Inverse Kohn-Sham using Moreau-Yosida proximal-point algorithm

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Density-functional theory

$$\psi : \mathbb{R}^{3N} \to \mathbb{C} \qquad \rho_\psi = N \int_{\mathbb{R}^{3(N-1)}} |\psi|^2 \qquad \langle v,\, \rho\rangle = \int v\rho$$

$$E(v) = \inf_\psi \langle \psi | \hat{H}_0 + \sum_j v(\mathbf{r}_j) | \psi \rangle = \inf_\rho \{F(\rho) + \langle v,\, \rho\rangle\}$$

$$F(\rho) = \sup_v \{E(v) - \langle v,\, \rho\rangle\} = \inf_{\Gamma\mapsto\rho} \mathrm{Tr}\, \hat{H}_0 \Gamma = \inf_{\sum_j p_j \rho_{\psi_j} = \rho} \sum_j p_j \langle \psi_j | \hat{H}_0 | \psi_j \rangle$$

Kohn–Sham (KS) scheme

Interacting electrons

$$\hat{H} = -\frac{1}{2}\sum_j \nabla_j^2 + \frac{1}{2}\sum_{k\neq j} \frac{1}{|\mathbf{r}_j - \mathbf{r}_k|} + \sum_j v_{\mathrm{ext}}(\mathbf{r}_j)$$

ρ_{gs}

Non-interacting electrons (KS system)

$$\hat{H}_{\mathrm{KS}} = -\frac{1}{2}\sum_j \nabla_j^2 + \sum_j [v_{\mathrm{ext}}(\mathbf{r}_j) + v_{\mathrm{H}}(\mathbf{r}_j) + v_{\mathrm{xc}}(\mathbf{r}_j)]$$

Unknown: v_{xc}

Approximation: $\tilde{E}_{\mathrm{xc}} \implies \tilde{v}_{\mathrm{xc}} = \frac{\delta}{\delta\rho}\tilde{E}_{\mathrm{xc}}$

Forward KS problem: Given approximate $\tilde{v}_{\mathrm{xc}}$, obtain ρ_{gs}

Inverse KS problem: Given ρ_{gs}, obtain exact v_{xc}

Moreau–Yosida Regularisation

Duality mapping $J_{\mathrm{dual}} : \mathcal{D} \to \mathcal{V}$

$\mathcal{D}$ = density space, $\mathcal{V} = \mathcal{D}^*$ potential space, $\mathcal{D} \ni \rho \mapsto J_{\mathrm{dual}}[\rho] \in \mathcal{V}$

Moreau–Yosida regularisation:

- $\mathcal{D}$ uniformly convex, $F : \mathcal{D} \to \mathbb{R} \cup \{+\infty\}$ convex, lower semicontinous
$$F^\varepsilon(\rho) = \inf_{\tilde\rho\in\mathcal{D}} \left\{F(\tilde\rho) + \tfrac{1}{2\varepsilon}\|\tilde\rho - \rho\|^2\right\}, \quad \varepsilon > 0$$
- $F^\varepsilon(\rho)$ is differentiable $\implies$
Every ρ is v-representable $\boxed{\varepsilon\text{-HK mapping: } v^\varepsilon[\rho] = -\frac{\delta F^\varepsilon}{\delta\rho}[\rho]}$
- Lossless regularization:
$$E^\varepsilon(v) = \inf_{\rho\in\mathcal{D}} \{F^\varepsilon(\rho) + \langle v, \rho\rangle\} \quad \implies \quad E(v) = E^\varepsilon(v) + \frac{\varepsilon}{2}\|v\|^2$$

Quantitative Hohenberg–Kohn

$\mathcal{D}$ is p-smooth if $r(t) \leq ct^p$,

$$r(t) = \sup\left\{\frac{1}{2}(\|\rho+\tilde\rho\| + \|\rho-\tilde\rho\|) - 1 : \|\rho\| = 1, \|\tilde\rho\| = t\right\}$$

Theorem. [1] $\mathcal{D}$ strictly convex, p-smooth with $p \leq 2$: The ε-HK mapping is Hölder continuous

$$\|v^\varepsilon[\rho] - v^\varepsilon[\tilde\rho]\| \leq \frac{C}{\varepsilon^{p-1}}\|\rho - \tilde\rho\|^{p-1}, \quad \varepsilon > 0$$

Proximal point

ρ^ε proximal point of density ρ

$$\rho^\varepsilon = \operatorname*{argmin}_{\tilde\rho\in\mathcal{D}} \left\{F(\tilde\rho) + \tfrac{1}{2\varepsilon}\|\tilde\rho - \rho\|^2\right\}, \quad \varepsilon > 0$$

- $\rho^\varepsilon \to \rho$ as $\varepsilon \to 0^+$, $\|\rho^\varepsilon - \rho\| = \mathcal{O}(\varepsilon^{1/2}) \quad (\varepsilon \to 0^+)$
- Non-expansiveness:
$$\|\rho^\varepsilon - \tilde\rho^\varepsilon\|_{\mathcal{D}} \leq \|\rho - \tilde\rho\|_{\mathcal{D}}, \quad \forall \rho, \tilde\rho \in \mathcal{D}$$

Moreau–Yosida inverse KS

$T(\rho)$ kinetic energy, $E_{\mathrm{H}}(\rho)$ Hartree energy

$$\mathcal{F}(\rho) = T(\rho) + E_{\mathrm{H}}(\rho) + \langle v_{\mathrm{ext}}, \rho\rangle$$

$\mathcal{F}^\varepsilon(\rho)$ differentiable $\implies$

$$v_{\mathrm{xc}}^\varepsilon = -\frac{\delta}{\delta\rho}\mathcal{F}^\varepsilon(\rho_{\mathrm{gs}}^\varepsilon) = \tfrac{1}{\varepsilon}J_{\mathrm{dual}}[\rho_{\mathrm{gs}}^\varepsilon - \rho_{\mathrm{gs}}]$$

Theorem. [2, 3] Exchange-correlation potential obtained from

$$v_{\mathrm{xc}}(\mathbf{r}) = \lim_{\varepsilon\to 0^+} v_{\mathrm{xc}}^\varepsilon(\mathbf{r}) = \lim_{\varepsilon\to 0^+} \tfrac{1}{\varepsilon}J_{\mathrm{dual}}\left[\rho_{\mathrm{gs}}^\varepsilon - \rho_{\mathrm{gs}}\right](\mathbf{r})$$

The form of J_{dual} depends on the choice of $\mathcal{D}$

Sketch of proof

Assumption: v_{KS} exists for given ρ_{gs}

$$\min_\rho (T(\rho) + \langle v_{\mathrm{KS}}, \rho\rangle) \iff \underline{\partial} T(\rho_{\mathrm{gs}}) + v_{\mathrm{KS}} \ni 0$$

$$\min_\rho (\mathcal{F}(\rho) + \langle v_{\mathrm{xc}}, \rho\rangle) \iff \underbrace{\underline{\partial}\mathcal{F}(\rho_{\mathrm{gs}}) + v_{\mathrm{xc}} \ni 0}_{(1)}$$

Moreau-Yosida regularisation:

$$\min_\rho \left\{\mathcal{F}(\rho) + \tfrac{1}{2\varepsilon}\|\rho - \rho_{\mathrm{gs}}\|_{\mathcal{D}}^2\right\} \iff \underbrace{\underline{\partial}\mathcal{F}(\rho_{\mathrm{gs}}^\varepsilon) + \tfrac{1}{\varepsilon}J_{\mathrm{dual}}(\rho_{\mathrm{gs}}^\varepsilon - \rho_{\mathrm{gs}}) \ni 0}_{(2)}$$

(1), (2) and $\rho_{\mathrm{gs}}^\varepsilon \to \rho_{\mathrm{gs}}$ as $\varepsilon \to 0^+ \implies v_{\mathrm{xc}} = \lim_{\varepsilon\to 0^+} \tfrac{1}{\varepsilon}J_{\mathrm{dual}}[\rho_{\mathrm{gs}}^\varepsilon - \rho_{\mathrm{gs}}]$

Obtaining the proximal point

Minimise $\mathcal{E}(\rho\,; \rho_{\mathrm{gs}}) = \mathcal{F}(\rho) + \frac{1}{2\varepsilon}\|\rho - \rho_{\mathrm{gs}}\|_{\mathcal{D}}^2 \qquad (\rho_{\mathrm{gs}}$ fixed)

Iterative procedure:

1. Fix $\{\varepsilon_k\}_k$, $\varepsilon_k \to 0$ as $k \to \infty$, mixing parameter $0 < \mu < 1$
2. Determine (e.g., with $v_0^{(k)} = 0$)
$$\rho_i^{(k)} = \operatorname{argmin}\left\{\mathcal{F}(\rho) + \langle v_i^{(k)}, \rho\rangle\right\}$$
and compute
$$v_{i+1}^{(k)} = (1-\mu)v_i^{(k)} + \tfrac{\mu}{\varepsilon_k}J[\rho_i^{(k)} - \rho_{\mathrm{gs}}]$$
3. Extrapolate $k \to \infty$

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