Screening procedure for structurally and electronically matched contact layers for high-performance solar cells: hybrid perovskites

Keith T. Butler*

Centre for Sustainable Chemical Technologies and Department of Chemistry, University of Bath, Claverton Down, Bath BA2 7AY, UK

Yu Kumagai

Materials Research Center for Element Strategy, Tokyo Institute of Technology, Yokohama 226-8503, Japan

Fumiyasu Oba

Materials Research Center for Element Strategy, Tokyo Institute of Technology, Yokohama 226-8503, Japan
Materials and Structures Laboratory, Tokyo Institute of Technology, Yokohama 226-8503, Japan and
Center for Materials Research by Information Integration, National Institute for Materials Science, Tsukuba 305-0047, Japan

Aron Walsh

Centre for Sustainable Chemical Technologies and Department of Chemistry, University of Bath, Claverton Down, Bath BA2 7AY, UK and
Global E^3 Institute and Department of Materials Science and Engineering, Yonsei University, Seoul 120-749, Korea

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I. BAND ENERGY DATA

The data used for estimation of band alignments is presented in table I. Where possible experimental photoemission data was used. The data from reference[1] was estimated based on Mulliken electronegativity and should be treated with some added caution. The data along with a simple python script for aligning to a given absorber material has been made available on an open repository[2], we encourage the interested reader to test the effects of varying sensitivity and band positions.

Species	Eg	EA	IP	Ref IP	Ref Eg	Ref EA
Al2O3	8.7	3.72	12.42		[3]	[4]
AlAs	2.36	1.71	4.07	[5]	[5]	
AlN	6.03	1.9	7.93	[6]	[7]	
AlP	2.42	2.51	4.93	[8]	[8]	
AlSb	1.6	3.6	5.2	[9]	[9]	
BaO	3.99	0.57	4.56		[10]	[11]
BaS	3.81	0.84	4.65		[10]	[11]
BaSe	3.66	0.95	4.61		[10]	[11]
BaTe	3.4	1.43	4.83		[12]	[11]
Bi2O3	2.77	4.63	7.4	[13]	[13]	
BN	6.2	4.5	10.7		[14]	[9]
C	5.48	0.37	5.85	[5]	[5]	
CaF2	12.1	-0.3	11.8		[14]	[15]
CaO	6.8	0.7	7.5		[14]	[11]
CaS	4.60	1.85	6.45		[16]	[11]
CaSe	4.87	2.32	7.19		[17]	[11]
CaTe	4.07	3.53	7.6		[17]	[11]
CdO	0.78	5.24	6.02		[18]	[18]
CdS	2.42	4.5	6.92		[8]	[8]
CdSe	1.74	4.56	6.3		[8]	[8]

CdTe	1.44	4.28	5.72		[8]	[8]
CsI	6.2	-0.3	5.9		[19]	[19]
Cu2O	2.1	3.56	5.66	[20]	[20]	
Fe2O3	2.1	4.85	6.95	[21]	[21]	
Ga2O3	4.8	3.2	8		[14]	[22]
GaAs	1.42	4.07	5.49		[23]	[8]
GaN	3.43	4.10	7.53		[7]	[10]
GaP	2.26	4.1	6.36		[8]	[14]
GaSb	0.73	4.06	4.79		[8]	[8]
Ge	0.74	4.01	4.75	[5]	[5]	
GeO2	5.35	2.93	8.28		[14]	[24]
HfO2	5.7	2	7.7		[25]	[26]
In2O3	2.7	4.3	7	[20]	[27]	
InAs	0.36	4.9	5.26		[8]	[8]
InN	0.65	5.8	6.45		[28]	[29]
InP	1.34	4.38	5.72		[8]	[8]
InSb	0.24	4.59	4.83		[30]	[9]
KBr	7.4	0.8	8.2		[19]	[19]
KCl	8.9	0.5	9.4		[19]	[19]
KF	10.7	0	10.7		[19]	[19]
KI	6	1.2	7.2		[19]	[19]
KTaO3	3.53	3.8	7.33	[13]	[13]	
La2O3	6	2.5	8.5		[31]	[31]
LiBr	7.6	0.2	7.8		[19]	[19]
LiF	13.6	1.35	14.95		[19]	[25]
MgO	7.8	-0.65	7.15		[5]	[5]
MgS	4.87	3.15	8.02		[25]	[11]
MgSe	4.05	4.5	8.55		[25]	[11]

MnO	3.28	1.8	5.08	[21]	[21]	
MoO3	2.8	2.20	5		[32]	[33]
NaBr	7.5	0.4	7.9		[19]	[19]
NaCl	8.5	0.5	9		[19]	[19]
NaF	11.6	1.35	12.95		[19]	[25]
NaNbO3	3.43	3.8	7.23	[13]	[13]	
NiO	3.6	1.73	5.33	[34]	[34]	
PbS	0.4	4.6	5		[14]	[35]
RbBr	7.4	0.4	7.8		[19]	[19]
RbCl	8.2	0.5	8.7		[19]	[19]
RbF	10.35	-0.1	10.25		[19]	[19]
RbI	6.1	1.2	7.3		[19]	[19]
Sb2O3	3	4.17	7.17	[13]	[13]	
Si	1.17	3.98	5.15	[5]	[5]	
Si3N4	5.3	2.1	7.4		[36]	[36]
SiC	2.36	4	6.36	[37]	[6]	
SiO2	9	0.9	8.1		[38]	[38]
SnO2	3.6	5.3	8.9		[21]	[21]
SnS2	2.1	4.2	6.3		[39]	[39]
SnSe2	1	4.35	5.35		[39]	[39]
SrO	5.2	0.67	5.87		[14]	[11]
SrS	4.3	1.35	5.65		[40]	[11]
SrSe	4.42	1.77	6.19		[17]	[11]
SrTe	3.73	2.4	6.13		[17]	[11]
SrTiO3	3.2	4.08	7.28	[13]	[13]	
Ta2O5	4.5	3.2	7.7		[3]	[41]
TiO2	3.05	4.2	7.21	[21]	[3]	
V2O5	2.30	4	6.3		[42]	[43]

WO3	2.95	3.33	6.28		[23]	[44]
Zn2TiO4	3.21	3.25	6.46	[13]	[13]	
ZnO	3.3	4.2	7.5	[45]	[46]	
ZnS	3.7	3.8	7.5	[5]	[14]	
ZnSe	2.82	4	6.82		[8]	[8]
ZnTe	2.26	3.53	5.79		[8]	[8]
ZrO2	5.8	2.6	8.4		[36]	[25]
CuI	3.37	2.43	5.8	[18]	[18]	[18]
CuI	2.18	3.02	5.2	[18]	[18]	[18]
CuI	4.62	2.28	5.3	[18]	[18]	[18]
BaTiO3	3.3	4.21	7.51	[1]	[1]	
CdFe2O4	2.3	4.89	7.19	[1]	[1]	
CuO	1.7	4.89	6.59	[1]	[1]	
FeTiO3	2.8	4.56	7.36	[1]	[1]	
Hg2Nb2O	1.8	5.05	6.85	[1]	[1]	
Nb2O5	3.4	4.16	7.56	[1]	[1]	
PbO	2.8	4.46	7.26	[1]	[1]	
PbFe12O19	2.3	5.2	7.5	[1]	[1]	
YFeO3	2.6	4.6	7.2	[1]	[1]	
FeS2	0.95	4.96	5.91	[1]	[1]	
HfS2	1.13	4.63	5.76	[1]	[1]	
HgS	2	4.74	6.74	[1]	[1]	
In2S3	2	3.69	5.69	[1]	[1]	
MoS2	1.17	4.43	5.6	[1]	[1]	
ZrS2	1.82	4.28	6.1	[1]	[1]	
Ag2O	1.2	4.69	5.89	[1]	[1]	
AlTiO3	3.6	3.64	7.24	[1]	[1]	
Ce2O3	2.4	4	6.4	[1]	[1]	

CoO	2.6	4.39	6.99	[1]	[1]
CoTiO3	2.25	4.64	6.89	[1]	[1]
CuTiO3	2.99	4.32	7.31	[1]	[1]
FeO	2.4	4.33	6.73	[1]	[1]
Fe3O4	0.1	5.73	5.83	[1]	[1]
FeOOH	2.6	5.08	7.68	[1]	[1]
HgO	1.9	5.13	7.03	[1]	[1]
Hg2Ta2O7	1.8	5.34	7.14	[1]	[1]
KNbO3	3.3	3.64	6.94	[1]	[1]
LaTi2O7	4	3.9	7.9	[1]	[1]
LiNbO3	3.5	3.77	7.27	[1]	[1]
LiTaO3	4	3.55	7.55	[1]	[1]
MgTiO3	3.7	3.75	7.45	[1]	[1]
MnO2	0.25	5.83	6.08	[1]	[1]
MnTiO3	3.1	4.04	7.14	[1]	[1]
Nd2O3	4.7	2.87	7.57	[1]	[1]
NiTiO3	2.18	4.7	6.88	[1]	[1]
PdO	1	5.29	6.29	[1]	[1]
Pr2O3	3.9	3.24	7.14	[1]	[1]
Sm2O3	4.4	3.07	7.47	[1]	[1]
SnO	4.2	3.59	7.79	[1]	[1]
Tb2O3	3.8	3.44	7.24	[1]	[1]
Tl2O3	1.6	4.55	6.15	[1]	[1]
Yb2O3	4.9	3.02	7.92	[1]	[1]
ZnTiO3	3.06	4.27	7.33	[1]	[1]
Ag2S	0.92	4.5	5.42	[1]	[1]
AgAsS2	1.95	4.51	6.46	[1]	[1]
AgSbS2	1.72	4.51	6.23	[1]	[1]

As2S3	2.5	4.58	7.08	[1]	[1]
Ce2S3	2.1	3.59	5.69	[1]	[1]
CoS	0	5.17	5.17	[1]	[1]
CoS2	0	5.49	5.49	[1]	[1]
CoAsS	0.5	4.96	5.46	[1]	[1]
CuS	0	5.27	5.27	[1]	[1]
Cu2S	1.1	4.44	5.54	[1]	[1]
CuS2	0	5.57	5.57	[1]	[1]
Cu3AsS4	1.28	4.75	6.03	[1]	[1]
CuFeS2	0.35	4.97	5.32	[1]	[1]
Cu5FeS4	1	4.55	5.55	[1]	[1]
CuInS2	1.5	4.06	5.56	[1]	[1]
CuIn5S8	1.26	4.09	5.35	[1]	[1]
Dy2S3	2.85	3.36	6.21	[1]	[1]
FeS	0.1	4.97	5.07	[1]	[1]
Fe3S4	0	5.18	5.18	[1]	[1]
FeAsS	0.2	5.01	5.21	[1]	[1]
Gd2S3	2.55	3.57	6.12	[1]	[1]
HgSb4S8	1.68	4.81	6.49	[1]	[1]
La2S3	2.91	3.25	6.16	[1]	[1]
MnS	3	3.31	6.31	[1]	[1]
MnS2	0.5	4.99	5.49	[1]	[1]
Nd2S3	2.7	3.3	6	[1]	[1]
NiS	0.4	5.03	5.43	[1]	[1]
NiS2	0.3	5.39	5.69	[1]	[1]
OsS2	2	4.74	6.74	[1]	[1]
Pb10Ag3Sb11S28	1.39	4.59	5.98	[1]	[1]
Pb2As2S5	1.39	4.71	6.1	[1]	[1]

PbCuSbS3	1.23	4.61	5.84	[1]	[1]
Pb5Sn3Sb2S14	0.65	4.95	5.6	[1]	[1]
Pr2S3	2.4	3.43	5.83	[1]	[1]
PtS2	0.95	5.53	6.48	[1]	[1]
Rh2S3	1.5	4.61	6.11	[1]	[1]
RuS2	1.38	4.89	6.27	[1]	[1]
Sb2S3	1.72	4.72	6.44	[1]	[1]
Sm2S3	2.6	3.39	5.99	[1]	[1]
SnS	1.01	4.66	5.67	[1]	[1]
Tb2S3	2.5	3.51	6.01	[1]	[1]
TiS2	0.7	4.76	5.46	[1]	[1]
TlAsS2	1.8	4.16	5.96	[1]	[1]
WS2	1.35	4.86	6.21	[1]	[1]
ZnS2	2.7	4.21	6.91	[1]	[1]
Zn3In2S6	2.81	3.59	6.4	[1]	[1]

TABLE I: Band energies used in the initial screening for contact materials. Material chemical formula, ionisation potential (IP), electron affinity (EA) and band gap (E_g) as well as the source of the data are presented. All numerical values are given in eV. All values from reference[1] are based on band gap and Mulliken electronegativity rather than experimental measurement of IP/EA.

II. VALIDATION OF SITE MATCHING

We can consider an illustrative example of the ASO factor to test its robustness. We look at the heterointerface between ZnO and GaN, this pairing has been reported with several different crystallographic and chemical interface compositions. Notably the (0001)/(0001) interface is commonly reported and in nanowires the (11-20)/(11-20) interface is observed[47].

GaN		ZnO		Strain		ASO	LS
Surface	Multiplicity	Surface	Multiplicity	u	v		
(0001)	(1,1)	(0001)	(1,1)	1.96	1.96	1.00	33.78
(0001)	(1,5)	(01-10)	(1,3)	1.96	1.97	0.36	12.14
(0001)	(1,2)	(01-11)	(1,1)	1.96	3.76	0.50	12.95
(10-10)	(1,1)	(10-10)	(1,1)	1.96	0.46	1.00	45.25
(11-20)	(1,1)	(11-20)	(1,1)	0.46	1.96	1.00	45.25
(11-21)	(3,1)	(10-11)	(5,1)	1.89	0.88	0.38	15.93
(11-21)	(1,1)	(11-21)	(1,1)	1.96	0.88	1.00	41.32

TABLE II. Lattice and site matching factors for the GaN/ZnO heterointerface. Miller indices and supercell expansion for each material is given as are the strains (in percentage) of the surface vectors, the ASO factor calculated as described in the main article and the ELS (here LS due to neglect of electronic offset) factor also calculated as in the main text.

Starting with the lattice matching criterion we identify a number of different possible combinations, they are listed in Table II along with the strain in u and v surface vectors. The experimentally observed interfaces are identified along with (0001)/(01-11), (0001)/(10-10), (10-10)/(10-10), (11-21)/(10-11) and (11-21)/(11-21); in total 7 possible interfaces.

Of these the final three are not reported to our knowledge, this is probably related to the lack of any GaN (10-10) or (11-21) oriented substrates on which they could grow. We calculate the ASO for the interfaces, they are also reported in Table II. By combining the ASO and lattice strain components of the ELS (here we ignore band offset as we are interested only in growth) we calculate values for each of the interfaces.

Of the four possible interfaces identified from the combination of known existing GaN substrates with the appropriate orientation and lattice matching the (0001)/(0001) and (11-20)/(11-20) interfaces have significantly larger LS numbers than the alternative interfaces. This matches with the experimentally reported interface structures.

* k.t.butler@bath.ac.uk

[1] Y. Xu and M. A. Schoonen, Am. Mineral. **85**, 543 (2000).

[2] "Bandalignmentfromdata," `https://github.com/keeeto/BandAliginingFromData/`, accessed: 2015-12-03.

[3] G. D. Wilk, R. M. Wallace, and J. M. Anthony, J. Appl. Phys. **89**, 5243 (2001).

[4] S. R. Desai, H. Wu, C. M. Rohlfing, and L.-S. Wang, J. Chem. Phys. **106**, 1309 (1997).

[5] A. Grüneis, G. Kresse, Y. Hinuma, and F. Oba, Phys. Rev. Lett. **112**, 096401 (2014).

[6] M. E. Levinshtein, S. L. Rumyantsev, and M. S. Shur, *Properties of Advanced Semiconductor Materials: GaN, AIN, InN, BN, SiC, SiGe* (John Wiley & Sons, 2001) p. 194.

[7] H. Teisseyre, P. Perlin, T. Suski, I. Grzegory, S. Porowski, J. Jun, A. Pietraszko, and T. D. Moustakas, J. Appl. Phys. **76**, 2429 (1994).

[8] S. Fonash, *Solar Cell Device Physics* (Elsevier, 2012) p. 352.

[9] S. Adachi, *Handbook on Physical Properties of Semiconductors, Volume 2* (Springer Science & Business Media, 2004).

[10] Y. Kaneko and T. Koda, J. Cryst. Growth **86**, 72 (1988).

[11] K. Y. Tsou and E. B. Hensley, J. Appl. Phys. **45**, 47 (1974).

[12] F. E. Martin and E. B. Hensley, Physical Review **163**, 219 (1967).

[13] Y. Matsumoto, J. Sol. State Chem. **126**, 227 (1996).

[14] O. Madelung, *Semiconductors: Data Handbook* (Springer Science & Business Media, 2012) p. 691.

[15] B. Quiniou, W. Schwarz, Z. Wu, R. M. Osgood, Q. Yang, and J. M. Phillips, Appl. Phys. Lett. **60**, 183 (1992).

[16] W. Lehmann, J. Electrochem. Soc. **117**, 1389 (1970).

[17] W. H. Strehlow and E. L. Cook, Journal of Physical and Chemical Reference Data **2**, 163 (1973).

[18] K. T. Butler, Phys. Status Solidi (a) **212**, 1461 (2015).

[19] R. T. Poole, J. G. Jenkin, J. Liesegang, and R. C. G. Leckey, Phys. Rev. B **11**, 5179 (1975).

[20] J. Deuermeier, J. Gassmann, J. Brotz, and A. Klein, J. Appl. Phys. **109**, 113704 (2011).

[21] V. Stevanović, S. Lany, D. S. Ginley, W. Tumas, and A. Zunger, Phys. Chem. Chem. Phys. **16**, 3706 (2014).

[22] T. Thomas, X. Guo, M. Chandrashekhar, C. B. Poitras, W. Shaff, M. Dreibelbis, J. Reiherzer, K. Li, F. J. DiSalvo, M. Lipson, and M. Spencer, J. Cryst. Growth **311**, 4402 (2009).

[23] K. J. Reynolds, J. A. Barker, N. C. Greenham, R. H. Friend, and G. L. Frey, J. Appl. Phys.

92, 7556 (2002).

[24] L.-S. Wang, H. Wu, S. R. Desai, J. Fan, and S. D. Colson, J. Phys. Chem. **100**, 8697 (1996).

[25] B. D. Pelatt, R. Ravichandran, J. F. Wager, and D. A. Keszler, J. Am. Chem. Soc. **133**, 16852 (2011).

[26] S. Monaghan, P. Hurley, K. Cherkaoui, M. Negara, and A. Schenk, Solid State Electron. **53**, 438 (2009).

[27] A. Walsh and C. R. A. Catlow, J. Mater. Chem. **20**, 10438 (2010).

[28] Q. Guo and A. Yoshida, Japanese J. Appl. Phys. **33**, 2453 (1994).

[29] S. X. Li, K. M. Yu, J. Wu, R. E. Jones, W. Walukiewicz, J. W. Ager, W. Shan, E. E. Haller, H. Lu, and W. J. Schaff, Phys. Rev. B **71**, 161201 (2005).

[30] L. I. Berger, *Semiconductor Materials* (CRC Press, 1996) p. 496.

[31] P. W. Peacock and J. Robertson, Appl. Phys. Lett. **83**, 2025 (2003).

[32] R. Juryska, Phys. Status Solidi (b) **72**, K161 (1975).

[33] A. Galtayries, S. Wisniewski, and J. Grimblot, J. Electron. Spectrosc. **87**, 31 (1997).

[34] J. He, H. Lindström, A. Hagfeldt, and S.-E. Lindquist, Sol. Ener. Mater. Sol. Cells **62**, 265 (2000).

[35] R. A. Knapp, Phys. Rev. **132**, 1891 (1963).

[36] J. Robertson, J. Vac. Sci. Tech. B **18**, 1785 (2000).

[37] M. Bozack, Phys. Status Solidi (b) **202**, 549 (1997).

[38] S.M.Sze, *Semiconductor Devices: Physics and Technology* (2008).

[39] R. H. Williams, R. B. Murray, D. W. Govan, J. M. Thomas, and E. L. Evans, Journal of Physics C: Solid State Physics **6**, 3631 (1973).

[40] J. M. Fitz-Gerald, J. Hoekstra, P. D. Rack, and J. D. Fowlkes, Appl. Phys. Lett. **82**, 3466 (2003).

[41] B. C.-m. Lai, J. Electrochem. Soc. **146**, 266 (1999).

[42] S. F. Cogan, N. M. Nguyen, S. J. Perrotti, and R. D. Rauh, J. Appl. Phys. **66**, 1333 (1989).

[43] H.-J. Zhai and L.-S. Wang, J. Chem. Phys. **117**, 7882 (2002).

[44] C. W. Walter, C. F. Hertzler, P. Devynck, G. P. Smith, and J. R. Peterson, J. Chem. Phys. **95**, 824 (1991).

[45] R. A. Powell, W. E. Spicer, and J. C. McMenamin, Phys. Rev. B **6**, 3056 (1972).

[46] L. Ley, R. A. Pollak, F. R. McFeely, S. P. Kowalczyk, and D. A. Shirley, Phys. Rev. B **9**, 600

(1974).

[47] C.-Y. Chang, H.-M. Huang, Y.-P. Lan, T.-C. Lu, L.-W. Tu, and W.-F. Hsieh, Crystal Growth & Design **13**, 3098 (2013), http://dx.doi.org/10.1021/cg400497r.