1.469965435)=-9.577351688

From this, **Eq. (1)** is obtained, **$2\mu_{BA}+\mu_{Pb}+4\mu_I=\Delta H\ (BA_2PbI_4)=-9.577$ eV**

According to Eq. (1), it can be concluded that the value of **A point and B point.**

$2\mu_{BA}+\mu_{Pb}+4\mu_I=-9.577$ eV, because on the μ_{Pb} axis, the value of μ_{BA} and μ_I are equal to 0, $\mu_{Pb}=-9.577$ (**A point**).

$2\mu_{BA}+\mu_{Pb}+4\mu_I=-9.577$ eV, because on the μ_I axis, the value of μ_{BA} and μ_{Pb} are equal to 0, $\mu_I=-2.394$ (**B point**).

According to Eq. (4), it can be concluded that the value of **C point** and **D point.**

$\mu_{Pb}+2\mu_I=-3.935$ eV, because on the μ_{Pb} axis, the value of μ_I are equal to 0, $\mu_{Pb}=-3.935$ (**C point**).

$\mu_{Pb}+2\mu_I=-3.935$ eV, because on the μ_I axis, the value of μ_{Pb} are equal to 0, $\mu_I=-1.967$ (**D point**).

According to Eq. (3), it can be concluded that the value of **E point and F point.**

$\mu_{Pb}+2\mu_I=-2.117$ eV, because on the μ_{Pb} axis, the value of μ_I are equal to 0, $\mu_{Pb}=-1.058$ (**E point**).

$\mu_{Pb}+2\mu_I=-2.117$ eV, because on the μ_I axis, the value of μ_{Pb} are equal to 0, $\mu_I=-1.058$ (**F point**).

Table S1 Distance of atoms

Atom 1	Atom 2	Count	$d_{1,2}$ (Å)
C1	H1D	1x	1.0969
	H1E	1x	1.0971
	N1	1x	1.5009
	C2	1x	1.5215
	H1A	1x	2.0923
	H1C	1x	2.128
	H1B	1x	2.1311
	H2A	1x	2.1566
	H2B	1x	2.1593
H1D	C1	1x	1.0964

	H1E	1x	1.777
	N1	1x	2.0995
	C2	1x	2.1799
	H1A	1x	2.3815
	H1B	1x	2.4431
H1E	C1	1x	1.0971
	H1D	1x	1.7773
	N1	1x	2.0985
	C2	1x	2.1754
	H1A	1x	2.3797
	H1C	1x	2.438
C2	H2A	1x	1.101
	H2B	1x	1.1013
	C1	1x	1.5216
	C3	1x	1.5331
	H3A	1x	2.156
	H3B	1x	2.1594
	H1E	1x	2.1755
	H1D	1x	2.1802
H2A	C2	1x	1.101
	H2B	1x	1.7687
	C1	1x	2.1566
	C3	1x	2.1672
H2B	C2	1x	1.1014
	H2A	1x	1.7692
	C1	1x	2.1594
	C3	1x	2.1707
C3	H3B	1x	1.1008
	H3A	1x	1.1008
	C4	1x	1.5259
	C2	1x	1.5327
	H2A	1x	2.1672
	H2B	1x	2.1707
	H4B	1x	2.1815
	H4A	1x	2.1818
	H4C	1x	2.1837
H3A	C3	1x	1.1007
	H3B	1x	1.7629
	C2	1x	2.1559
	C4	1x	2.1644
H3B	C3	1x	1.1003
	H3A	1x	1.763
	C2	1x	2.1588
	C4	1x	2.1662

C4	H4A	1x	1.0978
	H4C	1x	1.0982
	H4B	1x	1.0985
	C3	1x	1.5259
	H3A	1x	2.1645
	H3B	1x	2.1664
H4A	C4	1x	1.0978
	H4B	1x	1.7715
	H4C	1x	1.773
	C3	1x	2.1819
H4B	C4	1x	1.0985
	H4C	1x	1.7648
	H4A	1x	1.7709
	C3	1x	2.1815
H4C	C4	1x	1.0982
	H4B	1x	1.7649
	H4A	1x	1.7731
	C3	1x	2.1838
N1	H1A	1x	1.0327
	H1B	1x	1.0458
	H1C	1x	1.0465
	C1	1x	1.5013
	H1E	1x	2.0988
	H1D	1x	2.1
H1A	N1	1x	1.0326
	H1B	1x	1.6694
	H1C	1x	1.6717
	C1	1x	2.0923
	H1E	1x	2.3797
	H1D	1x	2.3815
H1B	N1	1x	1.0458
	H1A	1x	1.6695
	H1C	1x	1.6955
	C1	1x	2.1315
	H1D	1x	2.4436
H1C	N1	1x	1.0464
	H1A	1x	1.6714
	H1B	1x	1.6954
	C1	1x	2.128
	H1E	1x	2.438

Table S2 Atomic parameters

	Element	Atom	Wyck.	Site	x/a	y/b	z/c
1	C	C1	8	c	0.96535	0.4707	0.88132
2	H	H1D	8	c	0.85414	0.50676	0.89439
3	H	H1E	8	c	0.9888	0.35554	0.89575
4	C	C2	8	c	0.97396	0.47284	0.82631
5	H	H2A	8	c	1.08867	0.44203	0.81461
6	H	H2B	8	c	0.95202	0.59001	0.81286
7	C	C3	8	c	0.86038	0.36018	0.80416
8	H	H3A	8	c	0.88285	0.24427	0.81848
9	H	H3B	8	c	0.74628	0.39136	0.8163
10	C	C4	8	c	0.86709	0.35798	0.74891
11	H	H4A	8	c	0.78685	0.27498	0.73364
12	H	H4B	8	c	0.97983	0.32626	0.73593
13	H	H4C	8	c	0.84095	0.4713	0.73346
14	N	N1	8	c	1.07844	0.57748	0.90381
15	H	H1A	8	c	1.07024	0.57302	0.94113
16	H	H1B	8	c	1.06112	0.69207	0.89372
17	H	H1C	8	c	1.18913	0.54738	0.89483
18	I	I1	8	c	1.03301	0.98413	0.8832
19	I	I2	8	c	0.6867	1.18472	0.99747
20	Pb	Pb1	4	a	1	1	1