Supplementary Information: A Predictive Framework for Designing Chiral Charge Density Wave in Layered Quantum Materials

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TABLE S1. Detailed Structural information of various 2×2×3 and 2×2×4 CDW phases of CsV_3Sb_5

Phase	**Lattice parameters**	**Atomic coordinates (fractional)**			
(Space Group)	**($\AA$ ∘)**	**Atoms**	**X**	**Y**	**Z**
2×2×3	a=18.8554	Cs1(4l)	0	-0.5	-0.16549
(Cmmm)	b=10.8857	Cs2(8m)	0.25	0.25	-0.16661
	c=28.1078	Cs3(4k)	0	0	0.83223
	α=β=γ=90	Cs5(2c)	0	-0.5	0.5
		Cs4(4f)	0.25	0.25	0.5
		Cs5(2d)	0	0	0.5
		V1(16r)	0.12735	-0.38194	0.66674
		V2(16r)	0.37709	-0.37699	0.66671
		V3(8n)	0	-0.7542	0.66674
		V4(8o)	0.24538	-0.5	0.66671
		V5(8p)	0.37709	-0.12313	0
		V6(8p)	0.37265	-0.38186	0
		V7(4g)	0.25458	-0.5	0
		V8(4i)	0	-0.74578	0
		Sb1(16r)	0.41634	-0.24886	0.74657
		Sb2(16r)	0.08365	-0.25105	0.5869
		Sb3(16r)	0.41636	-0.25119	0.07984
		Sb4(4l)	0	-0.5	0.6668
		Sb5(4k)	0	0	0.66669
		Sb6(8o)	0.16665	-0.5	0.7489
		Sb7(8o)	0.33268	-0.5	0.5869
		Sb8(8o)	0.16735	-0.5	0.07977
		Sb9(8o)	0.16736	0	0.7465
		Sb10(8o)	0.33333	0	0.5846
		Sb11(8o)	0.16665	0	0.08214

		Sb12(8m)	0.25	-0.25	0.66673
		Sb13(4e)	0.25	-0.25	0
		Sb14(2b)	0	-0.5	0
		Sb15(2a)	0	0	0
2×2×3	a=b=10.8865	Cs1(3c)	0.5	0	0
($P6_422$)	c=28.1009	Cs2(6f)	0.5	0.5	0.00111
	α=β=90	Cs3(3a)	0	0	0
	γ=120	V1(12k)	0.7541	0.25425	0.8333
		V2(12k)	0.25452	0.00917	0.8333
		V3(6h)	0	0.25408	0.83333
		V4(6j)	0.49078	0.24539	0.83333
		Sb1(12k)	0.33332	0.16665	0.91549
		Sb2(12k)	0.83255	0.16524	0.91312
		Sb3(12k)	0.33484	0.66742	0.91319
		Sb4(12k)	0.83255	0.66731	0.91318
		Sb5(6f)	0.5	0	0.83325
		Sb6(3b)	0	0	0.83333
		Sb7(3d)	0	0.5	0.83333
2×2×3	a=b=10.8865	Cs1(3d)	0	0.5	0.16667
($P6_222$)	c=28.1009	Cs2(3b)	0	0	0.16667
	α=β=90	Cs3(6f)	0.5	0.5	0.16782
	γ=120	V1(12k)	0.25426	0.75411	-0.00002
		V2(12k)	0.99082	0.74547	-0.00003
		V3(6i)	0.75462	0.50924	0
		V4(6g)	0.25408	0	0
		Sb1(12k)	0.83332	0.66667	0.08219
		Sb2(12k)	0.33261	0.66514	0.07988
		Sb3(12k)	0.83477	0.16743	0.07981
		Sb4(12k)	0.3327	0.16748	0.07987
		Sb5(6c)	0	0.5	-0.00006
		Sb6(3c)	0.5	0	0
		Sb7(3a)	0	0	0

2×2×4	a= 18.8577	Cs1(4k)	0	0	0.12418
(Cmmm)	b= 10.8860	Cs2(4k)	0	0	0.37496
	c= 37.4829	Cs3(8m)	0.25	-0.25	0.12498
	α=β=γ=90	Cs4(8m)	0.25	-0.25	0.37495
		Cs5(4l)	0	-0.5	0.1258
		Cs6(4l)	0	-0.5	0.37498
		V1(16r)	-0.37264	-0.382	0.24997
		V2(16r)	-0.12284	-0.62292	0.24999
		V3(8q)	-0.37265	-0.38213	0.5
		V4(8q)	-0.12284	-0.62285	0.5
		V5(8p)	-0.37713	-0.37695	0
		V6(8p)	-0.12735	-0.61805	0
		V7(8n)	0	-0.74568	0.24996
		V8(8o)	-0.25467	-0.5	0.24999
		V9(4j)	0	-0.74573	0.5
		V10(4h)	-0.25474	-0.5	0.5
		V11(4i)	0	-0.75428	0
		V12(4g)	-0.24539	-0.5	0
		Sb1(16r)	-0.08364	-0.75103	0.30982
		Sb2(16r)	-0.08367	-0.751	0.55984
		Sb3(16r)	-0.08366	-0.75112	0.80988
		Sb4(16r)	-0.08364	-0.74882	0.05985
		Sb5(4k)	0	0	0.24992
		Sb6(8m)	0.25	-0.25	0.24997
		Sb7(4l)	0	-0.5	0.25
		Sb8(8o)	-0.33334	-0.5	0.31156
		Sb9(8o)	0.16731	-0.5	0.30982
		Sb10(8o)	-0.33333	-0.5	0.5616

		Sb11(8o)	0.16732	-0.5	0.55984
		Sb12(8o)	-0.33335	-0.5	0.81164
		Sb13(8o)	0.16736	-0.5	0.80983
		Sb14(8o)	-0.33266	-0.5	0.05981
		Sb15(8o)	0.16665	-0.5	0.06159
		Sb16(2d)	0	0	0.5
		Sb17(4f)	0.25	-0.25	0.5
		Sb18(2c)	0	-0.5	0.5
		Sb19(2a)	0	0	0
		Sb20(4e)	0.25	-0.25	0
		Sb21(2b)	0	-0.5	0
2×2×4	a=18.85590	Cs1(8i)	0.5	0.5	-0.24919
(Fmmm)	b= 10.88620	Cs2(8f)	0.25	0.25	-0.25
	c=37.48960	Cs3(4b)	0.5	0.5	-0.5
	α=β=γ=90	Cs4(8e)	0.25	0.25	-0.5
		Cs5(4a)	0	0	0
		V1(32p)	0.62735	0.38197	-0.37497
		V2(32p)	0.37712	0.12298	-0.37499
		V3(16m)	0.5	0.24576	-0.37497
		V4(16n)	0.74536	0.5	-0.37499
		Sb1(32p)	0.41635	0.25107	-0.43482
		Sb2(32p)	0.91634	0.75113	-0.68487
		Sb3(8i)	0.5	0.5	-0.37492
		Sb4(8i)	0.5	0.5	-0.87501
		Sb5(16n)	0.66667	0.5	-0.43654
		Sb6(16n)	0.16732	0.5	-0.43482
		Sb7(16n)	0.66666	0.5	-0.68662
		Sb8(16n)	0.66735	0.5	-0.18483
		Sb9(16j)	0.25	0.25	-0.37498
2×2×4	a= 18.85650	Cs1(4k)	-0.25	0.25	-0.24997
(C222)	b= 10.88760	Cs2(4k)	-0.25	0.25	-0.5

c= 37.47060	Cs3(4k)	-0.25	0.25	0.00083
α=β=γ=90	Cs4(4i)	0	0	-0.24996
	Cs5(4j)	0	0.5	-0.25081
	Cs6(4k)	0.25	0.25	-0.24913
	Cs7(2d)	0	0	-0.5
	Cs8(2c)	0	0.5	-0.5
	Cs9(2a)	0	0	0
	Cs10(2b)	0	0.5	0
	V1(8l)	-0.25471	0.49994	-0.37497
	V2(8l)	0.00003	0.25425	-0.37498
	V3(8l)	-0.24995	0.49574	-0.87502
	V4(8l)	0.00465	0.24998	-0.87498
	V5(8l)	-0.37265	0.38203	-0.37498
	V6(8l)	-0.12285	0.62282	-0.37496
	V7(8l)	-0.37267	0.38208	-0.62503
	V8(8l)	-0.1229	0.6228	-0.62502
	V9(8l)	-0.3729	0.37289	-0.875
	V10(8l)	-0.12265	0.63201	-0.875
	V11(8l)	0.12266	0.86804	-0.12498
	V12(8l)	-0.12715	0.62705	-0.12502
	Sb1(8l)	-0.33335	0.49999	-0.43658
	Sb2(8l)	-0.08366	0.24901	-0.43483
	Sb3(8l)	-0.08369	0.75099	-0.43483
	Sb4(8l)	0.16736	0.49997	-0.43483
	Sb5(8l)	-0.33333	0.49996	-0.68666
	Sb6(8l)	-0.08374	0.24895	-0.68489
	Sb7(8l)	-0.08369	0.75106	-0.68485
	Sb8(8l)	0.16742	0.50008	-0.68489
	Sb9(8l)	-0.33366	0.50111	-0.93486
	Sb10(8l)	-0.08261	0.25005	-0.93482
	Sb11(8l)	-0.08336	0.75002	-0.93662

		Sb12(8l)	0.1663	0.49884	-0.93486
		Sb13(8l)	-0.33366	0.49891	-0.18482
		Sb14(8l)	-0.08331	0.25003	-0.18662
		Sb15(8l)	-0.08261	0.74993	-0.18486
		Sb16(8l)	0.16627	0.50113	-0.18486
		Sb17(4k)	-0.25	0.25	-0.37497
		Sb18(4k)	-0.25	0.25	-0.875
		Sb19(4i)	0	0	-0.37497
		Sb20(4i)	0	0	-0.875
		Sb21(4j)	0	0.5	-0.375
		Sb22(4j)	0	0.5	-0.87495
		Sb23(4k)	0.25	0.25	-0.37493
		Sb24(4k)	0.25	0.25	-0.87505
2×2×4	a=18.85800	Cs1(8k)	-0.25	0.25	0.625
(Cccm)	b= 10.88760	Cs2(8k)	0.25	0.25	0.6242
	c= 37.47120	Cs3(8j)	0	0.5	0.62583
	α=β=γ=90	Cs4(8i)	0	0	0.62501
		V1(16m)	-0.37271	0.38197	0.75003
		V2(16m)	0.12289	0.37725	0.74997
		V3(8l)	-0.37296	0.37272	0
		V4(8l)	0.12272	0.36795	0
		V5(8l)	-0.24987	0.49583	0
		V6(8l)	-0.00467	0.75008	0
		V7(8l)	0.1227	0.86805	0.5
		V8(8l)	0.12718	0.37285	0.5
		V9(8g)	-0.25466	0.5	0.75
		V10(8h)	0	0.7458	0.75
		Sb1(16m)	-0.33334	0.50001	0.81162
		Sb2(16m)	0.16744	0.49999	0.80987
		Sb3(16m)	-0.08374	0.75104	0.80981
		Sb4(16m)	-0.08372	0.24895	0.80987
		Sb5(16m)	-0.33367	0.50102	0.05982

		Sb6(16m)	0.16621	0.49892	0.05987
		Sb7(16m)	-0.08333	0.75	0.06161
		Sb8(16m)	-0.08257	0.25014	0.05987
		Sb9(8k)	-0.25	0.25	0.75007
		Sb10(4b)	0	0.5	0.75
		Sb11(4a)	0	0	0.75
		Sb12(4f)	-0.25	0.25	0
		Sb13(4e)	0.25	0.25	0
		Sb14(4d)	0	0.5	0
		Sb15(4c)	0	0	0
2×2×4	a=18.85780	Cs1(16g)	0.25	1.25	0.12501
(Fddd)	b=10.88670	Cs2(16g)	0.25	1.25	-0.37421
	c=37.4658	V1(32h)	-0.12727	0.88192	-0.00003
	α=β=γ=90	V2(16e)	-0.24537	1	0
		V3(16f)	0.5	1.74588	-0.5
		V4(32h)	0.37708	1.37722	-0.49997
		Sb1(32h)	-0.16666	1.00002	-0.06161
		Sb2(32h)	0.33257	1.49998	-0.55989
		Sb3(32h)	0.08373	1.25104	-0.55983
		Sb4(32h)	0.08372	1.24896	-0.05989
		Sb5(16g)	0.25	1.25	-0.00007
		Sb6(8b)	0.5	1.5	-0.5
		Sb7(8a)	0	1	0

TABLE S2. Enthalpies and space groups of distorted 2×2×2 KV_3Sb_5 structures generated by incorporating layers with distinct interlayer phases φ_i^ℓ. + and - represent $\varphi_i^\ell = 0$ and $\varphi_i^\ell = \pi$, respectively.

Input φ_i^ℓ	**Output**		**Input φ_i^ℓ**	**Output**	
Layer 1; Layer 2	**Space Group**	**Enthalpy (eV/atom)**	**Layer 1; Layer 2**	**Space Group**	**Enthalpy (eV/atom)**
+ + + ; + + +	P6/mmm	-5.92266	+ + - ; + + -	P6/mmm	-5.92298
+ + + ; + + -	Cmmm	-5.92314	+ + - ; + - +	Fmmm	-5.92346
+ + + ; + - +	Cmmm	-5.92345	+ + - ; + - -	Cmmm	-5.92312
+ + + ; + - -	Fmmm	-5.92346	+ + - ; - + +	Fmmm	-5.92346
+ + + ; - + +	Cmmm	-5.92314	+ + - ; - + -	Cmmm	-5.92314
+ + + ; - + -	Fmmm	-5.92346	+ + - ; - - +	P6/mmm	-5.92305
+ + + ; - - +	**Fmmm**	**-5.92346**	+ + - ; - - -	Fmmm	-5.92346
+ + + ; - - -	P6/mmm	-5.92305	+ - + ; + - +	P6/mmm	-5.92298
+ - - ; + - -	P6/mmm	-5.92266	+ - + ; + - -	Cmmm	-5.92314
+ - - ; - + +	P6/mmm	-5.92305	+ - + ; - + +	Fmmm	-5.92346
+ - - ; - + -	Fmmm	-5.92346	+ - + ; - + -	P6/mmm	-5.92305
+ - - ; - - +	Fmmm	-5.92346	+ - + ; - - +	Cmmm	-5.92314
+ - - ; - - -	Cmmm	-5.92314	+ - + ; - - -	Fmmm	-5.92346
- + + ; - + +	P6/mmm	-5.92298	- + - ; - + -	P6/mmm	-5.92266
- + + ; - + -	Cmmm	-5.92314	- + - ; - - +	Fmmm	-5.92346
- + + ; - - +	Cmmm	-5.92314	- + - ; - - -	Cmmm	-5.92289
- + + ; - - -	Fmmm	-5.92346	- - + ; - - +	P6/mmm	-5.92266
- - - ; - - -	P6/mmm	-5.92266	- - + ; - - -	Cmmm	-5.92267

TABLE S3. Enthalpies and symmetries of 2×2×3 superstructures formed by incorporating various ISD layers for KV_3Sb_5. ISD layer indexes are the same as them defined in Extended Data Fig. 3. In Table, the layer index is in bottom-to-top sequence. Certain sequences of incorporating layers are identical to those we listed in the Table, thereby we do not show them in the Table. After fully relaxation, some structure go into the same phase.

Layer index	**Space Group**	**Enthalpy (eV/atom)**	**chirality**	**Layer index**	**Space Group**	**Enthalpy (eV/atom)**	**chirality**
112	Cmmm	-5.92327	achiral	124	$P6_222$	-5.92342	chiral
114				132			
122				143			
113				123	$P6_422$	-5.92342	chiral
121				134			
131				142			
133							
141							
144							

TABLE S4. Enthalpies and symmetries of 2×2×4 superstructures formed by incorporating various ISD layers for KV_3Sb_5. ISD layer indexes are the same as them defined in Extended Data Fig. 3. In Table, the layer index is in bottom-to-top sequence. Certain sequences of incorporating layer are identical to those we listed in the Table, thereby we do not show them in the Table. After fully relaxation, some structure go into the same phase.

Layer index	**Space Group**	**Enthalpy (eV/atom)**	**chirality**	**Layer index**	**Space Group**	**Enthalpy (eV/atom)**	**chirality**
1112	Cmmm	-5.92315	achiral	1122	Fmmm	-5.92311	achiral
1113				1133			
1114				1144			
1222				1123	C222	-5.92323	chiral
1333				1124			
1444				1132			
1213	Cccm	-5.92339	achiral	1134			
1214				1142			
1232				1143			
1242				1223			
1314				1224			
1323				1233			
1343				1244			
1424				1332			
1434				1334			
1234	Fddd	-5.92334	achiral	1344			
1243				1422			
1324				1433			
1342				1442			
1423				1443			
1432							

TABLE S5. Enthalpies and space groups of distorted 2×2×2 RbV_3Sb_5 structures generated by incorporating layers with distinct interlayer phases φ_i^ℓ. + and - represent $\varphi_i^\ell = 0$ and $\varphi_i^\ell = \pi$, respectively.

Input φ_i^ℓ	**Output**		**Input φ_i^ℓ**	**Output**	
Layer 1; Layer 2	**Space Group**	**Enthalpy (eV/atom)**	**Layer 1; Layer 2**	**Space Group**	**Enthalpy (eV/atom)**
+ + + ; + + +	P6/mmm	-5.92847	+ + - ; + + -	P6/mmm	-5.92924
+ + + ; + + -	Cmmm	-5.92916	+ + - ; + - +	Fmmm	-5.92976
+ + + ; + - +	Cmmm	-5.92916	+ + - ; + - -	Cmmm	-5.92916
+ + + ; + - -	Fmmm	-5.92976	+ + - ; - + +	Fmmm	-5.92976
+ + + ; - + +	Cmmm	-5.92916	+ + - ; - + -	Cmmm	-5.92916
+ + + ; - + -	Fmmm	-5.92976	+ + - ; - - +	P6/mmm	-5.92923
+ + + ; - - +	**Fmmm**	**-5.92976**	+ + - ; - - -	Fmmm	-5.92976
+ + + ; - - -	P6/mmm	-5.92923	+ - + ; + - +	P6/mmm	-5.92924
+ - - ; + - -	P6/mmm	-5.92847	+ - + ; + - -	Cmmm	-5.92314
+ - - ; - + +	P6/mmm	-5.92923	+ - + ; - + +	Fmmm	-5.92976
+ - - ; - + -	Fmmm	-5.92976	+ - + ; - + -	P6/mmm	-5.92923
+ - - ; - - +	Fmmm	-5.92976	+ - + ; - - +	Cmmm	-5.92916
+ - - ; - - -	Cmmm	-5.92916	+ - + ; - - -	Fmmm	-5.92346
- + + ; - + +	P6/mmm	-5.92924	- + - ; - + -	P6/mmm	-5.92851
- + + ; - + -	Cmmm	-5.92314	- + - ; - - +	Fmmm	-5.92980
- + + ; - - +	Cmmm	-5.92916	- + - ; - - -	Cmmm	-5.92916
- + + ; - - -	Fmmm	-5.92980	- - + ; - - +	P6/mmm	-5.92851
- - - ; - - -	P6/mmm	-5.92851	- - + ; - - -	Cmmm	-5.92916

TABLE S6. Enthalpies and symmetries of 2×2×3 superstructures formed by incorporating various ISD layers for RbV_3Sb_5. ISD layer indexes are the same as them defined in Extended Data Fig. 3. In Table, the layer index is in bottom-to-top sequence. Certain sequences of incorporating layers are identical to those we listed in the Table, thereby we do not show them in the Table. After fully relaxation, some structure go into the same phase. The same space group in the table represents the same structure.

Layer index	**Space Group**	**Enthalpy (eV/atom)**	**chirality**	**Layer index**	**Space Group**	**Enthalpy (eV/atom)**	**chirality**
112				124			
113				132	$P6_222$	-5.92983	chiral
114				143			
121				123			
122	Cmmm	-5.92967	achiral	134	$P6_422$	-5.92983	chiral
131				142			
133							
141							
144							

TABLE S7. Enthalpies and symmetries of 2×2×4 superstructures formed by incorporating various ISD layers for RbV_3Sb_5. ISD layer indexes are the same as them defined in Extended Data Fig. 3. In Table, the layer index is in bottom-to-top sequence. Certain sequences of incorporating layer are identical to those we listed in the Table, thereby we do not show them in the Table. After fully relaxation, some structure go into the same phase. The same space group in the table represents the same structure.

Layer index	**Space Group**	**Enthalpy (eV/atom)**	**chirality**	**Layer index**	**Space Group**	**Enthalpy (eV/atom)**	**chirality**
1112	Cmmm	-5.92958	achiral	1122	Fmmm	-5.92957	achiral
1113				1133			
1114				1144			
1222				1123	C222	-5.92967	chiral
1333				1124			
1444				1132			
1213	Cccm	-5.92980	achiral	1134			
1214				1142			
1232				1143			
1242				1223			
1314				1224			
1323				1233			
1343				1244			
1424				1332			
1434				1334			
1234	Fddd	-5.92981	achiral	1344			
1243				1422			
1324				1433			
1342				1442			
1423				1443			
1432							

TABLE S8. Detailed Structural information of pristine phase, 2×2×2 CDW phases of 1T-$TiSe_2$

Phase	**layer index**	**Space Group**	**Lattice parameters**	**Atomic coordinates (fractional)**			
[energy(eV)]			**($\AA$ ∘)**	**Atoms**	**X**	**Y**	**Z**
1×1×1		P-3m1	a=b=3.5338	Ti1 (1a)	0.0	0.0	0.0
(-6.02114)		(achiral)	c=6.6355	Se1 (2d)	0.33333	0.66667	0.233
			α=β=90				
			γ=120				
2×2×2	layer1:$q_1q_2q_3$	C2	a=13.4259	Ti1 (4c)	0.25621	-0.00544	0.01247
(-6.021849)	layer2:$q_1q_{\overline{2}}q_{\overline{3}}$	(chiral)	b=7.0582	Ti2 (2b)	0.5	-0.7383	0.5
			c=9.0787	Ti3 (2b)	0.5	-0.25079	0.5
			α=γ=90	Se1 (4c)	0.44975	-0.24644	0.16735
			β=137.7145	Se2 (4c)	0.70113	-0.00218	0.67005
				Se3 (4c)	0.44854	-0.75004	0.16676
				Se4 (4c)	0.69753	-0.50135	0.66279
2×2×2	layer1:$q_1q_2q_3$	P-3c1	a=b=7.0569	Ti1 (6f)	-0.48825	-0.48825	0.75
(-6.02177)	layer2:$q_{\overline{1}}q_{\overline{2}}q_{\overline{3}}$	(achiral)	c=13.264	Ti2 (2a)	0	0	0.75
			c=18.1469	Se1 (12g)	-0.33272	-0.16982	0.86745
			α=β=90	Se2 (4d)	-0.33333	0.33333	0.86673
			γ=120				
2×2×2	layer1:$q_1q_2q_3$	C2/c	a=12.2191	Ti1 (8f)	-0.25621	-0.25573	-0.25005
(-6.021844)	layer2:$q_1q_{\overline{2}}q_3$	(achiral)	b=7.0569	Ti2 (4e)	-0.5	-0.98816	-0.25
			c=18.1469	Ti3 (4e)	-0.5	-0.50042	-0.25
			α=γ=90	Se1 (8f)	-0.531	-0.25222	-0.36615
			β=132.315	Se2 (8f)	-0.53502	-0.75131	-0.36614
				Se3 (8f)	-0.78202	-0.00027	-0.36533
				Se4 (8f)	-0.78253	-0.49616	-0.36613

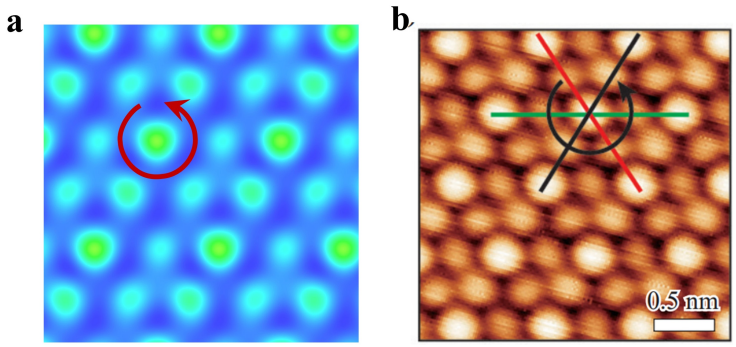

FIG. S1. **Chiral charge distribution. a,** Simulated surface charge distribution of our predicted chiral CDW and **b,** experimental observed STM image [1], respectively.

The energy regulation between coherent and incoherent CDW distortions.

To elucidate how incoherent CDW distortions can lower the system's total energy, we consider a simplified two-band diagram. In this diagram, the structure with coherent CDW distortions corresponds to a half-filled metallic state with degenerate bands (Fig. S2a, left). The introduction of incoherent distortions lifts this degeneracy, resulting in one band shifting upward in energy while the other shifts downward (Fig. S2a, right). If electrons occupy only the lower-energy band and leave the upper-energy band unfilled, the total energy of the system is lowered by the incoherent CDW distortions (Fig. S2a).

However, when the electron filling increases such that both bands are fully occupied, the total energy rises in both phases (Fig. S2b). Importantly,this energy increase is more significant for the phase with incoherent CDW distortions due to the occupation of the higher-energy band ($E_1 > E_0$).Then at a certain case, the total energy of the phase with incoherent CDW distortion may be higher than that of the phase with coherent CDW distortions, leading to a thermodynamically driven phase transition in which the system favors the coherent CDW distortions at low temperature.

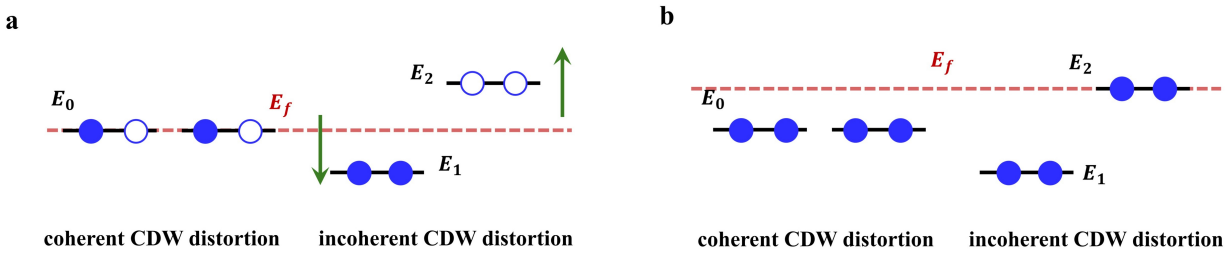

FIG. S2. **Energy tunable under CDW distortions. a,** Intrinsic electronic occupation in structures with coherent and incoherent CDW distortion. **b,** Electronic occupations after electron doping in structures with coherent and incoherent CDW distortions.

TABLE S9. Detailed Structural information of pristine phase, $\sqrt{13}\times\sqrt{13}\times1$, $\sqrt{13}\times\sqrt{13}\times2$ and $\sqrt{13}\times\sqrt{13}\times3$ CDW phases of 1T-$NbSe_2$

Phase	**layer index**	**Space Group**	**Lattice parameters**	**Atomic coordinates (fractional)**			
[energy(eV)]			**($\AA$ $\circ$)**	**Atoms**	**X**	**Y**	**Z**
1×1×1		P-3m1	a=b=3.4440	Nb1(1a)	0.63878	0.15317	0.00441
(-6.87480)		(achiral)	c=6.2141	Se1(2d)	0.71307	0.93042	0.00063
			α=β=90				
			γ=120				
$\sqrt{13}\times\sqrt{13}\times1$	layer1:$q_1q_2q_3$	P-3	a=b=12.4925	Nb1(6g)	0.00	0.00	0.00
(-6.88682)		(achiral)	c=6.2817	Nb2(6g)	0.33333	0.66667	0.27025
			α=β=90	Nb3(1a)	0	0	0
			γ=120	Se1(6g)	0.80344	0.28857	0.25664
				Se2(6g)	0.87632	0.05104	0.28245
				Se3(6g)	0.02828	0.59233	0.25088
				Se4(6g)	0.1029	0.35325	0.279
				Se5(2d)	0.33333	0.66667	0.25128
$\sqrt{13}\times\sqrt{13}\times2$	layer1:$q_1q_2q_3$	P-1	a=12.4919	Nb1(2i)	0.13758	0.03641	0.75239
(-6.8870)	layer2:$q_{\bar{1}}q_2q_3$	(achiral)	b=12.5201	Nb2(2i)	0.43766	0.81397	0.74982
			c=12.4963	Nb3(2i)	0.72362	0.59824	0.74974
			α=β=90	Nb4 (2i)	0.01564	0.37251	0.75199
			γ=60	Nb5 (2i)	0.36714	0.10182	0.74972
				Nb6 (2i)	0.65386	0.88434	0.75026
				Nb7 (2i)	0.93931	0.66856	0.75105
				Nb8 (2i)	0.29323	0.40195	0.74779
				Nb9 (2i)	0.58553	0.16988	0.75062
				Nb10 (2i)	0.87012	0.95342	0.74978
				Nb11 (2i)	0.16643	0.73198	0.74934
				Nb12 (2i)	0.50398	0.52085	0.74714

				Nb13 (2i)	0.80840	0.24479	0.75132
				Se1 (2i)	0.16789	0.17332	0.87648
				Se2 (2i)	0.47823	0.93537	0.89139
				Se3 (2i)	0.77713	0.70948	0.89363
				Se4 (2i)	0.08896	0.47836	0.87721
				Se5 (2i)	0.40351	0.23766	0.88785
				Se6 (2i)	0.70452	0.00812	0.89388
				Se7 (2i)	0.00529	0.78244	0.89134
				Se8 (2i)	0.32060	0.55166	0.87607
				Se9 (2i)	0.62591	0.32007	0.87515
				Se10 (2i)	0.94118	0.08115	0.87913
				Se11 (2i)	0.24623	0.85561	0.87683
				Se12 (2i)	0.55146	0.63367	0.88922
				Se13 (2i)	0.85034	0.39928	0.87759
				Se14 (2i)	0.30013	0.98749	0.61220
				Se15(2i)	0.60281	0.76059	0.60650
				Se16 (2i)	0.90350	0.53356	0.61009
				Se17 (2i)	0.21737	0.29342	0.62603
				Se18 (2i)	0.52948	0.05953	0.60853
				Se19 (2i)	0.82816	0.83374	0.60680
				Se20 (2i)	0.13800	0.59743	0.62177
				Se21 (2i)	0.45770	0.36944	0.61990
				Se22 (2i)	0.75652	0.13412	0.61034
				Se23 (2i)	0.06032	0.91293	0.62437
				Se24 (2i)	0.36463	0.68758	0.62301
				Se25 (2i)	0.68295	0.44881	0.62250
				Se26 (2i)	0.98643	0.21871	0.62381
$\sqrt{13}\times\sqrt{13}\times3$	layer1:$q_1q_2q_3$	$P3_1$	a=b=12.4908	Nb1 (3a)	0.09845	0.76881	0.16813
(-6.88701)	layer2:$q_{\bar{1}}q_2q_3$	(chiral)	c=18.7800	Nb2 (3a)	0.17456	0.54579	0.16771
	layer3:$q_{\bar{1}}q_{\bar{2}}q_3$		α=β=90	Nb3 (3a)	0.24495	0.32864	0.16716
			γ=120	Nb4 (3a)	0.31029	0.10126	0.16847

Nb5 (3a)	0.393	0.83269	0.16558
Nb6 (3a)	0.46205	0.61565	0.16687
Nb7 (3a)	0.53117	0.3989	0.16726
Nb8 (3a)	0.61646	0.1313	0.16539
Nb9 (3a)	0.67824	0.90034	0.16698
Nb10 (3a)	0.74697	0.68429	0.16648
Nb11 (3a)	0.82156	0.46262	0.166
Nb12 (3a)	0.94611	0.25111	0.16546
Nb13 (3a)	0.97484	0.97464	0.16753
Se1 (3a)	0.26529	0.90383	0.25127
Se2 (3a)	0.3379	0.6672	0.26101
Se3 (3a)	0.41026	0.44047	0.26244
Se4 (3a)	0.48998	0.20855	0.25159
Se5 (3a)	0.56401	0.96775	0.25862
Se6(3a)	0.63626	0.73895	0.26272
Se7 (3a)	0.71088	0.51317	0.26077
Se8 (3a)	0.79432	0.282	0.25088
Se9 (3a)	0.86855	0.0501	0.25051
Se10 (3a)	0.94544	0.81152	0.2527
Se11 (3a)	0.02486	0.58735	0.25137
Se12 (3a)	0.10785	0.36418	0.2599
Se13 (3a)	0.17237	0.1293	0.25195
Se14 (3a)	0.21039	0.71731	0.07473
Se15 (3a)	0.28575	0.49107	0.073
Se16 (3a)	0.35888	0.26265	0.07403
Se17 (3a)	0.43372	0.02378	0.08274
Se18 (3a)	0.51277	0.78998	0.07076
Se19 (3a)	0.58476	0.56378	0.0713
Se20 (3a)	0.65761	0.3272	0.08122
Se21 (3a)	0.74956	0.09971	0.0808
Se22 (3a)	0.81323	0.86442	0.07289
Se23 (3a)	0.89582	0.64314	0.08247
Se24 (3a)	0.97562	0.4185	0.0831

				Se25 (3a)	0.05399	0.17912	0.08242
				Se26 (3a)	0.12769	0.9488	0.0828
$\sqrt{13}\times\sqrt{13}\times3$	layer1:$q_1q_2q_3$	$P3_2$	a=b=12.4908	Nb1 (3a)	0.09999	0.77086	0.16843
(-6.8869)	layer2:$q_{\bar{1}}q_{\bar{2}}q_3$	(chiral)	c=18.7800	Nb2 (3a)	0.17503	0.54486	0.16596
	layer3:$q_{\bar{1}}q_2q_3$		α=β=90	Nb3 (3a)	0.24514	0.32883	0.16665
			γ=120	Nb4 (3a)	0.31116	0.10311	0.16834
				Nb5 (3a)	0.39224	0.83232	0.16738
				Nb6 (3a)	0.46085	0.61446	0.16693
				Nb7 (3a)	0.53073	0.39858	0.16731
				Nb8 (3a)	0.61749	0.13142	0.16577
				Nb9 (3a)	0.67822	0.90079	0.16738
				Nb10 (3a)	0.74677	0.68442	0.1665
				Nb11 (3a)	0.82303	0.46463	0.16583
				Nb12 (3a)	0.94675	0.25197	0.1648
				Nb13 (3a)	0.97544	0.97668	0.16775
				Se1 (3a)	0.26462	0.90433	0.25349
				Se2 (3a)	0.33626	0.66497	0.26115
				Se3 (3a)	0.41017	0.44001	0.26271
				Se4 (3a)	0.49048	0.20871	0.25169
				Se5 (3a)	0.56458	0.96836	0.26005
				Se6(3a)	0.63568	0.73867	0.26246
				Se7 (3a)	0.71105	0.51352	0.26043
				Se8 (3a)	0.79507	0.28242	0.25098
				Se9 (3a)	0.86867	0.05107	0.25064
				Se10 (3a)	0.94605	0.81269	0.25251
				Se11 (3a)	0.02534	0.58685	0.24927
				Se12 (3a)	0.10833	0.36509	0.25855
				Se13 (3a)	0.17266	0.12998	0.25189
				Se14 (3a)	0.21145	0.71898	0.07493
				Se15 (3a)	0.28653	0.49068	0.07085
				Se16 (3a)	0.3598	0.26358	0.07347
				Se17 (3a)	0.43339	0.02317	0.08464
				Se18 (3a)	0.5116	0.78995	0.0728

Se19 (3a)	0.58495	0.56401	0.07131
Se20 (3a)	0.65844	0.32725	0.0817
Se21 (3a)	0.74936	0.09971	0.08022
Se22 (3a)	0.8134	0.86501	0.07361
Se23 (3a)	0.89701	0.64455	0.08273
Se24 (3a)	0.97515	0.41801	0.08131
Se25 (3a)	0.05429	0.17929	0.08197
Se26 (3a)	0.12817	0.94967	0.08263

Chirality-locking responses

Structures with left and right chirality can be interconverted via mirror symmetry, such as M_z. Mirror symmetry ensures identical band structures in the same system with opposite chirality, but the spin texture in momentum space reverses, yielding an opposite spin polarization in the band structure. This reversal of spin polarization leads to an inversion in the direction of spin currents and the sign of spin accumulation at the boundaries (Fig. S3 step I). Consequently, the local magnetic field and the Berry curvature on the top and bottom surfaces of the material also reverse as the change of chirality (Fig. S3). This reversal will then reflect in the correlation-driven spin-anomalous dual-Hall effect, where the Hall conductivity exhibits opposite signs in material with right (σ^r_{xy}) and left chirality (σ^l_{xy}), given by $\sigma^r_{xy} = -\sigma^l_{xy}$. Therefore, controlling material chirality offers a means to tune the sign of the Hall conductivity, warranting further experimental investigation.

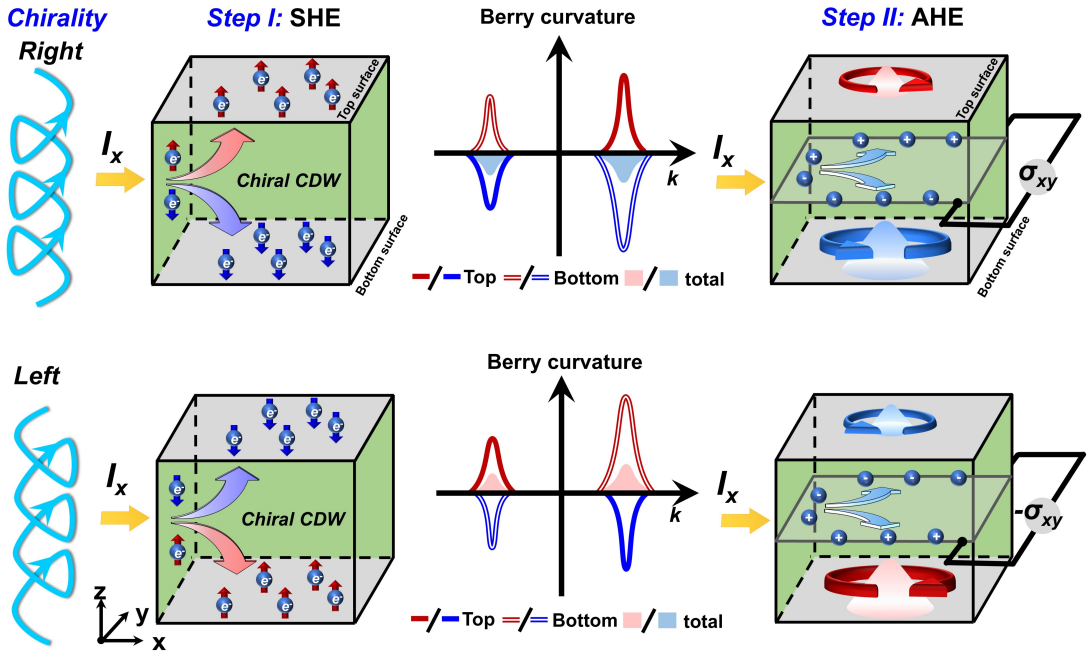

FIG. S3. **Chirality-locking dual-Hall effect.** A sketch illustration of correlation-driven spin-anomalous dual-Hall effect in nonmagnetic chiral CDW materials with opposite chirality. Step I represents spin Hall effect (SHE) of chiral system emerging from current injection along z-direction. The middle panel shows the Berry curvature distributions on the top and bottom surfaces, induced by the combination of spin accumulation and chiral symmetry breaking, which breaks P, T, and PT symmetry. The imblance distribution of Berry curvature on the top and bottom surface induces the anomalous Hall effect (AHE), shown in Step II. Top and bottom panels manifest two scenarios of dual-Hall effect with differect chirality.

[1] Kim, H. et al. Microscopic Origin of Chiral Charge Density Wave in $TiSe_2$. Preprint at https://arxiv.org/abs/2401.01669 (2024).