Electronic Supplementary Information for:

Copper Halide Diselenium: Predicted Two-Dimensional Materials with Ultrahigh Anisotropic Carrier Mobilities

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S1. van der Waals DFT methods

Table S1. Structural parameters and cleavage energy of bulk $CuXSe_2$ considering different van der Waals (vdW) DFT methods (distances in Å, angles in °, cleavage energy in J/m^2).

$CuXSe_2$	vdW DFT method	*a*	*b*	*c*	*γ*	Cleavage Energy
X=Cl	optB88	4.80	7.81	14.70	135.72	0.35
	SCAN-rVV10	4.65	7.66	14.39	135.71	0.28
	DFT-dDsC	4.75	7.75	14.61	135.44	0.26
	FIA	4.82	7.79	14.69	135.49	0.19
	PBE-D2	4.87	7.56	14.19	134.78	0.24
	Exp.	4.63	7.68	14.55	132.79	-
X=Br	optB88	4.82	7.87	11.90	113.91	0.33
	SCAN-rVV10	4.65	7.79	11.72	115.37	0.27
	DFT-dDsC	4.74	7.79	11.81	114.89	0.29
	FIA	4.84	7.79	12.14	114.28	0.18
	PBE-D2	4.88	7.86	12.38	114.53	0.23
	Exp.	4.64	7.88	11.18	103.44	-

references

optB88: J. Klimes, D. R. Bowler, and A. Michaelides, Journal of Physics: Condensed Matter 22, 022201 (2009).

SCAN-rVV10: H. Peng, Z.-H. Yang, J. P. Perdew, and J. Sun, Physical Review X 6, 041005 (2016).

DFT-dDsC: S. N. Steinmann and C. Corminboeuf, The Journal of Chemical Physics 134, 044117 (2011).

FIA: T. Gould, S. Lebegue, J. G. Angyann, and T. Bucko, J. Chem. Theor. Comput. 12, 5920 (2016).

S2. *ab initio* molecular dynamics (AIMD) simulations: computational details

ab initio (DFT) molecular dynamics simulations were performed with VASP for the periodic supercell of the $P2_1$ phases. A 4×3 supercell is used. Starting from each structure at ambient pressure and T=0K, AIMD calculations were performed at 300 K, and 450 K. The time step was 1 fs, and the total simulation time was as long as 5 ps. Brillouin zone integration is restricted to the Γ point of the supercell. A canonical NVT (N: constant number of particles, V: constant volume, and T: constant temperature) ensemble was adopted for the AIMD calculations using the algorithm of Nose, as implemented in the VASP code. In our DFT calculations on 2D CuXSe2 phases, the PBE functional is used throughout this work. The projector-augmented wave (PAW) method was used for the core-electron representation. After preliminary convergence tests, we used a cutoff energy E_{cut}= 400 eV for the valence basis set.

S3. Optimized bulk crystal structures of Se, CuCl and CuBr

POSCAR file of bulk Se

```
Se3
   1.0000000000000000
     4.2674004588667813   -0.0000000000031202    0.0000000000000000
    -2.1337002293888814    3.6956772055057616   -0.0000000000000000
     0.0000000000000000   -0.0000000000000000    5.0975389853209814
   Se
     3
Direct
  0.9999999987741006  0.2324636600416699  0.6666666980596716
  0.7675363270301615  0.7675363432286600 -0.0000000000000000
  0.2324636627098858  0.0000000000000000  0.3333333725943888
```

POSCAR file of bulk CuCl

```
Cu1 Cl1
   1.0000000000000000
     3.7550907217422735   -0.0000000000000000    0.0000000000000000
     1.8775453608711368    3.2520039585524723   -0.0000000000006158
     1.8775453608711368    1.0840013195496629    3.0660187353185049
   Cu   Cl
     1     1
Direct
  0.0000000000000000 -0.0000000000000000 -0.0000000000000000
  0.7499999964257142  0.7500000131744073  0.7500000563555744
```

POSCAR file of bulk CuBr

```
Cu1 Br1
   1.0000000000000000
     3.9622944839815908    0.0000000000000000    0.0000000000000000
     1.9811472419907954    3.4314476803461109   -0.0000000000005923
     1.9811472419907954    1.1438158934809199    3.2351998987764121
   Cu   Br
     1     1
Direct
  0.7500000501571833  0.7500000106859233  0.7500000071193540
 -0.0000000000000000  0.0000000000000000  0.0000000000000000
```

S4. Optimized bulk crystal structures of $CuClSe_2$ and $CuBrSe_2$

POSCAR file of bulk $CuClSe_2$

```
Cu2 Se4 Cl2
   1.0000000000000000
     4.8400000000000007    0.0000000000000000    0.0000000000000000
     0.0000000000000000    7.5599999999999996    0.0000000000000000
     0.0000000000000000    0.0000000000000000   14.2140407961451185
   Cl   Cu   Se
     2     2     4
Direct
  0.3891472879432872  0.3473208855960385  0.6604860956716724
  0.8874338283006332  0.3449078951755667  0.8692365288953942
  0.6428067609579973  0.5143621575782341  0.7644184646726834
  0.1417834418910231  0.1794970392781846  0.7647072704067747
  0.9738959846544886  0.7039969511795665  0.6942502212099648
  0.3080929326520869  0.7037548969978684  0.8350896847604974
  0.8081749032365443  0.9882526810089496  0.6947605979116637
  0.4733248852111773  0.9880698305049256  0.8354522551856378

  0.00000000E+00  0.00000000E+00  0.00000000E+00
  0.00000000E+00  0.00000000E+00  0.00000000E+00
  0.00000000E+00  0.00000000E+00  0.00000000E+00
  0.00000000E+00  0.00000000E+00  0.00000000E+00
  0.00000000E+00  0.00000000E+00  0.00000000E+00
  0.00000000E+00  0.00000000E+00  0.00000000E+00
  0.00000000E+00  0.00000000E+00  0.00000000E+00
  0.00000000E+00  0.00000000E+00  0.00000000E+00
```

POSCAR file of bulk $CuBrSe_2$

```
Cu2 Se4 Br2
   1.0000000000000000
     4.8600000000000003    0.0000000000000000    0.0000000000000000
     0.0000000000000000    7.7000000000000002    0.0000000000000000
     0.0000000000000000    0.0000000000000000   14.1794433052653055
   Br   Cu   Se
     2     2     4
Direct
  0.3871435868035642  0.8682823193372968  0.7070289924125482
  0.8871436109117818  0.8686106316154394  0.9363784886588796
  0.6362448852199248  0.0424167199400214  0.8217586277194405
  0.1362448851996816  0.6944763274003878  0.8216505563519897
  0.9699286077686596  0.2283604628058481  0.7518915935430005
  0.3028668754478406  0.2284800279181673  0.8917226141009730
  0.8028668754680840  0.5084129693322262  0.7516865699704572
  0.4699286561267914  0.5085325036398392  0.8915159555998567

  0.00000000E+00  0.00000000E+00  0.00000000E+00
  0.00000000E+00  0.00000000E+00  0.00000000E+00
  0.00000000E+00  0.00000000E+00  0.00000000E+00
  0.00000000E+00  0.00000000E+00  0.00000000E+00
  0.00000000E+00  0.00000000E+00  0.00000000E+00
  0.00000000E+00  0.00000000E+00  0.00000000E+00
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  0.00000000E+00  0.00000000E+00  0.00000000E+00
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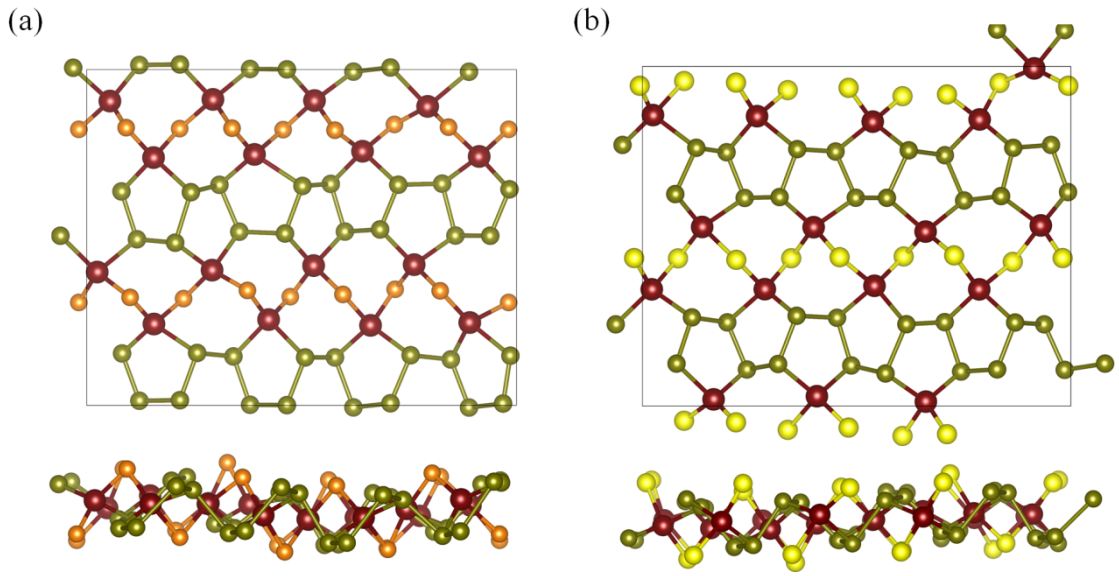

Figure S1. Snapshots of ab initio molecular dynamics (AIMD) simulation after 5 *p*s at 450 K for (a) $CuClSe_2$ and (b) $CuBrSe_2$ monolayers

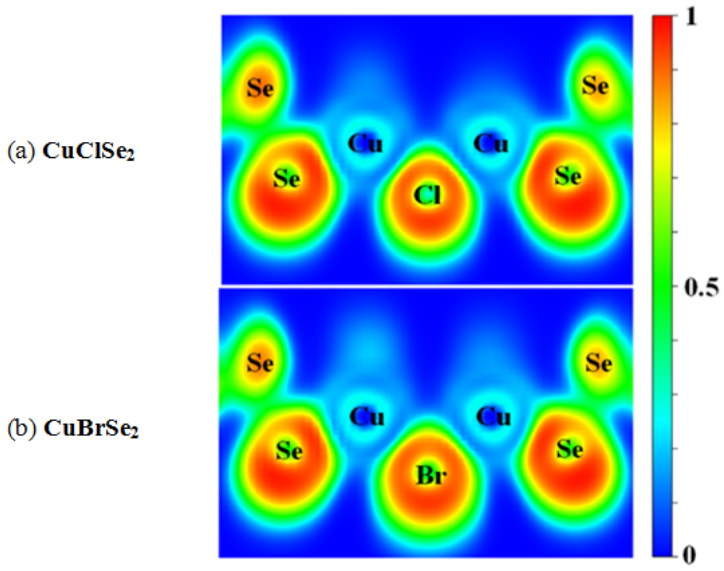

Figure S2. Electron localization function (ELF) contour maps sliced along planes containing directly connected Se-Cu-X-Cu-Se atoms for $CuClSe_2$ (a) and $CuBrSe_2$ (b) monolayers. The position of atoms is marked with the corresponding chemical symbols.