Can we improve energy density functionals?
—A perturbative method—

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Density Functional Theory (DFT) and Energy Density Functional (EDF)

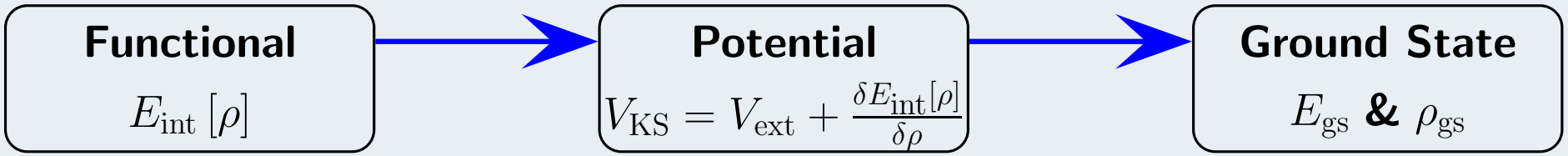
- Hohenberg-Kohn thm. guarantees existence of the unique EDF $E_{\rm int}$
- Kohn-Sham scheme provides a practical method
- Kohn-Sham orbital φ_j obays

$$\left[-\frac{\hbar^2}{2M}\Delta + V_{\rm KS}(\boldsymbol{r})\right]\varphi_j(\boldsymbol{r}) = \varepsilon_j\varphi_j(\boldsymbol{r})$$

$$V_{\rm KS}(\boldsymbol{r}) = V_{\rm ext}(\boldsymbol{r}) + \left.\frac{\delta E_{\rm int}[\rho]}{\delta\rho}\right|_{\rho=\rho_{\rm gs}(\boldsymbol{r})} \qquad \rho_{\rm gs}(\boldsymbol{r}) = \sum\left|\varphi_j(\boldsymbol{r})\right|^2$$

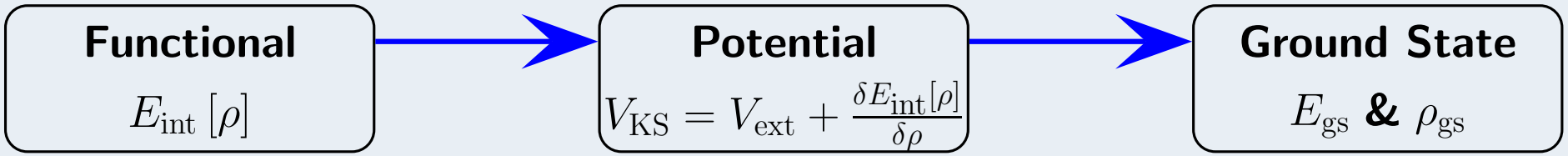
- If the exact $E_{\rm int}$ were known, DFT would provide the exact $\rho_{\rm gs}$ and $E_{\rm gs}$
  $\rightarrow$ Practically, approximation is needed to derive $E_{\rm int}$
- Accuracy of $E_{\rm int}$ governs accuracy of DFT calc.

Usual DFT Calculation



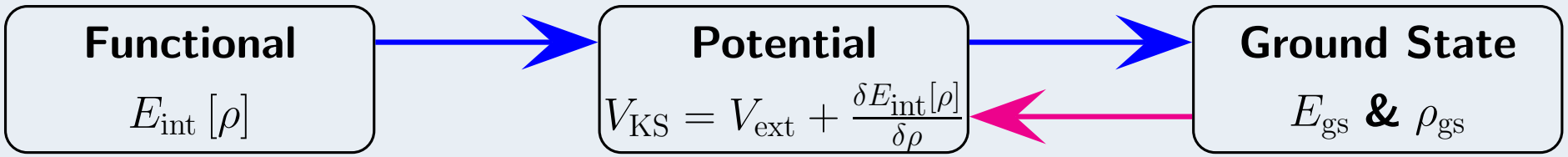
Introduction

Usual DFT Calculation



Q1. If we know ρ_{gs}, can we derive V_{KS}?

Inverse Kohn-Sham Method



Q1. If we know $\rho_{\rm gs}$, can we derive $V_{\rm KS}$?

A1. Yes! Inverse Kohn-Sham Method

Inverse Kohn-Sham Method

- Assume that $\rho_{\rm gs}$ of a system (e.g., ^{208}Pb) is known
- $V_{\rm KS}$ for the system (e.g., ^{208}Pb) reads

$$V_{\rm KS}(\boldsymbol{r}) = \frac{\sum \varphi_j^*(\boldsymbol{r}) \left(\varepsilon_j + \frac{\hbar^2}{2M}\Delta\right) \varphi_j(\boldsymbol{r})}{\rho_{\rm gs}(\boldsymbol{r})}$$

Practically, $V_{\rm KS}$ is derived iteratively

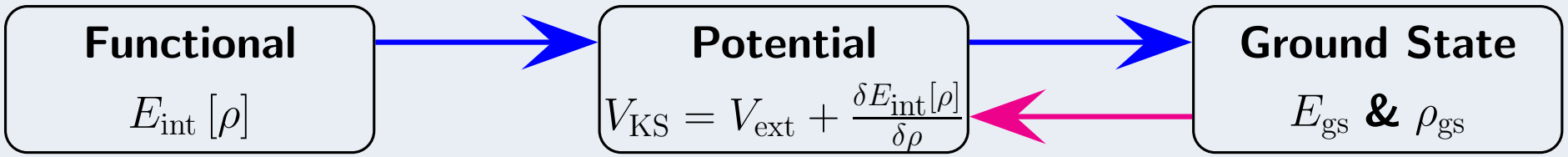
$$V_{\rm KS}^n(\boldsymbol{r}) = \frac{\sum \varphi_j^{n*}(\boldsymbol{r}) \left(\varepsilon_j^n + \frac{\hbar^2}{2M}\Delta\right) \varphi_j^n(\boldsymbol{r})}{\rho_{\rm gs}(\boldsymbol{r})} = \frac{\rho^n(\boldsymbol{r})}{\rho_{\rm gs}(\boldsymbol{r})} V_{\rm KS}^{n-1}(\boldsymbol{r})$$

$$\left[-\frac{\hbar^2}{2M}\Delta + V_{\rm KS}^{n-1}(\boldsymbol{r})\right] \varphi_j^n(\boldsymbol{r}) = \varepsilon_j^n \varphi_j^n(\boldsymbol{r}) \qquad \rho^n(\boldsymbol{r}) = \sum \left|\varphi_j^n(\boldsymbol{r})\right|^2$$

- This potential $V_{\rm KS}$ can only be used for the system (e.g., ^{208}Pb)
  $\rightarrow$ This $V_{\rm KS}$ cannot be used directly for the other systems (e.g., ^{48}Ca)

Wang and Parr. *Phys. Rev. A* **47**, R1591 (1993)

Inverse Kohn-Sham Method



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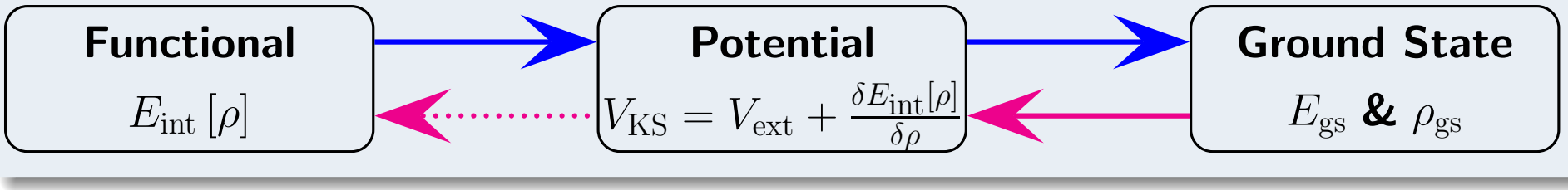
Inverse Kohn-Sham Method



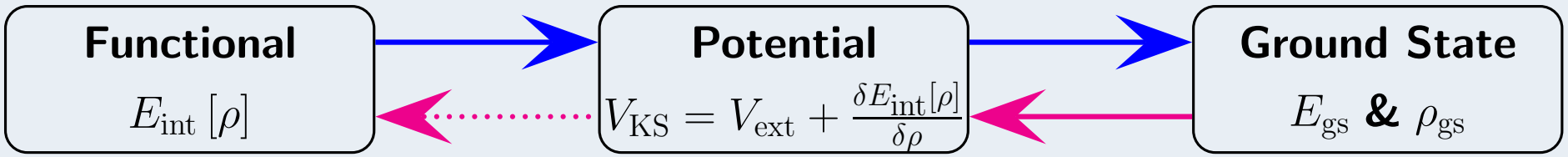
Q1. If we know $\rho_{\rm gs}$, can we derive $V_{\rm KS}$?

A1. Yes! Inverse Kohn-Sham Method

Q2. Can we derive $E_{\rm int}$ from $V_{\rm KS}$?

Introduction

Inverse Kohn-Sham Method



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A1. Yes! Inverse Kohn-Sham Method

Q2. Can we derive $E_{\rm int}$ from $V_{\rm KS}$?

A2. Today's Topic!! "IKS-DFPT"

Framework of IKS-DFPT

Brief Idea of IKS-DFPT

- DFT calculation is succeeded
  $\rightarrow$ We assume that a "known" EDF $\tilde{E}_{\mathrm{int}}$ is good enough
- Improve $\tilde{E}_{\mathrm{int}}$ using some "exact" G.S. densities
- Difference between $\tilde{E}_{\mathrm{int}}$ & $E_{\mathrm{int}}^{\mathrm{exact}}$ is treated as 1st order perturbation
- Idea of **Density Functional Perturbation Theory (DFPT)** is used
  Original DFPT is usually used for phonon calculation for solid
  with considering pertubation for V_{ext}
- E_{gs} is formally derived by two methods
  - Inverse Kohn-Sham method
  - Density functional perturbation theory
- Using the fact that two E_{gs} should be identical, $E_{\mathrm{int}}^{\mathrm{exact}} - \tilde{E}_{\mathrm{int}}$ is derived

Naito, Ohashi, and Liang. *J. Phys. B* **52**, 245003 (2019)

Perturbation Expansion for E_{int}, ρ, φ_j

$$E_{\mathrm{int}}^{\mathrm{exact}}[\rho] = \tilde{E}_{\mathrm{int}}[\rho] + \lambda E_{\mathrm{int}}^{(1)}[\rho] + O\left(\lambda^2\right)$$
$$V_{\mathrm{ext}}^{\mathrm{exact}}(\boldsymbol{r}) = \tilde{V}_{\mathrm{ext}}(\boldsymbol{r})$$
$$\rho_{\mathrm{gs}}^{\mathrm{exact}}(\boldsymbol{r}) = \tilde{\rho}_{\mathrm{gs}}(\boldsymbol{r}) + \lambda \rho_{\mathrm{gs}}^{(1)}(\boldsymbol{r}) + O\left(\lambda^2\right)$$
$$\varphi_j^{\mathrm{exact}}(\boldsymbol{r}) = \tilde{\varphi}_j(\boldsymbol{r}) + \lambda \varphi_j^{(1)}(\boldsymbol{r}) + O\left(\lambda^2\right)$$

Note: $\rho_{\mathrm{gs}}^{\mathrm{exact}}$ is known

Assumption: Orthogonal Condition

$$\int \tilde{\varphi}_j^*(\boldsymbol{r})\, \varphi_j^{(1)}(\boldsymbol{r})\, d\boldsymbol{r} = 0$$

Density

$$\tilde{\rho}_{\mathrm{gs}}(\boldsymbol{r}) = \sum \left|\tilde{\varphi}_j(\boldsymbol{r})\right|^2$$
$$\rho_{\mathrm{gs}}^{(1)}(\boldsymbol{r}) = \sum \left[\varphi_j^{(1)*}(\boldsymbol{r})\, \tilde{\varphi}_j(\boldsymbol{r}) + \tilde{\varphi}_j^*(\boldsymbol{r})\, \varphi_j^{(1)}(\boldsymbol{r})\right]$$

Ground-State Energy from IKS

$$E_{\mathrm{gs}}^{\mathrm{exact}} = \sum \varepsilon_j^{\mathrm{exact}} + E_{\mathrm{int}}^{\mathrm{exact}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right] - \int \frac{\delta E_{\mathrm{int}}^{\mathrm{exact}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right]}{\delta\rho\left(\boldsymbol{r}\right)} \rho_{\mathrm{gs}}^{\mathrm{exact}}\left(\boldsymbol{r}\right)\, d\boldsymbol{r}$$

$$=$$

$\varepsilon_j^{\mathrm{exact}}$ can be derived by inverse Kohn-Sham method

Ground-State Energy from IKS

$$E_{\mathrm{gs}}^{\mathrm{exact}} = \sum \varepsilon_j^{\mathrm{exact}} + E_{\mathrm{int}}^{\mathrm{exact}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right] - \int \frac{\delta E_{\mathrm{int}}^{\mathrm{exact}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right]}{\delta\rho\left(\boldsymbol{r}\right)} \rho_{\mathrm{gs}}^{\mathrm{exact}}\left(\boldsymbol{r}\right)\, d\boldsymbol{r}$$

$$= \sum \varepsilon_j^{\mathrm{exact}} + \tilde{E}_{\mathrm{int}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right] + \lambda E_{\mathrm{int}}^{(1)}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right]$$

$$- \int \frac{\delta \tilde{E}_{\mathrm{int}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right]}{\delta\rho\left(\boldsymbol{r}\right)} \rho_{\mathrm{gs}}^{\mathrm{exact}}\left(\boldsymbol{r}\right)\, d\boldsymbol{r} - \lambda \int \frac{\delta E_{\mathrm{int}}^{(1)}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right]}{\delta\rho\left(\boldsymbol{r}\right)} \rho_{\mathrm{gs}}^{\mathrm{exact}}\left(\boldsymbol{r}\right)\, d\boldsymbol{r} + O\left(\lambda^2\right)$$

$\varepsilon_j^{\mathrm{exact}}$ can be derived by inverse Kohn-Sham method

Ground-State Energy from DFPT

$$E_{\mathrm{gs}}^{\mathrm{exact}} = \tilde{E}_{\mathrm{gs}} + \lambda \left. \frac{dE_{\mathrm{int}}^{\mathrm{exact}}\left[\tilde{\rho}_{\mathrm{gs}}\right]}{d\lambda} \right|_{\lambda=0} + O\left(\lambda^2\right)$$

$$=$$

$$=$$

Please see our paper for the deviation of this equation, since it is complicated

Ground-State Energy from DFPT

$$E_{\mathrm{gs}}^{\mathrm{exact}} = \tilde{E}_{\mathrm{gs}} + \lambda \left. \frac{dE_{\mathrm{int}}^{\mathrm{exact}}\left[\tilde{\rho}_{\mathrm{gs}}\right]}{d\lambda} \right|_{\lambda=0} + O\left(\lambda^2\right)$$

$$= \tilde{E}_{\mathrm{gs}} + \lambda E_{\mathrm{int}}^{(1)}\left[\tilde{\rho}_{\mathrm{gs}}\right] + O\left(\lambda^2\right)$$

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Ground-State Energy from DFPT

$$E_{\mathrm{gs}}^{\mathrm{exact}} = \tilde{E}_{\mathrm{gs}} + \lambda \left. \frac{dE_{\mathrm{int}}^{\mathrm{exact}}\left[\tilde{\rho}_{\mathrm{gs}}\right]}{d\lambda} \right|_{\lambda=0} + O\left(\lambda^2\right)$$

$$= \tilde{E}_{\mathrm{gs}} + \lambda E_{\mathrm{int}}^{(1)}\left[\tilde{\rho}_{\mathrm{gs}}\right] + O\left(\lambda^2\right)$$

$$= \sum \tilde{\varepsilon}_j + \tilde{E}_{\mathrm{int}}\left[\tilde{\rho}_{\mathrm{gs}}\right] - \int \frac{\delta \tilde{E}_{\mathrm{int}}\left[\tilde{\rho}_{\mathrm{gs}}\right]}{\delta \rho\left(\boldsymbol{r}\right)} \tilde{\rho}_{\mathrm{gs}}\left(\boldsymbol{r}\right) d\boldsymbol{r} + \lambda E_{\mathrm{int}}^{(1)}\left[\tilde{\rho}_{\mathrm{gs}}\right] + O\left(\lambda^2\right)$$

Please see our paper for the deviation of this equation, since it is complicated

Equation for IKS-DFPT

Compare two expression and neglect $O\left(\lambda^2\right)$ term

$$\tilde{E}_{\mathrm{gs}} + \lambda E^{(1)}_{\mathrm{int}}\left[\tilde{\rho}_{\mathrm{gs}}\right] \simeq \sum \varepsilon_j^{\mathrm{exact}} + \tilde{E}_{\mathrm{int}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right] + \lambda E^{(1)}_{\mathrm{int}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right] - \int \frac{\delta \tilde{E}_{\mathrm{int}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right]}{\delta\rho\left(\boldsymbol{r}\right)} \rho_{\mathrm{gs}}^{\mathrm{exact}}\left(\boldsymbol{r}\right) d\boldsymbol{r} - \lambda \int \frac{\delta E^{(1)}_{\mathrm{int}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right]}{\delta\rho\left(\boldsymbol{r}\right)} \rho_{\mathrm{gs}}^{\mathrm{exact}}\left(\boldsymbol{r}\right) d\boldsymbol{r}$$

Therefore, We Should Solve

$$\lambda E^{(1)}_{\mathrm{int}}\left[\tilde{\rho}_{\mathrm{gs}}\right] - \lambda E^{(1)}_{\mathrm{int}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right] + \lambda \int \frac{\delta E^{(1)}_{\mathrm{int}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right]}{\delta\rho\left(\boldsymbol{r}\right)} \rho_{\mathrm{gs}}^{\mathrm{exact}}\left(\boldsymbol{r}\right) d\boldsymbol{r}$$

$$\simeq \sum \varepsilon_j^{\mathrm{exact}} + \tilde{E}_{\mathrm{int}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right] - \int \frac{\delta \tilde{E}_{\mathrm{int}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right]}{\delta\rho\left(\boldsymbol{r}\right)} \rho_{\mathrm{gs}}^{\mathrm{exact}}\left(\boldsymbol{r}\right) d\boldsymbol{r} - \tilde{E}_{\mathrm{gs}}$$

Equation for IKS-DFPT

Compare two expression and neglect $O\left(\lambda^2\right)$ term

$$\tilde{E}_{\mathrm{gs}} + \lambda E^{(1)}_{\mathrm{int}}\left[\tilde{\rho}_{\mathrm{gs}}\right] \simeq \sum \varepsilon_j^{\mathrm{exact}} + \tilde{E}_{\mathrm{int}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right] + \lambda E^{(1)}_{\mathrm{int}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right] - \int \frac{\delta \tilde{E}_{\mathrm{int}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right]}{\delta\rho\left(\boldsymbol{r}\right)} \rho_{\mathrm{gs}}^{\mathrm{exact}}\left(\boldsymbol{r}\right) d\boldsymbol{r} - \lambda \int \frac{\delta E^{(1)}_{\mathrm{int}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right]}{\delta\rho\left(\boldsymbol{r}\right)} \rho_{\mathrm{gs}}^{\mathrm{exact}}\left(\boldsymbol{r}\right) d\boldsymbol{r}$$

Therefore, We Should Solve

$$\lambda E^{(1)}_{\mathrm{int}}\left[\tilde{\rho}_{\mathrm{gs}}\right] - \lambda E^{(1)}_{\mathrm{int}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right] + \lambda \int \frac{\delta E^{(1)}_{\mathrm{int}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right]}{\delta\rho\left(\boldsymbol{r}\right)} \rho_{\mathrm{gs}}^{\mathrm{exact}}\left(\boldsymbol{r}\right) d\boldsymbol{r}$$

$$\simeq \sum \varepsilon_j^{\mathrm{exact}} + \tilde{E}_{\mathrm{int}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right] - \int \frac{\delta \tilde{E}_{\mathrm{int}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right]}{\delta\rho\left(\boldsymbol{r}\right)} \rho_{\mathrm{gs}}^{\mathrm{exact}}\left(\boldsymbol{r}\right) d\boldsymbol{r} - \tilde{E}_{\mathrm{gs}} = C\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right]$$

Right-hand side can be calculated exactly as $C\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right]$ (constant)

Framework of IKS-DFPT

Our Task

Deribe $\lambda E_{\mathrm{int}}^{(1)}$ from

$$\lambda E_{\mathrm{int}}^{(1)}\left[\tilde{\rho}_{\mathrm{gs}}^{\mathrm{exact}}\right]-\lambda E_{\mathrm{int}}^{(1)}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right]+\lambda\int\frac{\delta E_{\mathrm{int}}^{(1)}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right]}{\delta\rho\left(\boldsymbol{r}\right)}\rho_{\mathrm{gs}}^{\mathrm{exact}}\left(\boldsymbol{r}\right)\,d\boldsymbol{r}=C\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right]$$

This equation is a functional equation: Difficult to solve!!

How to Solve It?

Framework of IKS-DFPT

Our Task

Deribe $\lambda E_{\mathrm{int}}^{(1)}$ from

$$\lambda E_{\mathrm{int}}^{(1)}\left[\tilde{\rho}_{\mathrm{gs}}^{\mathrm{exact}}\right] - \lambda E_{\mathrm{int}}^{(1)}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right] + \lambda \int \frac{\delta E_{\mathrm{int}}^{(1)}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right]}{\delta\rho\left(\boldsymbol{r}\right)} \rho_{\mathrm{gs}}^{\mathrm{exact}}\left(\boldsymbol{r}\right) d\boldsymbol{r} = C\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right]$$

This equation is a functional equation: Difficult to solve!!

How to Solve It?

1. Assume the form of $\lambda E_{\mathrm{int}}^{(1)}$
2. If $\lambda E_{\mathrm{int}}^{(1)}$ has n parameters,
   substitute ρ_{gs} and ε_j of n kind of systems to the equation
3. Solve simultaniously

Benchmark Calculation (1): Atoms

Assumption for $\lambda E_{\mathrm{int}}^{(1)}$

$$\lambda E_{\mathrm{int}}^{(1)}[\rho] = \lambda A \int [\rho(\boldsymbol{r})]^{\alpha} \, d\boldsymbol{r}$$

Unknown parameters: λA and α

Equation for IKS-DFPT1

$$\lambda A \int \left\{ \left[\tilde{\rho}_{\mathrm{gs}}(\boldsymbol{r}) \right]^{\alpha} + (\alpha - 1) \left[\rho_{\mathrm{gs}}^{\mathrm{exact}}(\boldsymbol{r}) \right]^{\alpha} \right\} d\boldsymbol{r} = C\left[\rho_{\mathrm{gs}}\right]$$

To derive λA and α, two densities are required

Iterative IKS-DFPT: Calculated Functional at n-th Step

$$E_{\mathrm{int}}^{n\text{-th}}[\rho] = \tilde{E}_{\mathrm{int}}[\rho] + \sum_{j=1}^{n} \lambda A_j \int [\rho(\boldsymbol{r})]^{\alpha_j} \, d\boldsymbol{r}$$

EDF for Electronic Systems

$$E_{\mathrm{int}}[\rho] = E_{\mathrm{H}}[\rho] + E_{\mathrm{x}}[\rho] + E_{\mathrm{c}}[\rho]$$

E_{H} Coulomb Hartree EDF

E_{x} Coulomb exchange EDF (LDA is used in this work)

E_{c} Coulomb correlation EDF (LDA is used in this work)

Setup

- Noble gas atoms are used for ρ_{gs}
- Two patterns are tested (Today I only show pattern 1)

Pattern 1

Known $\tilde{E}_{\mathrm{int}}$ E_{H}

"Exact" E_{int} $E_{\mathrm{H}} + E_{\mathrm{x}}$

Can we reproduce E_{x}?

Pattern 2

Known $\tilde{E}_{\mathrm{int}}$ $E_{\mathrm{H}} + E_{\mathrm{x}}$

"Exact" E_{int} $E_{\mathrm{H}} + E_{\mathrm{x}} + E_{\mathrm{c}}$

Can we reproduce E_{c}?

Benchmark Calculation (1): Atoms

Energy: $E_{\mathrm{int}}^{\mathrm{exact}}$ is Hartree & LDA exchange (Hartree atomic unit)

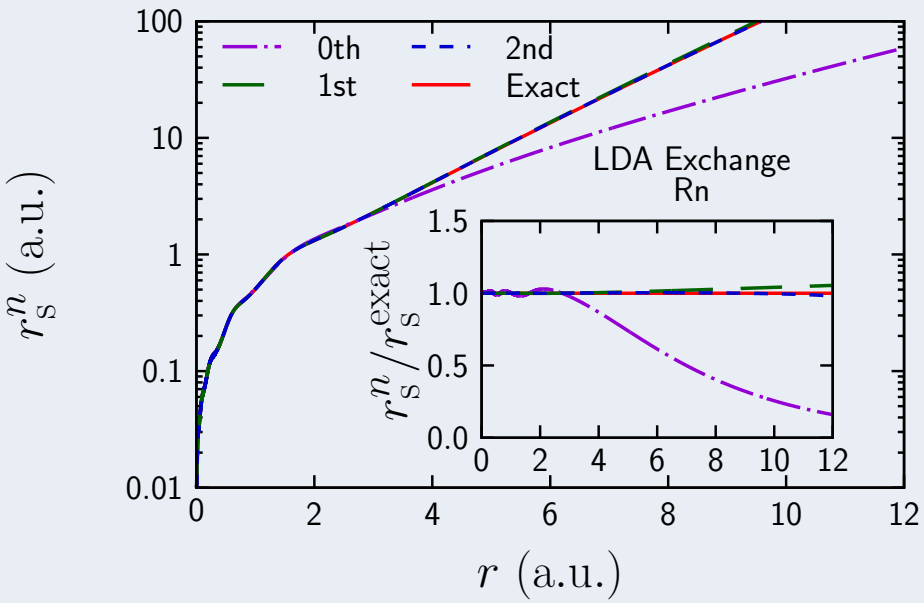
n	α_n	λA_n	$E_{\mathrm{gs}}^{n\text{-th}}$ of Xe	$E_{\mathrm{gs}}^{n\text{-th}}$ of Rn
0			−7054.6485	−21479.344
1	1.3311445	−0.7558229	−7224.9365	−21852.010
2	1.0436323	0.0306234	−7223.0601	−21848.894
Target	1.3333333	−0.7385588	−7223.1853	−21848.954

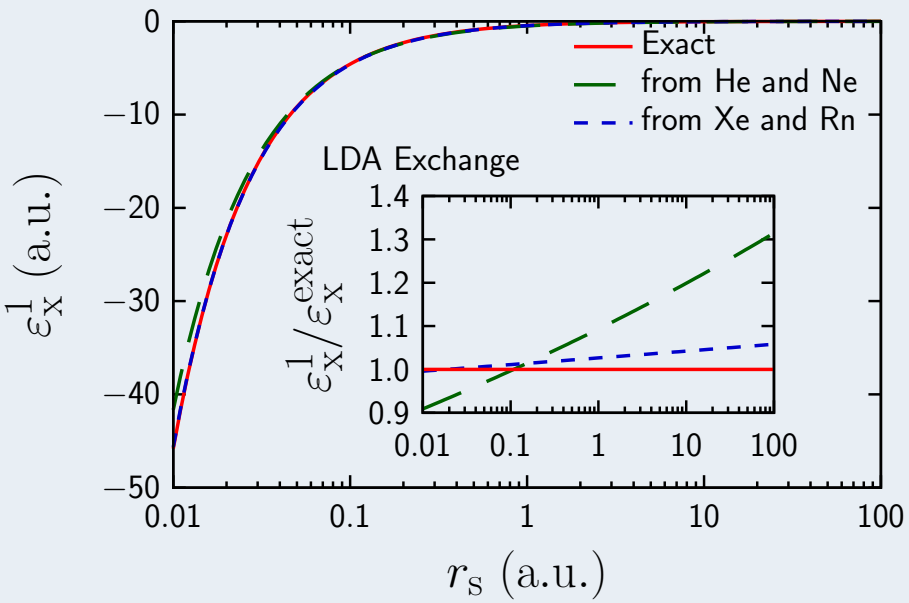
Hartree Atomic Unit

$$m_e = 1, \qquad \hbar = 1, \qquad e^2 = 1, \qquad 4\pi\varepsilon_0 = 1, \qquad c = \frac{1}{\alpha}$$

Benchmark Calculation (1): Atoms

Density and energy density: $E_{\mathrm{int}}^{\mathrm{exact}}$ is Hartree & LDA exchange





$$r_{\mathrm{s}} = \left(\frac{3}{4\pi\rho}\right)^{1/3}, \qquad E_{\mathrm{x}}[\rho] = \int \varepsilon_{\mathrm{x}}^{1}(r_{\mathrm{s}}(\boldsymbol{r}))\,\rho(\boldsymbol{r})\,d\boldsymbol{r}$$

Relativistic Extension

- There are scalar KS potential S_{KS} and vector KS potential V_{KS}
- There are scalar density ρ_{S} and vector density ρ_{V}

$$\rho_{\mathrm{S}}(\boldsymbol{r}) = \sum \varphi_j^{\dagger}(\boldsymbol{r}) \beta \varphi_j(\boldsymbol{r}) = \sum \overline{\varphi}_j(\boldsymbol{r}) \varphi_j(\boldsymbol{r}) \qquad \rho_{\mathrm{V}}(\boldsymbol{r}) = \sum \varphi_j^{\dagger}(\boldsymbol{r}) \varphi_j(\boldsymbol{r})$$

- S.P. orbital obeys $\left[\boldsymbol{\alpha} \cdot \boldsymbol{p} + \beta\left(m + S_{\mathrm{KS}}(\boldsymbol{r})\right) + V_{\mathrm{KS}}(\boldsymbol{r})\right] \varphi_j(\boldsymbol{r}) = \varepsilon_j(\boldsymbol{r})$

Non-Relativistic Inverse Kohn-Sham Method

$$V_{\mathrm{KS}}(\boldsymbol{r}) = -\frac{1}{\rho(\boldsymbol{r})} \sum \varphi_j^{*}(\boldsymbol{r}) \left(-\frac{\hbar^2}{2M} \Delta - \varepsilon_j\right) \varphi_j(\boldsymbol{r})$$

Wang and Parr. *Phys. Rev. A* **47**, R1591 (1993)

Relativistic Inverse Kohn-Sham Method

$$V_{\mathrm{KS}}(\boldsymbol{r}) + S_{\mathrm{KS}}(\boldsymbol{r}) = -M - \frac{1}{\rho_{\mathrm{V}}(\boldsymbol{r}) + \rho_{\mathrm{S}}(\boldsymbol{r})} \sum \left[\varphi_j^{\dagger}(\boldsymbol{r}) + \overline{\varphi}_j(\boldsymbol{r})\right] \left(\boldsymbol{\alpha} \cdot \boldsymbol{p} - \varepsilon_j\right) \varphi_j(\boldsymbol{r})$$

$$V_{\mathrm{KS}}(\boldsymbol{r}) - S_{\mathrm{KS}}(\boldsymbol{r}) = +M - \frac{1}{\rho_{\mathrm{V}}(\boldsymbol{r}) - \rho_{\mathrm{S}}(\boldsymbol{r})} \sum \left[\varphi_j^{\dagger}(\boldsymbol{r}) - \overline{\varphi}_j(\boldsymbol{r})\right] \left(\boldsymbol{\alpha} \cdot \boldsymbol{p} - \varepsilon_j\right) \varphi_j(\boldsymbol{r})$$

Accorto, Naito, Liang, Nikšić, and Vretenar. *Phys. Rev. C* **103**, 044304 (2021)

Non-Relativistic IKS-DFPT

$$\lambda E_{\mathrm{int}}^{(1)}\left[\tilde{\rho}_{\mathrm{gs}}\right] - \lambda E_{\mathrm{int}}^{(1)}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right] + \lambda \int \frac{\delta E_{\mathrm{int}}^{(1)}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right]}{\delta\rho\left(\boldsymbol{r}\right)} \rho_{\mathrm{gs}}^{\mathrm{exact}}\left(\boldsymbol{r}\right)\, d\boldsymbol{r}$$

$$\simeq \sum \varepsilon_j^{\mathrm{exact}} - \tilde{E}_{\mathrm{gs}} + \tilde{E}_{\mathrm{int}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right] - \int \frac{\delta \tilde{E}_{\mathrm{int}}\left[\rho_{\mathrm{gs}}^{\mathrm{exact}}\right]}{\delta\rho\left(\boldsymbol{r}\right)} \rho_{\mathrm{gs}}^{\mathrm{exact}}\left(\boldsymbol{r}\right)\, d\boldsymbol{r}$$

Naito, Ohashi, and Liang. *J. Phys. B* **52**, 245003 (2019)

Relativistic IKS-DFPT

$$\lambda E_{\mathrm{int}}^{(1)}\left[\tilde{\rho}_{\mathrm{V,\,gs}}, \tilde{\rho}_{\mathrm{S,\,gs}}\right] - \lambda E_{\mathrm{int}}^{(1)}\left[\rho_{\mathrm{V,\,gs}}^{\mathrm{exact}}, \rho_{\mathrm{S,\,gs}}^{\mathrm{exact}}\right]$$

$$+ \lambda \int \frac{\delta E_{\mathrm{int}}^{(1)}\left[\rho_{\mathrm{V,\,gs}}^{\mathrm{exact}}, \rho_{\mathrm{S,\,gs}}\right]}{\delta\rho_{\mathrm{V}}\left(\boldsymbol{r}\right)} \rho_{\mathrm{V,\,gs}}^{\mathrm{exact}}\left(\boldsymbol{r}\right)\, d\boldsymbol{r} + \lambda \int \frac{\delta E_{\mathrm{int}}^{(1)}\left[\rho_{\mathrm{V,\,gs}}^{\mathrm{exact}}, \rho_{\mathrm{S,\,gs}}\right]}{\delta\rho_{\mathrm{S}}\left(\boldsymbol{r}\right)} \rho_{\mathrm{S,\,gs}}^{\mathrm{exact}}\left(\boldsymbol{r}\right)\, d\boldsymbol{r}$$

$$\simeq \sum \varepsilon_j^{\mathrm{exact}} - \tilde{E}_{\mathrm{gs}} + \tilde{E}_{\mathrm{int}}\left[\rho_{\mathrm{V,\,gs}}^{\mathrm{exact}}, \rho_{\mathrm{S,\,gs}}^{\mathrm{exact}}\right]$$

$$- \int \frac{\delta \tilde{E}_{\mathrm{int}}\left[\rho_{\mathrm{V,\,gs}}^{\mathrm{exact}}, \rho_{\mathrm{S,\,gs}}^{\mathrm{exact}}\right]}{\delta\rho_{\mathrm{V}}\left(\boldsymbol{r}\right)} \rho_{\mathrm{V,\,gs}}^{\mathrm{exact}}\left(\boldsymbol{r}\right)\, d\boldsymbol{r} - \int \frac{\delta \tilde{E}_{\mathrm{int}}\left[\rho_{\mathrm{V,\,gs}}^{\mathrm{exact}}, \rho_{\mathrm{S,\,gs}}^{\mathrm{exact}}\right]}{\delta\rho_{\mathrm{S}}\left(\boldsymbol{r}\right)} \rho_{\mathrm{S,\,gs}}^{\mathrm{exact}}\left(\boldsymbol{r}\right)\, d\boldsymbol{r}$$

Accorto, Naito, Liang, Nikšić, and Vretenar. *Phys. Rev. C* **103**, 044304 (2021)

DD-PC1 (without Coulomb)

$$S_{\text{DD-PC1}}\left[\rho_{\text{V}}, \rho_{\text{S}}, \vec{\rho}_{\text{TV}}\right] = \alpha_{\text{S}}\left(\rho_{\text{V}}\right)\rho_{\text{S}} + \delta_{\text{S}}\,\Delta\rho_{\text{S}}$$

$$V_{\text{DD-PC1}}\left[\rho_{\text{V}}, \rho_{\text{S}}, \vec{\rho}_{\text{TV}}\right] = \alpha_{\text{V}}\left(\rho_{\text{V}}\right)\rho_{\text{V}} + \alpha_{\text{TV}}\left(\rho_{\text{V}}\right)\vec{\rho}_{\text{TV}}$$

$$\Sigma_{\text{R}}\left[\rho_{\text{V}}, \rho_{\text{S}}, \vec{\rho}_{\text{TV}}\right] = \frac{1}{2}\frac{\partial\alpha_{\text{S}}}{\partial\rho}\rho_{\text{S}}^2 + \frac{1}{2}\frac{\partial\alpha_{\text{V}}}{\partial\rho}\rho_{\text{V}}^2 + \frac{1}{2}\frac{\partial\alpha_{\text{TV}}}{\partial\rho}\rho_{\text{TV}}^2$$

$$\alpha_{\text{S}}\left(\rho_{\text{V}}\right) = a_{\text{S}} + \left(b_{\text{S}} + c_{\text{S}}\frac{\rho_{\text{V}}}{\rho_{\text{sat}}}\right)e^{-d_{\text{S}}\rho_{\text{V}}/\rho_{\text{sat}}}$$

$$\alpha_{\text{V}}\left(\rho_{\text{V}}\right) = a_{\text{V}} + b_{\text{V}}e^{-d_{\text{V}}\rho_{\text{V}}/\rho_{\text{sat}}}$$

$$\alpha_{\text{TV}}\left(\rho_{\text{V}}\right) = b_{\text{TV}}e^{-d_{\text{TV}}\rho_{\text{V}}/\rho_{\text{sat}}}$$

Nikšić, Vretenar, and Ring. *Phys. Rev. C* **78**, 034318 (2008)

Benchmark Calculation

Start "LDA terms (only a_{S} and a_{V})" of DD-PC1

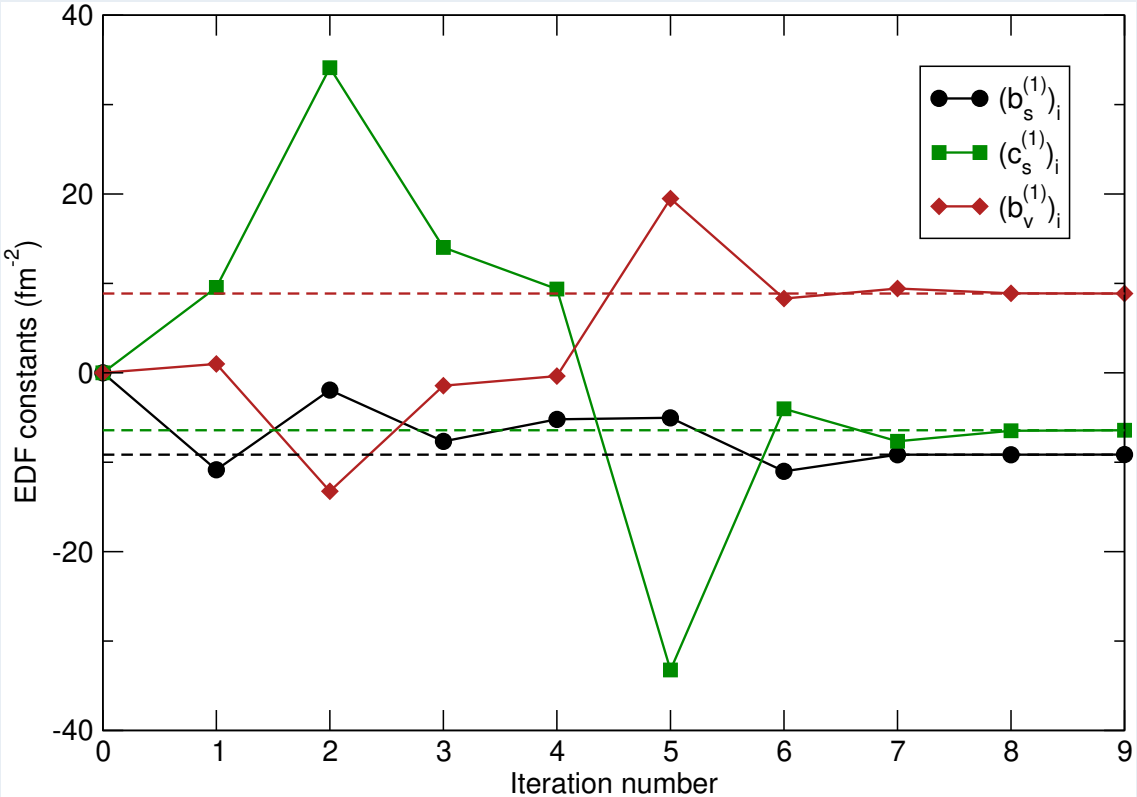
Goal All terms of DD-PC1 Can we repdoduce b and c? (d are fixed)

Systems: $N = Z$ nuclei w/o Coulomb (${}^{8}_{8}16$, ${}^{28}_{28}56$, ${}^{50}_{50}100$)

Benchmark Results

Iterative method is used

$$b_{\rm S} = \sum_k b_{\rm S}^{k\text{-th}} \qquad c_{\rm S} = \sum_k c_{\rm S}^{k\text{-th}} \qquad b_{\rm V} = \sum_k b_{\rm V}^{k\text{-th}}$$



Accorto, Naito, Liang, Nikšić, and Vretenar. *Phys. Rev. C* **103**, 044304 (2021)

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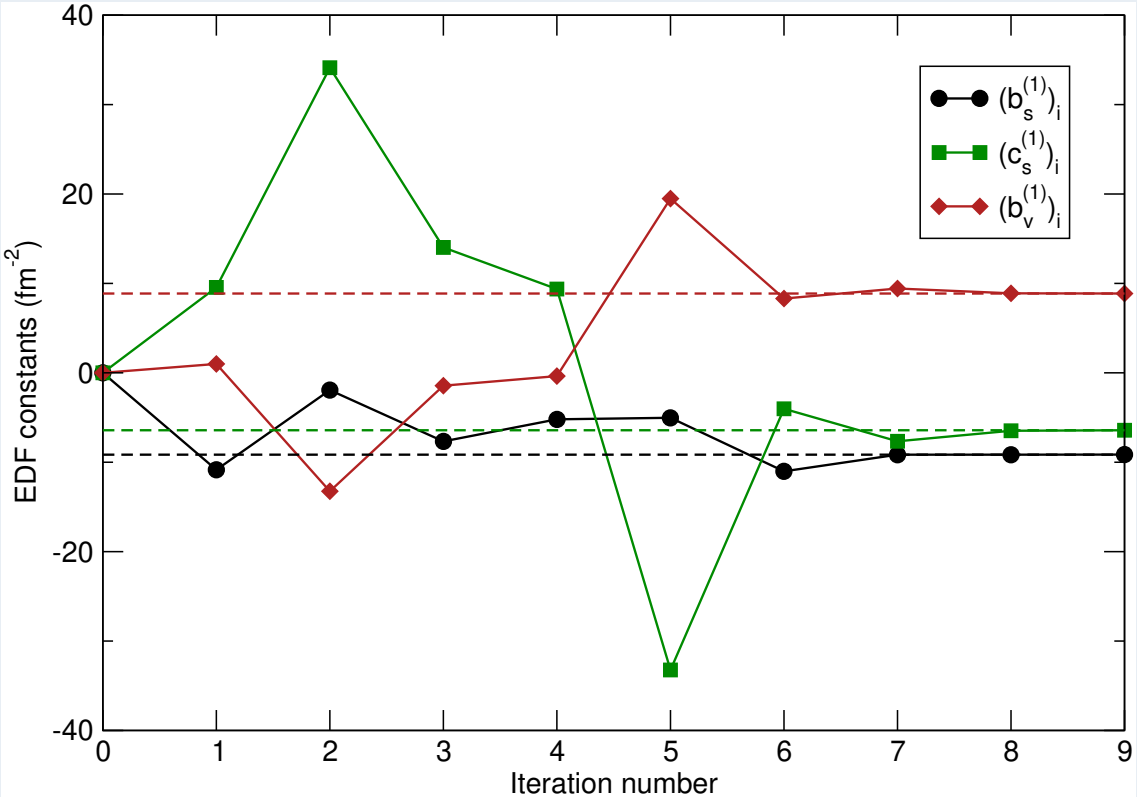
$$b_{\mathrm{S}} = \sum_k b_{\mathrm{S}}^{k\text{-th}} \qquad c_{\mathrm{S}} = \sum_k c_{\mathrm{S}}^{k\text{-th}} \qquad b_{\mathrm{V}} = \sum_k b_{\mathrm{V}}^{k\text{-th}}$$



Well reproduced!!

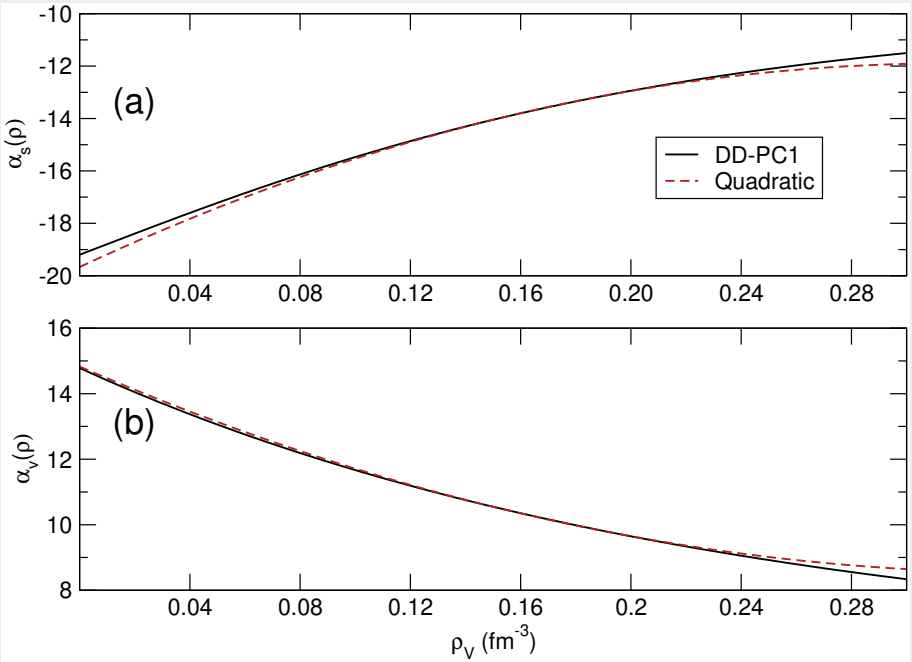
Accorto, Naito, Liang, Nikšić, and Vretenar. *Phys. Rev. C* **103**, 044304 (2021)

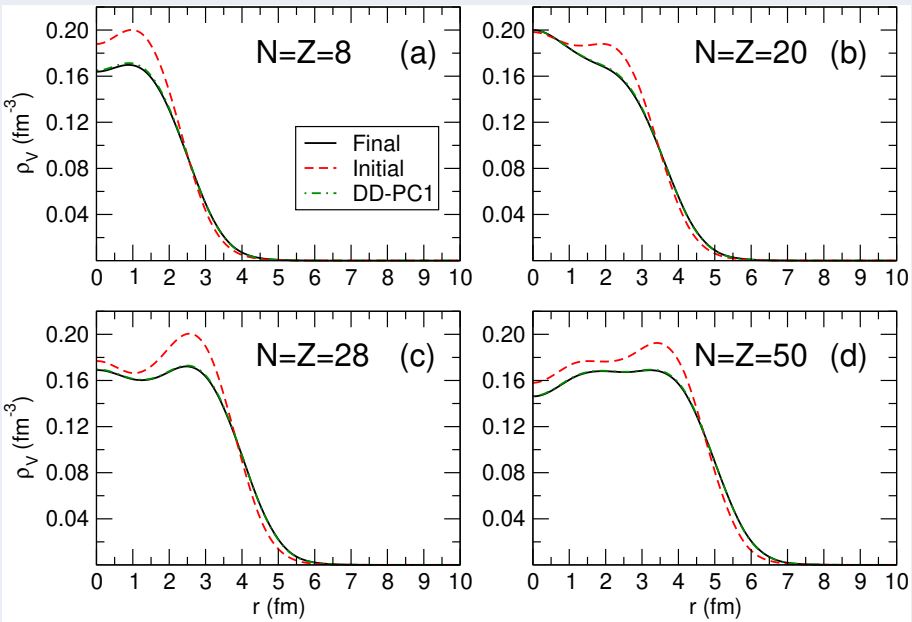
Benchmark Results (2)

Iterative method with **different ansatz** is used

$$\alpha_{\mathrm{S}}(\rho_{\mathrm{V}}) = a_{\mathrm{S}} + \sum_k b_{\mathrm{S}}^{k\text{-th}} \left(\frac{\rho_{\mathrm{V}}}{\rho_{\mathrm{sat}}} - 1\right) + \sum_k c_{\mathrm{S}}^{k\text{-th}} \left(\frac{\rho_{\mathrm{V}}}{\rho_{\mathrm{sat}}} - 1\right)^2$$

$$\alpha_{\mathrm{V}}(\rho_{\mathrm{V}}) = a_{\mathrm{V}} + \sum_k b_{\mathrm{V}}^{k\text{-th}} \left(\frac{\rho_{\mathrm{V}}}{\rho_{\mathrm{sat}}} - 1\right) + \sum_k c_{\mathrm{V}}^{k\text{-th}} \left(\frac{\rho_{\mathrm{V}}}{\rho_{\mathrm{sat}}} - 1\right)^2$$





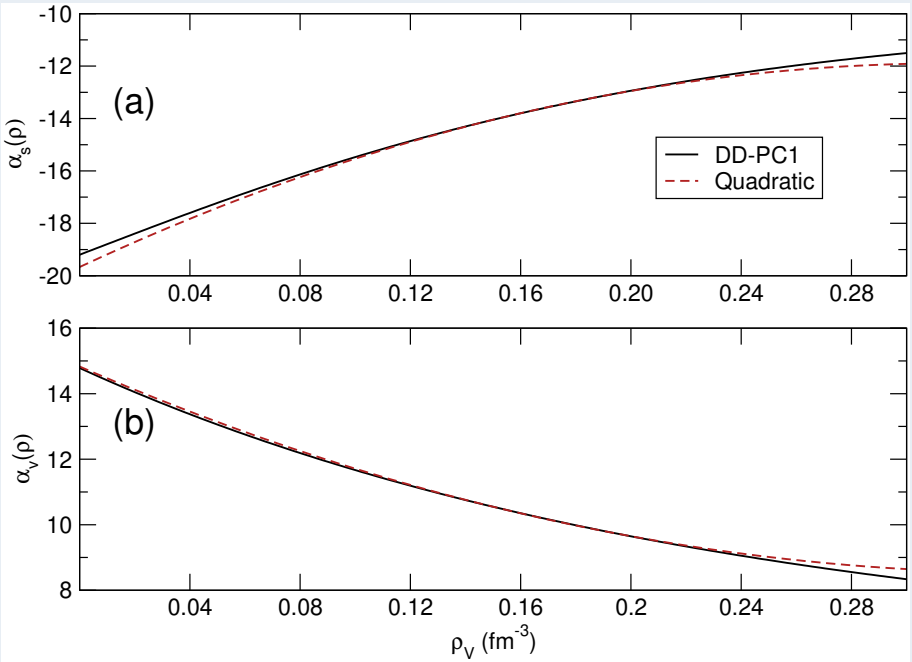
Accorto, Naito, Liang, Nikšić, and Vretenar. *Phys. Rev. C* **103**, 044304 (2021)

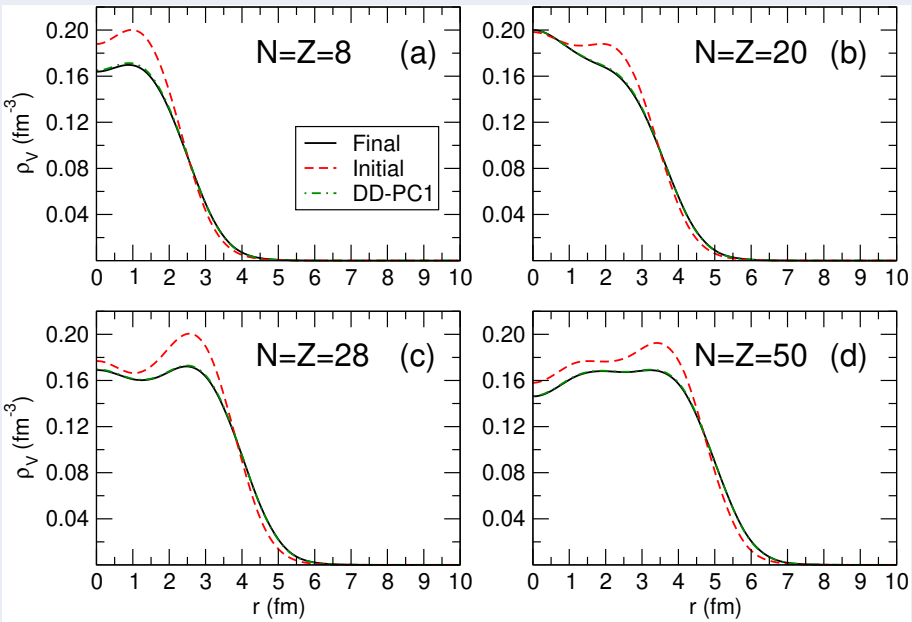
Benchmark Results (2)

Iterative method with **different ansatz** is used

$$\alpha_S(\rho_V) = a_S + \sum_k b_S^{k\text{-th}} \left(\frac{\rho_V}{\rho_{sat}} - 1\right) + \sum_k c_S^{k\text{-th}} \left(\frac{\rho_V}{\rho_{sat}} - 1\right)^2$$

$$\alpha_V(\rho_V) = a_V + \sum_k b_V^{k\text{-th}} \left(\frac{\rho_V}{\rho_{sat}} - 1\right) + \sum_k c_V^{k\text{-th}} \left(\frac{\rho_V}{\rho_{sat}} - 1\right)^2$$





Even with different ansatz, well reproduced!!

Accorto, Naito, Liang, Nikšić, and Vretenar. *Phys. Rev. C* **103**, 044304 (2021)

Conclusion and Perspectives

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- A method to improve an EDF using known *exact* densities is proposed **IKS-DFPT**
- Inverse KS method and IKS-DFPT are extended to relativsitc systems
- IKS-DFPT works well both for atomic systems and for nuclear systems in benchmark calculations

Perspectives

- How can we constrain spin-orbit potential?
- Can we go beyond benchmark calculations?
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Thank you for attention!!