Supplemental Material
A Simple Generalized Gradient Approximation for the Non-interacting Kinetic Energy Density Functional

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I. LITHIUM PSEUDO-ATOM

As noted in the main paper, the Li pseudo-atom has only a single orbital, thus must have vanishing t_θ, v_θ, and T_θ. This is essentially impossible behavior to reproduce exactly with a GGA KEDF, so it is of interest to see what a particular approximate KEDF does. The crucial test is scf, not post-scf. For perspective, in addition to the BLPS, we considered another local pseudo-potental (LPP), called "mod1", in its LDA version[1]. All the calculations were done with a locally modified version of the APE code[2].

Fig. 1 summarizes the results. First, one sees that the mod1 LPP is quite similar to the LPP obtained as the s-channel of the Hamann non-local pseudo-potential[3] (NLPP) while the BLPS[4,5] is quite different. However, the Hamann s-channel and BLPS pseudo-densities have a rough qualitative similarity. The mod1 pseudo-density is distinct. All, however, have zero slope at the origin, an important distinction from a cusped density in the context of a GGA KEDF. Second, one also sees that the Hamann calibration of the LKT a parameter is successful in the sense that the scf BLPS LKT and KS pseudo-densities are reasonably close. The LKT-KS comparison is not as good in the mod1 case, a sign of possibly limited transferability. Third, for both BLPS and mod1 LPPs, LKT delivers a non-vanishing v_θ^{LKT} over a substantial range of r and even, in the BLPS case, has $v_\theta^{LKT} < 0$ around $r = 2$ bohr.

Contributions to the LKT non-interacting kinetic energy are listed in Table I for BLPS and mod1. It can be seen that the Pauli energy is non-negligible (around 35%) relative to the VW energy. This non-zero result is certainly a limitation of the GGA KEDF form. However, the total non-interacting kinetic energies from LKT are close to the KS reference values. As must be true for an N-representable approximate KEDF, the exact KS kinetic energy is a lower bound to the LKT value in both cases.

TABLE I. Computed values (Hartree atomic units) of Pauli energy T_θ^{LKT}, von Weizsäcker energy T_W^{LKT}, and total non-interacting kinetic energy from LKT, T_s^{LKT}, together with the reference KS kinetic energy for both BLPS and mod1 LPPs.

	T_θ^{LKT}	T_W^{LKT}	T_s^{LKT}	T_s^{KS}
BLPS	0.018873	0.055018	0.073891	0.069254
mod1	0.025726	0.067051	0.092777	0.091551

II. COMPUTATIONAL TECHNICAL DETAILS - PERIODIC SYSTEMS

Conventional KS calculations were done with ABINIT vers. 8.4.3[6]. The OF-DFT calculations used PROFESS[7] and/or PROFESS@QUANTUM-ESPRESSO[8]. The KS-DFT calculations used a Fermi-Dirac smearing parameter of 0.01 eV for metals. A $20 \times 20 \times 20$ Monkhorst-Pack[9] k-point grid was used for all systems. The energy cutoff was 1600 eV in both KS and OF-DFT calculations. We used the "`dime=two`" option in PROFESS. This option forces both the real-space and Fourier transform integration grid size cardinalities in each crystalline direction to be powers of two (which is optimal in parallel calculations).

For LKT, $a = 1.3$ was used throughout. For the WGC KEDF, we used default parameter values $\alpha = (5 + \sqrt{5})/6$ and $\beta = (5 - \sqrt{5})/6$, as well as γ, 2.7 and 4.2 for metals and semiconductors, respectively, as recommended in Ref. [10]. For the HC KEDF, we used averaged parameters $\lambda = 0.01177$ and $\beta = 0.7143$.

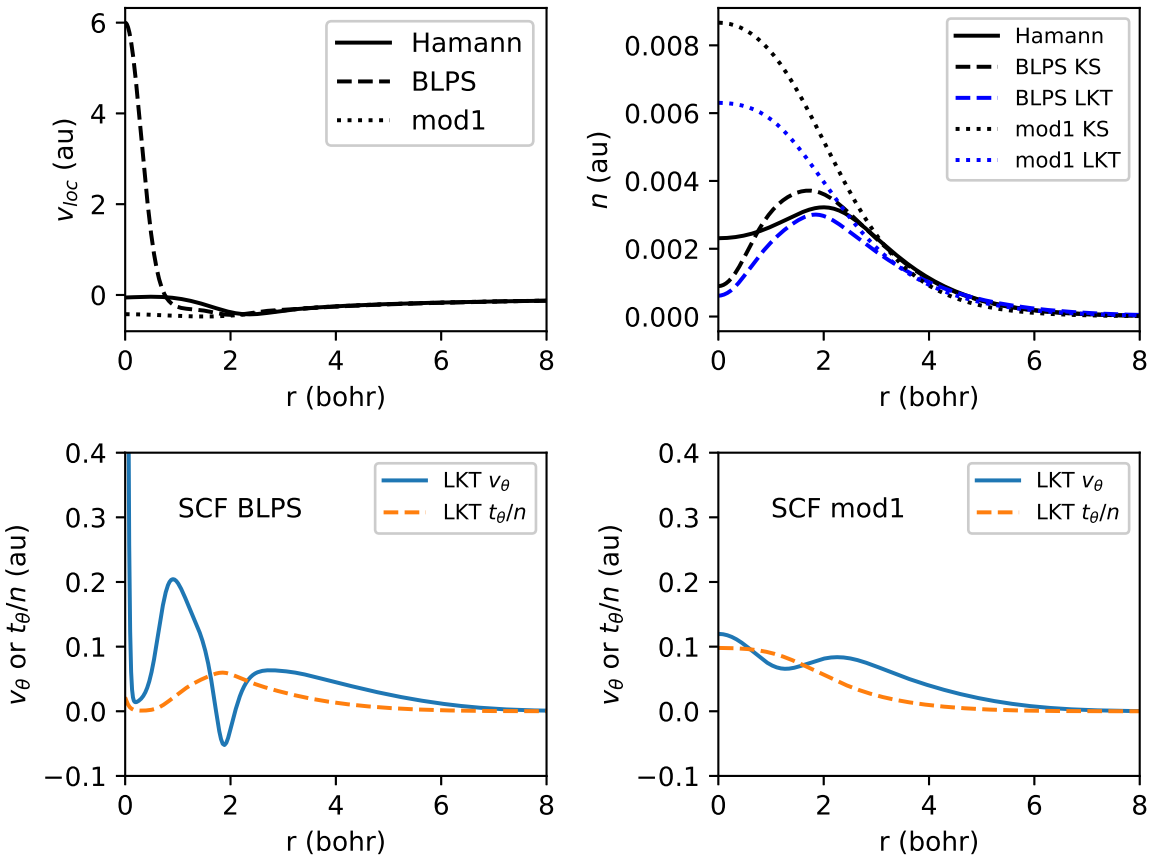

FIG. 1. Top left: BLPS and mod1 LPPs compared with Hamann NLPP s-channel potential; Top right: Li atom KS and LKT pseudo-densities for BLPS and mod1 LPPs along with the Hamann NLPP pseudo-density. Bottom panels: Li pseudo-atom LKT v_θ (solid, blue) and t_θ/n (dashed, orange) for BLPS (left) and mod1 (right) LPPs.

III. DETAILED TABULATION OF RESULTS ON SIMPLE SOLIDS

Detailed results from the validation calculations reported in the main paper are tabulated here. We used the BLPS of LDA type for both KS and OF calculations, including elements Li, Mg, Al, Ga, In, P, As and Sb. Table II gives the results for the metals Li, Mg, and Al for the four KEDFs compared to the KS reference values of V_0, E_0, and B_0. The structures include simple cubic (sc), body-centered cubic (bcc), face-centered cubic (fcc), and hexagonal close packed (hcp). Table III provides the corresponding results for the III-V semiconductors of zinc-blende structure we studied.

With the same data, Fig. 2 shows the differences of V_0, E_0 (either per atom or per cell), and B_0 between orbital-free calculations and conventional KS reference calculations for various KEDFs. The quantities shown are defined as

$$\Delta Q = Q_{OF} - Q_{KS}, \tag{1}$$

where Q is V_0, E_0, or B_0. The corresponding display for semiconductors is Fig. 3.

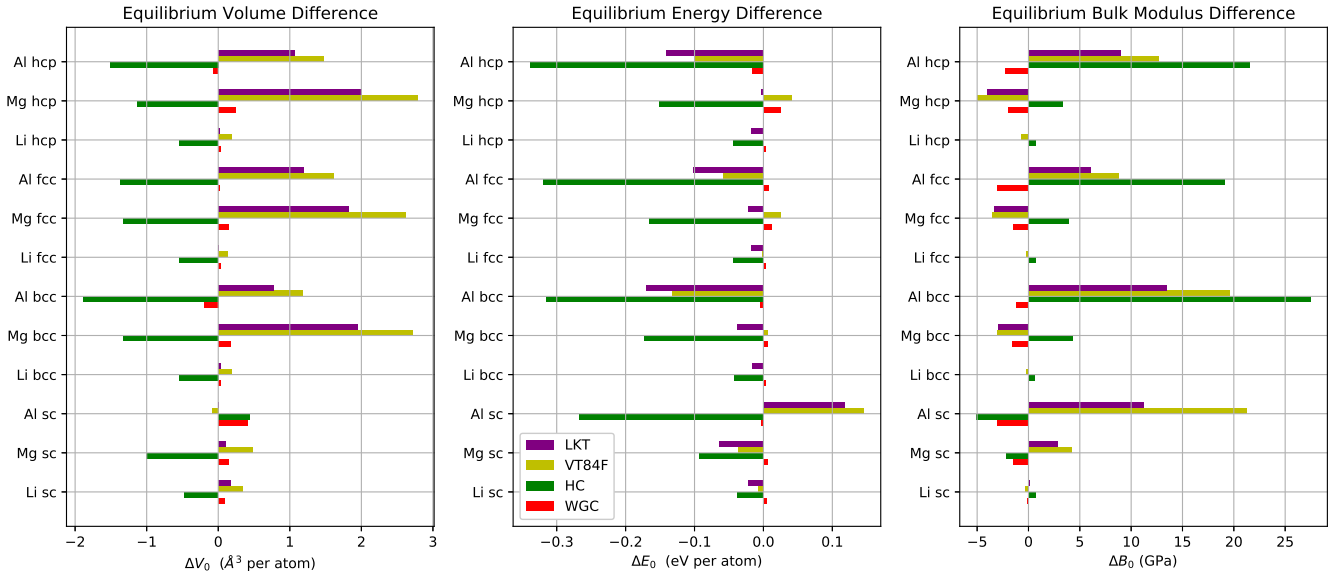

FIG. 2. Differences of equilibrium volume ΔV_0, energy ΔE_0, and bulk modulus ΔB_0 between orbital-free calculations with WGC (red), HC (green), VT84F (yellow), LKT (purple), and reference conventional KS calculations for metals. See definitions in Eq. (1).

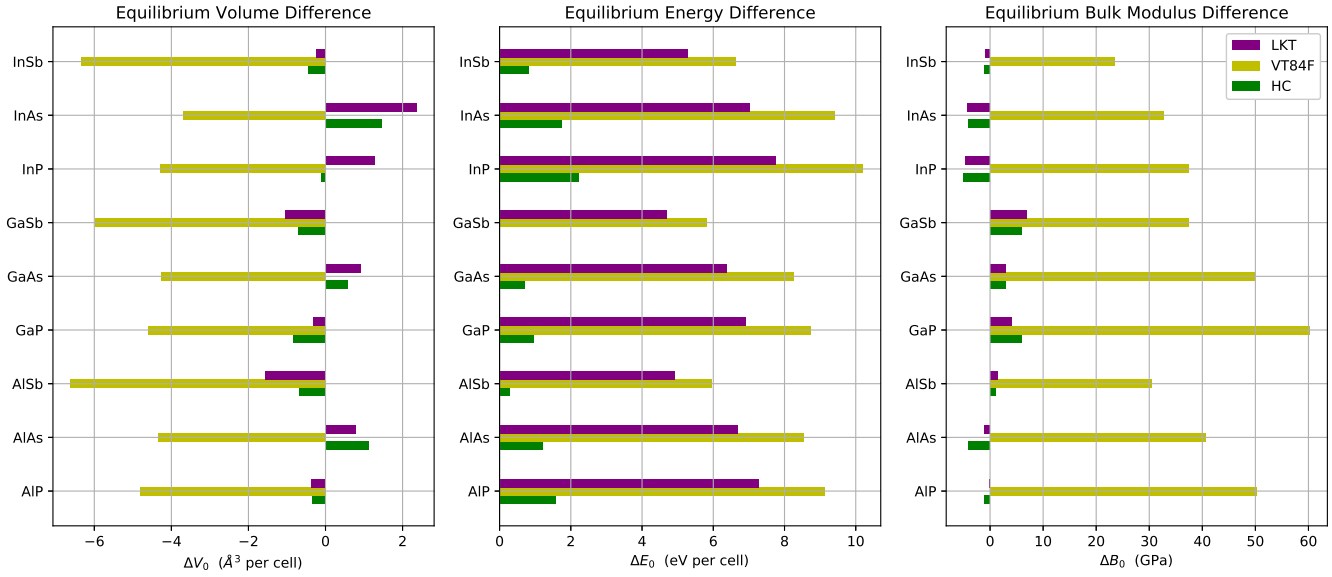

FIG. 3. Differences of equilibrium volume ΔV_0, energy ΔE_0 and bulk modulus ΔB_0 between orbital-free calculations with HC (green), VT84F (yellow), LKT (purple), and reference conventional KS calculations for semiconductors. Same quantities as in Fig. 2.

TABLE II. Equilibrium volume V_0 (Å^3/atom), energy E_0 (eV/atom), and bulk moduli B_0 (GPa) of simple elemental metals in sc, bcc, fcc, and hcp structures (see text for notation). For the energy, the differences with respect to the ordinarily lowest energy phases are shown. Those are bcc, hcp, and fcc respectively for Li, Mg, and Al.

	KEDF	Li				Mg				Al			
		sc	bcc	fcc	hcp	sc	bcc	fcc	hcp	sc	bcc	fcc	hcp
	KS	19.436	18.768	18.690	18.706	24.899	21.376	21.337	21.149	18.813	16.085	15.608	15.733
	WGC	19.529	18.809	18.727	18.743	25.052	21.551	21.481	21.398	19.226	15.887	15.632	15.666
V_0	HC	18.963	18.220	18.152	18.167	23.890	20.048	20.005	20.016	19.255	14.198	14.241	14.219
	VT84F	19.775	18.951	18.819	18.897	25.377	24.090	23.961	23.932	18.724	17.259	17.228	17.208
	LKT	19.605	18.805	18.700	18.726	25.006	23.320	23.167	23.160	18.825	16.860	16.803	16.803
	KS	0.139	-7.598	-0.001	-0.001	0.370	0.041	0.019	-24.672	0.363	0.091	-57.948	0.039
	WGC	0.141	-7.595	-0.001	-0.001	0.351	0.023	0.006	-24.647	0.352	0.078	-57.940	0.015
E_0	HC	0.144	-7.641	-0.002	-0.002	0.428	0.020	0.005	-24.824	0.415	0.095	-58.268	0.020
	VT84F	0.131	-7.598	-0.003	-0.002	0.291	0.005	0.002	-24.630	0.568	0.017	-58.007	-0.003
	LKT	0.134	-7.615	-0.002	-0.002	0.310	0.007	0.001	-24.676	0.583	0.023	-58.050	0.000
	KS	17.4	17.0	17.4	17.4	29.3	37.2	37.7	38.5	65.2	76.0	84.0	81.2
	WGC	17.3	16.9	17.4	17.3	27.8	35.6	36.2	36.6	62.1	74.7	80.9	78.9
B_0	HC	18.1	17.7	18.2	18.1	27.1	41.5	41.7	41.9	60.1	103.4	103.1	102.7
	VT84F	17.2	16.7	17.2	16.7	33.6	34.1	34.1	33.5	86.5	95.5	92.8	93.9
	LKT	17.6	16.9	17.4	17.4	32.2	34.2	34.3	34.5	76.4	89.4	90.1	90.2

TABLE III. Equilibrium volume V_0 (Å^3/cell), energy E_0 (eV/cell), and bulk moduli B_0 (GPa) of simple III-V zinc-blende semiconductors. Systems include AlP, AlAs, AlSb, GaP, GaAs, GaSb, InP, InAs, InSb. Conventional KS results are from Ref. [11] except the B_0 for GaP for which we use 88 GPa from our calculation. The HC data are taken from Ref. [11].

	KEDF	AlP	AlAs	AlSb	GaP	GaAs	GaSb	InP	InAs	InSb
	KS	40.637	43.616	56.607	37.646	40.634	52.488	46.040	49.123	62.908
	WGC	-	-	-	-	-	-	-	-	-
V_0	HC	40.290	44.746	55.917	36.795	41.214	51.779	45.930	50.596	62.461
	VT84F	35.827	39.264	49.983	33.027	36.372	46.502	41.755	45.412	56.560
	LKT	40.258	44.417	55.040	37.313	41.546	51.436	47.316	51.492	62.664
	KS	-239.182	-232.908	-206.606	-243.079	-235.799	-209.697	-235.722	-228.537	-202.387
	WGC	-	-	-	-	-	-	-	-	-
E_0	HC	-238.612	-231.702	-206.309	-242.113	-235.086	-209.686	-233.497	-226.775	-201.572
	VT84F	-231.055	-224.355	-200.638	-234.337	-227.528	-203.883	-225.530	-219.116	-195.738
	LKT	-232.910	-226.228	-201.676	-236.168	-229.411	-205.002	-227.971	-221.501	-197.113
	KS	90	80	60	88	75	56	73	65	50
	WGC	-	-	-	-	-	-	-	-	-
B_0	HC	89	76	61	94	78	62	68	61	49
	VT84F	140.3	120.6	90.5	148.3	124.9	93.4	110.4	97.7	73.5
	LKT	89.9	79.0	61.5	92.0	77.9	62.8	68.4	60.7	49.1

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