Supporting Information for: All-electron Gaussian-based G_0W_0 for Valence and Core Excitation Energies of Periodic Systems

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We present band gaps computed by G_0W_0@PBE using **k**-meshes ranging from $3 \times 3 \times 3$ to 6×6×6 without any finite size corrections, which were used for performing finite size extrapolations in Table 3. For wZnO and wAlN, we used $3 \times 3 \times 2$, $4 \times 4 \times 3$ and $6 \times 6 \times 4$ **k**-meshes. cc-pVTZ/cc-pVTZ-RI basis sets were used for all solids, except for Ne and Ar where we used the aug-cc-pVTZ/aug-cc-pVTZ-RI basis. The most diffuse *p* function of Mg in MgO was removed to avoid linear dependencies.

Table S1: Band gaps of semiconductors and rare gas solids from G_0W_0@PBE at various k-meshes without finite size corrections. cc-pVTZ basis is used unless specified. The experimental lattice constants used in this study are also listed. All band gap values are in eV.

System	$3 \times 3 \times 3$	$4 \times 4 \times 4$	$5 \times 5 \times 5$	$6 \times 6 \times 6$	Lattice Constant (Å)
Si	0.90	0.95	0.98	0.99	$a = 5.430$[1]
C	4.90	5.02	5.15	5.21	$a = 3.567$[1]
SiC	1.82	2.01	2.07	2.13	$a = 4.350$[1]
BN	5.34	5.63	5.75	5.88	$a = 3.615$[1]
BP	1.76	1.90	1.92	1.96	$a = 4.538$[2]
AlP	2.03	2.16	2.18	2.22	$a = 5.451$[1]
GaN	2.83	2.91	2.95	2.98	$a = 4.520$[1]
GaP	1.96	2.10	2.11	2.15	$a = 5.448$[2]
MgO	6.56	6.77	6.91	7.00	$a = 4.213$[1]
ZnO	2.61	2.70	2.73	2.75	$a = 4.580$[1]
ZnS	3.38	3.44	3.48	3.51	$a = 5.420$[1]
Ne	16.92	17.69	18.16	18.46	$a = 4.430$[1]
Ar	11.36	11.82	12.11	12.30	$a = 5.260$[1]
System	$3 \times 3 \times 2$	$4 \times 4 \times 3$	$6 \times 6 \times 4$		Lattice Constant (Å)
wAlN	5.25	5.44	5.56		$a = 3.110, c = 4.980, z_1 = 0, z_2 = 0.38213$[3]
wZnO	2.80	2.89	2.94		$a = 3.250, c = 5.207, z_1 = 0, z_2 = 0.3825$[4]

In Table S2, we present a comparison on LDA and PBE band gaps of zinc blende ZnO using different Gaussian basis sets at $4 \times 4 \times 4$ **k**-mesh. The tested basis sets are cc-pVTZ, cc-pVQZ (quadruple-zeta basis), cc-pVTZ-PP (pseudopotential for Zn), cc-pwCVTZ (extra core basis for Zn and O), aug-cc-pVTZ (augmented/diffuse basis for O), and GTH-TZV2P-MOLOPT (pseudopotential for Zn and O). We find that the four all-electron Gaussian basis sets predict almost the same PBE (0.93 eV) and LDA (0.64 eV) band gaps. Only the two basis sets with pseudopotentials give

smaller PBE and LDA band gaps.

Table S2: LDA and PBE band gaps of zinc blende ZnO in different Gaussian basis sets at $4 \times 4 \times 4$ k-mesh. All values are in eV.

Basis	PBE	LDA
cc-pVTZ	0.93	0.64
cc-pVQZ	0.93	0.63
cc-pwCVTZ	0.94	0.64
aug-cc-pVTZ (aug for O only)	0.92	0.62
cc-pVTZ-pp	0.78	0.45
GTH-TZV2P-MOLOPT	0.65	0.35

References

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