Supporting Information for

Local Spectral Formulation of the One-Determines-All (ODA) Principle for Multistate Density Functionals

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This supporting information contains all data used to make figure-plots in the text and the table-of-content graphic (6 pages). The two-site-two-electron Hubbard Model was adopted as an illustrative case for the computational procedure of the one-determines-all (ODA) principle and the spectral decomposition method (6 pages). The goal of this approach is to transform an inter-coupled matrix functional at the state level in multistate density functional theory into a computationally tractable, grid-based integration scheme at a cost comparable to that of Kohn-Sham density functional theory. We note that all computation including generation of target energies and parameter-fitting were carried out using an on-line large-language-model (LLM). The results have been selectively tested and validated. The entire training and computing process took no more than 30 minutes, including the time spent training the artificial intelligence (AI) model and typing keyboard instructions. The outcome also illustrates the integration of an AI model to assist theoretical and computational research. We invite interested readers to repeat and reproduce the results following the steps highlighted in the text which would also help gain a thorough understanding of the ODA principle and spectral decomposition computational procedure.

Table S1. Analytical and spectral decomposition energies (in units *1/t*) of a Hubbard 2-site-2-electron Hubbard model. Energies are given in units *1/t*, computed at different interaction strengths in the unitless scale U/t. Results from local spectral theorem (LST) are determined numerically using spectral decomposition at site 1 and site 2, which give identical data for this symmetrical case. The spectral channel interaction energy function is $g(\lambda_i, \{\lambda_j\}_{\{j\neq i\}}) = \frac{1}{2}U\lambda_i(\lambda_i - 1)$, which accounts for on-site electron interactions. In all calculations $t = 1$.

$\frac{U}{t}$	Analytical (exact)				Local Spectral Theorem (LST)			
	E_0	$E_1(T)$	E_2	E_3	E_0	$E_1(T)$	E_2	E_3
0	-2	0	0	2	-2	0	0	2
1	-1.56155	0	1	2.561553	-1.56155	0	1	2.561553
2	-1.23607	0	2	3.236068	-1.23607	0	2	3.236068
3	-1	0	3	4	-1	0	3	4
4	-0.82843	0	4	4.828427	-0.82843	0	4	4.828427
5	-0.70156	0	5	5.701562	-0.70156	0	5	5.701562
6	-0.60555	0	6	6.605551	-0.60555	0	6	6.605551
7	-0.53113	0	7	7.531129	-0.53113	0	7	7.531129
8	-0.47214	0	8	8.472136	-0.47214	0	8	8.472136
9	-0.42443	0	9	9.424429	-0.42443	0	9	9.424429
10	-0.38516	0	10	10.38516	-0.38516	0	10	10.38516
11	-0.35235	0	11	11.35235	-0.35235	0	11	11.35235

Table S2a. Analytical and spectral decomposition energies (in units *1/t*) of a Hubbard 2-site-2-electron Hubbard model. Energies are given in units *1/t*, computed at different interaction strengths in the unitless scale U/t. In all calculations $t = 1$. Results from local spectral theorem (LST) for the reference state are determined numerically using spectral decomposition at site 1 and site 2, which give identical data for this symmetrical case. The spectral channel interaction energy function is $g^{ref}\left(\lambda_i, \{\lambda_j\}_{\{j\neq i\}}\right) = \frac{1}{2}U\lambda_i(\lambda_i - 1) + 0.1\, U/(\lambda_i + 0.5)$, in which the second term is introduced to artificially distort the exact results to mimic a reference state, analogous to Hartree-Fock theory with respect to full configuration interaction.

$\frac{U}{t}$	Analytical (exact)				Reference State (ref)			
	E_0	$E_1(T)$	E_2	E_3	E_0	$E_1(T)$	E_2	E_3
0	-2	0	0	2.00000	-2.00000	0.00000	0	2.00000
0.45	-1.78762	0	0.45	2.23762	-1.70644	0.06000	0.558	2.32444
0.50	-1.76556	0	0.5	2.26556	-1.67571	0.06667	0.62	2.36238
0.55	-1.74382	0	0.55	2.29382	-1.64536	0.07333	0.682	2.40069
1	-1.56155	0	1	2.56155	-1.38847	0.13333	1.24	2.76180
2	-1.23607	0	2	3.23607	-0.91243	0.26667	2.48	3.65910
3	-1	0	3	4.00000	-0.53915	0.40000	3.72	4.65915
4	-0.82843	0	4	4.82843	-0.23643	0.53333	4.96	5.72976
5	-0.70156	0	5	5.70156	0.01947	0.66667	6.2	6.84719
6	-0.60555	0	6	6.60555	0.24413	0.80000	7.44	7.99587
7	-0.53113	0	7	7.53113	0.44746	0.93333	8.68	9.16588
8	-0.47214	0	8	8.47214	0.63583	1.06667	9.92	10.35084
9	-0.42443	0	9	9.42443	0.81340	1.20000	11.16	11.54660
10	-0.38516	0	10	10.38516	0.98298	1.33333	12.4	12.75035
11	-0.35235	0	11	11.35235	1.14650	1.46667	13.64	13.96017
12	-0.32456	0	12	12.32456	1.30533	1.60000	14.88	15.17467

Table S2b. Correlation-corrected energies (in units *1/t*) with respect to the reference state (see legend of Table S2a), using a fourth-order polynomial, channel-density-only function to approximate the missing correlation

$$g^c(\lambda) = 6.99793 + 1.45722\lambda - 3.42128\lambda^2 - 10.0951\lambda^3 + 4.77050\lambda^4$$

and a channel density and interaction strength coupled brute-force (polynomial) function

$$g^c(\lambda, U) = \sum_{m=0}^{2} \quad \sum_{n=0}^{4} a_{mn}\, U^m \lambda^n$$

for the Hubbard 2-site-2-electron Hubbard model with $t = 1$. Results from local spectral theorem (LST) for the reference state are determined numerically using spectral decomposition at site 1 and site 2, which give identical data for this symmetrical case. The spectral channel reference interaction energy function is $g^{ref}\left(\lambda_i, \{\lambda_j\}_{\{j\neq i\}}\right) = \frac{1}{2}U\lambda_i(\lambda_i - 1) + 0.1\, U/(\lambda_i + 0.5)$, in which the second term is introduced to artificially distort the exact results to mimic a reference state, analogous to Hartree-Fock theory with respect to full configuration interaction.

$\frac{U}{t}$	Density only $g^c(\lambda)$				Density-energy coupled $g^c(\lambda, U)$			
	E_0	$E_1(T)$	E_2	E_3	E_0	$E_1(T)$	E_2	E_3
0	-2.91891	-1.20765	-0.58146	1.12980	-2.00000	0.0	0.0	2.00000
0.45	-2.58658	-0.64965	-0.52146	1.41547	-1.78762	0.0	0.45	2.23762
0.50	-2.55155	-0.58765	-0.51480	1.44911	-1.76556	0.0	0.5	2.26556
0.55	-2.51691	-0.52565	-0.50813	1.48313	-1.74382	0.0	0.55	2.29382
1	-2.22226	-0.44813	0.03235	1.80649	-1.56155	0.0	1	2.56155
2	-1.67291	-0.31480	1.27235	2.63047	-1.23607	0.0	2	3.23607
3	-1.24581	-0.18146	2.51235	3.57670	-1.00000	0.0	3	4.00000
4	-0.90668	-0.04813	3.75235	4.61090	-0.82843	0.0	4	4.82843
5	-0.62666	0.08520	4.99235	5.70422	-0.70156	0.0	5	5.70156
6	-0.38585	0.21854	6.23235	6.83675	-0.60555	0.0	6	6.60555
7	-0.17143	0.35187	7.47235	7.99565	-0.53113	0.0	7	7.53113
8	0.02478	0.48520	8.71235	9.17278	-0.47214	0.0	8	8.47214
9	0.20804	0.61854	9.95235	10.36285	-0.42443	0.0	9	9.42443
10	0.38186	0.75187	11.19235	11.56236	-0.38516	0.0	10	10.38516
11	0.54861	0.88520	12.43235	12.76895	-0.35235	0.0	11	11.35235
12	0.70995	1.01854	13.67235	13.98094	-0.32456	0.0	12	12.32456

Table S3. Fitted polynomial coefficients for the density-only correlation-correction channel function $g^c(\lambda)$ and density-interaction energy coupled correlation function $g^c(\lambda, U)$. All parameters were fitted by minimizing the least square error function in the energy range of $[0, 4]$ and $[7, 10]$ as training, and plotted in the text in the full range of $[0, 12]$ for validation.

$$\min_{A} \sum_{U\in[0,4]\cup[7,10]} \sum_{I} \left(E_I^{\text{exact}}(U) - E_I^{\text{model}}(U; \boldsymbol{A})\right)^2$$

Note:

- The fit range was only 0–4 and 7–10 for U with $t = 1$,
- the target was the exact energies,
- the variables being changed were the coefficients in the matrix $\boldsymbol{A}$ (below),
- and the fitting was done through the energies of the reconstructed distorted-correlation corrected model, not by fitting directly to the exact $g^{exact}(\mathbf{D})$.

(a). For the density only channel function:

$$g^c(\lambda) = 6.99793 + 1.45722\lambda - 3.42128\lambda^2 - 10.0951\lambda^3 + 4.77050\lambda^4$$

(b). For the channel density and interaction strength coupled function

$$g^c(\lambda, U) = \sum_{m=0}^{2} \sum_{n=0}^{4} a_{mn}\, U^m \lambda^n$$

Optimized coefficient matrix $\boldsymbol{A}$:

a_{mn}	0	1	2	3	4
0	-0.02364	0.02599	0.06841	-0.10673	0.03597
1	-0.00733	-0.01029	-0.02098	-0.04103	0.01296
2	0.32041	0.31844	-0.22493	-0.78059	0.36667

Table S4. Numerical results on the channel correlation function $g^c(\lambda, U)$ at $U = 2, 6, and\ 9$. Due to the simplicity of the Hubbard model in which the eigenvalues of $\mathbf{D}(\boldsymbol{r}_s)$ are $\lambda = \{0, 1, 1, 2\}$ on both sites and throughout independent of U. Thus, even though continuous functions were fitted, the only relevant spectral channels are these four eigenvalues.

lambda	g2	g6	g9
0	1.2433	11.4671	25.8636
0.1	1.3594	12.5013	28.1922
0.2	1.4402	13.2202	29.8135
0.3	1.4715	13.5012	30.4529
0.4	1.4425	13.2534	29.9072
0.5	1.3462	12.4178	28.0450
0.6	1.1794	10.9676	24.8064
0.7	0.9421	8.9078	20.2033
0.8	0.6386	6.2753	14.3192
0.9	0.2763	3.1391	7.3091
1	-0.1333	-0.4000	-0.6000
1.1	-0.5754	-4.2091	-9.1099
1.2	-1.0312	-8.1235	-17.8504
1.3	-1.4784	-11.9463	-26.3799
1.4	-1.8911	-15.4489	-34.1848
1.5	-2.2395	-18.3708	-40.6804
1.6	-2.4904	-20.4191	-45.2098
1.7	-2.6067	-21.2695	-47.0448
1.8	-2.5479	-20.5655	-45.3854
1.9	-2.2694	-17.9184	-39.3601
2	-1.7235	-12.9081	-28.0256