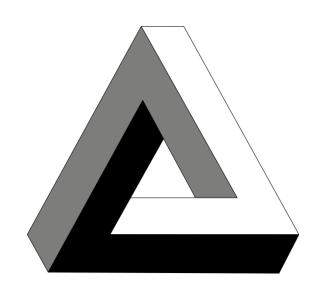
# THE DELTA PROJECT: CURRENT STATUS AND FUTURE DIRECTIONS



### Kurt Lejaeghere

Center for Molecular Modeling Ghent University, Belgium

together with
Stefaan Cottenier
the Delta collaboration







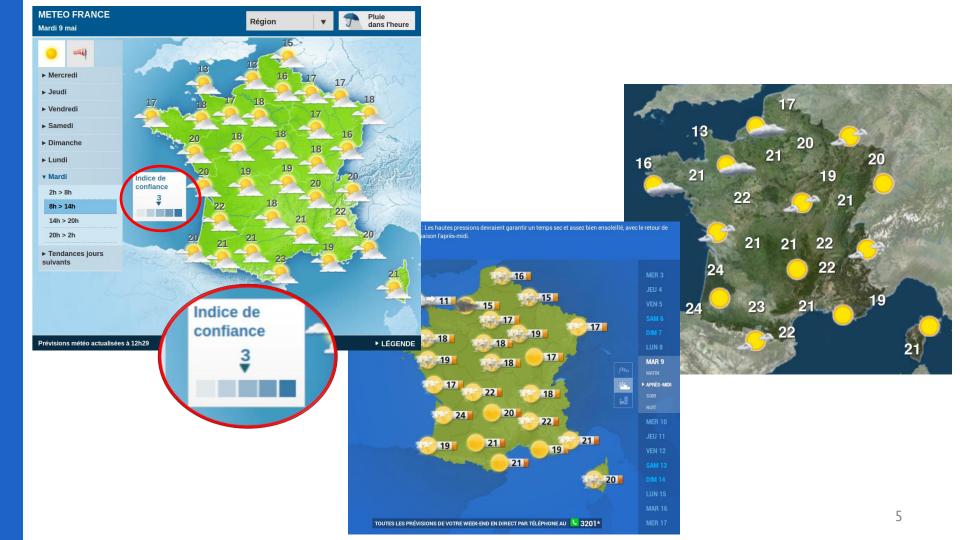


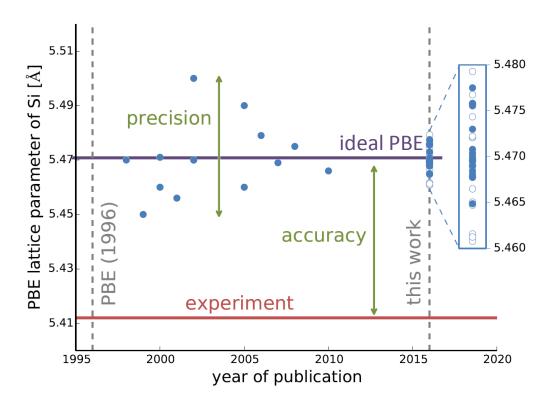






TOUTES LES PRÉVISIONS DE VOTRE WEEK-END EN DIRECT PAR TÉLÉPHONE AU 3201\*





KL and 68 others, Science 351 (6280), aad3000 (2016).

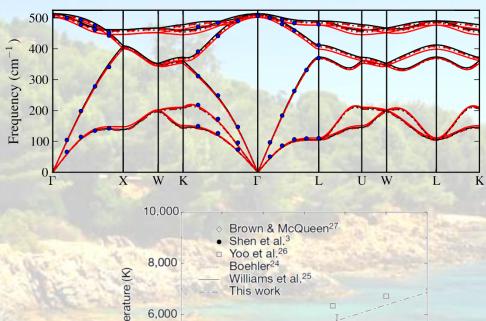


### **ACCURACY**





#### He et al., PRB 89, 064305 (2014)



# Shen et al.<sup>3</sup> Yoo et al.<sup>26</sup> Boehler<sup>24</sup> Williams et al.<sup>25</sup> This work

150

2,000 50

100

### Alfè et al., Nature 401, 462 (1999)

Pressure (GPa)

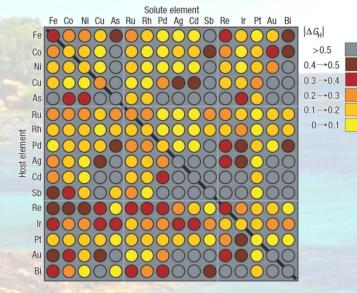
200

250

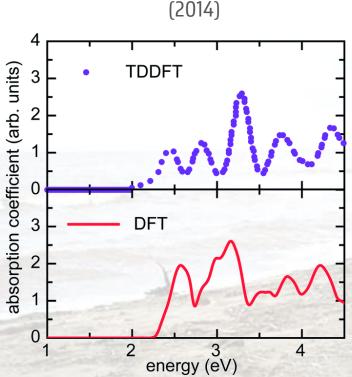
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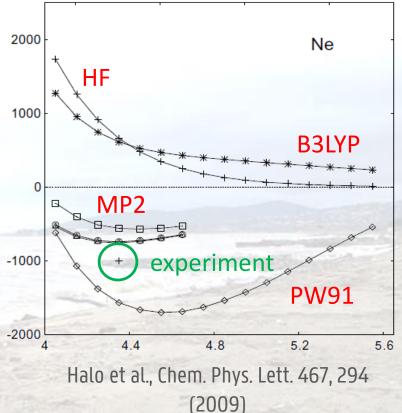
350

# Greeley et al., Nature Mater. 5, 909 (2006)



Hidalgo et al., Nanoscale 6, 3325 (2014)**TDDFT** 





(2009)



Tran, Stelzl, and Blaha

J. Chem. Phys. 144, 204120 (2016)

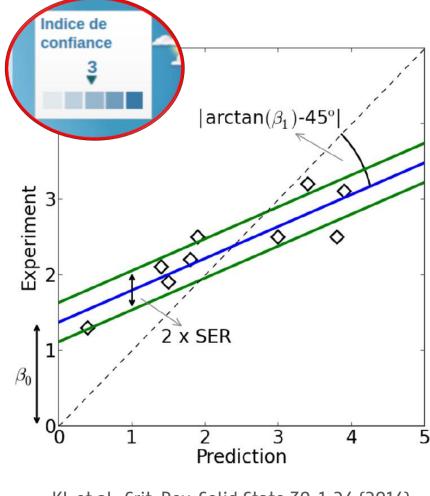
not reflect the trends for finite systems, as mentioned in Sec. III. Thus, in order to provide to the reader of the present work a more general view on the accuracy and applicability of the functionals, a very brief summary of some of the literature results for molecular systems is given below. To this end, we consider the atomization energy of strongly bound molecules and the interaction energy between weakly bound molecules, for which widely used standard testing sets exist.

#### A. Atomization energy of molecules

The atomization energy of molecules is one of the most

TABLE V. Results from the literature (reference in last column) for the MAE (in kcal/mol) on the S22 testing set.

Functional	MAE	Reference
LDA		
LDA <sup>80</sup>	2.3	48
GGA		
PBEsol <sup>83</sup>	1.8	48
PBE <sup>73</sup>	2.8	48
RPBE <sup>92</sup>	5.2	49
revPBE <sup>91</sup>	5.3	27
BLYP <sup>93,94</sup>	4.8, 8.8	106, 48



KL et al., Crit. Rev. Solid State 39, 1-24 (2014)

$$Y = \beta_0 + \beta_1 X \pm SER$$

$$\beta_0; \beta_1 - 1$$

$$systematic$$

$$deviation$$

$$SER$$

$$error bar$$

functional error



19

TOUTES LES PRÉVISIONS DE VOTRE WEEK-END EN DIRECT PAR TÉLÉPHONE AU 3201\*

### **PRECISION**



#### RESEARCH ARTICLE

#### **DFT METHODS**

# Reproducibility in density functional theory calculations of solids

Kurt Lejaeghere,1\* Gustav Bihlmayer,2 Torbjörn Björkman,3,4 Peter Blaha,5 Stefan Blügel, Volker Blum, Damien Caliste, 7,8 Ivano E. Castelli, Stewart J. Clark, 10 Andrea Dal Corso, 11 Stefano de Gironcoli, 11 Thierry Deutsch, 7,8 John Kay Dewhurst, 12 Igor Di Marco, 13 Claudia Draxl, 14,15 Marcin Dułak, 16 Olle Eriksson, 13 José A. Flores-Livas, 12 Kevin F. Garrity, 17 Luigi Genovese, 7,8 Paolo Giannozzi, 18 Matteo Giantomassi, 19 Stefan Goedecker, 20 Xavier Gonze, 19 Oscar Grånäs, 13,21 E. K. U. Gross, 12 Andris Gulans, 14,15 François Gygi, 22 D. R. Hamann, 23,24 Phil J. Hasnip,<sup>25</sup> N. A. W. Holzwarth,<sup>26</sup> Diana Iuşan,<sup>13</sup> Dominik B. Jochym,<sup>27</sup> François Jollet, 28 Daniel Jones, 29 Georg Kresse, 30 Klaus Koepernik, 31,32 Emine Kücükbenli, 9,11 Yaroslav O. Kvashnin, 13 Inka L. M. Locht, 13,33 Sven Lubeck, 14 Martijn Marsman, 30 Nicola Marzari, 9 Ulrike Nitzsche, 31 Lars Nordström, 13 Taisuke Ozaki, 34 Lorenzo Paulatto, 35 Chris J. Pickard, 36 Ward Poelmans, 1,37 Matt I. J. Probert, 25 Keith Refson, 38,39 Manuel Richter, 31,32 Gian-Marco Rignanese, 19 Santanu Saha, 20 Matthias Scheffler, 15,40 Martin Schlipf, 22 Karlheinz Schwarz, 5 Sangeeta Sharma, 12 Francesca Tavazza, 17 Patrik Thunström, 41 Alexandre Tkatchenko, 15,42 Marc Torrent, 28 David Vanderbilt, 23 Michiel J. van Setten, 19 Veronique Van Speybroeck, John M. Wills, 43 Jonathan R. Yates, 29 Guo-Xu Zhang,44 Stefaan Cottenier1,45\*

The widespread popularity of density functional theory has given rise to an extensive range

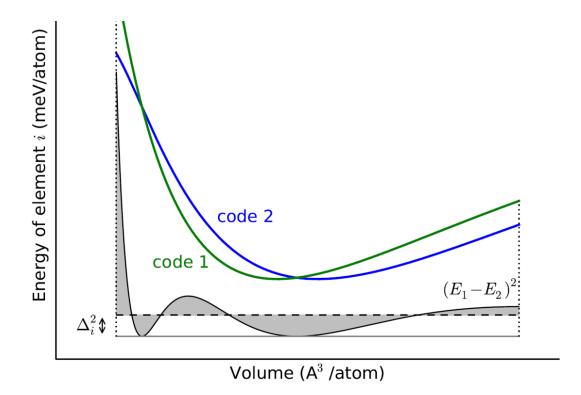
the fundamental variable. Although this reformulation is in principle exact, it is not fully known how the interaction between individual electrons should be transformed. As a result, the specific form of the unknown part of the interaction energy, the exchange-correlation functional, has been the focus of many investigations, leading to a plethora of available functionals in both solid-state physics (15–19) and quantum chemistry (15, 20–23).

Once a particular exchange-correlation function-

al has been chosen, the mathematical problem is completely specified as a set of Kohn-Sham equations, whose solution yields orbitals and energies from which the total electronic energy can be evaluated. A variety of such numerical solution schemes have been implemented in different computer codes. Comparisons of their performance are much less frequent or extensive than those of exchange-correlation functionals, however (21, 24-29). One might reasonably expect that because they solve the same equations, they all produce similar answers for a given crystal structure, but a glance at the literature shows that this assumption is by no means always true. Figure 1 demonstrates that even for a well-studied material such as silicon, deviations between predictions from different codes (the "precision") are of the same order of magnitude as the deviation from the 0 K experimental value (the "accuracy") (26, 30). Because all of the codes shown in Fig. 1 treat silicon at the same level of theory, using the same exchange-correlation functional, they rield the come common by definition Hermann

Н																	Не
194		1:														194	
hP4																	hP2
Li	Ве	B C N O F Ne															Ne
166	194	- large number of elements 166 194 205 12 15 2															225
hR9	hP2	hP26 hP4 cP8 m54 m58														cF4	
Na	Mg	- diverse set of crystal structures  Al Si P S CI A															Ar
166	194		225 227 64 70 64 22														
hR9	hP2							cF4	cF8	oS8	oF128	oS8	cF4				
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Со	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
229	225	194	194	229	229	217	229	194	225	225	194	64	227	166	152	64	225
cI2	cF4	hP2	hP2	cI2	cI2	c158	cI2	hP2	cF4	cF4	hP2	oS8	cF8	hR6	hP3	oS8	cF4
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Те	I	Xe
229	225	194	194	229	229	194	194	225	225	225	194	139	227	166	152	64	225
cI2	cF4	hP2	hP2	cI2	cI2	hP2	hP2	cF4	cF4	cF4	hP2	tI2	cF8	hR6	hP3	oS8	cF4
Cs	Ва	Lu	Hf	Та	W	Re	Os	lr	Pt	Au	Hg	TI	Pb	Bi	Ро	At	Rn
229	229	194	194	229	229	194	194	225	225	225	139	194	225	166	221		225
cI2	cI2	hP2	hP2	cI2	cI2	hP2	hP2	cF4	cF4	cF4	tI2	hP2	cF4	hR6	cP1		cF4





$$\Delta_i(1,2) = \sqrt{\frac{1}{\Delta V_i}} \int (E_{1,i}(V) - E_{2,i}(V))^2 dV$$
$$\Delta(1,2) = \langle \Delta_i(1,2) \rangle$$



			AE			PAW											USPP								NCPP dates														
		Elk	exciting	FHI-aims/tight	FHI-aims/really_t	FHI-aims/tier2	FLEUR	FPLO/default	FPLO/T+F	FPLO/T+F+s	RSPt	WIENZK/default	WIEN2k/enhanced	WIENZK/acc	GBRV12/ABINIT	GPAW06/GPAW	GPAW09/ABINIT	GPAW09/GPAW	JTH01/ABINIT	JTH02/ABINIT	PS1ib031/QE	PSlibl00/QE	VASP2007/VASP	VASP2012/VASP	VASPGWZ015/VASP	GBRV12/QE	GBRV14/CASTEP	GBRV14/QE	OTEG7/CASTEP	80/600	Vdb/CASTEP	Vdb2/DMCAPO	FHI98pp/ABINIT	HGH/ABINIT	HGH-NLCC/BigDFT	MBK2013/OpenMX	ONCVPSP(PD0.1)/ABINI	ONCVPSP(SG15)1/CASTE	ONCVPSP(SG15)1/QE
	Elk			0.6	0.6								18 0								1.6	0.9			_						.4 6.				1.1			1.5	1.4 1
	exciting	0.3	_	0.5									_	_					1.2	0.6	1.6	8.0			-	1.0					.3 6.		134		1.1	2.1	0.7	1.4	1.3 1
	FHI-aims/tight		0.5		0.0						0.7		1.8 0	- 1						0.7		1.0			_					7 0			136		1.2	2.0	0.8		1.4 1
	FHI-aims/really_tight		0.5			0.5	0.7						1.8 0	_		3.8					1.7				-		1.2			7 0			136		1.2	2.0	0.8	1.5	14 1
	FHI-aims/tier2		0.1										1.8	-				15	1.2	0.6		8.0			-	0.9					.3 6.		134		1.1	2.1	0.7	1.4	13 1
ш	FLEUR		0.5			0.5		_	0.8		_		1.5	-						0.6		8.0			_		1.0			7 0	_		132		1.0	19	0.6	1.3	13 1
₹	FPLO/default	39					3.6		31		-			_		31						3.9			_	4.0				1 3			-		3.6	3.2	37	4.1	41 4
_	FPLO/T+F FPLO/T+F+s		1.0				0.8	-	_	0.8						3.4			10	0.9		1.3					1.3			1 1					1.2	18	1.0		16 1
	RSPt		0.9				0.8		0.8	_				_		3.5			1.2		1.4	1.3	19 19		_	1.4	1.4		29 1						1.2		1.0	1.6	1.6 1
	WIENZk/default		1.7					-		-	1.3	-	1.3 0	-		3.4				0.7 1.5		1.1			-					0 0					1.1	1.8	0.8	1.5	15 1
	WIENZK/derault WIENZk/enhanced		1.7								13 (	_	0.9 1	- 1		3.2 2.6			1.3			1.8	1.7		- 1	1.9	2.0			.8 1					1.6	1.5	1.0	21	10 1
	WIEN2k/ennanced		0.2		_	_	0.4					1.7				3.8				_	1.6		_	0.7	-				_	_	_	4 6.2		2.0		2.0	27	1.9	1.9 1
	GBRV12/ABINIT		0.8			_	_	_		_	_		20 0	_	uo	_	_	1.6				1.1	23	_	_	0.9		_	28 1	_	.7 6.		15.1		1.5	24	11	18	1.7 1
	GPAW06/GPAW		3.8		3.8			3.1					26 3		41	-6.2				35		3.8			-	4.0				9 3			123		3.0	30	36	37	38 3
	GPAW09/ABINIT		1.3	13	1.3	1.3	1.3	4.1					21 1	_	15	36	-	0.6				1.5			_				25 1				136		1.7	23	12	1.7	17 1
	GPAW09/GPAW		1.5		1.6	1.5	1.5	4.1					2.2 1	-		35	_	0.0	16		2.1	1.6	2.5			1.7	1.7			5 1	.4 6.		136		1.8	2.3	1.5	1.8	18 1
_	JTH01/ABINIT		1.2			1.2	1.0	3.4					11 1				1.5	1.6				1.4					1.5		30 1				130		1.3	1.5	1.2	1.4	14 1
PAW	JTH02/ABINIT		0.6						0.9				15 0				1.4		09	u.s			19				1.2			7 0			134		1.2	-	0.7	14	14 1
χ	PSlib031/QE		1.6				1.5				_		16 1	-		30				14	2.0		15		-				31 1						1.6			21	22 2
_	PSlib100/QE	_	0.8				0.8		13				18 0	_			1.5			0.9	1.6		17		_		1.2			9 0		1 5.9			1.4	19	0.9	1.6	16 1
	VASP2007/VASP		2.1										14 2	-		28					1.5	1.7		18	_		2.2			1 1	-		124		2.2	17	1.9		24 2
	VASP2012/VASP	_	0.6				0.7						19 0	_			1.4				1.6		1.8		_		1.2			.8 0		5 6.3	134		1.2		0.9		1.5 1
	VASPGW2015/VASP		0.4				0.6		10		0.8		20 0	_			1.3			0.7		0.8	21	_	-					.5 0		6 6.2			1.1				14 1
	GBRV12/QE	1.1	1.0	1.1	1.1	0.9	1.0	4.0	13	1.4	1.2	1.9 :	20 0	19	0.7	4.0	1.6	1.7	15	1.2	2.0	1.1	21	11	1.1		0.4	11 :	26 1	.0 0	.8 6.	3 6.4	15.3	2.3	1.4	21	1.2	1.6	1.5 1
	GBRV14/CASTEP	1.1	1.1	1.2	1.2	1.0	1.0	4.0	13	1.4	13	1.9 2	20 1	LO	0.8	3.8	1.6	1.7	15	12	1.9	1.2	22	1.2	1.1	0.4		13 :	26 0	9 0	.9 6.	2 6.3	15.0	2.4	1.6	21	1.1	1.5	1.5 1
0	GBRV14/QE	1.0	1.0	1.1	1.1	0.9	1.0	4.1	1.3	1.4	13	1.9	20 1	LO	0.7	4.0	1.6	1.7	15	1.2	2.0	1.2	21	11	1.1	0.1	0.3		26 1	.0 0	.8 6.	3 6.3	15.2	2.3	1.4	2.1	1.2	1.6	1.5 1
JSPP	OTFG7/CASTEP	25	2.5	2.6	26	25	2.6	5.8	31	29	30	8.8	3.8 2	25	28	5.6	2.5	2.7	30	26	3.1	2.2	3.5	25	26	2.6	2.6	26	2	2 2	4 4	8 5.7	14.5	2.7	2.9	3.4	2.4	2.6	2.6 2
S	OTFG9/CASTEP	0.4	0.5	0.7	0.7	0.5	0.7	4.1	11	1.0	1.0	1.8	2.0	15	1.0	39	1.4	1.5	14	0.7	1.6	0.9	21	0.8	2.5	1.0	0.9	LO :	2.2	0	.6 6.	3 6.2	13.6	2.2	1.1	2.1	0.8	1.5	14 1
_	SSSP/QE	0.4	0.3	0.6	0.6	0.3	0.5	3.9	10	0.9	0.8	1.6	17 0	13	0.7	36	1.3	1.4	11	0.6	1.5	0.7	19	0.6	0.4	8.0	0.9	18	24 0	.6	6.	4 6.2	13.6	2.1	1.0	2.0	0.7	1.4	1.2 1
	Vdb/CASTEP	6.4	6.4	6.4	6.5	64	6.5	7.9	6.6	6.4	6.7	7.1	69 6	i4	64	7.4	6.5	6.5	6.5	6.3	6.1	6.1	6.5	6.5	56	6.3	6.2	i3 4	4.8 6	.3 6	.4	9.6	16.3	6.6	6.1	6.6	6.4	5.7	5.8 5
	Vdb2/DACAPO	6.3	6.3	6.3	6.3	6.3	6.3	7.2	6.4	6.4	6.5	7.0	6.9 6	i2	6.3	7.6	6.1	6.1	6.5	6.2	5.8	5.9	61	6.3	52	6.4	6.3	i3 !	5.7 6	.2 6	.2 9.	6	17.9	6.2	5.9	6.4	61	6.5	6.5 6
	FHI98pp/ABINIT	135	134	13.6	13.6	134	132	13.0	13.7 1	3.0	32 1	3.0 1	2.3 1	34	15.1	12.3	13.6	13.6	13.0	13.4	12.8	13.5	12.4	134 1	37	15.3	5.0 1	5.2 1	45 13	.6 1	3.6 16.	3 17.9		14.3	8.5	130	133	13.3	13.6 13
	HGH/ABINIT	2.2	2.2	2.2	2.2	22	2.0	4.9	24	2.3	2.2	2.8	2.8 2	2.1	25	4.5	2.3	2.5	2.2	2.2	2.4	2.1	3.0	2.2	2.2	2.3	2.4	2.3	27 2	2 2	.1 6.	6 6.2	14.3		0.9	26	2.0	2.0	2.0 2
٥	HGH-NLCC/BigDFT	1.1	1.1	1.2	1.2	1.1	1.0	3.6	1.2	1.2	11	1.7	1.6 1	LO	15	30	1.7	1.8	13	1.2	1.6	1.4	2.2	1.2	1.1	1.4	1.6	L4 :	29 1	1 1	.0 6.	1 5.9	85	0.9		1.8	1.1	1.5	1.4 1
NCPP	MBK2013/OpenMX	21	2.1	2.0	2.0	21	1.9	3.2	18	1.8	18		15 2	_	24	3.0	2.3	2.3	15	19	1.7	1.9			_	2.1			3.4 2	1 2	.0 6.	6 6.4	13.0	2.6	1.8		2.0	2.2	2.2 2
ĭ	ONCVPSP(PD0.1)/ABINIT		0.7								0.8		17 0	_		36				0.7		0.9	19		_		1.1		24 0		.7 6.		13.3		1.1			1.3	14 1
_	ONCVPSP(SG15)1/CASTEP		1.4			1.4	1.3	4.1					19 1	-		3.7				14	2.1	1.6	25		_		1.5		26 1	.5 1	.4 5.				1.5	2.2	1.3		0.3 0
	ONCVPSP(SG15)1/QE		1.3		1.4								19 1	-	17				14	14	2.2	1.6			1.4		1.5		26 1						1.4	2.2		0.3	0
	ONCVPSP(SG15)2/CASTEP	1.4	1.4	1.5	1.5	14	1.3	4.1	16	1.6	15	2.1	19 1	L4	18	3.7	1.7	1.8	14	14	2.1	1.6	25	16	1.5	1.6	1.5	L5 2	26 1	5 1	.3 5.	7 6.5	13.4	2.0	1.4	2.2	1.3	0.1	0.3



molmod.ugent.be/deltacodesdft

	version			meV/atom
VASP	5.2.12	plane waves	PAW 2015 GW-ready (5.4)	0.3 meV/atom
FHI-aims	081213	tier2 numerical orbitals	all-electron (relativistic zora scalar 1e-12)	0.3 meV/atom
Quantum ESPRESSO	5.1	plane waves	SSSP Accuracy (mixed NC/US/PAW potential library)	0.3 meV/atom
Elk	3.1.5	APW+lo	all-electron	0.3 meV/atom
ABINIT	7.8.2	plane waves	PAW JTH v1.0	0.4 meV/atom
FLEUR	0.26	LAPW (+lo)	all-electron	0.4 meV/atom
Quantum ESPRESSO	5.1	plane waves	SSSP Efficiency (mixed NC/US/PAW potential library)	0.4 meV/atom
CASTEP	9.0	plane waves	OTFG CASTEP 9.0	0.5 meV/atom
ABINIT	7.7.3	plane waves	PAW JTH v0.2	0.5 meV/atom
FHI-aims	081213	tight numerical orbitals	all-electron (relativistic atomic_zora scalar)	0.5 meV/atom
VASP	5.2.12	plane waves	PAW 2012	0.6 meV/atom
ABINIT	7.11.8	plane waves	pseudo_dojo_ONCVPSP 0.1	0.6

### what's next?

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[extendable]

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### test systems

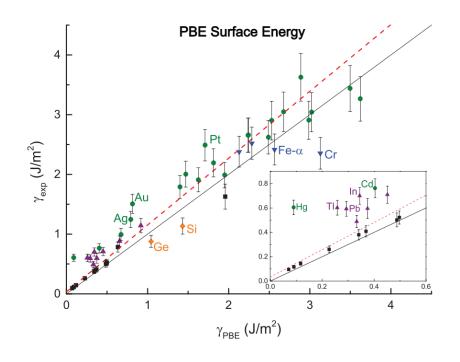
- structural diversity
   all elemental crystals with fcc, bcc, sc and diamond structure (12/8/6/4 neighbours)
- chemical diversity all oxides  $X_2O_1$ ,  $X_2O_3$ ,  $X_2O_5$  and  $XO_3$  (oxidation numbers +1 to +6)
- bond diversity (validation?)
  for each element an experimentally known binary metal, ionic and covalent compound

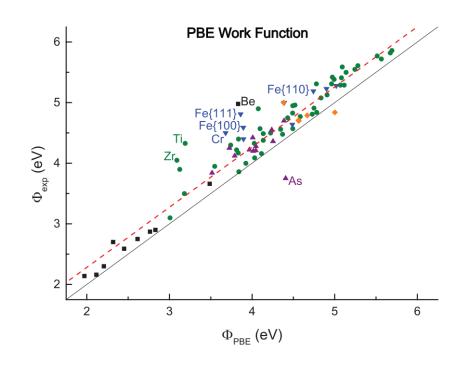
## properties

- formation energies
- band structures
- forces
- phonon frequencies
- magnetization

- ...

	β <sub>0</sub>	1 – β <sub>1</sub>	SER
Φ [eV]	0.30	1%	1.29
$\gamma$ [J/m <sup>2</sup> ]	0.03	-12 %	2.34





De Waele et al., PRB 94, 235418 (2016)

# methodologies

- hybrid-functional DFT
- meta-GGA DFT
- GW
- BSE
- RPA

- ...



#### RESEARCH ARTICLE SUMMARY

**DFT METHODS** 

### acknowledgments

# Reproducibility in density functional theory calculations of solids

Kurt Lejaeghere,\* Gustav Bihlmayer, Torbjörn Björkman, Peter Blaha, Stefan Blügel, Volker Blum, Damien Caliste, Ivano E. Castelli, Stewart J. Clark, Andrea Dal Corso, Stefano de Gironcoli, Thierry Deutsch, John Kay Dewhurst, Igor Di Marco, Claudia Draxl, Marcin Dułak, Olle Eriksson, José A. Flores-Livas, Kevin F. Garrity, Luigi Genovese, Paolo Giannozzi, Matteo Giantomassi, Stefan Goedecker, Xavier Gonze, Oscar Grånäs,





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