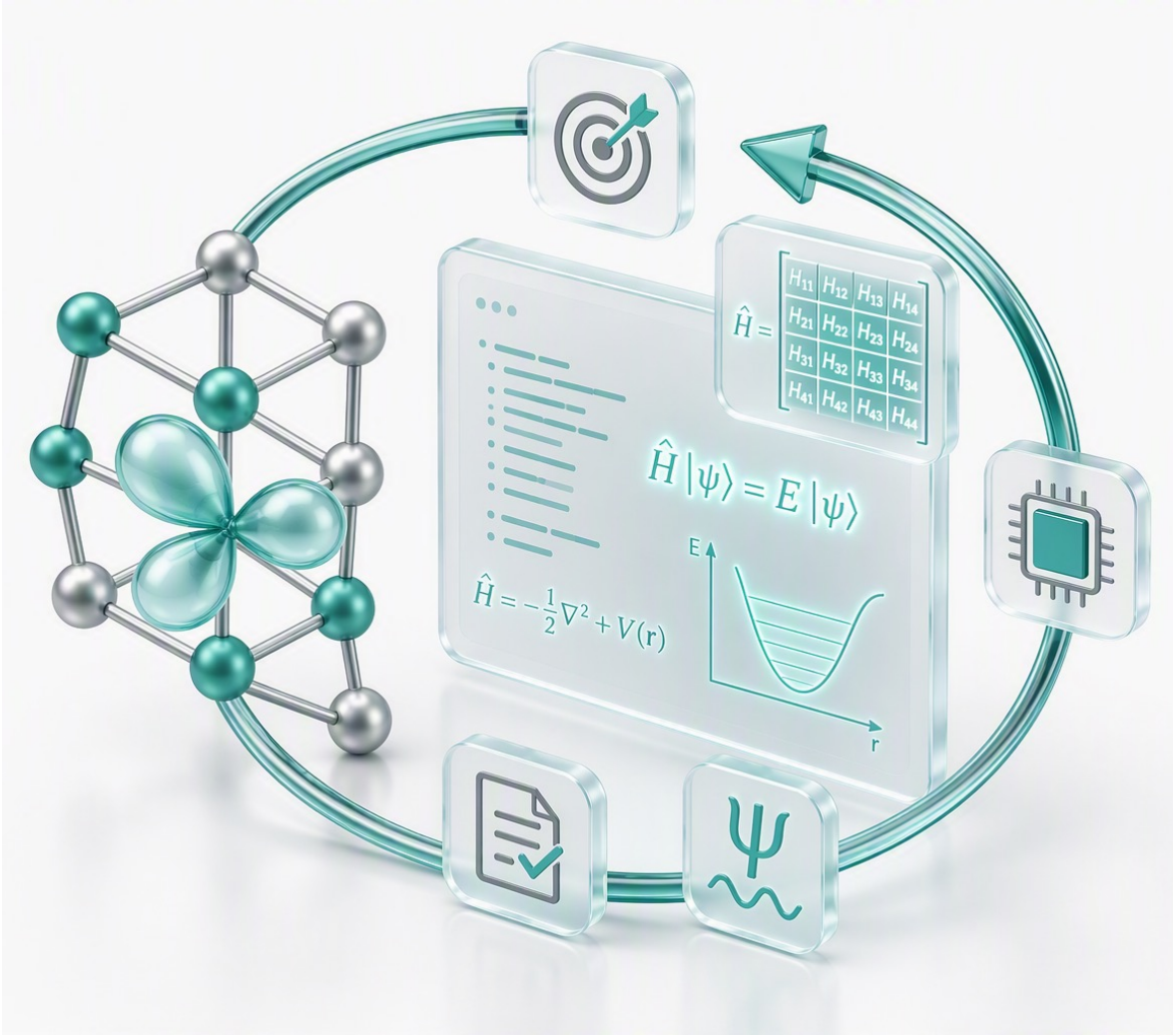
中国科学院大学 University of Chinese Academy of Sciences
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Variation Training of Environment-Adaptive Atomic Orbital Basis, Agentic Designing XC Functional with iKS and Differentiable Electronic Structure Program - GradSCF

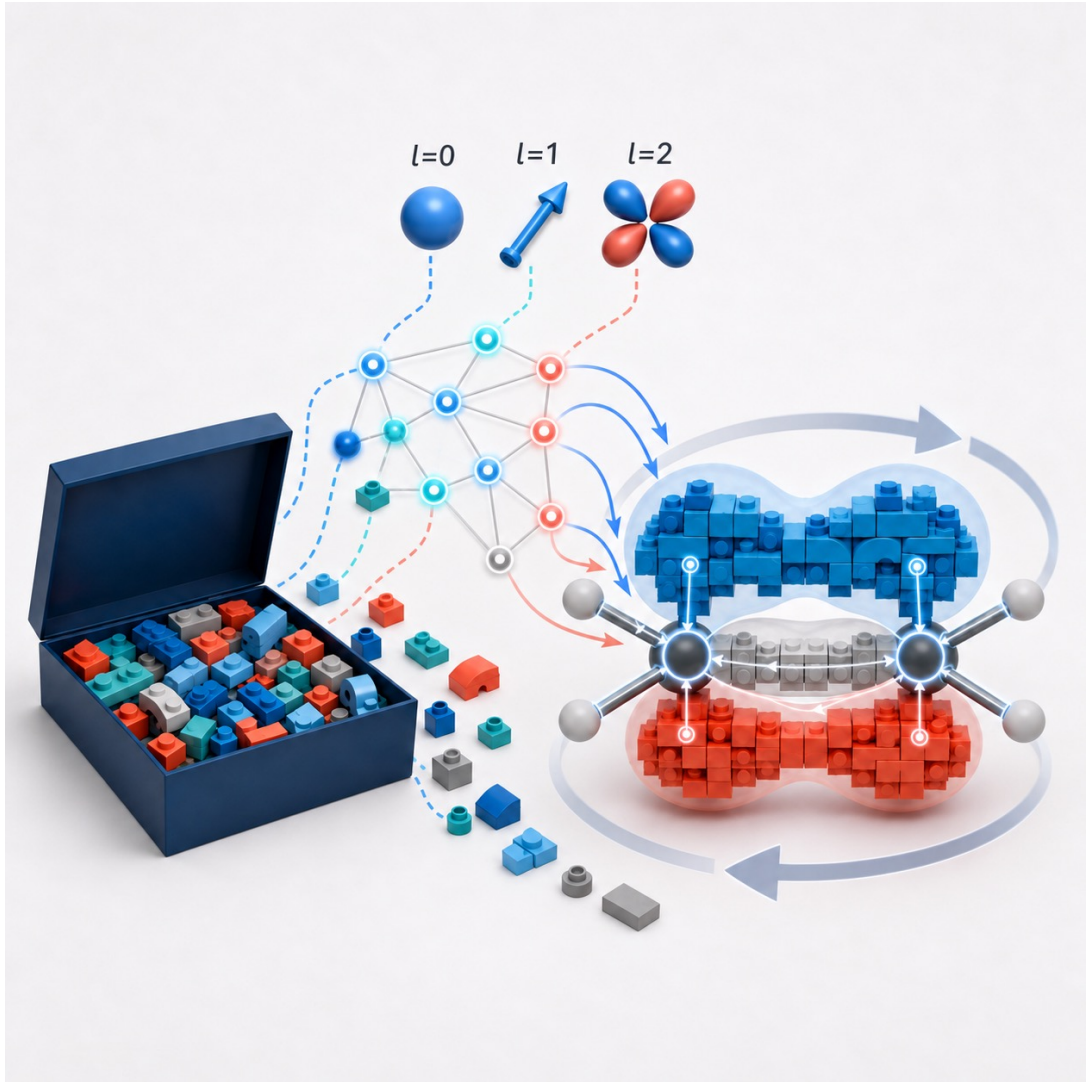


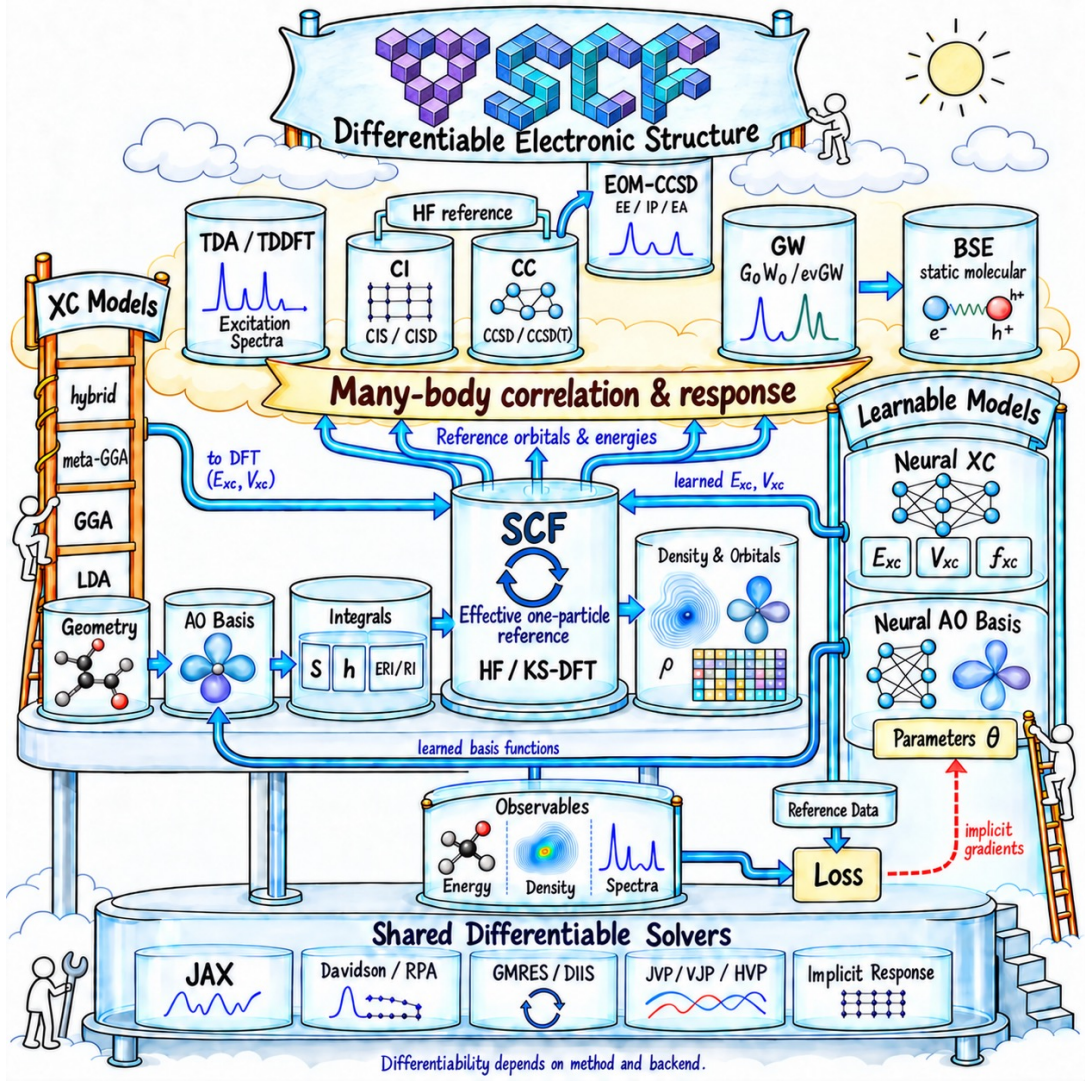
Yuan Jiao

School of Advanced Interdisciplinary Sciences, SAIS

University of Chinese Academy of Sciences

- **Variation Training of Environment-Adaptive Atomic Orbital Basis**
- **Inverse Kohn-Sham Scheme and Self-Consistent Functional Design**
- **From GradTDDFT to GradSCF – Basic Module of GradSCF**

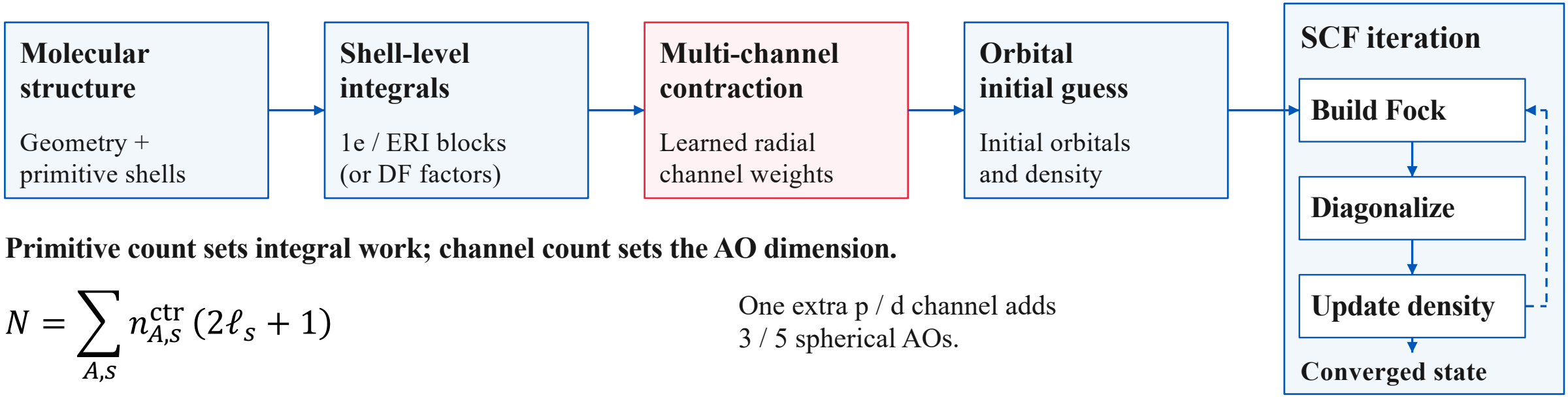




■ Variation Training of Environment-Adaptive Atomic Orbital Basis

- **Background: from molecular structure to self-consistency**

Contract shell integrals into the working AO basis before the SCF iterations.



Primitive count sets integral work; channel count sets the AO dimension.

$$N = \sum_{A,s} n_{A,s}^{\mathrm{ctr}} \, (2\ell_s + 1)$$

One extra p / d channel adds 3 / 5 spherical AOs.

AO size controls cost per cycle; cycle count also depends on the initial guess and conditioning.

Operation	Storage	Leading work
Matrices / diagonalization	$\mathcal{O}(N^2)$	$\mathcal{O}(N^3)$
Dense ERI / unscreened J-K	$\mathcal{O}(N^4)$	$\mathcal{O}(N^4)$
NNAO: DF factors / J and K	$\mathcal{O}(MN^2)$	J: $\mathcal{O}(MN^2)$, K: $\mathcal{O}(MN^2 n_{\mathrm{occ}})$

M: auxiliary functions; n_occ: occupied orbitals. Formal estimates; screening and direct builds change actual costs.

■ Variation Training of Environment-Adaptive Atomic Orbital Basis

- **One-Electron Integral**

$$S_{\mu\nu} = \langle \chi_\mu \mid \chi_\nu \rangle \qquad h_{\mu\nu} = \langle \chi_\mu \mid \hat{h} \mid \chi_\nu \rangle$$

$$\hat{h} = -1/2\,\nabla^2 - \sum_A Z_A\,|\mathbf{r} - \mathbf{R}_A|^{-1}$$

- **Two-Electron Integral**

$$(\mu\nu \mid \lambda\sigma) = \iint \frac{\chi_\mu(\mathbf{r}_1)\chi_\nu(\mathbf{r}_1)\chi_\lambda(\mathbf{r}_2)\chi_\sigma(\mathbf{r}_2)}{r_{12}}\, d\mathbf{r}_1\, d\mathbf{r}_2$$

- **From contracted functions to integral transformations**

$$g_{A,s,p,m}(\mathbf{r}) = N_{A,s,p}\, r_A^{\ell_s} e^{-\alpha_{A,s,p} r_A^2} Y_{\ell_s m}(\hat{\mathbf{r}}_A), \qquad m = -\ell_s, \ldots, \ell_s$$

$$\chi_{A,s,q,m}(\mathbf{r}) = \sum_p \bar{d}_{A,s,q,p}\; g_{A,s,p,m}(\mathbf{r}) = R_{A,s,q}(r_A) Y_{\ell_s m}(\hat{\mathbf{r}}_A)$$

$$\left[S_{\text{prim}}\right]_{ab} = \int g_a(\mathbf{r}) g_b(\mathbf{r})\, d\mathbf{r}, \qquad \left[h_{\text{prim}}\right]_{ab} = \int g_a(\mathbf{r}) \hat{h}\, g_b(\mathbf{r})\, d\mathbf{r}$$

$$(ab \mid cd)_{\text{prim}} = \iint \frac{g_a(\mathbf{r}_1) g_b(\mathbf{r}_1) g_c(\mathbf{r}_2) g_d(\mathbf{r}_2)}{r_{12}}\, d\mathbf{r}_1\, d\mathbf{r}_2$$

$$T_{A,s} = \bar{D}_{A,s} \otimes I_{2\ell_s+1}$$

$$S = T^{\mathsf{T}} S_{\text{prim}} T, \qquad h = T^{\mathsf{T}} h_{\text{prim}} T$$

$$(\mu\nu \mid \lambda\sigma) = \sum_{abcd} T_{a\mu}\, T_{b\nu} T_{c\lambda} T_{d\sigma} (ab \mid cd)_{\text{prim}}$$

At fixed exponents, training differentiates cached mixed-pool projections; calculations use density fitting.

■ Variation Training of Environment-Adaptive Atomic Orbital Basis

- **Self-consistent energy in the contracted basis**

Normalized network outputs define the active projection blocks; fixed core blocks are identities.

$$\mathbf{u}_\theta = f_\theta(\mathcal{R}), \qquad U_{A,s}(\theta) = \bar{D}_{A,s}(\mathbf{u}_\theta) \otimes I_{2\ell_s+1}$$

$$D_{\text{pool}} = U_\theta P U_\theta^{\mathsf{T}}, \qquad F_\theta(P) = U_\theta^{\mathsf{T}}\left[h_{\text{pool}} + J_{\text{pool}}\left(D_{\text{pool}}\right) - 1/2\, K_{\text{pool}}\left(D_{\text{pool}}\right)\right]U_\theta$$

$$P = 2C_{\text{occ}}C_{\text{occ}}^{\mathsf{T}}, \qquad C^{\mathsf{T}}SC = I \qquad\qquad F(P^\star)C^\star = SC^\star\varepsilon^\star$$

$$E[P] = \text{Tr}(Ph) + \frac{1}{2}\text{Tr}[PJ(P)] - \frac{1}{4}\text{Tr}[PK(P)] + E_{\text{nuc}}$$

- **Implicit derivative of the SCF**

$L = L(\theta, D_\theta)$ with $D_\theta = T_{SCF}^K(D_0)$

$$R(D_\theta, \theta) = F_\theta(D_\theta)D_\theta S - SD_\theta F_\theta(D_\theta) = 0 \qquad \nabla_\theta L = \partial_\theta L(\theta, D_\theta) + \partial_{\rho_\theta} L(\theta, D_\theta)\frac{\partial_\theta R}{\partial_{D_\theta} R}$$

Differentiate the converged root rather than the solver trajectory.
The overlap response is retained, including the metric (Pulay) contribution.

■ Variation Training of Environment-Adaptive Atomic Orbital Basis

Variational learning of contractions

MACE invariants predict signed, shell-wise radial coefficients.

$$\mathbf{u}_{A,s,q} = f_{\theta,Z_A}\left(\mathbf{h}_A^{\mathrm{inv}}\right)$$

$$\mathbf{u}_{A,s,q}(Q\mathcal{R}) = \mathbf{u}_{A,s,q}(\mathcal{R})$$

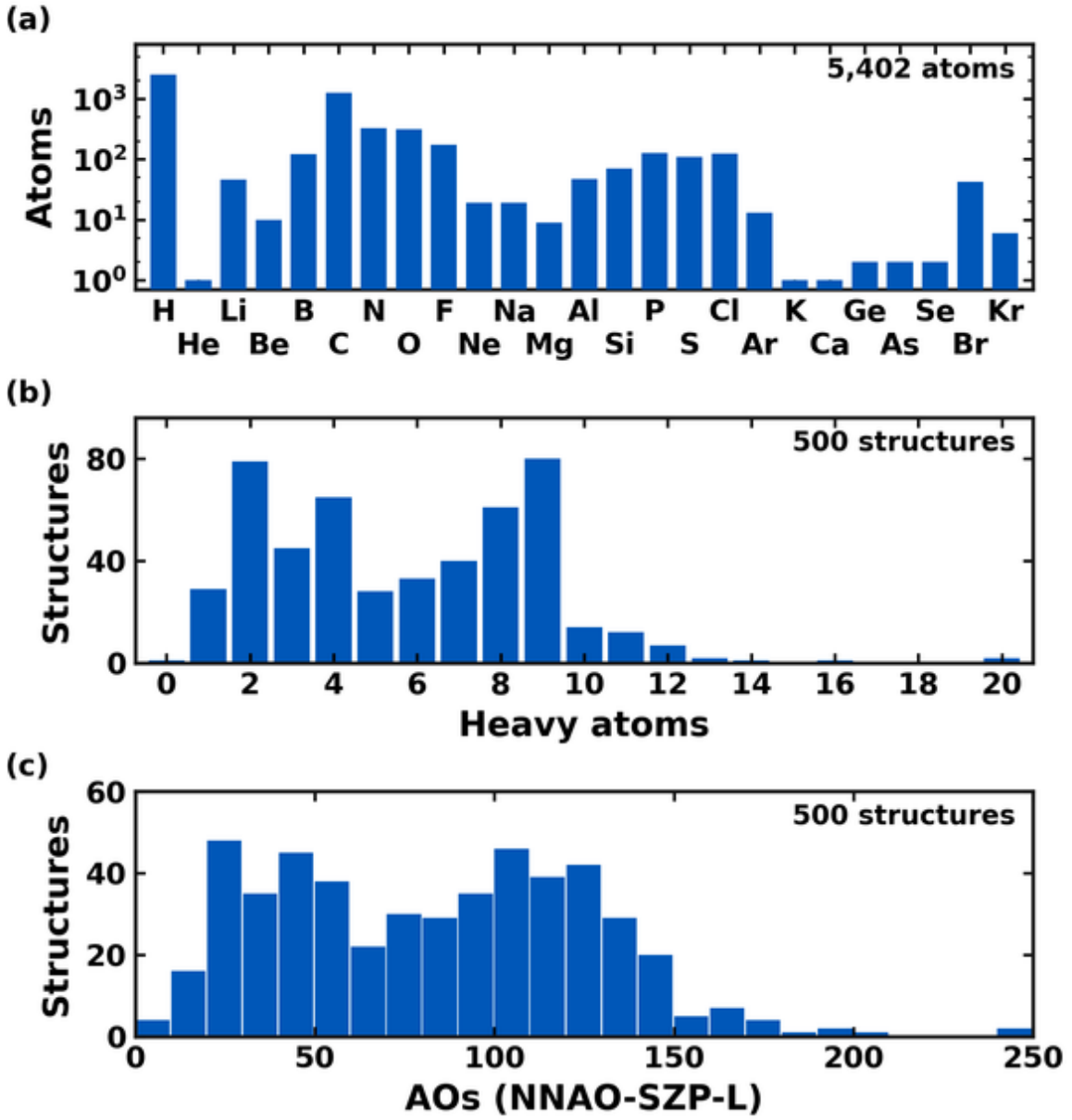
Shared radial coefficients preserve the rotational covariance of each complete spherical shell.

Energy-only objective

$$\mathcal{L}_{\mathrm{C}}\left(\theta;\hat{\beta}\right) = \frac{1}{N_{\mathrm{train}}}\sum_i E_i^{\mathrm{DF},\star}\left(\mathbf{d}_i(\theta),\hat{\beta}\right)$$

Calibrate the exponent pool first; hold it fixed during MACE training.

Free-contraction solutions are diagnostic references, not training labels.



■ Variation Training of Environment-Adaptive Atomic Orbital Basis

- **Environment adaptation** Fixed AO count; geometry-dependent radial contractions.

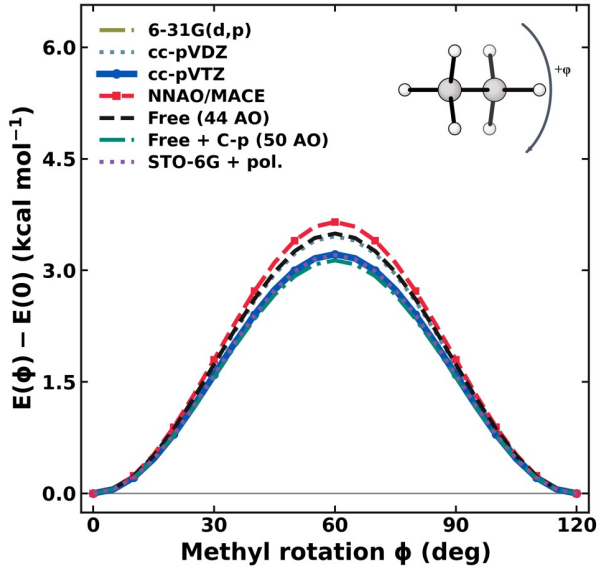
TABLE I. H and C shell structures of the comparison Gaussian basis sets.

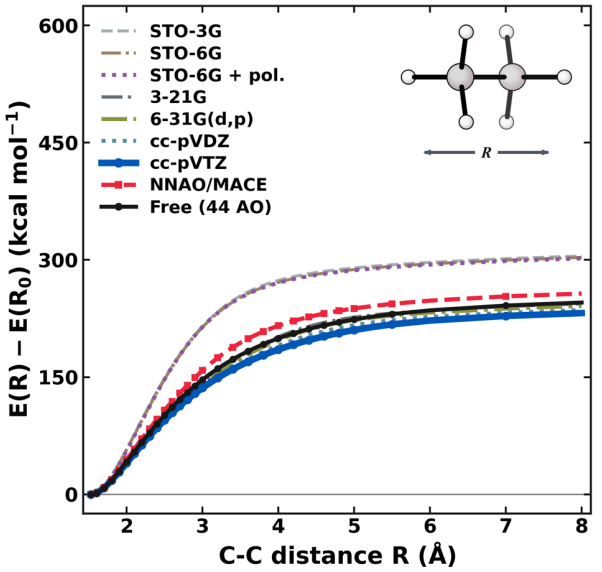
	Hydrogen (H)			Carbon (C)		
Basis	Primitives	Contracted	N_{AO}	Primitives	Contracted	N_{AO}
STO-3G	$(3s)$	$[1s]$	1	$(6s3p)$	$[2s1p]$	5
STO-6G	$(6s)$	$[1s]$	1	$(12s6p)$	$[2s1p]$	5
STO-6G+P[a]	$(6s1p)$	$[1s1p]$	4	$(12s6p1d)$	$[2s1p1d]$	10
3-21G	$(3s)$	$[2s]$	2	$(6s3p)$	$[3s2p]$	9
6-31G(d,p)	$(4s1p)$	$[2s1p]$	5	$(10s4p1d)$	$[3s2p1d]$	14
def2-SVP	$(4s1p)$	$[2s1p]$	5	$(7s4p1d)$	$[3s2p1d]$	14
cc-pVDZ	$(4s1p)$	$[2s1p]$	5	$(9s4p1d)$	$[3s2p1d]$	14
cc-pVTZ	$(5s2p1d)$	$[3s2p1d]$	14	$(10s5p2d1f)$	$[4s3p2d1f]$	30
ma-def2-QZVPP	$(7s3p2d1f)$	$[4s3p2d1f]$	30	$(16s9p3d2f1g)$	$[8s5p3d2f1g]$	61
NNAO-SZP-L[b]	$(6s2p)$	$[1s1p]$	4	$(12s6p3d)$	$[2s1p1d]$	10

Parentheses and brackets denote primitive and contracted radial shells. N_{AO} includes core functions and uses real spherical AOs ($s, p, d, f, g = 1, 3, 5, 7, 9$).

[a]STO-6G plus one H p and one C d shell. S22 uses $(\alpha_{H,p}, \alpha_{C,d}) = (0.8, 0.8)\, a_0^{-2}$ from def2-SVP; ethane uses $(1.1, 0.8)\, a_0^{-2}$.

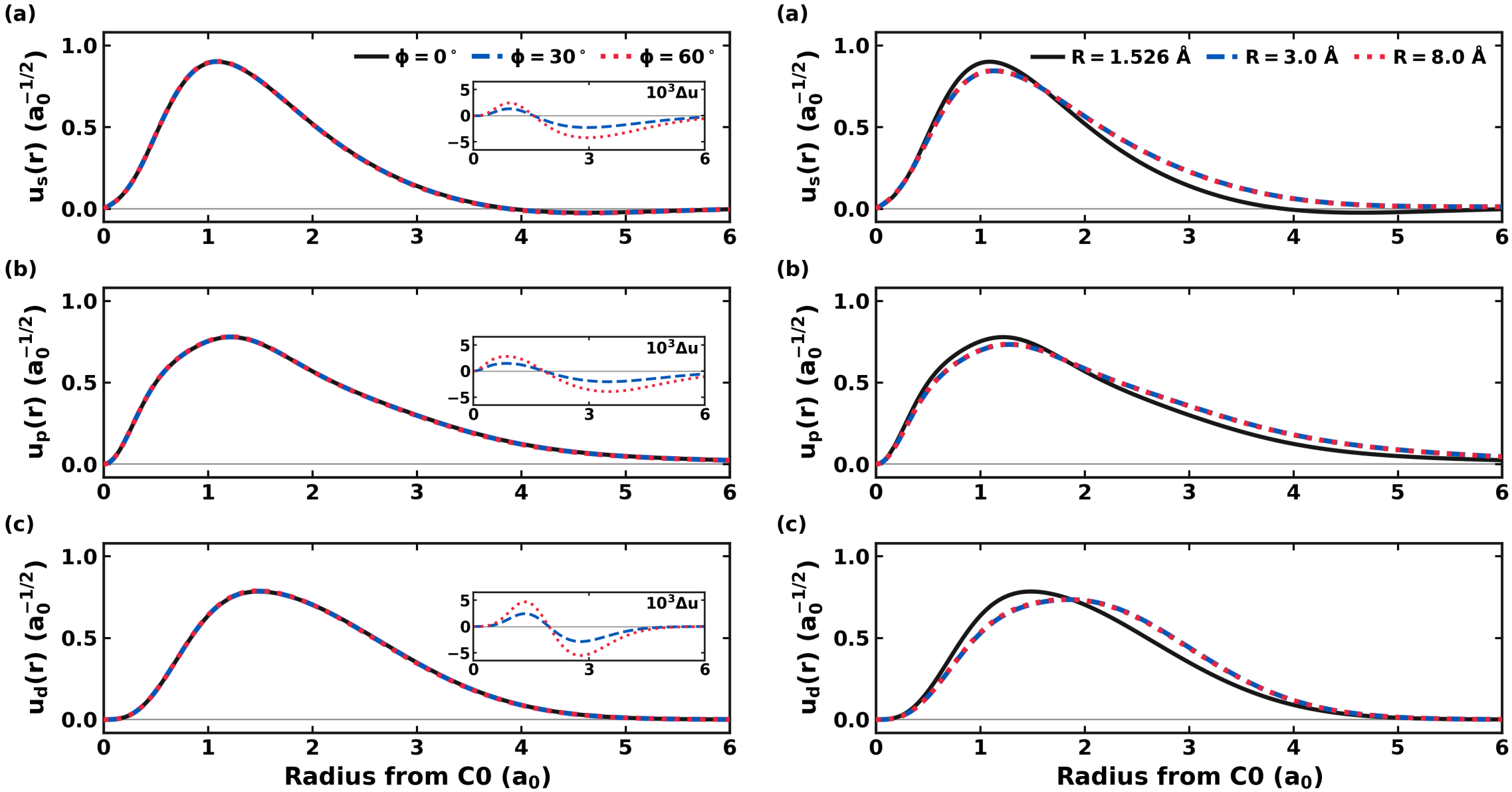
[b]Included for reference. Its C inventory includes one fixed six-primitive core s contraction; the other shells are active.





■ Variation Training of Environment-Adaptive Atomic Orbital Basis

- **Radial response: methyl rotation (left), C-C stretching (right)**

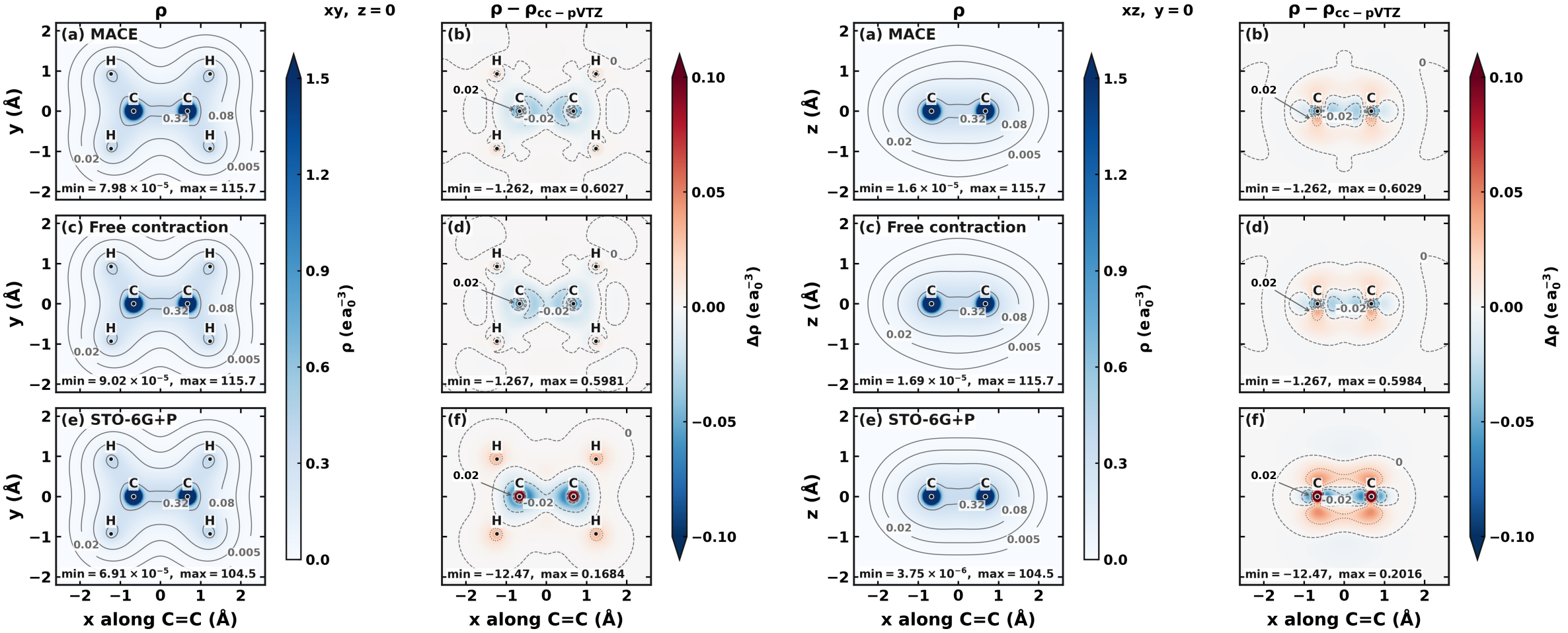


■ Variation Training of Environment-Adaptive Atomic Orbital Basis

- **Electronic density comparison**

$$\rho(\mathbf{r}) = \sum_{\mu\nu} P^{\star}_{\mu\nu}\, \chi_{\mu}(\mathbf{r})\chi_{\nu}(\mathbf{r})$$

Spin-summed SCF density; difference maps use cc-pVTZ.



Rows: MACE / free contractions / STO-6G+P. Each plane shows the density and its difference from cc-pVTZ.

■ Variation Training of Environment-Adaptive Atomic Orbital Basis

- **Basis-set superposition error**

At fixed H, an enlarged trial set can lower each monomer reference energy.

V: orbital space; M(V): many-electron trial set.

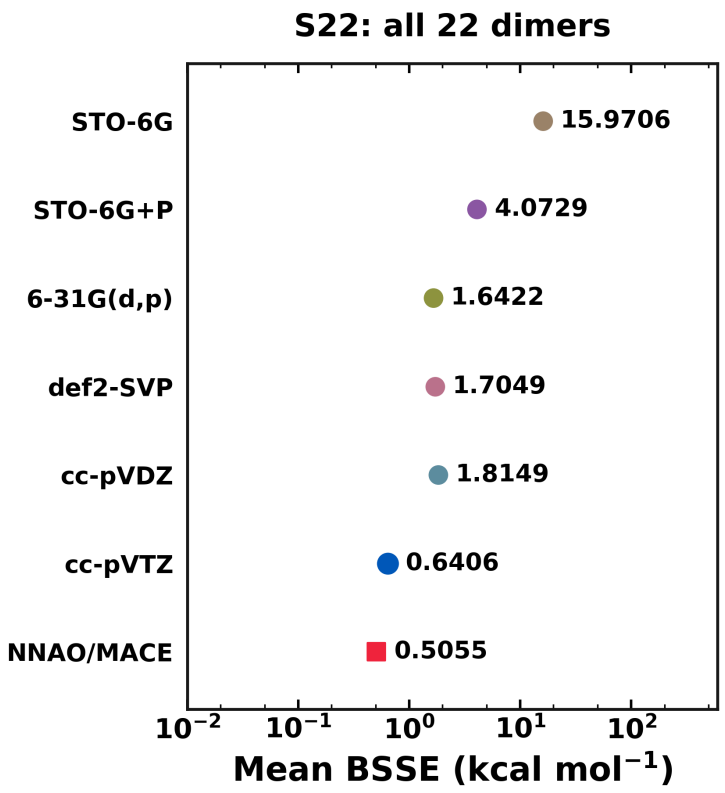
$$E(H,V) = \min_{0 \neq \Psi \in \mathcal{M}(V)} \frac{\langle \Psi | H | \Psi \rangle}{\langle \Psi | \Psi \rangle}$$

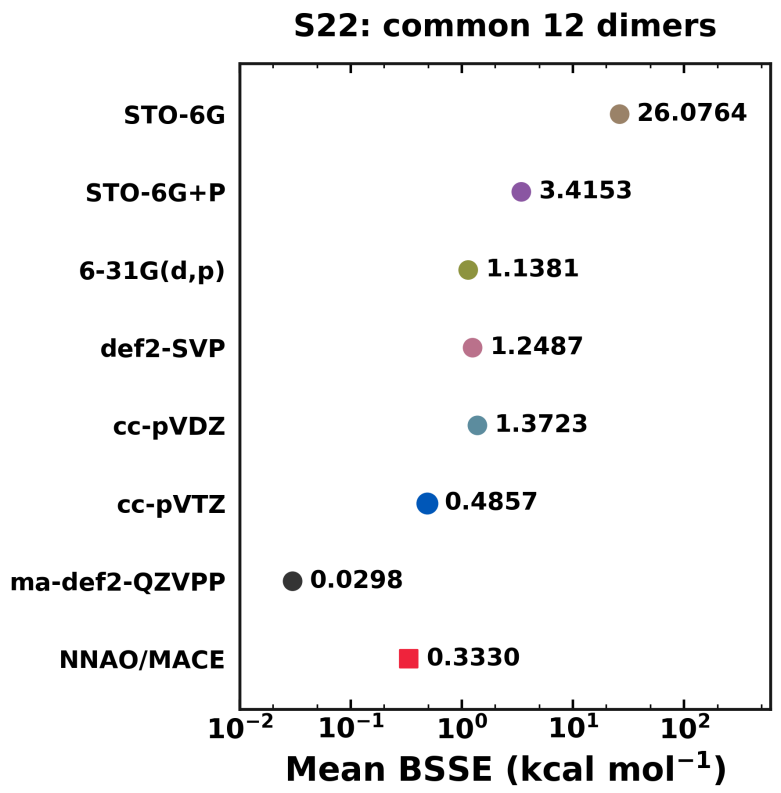
$$\begin{aligned} V_1 \subseteq V_2 \quad &\Rightarrow \\ E_0(H) \leq E(H,V_2) \quad &\leq E(H,V_1) \end{aligned}$$

$$\begin{aligned} U &= V_A + V_B, \qquad V_X \subseteq U \\ d_X &= E_X(V_X) - E_X(U) \geq 0 \end{aligned}$$

$$\begin{aligned} I_{\text{raw}} &= E_{AB}(U) - E_A(V_A) - E_B(V_B) \\ I_{\text{CP}} &= E_{AB}(U) - E_A(U) - E_B(U) \end{aligned}$$

$$I_{\text{raw}} = I_{\text{CP}} - (d_A + d_B)$$



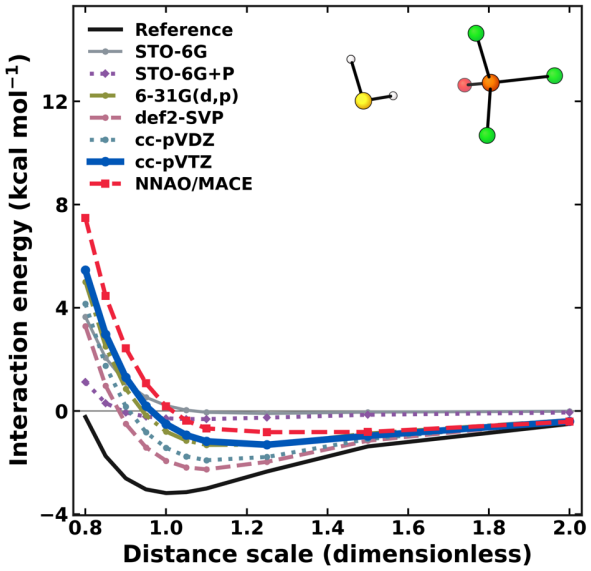


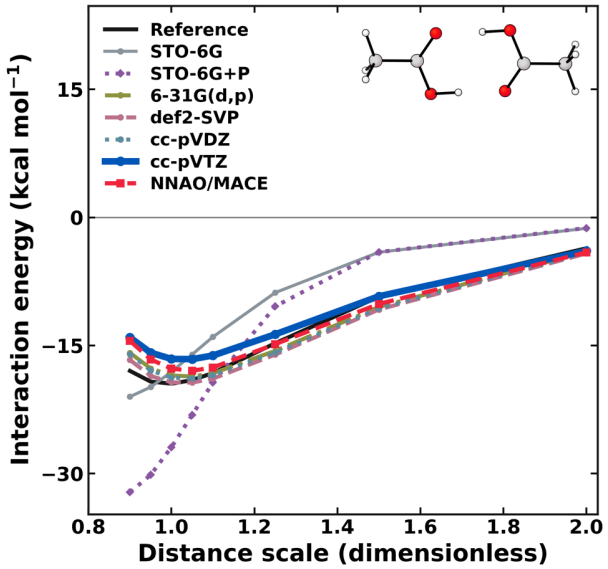
BSSE magnitude = monomer energy lowering.
Ghost functions add flexibility, not interactions.

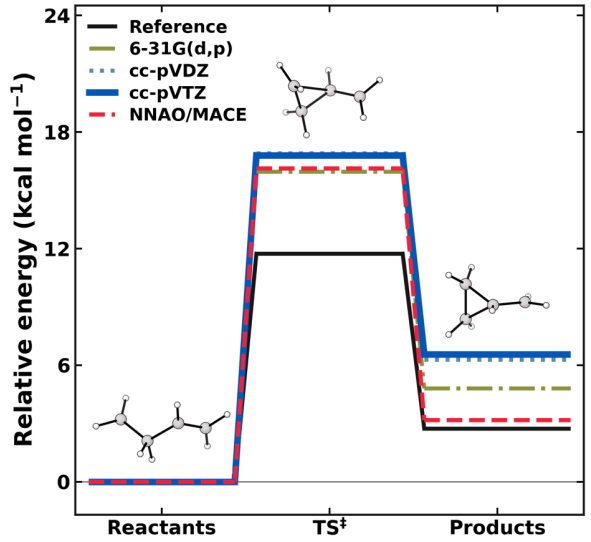
Freeze learned basis parameters across all five energies. CP balances spaces; it does not guarantee the CBS limit.

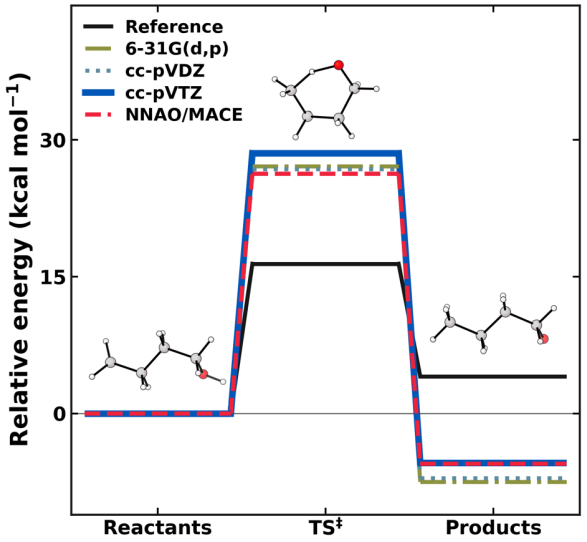
■ Variation Training of Environment-Adaptive Atomic Orbital Basis

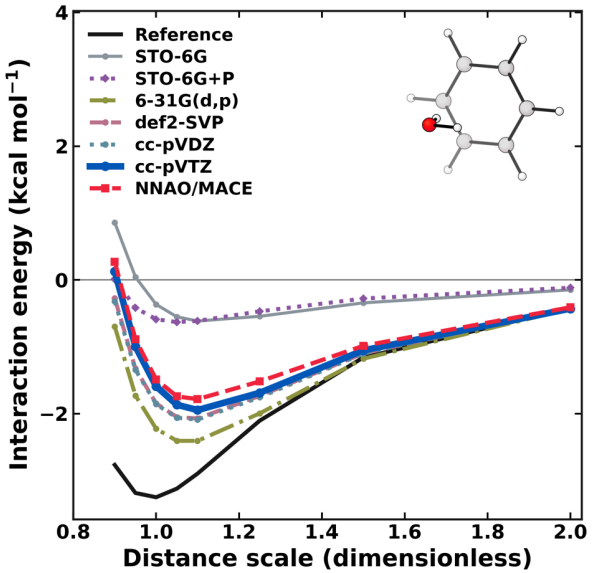
- Interaction curves (left) and reaction barriers (right)

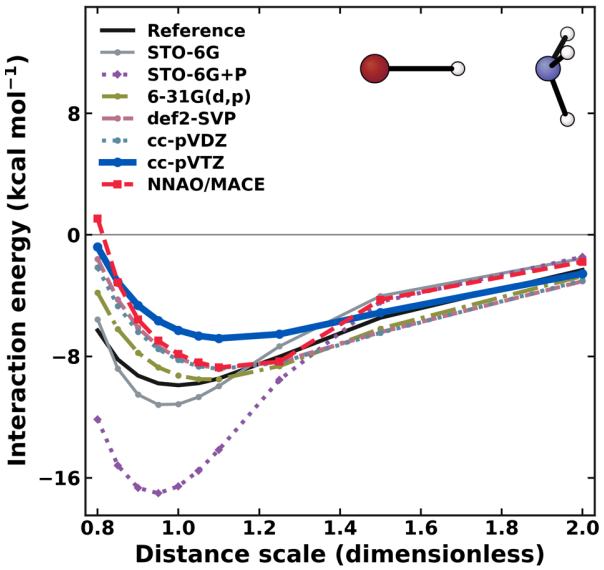


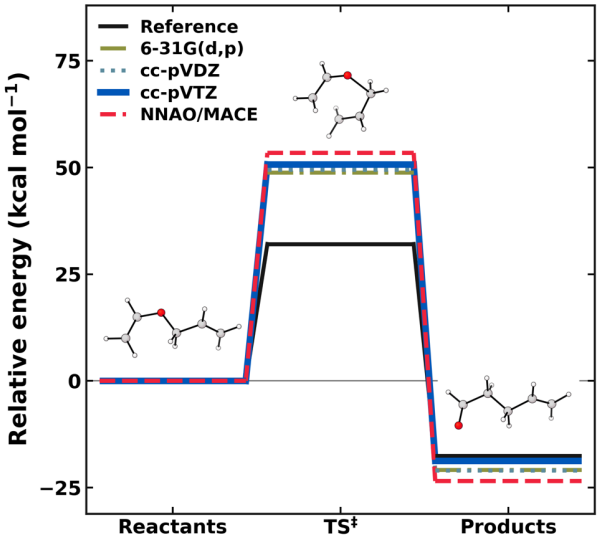


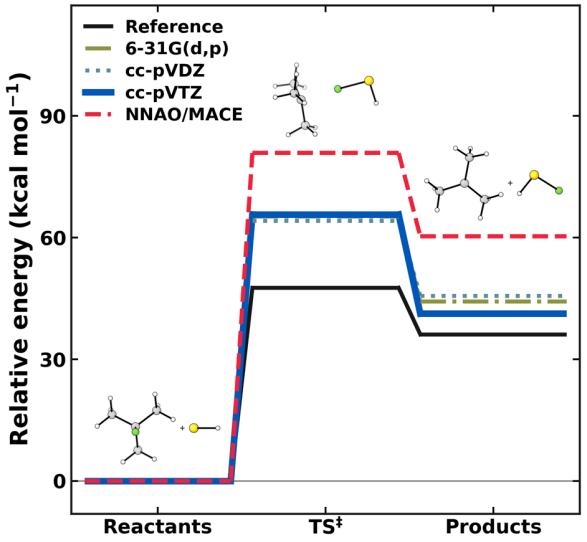












■ Variation Training of Environment-Adaptive Atomic Orbital Basis

- **Benchmark in different methods via gpu4pyscf**

TABLE I. Active-shell structures of the nine NNAO basis families. Parentheses list primitive radial shells and square brackets list contracted radial shells. N_{act} counts contracted active real spherical orbitals per atom; fixed cores are given separately in Table II.

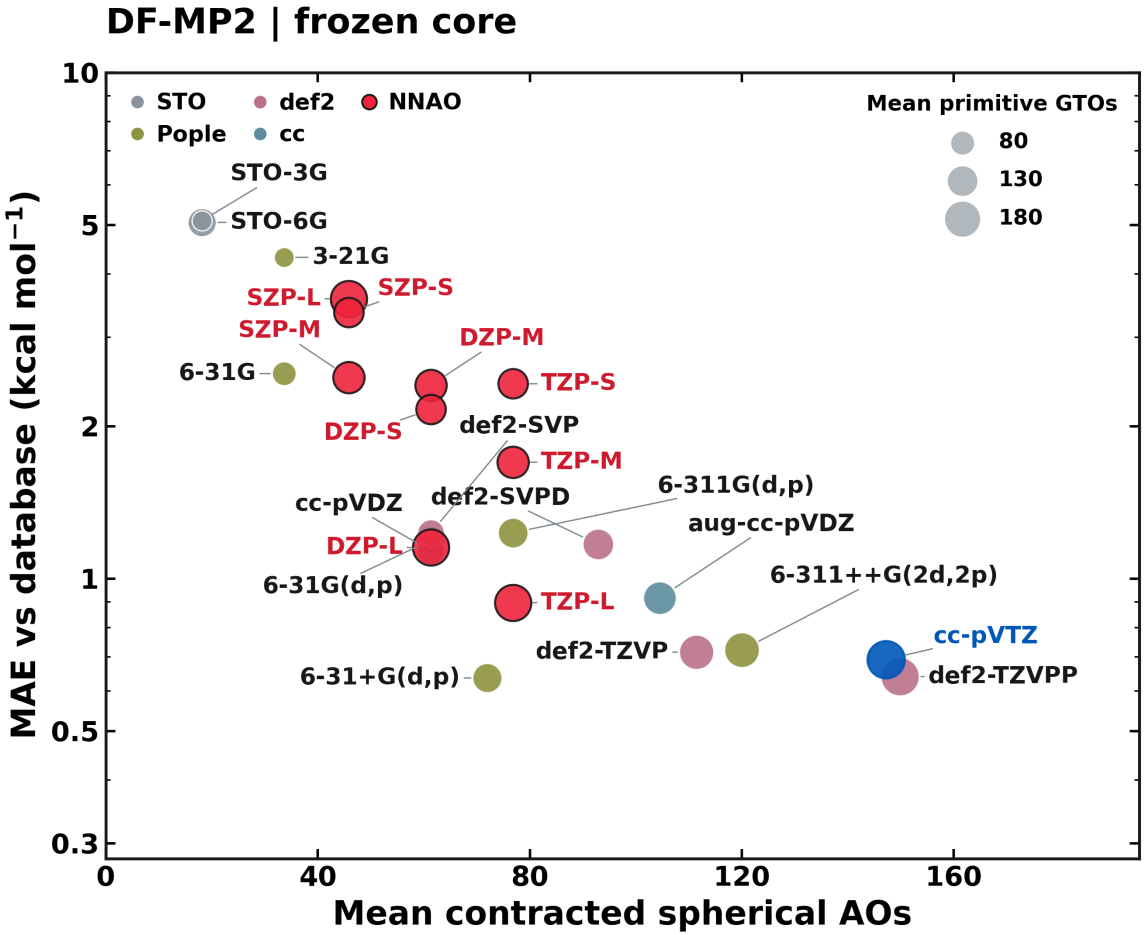
	s-valence templates[a]			*sp*-valence templates[a]		
Basis	Primitives	Contracted	N_{act}	Primitives	Contracted	N_{act}
NNAO-SZP-S	$(3s2p)$	$[1s1p]$	4	$(3s3p2d)$	$[1s1p1d]$	9
NNAO-SZP-M	$(4s2p)$	$[1s1p]$	4	$(4s4p2d)$	$[1s1p1d]$	9
NNAO-SZP-L	$(6s2p)$	$[1s1p]$	4	$(6s6p3d)$	$[1s1p1d]$	9
NNAO-DZP-S	$(3s2p)$	$[2s1p]$	5	$(3s3p2d)$	$[2s2p1d]$	13
NNAO-DZP-M	$(4s2p)$	$[2s1p]$	5	$(4s4p2d)$	$[2s2p1d]$	13
NNAO-DZP-L	$(6s2p)$	$[2s1p]$	5	$(6s6p3d)$	$[2s2p1d]$	13
NNAO-TZP-S	$(3s2p)$	$[3s1p]$	6	$(3s3p2d)$	$[3s3p1d]$	17
NNAO-TZP-M	$(4s2p)$	$[3s1p]$	6	$(4s4p2d)$	$[3s3p1d]$	17
NNAO-TZP-L	$(6s2p)$	$[3s1p]$	6	$(6s6p3d)$	$[3s3p1d]$	17

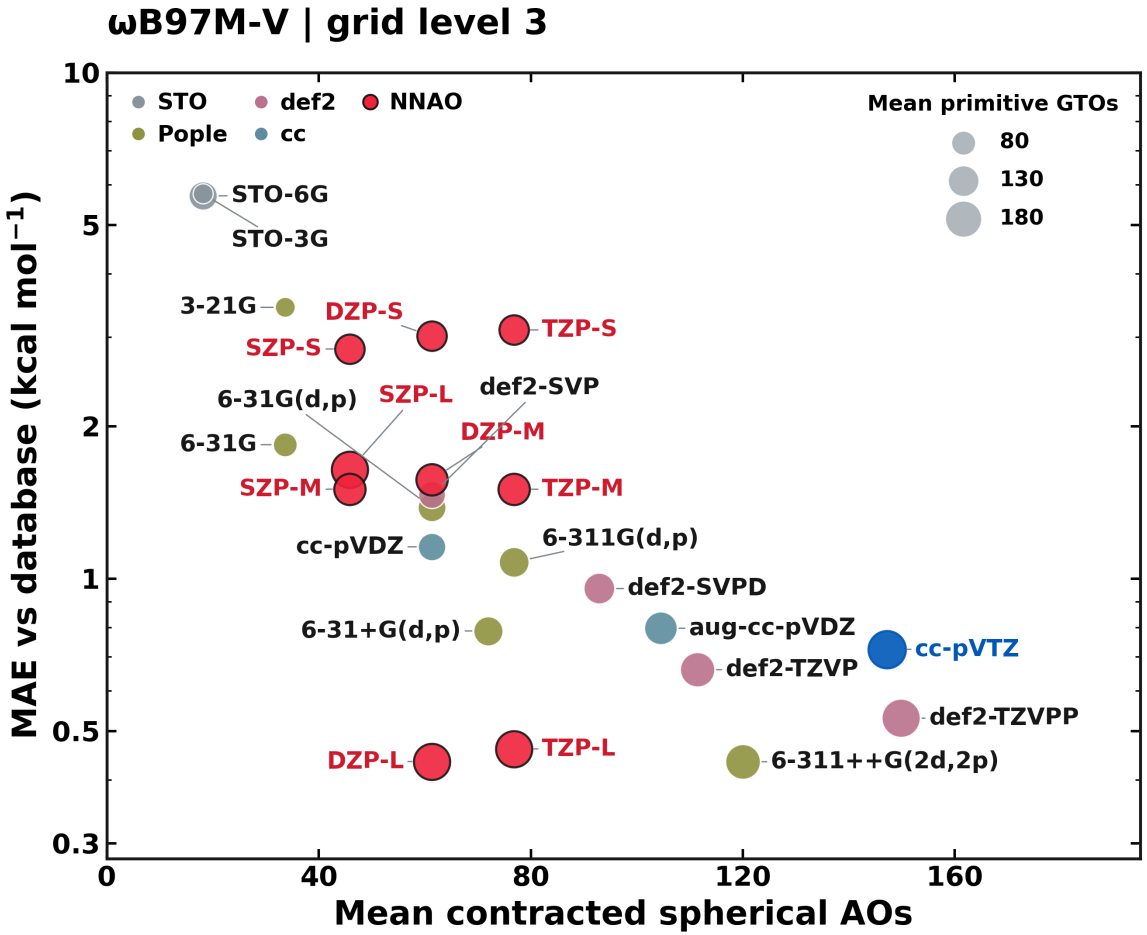
TABLE II. Element coverage and fixed reference cores of the NNAO families. V identifies the active valence channels. The total real spherical AO counts per atom include the fixed core and one active polarization radial function and are independent of the S/M/L pool tier.

				N_{AO}		
Elements	Core primitives[a]	Contracted core	V	SZP	DZP	TZP
H, He	—	—	*s*	4	5	6
Li, Be	$(6s)$	$[1s]$	*s*	5	6	7
B–Ne	$(6s)$	$[1s]$	*sp*	10	14	18
Na, Mg	$(12s6p)$	$[2s1p]$	*s*	9	10	11
Al–Ar	$(12s6p)$	$[2s1p]$	*sp*	14	18	22
K, Ca	$(18s12p)$	$[3s2p]$	*s*	13	14	15
Ga–Kr	$(18s12p3d)$	$[3s2p1d]$	*sp*	23	27	31

■ Variation Training of Environment-Adaptive Atomic Orbital Basis

- Benchmark in different methods via gpu4pyscf

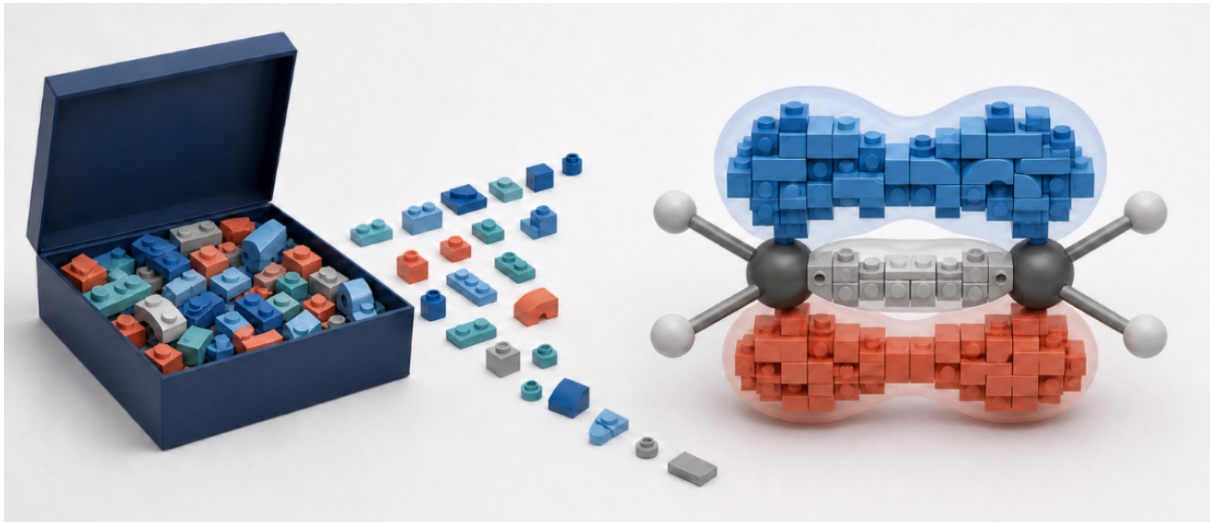




■ Variational Training of Environment-Adaptive Atomic Orbital Basis

- **Outlook**

  - Benchmark of other basis functions
  - IR or Raman Spectrum for Large Systems
  - Wrapping up this work



- **Functional**

  - Neural Network Basis Function for Molecular Systems
  - Variation Module for DFT and HF
  - Form Primitives Gaussian Integral to Differentiable SCF Module
  - Custom Basis Function

■ Inverse Kohn-Sham Scheme and Self-Consistent Functional Design

- **Background**
  - **Fixed-Density Mode**

$$D_\theta = D_{\text{ref}}$$

$$L = L(\theta, D)\Big|_{D=D_{ref}}$$

$$\nabla_\theta L = \partial_\theta L(\theta, D)\Big|_{D=D_{ref}} \qquad \text{with } \partial_\theta D = 0$$

  - **Explicit SCF Differential Mode**

$$L = L(\theta, D_\theta) \qquad \text{with } D_\theta = T^\theta_{SCF}(D_0)$$

$$\nabla_\theta L = \partial_\theta L(\theta, D_\theta) + \partial_{D_\theta} L(\theta, D_\theta) \cdot \partial_\theta D_\theta$$

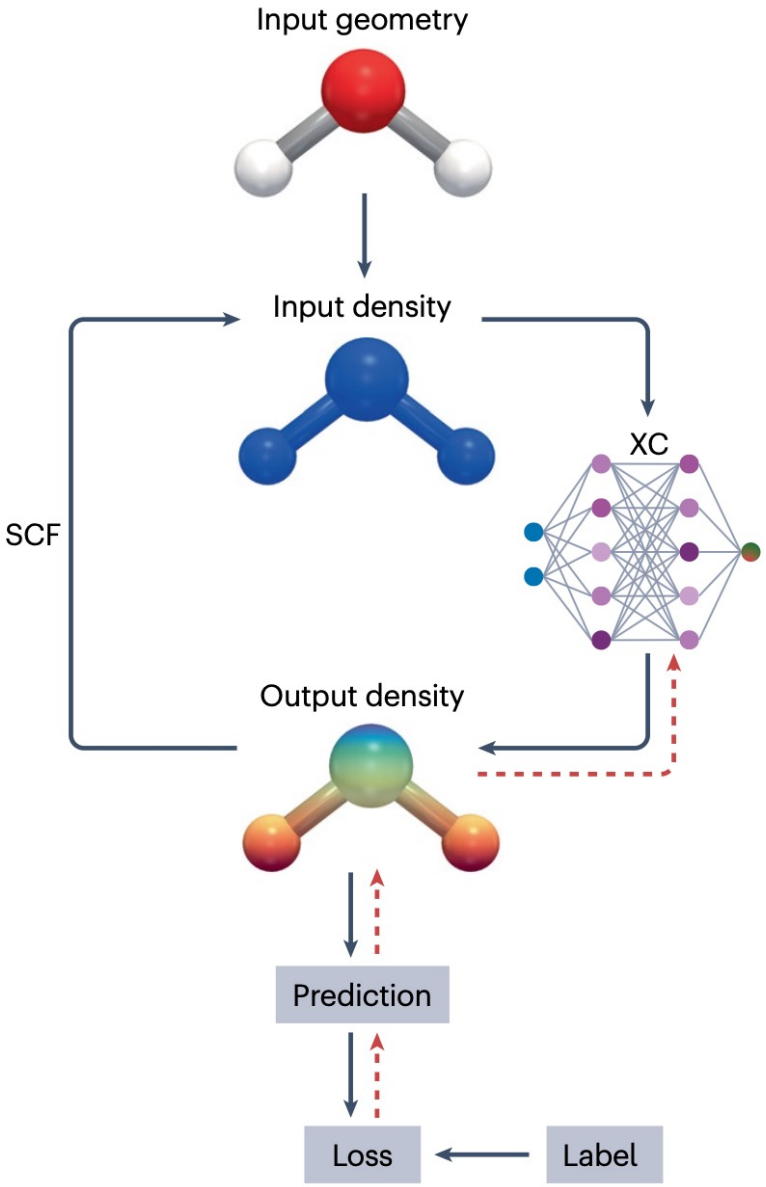
$$\text{with } \partial_\theta D_\theta = \sum_{K-1}[\Pi^{K-1}_{s=t+1} \partial_D T^\theta_{SCF}(D^s)]\partial_\theta T^\theta_{SCF}(D^t)$$

  - **Implicit SCF Differential Mode**

$$L = L(\theta, D_\theta) \qquad \text{with } D_\theta = T^K_{SCF}(D_0)$$

$$R(D_\theta, \theta) = F_\theta(D_\theta)D_\theta S - SD_\theta F_\theta(D_\theta) = 0$$

$$\nabla_\theta L = \partial_\theta L(\theta, D_\theta) + \partial_{\rho_\theta} L(\theta, D_\theta)\frac{\partial_\theta R}{\partial_{D_\theta} R}$$



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■ Inverse Kohn-Sham Scheme and Self-Consistent Functional Design

- **Inverse KS: reconstruct a local potential from a known density**

Forward KS predicts a density from an XC model; inverse KS reconstructs the potential from a target density.

1. Forward KS: functional → density

$$E[n] = T_s[n] + E_{\text{ext}}[n] + E_{\text{H}}[n] + E_{\text{xc}}[n] + E_{\text{NN}}$$

$$[-1/2\,\nabla^2 + v_s[n](\mathbf{r})]\phi_i = \epsilon_i \phi_i$$

$$v_s[n] = v_{\text{ext}} + v_{\text{H}}[n] + \frac{\delta E_{\text{xc}}[n]}{\delta n}$$

$$n(\mathbf{r}) = \sum_i f_i\,|\phi_i(\mathbf{r})|^2, \quad \sum_i f_i = N$$

An approximate XC functional closes the loop.
Solve the orbitals and density self-consistently.

2. Inverse KS: density → potential

Input: a reference density n_0 from CI / CC.

$$[-1/2\,\nabla^2 + v_s(\mathbf{r})]\phi_i(\mathbf{r}) = \epsilon_i \phi_i(\mathbf{r})$$

$$\sum_i f_i\,|\phi_i(\mathbf{r})|^2 = n_0(\mathbf{r})$$

Seek a local potential whose noninteracting ground state reproduces the target density.

$$v_{\text{xc}}(\mathbf{r}) = v_s(\mathbf{r}) - v_{\text{ext}}(\mathbf{r}) - v_{\text{H}}[n_0](\mathbf{r})$$

No analytical exact XC functional is required.
The result is the XC potential at this reference density.

Each system / density requires its own inversion; learning a transferable XC functional is a subsequent task.

Assume noninteracting ground-state v-representability; fix the constant potential gauge. Finite-space inversion is approximate. KS-pies, Sec. II i.

■ Inverse Kohn-Sham Scheme and Self-Consistent Functional Design

- **Wu–Yang inversion: the trial-system energy and the dual functional**

Given the target density n_0, vary the trial potential v; freeze the inverse reference only after inversion.

1. Noninteracting ground-state energy

$$E_s[v] = \min_{\Phi}\left\{T_s[\Phi] + \int v(\mathbf{r}) n_{\Phi}(\mathbf{r})\, d\mathbf{r}\right\}$$
$$= \sum_i f_i\, \epsilon_i[v]$$

E_s includes kinetic and trial-potential energies.

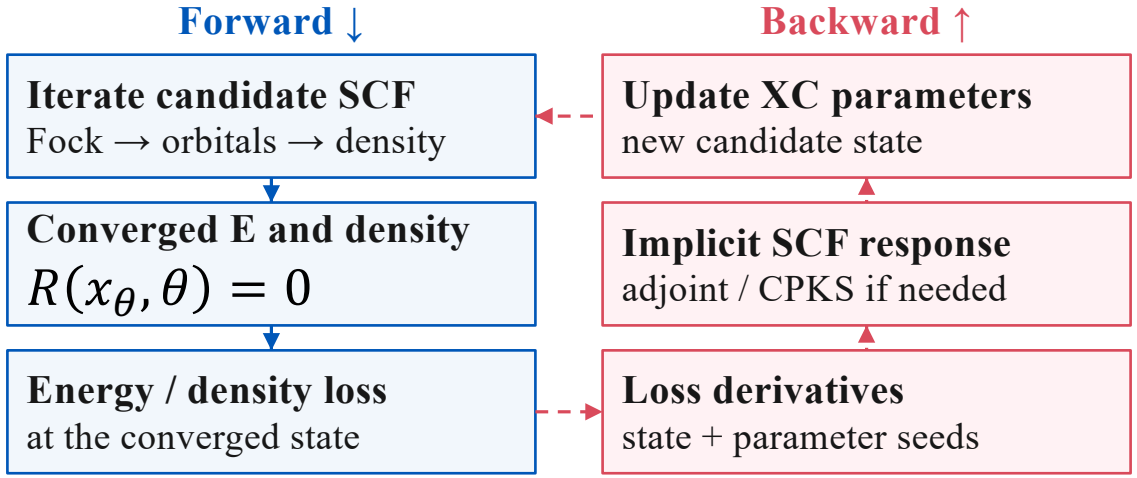
The density mismatch is the gradient of W.

2. Construct and maximize W[v]

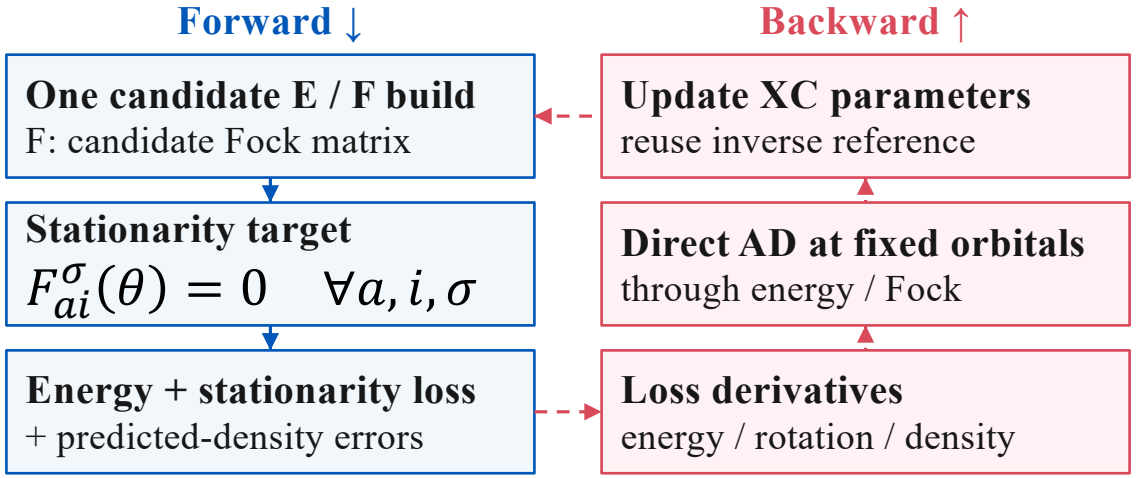
$$W[v] = E_s[v] - \int v(\mathbf{r}) n_0(\mathbf{r})\, d\mathbf{r}$$
$$= T_s[\Phi_v] + \int v(\mathbf{r})[n_v(\mathbf{r}) - n_0(\mathbf{r})]\, d\mathbf{r}$$

$$\max_v W[v] \quad \Leftrightarrow \quad \min_v\{-W[v]\}$$

SCF-based XC training



iKS-based XC training (reference fixed)



Zero Fock residual certifies reference stationarity without candidate SCF; final SCF tests stability and physical accuracy.

Wu & Yang, JCP 118, 2498 (2003); Nam et al., arXiv:2008.08783v2, Sec. II iii; Chen et al., arXiv:2610.30022, pp. 2–3.

■ Inverse Kohn-Sham Scheme and Self-Consistent Functional Design

- Functional
- Inverse Kohn-Sham Module(iGKS, iUKS, iUKS but undertesting)
- Agent (Codex, Claude Code…)
- XC100(Single-Particle Density Matrix, Total Energy in CCSD level)
- Differentiable TDDFT for Excited States

ByteDance | Seed

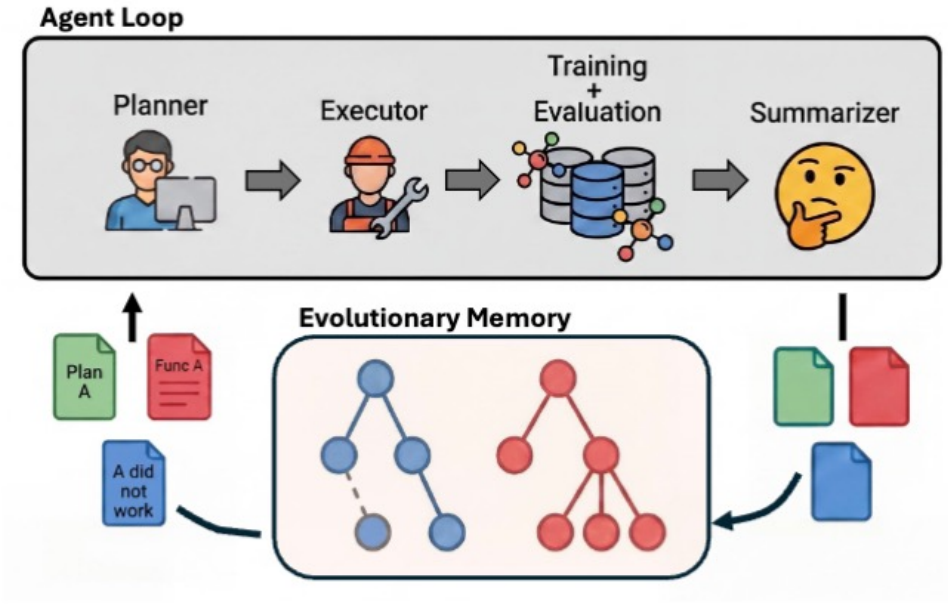
Agentic Discovery of Exchange–Correlation Density Functionals

Titouan Duston[1,2,*], Jiashu Liang[1], Yuanheng Wang[1], Weihao Gao[1], Xuelan Wen[1], Nan Sheng[1], Weiluo Ren[1], Yang Sun[1,†], Yixiao Chen[1,†]

[1]ByteDance Seed, [2]Princeton University

*Work done at ByteDance Seed, †Corresponding authors

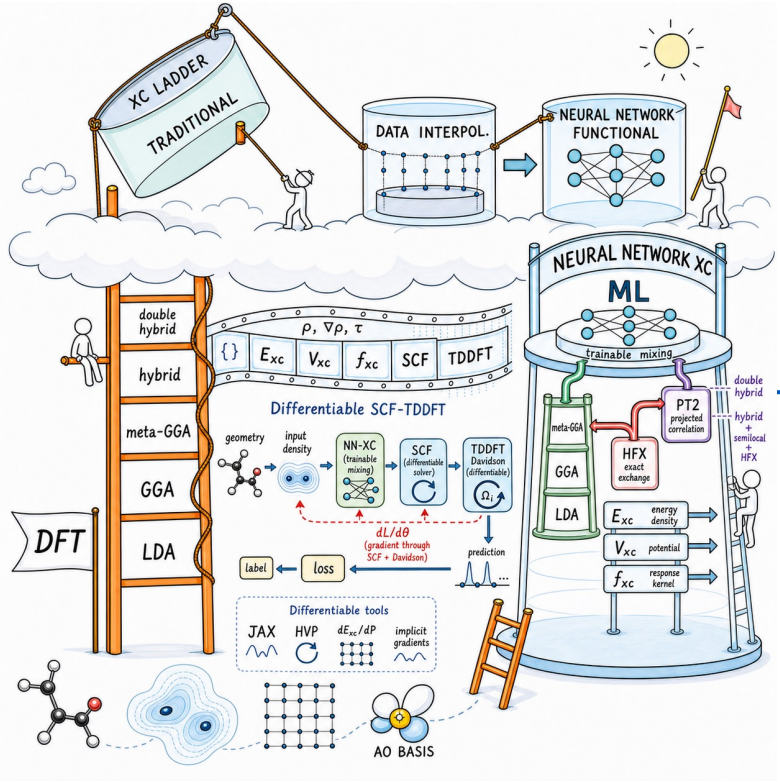
- Outlook
- Reward and Dataset
- iGKS for Orbital Dependent XC Functional
- Agentic Discovery of Self-Consistent XC Functional



■ From GradTDDFT to GradSCF – Background

A shared HF/DFT foundation, differentiable integrals and a broader method space

GradTDDFT



Reusable foundation

Reuse HF / KS-DFT

SCF, XC and TD response

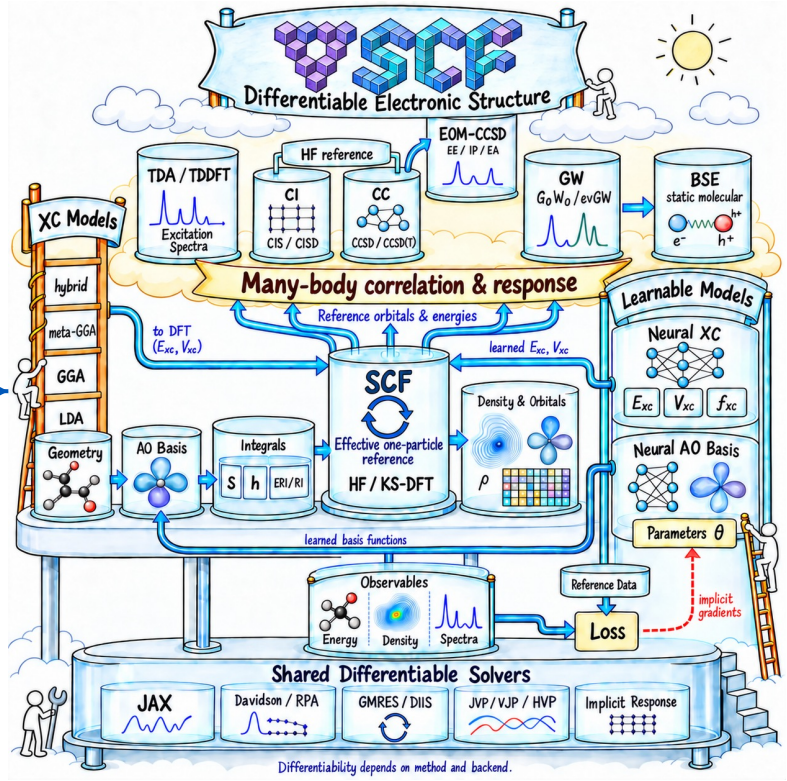
Develop integral + AD layer

One- and two-electron inputs
Coordinate / basis JVP / VJP

Unify numerical solvers

Linear / eigen / nonlinear
Shared implicit-response rules

GradSCF



Extend beyond HF/DFT: MPn · CI/FCI · CC · EOM-CC · TDHF/TDDFT · GW/BSE

The same integral and response infrastructure connects energies, spectra, forces and learnable models.

Gaussian kernels: libcint; integral assembly and AD rules: GradSCF. Derivative coverage is method specific.

■ From GradTDDFT to GradSCF – Core Modules of GradSCF

A modular JAX platform for electronic structure, response and learning.

Inputs and integrals
gto / integrals / ao2mo

Geometry, AO integrals, grids and AO–MO blocks.

Ground states and DFT
scf / dft / ofdft/ iks

HF / KS, orbital-free DFT and custom XC functionals.

Wavefunction correlation
mp / ci / fci / cc

MPn, CI / FCI, CC / QCI and perturbative corrections.

Excited states and quasiparticles
tdscf / cc.eom / gw / bse

TDHF / TDDFT, EOM-CC, GW and BSE optical response.

Shared numerical solvers
solvers

Linear, eigen and nonlinear solves; implicit JVP / VJP.

Response and properties
dfpt / scf.forces / tools.spectra

Density / field response, forces and higher derivatives.

Periodic systems(Under Testing)
pbc

Cells, k points, GTH / FFT, bands and phonon workflows.

Learnable physical models
model.neural_xc / nnao / neural_d

Neural XC, neural Gaussian bases and dispersion models.

Shared JAX / AD infrastructure connects these modules; derivative coverage follows each method.

■ From GradTDDFT to GradSCF – Workflow for New Methods

Define the physics once; reuse differentiable integrals, numerical solvers and response.

Inputs: geometry / basis / external fields / method parameters

SHARED INFRASTRUCTURE

Integral input

libcint + native AD

$S,\ h,\ (\mu\nu|\kappa\lambda)$

DF / AO2MO blocks

Coordinate / basis JVPs and VJPs

→

METHOD DEFINITION

New method

Spaces and tensor structure

Wavefunction Ansatz

Hamiltonian / kernel
Energy and correction theory

$R(x, I; \theta),\quad Av$

→

SHARED INFRASTRUCTURE

Shared solvers

Forward
Linear / eigen / nonlinear
Converged state / roots

Backward

Implicit JVP / VJP
Adjoint / transpose solve

→

MODEL + RESPONSE

Properties

Energy and corrections

Densities / observables

Forces / field response

Vibrations / spectra

$O(x^\star, I(\theta), \theta)$

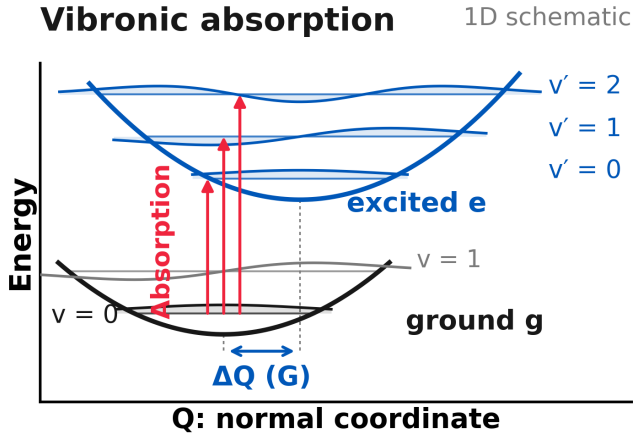
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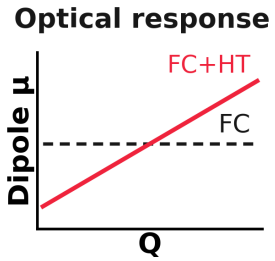
Backward: property → implicit solve → method expressions → integral parameters

x: method variables; I: integral data; θ: physical / model parameters. AD / response coverage is method specific.

■ From GradTDDFT to GradSCF – Automatic Differentiation for Spectroscopy

Benzene CIS: vibronic structure and HT intensity borrowing





K: dipole slope

FC: overlap
HT: dipole variation

Calculation workflow

RHF optimization + ground-state Hessian
30 mass-weighted internal modes

↓

CIS Davidson → 20 singlet states
Select 5 complete groups; keep S3/S4 together

↓

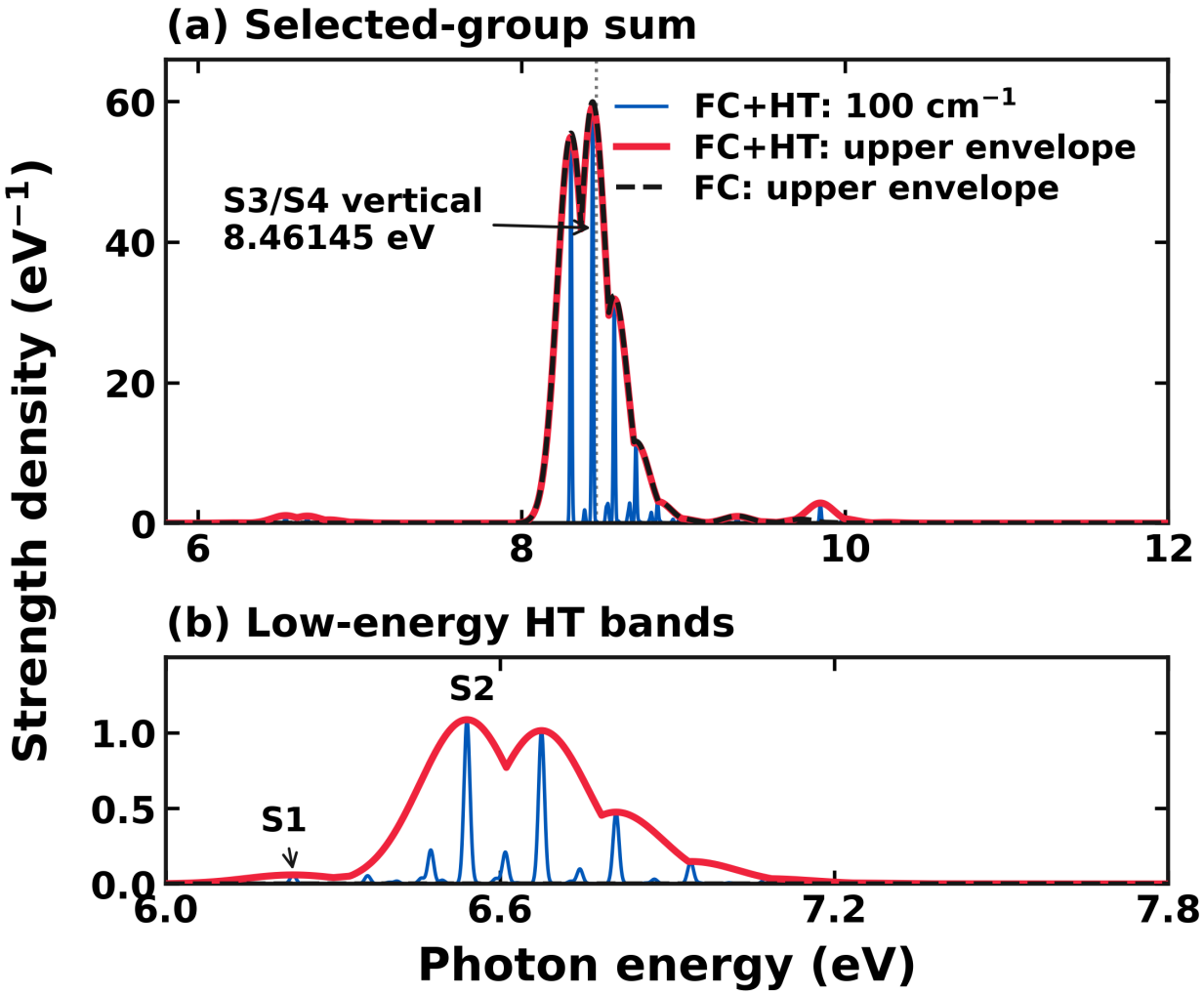
AD of Hamiltonian and transition dipoles
Integral → SCF → CIS-subspace nuclear response

↓

Linear vibronic model → FC / HT spectrum
Sum groups; retain FC–HT interference

0 K; equal curvature; no Duschinsky mixing.

CIS / Cartesian def2-SVP · 30 modes



S1, S2, S3/S4, S7, S11 only (6 states).
Upper envelopes guide peak heights; their areas are not oscillator strengths.

■ From GradTDDFT to GradSCF –Outlook

- **Outlook**

  - **Integral Module for Differential Input**
  - **Large Systems and High Efficient**
  - **Multireference Self-Consistent Methods**

GradSCF Tutorials

From public method calls to property derivatives, method development, and machine learning.

Start with GradSCF, computational graphs, AD, and JAX foundations, then follow three core parts and a machine-learning part. Every method follows **theory → computational graph and AD formulation → high-level API → results and validation**.

Choose an entry point

- Introducing GradSCF: scope, features, and learning paths.
- First calculation: common environment and minimal HF.
- Electronic-structure methods: isolated/periodic methods and variants.
- Properties: optimization, vibrations, spectra, response, phonons.
- Develop a method: equations, infrastructure, solvers, backward.
- Develop post-processing: frozen partial versus end-to-end total derivatives.
- Machine learning: model use, training, and parameter gradients.
- Work through an Agent: task routing, templates, and evidence.

- **Functional**

  - **Basic Electronic Structure Calculation for Single-Reference Methods**
  - **Full Differentiable Electronic Structure with Public Solver**
  - **Ground State, Excited State Spectrum with Automatic Differentiation**
  - **Neural Network Training for Electronic Structure Situation**

