Supporting Information for:

A computational study of the properties of low- and high-index Pd, Cu and Zn surfaces

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I . Figures

Figure S1. The Pd (111) surface facet (FCC). Left: View of the surface *xy*-plane; Right: View perpendicular to the surface along the *yz*-plane. The 1x1x7 unit cell is shown with a dotted line, including 20 Å vacuum above and below the surface; to aid viewing of the surface, the model has been replicated twofold, giving a supercell view, in the directions of the surface plane.

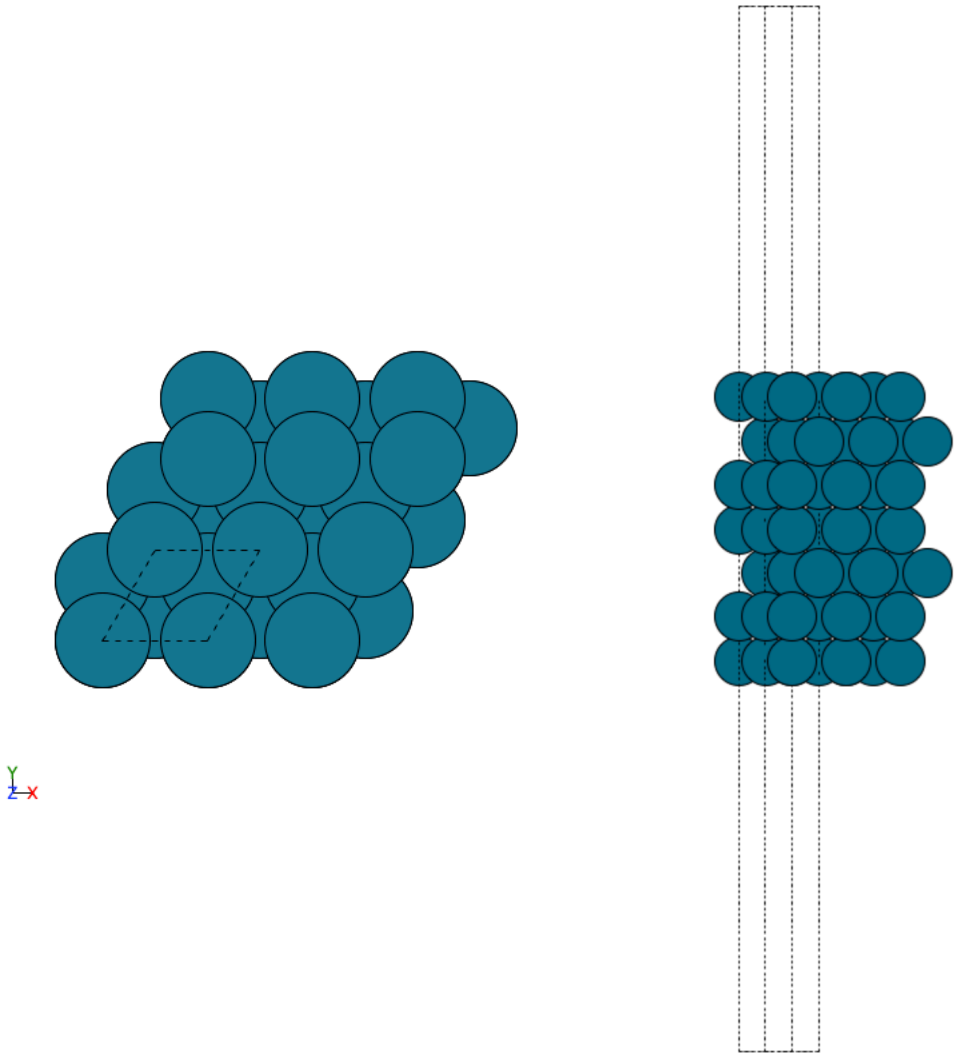


Figure S2. The Pd (100) surface facet (FCC). Left: View of the surface *xy*-plane; Right: View perpendicular to the surface along the *yz*-plane. The 1x1x7 unit cell is shown with a dotted line, including 20 Å vacuum above and below the surface; to aid viewing of the surface, the model has been replicated twofold, giving a supercell view, in the directions of the surface plane.

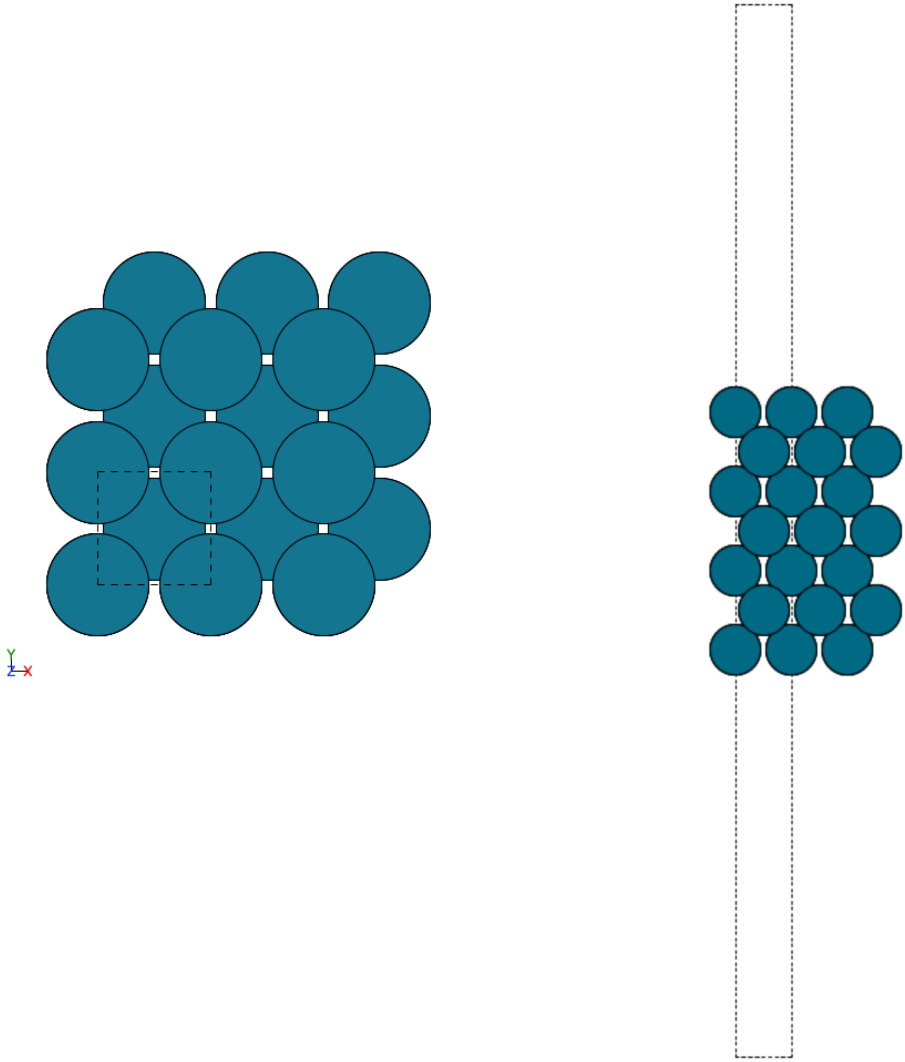


Figure S3. The Pd (110) surface facet (FCC). Left: View of the surface *xy*-plane; Right: View perpendicular to the surface along the *yz*-plane. The 1x1x7 unit cell is shown with a dotted line, including 20 Å vacuum above and below the surface; to aid viewing of the surface, the model has been replicated twofold, giving a supercell view, in the directions of the surface plane.

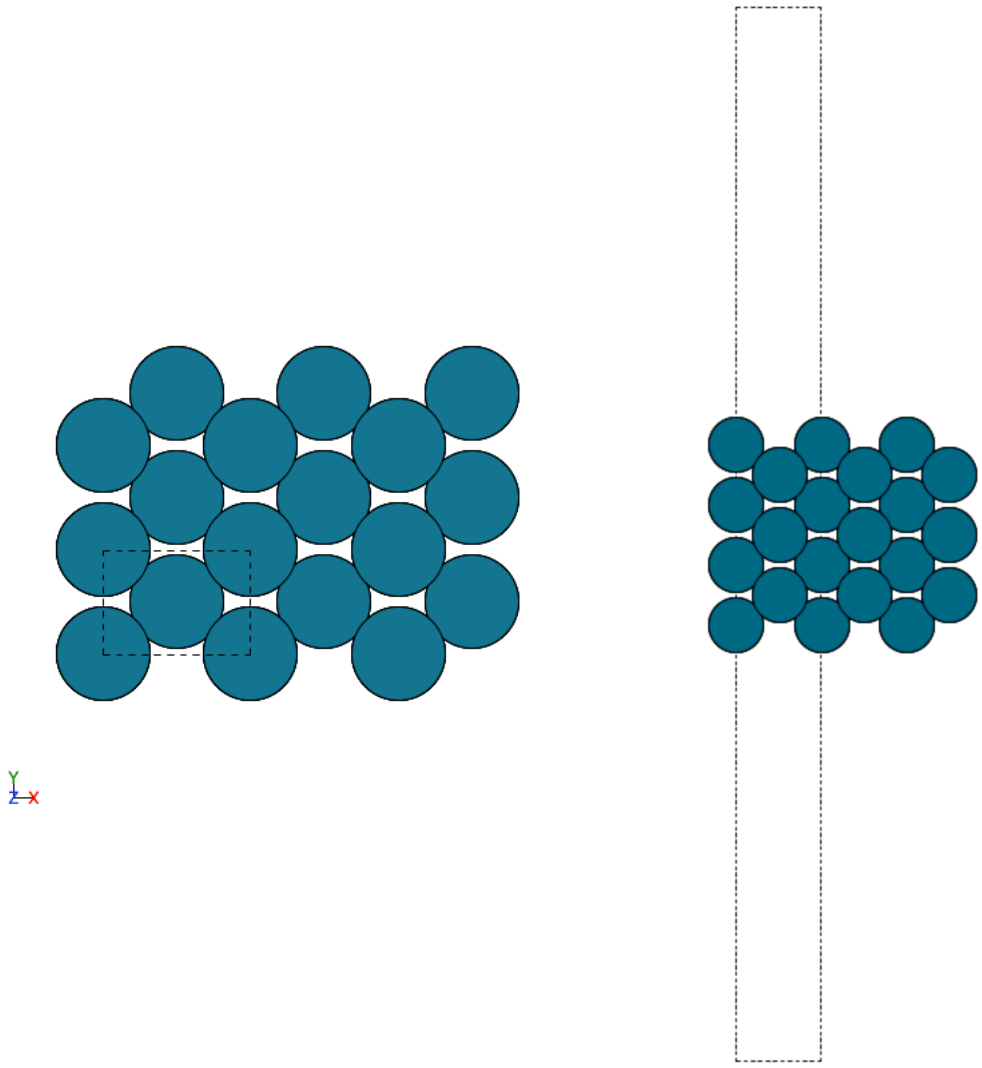


Figure S4. The Zn (0001) surface facet (HCP). Left: View of the surface *xy*-plane; Right: View perpendicular to the surface along the *yz*-plane. The 1x1x7 unit cell is shown with a dotted line, including 20 Å vacuum above and below the surface; to aid viewing of the surface, the model has been replicated twofold, giving a supercell view, in the directions of the surface plane.

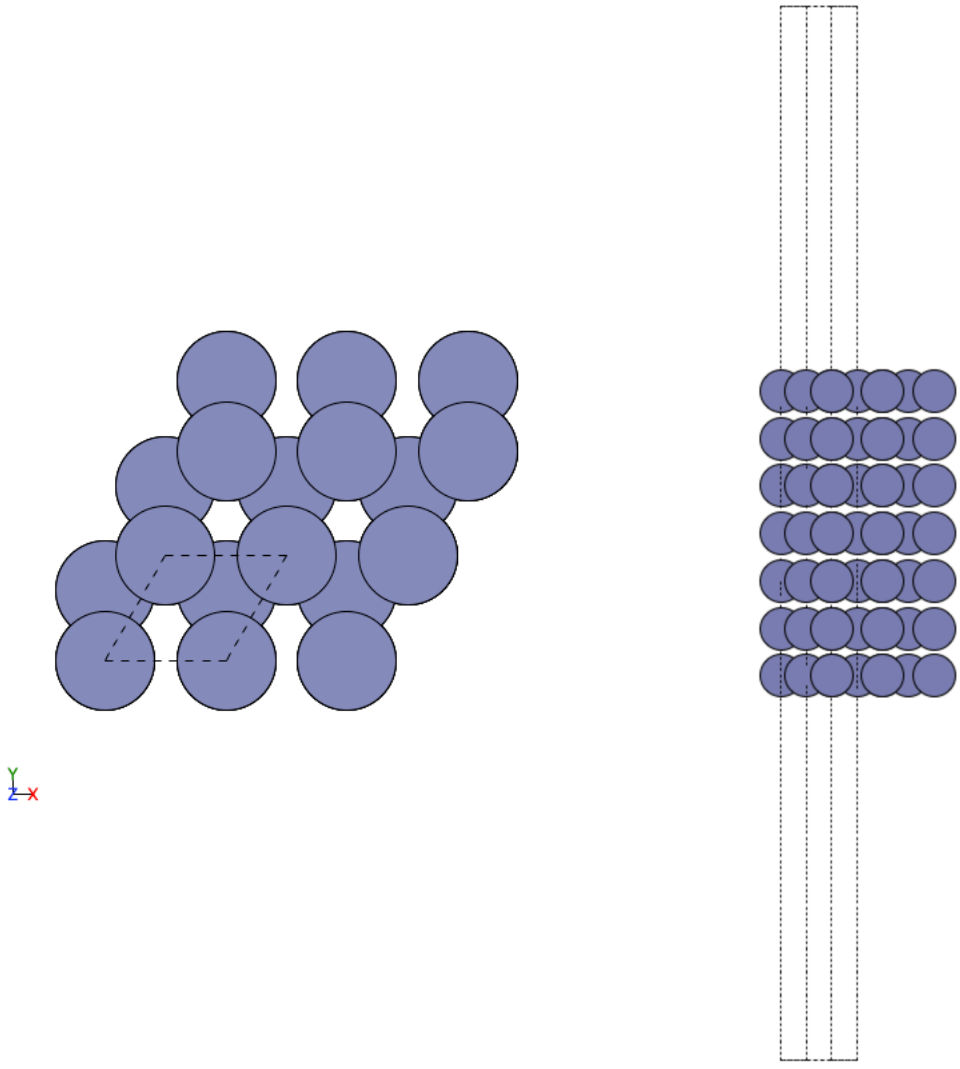


Figure S5. The Zn (10-10) surface facet (HCP). Left: View of the surface *xy*-plane; Right: View perpendicular to the surface along the *yz*-plane. The 1x2x7 unit cell is shown with a dotted line, including 20 Å vacuum above and below the surface; to aid viewing of the surface, the model has been replicated twofold, giving a supercell view, in the directions of the surface plane.

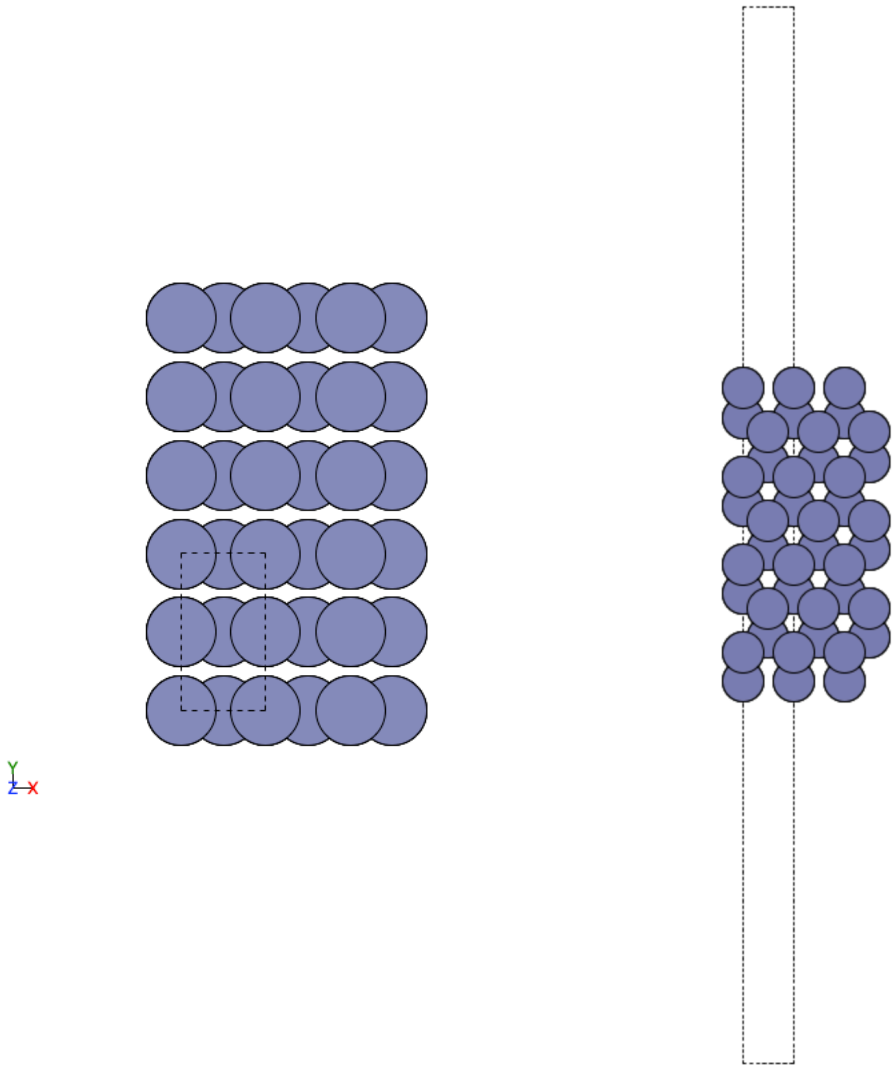


Figure S6. The surface energy (Jm^{-2}) for Cu (111), (100) and (110) facets, given in the top, middle and bottom graphs, respectively, as function of **k**-grid density ($Å^{-1}$) in the *x*- and *y*-directions. A key is given to show how many layers (5-10) were considered in the respective models.

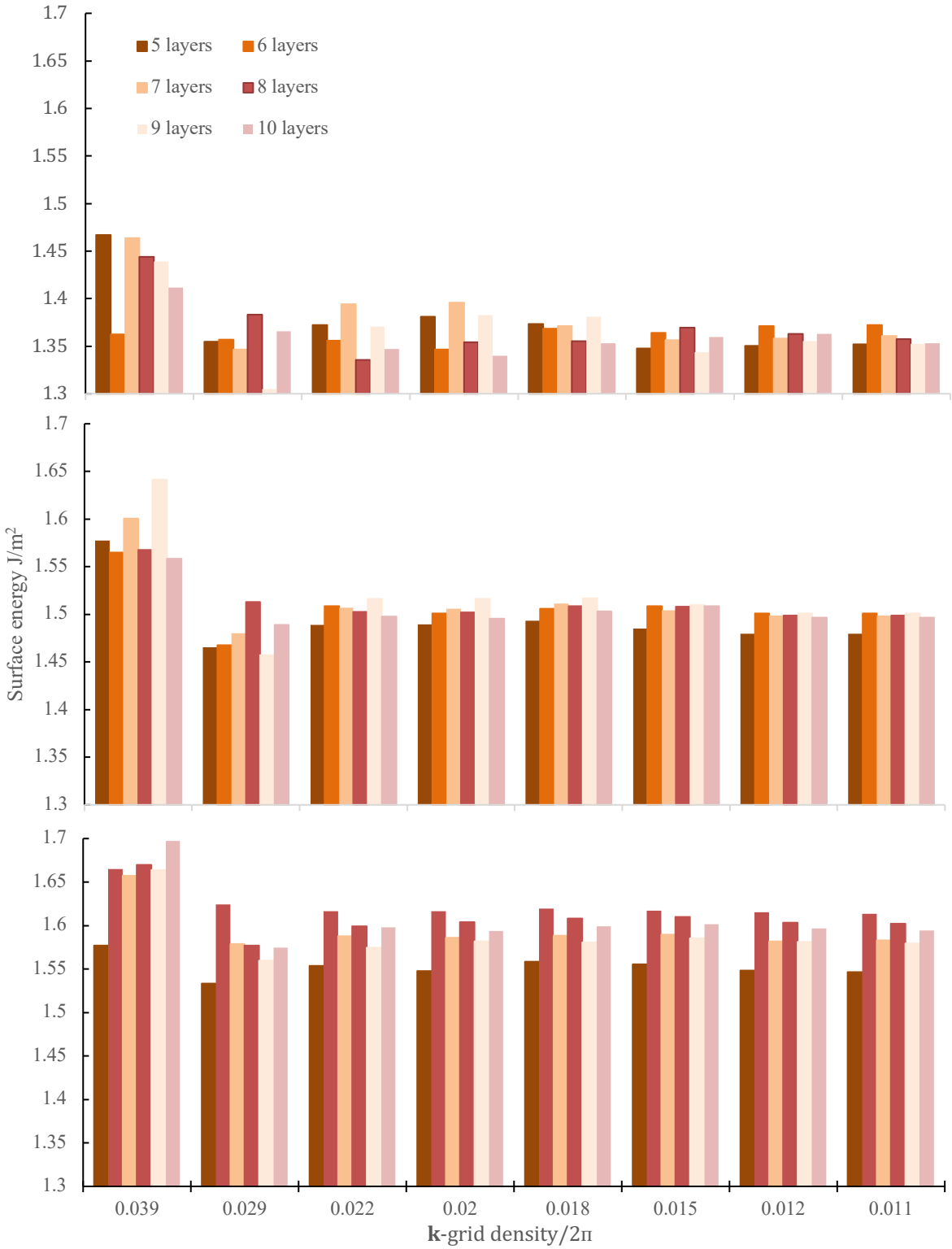
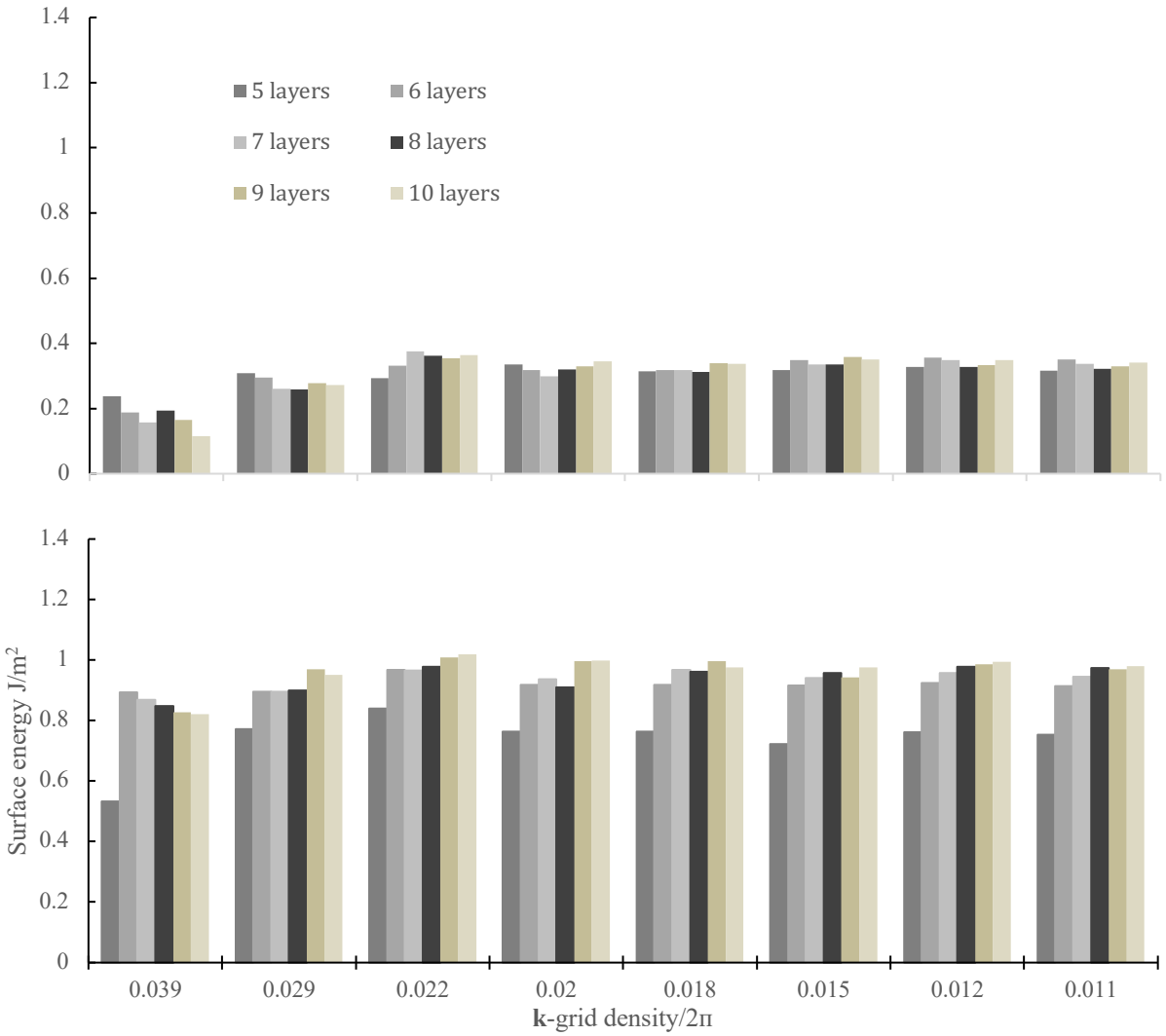


Figure S7. The surface energy (Jm^{-2}) for Zn (0001) and (10-10) facets, given in the top and bottom graphs, respectively, as function of **k**-grid density ($Å^{-1}$) in the x- and y-directions. A key is given to show how many layers (5-10) were considered in the respective models.



II. Tables

Table S1. Lattice parameters (a_0, given in Å) and cohesive energies (E_{coh}, given in eV/atom) calculated for Pd with different exchange functionals. Δa_0 and ΔE_{coh} are differences (%), and defined such that a negative sign indicates that the calculated value is lower than experiment. The MPAE is as defined in the main manuscript.

		a_0	Δa_0	E_{coh}	ΔE_{coh}	MPAE
GGA	PBE (+TS) (+MBD-NL)	3.93 (3.90) (3.92)	1.18 (0.41) (0.80)	3.73 (4.00) (3.98)	-4.11 (2.82) (2.54)	2.65 (1.61) (1.67)
	PBEsol	3.87	-0.53	4.48	15.29	7.91
	PBEint	3.97	2.106	3.31	-14.83	8.47
	revPBE	3.96	1.84	3.22	-17.22	9.53
	PW91	4.03	3.52	2.99	-23.11	13.31
	RPBE	3.98	2.132	3.10	-20.20	11.17
	R48PBE	4.05	4.13	2.66	-31.44	17.78
	B97-D	3.99	2.414	2.69	-30.74	16.58
	BLYP	4.02	3.40	3.01	-22.62	13.01
	HCTH-407	3.96	1.85	2.30	-40.87	21.36
Meta-GGA	mBEEF	3.88	-0.13	3.57	-8.15	4.14
	TPSS	3.89	0.12	4.35	11.90	6.01
	revSCAN	3.90	0.20	4.30	10.54	5.37
	SCAN	3.88	-0.12	4.35	11.90	6.01
	revTPSS	3.87	-0.53	4.39	12.98	6.76
	TPSSloc	3.82	-1.85	4.96	27.48	14.66
Hybrid	HSE06	3.92	0.87	3.05	-21.59	11.23
	PBE0 (+TS)	3.90 (3.87)	0.30 (-0.41)	2.94 (3.20)	-24.42 (-17.73)	12.36 (9.07)
	B3LYP	3.98	2.36	2.50	-35.73	19.04
Exp.	Gražulis *et al*. [1]	3.893	-	-	-	-
	Kittel [2]	-	-	3.89	-	-

Table S2. Lattice parameters (a_0, given in Å) and cohesive energies (E_{coh}, given in eV/atom) calculated for Cu with different exchange functionals. Δa_0 and ΔE_{coh} are differences (%), and defined such that a negative sign indicates that the calculated value is lower than experiment. The MPAE is as defined in the main manuscript.

		a_0	Δa_0	E_{coh}	ΔE_{coh}	MPAE
GGA	PBE (+TS) (+MBD-NL)	3.63 (3.54) (3.60)	0.47 (1.83) (-0.49)	3.53 (4.14) (3.86)	1.14 (18.62) (10.60)	0.80 (10.22) (5.54)
	PBEsol	3.56	-1.38	4.09	17.19	9.29
	PBEint	3.55	-1.77	4.19	20.03	10.90
	revPBE	3.66	1.32	3.13	-10.31	5.82
	PW91	3.60	-0.22	3.87	10.86	5.54
	RPBE	3.67	1.66	3.05	-12.60	7.14
	R48PBE	3.61	0.02	3.62	3.75	1.89
	B97-D	3.68	2.01	2.82	-19.23	10.62
	BLYP	3.70	2.51	2.51	-28.08	15.30
	HCTH-407	3.66	1.33	2.63	-24.64	12.98
Meta-GGA	mBEEF	3.57	-1.05	3.52	0.80	0.93
	TPSS	3.57	-1.05	3.83	9.77	5.41
	revSCAN	3.56	-1.35	3.91	12.09	6.72
	SCAN	3.55	-1.57	3.97	14.01	7.79
	revTPSS	3.55	-1.77	4.18	19.74	10.76
	TPSSloc	3.50	-2.99	4.58	31.23	17.11
Hybrid	HSE06	3.63	0.50	3.18	-8.88	4.69
	PBE0 (+TS)	3.63 (3.83)	0.39 (6.09)	3.14 (3.71)	-10.03 (6.30)	5.21 (6.19)
	B3LYP	3.61	-0.14	2.65	-24.07	12.10
Exp.	Gražulis *et al*. [1]	3.614	-	-	-	-
	Kittel [2]	-	-	3.49	-	-

Table S3. Lattice parameters (a_0 and c_0, given in Å) and cohesive energies (E_{coh}, given in eV/atom) calculated for Zn with different exchange functionals. Δa_0, Δc_0 and ΔE_{coh} are differences (%), and defined such that a negative sign indicates that the calculated value is lower than experiment. The MPAE is as defined in the main manuscript.

		a_0	Δa_0	c_0	Δc_0	E_{coh}	ΔE_{coh}	MPAE
GGA	PBE (+TS) (+MBD-NL)	2.78 (2.71) (2.75)	4.59 (1.93) (3.45)	5.17 (5.04) (5.12)	4.54 (1.88) (3.40)	1.09 (1.51) (1.36)	-19.26 (11.70) (0.74)	9.46 (5.17) (2.53)
	PBEsol	2.72	2.20	5.06	2.15	1.59	17.92	7.42
	PBEint	2.78	4.70	5.18	4.65	1.14	-15.41	8.25
	revPBE	2.81	5.79	5.23	5.73	0.78	-42.15	17.89
	PW91	2.84	6.91	5.29	6.85	0.87	-35.55	16.43
	RPBE	2.83	6.27	5.26	6.22	0.72	-46.44	19.46
	R48PBE	2.87	7.81	5.33	7.76	0.69	-48.59	21.38
	B97-D	2.88	8.42	5.36	8.37	0.33	-75.55	30.78
	BLYP	2.88	8.40	5.36	8.34	0.31	-76.96	31.23
	HCTH-407	2.82	6.08	5.25	6.03	0.38	-71.48	27.86
Meta-GGA	mBEEF	2.73	2.81	5.09	2.76	1.32	-2.22	2.59
	TPSS	2.73	2.70	5.08	2.66	1.35	0.00	1.78
	revSCAN	2.75	3.26	5.11	3.21	1.39	2.67	3.04
	SCAN	2.71	1.80	5.04	1.76	1.47	9.26	4.27
	revTPSS	2.70	1.72	5.03	1.67	1.62	20.00	7.79
	TPSSloc	2.67	0.29	4.96	0.24	2.03	50.59	17.03
Hybrid	HSE06	2.78	4.67	5.18	4.62	1.14	-15.40	8.23
	PBE0 (+TS)	2.78 (2.71)	4.54 (1.93)	5.17 (5.04)	4.49 (1.88)	1.14 (1.54)	-15.70 (13.70)	8.24 (5.84)
	B3LYP	2.85	7.14	5.30	7.09	0.44	-67.55	27.26
Exp.	Gražulis *et al*. [1]	2.665	-	4.947	-	-	-	-
	Kittel [2]	-	-	-	-	1.35	-	-

Table S4: MPAE for each density functional when applied to calculate equilibrium bulk structures, and associated cohesive energy, of Pd, Cu and Zn. The results are ordered descending, from best to worst, with respect to the MPAE within each DF approximation sub-category (GGA, meta-GGA, hybrid).

DF		**MPAE**			
		Pd	**Cu**	**Zn**	**Mean**
GGA	PBE (+TS) (+MBD-NL)	2.65 (1.61) (1.67)	0.80 (10.22) (5.54)	9.46 (5.17) (2.53)	4.30 (5.67) (3.25)
	PBEsol	7.91	9.29	7.42	8.21
	PBEint	8.47	10.90	8.25	9.21
	revPBE	9.53	5.82	17.89	11.08
	PW91	13.31	5.54	16.43	11.76
	RPBE	11.17	7.14	19.46	12.59
	R48PBE	17.78	1.89	21.38	13.68
	B97-D	16.58	10.62	30.78	19.33
	BLYP	13.01	15.30	31.23	19.85
	HCTH-407	21.36	12.98	27.86	20.73
Meta-GGA	mBEEF	4.14	0.93	2.60	2.56
	TPSS	6.01	5.41	1.78	4.40
	revSCAN	5.37	6.72	3.04	5.04
	SCAN	6.01	7.79	4.27	6.02
	revTPSS	6.76	10.76	7.79	8.44
	TPSSloc	14.66	17.11	17.03	16.27
Hybrid	HSE06	11.23	4.69	8.23	8.05
	PBE0 (+TS)	12.36 (9.07)	5.21 (6.19)	8.24 (5.84)	8.60 (7.03)
	B3LYP	19.04	12.10	27.26	19.47

Table S5. Surface energy (J/m^2) and work function (Φ, eV), calculated with the mBEEF density functional, for low- and high-index surfaces of Pd and Cu.

Facet	Surface energy	Φ	Surface energy	Φ
	Pd		Cu	
(111)	1.32	5.17	1.43	4.83
(100)	1.50	4.99	1.56	4.53
(110)	1.58	4.80	1.63	4.33
(332)	1.41	4.99	1.57	4.60
(322)	1.45	5.00	1.55	4.58
(221)	1.49	4.61	1.58	4.12
(210)	1.59	4.92	1.70	4.49

References

[1] Gražulis S, Chateigner D, Downs RT, Yokochi AF, Quirós M, Lutterotti L, Manakova E, Butkus J, Moeck P, Le Bail A. Crystallography Open Database - an open-access collection of crystal structures. *J Appl Crystallogr.* **42**, 726–729 (2009).

[2] Kittel C, Introduction to Solid State Physics, 8th Edition, John Wiley & Sons (2005)