Electronic Supplementary Material (ESI) for Nanoscale.
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Electronic supplementary information for

Highly-efficient heterojunction solar cells based on 2D Janus transition-metal nitride halide (TNH) monolayers with ultrahigh carrier mobility

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Table S1 Lattice constants l_a (Å), band gaps E_g (eV) calculated from PBE ($E_{g,PBE}$), HSE06 ($E_{g,HSE}$) and HSE06+SOC ($E_{g,HSE+SOC}$) for Janus TNH monolayers.

Material	l_a (Å)	$E_{g,PBE}$ (eV)	$E_{g,HSE}$ (eV)	$E_{g,HSE+SOC}$ (eV)
TiNCl	3.19	2.53	3.64	3.54
ZrNCl	3.45	3.18	4.25	4.33
HfNCl	3.39	3.82	4.93	4.74
FeNCl	3.03	0.57	1.48	1.71
FeNF	2.86	1.23	2.20	2.72
PdNCl	3.14	0.52	1.34	0.93
PdNF	3.02	0.99	2.01	2.08
PtNCl	3.15	0.72	1.55	1.05
PtNF	3.06	0.93	1.94	1.82
OsNCl	3.15	0.97	1.67	1.38
ReNCl	3.10			

Table S2 Calculated elastic constants (C_{11}, C_{12}, and C_{66}) of TNH monolayers.

Material	C_{11}	C_{12}	C_{66}
TiNCl	129.00	47.12	40.92
ZrNCl	121.28	50.38	35.44
HfNCl	128.58	50.02	39.26
FeNCl	157.20	50.92	53.12
FeNF	164.98	52.20	56.38
PdNCl	83.90	23.44	30.22
PdNF	69.60	13.26	30.52
PtNCl	89.90	26.90	31.92
PtNF	69.32	20.32	28.90
OsNCl	160.50	62.05	49.22
ReNCl	123.20	-80.02	-78.32
OsNCl/FeNCl	315.82	109.38	104.34

Table S3 Bader charges of all atoms constituting the janus TNH monolayers, and the electrostatic potential difference ($\Delta\Phi$) of them.

System	Bader charge						$\Delta\Phi$ (eV)
TiNCl	Ti	-1.627	N	1.183	Cl	0.444	0.218
ZrNCl	Zr	-2.150	N	1.545	Cl	0.605	0.110
HfNCl	Hf	-1.682	N	1.189	Cl	0.493	0.153
FeNCl	Fe	-1.045	N	0.771	Cl	0.274	0.484
FeNF	Fe	-1.354	N	0.728	F	0.626	1.620
PdNCl	Pd	-0.847	N	0.571	Cl	0.276	-1.217
PdNF	Pd	-1.057	N	0.483	F	0.573	0.184
PtNCl	Pt	-0.898	N	0.602	Cl	0.296	-1.546
PtNF	Pt	-1.292	N	0.599	F	0.693	-0.156
OsNCl	Os	-1.216	N	0.818	Cl	0.398	0.490
ReNCl	Re	-1.501	N	1.133	Cl	0.367	0.699

Table S4 The calculated DP constants E_1 (eV), elastic moduli C_{2D} (N m^{-1}), effective masses m^*/m_0, relaxation-time τ (10^2 s) and carrier mobility μ (10^2 cm^2 V^{-1} s^{-1}) of the 2D TNHs for electrons (e) and holes (h) along the *a*- and *b*-directions.

System	Carrier type	E_1 (eV)	C_{2D} (N m^{-1})	m^*/m_0	τ (10^2 s)	μ (10^2 cm^2 V^{-1} s^{-1})
TiNCl	e(a-axis)	1.39	129.74	0.33	16.38	83.22
	h(a-axis)	2.97		0.98	1.21	2.07
	e(b-axis)	2.05	128.07	1.15	2.14	3.12
	h(b-axis)	6.20		4.39	0.06	0.02
ZrNCl	e(a-axis)	1.5	118. 13	0.65	6.53	16.90
	h(a-axis)	0.22		2.44	81.04	55.98
	e(b-axis)	3.09	116.76	0.57	1.74	5.18
	h(b-axis)	0.79		1.41	10.74	12.85
HfNCl	e(a-axis)	1.02	129. 41	1.03	9.70	15.79
	h(a-axis)	4.57		1.82	0.27	0.25
	e(b-axis)	1.81	129.32	0.74	4.32	9.87
	h(b-axis)	4.43		3.00	0.18	0.10
FeNCl	e(a-axis)	0.65	156.27	0.71	41.82	98.51
	h(a-axis)	1.21		2.15	4.01	3.14
	e(b-axis)	3.91	168.14	0.70	0.27	3.05
	h(b-axis)	5.42		3.35	0.14	0.07
FeNF	e(a-axis)	0.32	164.84	2.31	56.16	40.82
	h(a-axis)	2.11		1.40	2.13	2.55
	e(b-axis)	0.78	162.26	0.84	25.54	50.96
	h(b-axis)	0.95		4.02	3.61	1.51
PdNCl	e(a-axis)	1.63	81.58	3.47	0.71	0.35
	h(a-axis)	0.87		1.12	7.79	11.73
	e(b-axis)	1.8	81.56	1.70	1.20	1.19
	h(b-axis)	1.64		1.01	2.43	4.06
PdNF	e(a-axis)	4.85	83.86	2.26	0.13	0.09
	h(a-axis)	5.37		1.61	0.15	0.15
	e(b-axis)	0.83	91.44	1.25	8.57	11.52
	h(b-axis)	5.33		3.54	0.07	0.03
OsNCl	e(a-axis)	0.62	159. 57	0.29	115.68	670.97
	h(a-axis)	5.23		0.25	1.90	12.89
	e(b-axis)	1.79	159.73	0.31	12.98	70.44
	h(b-axis)	4.39		2.80	0.24	0.14
PtNCl	e(a-axis)	2.13	90.99	2.62	0.62	0.40
	h(a-axis)	0.74		0.88	15.21	29.00
	e(b-axis)	1.41	90.99	0.75	4.94	11.11
	h(b-axis)	0.79		5.42	2.17	0.67

PtNF	e(a-axis)	2.13	83.75	0.97	1.53	2.65
	h(a-axis)	0.74		0.60	20.75	58.66
	e(b-axis)	2.28	83.60	1.19	1.11	1.57
	h(b-axis)	1.39		6.58	0.54	0.14

Table S5: The reduced effective mass of exciton μ_{ex}, 2D macroscopic static dielectric constant ε_r, exciton binding energy E_{exc}（eV）and exciton Bohr radius a^*（Å）of OsNCl and FeNCl monolayers.

system	μ_{ex}		ε_{2D}		E_{exc} (eV)		a^*(Å)	
	μ_{ex}^x	μ_{ex}^y	ε_{2D}^x	ε_{2D}^y	E_{exc}^x	E_{exc}^y	a_x^*	a_y^*
OsNCl	0.13	0.27	7.17	7.17	0.14	0.28	7.29	3.26
FeNCl	0.53	0.57	5.39	5.39	0.99	1.08	1.34	1.25

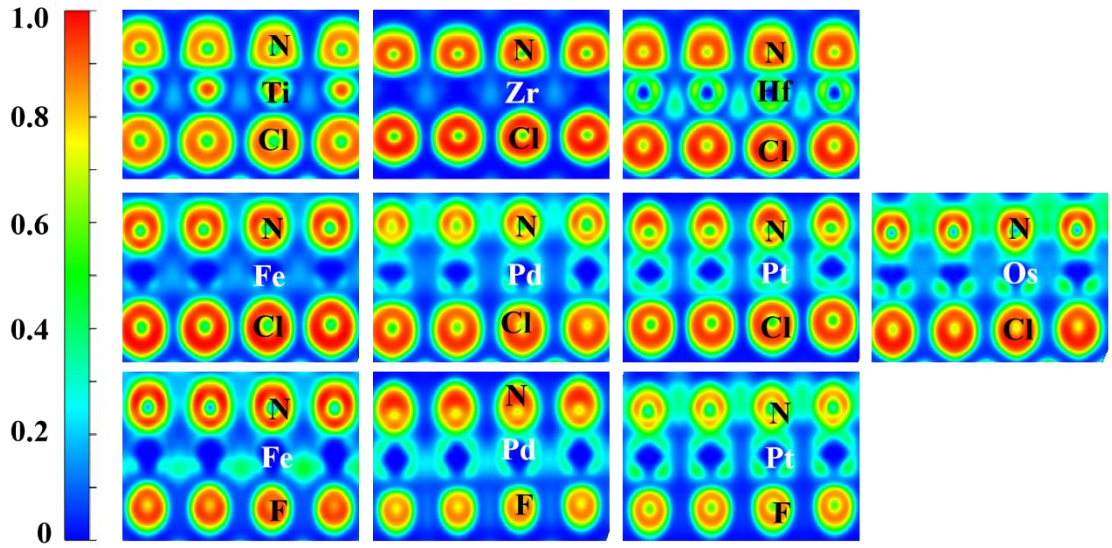

Fig S1: The electron localization functions (ELFs) of TNH monolayers along the T-N-H planes.

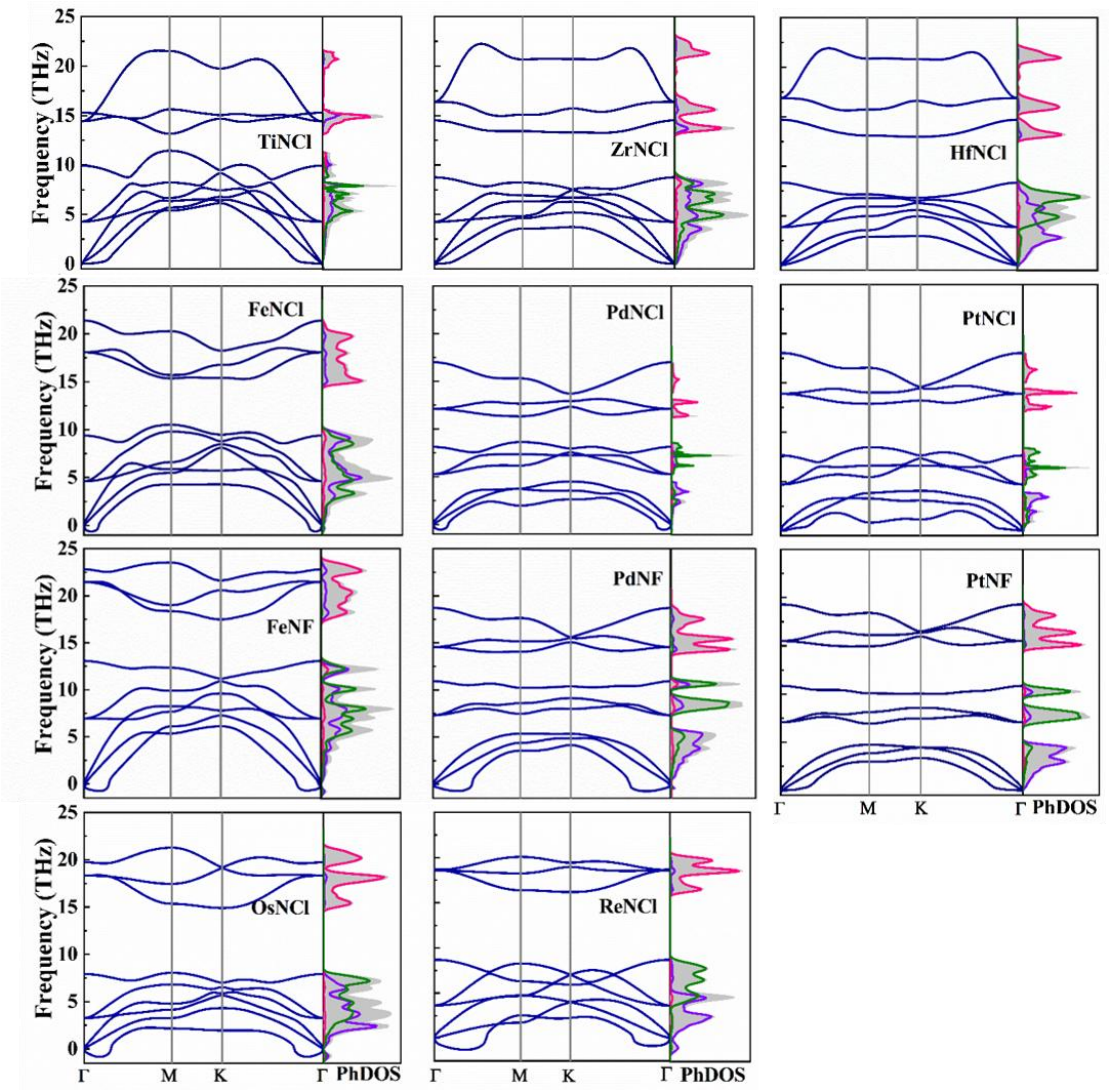

Fig. S2 Phonon band dispersion curves and projected phonon state density (PhDOS) of monolayer TNHs (T=Ti, Zr, Hf, Fe, Pd, Pt, Os and Re, H = Cl and F), slight imaginary frequency near the G-spot doesn't mean the structure is unstable. For PhDOS, the purple, green and red lines are contributed by transition metal (T), nitrogen (N) and halogens (H) atoms, respectively.

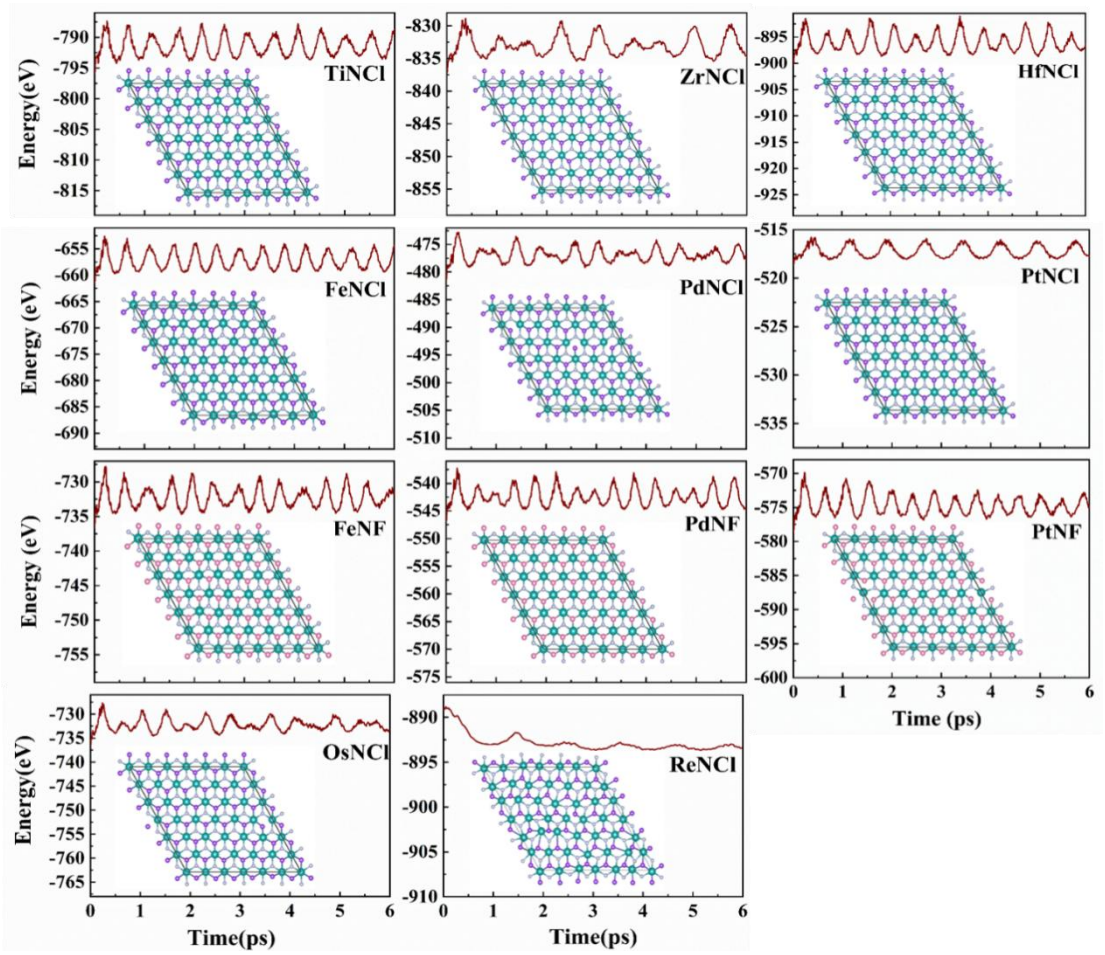

Fig. S3 Total energy fluctuation for Janus TNH monolayers during the AIMD simulation, the illustrations are the atomic structures after 6 ps at 300 K. All non-magnetic structures at 300 K to confirm the thermodynamic stability while 1L-ReNCl is thermodynamically stable at 50K, the supercell of 6 × 6 × 1 shows slight deformation and bond breakage for 1L-ReNCl. Here, the transition metal (T) atom, N atom, Cl atom and F atom are labeled cyan, gray, purple and pink.

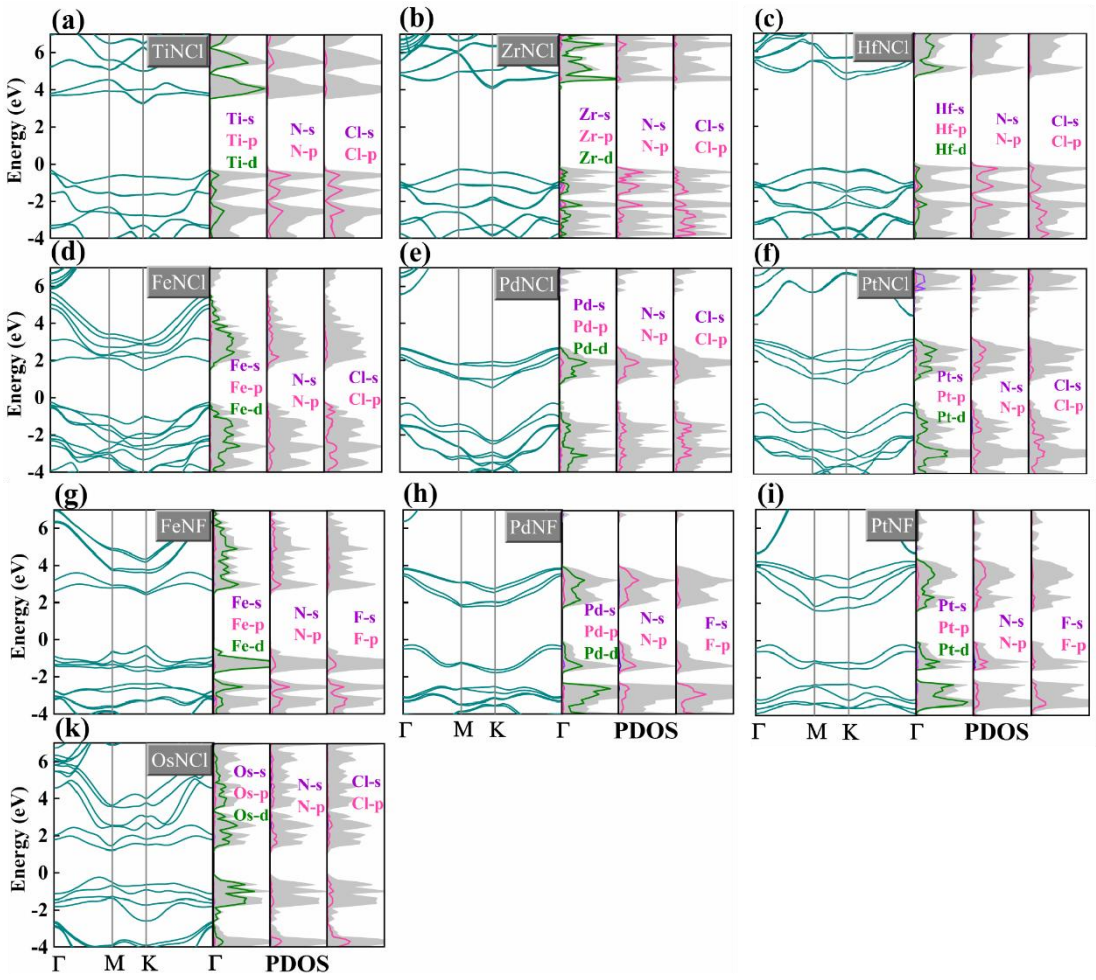

Fig. S4 Electronic band structures and projected density of states (PDOS) of janus TNH monolayers by using HSE+SOC. The Fermi level is set at 0.

Atomic lattices in VASP POSCAR format.

TiNCl
1.00000000000000
3.1892767007098541 -0.0000004909871754 0.0000000000000000
-1.5946387792294885 2.7619956940458423 0.0000000000000000
0.0000000000000000 0.0000000000000000 20.0000000000000000
Ti N Cl
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Direct
0.0000002752165482 0.9999998274913051 0.6157957050996479
0.6666669089281024 0.3333332393291135 0.6468319397900241
0.3333328448553412 0.6666669621795944 0.5268423721103203

ZrNCl
1.00000000000000
3.4454166427422348 -0.0000001081111996 0.0000000000000000
-1.7227084155136612 2.9838183585528668 0.0000000000000000
0.0000000000000000 0.0000000000000000 20.0000000000000000
Zr N Cl
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0.3333331361177584 0.6666668925470852 0.5237641882479112

HfNCl
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Hf N Cl
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0.3333324380763187 0.6666675982534613 0.5239306778497550

FeNCl
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Fe N Cl
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FeNF
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PdNCl
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0.3333332300504321 0.6666668512717494 0.5352620160637699

PtNCl
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-1.5748300113480493 2.7276846710202465 0.0000000000000000
0.0000000000000000 0.0000000000000000 20.0000000000000000
Pt N Cl
1 1 1
Direct
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0.6666666867635707 0.3333332268781711 0.6599016941684325
0.3333333402410841 0.6666668324491171 0.5202472689443871

OsNCl
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0.0000000000000000 0.0000000000000000 20.0000000000000000
Os N Cl
1 1 1
Direct
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0.6666675541191438 0.3333325185448004 0.6480957572409665
0.3333318505104970 0.6666680994539860 0.5248279017644748